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Bioaccumulation and health risk of trace elements in shellfish and their leachates from two coastal areas of Nigeria

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Abstract

Trace elements (Cd, Cu, Fe, Ni, Pb and Zn) were determined in tissues of two periwinkle species (Pachymelania fusca and Tympanotonus fuscatus) harvested in Ibeno and Ishiet coastal areas in Akwa Ibom State, Nigeria; their leachates, water, and sediments using atomic absorption spectrophotometry. The trace elements in periwinkle, except for Cd and Ni (in both species), Zn and Pb (in Tympanotonus fuscatus) were below FAO/WHO dietary allowances. Pachymelania fusca contained high levels Ni, Cd, Cu, and Fe. Leachates from both species contained varying amount of elements, and Cd exceeded FAO/WHO dietary allowance. Par-boiling thus reduced the health risk through consumption of contaminated periwinkles. Cd exceeded the recommended level for healthy fish cultivation in water and sediment. The Pachymelania fusca specie showed higher bioaccumulation than Tympanotonus fuscatus. The non-carcinogenic risk assessment revealed Cd intake as a major contributor to the health hazard of the adult population (HQ and THQ above one). The combined chemical exposures also indicated high health risk (HI>1). The study revealed further the presence of trace elements at toxic levels in leachates from Pachymelania fusca species (HQ>1). Consumption of periwinkles from the two locations portends health risk due to Cd.

Keywords: Accumulation; Hazard Assessment; Leachate; Periwinkles; Trace Elements

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1. Introduction

Fish and shellfish are consumed in many parts of the world, and are important source of nutrients for humans, especially for those in the coastal regions. Over the years, the pollution of the aquatic environment by toxic elements due to industralisation and urbanization has become a global issue. Trace metals are ubiquitous and being persistent pollutants can be detrimental to human and animal health (Udoh, 2000; Marcus et al., 2013).

Fish take up essential and non-essential metals from water, sediment and food, which can accumulate in the tissue and are biomagnified in the food chain (Alam et al., 2002; Subotić et al., 2013; Javed and Usmani, 2013; Alemdaroglu et al., 2003). Nickel is a carcinogenic element, and over exposure can cause skin irritation, damage to the liver and heart, as well as reduction in body weight. Lead is among the most toxic metals. It affects the brain, the cardiovascular system, thyroid gland, blood, kidney, bones and the reproductive system (Homady et al., 2002). Cadmium toxicity affects the brain, lungs, kidney, bones as well as the central nervous system (Alina et al., 2012). High exposure to Cu results in liver damage and gastrointestinal illness including abdominal pain, cramps and nausea; and high level of Zn causes acute renal failure, pancreatitis, anaemia and muscle pain (NAP, 1989).

Periwinkles constitute a significant source of protein and minerals for people of the oil-producing Niger Delta region of Nigeria (Udoh, 2000). People that suffer from medical conditions like gouty arthritis tend to avoid beef and depend more on seafood (fish, shellfish, among others). Due to incidents of aquatic contamination within the coastal areas of Nigeria, various attempts have been made to ascertain the levels and accumulation of trace micro-pollutants in periwinkles in the region (Marcus et al., 2013; Otitoloju and Don-Pedro, 2004; Ololade et al., 2011; Tongo, et al., 2017). These studies are strongly motivated by concerns regarding the possible transfer of toxic chemical contaminants from seafood to humans via dietary consumption (Türkmen et al., 2005; Chen et al., 2011; Rahman, et al., 2012).

Despite the number of studies on periwinkles in the Niger Delta region (Udoh, 2000; Marcus et al., 2013; Otitoloju and Don-Pedro, 2004; Ololade et al., 2011), risk assessment based on human exposures to the contaminants have not been given due attention. Periwinkles form part of the daily staple food in the region, and chronic exposures to contaminants through consumption may lead to adverse health effects, including illness and death.

It has been reported, and commonly held that cooking does not affect contaminant levels in consumed seafood (Liao and Ling, 2003; Wang et al., 2005; Chen-Wuing et al., 2006; Yi et al., 2011; Yuan et al., 2014). Some studies, however, have highlighted the effect of processing methods on contaminants in seafood (Lee et al., 2006; Urban et al., 2009). The periwinkle, a highly nutritive seafood consumed in the Niger Delta region of Nigeria has two methods of processing: (a) the periwinkle is trimmed, washed and initially boiled. The leachate water is then drained, the periwinkle rinsed and ready to be added to the cooking dish: either in-shell, for the tissue to be sucked out at the table; or out-shell, the tissue is needle-picked out of the shell for the meal preparation; (b) the periwinkle is trimmed, washed and added directly to the cooking dish; that is, without the initial par-boiling. Leachate water from the periwinkle which, invariably is part of the food in the second method, could harbour contaminants. The potential contribution of periwinkle leachates to health risk or otherwise, however, has not been documented.

The objective of the present study is to assess: the levels of selected trace elements (Cd, Cu, Fe, Ni, Pb and Zn) in water, sediment, and periwinkles (*Pachymelania fusca* mutans (var) and *Tympanotonus fuscatus* radula (var) collected from two important fishing areas in Akwa Ibom State; the health risk posed by consumption of contaminated periwinkles; and the effect of processing (par-boiling), if any, on contaminants in periwinkles via analysis of leachates.

2. Materials and methods

2.1. Study area

The study covered two stations namely: Ishiet Inua Akpa and Okposo-1 in Uruan and Ibeno Local Government Areas, respectively (Fig. 1). The two locations will simply be referred to as Ishiet and Ibeno, respectively. They are fishing communities in Akwa Ibom State, located in the southeast of Nigeria. The communities are rich in seafood such as fishes, shrimps, lobsters, crabs, periwinkles and oyster, among others. Ishiet has a market located close to the shores. The Ishiet beach environment is polluted as a result of the incessant disposal of waste such as human faeces, petroleum product from engine boats, dirt from the market, shops and surface run-offs. The lbeno river takes it source from Qua lboe river which links to the Atlantic ocean. The location of the Qua Iboe crude oil terminal (QIT), in addition to uncontrolled disposal of human wastes at the shores of the Ibeno beach, make contamination of the water body inevitable.

2.2. Samples collection, preparation and digestion

The periwinkle samples were collected at low tide in March 2006 from two fishing beaches – Ishiet and Ibeno in Akwa Ibom State, Nigeria (Fig. 1). Boats were used to access the two shorelines during sample collection. The *Tympanotonus fuscatus* radula (var) sampled from Ishiet are brownish in colour with spikes, sharp and pointed shells while *Pachymelania fusca* mutans (var) from Ibeno, have rather smooth shells and are dark (Fig. 2). The *Tympanotonus fuscatus* have larger shell sizes than the *Pachymelania fusca*. The periwinkles were purchased from fishermen as soon as they were harvested. They were rinsed and stored in self-sealable polyethylene bags, and transported to the laboratory in an ice-chest.

Water and sediment samples were collected at the same locations as the periwinkles. Fifteen grab samples of 200 ml each were collected 15-20 cm above the surface sediment to obtain a composite water sample of 3 L in polyethylene bottle. Three sets were sampled for each location, and each acidified with 3 ml of concentrated nitric acid and kept in an ice-chest and transported to the laboratory. Surface sediments (1-20 cm depth) were collected in three sets of composite samples at each location. Each comprised fifteen 300 g of randomly sampled sediments combined to obtain about 4.5 kg sample. The sediment samples were stored in self-sealable polyethylene bags and kept in an ice-chest until it reached the laboratory.

In the laboratory, the periwinkle samples were thoroughly washed with tap water and allowed to drain. Samples of *Tympanotonus fuscatus* and *Pachymelania fusca* were randomly selected and transferred into a pot of boiling water (100°C) in the ratio 4:1 (water: periwinkle). The periwinkles were allowed to boil for twenty minutes, then turned into a sieve to collect the leachate. The leachate (i.e. the drained solution after boiling) was allowed to cool, transferred into polyethylene bottles, and acidified with 1 ml concentrated nitric acid/litre of leachate prior to analysis. The tissue (edible portion) was removed from the shells with a needle, and dried in an oven at 60 °C to constant weight.

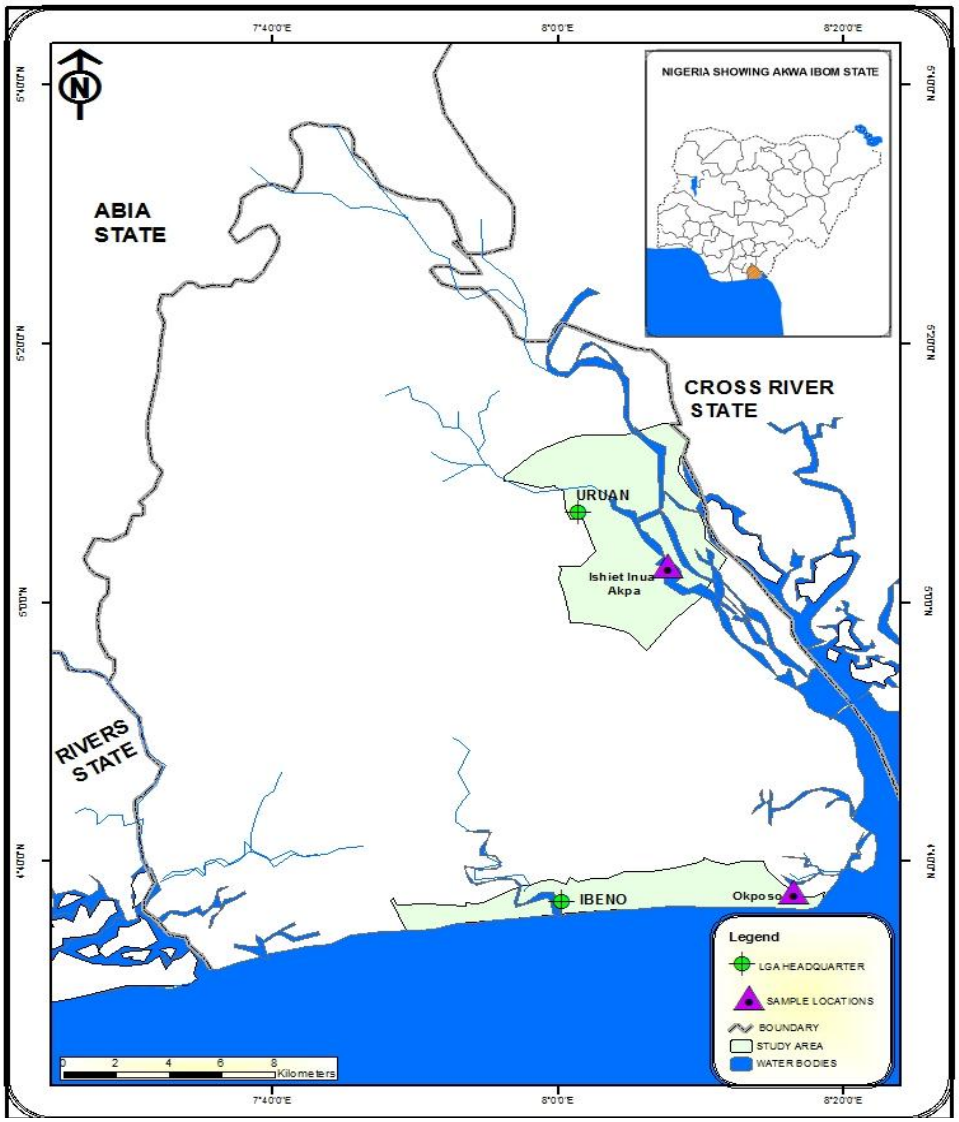

Figure 1. Map of Akwa Ibom State showing sample locations

Figure 2. Shells and tissues of *Pachymelania fusca* mutans (var)-(A) and *Tympanotonus fuscatus* radula (var)-(B)

The dried samples were ground to powder with a plastic mortar and pestle, passed through 0.5 mm mesh sieve, and stored in well-labelled plastic containers for digestion. The sediment samples were air-dried, and each sample was ground with a plastic mortar and pestle, passed through 2 mm mesh sieve and stored in well-labelled plastic containers for digestion.

2.2.1. Digestion of leachates and surface water samples

One hundred millilitres (l00 ml) of each water sample and leachate were measured into Teflon beakers and digested with 10 ml of concentrated HNO_3 on a hot plate to near dryness in a fume cupboard (APHA, 2005). The residue was dissolved with 0.5 M HNO_3, filtered quantitatively into 50 m1 standard flask using Whatman No. 42 filter paper and made to mark with dilute nitric acid (0.5 M). The same procedure was used for blank digest without the addition of the sample (Alam et al., 2002).

2.2.2. Digestion of sediment samples

Two grammes (of sediment samples (dry wt.) was digested with a mixture of hydrochloric acid and nitric acid (3:1) according to the ISO 11466 procedure on a temperature regulated hot plate (ISO, 1995). The blank digest was treated the same way.

2.2.3. Digestion of periwinkles tissue

A modified method of De Wolf et al. (2000) was used for the digestion of periwinkle tissues. Two grammes of periwinkle was weighed into 50 ml Teflon beaker. The digestion was done using 20 ml mixture of nitric acid and perchloric acid (2:1) boiled on a hot plate in a reflux system to obtain a clear solution, which was then evaporated to near dryness in a fume cupboard. The cooled residue was dissolved in 0.5 M HNO_3, filtered quantitatively into 50 m1 standard flask using Whatman No. 42 filter paper and made up to mark with dilute nitric acid (0.5 M). The same procedure was used for the blank digest.

2.3. AAS analysis of samples

The digested samples of water, sediment, periwinkle tissue, and leachate were analysed for cadmium, nickel, zinc, iron, copper and lead using atomic absorption spectrophotometer (UNICAM SOLAAR 969 model).

The content (w) of the elements in the sediment, periwinkle tissue, water and leachate samples was calculated using equation 1.

$$w_{(M)} = (\rho_1 - \rho_0)*f*V/m \text{ (or v)} \quad (1)$$

Where, $w_{(M)}$ is the mass fraction of the element M in the sample, in mg/kg or mg/l; ρ_1 is the concentration of the element, in milligrams per litre, corresponding to the absorbance of the diluted test portion; ρ_0 is the concentration of the element, in milligrams per litre, corresponding to the absorbance of the blank test solution; f is the dilution factor of the diluted test portion, if applicable; V is the volume, in litres, of the test portion taken for the analysis; m is the mass of the sample, in kilograms, corrected for water content (for solids); and v is the volume of sample digested, in litres (for liquid samples).

2.4. Quality control

Samples were analysed in triplicates alongside method blanks. All glassware and polyethylene containers were thoroughly washed with warm soapy water, rinsed with tap water, soaked in 10% nitric acid for 24 hours, and rinsed with deionised water. They were dried and kept in airtight containers prior to use. All the reagents were analytical grade purchased from British Drug House Chemicals (BDH, UK). Standard solutions of elements for calibration were prepared from 1000 ppm stock standard solutions (BDH, UK). Analytical procedures were validated by carrying out spiked recovery studies on the samples with known concentrations of the standard solutions in five replicates (Onianwa et al., 2001; Akinyele and Shokunbi, 2015; Ihedioha et al., 2017). The percentage recoveries of elements in samples were computed using equation 2.

$$\text{Recovery (\%)} = \frac{C_{spiked} - C_{unspike}}{C_{spiked}} \times 100 \quad (2)$$

The mean recoveries (±SD) % were 93±0.99, 78±2.36, 102±0.78, 83±1.11, 98±1.42 and 90±1.58 for Cd, Ni, Zn, Fe, Cu, and Pb, respectively. Sample blanks were included after every 10 samples.

Standard reference material - SRM 2710A (Montana 1 Soil) from the National Institute of Standards and Technology (NIST) was also analysed with the same protocol for sediment in three replicates. The recoveries of elements in SRM 2710A were computed using the relationship: measured value/Certified value x 100%. The results are presented in Table 1; and was in good agreement with NIST certified values.

The instrument's limits of detection (LOD) and limit of quantification (LOQ) for each element were determined as 3.0σ and 10σ, respectively for ten independent measurements of reagent blanks and expressed in mg/l (Table 1). Sample blank were included after every 10 samples.

Table 1. Measured and Certified Values (dry weight) for selected elements, LOD and LOQ

Element	NIST SRM 2710A – Montana 1 soil			LOD (mg/l)	LOQ (mg/l)
	Measured Value (mg/kg)	Certified Value (mg/kg)	% Recovery		
Cd	13.1 ± 0.9	12.3 ± 0.3	106	0.63	2.08
Ni	7.05 ± 0.8	8.0 ± 1.0*	88	0.07	0.24
Zn	3805.9 ± 115	4180 ± 150	91	0.21	0.69
Fe	34752 ± 332	43200 ± 800	80	1.14	3.78
Cu	2800 ± 35	3420 ± 50	82	0.21	0.69
Pb	4949.6 ± 19	5520 ± 30	90	0.61	0.20

** Reference value*

2.5. Estimation of bioaccumulation factor

Bioaccumulation factor (BAF) describes the absorption and distribution of a substance in an organism after exposure in a given environmental matrix (Subotić et al., 2013); and was used here to ascertain the periwinkle species with greater health hazard (DeForest et al., 2007). It is calculated as the ratio of the contaminant levels in the periwinkle tissues to those in sediment (Liao and Ling, 2003; Javed and Usmani, 2013), equation 3.

$$BAF = C_{(periwinkle)}/C_{(sediment)} \quad (3)$$

2.6. Health risk assessment of trace elements from seafood

2.6.1. Estimated daily intake

The exposure assessment for ingestion of contaminated periwinkles was first calculated using equation 4 (Huang et al., 2013):

$$EDI_{periwinkle} = \frac{C_m \times IRP \times CF \times EF \times ED}{BW \times AT} \quad (4)$$

where, EDI is the estimated daily intake of trace elements in periwinkles (mg/kg/day; *Pachymelania fusca* or *Tympanotonus fuscatus*); C_m is the trace elements concentration in periwinkles (mg/kg); IRP is ingestion rate, and is 0.036 Kg/person/day for periwinkles [Periwinkle ingestion rate (IRP) was obtained from one-on-one interview and questionnaire survey of 110 adult participants, aged 17 to 60 years (Onianwa et al., 2001; Chien et al., 2002). The number of periwinkles consumed for breakfast, lunch and dinner were averaged and used to compute the ingestion rate per day]; CF is the conversion factor of fresh weight to dry weight intake rates of periwinkle tissues, using moisture percentage in periwinkle species (Urban et al., 2009) and is 0.2 for

Pachymelania fusca and 0.17 for *Tympanotonus fuscatus*; EF is exposure frequency (365 days/year); ED is exposure duration (70 years); BW is body weight (70 kg); and AT is the averaging time (ED x EF, in days).

In the case where the dish is prepared with the periwinkle shells without par-boiling, (as described in the introduction, method b), the consumers will be exposed to the trace elements in the leachates as well. The intake of elements from leachate calculated using equation 5 (Liu et al., 2013).

$$EDI_{leachate} = \frac{C_m \text{ x } IRL \text{ x } EF \text{ x } ED}{BW \text{ x } AT} \quad (5)$$

Where, EDI is the estimated daily intake of trace elements in periwinkles leachate (mg/L/day; from *Pachymelania fusca* or *Tympanotonus fuscatus*); C_m is the concentrations of trace elements in the leachates, IRL is the ingestion rate estimated as 0.10 L/day of leachates (given an average of 0.05 L water content per plate of food and considering the Nigerian feeding habit (Latunde-Dada, 1990). It is also important to note that the moisture contents may vary widely across different varieties of dishes (Onabanjo and Oguntona, 2003); other parameters in equation 5 were as described in equation 4. The interactions from other recipes in the meal were assumed to be of no effect in this study.

2.6.2. Hazard quotient

The non-carcinogenic risk – the hazard quotient (HQ), for individual elements in periwinkle tissues and leachates was calculated as the ratio of the exposure level (intake) to a reference dose (RfD) for the element derived from a single exposure period (USEPA, 1989; 2000), equation 6.

$$HQ = EDI/RfD \quad (6)$$

Where, RfD is the oral reference doses (mg/kg/day), for Cd=1.00E-03; Ni=2.00E-02; Zn=3.00E-01; Fe=7.00E-01; Cu=4.00E-02 (USEPA, 2017); and Pb=4.00E-03 (USEPA< 2007). HQ<1 means there is no obvious adverse effect on the exposed population, whereas HQ≥1 implies potential health risk.

2.6.3. The target health quotient

The target health quotient (THQ) for combined risk of trace elements is given by the sum of each individual element (Hallenbeck, 1993; Lee et al., 2006), (equations 7a and b)

$$THQ_{(periwinkle\ tissue)} = HQ_{(element1)} + HQ_{(element\ 2)} + \ldots\ldots + HQ_{(element\ n)} \quad (7a)$$

$$THQ_{(leachate)} = HQ_{(element1)} + HQ_{(element\ 2)} + \ldots\ldots + HQ_{(element\ n)} \quad (7b)$$

THQ=1.0 is the threshold value for non-carcinogenic risks recommended by USEPA (2017); THQ>1, implies potential non-carcinogenic health risk.

2.6.4. Hazard index

The hazard index (HI) developed by USEPA (1986), was used to assess the overall potential non-carcinogenic risk from more than one element. When HI≥1, there is a potential health risk. HI, for combined pathway was calculated as follows (Equation 8):

$$HI = THQ_{(periwinkle\ tissue)} + THQ_{(leachate)} \quad (8)$$

3. Results and discussion

3.1. Levels of trace elements in monitored matrices

The levels of trace elements (Cd, Ni, Zn, Fe, Cu, and Pb) in the periwinkle species -*Pachymelania fusca* and *Tympanotonus fuscatus*, their leachates, sediment and water samples are presented in Table 2. Trace elements levels varied widely between the studied matrices. Bottom sediment acts as sink to chemical compounds deposited in the aquatic environments, and recently considered an active aquatic compartment that performs a fundamental role in the redistribution of chemical compounds to the aquatic biota (Idris et al., 2007; Pasos et al., 2010). In most cases, the periwinkle samples recorded higher concentrations of trace elements compared to sediment, water and leachate samples. Periwinkles being bottom feeders, the trend is in agreement with previous reports that at lower exposure concentrations in environmental media, element uptake by biota increases (DeForest et al., 2007). Variations observed in the periwinkle species could be explained by inherent aquatic environment dynamics, the physiological characteristics of *Pachymelania fusca* and *Tympanotonus fuscatus* as well as the anthropogenic input from such elements as are present in the surrounding locations (Alemdaroglu et al., 2003; Suffern et al., 1981; Zhang and Wong, 2007). Copper was below detection limit in the water samples at Ibeno station. The least amount of quantifiable trace elements was recorded in water and leachate samples.

Higher concentrations (mg/kg) of Ni (48.3), Cd (39.5), Cu (22.1), and Fe (5.60) were found in the tissue of *Pachymelania fusca*, and Zn (57.2) and Pb (20.2) in *Tympanotonus fuscatus* (Table 2). Concentrations of trace elements (mg/kg) in periwinkles have been reported as 0.31 (Cd), 3.80 (Ni), 22.9 (Zn) and 12.1 (Cu) (Onianwa et al., 2000; 2001); ND (Cd), 25.0 (Ni), 710 (Zn), 3500 (Fe), 230 (Cu) and 10.0 (Pb) (Udoh, 2000); and 0.391 (Cd), 11.058 (Ni) (Table 3), and 1.241 (Pb) (Marcus et al., 2013). The levels of Cd, Zn, Fe and Cu in *Pachymelania fusca* and *Tympanotonus fuscatus* tissues were lower, while Ni in *Pachymelania fusca* from Ibeno and Pb in *Tympanotonus fuscatus* from Ishiet were higher than that reported in the Niger Delta region (Udoh, 2000). Zn in periwinkles from Ibadan, southwest Nigeria (Onianwa et al. 2001), and Pb in shellfish (Marcus et al., 2013) was higher than that observed in *Pachymelania fusca* from Ibeno in this study (Table 3). The trends in trace elements' levels were Ni>Cd>Cu>Zn>Fe>Pb (*Pachymelania fusca* from Ibeno); Zn>Cd>Cu>Ni>Pb>Fe (*Tympanotonus fuscatus* from Ishiet). Cd in tissues of both species were higher than various agencies' recommended limits (FAO, 1983; FDA, 2001; EC, 2001; Javed and Usmani, 2013; Taweel et al., 2013) (Table 2). Ni in both species, and Zn and Pb in *Tympanotonus fuscatus* were higher than the FAO/WHO permissible limit (Javed and Usmani, 2013; Taweel et al., 2013). Comparison of periwinkle species in this study with other shellfish (Ololade et al., 2008; Kang et al., 2000; Gbaruko and Friday, 2007; Sarkar et al., 2008; Rumisha et al., 2012; Sivaperumal et al., 2007; Hedouin et al., 2009; De Mora et al., 2004) are presented in Table 3.

Leachate, which is the solution obtained after par-boiling the periwinkles (as described in section 2.2) was analysed to determine the amount of elements leached from the two species of periwinkles. The pattern of elements in leachate was in the order Cd>Ni>Pb>Cu>Zn>Fe for *Pachymelania fusca* and Ni>Fe>Zn>Cd>Pb>Cu for *Tympanotonus fuscatus*. Of all the leached elements, only Cd (1.39 mg/kg) in *Pachymelania fusca* was greater than FAO/WHO dietary allowance of 1.0 mg/kg (Javed and Usmani, 2013; Taweel et al., 2013) (Table 2). Par-boiling appeared to reduce health risk in Cd-contaminated periwinkles: the periwinkles freed some amount of elements in the leachate; hence, par-boiling and draining off the water could be recommended as an effective method of preparation. This may reduce the health risk to humans, if not in the short then the long-term. The concentration of elements in leachates from *Pachymelania fusca* and *Tympanotonus fuscatus* were compared with the WHO recommended standards for drinking water, and Cd, Ni, Pb in leachates from both *Pachymelania fusca* and *Tympanotonus fuscatus* and Fe from *Tympanotonus fuscatus* were above the recommended levels (WHO, 2008) (Table 2).

In water, the highest concentrations (mg/kg) of Cd (1.27) and Ni (0.8) were found in Ibeno; and Zn (1.33), Fe (0.04) and Cu (0.03) in Ishiet. The elements trend was in the order Cd>Ni>Zn>Pb>Fe>Cu (Ibeno) and Zn>Cd>Ni>Pb>Fe>Cu (Ishiet). The levels of elements in Ibeno and Ishiet water were compared with permissible limits for aquaculture of USA, Kenya, India, and FAO (PHILMINAQ, 2017) (Table 2). Cd levels in the two locations were much higher than all permissible limits for aquaculture. Ni from Ibeno was 2-folds greater than the Kenyan limit but lower than for USA. Pb was above the Kenyan and Indian limits, but 3-folds lower than the USA limit. Zn and Pb in the two coastal waters (Ibeno and Ishiet), and Cu in Ishiet water were above the stipulated range for healthy fish cultivation set by FAO (Svobodová et al., 1993). These high levels may be due to the peculiar anthropogenic activities including oil and gas exploitations within the Niger Delta region. Cd, Ni and Fe were lower than the limits.

The health and diversity of the aquatic system depend on clean water and sediment, hence the need for the protection of sediment dwellers or organisms that depend on them as source of food, and humans who consume fish and shellfish. The mean levels (mg/kg) of trace elements in sediment ranged from 0.01 (Fe) to 48.5 (Ni) in Ibeno and 1.0 (Cu) to 57.2(Ni) in Ishiet; and trend in the order Ni>Pb>Cd>Zn>Cu>Fe (Ibeno) and Zn>Ni>Cd>Pb>Fe>Cu (Ishiet). Cd in sediment exceeded the recommended standards; Zn, Cu, and Pb were within limit (Washington State Department of Ecology, 2017).

Table 2. Levels of trace elements in periwinkles (*Pachymelania fusca* and *Tympanotonus fuscatus*), leachate, water and sediment samples

Location	Sample / Standard	Cd	Ni	Zn	Fe	Cu	Pb	Reference
Ibeno	*Pachymelania fusca* (mg/kg dw)	39.5±0.5	48.3±1.4	14.1±6.1	5.6±0.2	22.1±2.0	0.8±0.4	Study
Ishiet	*Tympanotonus fuscatus* (mg/kg dw)	34.3±0.7	20.3±0.4	57.2±6.1	5.1±2.5	21.5±12.3	20.2±2.3	Study
Permissible limits	Food (mg/kg)	1.0	2.0	50.0	263.0	30.0	1.30	Javed and Usmani, 2013; Taweel et al., 2013
Permissible limit for shellfish (μg/g dry wt)	FAO (1983)	0.5	na	40	na	30	0.5	FAO, 1983
	FDA (2001)	4.0	80	na	na	na	na	FDA, 2001
	EC (2001)	1.0	na	na	na	na	1.0	EC, 2001
Ibeno	Leachate (mg/L)	1.39±0.8	1.30±0.8	0.8±0.6	0.05±0.1	0.11±0.2	1.18±0.3	Study
Ishiet	Leachate (mg/L)	0.35±0.4	1.91±0.9	0.40±0.2	1.33±0.08	0.04±0.0.02	0.06±0.9	Study
Permissible limits	Drinking water (mg/L)	0.003	0.07	3.0	0.3	2.0	0.01	WHO, 2008
Ibeno	Water (mg/L)	1.27±0.8	0.8±1.0	0.43±0.4	0.02±5.3	BDL	0.06±0.7	Study
Ishiet	Water (mg/L)	1.0±0.04	0.13±1.6	1.33±0.08	0.04±3.5	0.03±0.02	0.06±0.9	Study
Permissible limits	Marine Water (mg/L)							
	USA	0.042	0.074	na	na	na	0.21	PHILMINAQ, 2017
	Kenya	0.01	0.3	na	na	na	0.01	PHILMINAQ, 2017
	India	0.001	na	na	na	na	0.001	PHILMINAQ, 2017
	FAO 1993	2.0-20	10	0.01-0.1	0.2	0.001-0.01	0.004-0.008	Svobodová et al., 1993
Ibeno	Sediment (mg/kg dw)	27.1±0.6	48.5±7.8	20±7.2	0.01±1.1	0.1±0.1	27.7±7.5	Study
Ishiet	Sediment (mg/kg dw)	23.9±0.1	57.2±0.9	157±6.2	1.2±0.2	1.0±0.06	7.4±1.4	Study
Permissible limits	Marine Sediment (mg/kg dw)	5.1	na	410	na	390	450	Washington State Department of Ecology, 2017

BDL = below detection limit; na = not available; dw = dry wet.

Table 3. Levels of trace elements in periwinkles and other shellfishes reported in the literature (mean values in mg/kg dry)

Location	Species	Cd	Ni	Zn	Fe	Cu	Pb	References
Ibeno, Nigeria	*Pachymelania fusca*	39.5	48.3	14.1	5.6	22.1	0.80	Present study
Ishiet, Nigeria	*Tympanotonus fuscatus*	34.3	20.3	57.2	5.10	21.5	20.2	Present study
Niger Delta, Nigeria	*Pachymelana byronensis*	BDL	25	710	3500	220	10	Udoh, 2000
Ibaban, Nigeria	*Periwinkle*	0.13	3.80	22.9	na	12.1	na	Onianwa et al., 2000 and 2001
Scheldt (Bath), Belgium	*Littorina litorea*	5.23	4.6	106	1214	138	1.6	De Wolf et al., 2000
Ondo coastal region, Nigeria	*Littorina litorea*	0.64–2.91	0.20 – 4.10	0.10	1315.5	310.2	0.02–0.52	Olalade et al., 2008 and 2011
Korean coast	*Littorina brevicula*	0.07–2.85	BDL–36.1	51– 186	BDL–2600	25–264	0.05–9.02	Kang et al., 2000
Niger Delta, Nigeria	*Littorina litorea*	na	3.48	30	na	7.8	5.81	Gbaruko and Friday, 2007
Northeast Coast of Bay of Bengal – India	*Anadara granosa* (bivalve)	na	na	495	na	150.6	72.61	Sarkar et al., 2008
Dar es Salaam coast, Tanzania	*Mactra ovalina* (bivalves)	0.1	5.0	65.0	1425	15	4.2	Rumisha et al., 2012
Cochin, India	*Oreochromis mossambiscus*	na	0.62	4.56	na	1.12	0.2	Sivaperumal et al., 2007
Boulari Bay, New Caledonia	*Isognomon isognomon* (oyster)	1.28	16.0	1,718	na	9.8	na	Hedouin et al., 2009
Gulf of Gulf, Oman	*Saccostrea cucullata* (oyster)	8.99–21.9	0.80–3.14	391–1610	na	0.69–276	0.384-0.673	De Mora et al., 2004

Note: BDL = below detection limit; na = not available

3.2. Bioaccumulation factor (BAF) of trace elements in *Pachymelania fusca* and *Tympanotonus fuscatus*

The mobility of trace elements from sediment to the edible parts of periwinkle species, bioaccumulation factor (BAF), was estimated as the ratio of the concentration of trace element in periwinkle to concentration in sediment. BAF of the trace elements in the periwinkle species (which are bottom feeders) are presented in Table 4. BAF<1 indicates no contamination of the periwinkle; 1>BAF≤10, the periwinkle is tolerant and BAF>10, a hyperaccumulator (Ávila et al., 2017). Based on the classification, there was no contamination to Zn and Pb *in Pachymelania fusca*, and Ni and Zn in *Tympanotonus fuscatus*. *Pachymelania fusca* was tolerant to Cd and Ni while *Tympanotonus fuscatus* was tolerant to Cd, Fe and Pb. *Pachymelania fusca* hyperaccumulated Fe and Cu, and *Tympanotonus fuscatus*, hyperaccumulated Cu. The high BAFs could be explained on local environmental pollution and feeding habits of the two species of periwinkles. The BAF of trace elements in the periwinkle was in the order: Fe>Cu>Cd>Ni>Zn>Pb (*Pachymelania fusca*), and Cu>Fe>Pb>Cd>Zn>Ni (*Tympanotonus fuscatus*).

Kang et al. (2000) observed a correlation between body weight and trace element concentrations. It was not the case in this study as *Tympanotonus fuscatus* with relatively bigger body sizes accumulated lower concentration of trace elements, except for Pb. This study corroborated the findings of De Wolf et al. (2000), of no correlation between body sizes and trace element concentrations. The differences in correlation may well be explained by the physiological characteristics and the trophic levels across the biological species.

Table 4. Calculated bioaccumulation factors of trace elements for *Pachymelania fusca* and *Tympanotonus fuscatus*

Trace elements	Bioaccumulation factor (BAF)	
	Pachymelania fusca	*Tympanotonus fuscatus*
Cd	1.46	1.44
Ni	1.00	0.35
Zn	0.71	0.36
Fe	560.00	4.25
Cu	221.00	21.50
Pb	0.03	2.73

3.3. Health risk assessment of trace elements from consumption of *Pachymelania fusca* and *Tympanotonus fuscatus*

3.3.1. Estimated daily intake

The *EDI* (mg/kgbw/day) of six trace elements (Cd, Ni, Zn, Fe, Cu, and Pb) were computed based on daily consumption of 0.036 kg/day *Pachymelania fusca* and *Tympanotonus fuscatus* and 0.1 L of their leachate from the two coastal locations, Ibeno and Ishiet. Presented in Table 5 are the EDI through the consumption of the two species of periwinkles and their leachates (if consumed without par-boiling) and the recommended oral

RfD values (USEPA, 2007; 2017). The RfD values state the estimated daily exposure to which the human population may continually be exposed over a life time without appreciable health risk (Saha and Zaman, 2013). The EDIs of Ni, Zn, Fe, Cu, and Pb were lower than the oral RfDs value suggesting that the elements in periwinkle tissues posed no health risk. EDIs of Cd in the tissues were 4-folds in *Pachymelania fusca* and 2-folds in *Tympanotonus fuscatus* higher than the oral RfD; also, higher for leachate from *Pachymelania fusca*. The total daily intake, the sum of the EDI (tissue) and EDI (leachate), described the total daily intake if the periwinkles were not par-boiled before use for meal preparation. These were 6-folds (*Pachymelania fusca*) and 3-folds (*Tympanotonus fuscatus)* higher than the reference point, and might pose a health risk to consumers. The result indicated more daily intake of Cd if the periwinkles were not par-boiled before use. The daily intake of elements varied in the two coastal areas (Table 5). The estimated daily intake of Cd, Ni, Fe and Cu were higher through the consumption of *Pachymelania fusca* from Ibeno, while Zn and Pb from *Tympanotonus fuscatus* consumption were higher for Ishiet.

3.3.2. Non-carcinogenic risks of trace elements through consumption of Pachymelania fusca and Tympanotonus fuscatus and their leachates

Hazard Quotient. Health risks by the adult population from consumption of contaminated *Pachymelania fusca* and *Tympanotonus fuscatus* harvested from the study area were determined from the HQ. If HQ>1, the consumers were likely to experience obvious adverse effects. The estimated HQs and the cumulative risks (THQs) are presented in Table 6. The HQ of Cd was greater than one in both tissues of *Pachymelania fusca* and *Tympanotonus fuscatus*, and leachate from *Pachymelania fusca*; and indicated considerable health hazard of Cd from consumption of these seafood in the study area. HQs of other individual trace elements, Ni, Zn, Fe, Cu, and Pb were less than one, implying no non-carcinogenic risk of these elements through the consumption of both *Pachymelania fusca* and *Tympanotonus fuscatus* tissues and leachate. The total diet HQs of Cd, the sum of the HQ (tissue) and HQ (leachate) were 6.049E+00 and 3.499E+00 for *Pachymelania fusca* and *Tympanotonus fuscatus*, respectively and signified the risk in the population that did not treat (par-boil) periwinkles before use. The consumption of high Cd containing *Pachymelania fusca* and *Tympanotonus fuscatus* posed threat to health of the adult population (Table 6).

TABLE 5. Estimated daily intake of trace elements (mg/kg bw/day) through the consumption of *Pachymelania fusca and Tympanotonus fuscatus* compared with the oral reference doses (RfDo)

Periwinkle/Daily Reference intake	EDI (mg/kg/day) of trace elements					
	Cd	Ni	Zn	Fe	Cu	Pb
Pachymelania fusca tissue	4.063E-03	4.968E-03	1.450E-03	5.760E-04	2.273E-03	8.23E-05
Pachymelania fusca leachate	1.986E-03	1.857E-03	1.143E-03	7.143E-05	1.570E-04	1.686E-03
Total intake	6.049E-03	6.825E-03	2.593E-03	6.474E-04	2.430E-03	1.768E-03
Tympanotonus fuscatus tissue	2.999E-03	1.775E-03	5.001E-03	4.459E-04	1.880E-03	1.766E-03
Tympanotonus fuscatus leachate	5.000E-04	2.729E-03	5.714E-04	1.900E-03	5.71E-05	8.57E-05
Total intake	3.499E-03	4.503E-03	5.572E-03	2.346E-03	1.937E-03	1.852E-03
RfDo (mg/kg/day) [38,39]	1.000E-03	2.000E-02	3.000E-01	7.000E-01	4.000E-02	4.000E-03

RfDo *is the oral reference dose*

Table 6. HQ of individual trace elements, the THQ and HI from the consumption of *Pachymelania fusca and Tympanotonus fuscatus* tissues and leachates

Periwinkle	HQ						THQ
	Cd	Ni	Zn	Fe	Cu	Pb	
Pachymelania fusca tissue	4.063E+00	2.484E-01	4.834E-03	8.229E-04	5.683 E-02	2.057 E-02	4.394E+00
Pachymelania fusca leachate	1.986E+00	9.286E-02	3.810E-03	1.020E-04	3.929 E-03	4.214 E-01	2.508E+00
Total Diet HQ	6.049E+00	3.413E-01	8.644E-03	9.249E-04	6.076 E-02	4.420 E-01	6.902E+00* (HI)
Tympanotonus fuscatus tissue	2.999E+00	8.874E-02	1.667E-02	6.370E-04	4.699 E-02	4.415 E-01	3.593E+00
Tympanotonus fuscatus leachate	5.000E-01	1.364E-01	1.905E-03	2.714E-03	1.429 E-03	2.143 E-02	6.639E-01
**Total Diet HQ	3.499E+00	2.252E-01	1.857E-02	3.351E-03	4.842 E-02	4.629 E-01	4.257E+00* (HI)

***HQ** is the hazard quotient / **THQ** is the target hazard quotient*
** **HI (hazard index)** is the sum of THQ values for the combined (tissue and leachate) non-carcinogenic effects of multiple trace elements*
*** **Total diet HQ** would be the risk value when the periwinkles are not treated (i.e. Par-boil sufficient water for about 20 minutes) before use in preparation of a meal.*

Target Hazard Quotient. The cumulative risks of trace elements (THQ) through consumption of *Pachymelania fusca* and *Tympanotonus fuscatus* tissues was greater than one. The values were 4.394E+00, 2.508E+00, 3.593E+00, and 6.639E-01 for *Pachymelania fusca* tissue, *Pachymelania fusca* leachate, *Tympanotonus fuscatus* tissue, and *Tympanotonus fuscatus* leachate, respectively (Table 6). The high values suggested that the adult population in Akwa Ibom State might have significant non-carcinogenic health risk through consumption of high level Cd in *Pachymelania fusca* and *Tympanotonus fuscatus*; with the greater risk from *Pachymelania fusca*.

The tissue and leachates of *Pachymelania fusca* contributed 64% and 36%, respectively of the combined trace elements to the THQ, while *Tympanotonus fuscatus* contributed 84% and 16%, respectively. This implied that the total health risk could be reduced by16 - 36 % if periwinkles were par-boiled before use in meal preparation. It is recommended for periwinkles to be par-boiled to leach out some amount of trace elements before use in meal preparation. The percentage contribution of single elements to the total diet HQ (TDHQ) which is the sum of HQ_{tissue} and $HQ_{leachate,}$ represented the population that used periwinkles directly without par-boiling; and was in the order Cd>Pb>Ni>Cu>Zn>Fe: 88%, 6%, 5%, 0.9%, 0.1% and 0.01%, respectively (for *Pachymelania fusca*) and 82%, 11%, 5.3%, 1.1%, 0.4% and 0.08%, respectively (for *Tympanotonus fuscatus*); the main cause of risk being Cd.

Hazard Index. HI, the sum of THQ in tissue and leachate is shown in Table 6. The HI values through the consumption of *Pachymelania fusca* from Ibeno and *Tympanotonus fuscatus* from Ishiet were 6.902E+00 and 4.257E+00, respectively. Cd was the main contributor to health risk. The result further revealed that the population, which do not par-boil the seafood before use for meal preparation, was at higher risks, particularly from consumption of *Pachymelania fusca*.

4. Conclusion

The levels of trace elements were assessed in two periwinkle species (*Pachymelania fusca* and *Tympanotonus fuscatus*) collected from two coastal locations, Ibeno and Ishiet in Akwa Ibom State, Nigeria; their leachates (after par-boiling), water and sediment. Bioaccumulation factor and human health risk assessment of periwinkles consumption were also determined. The ingestion rate of *Pachymelania fusca* and *Tympanotonus fuscatus* was 0.036 kg/person/day, obtained from a survey conducted among 110 adult participants aged 17-60 years.

Trace element levels varied between species of the periwinkles. Higher levels of Ni, Cd, Cu, and Fe were recorded for *Pachymelania fusca*, and Zn and Pb for *Tympanotonus fuscatus*. Par-boiling the periwinkles prior to meal preparation leached out some amount of trace elements. Leachate from *Pachymelania fusca* contained more Cd, Zn, Cu and Pb while *Tympanotonus fuscatus* had more Ni and Fe. None of the elements in leachates from both species, except Cd in *Pachymelania fusca* exceeded the FAO/WHO recommended value for food. Par-boiling, as a method of preparation could reduce the overall health risk of trace elements through the consumption of periwinkles, if not in the short then the long-term.

Higher values of Cd and Ni (Ibeno) and Zn, Fe, and Pb (Ishiet) were found in the coastal water; but Cd alone in the two locations exceeded the recommended range for aquaculture. Trace element levels (mg/kg) in coastal sediment ranged from 0.01 (Fe) to 48.5 (Ni) in Ibeno and 1.0 (Cu) to 57.2 (Ni) in Ishiet; with Cd above the recommended limit in both locations.

The BAF results revealed that *Pachymelania fusca* was not contaminated with Zn and Pb, tolerated Cd and Ni, and hyperaccumulated Fe and Cu; while *Tympanotonus fuscatus* was not contaminated with Ni and Zn, tolerated Cd, Fe and Pb, and hyperaccumulated Cu. *Pachymelania fusca* species bioaccumulated more of the trace elements.

Risk assessment revealed exposure to trace elements through the consumption of periwinkles. The HQ of the single elements - Ni, Zn, Cu, Fe and Pb showed non-carcinogenic health effect for consumption of *Pachymelania fusca* and *Tympanotonus fuscatus* tissues and their leachates. However, with hazard quotient above one, Cd was the main contributor to health risk in both periwinkle species and leachate of *Pachymelania fusca*. The combined elements exposures indicated high health risk (THQ > 1). Consumption of *Pachymelania fusca* species portends more risks than *Tympanotonus fuscatus*. Regular monitoring of edible periwinkles in the region, particularly Ibeno is highly recommended to prevent adverse health issues in the future. In addition, par-boiling which reduced the amount of trace elements in the periwinkles could be an effective method of processing shellfish from contaminated environment.

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