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Research article

Chemometrics, health and environmental risk assessments of commonly consumed biscuits in Lagos and Ibadan metropolises, Southwestern Nigeria

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ABSTRACT

The United Nations' Agenda 2030 for sustainable development calls, amongst others, for universal action toward ending malnutrition and ensuring healthy living and well-being for all. So, efforts have intensified to attain the sustainable development goal-2 targets on stunting and wasting in children. Reported herein, therefore, is the quantification of metals in biscuits. Biscuits are commonly consumed snacks world-over and have become sources of nourishment for children and adults due to growing sedentary lifestyles and hectic school/work schedules. Nine metals (Pb, Ni, Cu, Co, Zn, Fe, Na, Mg and Ca) were assayed in six biscuit types (crackers, cookies, shortcakes, digestives, cabins and wafers) via wet digestion and flame atomic absorption spectrophotometry, and the ensuing data subjected to multivariate analyses (analysis of variance, Tukey's test, Pearson correlation, and principal component and hierarchical cluster analyses). The highest concentrations of macrominerals were found in the wafers (Ca), crackers (Na) and cabins (Mg) whereas the micronutrients peaked in the cookies (Fe, Zn), crackers (Cu), shortcake (Co) and wafers (Ni), respectively. The metal levels in the sampled biscuits were all safe for consumption, except for Pb at 0.83 ± 0.76–2.3 ± 1.3 mg/kg. Similarly, the health risk assessments of ingesting metals from the biscuits exposed Pb as potentially liable to cause adverse non-carcinogenic and carcinogenic health effects in children (aged 4–20 years) but Co and Ni exhibited borderline non-carcinogenic and carcinogenic health risks, respectively, in children. Gratifyingly, the ecological risk assessments to evaluate the likelihood of wastes, from expired and/or egested potentially toxic metals-contaminated biscuits, to cause damage to ecology were categorized as low. Nonetheless, constant evaluation and monitoring remain germane.

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1. Introduction

The United Nations' sustainable development goals, specifically goal 2 (SDG-2: Zero Hunger) and goal 3 (SDG-3: Good Health and Well-Being), which aim at ending hunger by achieving food security and improved nutrition as well as promoting sustainable agriculture; and ensuring healthy living and well-being for all ages, respectively, reinforce the global call to universal action towards making the world a better place for all [1,2]. It is envisaged that the internationally agreed targets on stunting and wasting in children (<5 years old), for example, would be achieved by 2025 and, by 2030; end preventable deaths of 5-year-olds, and younger, by reducing their mortalities to 12 per 1000 and 25 per 1000 live births for neonatal and under-5-year-olds, respectively [1]. Nonetheless, it has been reported [2] that, in 2020, about 10 % of the world's population was undernourished, with 6.7 % and 22 % of malnourished children, under 5 years of age, experiencing wasting and stunting, respectively, whereas 33 % of women of child-bearing age suffered nutrition deficiency-related anemia.

Globally, snacks, such as biscuits, are increasingly becoming commonplace sources of nourishment for children and adults alike due to rising sedentary lifestyles amongst many urban-dwellers and hectic work/school schedules [3,4]. They can augment daily energy balances as well as improve mental acuity. Biscuits are typically pastry-based foods, which are mainly composed of flour, water, sugar, fat and salt, and are classified by their texture and constituents' ratios as: crackers, cabins, shortcakes, wafers, digestives and cookies [5]. They are ready-to-eat, portable and affordable. They also have long shelf-lives and a wide range of tastes and nutritional ingredients [6]. In addition, biscuits are less prone to microbial spoilage and, therefore, find appeal with many consumers. The biscuits industry in Nigeria, for instance, is estimated to be worth over $1.1 billion, with a total annual consumption of over 0.5 million metric tons [7]. Besides, the sales of snacks are projected to increase into 2024 at 17.9 % per annum [8].

Pollutants in the air, soil, water and wastewaters are of grave concern worldwide because they are harmful to humans, microbes and the environment [9,10] as are food contaminants [11]. Moreso, the occurrence of heavy metals' contamination in biscuits can be linked to environmental and industrial factors, including contaminants from raw materials, production/processing lines and packaging [12]. Heavy metals are elements (metals or metalloids) with atomic densities of >4 g/cm^3 that are non-biodegradable, but destroyable, and persistent with long biological half-lives [13]. They can accumulate in vital human organs (e.g., liver and kidney) through different mechanisms, such as oxidative stress, interference with essential metals and interaction with cellular macromolecules, with concomitant progressive toxic effects [10,14].

It is notable that health problems, such as cancers of the lungs, kidneys, liver and upper gastrointestinal tract, and disabilities associated with malnutrition as well as impaired psycho-social behaviors, intrauterine growth retardation and decreased immunological defenses; due to the depletion of essential nutrients in the body, have been attributed to the consumption of heavy metal-contaminated foods [15]. Chronic human exposure to heavy metals is also reported to lead to asthma, kidney damage, osteoporosis, type II diabetes and Parkinson's disease, amongst others [16]. It is significant that certain heavy metals (e.g., copper, zinc and manganese) act as micronutrients at lower concentrations but become harmful at higher concentrations whereas other heavy metals, such as lead, mercury and cadmium, have no known biological functions in the human body but are typically toxic at trace and/or low concentrations [13,17–19].

Chemometrics use statistical and mathematical methods to achieve objective data evaluation and find applications in quality control and in the quantitative and qualitative determinations of chemical parameters for assessing the authenticity of foods and related products [20,21]. Human and ecological health risk assessments, on the other hand, are appropriate tools for assessing and quantifying probable adverse effects of different pollutants on human health and the environment [22]. This is because the calculations of risks' values can aid researchers and policymakers to plan and strategize on combating adverse health effects by removing or mitigating pollution sources, eliminating receptors and disconnecting the nexuses between the sources of pollution and their receptors. It is noteworthy that the probable health risks associated with heavy metals from the consumption of biscuits are typically influenced by the concentrations of the heavy metals therein and daily rates of biscuit consumption as well as the consumer's body weight and oral reference doses, amongst others [23].

It is, therefore, pivotal that in a world preoccupied with controlling energy calories from the dietary intakes of proteins, fats and sugars, comparable attention must also be paid to the ingestion of essential elements; which make metabolism possible, as well as to the presence of apparently toxic, non-essential ones. Consequently, and in line with the global quest to meet the sustainable development goals of healthy living and well-being for all, we report on the quantification of some metals in different brands of commonly consumed biscuits in the metropolises of Lagos and Ibadan (Nigeria) and environs. This is not only pertinent in view of the myriad sources of food contamination but also due to the complicated interactions of heavy metals with the biological and physical environments. Essentially, it is vital to evaluate and continuously monitor the affected media in order to mitigate and/or eliminate related deleterious effects. This study underscores the aforementioned salient issues and volunteers plausible recommendations. The health risks of some of these heavy metals to children and adults, as well as to the environment, are also assessed.

2. Materials and methods

2.1. Materials

Water (Milli-Q) and analytical grade reagents were purchased from Sigma Aldrich and were used without purification unless otherwise stated. The reagents included nitric acid, hydrogen peroxide, perchloric acid and lyophilized brown bread (BCR–191).

2.2. Study areas

Lagos is the most populous city and commercial epicenter of Nigeria; located in the west of Africa, between latitude 6° 27′ 14.65″ N and longitude 3° 23′ 40.81″ E [24], at an elevation of 11 m above sea level. It is the largest city in Sub-Saharan Africa with an estimated population of 13.5 million (2018) and 20.6 million (2030) [25], and projected to exceed 32.6 million by 2050 [26] but a 2024 estimate has the population at about 16.5 million [27]. Lagos consists of rural, peri-urban and urban settlements [28], situated on the Atlantic littoral of Nigeria. Its climate is tropical, with an average temperature of 27–30 °C, high (≥80 %) humidity and average monthly and annual rainfall of 244 mm and 1532 mm, respectively [29]. There are two major seasons: rainy and dry seasons; from April to October; with a short break in mid-August, and November to March, respectively [30]. Additionally, the vegetation in Lagos is characterized by a freshwater swamp and, to some extent, mangrove swamp as well as wet and dry lowland rainforests and southern guinea savanna [31]. The area is also drained by several river systems, such as the Ona and Osun rivers; in the east, Ogun river in the center and Yewa river in the west [28]. Most of them flow southwards into the Atlantic coastal lagoons.

Ibadan, on the other hand, is the third most populous city in Nigeria, with an estimated population of 3.4 million (2018) [25], 4.0 million (2024) [32] and 5 million (2030) [25], respectively. It is located 128 km inland northeast of Lagos and 530 km southwest of Abuja; the Federal Capital of Nigeria [33], between latitude 7° 22′ 39.22″ N and longitude 3° 54′ 21.28″ E, at 181 m above sea level [24]. Ibadan is characterized by a tropical climate with two distinct seasons, an average annual rainfall and temperature of 1205 mm and 28 °C, respectively, and a relative humidity of 75 % [34]. Its vegetation is that of tropical rainforest [35] and the geology is categorized by metamorphic pre-Cambrian basement complex rocks with gneisses as the predominant rock type [34] and drained by the Ogunpa, Kudeti and Ona rivers and their tributaries [36].

The sampling and manufacturing/storage sites of the biscuits in this study are delineated in Fig. 1, showing study areas in Lagos and Ibadan and environs.

2.3. Sample collection and preparation

Three different brands were each selected from six different types of biscuits (viz: crackers, cookies, shortcakes, digestives, cabins and wafers) obtained from different locations within and outside Lagos, Nigeria (Fig. 1). A total of eighteen representative samples were assayed (*cf*. Table 1) in duplicates. These samples were selected based on their popularity, availability and affordability. They were transported to the laboratory, where the samples were manually pulverized, in a porcelain crucible, and immediately oven-dried at 60 °C for 1 h. The dried samples were sieved (2 mm mesh) and stored in air-tight bags until analysis.

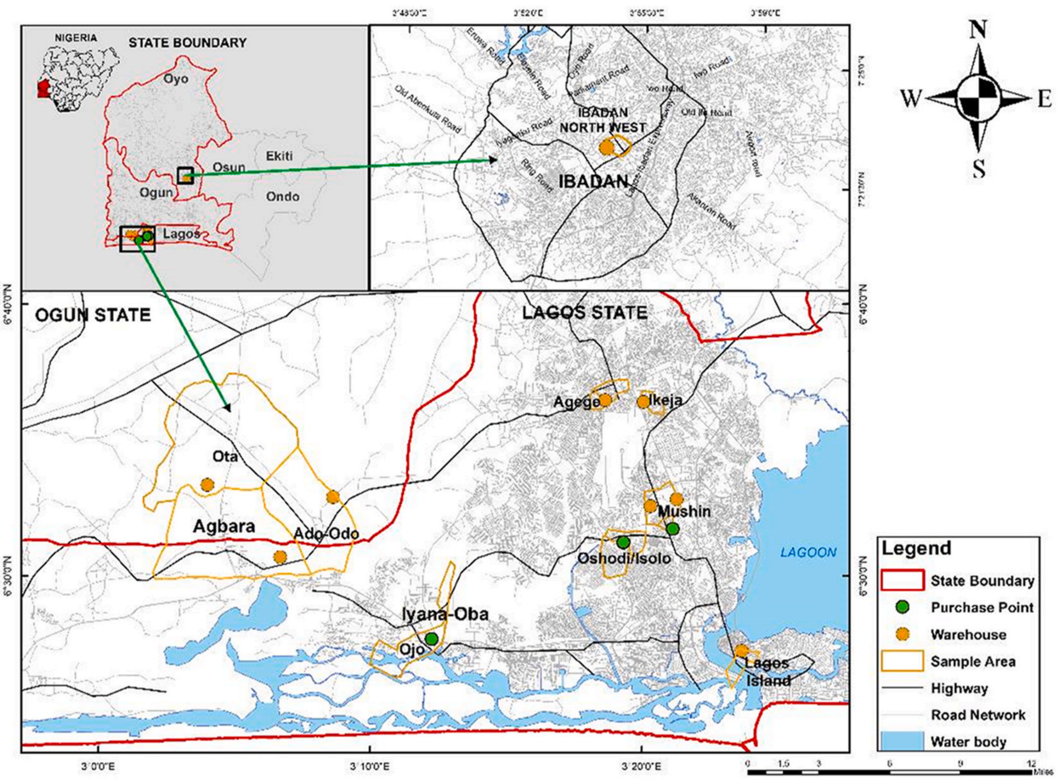

Fig. 1. Map of Southwestern Nigeria showing the study areas' sampling and storage sites.

Table 1
Types and sources of biscuits sampled.

S/N.	Biscuit Type	Sample Code	Warehouse/Manufacturer's Location	Purchase Point
1	**Crackers**	AA	Imported[a] (China)	Oshodi
		AB	Agbara (Ogun State)	Isolo
		AC	Lagos Island (Lagos State)	Iyana-Oba
2	**Cookies**	BA	Imported[a] (Denmark)	Iyana-Oba
		BB	Ota (Ogun State)	Ojo
		BC	Ibadan (Oyo State)	Mushin
3	**Shortcakes**	CA	Ibadan (Oyo State)	Mushin
		CB	Mushin (Lagos State)	Oshodi
		CC	Agege (Lagos State)	Mushin
4	**Digestives**	DA	Mushin (Lagos State)	Isolo
		DB	Ibadan (Oyo State)	Mushin
		DC	Lagos Island (Lagos State)	Ojo
5	**Cabins**	EA	Agege (Lagos State)	Mushin
		EB	Agbara (Ogun State)	Iyana-Oba
		EC	Imported[a] (China)	Mushin
6	**Wafers**	FA	Mushin (Lagos State)	Isolo
		FB	Imported[a] (Thailand)	Oshodi
		FC	Imported[a] (India)	Ojo

[a] Outside the study area.

2.4. Pseudo-total metal determination

The samples were wet-digested using a modified method [37,38] thus: a pulverized, dried and sieved sample (5.0 g) was weighed into a 250 mL beaker; into which a 15 mL solution of concentrated nitric acid (HNO_3), 30 % aqueous hydrogen peroxide (H_2O_2) and perchloric acid ($HClO_4$) in a 10:4:1 ratio had been added. The mixture-containing beaker was then covered with a wash glass and left standing overnight, at ambient temperature. After 22 h, its contents were gently heated to 60 °C, in a fume hood, until the brown effervescence ceased; leaving a colorless solution, which was allowed to cool to ambient temperature. Whence deionized water (20 mL) was added, filtered (125 mm Whatman) into a 50 mL volumetric flask and made up to mark with more deionized water.

Subsequently, the filtrate was transferred into labeled sample vials and analyzed for lead (Pb), cadmium (Cd), nickel (Ni), copper (Cu), cobalt (Co), zinc (Zn), iron (Fe), sodium (Na), magnesium (Mg) and calcium (Ca), using a flame atomic absorption spectrophotometer (FAAS), with an air–acetylene flame (Model 210VGP), under optimized operating conditions. Blanks were similarly analyzed.

2.5. Chemometrics

The data obtained were processed using multivariate statistical methods to reduce the variables to a smaller number of orthogonal factors [39]. The distribution patterns of the metals in the different biscuits were determined by analysis of variance (ANOVA), Tukey's honest significant difference (HSD) test, Pearson correlation, principal component analysis (PCA) and hierarchical cluster analysis (HCA). The varimax standardized rotation and score and loading plots as well as the dendrogram and heatmap strategies were utilized to perform PCA and HCA, respectively.

2.6. Health risk assessment

The data from metal analyses were used to determine the biscuits' estimated daily intake (EDI), target hazard quotients (THQ) and hazard indices (HI) for children and adults.

2.6.1. Estimated daily intake

The estimated daily intake (mg/kg body weight per day) of metals was calculated using equation (1) [40–42].

$$EDI = \frac{M_c \ x \ CR}{BW} \tag{1}$$

where M_c is the metal concentration in the biscuit, CR is consumption rate and BW is body weight.

The average consumption rates were 150 g/day and 50 g/day for children and adults, respectively, whereas the average body weights for children and adults were 40.9 kg [43,44] and 60.7 kg [45], respectively.

2.6.2. Target hazard quotient

A non-carcinogenic risk assessment method was used in the determination of THQ, using the ratio of the estimated contaminant dose to the reference dose [46], as expressed in equation (2) [47].

$$THQ = \frac{M_c \; x \; CR \; x \; EF \; x \; ED}{RfD \; x \; BW \; x \; AET} \; x \; 10^{-3}$$

where M_c is metal concentration, *CR* is consumption rate, *EF* is exposure frequency, *ED* is exposure duration, *RfD* is oral reference dose, *BW* is body weight and *AET* is the averaged exposure time for non-carcinogens while 10^{-3} is the unit conversion factor.

CR is 150 g/day (children) and 50 g/day (adults), *EF* is 365 days/year whereas *ED* is 61.3 years, which is the average life expectancy in Nigeria [48], and *AET* is 365 days multiplied by 61.3 years. The *RfD* (mg/kg/day) values for the metals are 0.0035 (Pb), 0.0001 (Cd), 0.02 (Ni), 0.04 (Cu), 0.0003 (Co), 0.3 (Zn), 0.7 (Fe), 34 (Na) and 11 (Mg) [49–51].

The limit of acceptance for the non-carcinogenic health risk of ingesting a particular element is one (THQ = 1) [52]. This implies that there is the potential for adverse non-carcinogenic health effects and, therefore, cause for bother when the non-carcinogenic health risk of ingesting a particular element is above the acceptable limit (i.e., THQ >1) whereas with THQ <1, the non-carcinogenic risk falls within the acceptance limit.

2.6.3. Total hazard index

The metal contents in the different biscuit types were calculated as the total hazard index (HI), according to equation (3), as delineated in equation (4) [40,53–55].

$$HI = \sum_{N=1}^{i} THQ_n \tag{3}$$

That is

$$HI = THQ_{Pb} + THQ_{Cd} + THQ_{Ni} + THQ_{Cu} + THQ_{Co} + THQ_{Zn} + THQ_{Fe} + THQ_{Na} + THQ_{Mg} \tag{4}$$

Therefore, HI is the total hazard index for all the metals under study (cf. eqn. (4); $n = 9$). The limit of acceptance for the non-carcinogenic health risk of ingesting all the metals present in the sample is one (i.e., HI = 1) [52,54]. This implies that there is cause for concern and a potential for adverse non-carcinogenic health effects when the non-carcinogenic health risk, of ingesting all the metals present, is above the acceptable limit (HI > 1) whereas when HI < 1, the non-carcinogenic risk falls within the acceptance limit. Non-carcinogenic risks are classified into negligible, low, medium and high risks, based on their HI values [49,54] (*cf.* Table 2).

2.6.4. Target cancer rate

The target cancer rate (TCR) was estimated using equation (5) [47,56].

$$TCR = \frac{M_c \; x \; CR \; x \; EF \; x \; ED \; x \; CSP_o}{BW \; x \; AET_c} \; x \; 10^{-3} \tag{5}$$

where M_c is metal concentration, *CR* is consumption rate (150 g/day; children, 50 g/day; adults), *EF* is exposure frequency (365 days/year), *ED* is exposure duration (61.3 years), CSP_o is the carcinogenic potency slope factor, *BW* is body weight and AET_c is the averaged exposure time for carcinogens while 10^{-3} is the unit conversion factor. The CSP_o values, $(mg/kg/day)^{-1}$, for Pb, Cd and Ni are 0.0085, 0.38 and 0.91, respectively [53,56,57].

2.7. Ecological risk assessment

The data from metal analyses were used to determine the potential ecological risks of the expired and/or egested biscuit wastes

Table 2
Classification of some parameters.

	Parameter	Range	Classification
(a)	**Hazard Index (HI)**	<0.1	Negligible
		≥0.1 < 1	Low
		≥1 < 4	Medium
		≥4	High
(b)	**Factor Loading**	0.3–0.5	Weak
		0.5–0.75	Moderate
		>0.75	Strong
(c)	**Potential Ecological Risk (RI)**	<40	Low
		40 ≤ RI < 80	Moderate
		80 ≤ RI < 160	Considerable
		160 ≤ RI < 320	High
		≥320	Very High
(d)	**Ecological Risk Index (ERI)**	<150	Low
		150 < RI < 300	Moderate
		300 < RI < 600	Considerable
		>600	Very High

[58–60].

The potential ecological risks (RI) and ecological risk indices (ERI) for the potentially toxic metals (PTM) in the biscuit wastes were calculated using equation (6) [61].

$$ERI = \sum RI = \sum T_i \, x \, PI = \sum T_i \, x \, \frac{C_s}{C_b} \tag{6}$$

where RI is the potential ecological risk factor of each PTM, T_i is the toxic-response factor of the PTM and PI is the pollution index. C_s is the concentration of PTM in the sample and C_b is the resultant background value. The toxic-response factors of some potentially toxic metals are given as: 5 (Pb), 30 (Cd), 5 (Ni), 5 (Cu), 5 (Co), 1 (Zn) and 1 (Fe) [62].

2.8. Data analysis

Statistical analysis was completed using Microsoft Excel and SPSS (version 19), using one-way analysis of variance (ANOVA). The Pearson correlation coefficient was adopted to assess the relationships amongst the metals in the biscuit samples. IBM SPSS statistic 20 was used to perform the multivariate statistical analysis whereas GraphPad Prism (v. 9.5) and PHeatmap R (v. 4.3.3) were used for the principal component and hierarchical cluster analyses, respectively. Significant differences were considered if $p < 0.05$.

2.9. Quality control

Glassware used were soaked in 2 M nitric acid for 12–24 h, washed with distilled water followed by deionized water prior to use. Analar grade reagents were used for analyses. The analyses were carried out in duplicates and with blanks. A certified reference material, BCR–191 (lyophilized brown bread), was also used to determine analytical performance. The CRM solutions used were prepared from the standard solutions with Milli-Q water and calibrations were accomplished by analyzing the prepared CRM standard solutions and blank samples. The blank samples were digested in the same way as the biscuit samples and the limits of detection (LOD) were calculated based on the blanks' standard deviations. Method accuracy was also validated by two replicate measurements.

3. Results

The quantification of the metal levels in different brands of commonly consumed biscuits in Lagos and Ibadan (Nigeria) and their environs is crucial because of their nutritional benefits as well as potential toxicity to ecology and environment [4,37]. Accordingly, the concentrations of select metals in six different types of biscuits were determined and the resulting data analyzed using chemometrics. The human and ecological health risk implications of the presence of heavy metals in the biscuits as well as their expired and/or egested wastes were equally assessed [58–60].

3.1. Determination of metals

Metal contents were analyzed in the certified reference material (CRM) and biscuits. The results of the certified reference material (CRM) for the pseudo-total concentrations, using a certified reference material of lyophilized brown bread (BCR-191) are shown in Table 3, with standard deviations of 0.1–0.3 of the certified values and <15 % precision for the pseudo-total digestion.

Table 4 shows the metal levels (mg/kg) detected in the different biscuit types post-wet digestion method by flame atomic absorption spectrophotometry (FAAS). Pb recorded the highest level of the potentially toxic metals (PTM) with 2.3 ± 1.3 mg/kg in the cracker biscuits. Conversely, Ca at 1174 ± 680 mg/kg, in the wafer biscuits, was the most abundant of the metals determined whereas nickel showed the lowest level, with 0.05 ± 0.05 mg/kg, in the shortcake biscuits. Notably, Cd was below the detection level of <0.02 mg/kg in any of the biscuit types investigated.

3.2. Chemometrics

In recent times, multivariate statistical analyses have been applied widely to examine environmental phenomena relating to food systems [63]. Multivariate statistical techniques are used to simplify and organize large data set to provide meaningful understanding. In the present study, six multivariate statistical techniques (mean, analysis of variance, Tukey's HSD test, Pearson correlation, factor

Table 3
Certified reference material analysis of CRM BCR–191.

Metal		Certified	Found[a]
Copper	Cu (mg/kg)	2.63 ± 0.07	2.97 ± 0.03
Iron	Fe (mg/kg)	40.70 ± 2.30	40.70 ± 2.80
Zinc	Zn (mg/kg)	19.50 ± 0.50	19.90 ± 1.80
Calcium	Ca (g/kg)	0.41	0.42 ± 0.02
Magnesium	Mg (g/kg)	0.50	0.52

[a] Mean values ± standard deviation for n = 3.

Table 4
Metal levels (mg/kg) in biscuits (wet digestion).

	Biscuit		Metal (mg/kg)									
			Lead	Cadmium	Nickel	Copper	Cobalt	Zinc	Iron	Sodium	Magnesium	Calcium
1.	**Crackers**	Mean	2.3 ± 1.3	<0.02	0.13 ± 0.12	1.37 ± 0.28	0.3 ± 0.2	6.0 ± 2.2	21.6 ± 8.7	710 ± 58	91.5 ± 3.6	368 ± 150
		Range	0.7–4.3		0.1–0.3	1.0–1.7	0.1–0.6	3.6–7.8	13.6–33.6	614–785	87–94	210–530
2.	**Cookies**	Mean	1.08 ± 1.02	<0.02	0.10 ± 0.15	1.30 ± 0.47	0.33 ± 0.12	13.0 ± 5.6	35.6 ± 7.0	563 ± 87	95.9 ± 2.7	477 ± 65
		Range	0.1–2.9		<0.02–0.4	0.7–2.0	0.2–0.5	5.8–18.3	28.6–47.7	468–670	92–100	510–580
3.	**Shortcakes**	Mean	0.83 ± 0.76	<0.02	0.05 ± 0.05	1.10 ± 0.32	0.5 ± 0.22	4.4 ± 0.61	22.4 ± 7.4	530 ± 29	92.2 ± 5.5	418 ± 150
		Range	0.2–1.9		<0.02–0.1	0.7–1.6	0.3–0.9	3.2–4.8	13.6–30.6	500–581	83–98	320–420
4.	**Digestives**	Mean	1.80 ± 0.65	<0.02	0.18 ± 0.23	1.37 ± 0.26	0.45 ± 0.43	5.23 ± 1.7	21.6 ± 6.1	580 ± 120	84.5 ± 10.6	187 ± 75
		Range	0.9–2.8		0.1–0.6	1.0–1.7	0.1–1.1	2.9–7.2	15.6–32.6	435–720	76–104	110–210
5.	**Cabins**	Mean	1.35 ± 0.49	<0.02	0.10 ± 0.10	1.00 ± 0.30	0.25 ± 0.16	3.9 ± 1.3	27.8 ± 4.0	475 ± 47	96.8 ± 5.8	643 ± 220
		Range	0.8–2.0		<0.02–0.3	0.8–1.6	0.1–0.5	2.0–5.6	21.6–32.6	427–549	86–103	430–790
6.	**Wafers**	Mean	1.57 ± 0.52	<0.02	0.25 ± 0.10	0.62 ± 0.16	0.22 ± 0.24	3.43 ± 0.28	13.8 ± 1.3	470 ± 210	92.1 ± 4.5	1174 ± 680
		Range	1.0–2.4		0.1–0.4	0.5–0.9	<0.06–0.64	3.1–3.8	12.6–15.0	224–736	86–100	340–1720

analysis/principal component analysis and hierarchical cluster analysis) were applied to evaluate the concentration of metals in the biscuit types analyzed.

Table S1 (supplementary data) shows the one-way analysis of variance (ANOVA) test of mean differences for the metals found in the biscuits. Ni (0.65) and Co (0.45) both furnished F-ratios less than one whereas the other metals ranged from 1.47 (Fe) to Ca (7.52). In contrast, Na, Mg and Cu recorded P-level values of 0.007, 0.021 and 0.001, respectively, at a confidence level of 0.05 while Zn and Ca gave zero.

The Tukey's honest significant difference (HSD) test was also invoked to determine the statistical significance of the differences between pairs of group means of the six biscuit types. The results are collated in Tables S2–S10 (supplementary data), showing significant mean differences in the Pb, Ni, Co and Fe contents between all the six different types of biscuits analyzed.

Conversely, Pearson correlation coefficients at 95 % (0.05) confidence level for the metal analyses is displayed in Table S11, showing non-correlation between the metals determined; except between Co and Na.

The data was subjected to a four-component factor analysis (principal component) to identify the distribution of the metals in the biscuits analyzed and presented in Fig. 2 as score and loading plots. Notably, the first four principal components resolved 69.08 % of the total data variance, with eigenvalues >1. The score plots (**a**, **c** and **e**) show the sample groupings based on the compositional similarities of the biscuit types while the loading plots (**b**, **d** and **f**) display variable correlations of the metals. The plots of principal component 1 (PC1) against each of principal components 2 (PC2), 3 (PC3) and 4 (PC4) are designated as **(a)** and **(b)**, **(c)** and **(d)**, and **(e)** and **(f)**, respectively.

In the score plots, samples of crackers, cookies and digestives were mainly gathered in the positive region of the PC1 axis while the cabin and wafer biscuit samples were majorly found in the negative region of the PC1 axis. It is notable that in the PC1 v PC2 plot, all samples of cookies occupied the positive quadrant in contrast to the PC1 v PC3 and PC1 v PC4 plots. The loading plots, on the other hand, showed Ni, Fe, Zn and Na in the positive quadrant of the PC1 v PC2 graph whereas the PC1 v PC3 and PC1 v PC4 graphs both had Fe, Co, Cu and Zn, and Ca in their positive and negative quadrants, respectively. Interestingly, Pb was observed on the negative *y* axis (PC1 = 0) in all three loading plots: **(b)**, **(d)** and **(f)** (*cf*. Fig. 2).

The result of the hierarchical cluster analysis (HCA), using Ward's linkage, is illustrated in Fig. 3; showing three clusters. Most of the metals: Ni, Co, Cu, Pb, Zn, Fe and Mg are collected in the first cluster (cluster 1) whereas Fe and Mg, which overlap from cluster 1, are also found in cluster 2 with Na. Cluster 3, in contrast, is occupied by Ca and Na; which sits at the boundary of clusters 2 and 3. This is corroborated by the results of the two-dimensional hierarchical clustering heatmap, shown in Fig. 4, which exhibited similar clusters for the metals. Fig. 4 also highlights the various clusters and subclusters inherent in the different biscuit samples assayed. The red and blue colors indicate the highest and lowest levels of metals, respectively, found in the biscuit types. Notably, Na and Ca exhibited the highest intensities of 1 while Ni, Co, Pb and Cu were close to zero intensity (blue). It can also be observed that the samples of the digestives (DA1, DA2, DB1, DB2, DC1 & DC2) and crackers (AB1, AB2, AC1 & AC2), and cookies (BA2, BB1, BB2 & BC1), respectively, predominated the two red Na clusters of biscuits whereas the only red Ca cluster was predominated by the wafers (FA1, FB1, FB2, FC1 & FC2). All the biscuit types also showed low (close to zero) intensities for Ni, Co, Pb and Cu (blue cluster; Fig. 4).

3.3. Health risk assessment

Contained in Table 5, are the data for total detected metal contents and some of the health risk assessment results of the six biscuit

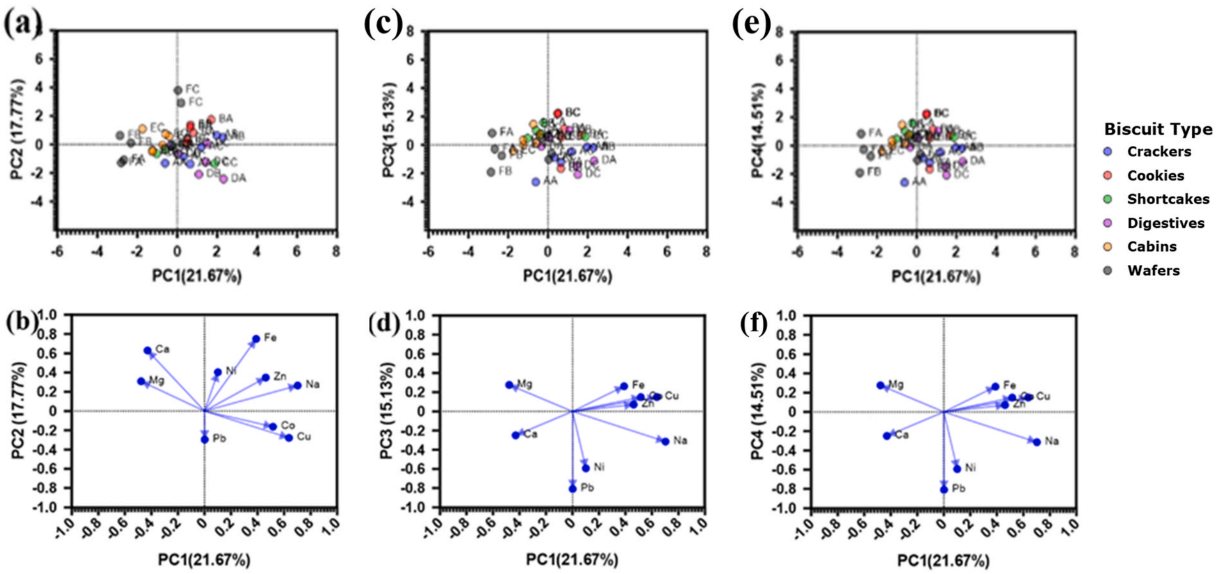

Fig. 2. Principal component analysis (PCA) showing score plots: **(a)**, **(c)** and **(e)**, and loading plots: **(b)**, **(d)** and **(f)**.

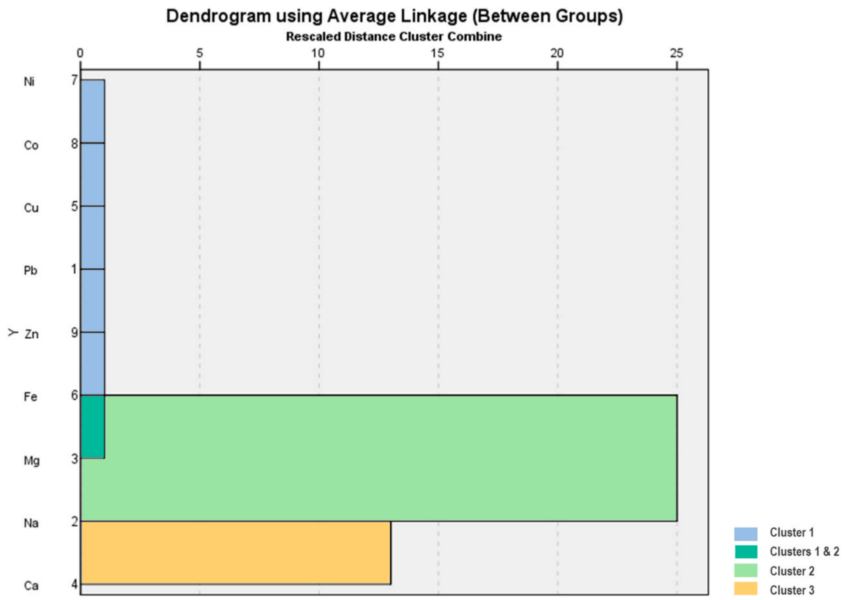

Fig. 3. Hierarchical cluster analysis (HCA) using Ward's linkage.

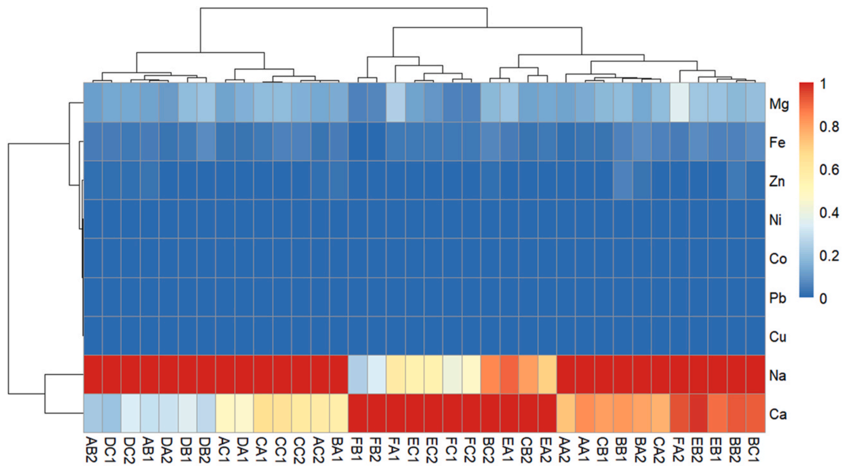

Fig. 4. Two-dimensional hierarchical clustering heatmap using Pheatmap in R.

types analyzed. Na and Ca were the most abundant of the metals determined at concentrations of 556 ± 130 mg/kg and 545 ± 420 mg/kg, respectively, whereas Ni and Co gave the lowest concentrations of 0.12 ± 0.15 mg/kg and 0.33 ± 0.33 mg/kg, respectively. Cu (1.13 ± 0.40), Pb (1.49 ± 0.92), Zn (6.44 ± 5.60), Fe (25.8 ± 11.3) and Mg (92.2 ± 6.9) were also detected but Cd was below detection level in all the biscuit types assessed.

Conversely, Table 6 compares the metal limits per week, stipulated by the Joint FAO/WHO Expert Committee on Food Additives (JECFA) [64,65], for children and adults of body weights of 40.9 kg and 60.7 kg, respectively [43–45], to the metal contents of the six types of biscuits (crackers, cookies, shortcakes, digestives, cabins and wafers) examined in this study. The wafers gave the highest levels for Cu (230.9; 342.7), Co (130.1; 193.1), Zn (83.5; 123.9), Fe (16.6; 24.6) and Na (1522.9; 2260.1), for children (4–20 years) and adults, respectively, whereas digestives and shortcakes recorded the highest levels for Mg (1219.7; 1810.2) and Ca (1990.3; 2953.9), and Pb (1.23; 1.83) and Ni (28.6; 42.5), respectively, for both 4–20-year-old children and adults. It is noteworthy that the following biscuits gave the lowest values for the respective metals: crackers (Pb, Cu & Na), cookies (Zn & Fe), shortcakes (Co), digestives (Cu), cabins (Mg) and wafers (Ni & Ca), for children and adults alike.

The results from the calculation of the target cancer rates (TCR) of the carcinogenic metals, using equation (5), are collected in Table 7. For children, the TCR of Pb was highest in the digestives (6.6×10^{-3}) and lowest in the crackers (7.17×10^{-5}) but Ni gave 9.17×10^{-4} (wafers) and 1.83×10^{-4} (shortcakes) as its highest and lowest values, respectively. Pb gave 1.89×10^{-3} (crackers) and 1.29×10^{-4} (wafers) as its highest and lowest values, respectively, for adults whereas Ni recorded the highest (2.06×10^{-4}) and lowest

Table 5
Health risk assessment parameters of biscuits.

		Metal										
		Pb	Cd	Ni	Cu	Co	Zn	Fe	Na	Mg	Ca	HI
Concentration[a] (mg/kg)		1.49 ± 0.92	BDL	0.12 ± 0.15	1.13 ± 0.40	0.33 ± 0.33	6.44 ± 5.60	25.8 ± 11.3	556 ± 133	92.2 ± 6.9	545 ± 425	
Children	THQ	1.56	ND	0.022	0.103	4.0410	0.072	0.13514	0.0599	0.0307	–	6.0238
	EDI (mg/kg BW/day)	0.00545	ND	0.00045	0.00412	0.00121	0.0236	0.09460	2.0373	0.338	1.997	
	PTWI (mg/kg BW)	0.025	0.007	0.035	3.5	0.7	7	5.6	17,500	2520	9100	
Adult	THQ	0.350	ND	0.0050	0.0232	0.90762	0.01607	0.03035	0.01346	0.00690	–	1.3526
	EDI (mg/kg BW/day)	0.00123	ND	0.0001	0.00093	0.00027	0.00530	0.02125	0.45759	0.07592	0.44863	
	PTWI (mg/kg BW)	0.025	0.007	0.035	3.5	0.7	7	5.6	17,500	2240	7000	
R*f*D[b]	(mg/kg/day)	0.0035[c]	0.0001	0.02	0.04	0.0003	0.3	0.7	34[d]	11[d]	–	

[a] Concentration of metal in biscuits.
[b] Ref. [50].
[c] Ref. [49].
[d] Ref. [51]; THQ: target hazard quotient, EDI: estimated daily intake, BW: body weight, PTWI: provisional tolerable weekly intake, R*f*D: oral reference dose, HI: hazard index, BDL: below detectable level, ND: not determined.

Table 6
Consumable quantities of biscuits using the Joint FAO/WHO Expert Committee on Food Additives (JECFA) limits [64,65].

	Biscuit		Metal									
			Pb	Cd	Ni	Cu	Co	Zn	Fe	Na	Mg	Ca
Children		(mg/week/kg BW)[a]	0.025	0.007	0.035	3.5	0.7	7	5.6	17,500	2520	9100
		(mg/week/40.9 kg)	1.005	0.281	1.407	140.7	28.14	281.4	225.12	703,500	101,304	365,820
	Crackers	(kg)	0.44	NA	11.01	104.5	95.4	47.7	10.6	1008.1	1126.4	1011.4
	Cookies	(kg)	0.95	NA	14.32	110.1	86.8	22.0	6.4	1271.3	1074.7	780.3
	Shortcakes	(kg)	1.23	NA	28.63	130.1	57.3	65.1	10.2	1350.5	1117.9	890.4
	Digestives	(kg)	0.57	NA	7.95	104.5	63.6	54.7	10.6	1234.1	1219.7	1990.3
	Cabins	(kg)	0.76	NA	14.32	143.2	114.5	73.4	8.2	1506.8	1064.8	578.8
	Wafers	(kg)	0.65	NA	5.73	230.9	130.1	83.5	16.6	1522.9	1119.1	317.0
Adult		(mg/week/kg BW)[a]	0.025	0.007	0.035	3.5	0.7	7	5.6	17,500	2240	7000
		(mg/week/60.7 kg)	1.518	0.424	2.123	212.5	42.5	427.9	339.9	1,060,000	150,000	550,000
	Crackers	(kg)	0.66	NA	16.3	155.1	141.6	70.8	15.7	1496.1	1671.7	1501.0
	Cookies	(kg)	1.41	NA	21.2	163.4	128.8	32.7	9.5	1886.8	1595.0	1158.0
	Shortcakes	(kg)	1.83	NA	42.5	193.1	85.0	96.6	15.2	2004.2	1659.0	1321.5
	Digestives	(kg)	0.84	NA	11.8	155.1	94.4	81.2	15.7	1831.5	1810.2	2953.9
	Cabins	(kg)	1.12	NA	21.2	212.5	170.0	108.9	12.2	2236.3	1580.2	859.1
	Wafers	(kg)	0.97	NA	8.5	342.7	193.1	123.9	24.6	2260.1	1660.8	470.5

[a] The quantity of biscuit (in kg) to be consumed per week per body weights to reach the JECFA limit, FAO: Food and Agriculture Organization, WHO: World Health Organization, NA: not applicable.

Table 7
Target cancer rate (TCR) of carcinogenic metals.

Biscuit	Target cancer rate (TCR)					
	Children			Adult		
	Lead	Cadmium	Nickel	Lead	Cadmium	Nickel
Crackers	7.17×10^{-5}	NA	4.77×10^{-4}	1.89×10^{-3}	NA	1.07×10^{-4}
Cookies	3.96×10^{-3}	NA	3.67×10^{-4}	8.9×10^{-4}	NA	8.24×10^{-5}
Shortcakes	3.04×10^{-3}	NA	1.83×10^{-4}	6.84×10^{-4}	NA	4.12×10^{-5}
Digestives	6.60×10^{-3}	NA	6.60×10^{-4}	1.48×10^{-3}	NA	1.48×10^{-4}
Cabins	4.95×10^{-3}	NA	3.67×10^{-4}	1.11×10^{-3}	NA	8.24×10^{-5}
Wafers	5.76×10^{-3}	NA	9.17×10^{-4}	1.29×10^{-3}	NA	2.06×10^{-4}

*United States Environmental Protection Agency (USEPA)'s maximum threshold level [56] ≥ 1 × 10−4.

(4.12×10^{-5}) rates in the wafer and shortcake biscuits, respectively.

3.4. Ecological risk assessment

Ecological risk assessments evaluate the likelihood of adverse ecological damages occurring to ecosystems exposed to one or more chemical, physical and/or biological stressors [66,67]. Subsequently, the process is used to systematically evaluate and organize data as well as assumptions, uncertainties and information towards understanding and predicting the relationships between stressors and ecological effects in order to enable sustainable environmental decision-making, amongst others [66]. Notably, wastes from foods comprise significant portions of waste streams in industrialized countries and can, therefore, contribute to ecological damages as well as nutritional losses [52,68].

Herein, the probability of wastes from potentially toxic metals-contaminated biscuits causing adverse ecological damages is assessed. The potential ecological risks (RI) of six metals (Pb, Ni, Cu, Co, Zn & Fe) were calculated and collated in Table 8 before

Table 8
Potential ecological risks and ecological risk indices.

Biscuit	Potential ecological risks (RI)							ERI[a]
	Lead	**Cadmium**	**Nickel**	**Copper**	**Cobalt**	**Zinc**	**Iron**	**(∑RI)**
Crackers	5.750	ND	0.130	0.685	0.429	0.120	1.080	8.194
Cookies	2.700	ND	0.100	0.650	0.471	0.260	1.780	5.961
Shortcakes	2.075	ND	0.050	0.550	0.714	0.088	1.120	4.597
Digestives	4.500	ND	0.180	0.685	0.643	0.105	1.080	7.192
Cabins	3.375	ND	0.100	0.500	0.357	0.078	1.390	5.800
Wafers	3.925	ND	0.250	0.310	0.314	0.069	0.690	5.558
Mean	3.721	–	0.135	0.563	0.488	0.120	1.190	6.217

[a] ERI: Ecological risk index, ND: not determined.

determining the ecological risk indices (ERI) for the biscuit samples. The crackers gave the highest RI value for Pb at 5.75 and ERI of 8.194 whereas the lowest RI and ERI values of 0.05 and 4.597, respectively, were recorded for Ni in shortcakes. Comparatively, the Pb and Fe contents of the biscuits were high, with average RI values of 3.72 and 1.19, respectively, while Ni (0.135) and Zn (0.12) gave low values; with Cu (0.563) and Co (0.488) values in between.

4. Discussion

The consequences of heavy-metals' contamination of biscuits to health and the environment are increasingly becoming tangible, world-over, due to the progressive rise in daily biscuit consumption and production [38,69–72]. Herein, the quantification of some potentially toxic metals in different brands of commonly consumed biscuits in the southwestern Nigerian metropolises of Lagos and Ibadan (cf. Fig. 1) was embarked upon to evaluate the affected media, with a view to highlighting, mitigating and/or eliminating as well as monitoring their associated harmful effects. It is notable that with an estimated 2024 combined population of about 20.5 million [27,32], the conurbations of Lagos and Ibadan are representative.

Six biscuit types: crackers, cookies, shortcakes, digestives, cabins and wafers, were purchased from different locations in Lagos and Ibadan, based on their availability, affordability and popularity, and assessed. Cognizance was also taken of the locations of the biscuits' manufacturers' plants/warehouses as PTM contaminants can be introduced into biscuits from raw materials and processing or packaging lines [12]. Most of the biscuits evaluated were manufactured in the study areas but representative samples made in China, Denmark, India and Thailand were also included (*cf.* Table 1).

The samples were successfully digested as described; with <15 % precision for the pseudo-total digestion for all the metals assessed. Also, the result of the certified reference material for the pseudo-total concentrations, analyzed using BCR–191, were generally within a 0.1–0.3 standard deviation of the certified values (*cf.* Table 3).

The post-wet digestion concentrations in dry weight of metals in crackers, cookies, shortcakes, digestives, cabins and wafers, as collated in Table 4, show Ca; found in wafers, as the most abundant metal determined with 1174 $\pm$ 680 mg/kg while the lowest metal level was found in shortcakes (Ni; 0.05 $\pm$ 0.05 mg/kg). More significantly, the highest level of potentially toxic metals (PTM) was recorded by Pb (2.3 $\pm$ 1.3 mg/kg; crackers) whereas Cd was not detected (<0.02 mg/kg) in any of the biscuit types investigated. It is worthy of note that only the shortcakes (0.83 $\pm$ 0.76 mg/kg) and cookies (1.08 $\pm$ 1.02 mg/kg) gave (lower boundary) values proximal to the European Commission's (EC) Pb limit of 0.2 mg/kg, for foods (cereals and pulses) [73,74]. Incidentally, Iwegbue's assessment, based on a varied procedure, yielded lower values (0.54 $\pm$ 0.92 mg/kg) for Pb but higher readings (0.05 $\pm$ 0.02 mg/kg) for Cd [38], in comparison to the samples herein. A similar scenario was reported by Arnich et al. [75] from the second French national total diet study. The varying metal levels detected in these studies could be, plausibly, ascribed to the sources of raw materials used and the processing regime. Conversely, the >100 % dry weight of Ca, found in the wafers; relative to the other biscuit types, may be connected to their production process. It is noteworthy that only the wafer biscuits furnished a value higher than the World Health Organization (WHO) recommended daily intake (RDI) of 1000 mg Ca per day for young adults [76].

It is pertinent to note that the highest levels of Cu (1.37 $\pm$ 0.28 mg/kg; crackers), Co (0.5 $\pm$ 0.22 mg/kg; shortcakes), Zn (13.0 $\pm$ 5.6 mg/kg; cookies) and Fe (35.6 $\pm$ 7.0 mg/kg; cookies) detected (*cf.* Table 4) were lower than the EC recommended limits of 5.0 mg/kg [77], 0.6 mg/kg [78], 25 mg/kg [79], and 48 mg/kg [80], respectively. The six different biscuit types assessed in this study can be, therefore, considered to be free of Cu, Co, Zn and Fe contaminants. Also noteworthy is that these metals are essential micronutrients, which deficiencies and/or excessive exposure to humankind can cause adverse health and environmental effects [81]. Cu is a co-factor for enzymes and possesses antioxidant properties. It is essential for metabolic and biochemical processes, such as hemoglobin synthesis and bone development, and can prevent cell structure damage [82]. Deficiency of Cu can lead to anemia, cardiovascular diseases and colon cancer whereas Cu poisoning can cause diarrhea, nausea, liver and kidney damage in man [83]. Fe, on the other hand, is an essential component of hemoglobin and myoglobin, and participates in many functions in the body, including oxygen and electron transfer, cell division and differentiation, immunity and energy metabolism as well as hormone synthesis and neurological development [84]. In addition, Fe deficiency is implicated in anemia, chronic bleeding, severe fatigue, eye defect, hair loss and irritability while long-term exposure to higher Fe concentrations could also result in nausea, diarrhea and hepatic failure [85].

Unlike Fe, which is primarily found in the blood, Zn is present in all human tissue and fluids; in the muscles, reproductive tract, hair and bones, for examples, at varying concentrations [86]. Besides, cause-and-effect relationships have been established between the dietary intake of Zn and its essential structural, regulatory and catalytic roles in enzymes and non-enzymatic proteins, and hormones [87]. It also facilitates DNA synthesis and cell division, and protects proteins, lipids and DNA from oxidative damage as well as maintains normal bone and vision, reproductive, metabolic and cognitive functions [86,87]. Zn deficiency symptoms include depressed immunities, retarded growth, skin lesions, skeletal anomalies and impaired reproduction as well as behavioral abnormalities, such as mood swings, smell and taste dysfunctions, irritability, depression and anorexia [87,88]. Similarly, Co is an essential metal with ubiquitous dietary and incremental exposures; from food supplements, occupation and medical devices [89]. It is a constituent of cobalamin, an important cofactor in vitamin B_{12}-dependent enzymes and can induce neurological, reproductive, immunological and endocrine responses as well as DNA damage, cardiomyopathy, polycythemia and bronchial asthma [90,91]. Co is also essential for nitrogen fixation by free-living bacteria, blue-green algae and symbiotic systems [90].

Nickel is a heavy metal and satisfies many of the criteria for the essentiality of trace elements to be categorized as an essential micronutrient [92]. It is ubiquitous in human nutrition and the environment, and even at an orally ingestible maximum of 0.5 mg/day has low toxicity because it is poorly absorbed and rapidly cleared from serum [93,94]. Evidently, Ni deficiencies are rare but can be lethal where they occur; just as oral exposures can affect the gastrointestinal, hematological, neurological or immunological systems; with acute exposures being primarily associated with gastrointestinal and neurological symptoms [94–96]. Recent human studies, however, suggest an association between Ni exposure and adverse reproductive and developmental outcomes [97]. There is no EC recommended maximum level for Ni in foods but a parametric value of 0.02 mg/L, which is within the WHO guideline value of 0.07 mg/L [94], has been recommended for portable water [95,97], where soluble Ni is more easily absorbed than food [93–95]. Interestingly, Ni was the least detected heavy metal of the elements assessed herein with the wafer and shortcake biscuits recording the highest and lowest mean values of 0.25 ± 0.10 mg/kg and 0.05 ± 0.05 mg/kg, respectively. Although Ni is prevalent in Nature, the quantities found in biscuits could be linked to the storage/production vessels, raw materials, such as cocoa additives, and/or hydrogenation catalysts [38].

Na and Mg macrominerals were also detected (Table 4) between 470 ± 210 mg/kg (wafers) and 710 ± 58 mg/kg (crackers), and 84.5 ± 10.6 mg/kg (digestives) and 96.8 ± 5.8 mg/kg (cabins), respectively; below the recommended dietary allowances (RDA) of 2000 mg/day (Na) [98] and 310 mg/day (Mg) [99], respectively. Instructively, Na is the principal cation in human extracellular fluid and it is essential for electrolyte/fluid balances and blood-pressure maintenance. Elevated sodium intake has been linked with a number of non-communicable diseases, such as hypertension and stroke as well as cardiovascular diseases whereas decreasing sodium intake may reduce blood pressure and associated risks [98]. Relatedly, Mg is also involved in the regulation of blood pressure and acts as a cofactor in about 350 enzymatic reactions in the body [100]. It is essential in the intermediary metabolism for the syntheses of proteins, carbohydrates, lipids and nucleic acids, and the active transport of calcium and potassium ions across cell membranes [101]. Notably, Mg deficiency can cause hypocalcemia and hypokalemia, leading to neurological or cardiac symptoms when associated with marked hypomagnesaemia [99–101]. Most of the body's Mg is stored in bones (60 %) and muscles (25 %) [99].

Besides, almost 99 % of the body's Ca stores are found in bones and teeth where it plays crucial roles in bone structure and muscle contractions as well as nerve impulse transmissions and glandular secretions [101,102]. Consequently, Ca deficiencies can cause rickets and osteoporosis, with associated increases in fracture risks, in children and adults, respectively [100]. Inadequate Ca intake has also been linked to increased risks of nephrolithiasis (kidney stones), colorectal cancer, hypertension and stroke as well as coronary artery diseases, insulin resistance and obesity [100,101]. A cause-and-effect relationship has also been established between Ca and the maintenance of normal digestive enzymes' functions, blood coagulation and energy-yielding metabolisms [102]. Detectable Ca in the biscuits herein ranged from 187 ± 75 mg/kg (digestives) to 1174 ± 680 mg/kg (wafers; Table 4); below the WHO's RDA of 1300 mg/day for children (9–18 years), post-menopausal women and over-65-year-old men [76,100].

A corollary of the metals' assays shows that the biscuits sampled had high concentrations of the macrominerals in the wafers (Ca), crackers (Na) and cabins (Mg) whereas the micronutrients (Fe, Zn, Cu, Co & Ni) were highest in the cookies (Fe, Zn), crackers (Cu), shortcake (Co) and wafers (Ni), respectively, and were all safe for consumption. In contrast, none of the six types of biscuits investigated recorded values below the EC's set maximum levels for Pb [74,103]; with the lowest Pb content detected in the shortcake brands at 0.83 ± 0.76 mg/kg (*cf.* Table 4). Pb remains a toxic metal of great public health concern as it has been reported to induce developmental neurotoxicity in young children as well as cardiovascular effects and nephrotoxicity in adults [73]. It is carcinogenic and can lead to adverse musculoskeletal, immunological and reproductive problems [104]. The Pb levels detected in the biscuits sampled herein are worrying and should because for alarm to the requisite regulatory authorities. It is also significant to note that <0.2 mg/kg Pb have been reported in similar studies in Ibadan [69] and Lagos [70] but higher (>0.2 mg/kg) in some other parts of Nigeria [38,71,72]. These indicate an increase in the Pb content of biscuits in Ibadan and Lagos metropolises, in the last decade, in comparison to the results obtained herein and are congruous with recent findings that Pb sources in foods are majorly anthropogenic [105]. Corroboratively, somewhat, the Pb levels recorded in biscuits in Asia [4] and Europe [106] are within the EC Pb limit of 0.2 mg/kg [74] although no level of Pb exposure is known to be without harmful effects [107].

The concentrations (mg/kg) of the nine metals determined from the six different biscuit types, in this study, are grouped into low, medium and high, and graphically represented in Fig. 5. Pb, Co and Ni are in the "low" category with concentrations <2.5 mg/kg (Fig. 5(a)) and followed by the micronutrients (Fe, Zn and Cu); with Fe recording the highest concentration of 36 mg/kg in the cookies (*cf.* Fig. 5(b)). The macrominerals (Ca, Na and Mg; Fig. 5(c)) fell into the third ("high") category with concentrations ($40 < x < 1200$). It is interesting to note that the six biscuit types showed congruous concentrations (92.2 ± 4.4 mg/kg) of Mg (brown color; Fig. 5(c)), which is below the RDA (Mg) of 310 mg/day [99]. Furthermore, Fig. 5(a) accentuates the Pb levels in the biscuits relative to those of Co and Ni, and leaves great cause for worry [107].

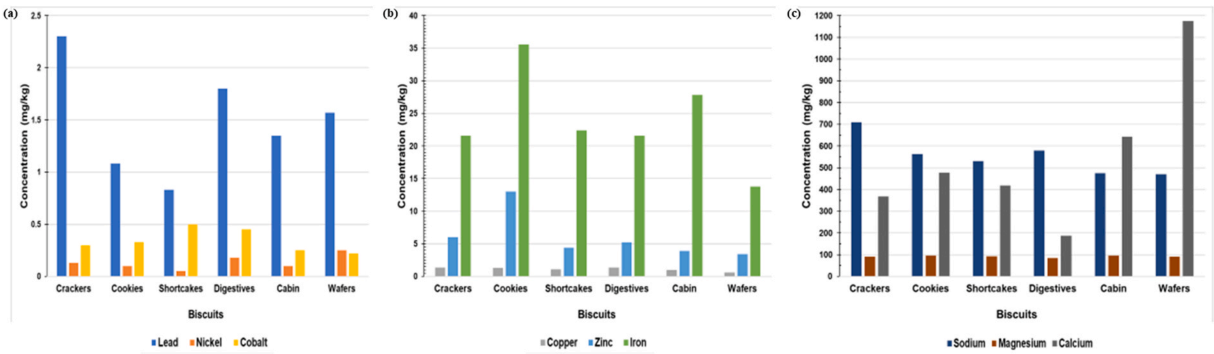

Fig. 5. Different metal concentrations found in the biscuits: (a) low; <2.5 mg/kg, (b) medium; <40 mg/kg, and (c) high; <1200 mg/kg.

Furthermore, the variations in the concentrations of metals found in biscuits globally are highlighted in Table 9. It is striking that the Pb levels observed in the present study were higher than those reported elsewhere; except for the works of Taha et al. [109] in Egypt (0.828 mg/kg) and Arigbede et al. [71] in southwestern Nigeria (3.11–92.0 mg/kg), respectively, whereas this study recorded the lowest metal concentrations for Ni; with the highest value of 5.70–9.85 mg/kg recorded in India [37]. The results obtained herein for the metals assayed were varied, within and without Nigeria, in comparison to previous works [108–115] (*cf.* Table 9) but comparable in some instances: as illustrated by results from Egypt [108,109] for Cu and Turkey [111] for Zn. In the case of the macrominerals, the lowest concentrations of 224–785 mg/kg (Na) and 76.0–104 mg/kg (Mg), and highest concentration of 110–1720 mg/kg (Ca), respectively, were observed in the present study.

The chemometric techniques employed to evaluate the data generated in this study yielded varying results. The one-way ANOVA test of mean differences for the metals in the biscuits, collected in Table S1, showed significant differences, at a confidence level of 0.05, with values (P-level) of 0.001, 0.007 and 0.021 for Cu, Na and Mg, respectively, and zero for Zn and Ca. On the other hand, the mean differences, determined using the Tukey's honest significant difference (HSD) test, were also used to identify metals that showed significant differences in the six biscuit types analyzed. Interestingly, significant mean differences were observed between pairs of all the biscuit types analyzed for Pb, Ni, Co and Fe whereas some of the biscuit pairs showed mean differences of <0.05 significance; ranging from zero to 0.029, for Cu, Zn, Na, Mg and Ca (Table 10). For instance, zero significance was recorded with Zn and Ca for the biscuit pairs of cookies–wafers and digestives–wafers, respectively, while Mg gave a 0.029 significance value for the cookies–digestives pair. It is noteworthy that both the one-way ANOVA and Tukey's HSD tests gave zero values for Zn and Ca, and are also in concurrence regarding the non-significance (<0.05) of the mean differences of Cu, Na and Mg in the biscuits analyzed whereas the mean differences of Pb, Ni, Co and Fe were significant.

The Pearson's correlation matrix, which was effectively used to display the inter-relationships between the metals assayed in the biscuits, at a 95 % confidence level, is contained in Table S11 (supplementary data). Non-correlations were observed between the metals determined except for the pair of Na and Co, which gave a significant positive correlation value of 0.389.

The data obtained for the metal analysis was also subjected to factor analysis to identify the distribution of the metals in the biscuits analyzed. The principal component analysis (PCA) was modeled on the first four components, which explained 69.1 % of the total variance; depicted as score and loading plots of the biscuits and metals, respectively, in Fig. 2. The first to fourth components

Table 9
Metal levels (mg/kg) reported in biscuits from different countries.

Metal	Concentration (mg/kg)	Country	Ref.	Metal	Concentration (mg/kg)	Country	Ref.
Lead	ND–0.13	India	37	Zinc	ND–13.4	India	37
	<0.001–1.90	Nigeria	38		7.9–73.3	Nigeria	38
	ND–0.08	Nigeria	69		5.64–157	Nigeria	71
	3.11–92.0	Nigeria	71		2.35–4.75	Egypt	108
	0.126–0.127	Egypt	108		2.12–14.89	Egypt	109
	0.828	Egypt	109		1.65–14.80	Bangladesh	110
	BDL–1.21	Bangladesh	110		3.1–16.1	Turkey	111
	0.10–4.30	Nigeria	PS		2.0–18.3	Nigeria	PS
Cadmium	0.02–0.05	Nigeria	38	Iron	ND–36.3	India	37
	0.003–0.09	Nigeria	69		3.50–364.2	Nigeria	38
	0.013–0.122	Egypt	108		1.02–2.07	Nigeria	69
	0.096	Egypt	109		99.4–296	Nigeria	71
	BDL–0.47	Bangladesh	110		27.14	Egypt	109
	<0.02	Nigeria	PS		8.9–33.47	Bangladesh	110
					6.9–35.4	Turkey	111
Nickel	5.70–9.85	India	37		50–86	Brazil	112
	0.5–6.5	Nigeria	38		12.6–33.6	Nigeria	PS
	1.462	Egypt	109				
	BDL	Bangladesh	110	Sodium	4173	Egypt	109
	<0.02–0.6	Nigeria	PS		0–17000	UK	113
					140–9350	China	114
Copper	<0.001–7.5	Nigeria	38		3910	Latin America/Caribbean	115
	0.08–0.51	Nigeria	69				
	ND	Nigeria	71		224–785	Nigeria	PS
	0.79–1.39	Egypt	108				
	1.815	Egypt	109	Magnesium	114.3–128.4	Nigeria	38
	0.35–2.55	Bangladesh	110		328.2	Egypt	109
	<1–42	Turkey	111		76.35–165.59	Bangladesh	110
	0.50–2.0	Nigeria	PS		76.0–104	Nigeria	PS
Cobalt	0.8–1.85	India	37	Calcium	87.3–696.0	Nigeria	38
	<0.001–2.84	Nigeria	38		301.1	Egypt	109
	0.202	Egypt	109		19.58–138.06	Bangladesh	110
	BDL	Bangladesh	110		110–1720	Nigeria	PS
	<0.06–1.1	Nigeria	PS				

Ref.: reference, ND: not determined, BDL: below detectable level, PS: present study.

Table 10
Tukey's HSD test showing non-significant mean-difference data of metals analyzed in the biscuits.

Dependent Variables			Mean Difference	Standard Error	Significance[b] (>0.05)	95 % Confidence Interval	
Metal[a]	Biscuits					Lower Bound	Upper Bound
Copper	Crackers	Wafers	0.75000	0.17997	0.003	0.2026	1.2974
	Cookies	Wafers	0.68333	0.17997	0.008	0.1359	1.2307
	Digestives	Wafers	0.75000	0.17997	0.003	0.2026	1.2974
	Wafers	Crackers	−0.75000	0.17997	0.003	−1.2974	−0.2026
		Cookies	−0.68333	0.17997	0.008	−1.2307	−0.1359
		Digestives	−0.75000	0.17997	0.003	−1.2974	−0.2026
Zinc	Cookies	Shortcakes	10.2833	2.4200	0.002	2.923	17.644
		Digestives	9.4500	2.4200	0.006	2.089	16.811
		Cabins	10.7833	2.4200	0.001	3.423	18.144
		Wafers	11.9500	2.4200	0.000	4.589	19.311
	Shortcakes	Cookies	−10.2833	2.4200	0.002	−17.644	−2.923
	Digestives	Cookies	−9.4500	2.4200	0.006	−16.811	−2.089
	Cabins	Cookies	−10.7833	2.4200	0.001	−18.144	−3.423
	Wafers	Cookies	−11.9500	2.4200	0.000	−19.311	−4.589
Sodium	Crackers	Cabins	241.5000	64.4391	0.009	45.502	437.498
		Wafers	247.2333	64.4391	0.007	51.236	443.231
	Cabins	Crackers	−241.5000	64.4391	0.009	−437.498	−45.502
	Wafers	Crackers	−247.2333	64.4391	0.007	−443.231	−51.236
Magnesium	Cookies	Digestives	11.3650	3.4724	0.029	0.803	21.927
	Digestives	Cookies	−11.3650	3.4724	0.029	−21.927	−0.803
		Cabins	−12.3333	3.4724	0.015	−22.895	−1.772
	Cabins	Digestives	12.3333	3.4724	0.015	1.772	22.895
Calcium	Crackers	Wafers	−805.833	176.512	0.001	−1342.71	−268.95
	Cookies	Wafers	−697.500	176.512	0.005	−1234.38	−160.62
	Shortcakes	Wafers	−755.833	176.512	0.002	−1292.71	−218.95
	Digestives	Wafers	−987.167	176.512	0.000	−1524.05	−450.29
	Wafers	Crackers	805.833	176.512	0.001	268.95	1342.71
		Cookies	697.500	176.512	0.005	160.62	1234.38
		Shortcakes	755.833	176.512	0.002	218.95	1292.71
		Digestives	987.167	176.512	0.000	450.29	1524.05

[a] Pb, Ni, Co and Fe (excluded) showed significant mean differences.
[b] Mean difference is significant at the 0.05 level.

(PC1–PC4) explained 21.7 %, 17.8 %, 15.1 % and 14.5 %, respectively, of the total variabilities and gave eigenvalues >1. The score and loading plots were used to display the sample groupings, based on the biscuits' compositional similarities, and variable correlations of the assayed metals, respectively. In the score plots, in Fig. 2(a)–(c) and (e), it can be observed that the biscuits differed among themselves appreciably, with the cracker and digestive biscuits affording the highest values (PC1) while the wafers gave the lowest values (PC1). For the PC2 axis, wafers and digestives gave the highest and lowest values, respectively, whereas on the PC3 and PC4 axes, cookies and crackers were the farthest apart. These results allude to the compositional dissimilarities of the different types of biscuits assessed. Likewise, the differences observed among the biscuit samples of the same class can be rationalized based on their

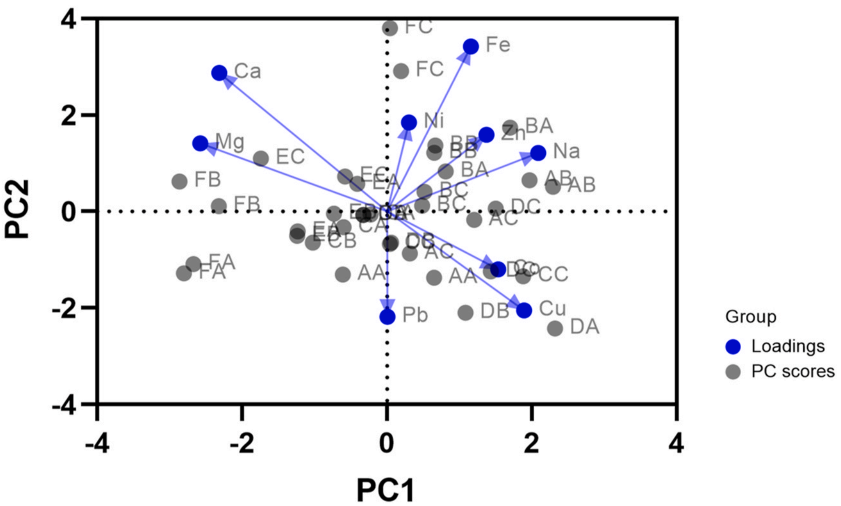

Fig. 6. Principal component analysis (PCA) score–loading biplot.

differences in manufacturer and location. Nevertheless, in the loading plots shown in Fig. 2(b)–(d) and (f), Na and Cu were revealed as the most dominant metals while Ca and Mg were the least dominant; with Pb maintaining the baseline (PC1 = 0), along the PC1 axes. The other four metals (Co, Zn, Fe and Ni) were positively correlated. Significantly, Na, Zn, Fe and Ni (PC2) and Fe, Co, Cu and Zn (PC3 and PC4) were in the positive quadrants of the loading plots while only Ca was present in the negative quadrants of the PC1 v PC3 and PC1 v PC4 graphs. No metal was observed in the negative quadrant of the PC1 v PC2 graph, but Pb was detected at the PC1 (=0) axis, between quadrants, in all the plots (*cf*. Fig. 2). In addition, Fig. 2(b) shows Fe as the most dominant metal along the PC2 axis whereas both Mg and Fe dominated on the PC3 and PC4 axes (*cf*. Fig. 2(d) and (f)).

Fig. 6 shows a representative score–loading biplot of PC1 vs. PC2, indicating that biscuit distributions, along the PC1 axis, were mainly based on metal contents. It is also evident that the highest levels of Fe and Zn, and Na and Pb are present in the cookies (BA & BC) and crackers (AA & AB), respectively; which are in agreement with the metals' digestion data in Table 4. In addition, it can be gleaned that digestives (DA & DB) and cookies (BB) are high in Cu and Ni, respectively. Therefore, the consumption of these types of biscuits will probably increase the levels of these metals in the body. In the same vein, the biplot PC3 and PC4 axes show the separation of the cabin and wafer biscuits; predominately distributed between the positive and negative axes, based on their high Mg and Ca contents, respectively.

Furthermore, the varimax-rotated PCA (with Kaiser normalization) generated four factors, with eigenvalues greater than one, which also explained 71.2 % of the total variance. The first factor loading (component 1) explained 21.8 % of the total variability of the metals studied and was attributed to Na (0.54), Mg (0.75) and Co (0.86), which furnished values > 0.5. Subsequently, they were classified with moderate (Na & Mg) and strong (Co) factor loadings, respectively (*cf*. Table 2), according to Belkhiri and Narany [116]. Similarly, strong factor loadings were exhibited by Cu (0.79) and Ca (0.87); in component 2, Fe (0.85); in component 3, and Ni (0.76); in component 4 while Pb (0.65; 0.54), in components 3 and 4, respectively, and Na (0.52), in component 4, were of moderate factor loadings. Significantly, all the four components for Zn gave weak factor loadings, with the first factor loading (component 1) recording an unclassified value (<0.3) of −0.1514. The second and third factor loadings explained 18.5 % and 16.1 %, respectively, of the total variance of the metals.

Conversely, the hierarchical cluster analysis (HCA), using Ward's linkage, and two-dimensional hierarchical clustering heatmap, depicted in Figs. 3 and 4, both grouped the metals into clusters. The HCA dendrogram (Fig. 3) classified the metals into three clusters; grouping most of the metals assessed (Ni, Co, Cu, Pb, Zn, Fe and Mg) in cluster 1 whereas Fe, Mg and Na were located in cluster 2, and Na and Ca found in cluster 3. Significantly, the dendrogram's rescaled distance was in the order of cluster 2 > cluster 3 > cluster 1. In other words, Fe and Mg were present in clusters 1 and 2 whereas Na was found in clusters 2 and 3. It is plausible that the clustered metals may have been similarly originated.

The two-dimensional hierarchical clustering heatmap (Fig. 4), on the other hand, grouped both metals and biscuits into clusters and subclusters, with a heatmap of 0–1 signifying metal levels. The three clusters consisted of Mg, Fe and Zn (cluster 1), and Na and Ca (cluster 3) while cluster 2 had Fe, Zn, Ni, Co, Pb, Cu and Na, with three subclusters of Fe, Zn and Ni; Zn, Ni and Co; and Ni, Co, Pb and Cu, respectively. The different biscuit types also exhibited two clusters of high intensities (red; Fig. 4) with Na but only one similar cluster with Ca; indicating high levels of the respective metals for the digestives and crackers (Na), and wafers (Ca), respectively. The blue cluster consisting of all the different biscuit samples as well as Ni, Co, Pb and Cu metals is characteristic of low intensities. It is noteworthy that the high levels of Na reported in the digestives, crackers and cookies as well as the high Ca content found in wafers are in agreement with the results from the wet digestion assays (*cf*. Table 4).

Next, the potential health risks of consuming the biscuits, under study, were assessed using such parameters as the biscuits' total detected metal contents, target hazard quotient (THQ), estimated daily intake (EDI) and total hazard index (HI) for children (aged 4–20 years) and adults, and collated in Table 5. The results show that Na and Ca were the most abundant metals at concentrations of 556 ± 130 mg/kg and 545 ± 420 mg/kg, respectively, whereas Ni gave the lowest concentration of 0.12 ± 0.15 mg/kg. Significantly, Cd was below the instrument's detection level (<0.2 mg/kg) but 1.49 ± 0.92 mg/kg Pb was detected in the biscuits.

The EDI values ranged from 0.0005 mg/kg BW/day (Ni) to 2.04 mg/kg BW/day (Na), for children, and 0.0001 mg/kg BW/day (Ni) to 0.46 mg/kg BW/day (Na), for adults. The EDI of metals recorded from biscuits' consumption decreased in the order of Na > Ca > Mg > Fe > Zn > Pb > Cu > Co > Ni for both children and adults. Gratifyingly, the calculated EDI of six of the metals assayed (Ni, Cu, Zn, Fe, Na and Mg) were below the recommended oral reference doses (R*f*D) for children and adults (*cf*. Table 5); indicating that the consumption of the biscuit brands examined did not pose any risks to health. Contrastingly, the EDI values recorded for Pb (0.0055 mg/kg BW/day) and Co (0.0012 mg/kg BW/day) in children were above the stipulated R*f*D for Pb (0.0035 mg/kg/day) [49] and Co (0.0003 mg/kg/day) [50] but the adult EDI of Pb (0.0012 mg/kg BW/day) was within limit whereas that of Co (0.00027 mg/kg BW/day) was borderline. It can, therefore, be surmised from the foregoing that the Pb and Co contents of these biscuits are sources of health risks to children. Conversely, the EDI values of the metals investigated were lower than the provisional tolerable weekly intake (PTWI) collated in Table 5, for both children and adults. Nevertheless, children are more prone to the toxic effects of PTM than adults [117,118], and their consumption of Pb-contaminated biscuits may result in neuro-developmental/behavioral impairments and reduction in the absorption of essential nutrients [119]. Moreover, in excess, Co has been reported to be mutagenic in somatic and germ cells, decrease glucose metabolism, and cause reproductive and developmental effects in animals [90]. It is hypothesized to affect heme synthesis. Studies have also indicated polycythemia and cardiomyopathy; in above-average beer consumers [91,120]. Neurologic effects, such as reversible hearing and vision impairments, in isolated cases have also been described as well as effects on the peripheral nervous system and potential cancer risk [90,120,121].

The non-carcinogenic health risks of ingesting any of the metals in the biscuits assayed were determined via the target hazard quotient (THQ) calculations [52]. Interestingly, all the metals in the biscuits sampled were within the acceptance limit (THQ <1) and posed no health risks, in both children and adults; except for Pb (1.56) and Co (4.04) in children, where THQ >1 (*cf*. Table 5) and, therefore, potentially liable for adverse non-carcinogenic health effects. Furthermore, the total hazard index (HI), calculated by adding the THQ values of the eight metals determined, gave values of 6.02 and 1.35 for children and adults, respectively. This implies that the total hazard indices for children and adults, in this study, were both above the acceptable limit (HI = 1). Consequently, the adults consuming the biscuits sampled herein fell within the medium range (HI ≥ 1 < 4; *cf.* Table 2) whereas the non-carcinogenic health risk of ingesting all eight metals present, in children, was categorized as high (HI ≥ 4) [49,54]. These results are reason for grave concerns, especially for children.

The consumable metal limits per week in the biscuits were also calculated, according to JECFA [64,65], with representative body weights of 40.9 kg for children [43,44] and 60.7 kg for adults [45], respectively. Shown in Table 6 are the JECFA limits and the quantities of the different biscuit types that must be consumed per week per body weight to reach these limits. It is notable that the cracker biscuits recorded the lowest values for Pb in children (0.44 kg) and adults (0.66 kg), respectively, whereas shortcakes gave the highest values of 1.23 kg (children) and 1.83 kg (adults), respectively. These computed consumable quantities of biscuits vis-à-vis the JECFA limits are significant taking cognizance of the facts that no level of Pb exposure is without health risks [107] and that the US Food and Drug Administration (FDA)-recommended serving size (i.e., how much an individual should be consuming per sitting) for biscuits is 30–55 g [122].

Besides, the highest levels of consumable biscuits (*cf.* Table 6) in the three macrominerals determined were recorded, in children and adults, respectively, thus {metal (children, adults; biscuit)}: Ca (1990.3, 2953.9; digestives), Na (1522.9, 2260.1; wafers) and Mg (1219.7, 1810.2; digestives) whereas the five micronutrients afforded the following: Cu (230.9, 342.7; wafers), Co (130.1, 193.1; wafers), Zn (83.5, 123.9; wafers), Fe (16.6, 24.6; wafers) and Ni (28.6, 42.5; shortcakes). To surmise, therefore, the results showed that the quantities of biscuits consumable per week per person (body weight) in order to exceed the JECFA limits for the metals assayed, in this study, increased in the order of Ni < Fe < Zn < Co < Cu < Mg < Na < Ca in children and adults. It was also evident that the wafers were the more prevalent and safer biscuit type since relatively larger quantities are required to reach the JECFA limits.

The carcinogenic health risks of consuming the metals (Pb and Ni) in the biscuit samples were also evaluated using the target cancer rate (TCR) calculations as shown in equation (5) [56] and collated in Table 7. The results for Pb, in children, revealed the digestives as having the highest TCR of 6.6×10^{-3} whereas the crackers furnished the lowest TCR of 7.17×10^{-5}. However, for adults, the highest calculated TCR (Pb) was unexpectedly lower at 1.89×10^{-3} (for crackers) in comparison to the TCR calculated for children. Nonetheless, the lowest adult TCR (Pb) of 6.84×10^{-4} (shortcakes) was higher than the lowest children's TCR for Pb (7.17×10^{-5}; crackers). On the contrary, the TCR for Ni ranged from 1.83×10^{-4} (shortcakes) to 9.17×10^{-4} (wafers), for children, and 4.12×10^{-5} (shortcakes) to 2.06×10^{-4} (wafers), for adults. It is vital to note that all the biscuits (except crackers; in children) gave TCR (Pb) values above the United States Environmental Protection Agency (USEPA)'s maximum threshold [54,56] of 1×10^{-4}. In the same vein, most of the TCR (Ni) were borderline for children whereas, in adults; cookies, shortcakes and cabins recorded TCR (Ni) values below 1×10^{-4} (*cf.* Table 7). It can be garnered, therefore, that the consumption of biscuits with these levels of Pb can lead to carcinogenic health risks in children as well as adults. In contrast, the risk of exposure to cancer, from Ni, to the children and adult populations consuming these biscuits is lower but may require further investigation because of the values' proximities to the threshold of acceptable risk for the potential development of cancer [47].

At this juncture, it may be somewhat remiss not to note that food wastes have been reported to occur at all stages of the supply chain and consumption process and, therefore, remain a systemic problem [123,124]. Globally, the generation of food wastes poses a significant source of burden to the environment, communities and society, at large [125]. Moreso, about 8 % of global warming, due to annual greenhouse gas emissions, has been attributed to food-based wastes [124,126]. PTM-containing foods and food wastes have also been reported to cause ecological damages [58,60]. Accordingly, it is plausible to posit that improperly warehoused, unused and/or expired biscuits as well as concomitant wastes from their human consumption can indirectly introduce deleterious PTM into the environment and, by extension, cause damage to ecosystems [127,128].

Collated in Table 8, therefore, are the values calculated for the potential ecological risks (RI) and ecological risk indices (ERI) of six of the potentially toxic metals (Pb, Ni, Cu, Co, Zn & Fe) assessed. Expectedly, Pb recorded the highest RI of 5.75 (crackers) while Ni (in shortcakes) posed the lowest RI with 0.05. The RI (mean) values decreased from Pb (3.72) in the order: Pb > Fe > Cu > Co > Ni > Zn; indicating Zn (0.12) as the PTM with the least potential to cause ecological damage, from biscuit wastes, amongst the metals determined. Conversely, the cracker and shortcake biscuits showed the highest and lowest ERI of 8.194 and 4.597, respectively. It is instructive to note that the highest and lowest RI values for Pb were also recorded for crackers and shortcakes, respectively. Gratifyingly, the ecological risks attributable to expired and/or egested biscuit wastes were categorized as low since the RI and ERI values were less than 40 and 150, respectively (*cf.* Table 2). Thus, it is plausible to surmise that the PTM-contaminated biscuit wastes, in this study, posed minimal risks to ecology.

5. Conclusion

The quantification of nine metals in six different types of commonly consumed biscuits in southwestern Nigeria has been successfully carried out, and multivariate statistical techniques used to simplify and organize the ensuing data. The human health risks effects in children and adults as well as ecological risks attributable to expired and/or egested biscuit wastes were also assessed. The metals' determinations on the biscuits afforded lower concentration values than the permissible limits, with the exception of Pb. Remarkably, only the wafer biscuits recorded a recommended daily intake (RDI) value above the World Health Organization's recommendation [76] of 1000 mg per day for Ca, for young adults.

Furthermore, the chemometric analyses revealed non-correlations between the metals; via the Pearson correlation coefficient at 95 % confidence level, generated a four-component factor analysis with eigenvalues greater than one in the principal component analysis

(PCA); which was used to assess the distribution of the metals in the biscuits, via score and loading plots. Also, the hierarchical cluster analysis (HCA), using Ward's linkage, furnished three clusters whereas the two-dimensional hierarchical clustering heatmap generated three main clusters and subclusters; for the metals, and multiple cluster dendrites; for the biscuit samples. It is apposite to note that the one-way ANOVA and Tukey's honest significant difference (HSD) tests of mean differences were also in agreement on the significances of the metals assayed.

The human health risk assessments of metals in the biscuits, using estimated daily intakes (EDI), target hazard quotients (THQ), hazard indices (HI) and target cancer rates (TCR), revealed the EDI values for Pb and Co to be above the oral reference doses (R*f*D) for children whereas, in adults, EDI (Pb) was below its R*f*D while that of Co was borderline. These were similarly replicated in the THQ wherein the values for Pb and Co were above the acceptance limit for children. Consequently, the HI of the metals were >1 and categorized as high and medium in children and adults, respectively. In the same vein, the TCR calculations, for children and adults, for Pb gave values above the United States Environmental Protection Agency (USEPA)'s maximum threshold [54,56] of 1×10^{-4} whereas for Ni, most of the biscuits were borderline; with 50 % of the biscuits assayed in the adult category falling below the USEPA maximum. Conversely, the consumable quantities of biscuits needed to exceed the Joint FAO/WHO Expert Committee on Food Additives (JECFA)'s metal limits decreased from Ca to Ni (i.e., Ca > Na > Mg > Cu > Co > Zn > Fe > Ni) in both children and adults. A corollary of the health risk assessments of ingesting the metals assayed in the biscuits is therefore that Pb is potentially liable to cause adverse non-carcinogenic and carcinogenic health effects in children whereas Co and Ni possessed borderline non-carcinogenic and carcinogenic health risks, respectively, in children.

Lastly, the raw materials accessed, poor packaging practices, industrial and automobile emissions, and ineffectual storage conditions are some of the conceivable sources of contaminants in the biscuits sampled. Nevertheless, the enforcement of strict quality assurance/controls and hazard analysis critical control point (HACCP) protocols pre/post-production can prove ameliorating. It is also gratifying that the ecological risk assessments to evaluate the likelihood of wastes from improperly warehoused, unused and/or expired potentially toxic metals (PTM)-contaminated biscuits, which could cause adverse harm to the ecosystem, were of low risks. The importance of constant monitoring cannot be, however, overemphasized.

Data availability statement

Other data related to this work can be obtained upon reasonable request from CTO (chionyedua.onwordi@lasu.edu.ng).

CRediT authorship contribution statement

Chionyedua T. Onwordi: Writing – review & editing, Writing – original draft, Validation, Project administration, Methodology, Funding acquisition, Data curation, Conceptualization. **Josephat U. Izunobi:** Writing – review & editing, Writing – original draft, Visualization, Resources, Formal analysis, Data curation. **Chukwudi N. Adiele:** Writing – original draft, Visualization, Resources, Investigation. **Aderonke O. Oyeyiola:** Writing – original draft, Methodology, Investigation. **Adelani J. Bamtefa:** Writing – original draft, Validation, Methodology, Investigation. **Adebola I. Akinjokun:** Writing – review & editing, Writing – original draft, Validation. **Leslie F. Petrik:** Writing – review & editing, Supervision, Project administration.

Declaration of competing interest

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

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.heliyon.2024.e34958.

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