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Dibutyl Phthalate an Emerging Contaminant in Water-Based Paints

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Abstract. Seven different manufacturers of paints were purchased from two major cities in southwest Nigeria. At least one sample from the same manufacturer and colour, was identified and analyzed for dibutyl phthalate (DBP). GC/MS method was developed for the identification and quantification of dibutyl phthalate in water-based paints after solvent extraction. The highest level of DBP found was 47,100 ppb (unregistered manufacturer), followed by 45,100 and 19,400 in a white, pink and cream coloured paint, respectively, while the lowest concentration was 721 ppb in a cream coloured paints, unregistered manufacturer. The PCA results were in good accordance with the findings of the correlation coefficient. DBP is used by all manufacturers for flexibility and better spreadability. It is a cheap alternative and readily available. It's an emerging contaminant newly reported in paints and its toxicological effects should not be underestimated, since there is no regulation in place.

Keywords: emerging contaminant, dibutyl phthalate, environmental toxicology, gas chromatography-mass spectrometry, multivariate statistical analysis

Introduction

Many articles have reported the presence of emerging compounds in consumer products such as phthalates, in the human and aquatic environments and wastewaters. Emerging pollutants are new chemicals without regulatory status and which impact on environment and human health are poorly understood (Lioy *et al*., 2015; USEPA, 2012).

Phthalates are produced in excess of million pounds per year (USEPA, 2012; EPA, 2006). Phthalates are used in several industrial and consumer products, basically as plasticizers (on Earth and NASEM, 2017; Lioy *et al*., 2015). They are released into the environment from different sources such as industrial releases, the disposal of productions, processing and industrial wastes, municipal wastes, sewage sludge and release from commodities containing phthalates Braun *et al*. (2013). The exact phthalate or combination of phthalates utilized in a specific product's formulation relies upon the properties the phthalates impart as well as their cost. Many phthalates can doubtlessly result in excessive exposure, both exclusively and together with other phthalates (Wang *et al*., 2019; Lioy *et al*., 2015; USEPA, 2012; Swan, 2008). They can frequently substitute for each other in products. U.S. Environmental Protection Agency's (EPA's) cutting-edge management plan considered the toxicity of phthalates, their occurrence in the environment and their considerable use and human exposure on Earth and (NASEM, 2017; CDC, 2005). Eight phthalates were considered: dibutyl phthalate (DBP), di-isobutyl phthalate (DIBP), butyl benzyl phthalate (BBP), di-*n*-pentyl phthalate (DnPP), di (2-ethylhexyl) phthalate (DEHP), di-*n*-octyl phthalate (DnOP), di-isononyl phthalate (DINP), and di-isodecyl phthalate (DIDP), USEPA (2012). These phthalates shows adverse effects to aquatic organisms in ecotoxicity studies Oehlmann *et al*. (2009). Toxic effects were observed at environmentally relevant exposures at the low concentration that is, ng/L to µg/L range Oehlmann *et al*. (2009). Phthalates exposures can occur from releases that result from the production, processing or industrial use as well as contamination to air, water, food and dust resulting from the use or disposal of commodities containing phthalates (Wang *et al*., 2019; Lioy *et al*., 2015; Russo *et al*., 2015; Shaikh *et al*., 2012). Exposure sources of phthalates in the order of prevalence is food, cosmetics, consumer products (other than toys) and toys (Zota *et al*., 2016; Serrano *et al*., 2014; Hubinger, 2010; HSDB, 2009; Koo and Lee, 2004).

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Dibutyl phthalate (DBP) is one of the main sources of indoor semi-volatile organic compounds (SVOCs) and topmost persistent organic contaminants Wang *et al*. (2010). It is used as softener for better spreadability, flexibility, to retain scents and dispensability (Larsson *et al*., 2014; Chang *et al*., 2013; Green *et al*., 2005) as a solvent and fixative; a suspension agent; a lubricant for aerosol valves; an anti-foamer; a skin emollient, a plasticiser, fingernail elongators (extensions), in medical devices and textiles, as propellants, in food packaging, as dental materials, and in paper production (Wittassek *et al*., 2011; OSHA, 2009). However, in few illegal food industry applications, phthalates have been used as a low-fee alternative to update palm oil as a clouding agent added to juices, yogurt and drinks Chang *et al*. (2013). Dibutyl phthalate is one of the most popular phthalates used as clouding agent in drinks, which has been detected by the Taiwan Food and Drug Administration in 2011 (USEPA, 2012; HSDB, 2009).

Phthalates can be released from products and exposure may occur in humans through food, dust, air and direct use of personal care products (Janjua *et al*., 2008; Wittassek and Angerer, 2008; Wormuth *et al*., 2006). DBP is very stable and can enter to all primary environmental media. It causes the most lethally to terrestrial organisms, fish, and aquatic invertebrates Staples *et al*. (1997). Besides its role as endocrine disruptor, DBP has been shown to alter the expression of a number of genes, increased DNA damage in sperm, premature breast development, shortened pregnancy in women, and decrease in anogenital distance among male infants (Zare *et al*., 2018; Monneret, 2017; Swan *et al*., 2005; Duty *et al*., 2003; Colón *et al*. 2000). It exposure is of potential concern for children's health Craig *et al*. (2013). The National Toxicology Program concluded that high levels of di-*n*-butyl phthalate, may adversely affect human reproduction or development (Monneret, 2017; Sedha *et al*., 2015; Lin *et al*., 2011; Jahnke *et al*., 2005; NTP, 2003). They have also been measured in foods, milk and drinking water. The relative contribution from the various sources and routes of exposure to phthalates is unknown (Schecter *et al*., 2013; Fierens *et al*., 2012; Cao, 2010; Kolarik *et al*., 2008; Wormuth *et al*., 2006). Despite the enormous health effects, there remain no regulations on DBP in water-based paints manufactured and marketed in Nigeria. The objective of this work was to identify and access the levels of dibutyl phthalate, an emerging contaminants in water-based paints and their associated health effects.

Materials and Methods

Paint sampling. Paint samples of different colours and 14 manufacturers were purchased in popular paint markets in Ibadan and Lagos, Nigeria. A total of 174 paint samples were collected. These samples were stored in air-tight plastic containers and analysed at the Council of Scientific and Industrial Research-National Environmental Engineering Research Institute Laboratory, Nagpur – Maharashtra, India (Idayat Apanpa-Qasim and Adeyi, 2018).

Samples pre-treatment and analysis. Approximately 2.5 mL paint samples were carefully measured into 50 mL polypropylene radiation sterilized centrifuge tubes and 20 mL of ethyl acetate was then added ((Idayat Apanpa-Qasim and Adeyi, 2018). The centrifuge tubes were shaken and mixed on a cyclo mixer at 50 cycles (CM 101) for homogeneity of the samples. The tubes were centrifuged at 5000 rpm at 20 °C for 20 min. The supernatant was filtered with PTFE micro-fibre syringe filter of 13 mm diameter and 0.22 micron pore size before analysis. The extracts were stored in 2 mL maxipense plastic vials analyzed using GC/MS. The internal standard (benzlyl benzoate) was added to all samples, blanks, and calibrators at known concentration.

Table 1. Instrument operating conditions

Operating condition	GC-MS	GC-MS Monitoring ions (m/z) used for the identification
Analytical column	DB 5 (30m× 0.25mm × 0. 25µm)	149, 150, 41
Carrier gas	Helium	
Gas flow rate	1.2mL/ min	
Injection volume	1µL	
Injection temperature	200ºC	
Detector temperature	280ºC	
Column temperature	180ºC (0.5 min) -20ºC/min-280ºC 7 mins)	
Inlet source temperature	300ºC	
Injection mode	split	
Solvent delay	4.2 mins	
Detector gain mode	relative	

Instrument operating conditions is presented in Table 1. Recovery study was carried out and DBP recovery was of 86%.

Statistical analysis. All the analyses were done in duplicate and the results were expressed in minimum and maximum values using MS-Excel. The data were also analyzed by using principal component analysis (PCA), cluster analysis (CA) and correlation coefficient using SPSS (Kumar *et al*., 2016; Trindade *et al*., 2015; Li *et al*., 2009).

Results and Discussion

Only 27 out of the 174 paint samples had DBP identified. The chromatogram and mass spectra of DBP are given in Fig. 1 and 2. This was confirmed based on the retention times and the molecular formula.

Concentrations of dibutyl phthalate in the different colours of paint samples by manufacturers based on ISO certification. Seven paint manufacturers with at least one sample each of the same colour were involved in this study. Two of the manufacturers were registered with Nigerian Industrial Standard (NIS) and International Organization for Standardization (ISO) and five unregistered manufacturers without NIS-ISO certification, producing different colours of water-based paints. The concentrations of the dibutyl phthalate in the paint samples with respect to manufacturers are presented in Table 2. Variation in dibutyl phthalate concentrations of paint samples with respect to colours produced by manufacturers are shown in Fig. 3.

The range of levels of dibutyl phthalate in all the paint samples with respect to colour was 721 (cream)–47,100 ppb (white). The highest concentrations of dibutyl phthalate in all the 27 paint samples was 47,100 ppb in a white coloured paint by manufacturer A, a registered manufacturer. This was followed by 45,100 ppb in a pink coloured paint and 19,400 ppb in a cream coloured paint by the same manufacturer, while the lowest concentration was 721 ppb in a cream coloured paints produced by manufacturer D, an unregistered manufacturer. The order of dibutyl phthalate with respect to paint colours where present is:

DBP: pink > white > orange > green > blue > cream > yellow > chocolate.

Correlation coefficient. The data obtained in this study was subjected to Pearson correlation coefficient and it was found out that the registration status (registered and unregistered status of the manufacturers) had a

Table 2. Concentrations (ppb) of dibutyl phthalate in paint samples with respect to manufacturers

Colour grouping	Manufacturer codes	NIS-ISO registration	Primary colours in the paint samples	Sample 1	Sample 2
NG 01	A	RG	blue	4900	5120
NG 03	A	RG	yellow	4880	-
NG 04	A	RG	white	47100	-
NG 05	A	RG	pink	45100	-
NG 06	A	RG	cream	19400	-
NG 07	A	RG	green	17600	-
NG 08	B	RG	white	3400	-
NG 09	B	RG	blue	8910	-
NG 10	C	RG	cream	2140	-
NG 11	D	WRG	cream	721	-
NG 12	E	WRG	green	5660	-
NG 14	E	WRG	orange	8190	7340
NG 15	E	WRG	cream	4080	-
NG 17	E	WRG	green	6120	5850
NG 18	E	WRG	blue	8860	-
NG 19	E	WRG	cream	3920	-
NG 20	E	WRG	blue	8050	-
NG 22	E	WRG	white	15500	15100
NG 23	F	WRG	chocolate	1470	-
NG 24	F	WRG	blue	3220	-
NG 25	F	WRG	green	1550	-
NG 27	G	WRG	cream	2610	4190

where RG means registered, WRG means without registration.

positive correlation with manufacturers and dibutyl phthalate versus colour (Table 3). This signifies that the toxicity of dibutyl phthalate should not be underestimated in its usage by manufacturers of paints of different colours.

Principal components analysis (PCA). Two principal components were extracted using the PCA with rotated component matrixes. Corresponding components, variable loadings, and the variances are presented in Table 4.

Only PCs with eigenvalues greater than 1 were considered. PCA of the whole data set yielded 2 data set explaining 86.25 % of the total variance. The first component was responsible for 60.40 % variance and

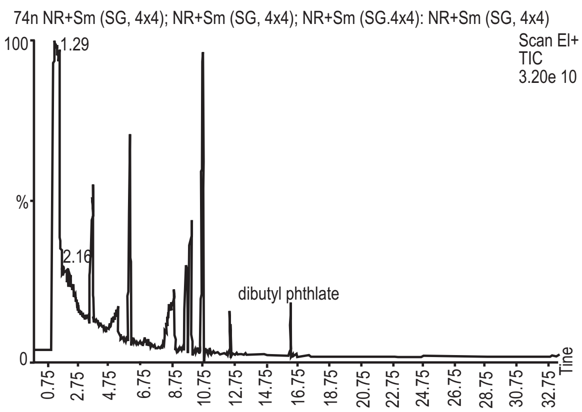

Fig. 1. Total ion chromatogram of dibutyl phthalate using GC-MS analysis.

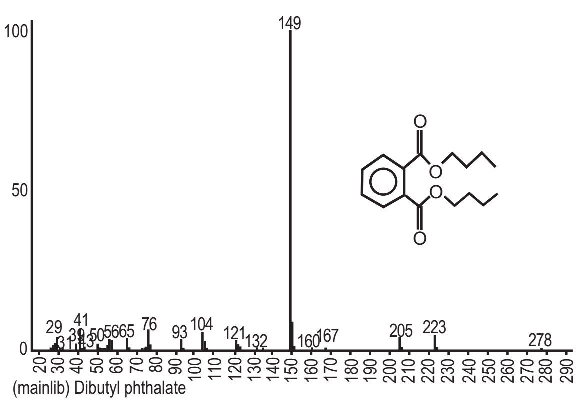

Fig. 2. Mass spectrums of dibutyl phthalate obtained after GC-MS analysis.

Table 3. Correlation coefficient of dibutyl phthalate in the paint samples

	Manu-facturers	Dibutyl phthalate	Colours	Registration status
Manufacturers	1	-0.492**	-0.186	0.923**
Dibutyl phthalate		1	0.429*	-0.479*
colours			1	-0.161
Registration status				1

Note: ** = significant at 0.01, * = significant at 0.05.

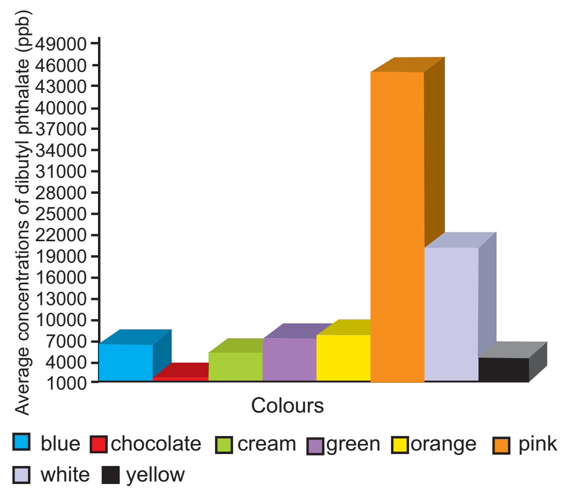

Fig. 3. Variation in concentrations of dibutyl phthalate in paints with respect to colours.

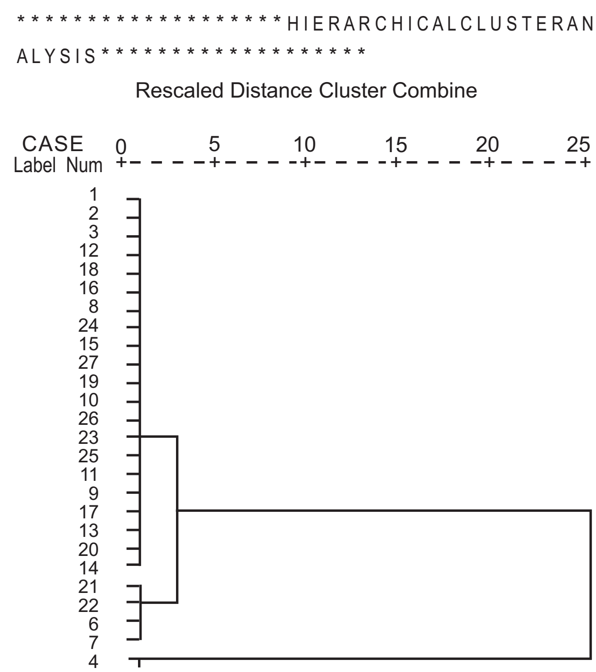

Fig. 4. Dendrogram showing the clusters of levels of dibutyl phthalates in paint samples.

was best represented by the registration status of manufacturers. The second principal component includes all the manufacturers which accounts for 25.85 % of the total variance.

The initial component matrix for dibutyl phthalates indicates that registration status and manufacturers are associated, showing high values in the first principal component (PC1) which explains 52.3% of the total variance and loads heavily on registration status (0.964), manufacturers (0.959). Registration status and manufacturers values are controlled by the producers of paints either registered companies or not. The second principal component (PC2) includes univocally colours and dibutyl phthalates, which accounts for 33.9% of the total variance. Common sources of colours in paints are majorly from pigments, and dibutyl phthalates influences the flexibility of paint produced. The analyzed results are in good accordance with the findings of the correlation coefficient.

Cluster analysis (CA). Cluster analysis (CA) was performed on the data using between-groups linkage method and squared Euclidean distance using the hierarchical clustering with SPSS software (Trindade *et al*., 2015; Li *et al*., 2009). Figure 4 shows the CA of dibutyl phthalate in the paint samples as a dendrogram. Two major clusters were obtained. Cluster 1: identified cases 1-3, 8-20, 23-27 and Cluster 2: cases 4 and 5. Cluster analysis revealed a division of the studied parameters into their similar class with respect to their normalized concentration levels.

Conclusion

This study predicts the possible human exposure to dibutyl phthalates in water-based paints, the levels were determined by GC/MS in 27 water-based paint samples. The low levels of DBP in water-based paints coincides with its physico-chemical characteristics (high volatility) that make DBP inappropriate for use in non-plastic products such as paints and probably as plasticizers in PVC found in packing materials. Chemicals such as DBP are used in a wide variety of consumer products (USEPA, 2012) they are suspected endocrine disrupters but their toxicity are poorly understood in paints. Combined exposure may occur through ingestion, inhalation and dermal exposure and their toxic as well as combined effects are poorly understood Larsson *et al*. (2014). Also, results from multivariate statistical analysis clearly show the use of DBP by all manufacturers for its flexibility and better spreadability in paint production in Nigeria despite its health effects. Hence, a need for stringent regulations to safe-guard public health from occupational exposure to these toxic and prohibited compounds is imperative.

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Table 4: Total variance and component matrixes extracted by PCA

Total variance explained

	Initial eigen values			Extraction sums of squared loadings			Rotation sums of squared loadings		
Component	Total	% of Variance	Cumulative %	Total	% of Variance	Cumulative %	Total	% of variance	cumulative %
Registration status	2.416	60.40	60.40	2.416	60.40	60.40	2.094	52.342	52.342
manufacturers	1.034	25.85	86.25	1.034	25.85	86.25	1.356	33.911	86.254
colours	0.473	11.83	98.09						
Dibutyl phthalates	0.077	1.914	100.00						

Extraction method: principal component analysis

(b) Component matrixes

Component matrix	Rotated component matrix		Compound	
	PC1	PC2	PC1	PC2
Registration status	**0.901**	0.362	**0.964**	-0.118
manufacturers	**0.910**	0.336	**0.959**	-0.145
colours	-0.451	**0.818**	-0.00005	**0.934**
Dibutyl phthalates	**-0.758**	0.347	-0.496	**0.670**

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Conflict of Interest. The authors declare no conflict of interest.

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