



Concentrations of Phthalates and Metals in Commercially Packaged Sachet and Plastic Bottled Water Sold in Lagos, Nigeria

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HIGHLIGHTS

- Mean concentrations of dimethylphthalate, diethylphthalate, and dibutylphthalate in bottled waters were 0.564±0.074, 0.248±0.166, and 0.042±0.049 mg/L, respectively.
- Mean concentrations of dimethylphthalate, diethylphthalate, and dibutylphthalate in sachet waters were 0.803±0.049, 0.243±0.035, and 0.160±0.073 mg/L, respectively.
- Drinking sachet waters may have higher risk of phthalates exposure compared with the bottled water.

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Acronyms and abbreviations

DMP=Dimethylphthalate
DEP=Diethylphthalate
DBP=Dibutylphthalate
Zn=Zinc
Cr=Chromium
Pb=Lead
Cd=Cadmium
HPLC=High Performance Liquid
Chromatography

ABSTRACT

Background: In many developing countries, numerous brands of bottled water and the relatively cheaper counterpart, sachet water, can be found in all cities, towns, and even villages. This study assessed the concentrations of some phthalates and metals in bottled and sachet water sold in Lagos, Nigeria.

Methods: Fifteen pieces of plastic bottled water and 15 pieces of sachet water were randomly obtained from different street vendors in Lagos, Nigeria. High Performance Liquid Chromatography (HPLC) was used to determine the levels of dimethylphthalate (DMP), diethylphthalate (DEP), and dibutylphthalate (DBP). Also, atomic absorption spectroscopy assay was applied in order to assess the contents of metals, including zinc (Zn), chromium (Cr), lead (Pb), and cadmium (Cd). All statistical analyses were carried out using the SPSS (version 20).

Results: The mean concentrations of DMP, DEP, and DBP in bottled water samples were 0.564±0.074, 0.248±0.166, and 0.042±0.049 mg/L, respectively; these rates for sachet water samples were 0.803±0.049, 0.243±0.035, and 0.160±0.073 mg/L, respectively. Some significant differences ($p<0.01$) were found between phthalates concentrations of various water brands. The mean DMP concentration of sachet water samples was significantly higher ($p<0.01$) than that of bottled waters. The concentrations of Zn, Cr, Pb, and Cd in the samples were within the acceptable limits.

Conclusion: The higher concentrations of phthalates in sachet water relative to bottled water indicate that drinking sachet water may pose higher risk of phthalates exposure.

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Introduction

Water is a major necessity of life, not only for man, but for every living system. It is in realization of this that the World Health Organization (WHO) has made “access to

adequate supply of safe drinking water to all people” as one of the primary goals (WHO, 2003a). In many countries, the primary responsibility of providing safe drink-

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ing water to the people rests mainly on the country's public water corporation. Access to safe potable water has globally remained inadequate especially in developing countries (Kumar and Puri, 2012; Oghenekohwiroro et al., 2016). This gap in demand and supply of safe drinking water has encouraged, in no small way, the present day increasing production of commercial packaged drinking water (Al-Saleh et al., 2011). Packaged water usually comes in two forms, depending on the packaging materials; (1) bottled water, and (2) sachet water. Bottled water is usually packaged in plastic bottles, while sachet water is typically packaged in plastic sleeves. In addition to the short supply of drinking water, the production of high volumes of packaged water is a safety concern. Many people are of the opinion that packaged water offers better bacteriological and physicochemical qualities, in comparison with corporation-supplied tap water (Al-Saleh et al., 2011; Doria, 2006); however, researches have proved that this may not be necessarily so.

Several investigations have evaluated the microbial, chemical, and physical properties of bottled and sachet water and found them wholesome and fit for drinking (Danso-Boateng and Frimpong, 2013; Onweluzo and Akuagbazie, 2010), but some found the microbiological quality unacceptable (Fisher et al., 2015). It is known that some metals induce toxic effects in mammals and they could be serious concerns threatening the safety of water supply. Also, there is a growing global concern that the chemical components of the packaging materials, especially phthalate esters, may leach out of the matrix to contaminate the water inside the container. Some phthalates that have been found at different concentrations in bottled and sachet water are dimethylphthalate (DMP), diethylphthalate (DEP), dibutylphthalate (DBP), di-n-hexyl-phthalate, as well as butyl benzyl phthalate (Kanchanamayoon et al., 2012; Keresztes et al., 2013; Oghenekohwiroro et al., 2016; Prapatpong and Kanchanamayoon, 2010;). Meanwhile, the United States Environmental Protection Agency (US EPA) has set the threshold limit values of 0.55, 0.45, and 5.0 mg/L for DEP, DBP, and DMP, respectively (US EPA, 1991). Excessive exposure to phthalates may result in some disorders such as cancer, endocrine system disruption, developmental abnormalities, and polynuropathy (Lee and Koo, 2007; WHO, 2003b).

Though relevant data on packaged water in Nigeria are not readily available, the production of packaged water in the country dates back to the 1990s and attained a rapid growth in the mid-2000s (Uduma and Uduma, 2014). Today, in many developing countries, including Nigeria, numerous brands of bottled water and the relatively cheaper counterpart, sachet water, can be found in all cities, towns, and even villages. This study assessed the concentrations of DMP, DEP, and DBP, along with some

metals, including zinc (Zn), chromium (Cr), lead (Pb), and cadmium (Cd) in bottled and sachet water sold in Lagos, Nigeria.

Materials and methods

Collection of packaged water samples

On September 2017, a total of 30 pieces of packaged water, made up of 15 pieces of plastic bottled water and 15 pieces of sachet water, were randomly bought from different street vendors in Lagos, Nigeria, taken to the laboratory, and kept in refrigerator overnight at 4 °C. For each brand of bottled water, its counterpart sachet water produced by the same company was bought. To ensure a wide sampling distribution, the five brands of packaged water were produced by five companies located in five different areas (Ikorodu, Yaba, Lagos Island, Ikeja, and Ebute-metta) of Lagos, Nigeria. To protect the identities of the brands, the bottled water samples were coded LAM, LAO, RDM, KOL, and VEL; while the sachet water samples were coded LAMS, LAOS, RDMS, KOLS, and VELS.

Analysis of phthalates in water samples

The phthalates content in the samples were determined by High Performance Liquid Chromatography (HPLC) apparatus (Shimadzu Nexera mx, Japan) as described in Dada and Ikeh (2018) with modifications. Care was taken to minimize sample contamination and maintain low background phthalate concentration. All the laboratory glassware or materials used in sample handling and preparation were either made of Teflon or stainless steel. The materials were prewashed with methanol and calcinated overnight at 500 °C.

-Extraction

Extraction was first carried out by measuring 20 ml of water sample into separation funnel and adding 20 ml of acetonitrile/methanol (3:1) into it. The mixture was shaken vigorously for 30 min, while releasing the funnel cap intermittently to release pressure build-up. The aqueous end was run off while the hydrocarbon (as non-polar) end was obtained in a 25 ml flask and made up to the mark with acetonitrile/methanol solution which used for phthalate analysis.

-Calculation

At first, a standard form of individual pure phthalate of known concentration was injected into the HPLC apparatus, providing a chromatogram with a given peak area as well as peak profile. These were used to create a

window in the HPLC in preparation for the test sample analysis. An aliquot of the extracted test sample was also injected into the HPLC to obtain a corresponding peak area and peak profile in a chromatogram. To obtain the concentration of the sample, the peak area of the sample was compared with that of the standard relative to the standard concentration. The HPLC was set to accommodate the dilutions made during extraction. The equation indicated below was used to calculate the concentration of the sample.

$$\text{Concentration of sample} = \frac{\text{Peak area of sample} \times \text{Concentration of standard}}{\text{Peak area of standard}}$$

-Programming of HPLC

The programming of HPLC was as follows: Column (stationary) phase: uBondapak C18; Length=100 mm; Internal Diameter (ID)=4.6 mm; Thickness=7 µm; Mobile phase: Acetonitrile/water (3:1); Sample injection volume: 1 ml; Flow rate: 0.10 ml/min; Detector: UV e 254 nm; Temperature: room temperature (28-30 °C).

Quantification of metals in water samples

The concentrations of metals in the water samples were determined as indicated previously by Dada and Ikeh (2018). First, 20 ml of water sample was measured into a conical flask. Dilute HNO₃ (HNO₃:deionized water=1:3) was added to the sample in a conical flask; then it was heated in a Bunsen burner until all the reddish yellow fumes were expelled. The solution was brought down, allowed to cool, and was transferred into a 10 ml standard flask. The standard flask was made up to mark with distilled water. Then, analysis of metal content was done using atomic absorption spectrometer (Perkin Elmer model 460, USA).

A suitable standard curve was generated in the atomic absorption spectrometer using standard solutions of metals of interest. Standard concentrations of 6, 12, 18, 24, and 30 mg/L were used. Adequate standard was generated for each metal passing through the origin. After a good curve was achieved, the digested sample solutions were aspirated into the apparatus for the analysis. For each metal, a unique cathode lamp was used for the analysis, operating at its peculiar maximum operational wavelength. A digital concentration read-out (meter) gave the direct concentration of the metals as contained in the test sample.

Statistical analysis

Multivariate Analysis of Variance (MANOVA) was used to determine the effect of packaged water type on the concentrations of phthalates and metals in water samples. The data generated from bottled water and sachet water were then separately processed by MANOVA to

determine if there were differences in the concentrations of phthalates, and of metals in different packaged water brands. The mean differences were compared for statistical significance using the Least Significant Difference (LSD) post hoc test. The mean concentrations of each phthalate in bottled water and sachet water; and of each metal in bottled water and sachet water were separately compared for significant difference by T-test analysis. All statistical analyses were carried out using the SPSS software (version 20).

Results

According to Table 1, the mean concentrations of DMP, DEP, and DBP in bottled water samples were 0.564±0.074, 0.248±0.166, and 0.042±0.049 mg/L, respectively; these rates for sachet water samples were 0.803±0.049, 0.243±0.035, and 0.160±0.073 mg/L, respectively. Some significant differences ($p<0.01$) were found between phthalates concentrations of various water brands. Also, the T-test analysis revealed that the mean DMP concentration of sachet water samples was significantly higher ($p<0.01$) than that of bottled waters.

The concentrations of Zn, Cr, Pb, and Cd in bottled water and sachet water sampled in this study were generally low and within the limits set by the WHO (Table 2). The highest concentrations recorded for Zn, Cr, Pb, and Cd were 0.079±0.105, 0.003±0.002, 0.100±0.124, and 0.001±0.002 mg/L respectively. The differences in the mean total concentrations of Zn, Cr, Pb, and Cd in bottled water and sachet water were not significant.

Discussion

A lot of concern has been raised about the sustainability of packaged water production and consumption, especially in some developing countries, in view of its inadequate regulation, monitoring, safety rule compliance, as well as its potential environmental and health impact. Packaged water has been linked to possible outbreak of water-borne diseases like cholera, found questionable in microbiological qualities (Fisher et al., 2015; Venkatesan et al., 2014), and has already been established as a major contributor to solid waste pollution in some West African countries including Nigeria (Mojekeh and Eze, 2011). However, the association of drinking packaged water with increased phthalate load as found in our study and also previous researches (Kanchanamayoon et al., 2012; Keresztes et al., 2013; Oghenekohwiroro et al., 2016) present another dimension to the worry about packaged water. In a study conducted in another state of Nigeria, phthalates occurred in concentrations of 0.03-0.09 µg/L for DMP, 0.06-0.20 µg/L for DEP, and 0.00-0.03 µg/L

Table 1: Phthalate concentrations in different brands of bottled and sachet water sold in Lagos, Nigeria

Brand code	Type	Concentrations of phthalates (mg/L)		
		DMP	DEP	DBP
LAM	Bottled	0.635±0.054 ^a	0.174±0.007 ^a	0.021±0.002
	Sachet	0.857±0.007 ^d	0.283±0.009 ^d	0.210±0.006 ^d
LAO	Bottled	0.621±0.023 ^a	0.175±0.006 ^a	0.040±0.052
	Sachet	0.761±0.002	0.209±0.020	0.153±0.006 ^d
RDM	Bottled	0.591±0.011 ^a	0.122±0.021	0.021±0.001
	Sachet	0.746±0.014	0.203±0.008	0.033±0.006
KOL	Bottled	0.519±0.027 ^a	0.563±0.005 ^a	0.003±0.001
	Sachet	0.852±0.020 ^d	0.272±0.006 ^d	0.237±0.011 ^d
VEL	Bottled	0.457±0.015	0.203±0.006 ^a	0.125±0.022 ^a
	Sachet	0.801±0.018 ^d	0.250±0.003 ^d	0.168±0.009
Mean concentration	Bottled	0.564±0.074	0.248±0.166	0.042±0.049
	Sachet	0.803±0.049 [*]	0.243±0.035	0.160±0.073
T-test	Bottled	t=10.472, p<0.01	t=0.093, p>0.05	t=-5.215, p>0.05
	Sachet			

DMP=dimethylphthalate; DEP=diethylphthalate; DBP=dibutylphthalate

*=significantly high (p<0.01) relative to the total mean concentration of bottled water

a=significantly high (p<0.01) relative to the concentration of the same phthalate type in one or more bottled water brands

d=significantly high (p<0.01) relative to the concentration of the same phthalate type in one or more sachet water brands

Table 2: Metal concentrations in different brands of bottled and sachet water sold in Lagos, Nigeria

Brands code	Type	Concentration of metals (mg/L)			
		Zinc	Chromium	Lead	Cadmium
LAM	Bottled	0.014±.001	0.002±0.001 ^b	0.004±0.005	0.001±0.001
	Sachet	0.079±0.105	0.002±0.001 ^c	0.001±0.001	0.001±0.001
LAO	Bottled	0.014±0.001	0.001±0.001	0.002±0.001	0.001±0.001
	Sachet	0.019±0.002	0.001±0.001	0.004±0.005	0.001±0.002
RDM	Bottled	0.044±0.057	0.001±0.000	0.004±0.005	ND
	Sachet	0.014±0.003	ND	0.004±0.002	ND
KOL	Bottled	0.010±0.001	ND	0.100±0.124	ND
	Sachet	0.019±0.002	0.003±0.002 ^c	0.002±0.001	0.001±0.001
VEL	Bottled	0.012±0.001	ND	0.004±0.005	ND
	Sachet	0.014±0.004	0.001±0.001	0.021±0.002 ^c	0.001±0.001
Mean concentration	Bottled	0.019±0.025	0.001±0.001	0.022±0.060	<0.001
	Sachet	0.029±0.047	0.001±0.001	0.007±0.008	<0.001
T-test	Bottled	t=-0.702, p>0.05	t=-0.632, p>0.05	t=0.984, p>0.05	t=-1.426, p>0.05
	Sachet				
WHO limit		Not given	0.050	0.010	0.003

ND=Not detected

b=significantly high (p<0.01) relative to the concentration of the same metal type in one or more other bottled water brands

c=significantly high (p<0.01) relative to the concentration of the same metal type in one or more other sachet water brands

for DBP in samples of sachet water (Oghenekohwiroro et al., 2016). Similarly, a survey carried out on samples of bottled water in Thailand showed presence of DMP, DEP, and DBP in concentrations of 0.16-0.53, 0.11-0.54, and 0.17-0.33 mg/L, respectively. Relative to the works of Oghenekohwiroro et al. (2016) and Kanchanamayoon et al. (2012), the concentrations of DMP, DEP, and DBP in the sampled packaged water in this study were higher.

In addition, the presence of all the three investigated phthalates (DMP, DEP, and DBP) in all the water samples assessed in the present work might be an indication that other phthalates not assessed were also possibly present.

Apart from the general sources of phthalate contamination which both surface and groundwater water are usually exposed to, packaged water (bottled and sachet) are

additionally exposed to phthalate entry from many other potential sources in the course of their production. For instance, packaged water could be exposed to phthalate contamination from treatment facilities such as pipes, storage tanks, as well as filtering systems (Leivadara et al., 2008). Hence, consumers of packaged water are potentially exposed to increased levels of phthalate intake. The occurrence of DMP in higher concentrations compared to either DEP or DBP in this study may be partly attributed to the extra water hydrophilic and solubility properties associated with DMP relative to other phthalate esters (Staples, 2003). The results showed that the type of packaged water and the brand of packaged water both have significant influence on the concentration of phthalates imply that consumer's choice of brand, and type of packaged water (whether bottled or sachet) will have significant influence on their level of exposure to phthalate.

The higher concentrations of phthalates in sachet water compared to bottled water in this study may be attributed to the quality of packaging material. Studies have shown that water packaged in plastic sachet is potentially more prone to contamination by phthalate molecules that are leached from the polymer matrix (Keresztes et al., 2013; Oghenekohwiroro et al., 2016). Another probable reason that may be adduced for the higher phthalate levels in sachet water in this study is the volume of packaging container. In Nigeria, sachet water is usually packaged in sealed 500 ml plastic sleeves as opposed to the bottled counterpart that may come in volumes of 75 ml, 100 ml, or higher; but more commonly in 75 ml. Since 500 ml sachet and 75 ml bottled water samples were used for this study, the factor of higher surface/volume ratio might have induced increased phthalate leaching and migration from the 500 ml polymer matrix to the water content.

Due to the relative low cost of sachet water, and the low disposable income of a majority of the masses, sachet water enjoys far higher patronage than bottled water in Nigeria and many developing countries. This implies that more people are at higher risks of phthalate exposure through sachet water consumption. This situation calls for concern and intervention by government and appropriate authorities since epidemiological studies have shown that phthalates have the ability of inducing adverse health effects like male reproductive system disorders, breast and testicular cancers, as well as neuro-endocrine system disruption among others (Matsumoto et al., 2008).

While there is a need to address the occurrence of increasing levels of phthalate and other contaminants in packaged and other forms of drinking water supply, the case of sachet water is a complex and special problem that requires immediate and holistic intervention in developing countries, especially in West Africa. Sachet

water industry has continued to provide near-indispensable roles in the social and economic activities of many West African countries where it provides many jobs and improves access to clean drinking water. However, when these seemingly positive roles are juxtaposed with its adverse health and environmental effects, its sustainability becomes doubtful. Studies have identified sachet water as a potential medium of cholera and other microbial disease (Fisher et al., 2015; Venkatesan et al., 2014). Sachet water has constituted a major environmental menace in many developing West African countries where street roads, gutters, and storm drains get clogged from mainly the plastic sleeves of discarded sachets (Oghenekohwiroro et al., 2016). When these undesirable drawbacks of sachet water are considered along with its likelihood of exposing consumers to increased levels of phthalates as found in this and previous studies, the need to undertake a comprehensive review of sachet water industry will be obvious by government and relevant stakeholders.

The concentrations of trace/toxic metals assayed in this study were lower or within the limits set by WHO (WHO, 2003a,c; 2011a,b). However, care should be taken when assessing the effects or toxicity of metals, phthalates, and other contaminants. Toxicity evaluation based on each chemical contaminant separately may underestimate the potential risk of contaminants, as individuals are often exposed to more than one chemical contaminant type at the same time and from many sources (Dada and Ikeh, 2018; Wittassek and Angerer, 2008).

Conclusion

This study evaluated the concentrations of some phthalates and metals in selected bottled and sachet water samples. The concentrations of metals were within the regulatory limits set by WHO; however the water samples contained different concentrations of phthalates. The higher concentrations of phthalates in sachet water relative to bottled water indicate that drinking sachet water may pose higher risk of phthalates exposure. Nigerian government should map out holistic policies and embark on programmes that would discourage the production and consumption of sachet water.

Author contributions

E.O.D. designed the study and wrote the manuscript; V.A.O., K.E.I., and S.O.I. conducted the experimental work; M.O.A. analyzed the data and read the manuscript for correctness. All authors revised and approved the final draft of the manuscript.

Conflicts of interest

There are no conflicts of interest.

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