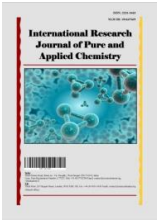

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Distribution and Health Risk Assessment of Phthalate Acid Esters in Bottled Drinking Water from Selected Brands in Delta State, Southern Nigeria

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Authors' contributions

This work was carried out in collaboration among all authors. Author OE designed the study, wrote the protocol and managed the analyses of the study. Authors OEO and ACI managed the literature searches and wrote the first draft of the manuscript. Author OEO performed the statistical analysis. All authors read and approved the final manuscript.

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ABSTRACT

Phthalate acid esters (PAEs) are widely used as plasticizers. However, they have the potential to migrate from plastic materials into the environment where they pose detrimental health impacts. Hence, they have gained wide attention. This study was therefore carried out to investigate the distribution and health risk assessment of PAEs in different bottled drinking water brands in Delta State, Southern Nigeria. Samples were collected from ten (10) bottled drinking water brands across eight (8) local government areas of the State. The 6-EPA controlled PAEs (DMP, DEP, DBP, BBP, DEHP and DNOP) and one uncontrolled PAEs (BEP) were investigated. The individual PAE concentrations ranged from 0.005 – 2.738 µg/L while the total concentration of PAEs in the different brands ranged from 0.023 - 4.361 µg/L. The predominant PAEs across the ten brands were DMP and BEP however DBP had the highest total concentration (4.392 µg/L) making it the main contributor to PAE levels in bottled drinking water brands. Analysis of variance showed no significant difference in the concentration of PAEs detected in the different bottled drinking water

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brands (p>0.05). Health risk assessment indicated little non-carcinogenic risks from five controlled PAEs and little carcinogenic risk from DEHP. Continuous monitoring is however recommended to ensure than PAE levels in bottled drinking water remain within permissible limits. Our results therefore provides important information for the understanding of the distribution and potential health risks of PAEs in different bottled drinking water brands in Delta State.

Keywords: Phthalates acid esters; bottled drinking water; health risk assessment; GC-MS analysis.

1. INTRODUCTION

The consumption of bottled drinking water has increased rapidly in many developing countries because urbanization, intensive agriculture, recreation, and the manufacturing industry have continued to affect global water quality. In Nigeria, pollution of inland water bodies and coastal waters, poor waste disposal, oil drilling activities as well as other industrial activities affects suitability of drinking water sources. This has resulted in people purchasing sachet and bottled drinking water from private companies on the assumption that they offer safe drinking water. Safe drinking water is that which does not present any significant health risk over a lifetime consumption, including any sensitivities that may occur in different stages of life. It is water which is free from pathogenic microbes, hazardous chemicals/substances and aesthetically acceptable (i.e. pleasing to sight, odorless and tasteless). It is important that this type of water should not only be available, but also be available in enough quantity all the time [1].

Polyethylene terephthalate (PET) is a thermoplastic polymer widely used in commercial bottles for drinking water due to its cheaper cost of production [2]. However, studies have shown that PET bottling could leach endocrine disruptors such as phthalate acid esters (PAEs) under daily use conditions [3,4]. PAEs are non-reactive plasticizers, produced industrially and used in the manufacture of plastics to increase their durability, transparency, flexibility, and longevity [5]. PAEs are also used in a wide range of PVC applications including building and construction, flooring, and electric cables, and non-PVC applications such as paints, coatings, rubber products, adhesives and sealants [6,7,8]. They accounted for 65% of the world consumption of plasticizers in 2017. PAEs also serve as solvents and emulsifiers used in pharmaceuticals, pesticides, health and beauty products as well as children's toys. PAEs have become one of the highest yielding chemicals in the world with global production approximately amounting to 6 million tons per year, because of their wide applications and the growing demand [9,10].

Many PAEs and their metabolites cause adverse effects on reproduction and development such as testicular and sexual differentiation effects in humans and animals [11]. Epidemiological and toxicity studies have shown that PAEs have adverse effects, such as estrogenicity, reproductive, hepatic, neurobehaviour, and immune toxicity [12,13]. Eight PAEs, namely diethyl phthalate (DEP), dibutyl phthalate (DBP), butyl benzyl phthalate (BBP), bis(2-ethylhexyl) phthalate (DEHP), dicyclohexyl phthalate (DCHP), dipentyl phthalate (DPP), dihexyl phthalate (DHXP), and di-n-propyl phthalate (DPrP), have been recognized as endocrine disruptors. A main route of exposure to phthalates is via water use. Several studies have occasionally detected these compounds in underground water [14,15,16,17] and also bottled drinking water [18,19]. Phthalate esters are often found in water, soil, air, food products and the human body [20]. The fact that some phthalates have been found in environmental samples, shows they can be released during use and migrate from plastic materials to contaminate their contents such as water, food and beverages [21,22,23,24].

Considering the environmental and human adverse effects, six PAEs, including dimethyl phthalate (DMP), dinoctyl phthalate (DNOP), DEP, DBP, BBP, and DEHP, have been added to the priority pollutant list of the United States Environmental Protection Agency (USEPA) [25]. There is a need for efforts to monitor these and other phthalates in the environment, especially in drinking water. One of the targets of the millennium development goals (MDG's) in terms of healthy living for the masses can be achieved through the supply of safe water [26]. Therefore, the aim of this survey was to investigate the distribution and health risk of 6 controlled and 1 uncontrolled PAEs in PET bottled drinking water cross Delta State, Southern Nigeria. It should be noted that there is paucity of comprehensive data on the occurrence of PAEs in bottled drinking

water in Delta State specifically and Nigeria at large.

2. MATERIALS AND METHODS

2.1 Study Area

The study was carried out in Delta State. The State lies approximately between longitude 5’00 and 6’ 45 East and Latitude 5’00 and 6’30 North. It is generally a low-lying without remarkable hills and lies within the humid tropical zone with defined dry seasons (November – March) and rainy seasons (April - October). The rainy season is brought by South-West Trade Wind blowing across the Atlantic Ocean, while the dry dusty and often referred to the North- East Trade Wind blowing across the Sahara Desert dominates the dry season. The state is made up of about 25 local government areas. It is situated in the south-south geo-political zones with a land mass of 18,050 km^2 of which more than 60% consist of lands and the rest consist basically of river, creeks and mangroves.

2.2 Sample Collection and Storage

Sampling was carried out in May to June, 2019. Bottled drinking water samples were collected from ten (10) bottled drinking water factories located at Asaba, Agbor, Ekpan, Sapele, Oshimili, Effurun, Udu and Ughelli town, spreading across eight (8) local government areas of the State namely Oshimili North and South, Ika South, Uvwie, Sapele, Udu, Ughelli North and South local government areas. Details about the sampling locations are presented in Table 1.

2.3 Sample Handling and Pre-Treatment

Water samples were collected in amber glass containers. Conventional sampling practices were followed. No special sample preservations and storage steps were taken since phthalate are stable at pH 7. Samples were collected and sealed with a glass stopper followed by a metal clip. The samples were refrigerated at 4°C free from light from the time of collection until extraction.

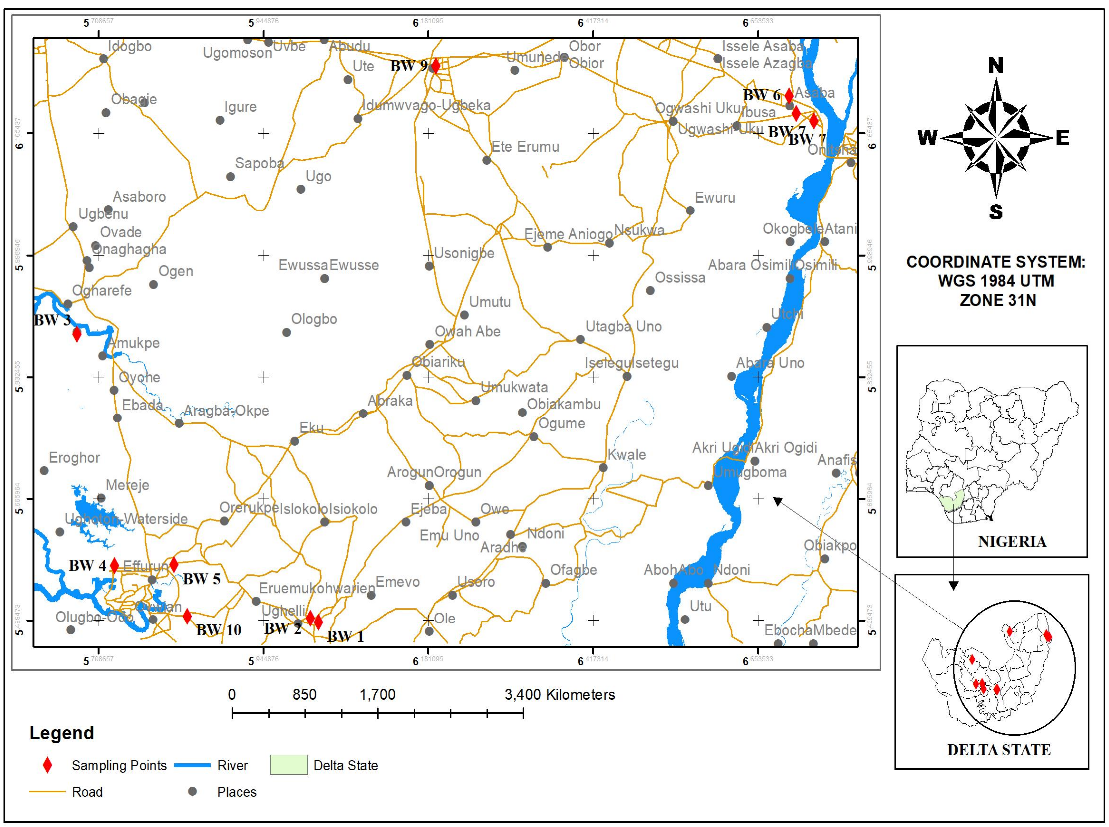

Fig. 1. GIS based Map of Study Area

2.4 Safety Measures

All procedures and associated reagent used during this research followed Standard risk assessment protocols specified by Bachema Analytical Laboratories, Switzerland and where this work was carried out. The new method for analyses of Phthalates in water samples using GC/MS was validated according to Institute Bachema AG Quality Management Guidelines as certified by ISO 17025 (Institute of Bachema QRL-Verziechnis, Verse 04).

2.5 Reagents

Anhydrous Sodium Sulphate oven heated at 400˚C (Fluka), cyclohexane distillated in an all glass (Scharalau), Acetone GC (Scharalau), 1,11-Dibromoundecan ISTD(Labor Dr. Ehrenstofer), Reagent water (Nanopure/water-Barnstead), ISTD- solution: 25µL Dibromoundecan in 250 ml cyclohexane. (1ml of this solution contains Approximate 10µg ISTD). Phthalate Esters were mix 0.2mg/ml in methanol (Accu standards M606), Phthalates and Adipate Esters mix 1, 100ng/l (Labor Dr. Ehrenstofer). All standards were stored at 4°C.

2.6 Extraction of Water Samples

A subsample of 1 L was weighed into a calibrated Erlenmeyer flask of 1 L. To this is added 1ml Cyclohexane internal Standard solution and then 9 ml Cyclohexane. With the aid of a glass coated magnet the sample was then liquid / liquid intensively extracted for a minimum of an hour. Using an upward delivery glass-separating funnel, the solvent Extract was then isolated in a 10 ml vial. Sample volume is 1 L in 10 ml Cyclohexane Extract [27].

2.7 Blank Samples

A 1 L aliquot nanopure water is treated exactly as a sample including exposure to all glass wares, equipment, solvents. This is done by the injection of the extracted blanks at the beginning of all the analytical series.

2.8 Chemicals and Materials

Anhydrous sodium sulphate oven heated at 400°C; cyclohexane distillate in an all glassware; acetone; 1,11-Dibromoundecane (ISTD); reagent water; Agilent gas chromatograph 6890N coupled with MS; sample collection bottles; 1l Erlenmeyer flasks; glass coated metal magnetic mixer; Upward delivery glass separating funnel; 10 ml vials with septum seals (Silicon/PTFE); Balance (3 decimal places); glass beakers; spatulas; forceps; aluminum foil; solvent reservoir bottle; calibrated syringe; ultrasonic bath; Bunsen burner; drying oven; muffle furnace; desiccators; measuring cylinders; test tubes; Pasteur pipettes; calibrated pipette.

2.9 Quality Control

Distilled water used during experimental process was analyzed separately for all phthalates acid esters and used as control.

2.10 Analysis of Phthalates in Water Using GC/MS

Analysis was carried out using gas chromatography with mass spectrometry (GC/MS). The analytes in the extract were separated by means of capillary gas chromatography using temperature programming. The chromatographically separated phthalate acid esters (Table 2) were detected and measured with mass spectrometer. This process is made possible by large volume injection (LVI) technique used to introduce 10µl of the extract at a rate of approx. 4.6ul/sec into the injector. The injector is kept at an initial temperature of 78°C and rises gradually at the rate of 5°C/sec to 300°C so as to enable complete volatilization of solvent into the analytical column. Oven initial temperature was kept at 70°C and on injection rises to 300°C at a rate of 20°C/min. Injector's temperature is brought back quickly to normal by the use of a liquid Nitrogen cryo-cooling unit. The new method for analysis of Phthalate in water samples using GC/MS has been validated [28,29,30].

2.11 Statistical Analysis

Significant differences in PAE concentrations among samples were analyzed using one-way analysis of variance in Microsoft Excel. Significant difference was considered at ($p < 0.05$).

2.12 Health Risk Assessment

Non-cancer hazard quotient (HQ) and excess cancer risks (ECR) via drinking water ingestion for adults [31,32,33] were assessed by the equations:

$$EDI = MC \times WC \quad (1)$$

$$HQ = \frac{EDI}{RfD} \quad (2)$$

$$ECR = \text{drinking water unit risk} \times MC \quad (3)$$

where EDI was the estimated daily intake through drinking water (µg/kg body weight/day) and MC was the maximum concentration of PAEs in µg/L determined in PET-bottled drinking water samples. The daily water consumption (WC) was calculated based on the body weight and the dietary reference values for water. An average body weight of 60 kg for adults and 2 L/day as the dietary reference value for water consumption were assumed [34]. The ingestion reference dose values (RfD) (µg/kg body weight/day) were obtained from the USEPA Integrated Risk Information System [35]. Among the phthalates considered, DEHP was the only one to be probably carcinogenic for humans. The reference carcinogenic unit risk from drinking water was 4.0×10^{-7} per µg/L. An HQ of 1 or less in the event that only one contaminant and/or exposure route was assessed indicates that the receptor's exposure was equal to or less than "allowable" exposure and adverse health effects were considered unlikely to occur. An ECR less than 10^{-6} is typically considered negligible.

3. RESULTS AND DISCUSSION

3.1 PAE Levels in Bottled Drinking Water

Seven (7) phthalate acid esters in ten (10) bottled drinking water brands in Delta State were investigated in this study, and the concentrations are presented in Table 3 and Fig. 2. The range, mean and total concentrations for the seven PAEs across all the brands are presented in Table 4.

Of the seven (7) PAEs, only one - DMP - was detected in all the bottled drinking water brands. BEP was detected in 7 brands, BBP was detected in 5 brands, DEHP and DNOP were detected in 4 brands, while DBP and DEP were detected in 3 brands. All the PAEs were detected in brands BW 1 and BW 5 while all except DMP were below the limits of detection in BW 4, 8 and 9.

The individual PAE concentrations ranged from 0.005 – 2.738 µg/L in all the brands with DPB in BW 1 having the highest value and DEHP in BW 7 having the least value.

Table 1. Sample locations and their GPS coordinates

S/No	Sample area	LGA	Sample code	Sampling coordinates	
				Northings (N)	Eastings (E)
1	Ughelli	Ughelli North	BW 1	05˚30'03.6"	006˚00'46.2"
2	Ughelli	Ughelli South	BW 2	05˚30'07.8'	006˚00'46.2"
3	Sapele	Sapele	BW 3	05˚53'27.4'	005˚40'41.6"
4	Ekpan	Uvwie	BW 4	05˚34'27.4"	005˚43'57.2"
5	Effurun	Uvwie	BW 5	05˚34'33.0"	005˚49'3.0"
6	Asaba	Oshimili South	BW 6	06˚12'59.2"	006˚41'55.5"
7	Asaba	Oshimili South	BW 7	06˚10'57.7"	006˚44'4.4"
8	Oshimili	Oshimili North	BW 8	06˚11'33.1"	006˚42'33.9"
9	Agbor	Ika South	BW 9	06˚15'28.0"	006˚11'35.0"
10	Udu	Udu	BW 10	05˚30'18.0	005˚50'12.0"

Table 2. Target compounds for laboratory analysis

Name	Abbreviation
Butyl benzyl phthalate	BBP
Butyl ethyl phthalate	BEP
Di(2-ethylhexyl) phthalate	DEHP
Dibutyl phthalate	DBP
Diethyl phthalate	DEP
Dimethyl phthalate	DMP
Di-n-octyl phthalate	DNOP

**Abbreviation created for reference only in this study, no known abbreviation found in literature*

Table 3. Concentration (µg/L) of phthalate esters in different bottled drinking water brands in Warri Metropolis

S/N	Samples	BBP	BEP	DEHP	DBP	DEP	DMP	DNOP	Min	Max	Sum
1	BW 1	0.064	1.360	0.056	2.738	0.103	0.027	0.013	0.027	2.738	4.361
2	BW 2	0.021	1.357	<DL	<DL	<DL	0.033	0.016	0.021	1.357	1.427
3	BW 3	0.021	0.150	0.012	1.252	<DL	0.040	<DL	0.012	1.252	1.475
4	BW 4	<DL	<DL	<DL	<DL	<DL	0.035	<DL	-	0.035	0.035
5	BW 5	0.014	0.699	0.008	0.402	0.023	0.053	0.021	0.008	0.699	1.220
6	BW 6	<DL	0.063	0.005	<DL	0.010	0.034	0.006	0.005	0.063	0.118
7	BW 7	<DL	0.065	<DL	<DL	<DL	0.035	<DL	0.035	0.065	0.100
8	BW 8	<DL	<DL	<DL	<DL	<DL	0.027	<DL	-	0.027	0.027
9	BW 9	<DL	<DL	<DL	<DL	<DL	0.023	<DL	-	0.023	0.023
10	BW 10	0.008	0.308	<DL	<DL	<DL	0.030	<DL	0.008	0.308	0.346

BW 1 – 10 represents the different bottled drinking water brands in Delta State; <DL represents concentrations below the limits of detection

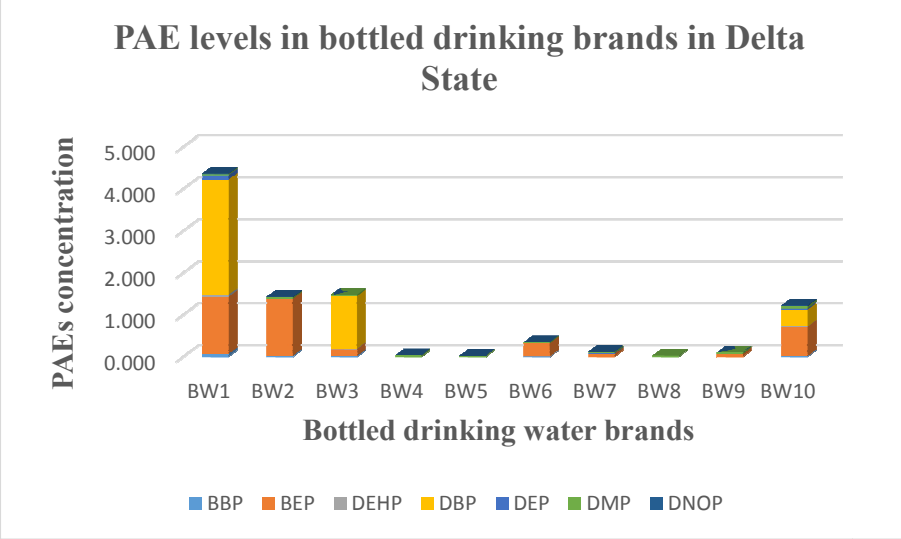

Fig. 2. Concentration of PAEs in ten bottled drinking water brands in Delta State

The single factor analysis of variance (ANOVA) showed that there is no significant difference in the concentration of PAEs detected in the different bottled drinking water brands ($p > 0.05$) (Table 5). The total PAEs in the different brands ranged from 0.023 - 4.361 µg/L with the highest recorded in BW 1 and the lowest in BW 9. The total concentration of PAEs in the various brands in descending order were BW 1 > BW 3 > BW 2 > BW 5 > BW 10 > BW 6 > BW 7 > BW 4 > BW 8 > BW 9 (Table 3).

The percentage (%) composition of the different PAEs in the different bottled drinking water brands in Delta State was determined. The result showed that the most predominant PAE in BW 1 was DBP (62.79%) while the least was DNOP (0.30%). In BW 2, the highest was BEP (95.12%) while the lowest was DNOP (1.12%). For BW 3, the most predominant was DBP (84.90%) while the least was BBP (1.40%). Moreover, in BW 4, BW 5 and BW 8, DMP was the only detected PAE hence it accounted for 100% of the composition. For BW 6, the most predominant PAE was BEP (89.10%) while the least predominant was BBP (2.22%). In BW 7, the highest was BEP (53.24%) while the lowest was DEHP (4.23%). In BW 9, the highest was BEP (65.0%) while the lowest was DMP (35.0%). In BW 10, the highest was BEP (57.28%) while the lowest was DNOP (1.12%). Among the PAEs, BEP was the most abundant having the highest percentage composition in 5 brands namely BW 2, BW 6, BW 7, BW 9 and BW 10; DMP had the highest predominance in 3 brands which were BW 4, BW 5 and BW 8, while DBP

was the highest in 2 brands namely BW 1 and BW 3.

For the six EPA-controlled PAEs (DMP, DEP, DBP, BBP, DEHP and DNOP), the total concentrations (Σ_6PAEs) were 0.023 – 3.001 µg/L, accounting for 10.98 – 100% of Σ_7PAEs. Among them, DBP had the highest total concentration (4.392 µg/L) making it the main contributor to PAEs in bottled drinking water in Delta State. It ranged from 0.402 to 2.378 µg/L (Table 4), accounting for 7.84 – 53.87% of the total amount of the six EPA-controlled PAEs (5.13 µg/L) which is lower than previous reports [36,2,37,38]. The maximum concentration of 2.738 µg/L was higher than Guo et al. [32], Zaki [39], Keresztes et al. [37], and Yang et al. [40]. The concentrations of DMP ranged from 0.023 - 0.053 µg/L, with a mean value (0.034 µg/L) (Table 4) lower than that reported by Li et al. [38]. The maximum concentration of 0.053 µg/L was higher than Yang et al. [40] but lower than Li et al. [38] and Montuori et al. [41]. DEP levels ranged from 0.01 to 0.103 µg/L with mean value (0.045 µg/L) (Table 4) which is lower than those reported in Italy [41] and Spain [42] but higher than that reported in China [38]. The maximum concentration of 0.103 µg/L were similar to Cao [36] and Psillakis and Kalogerakis [43], higher than Amiridou and Voutsa [31], Li et al. [38], and Yang et al. [40] but lower than Montuori et al. [41] and Guart et al. [42]. DNOP levels ranged from 0.006 - 0.021 µg/L with mean value (0.014 µg/L) (Table 4). The maximum concentration of 0.021 µg/L was lower than those reported in China [38, 42]. The levels of BBP and DEHP were the lowest. Table 4 shows that BBP levels ranged from 0.008 - 0.064 µg/L with mean value (0.026 µg/L) similar to that although reported by Li et al. [38] and much lower than the US EPA standard of 100 µg/L, respectively [44]. The maximum concentration of 0.064 µg/L was higher than Li et al. [38] but lower than Keresztes et al. [37], Yang et al. [40] and Guart et al. [42]. DEHP levels ranged from 0.005 - 0.056 µg/L with mean value (0.020 µg/L) (Table 4). The maximum concentration of 0.056 µg/L was higher than that reported by Li et al. [38] but lower than those reported by Zaki [39], Yang et al. [40] and Guart et al. [42]. The mean concentration of DEHP was lower than USEPA standards of 6 µg/L [44] and WHO standards of 8 µg/L [45].

The uncontrolled PAE (BEP), the concentration ranged from 0.063 - 1.360 µg/L with a mean value of 0.572 µg/L (Table 4). It is important to note that the mean and maximum values of BEP were higher than those of the EPA controlled PAEs except DBP. This implies that BEP was the second most significant PAEs detected in bottled drinking water brands. This emphasizes the assertion by Li et al. [38] that human exposure to the uncontrolled PAEs should be given more attention in future researches.

3.2 Health Risk Assessment

The human health risks could only be estimated for five controlled PAEs for which toxicity data were available. The estimated hazard quotient (HQ) and excess cancer risk (ECR) of these five PAEs are summarized in Table 6.

HQ values ranging from 4.10×10^{-6} – 8.76×10^{-4} were far less than the recommended limit of 1, indicating little non-carcinogenic health risks from the five controlled PAEs. This was is in agreement with the study of Li et al. [38]. Furthermore, according to the International Agency for Research on Cancer, only DEHP is possibly carcinogenic to humans (IARC Group2B). The ECR for DEHP (2.24×10^{-8}) was below the acceptable risk level of 10^{-6}, suggesting the carcinogenic health risk is extremely low for consumers exposed to this

Table 4. PAEs levels (µg/L) in bottled drinking water in Delta State (n=10)

PAEs	Range	Mean	Sum	Maximum levels in other researches
BBP	0.008 - 0.064	0.026	0.128	0.032[a], 0.11[c], 0.10[e], 1.28[j]
DEHP	0.005 - 0.056	0.020	0.081	0.021[a], 4.39[c], 0.30[g], 1.52[j]
DBP	0.402 - 2.738	1.464	4.392	1.02[c], 0.80[e], 0.046[f], 0.17[g]
DEP	0.01 - 0.103	0.045	0.136	0.071[a], 0.35[b], 0.024[c], 0.070[d], 0.10[h], 0.12[i], 20.5[j],
DMP	0.023 - 0.053	0.034	0.337	0.41[a], 0.10[b], 0.023[c]
DNOP	0.006 - 0.021	0.014	0.056	0.039[a], 0.27c
BEP	0.063 - 1.360	0.572	4.002	-

[a]*Li* et al. *(2019);* [b]*Montuori* et al. *(2008);* [c]*Yang* et al. *(2017);* [d]*Amiridou and Voutsa (2011);* [e]*Keresztes* et al. *(2013);* [f]*Guo* et al. *(2012);* [g]*Zaki (2015);* [h]*Cao (2008);* [i]*Psillakis and Kalogerakis (2003);* [j]*Guart* et al. *(2014)*

Table 5. Single factor ANOVA

Source of Variation	*SS*	*df*	*MS*	*F*	*P-value*	*F crit*
Between Groups	2.3611	9	0.2623	1.5718	0.1445	2.0401
Within Groups	10.0141	60	0.1669			
Total	12.3752	69				

Table 6. Health risk assessment of PAEs (µg/L) for adults

	DEHP	DEP	DBP	BBP	DNOP
MC	0.056	0.103	2.738	0.064	0.021
WC	0.032	0.032	0.032	0.032	0.032
EDI	0.0018	0.0033	0.0876	0.0020	0.0007
RfD	20	800	100	200	10
HQ	8.96×10^{-5}	4.10×10^{-6}	8.76×10^{-4}	1.02×10^{-5}	6.72×10^{-5}
UR	4.0×10^{-7}	-	-	-	-
ECR	2.24×10^{-8}	-	-	-	-

MC - Maximum Concentration of PAEs in µg/L determined in bottled drinking water samples; WC - Water Consumption; EDI: Daily intake via drinking water (µg/kg body weight/day); IRIS RfD – Reference dose (µg/kg bw/day); HQ - Hazard quotient; UR: US EPA drinking water unit risk of carcinogenicity (per µg/L); ECR - Excess cancer risks

compound in the bottled drinking water brands in Delta State, which is concurrent with the findings of Jeddi et al. [33], Li et al. [38] and Xu et al. [4]. But to this, conclusion should be noted that the PAEs evaluated in this study represented only a small proportion of the total PAEs in drinking water.

4. CONCLUSION

In this study, we measured the concentrations of PAEs in ten bottled drinking water brands across eight (8) local government areas in Delta State. Seven PAEs were investigated (six USEPA-controlled and 1 uncontrolled). DPB in BW 1 recorded the highest value while DEHP in BW 7 recorded the least value. Moreover, BW 1 had the highest total concentration of PAEs in the different brands while BW 9 had the lowest. In addition, the predominant PAEs across the ten brands were DMP and BEP. DMP was detected in all the brands while BEP was detected in seven. However, DBP had the highest total concentration making it the main contributor to PAE levels in the bottled drinking water brands. Moreover, health risk assessment was carried out and the results indicated little non-carcinogenic and carcinogenic health risks. However, continuous monitoring is recommended to ensure than PAE levels in bottled drinking water remain within permissible limits.

Our results therefore provides important information for the understanding of the distribution and potential health risks of PAEs in different bottled drinking water brands in Delta State.

COMPETING INTERESTS

Authors have declared that no competing interests exist.

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