

# Evaluation of Heavy Metals Concentration in Imported Frozen Fish *Trachurus Murphyi* Species Sold in Zaria Market, Nigeria

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**Abstract** The aim of the study is to assess the quality of fish imported into Nigeria with samples collected in Zaria. Concentrations of cadmium, lead, mercury, iron and nickel (Cd, Pb, Hg, Fe and Ni) were determined in the tissues/organs (skin, muscles, gills, liver, intestine, kidneys, brain and bones) of *Trachurus Murphyi* fish species on the basis of their import batches and across two major fishing origins. Health risks to human via dietary intake of fish were determined by the assessments of hazard quotients (HQ), daily intake of metals (DIM) and the individual's health risk index (HRI). Elemental determinations of the metals were performed by AAS via microwave digestion method of the fish tissues. The analysis of heavy metals variations (Post-Hoc) shows there are no significant differences ( $p < 0.05$ ) across the batches of most tissues. However, significant correlations among the tissues for each of the heavy metals were observed. Investigated results indicate high concentrations of heavy metals exist in each tissue and were above the safety limits recommended by FAO/WHO. The levels of heavy metals across the entire tissues were given in an increasing order as: Fe > Hg > Pb > Cd > Ni. However, individuals consuming fish above the recommended daily intake might be at risk from ingestion of heavy metals at unacceptable concentrations.

**Keywords** Frozen fish, *Trachurus Murphyi*, Heavy metals, Risk assessment, Safety

## 1. Introduction

Contaminations of fish species with toxic heavy metals are potential ways of human exposure. Consumption of imported frozen fish was found to supersede many source of animal protein such as beef, mutton and chickens among others in Nigeria [1]. Massive importation of frozen fish in the country has ranked Nigeria the largest importer of frozen fish in Africa [1]. With this regard, fish represent a valuable source of protein and nutrients of fundamental importance for diversified and healthy diets such as vitamins (D, A and B), minerals (calcium, iodine, zinc, iron and selenium) and polyunsaturated Omega-3 fatty acids (docosahexaenoic acid and eicosapentaenoic acid) [2]. Consequently, different variety of frozen fish species are imported and sold, although the species *Trachurus Murphyi* is the most common and more consumables among the consumers of fish in Zaria Metropolis. This fish species (*Trachurus Murphyi*) is particularly reported as wild captured and hence can either be *epipelagic* or *Benthopelagic* schooling species.

However, Benthos fish species are reported the most susceptible to water pollutions [3].

Fish bio-accumulates heavy metals to a much higher degree in which analysis of water along, cannot track out the exact concentrations. Fish and shellfish bio-accumulate metals in varied concentrations many times higher than the level present in water body and its sediment [4]. Fish take up metals from their surrounding habitat and concentrated to ranges of varied levels in their body tissues [4, 5]. The concern about the high levels of trace metals in seafood has prompted several researches and particularly frozen fish are of greater concern.

Heavy metal pollution of water has become a major environmental problem, almost since the advent of agricultural and industrial revolution. Today, most water resources are still being contaminated with heavy metals released from natural and anthropogenic activities such as weathering/erosion, volcanic eruption, mining activities, oceanic surge, domestic activities, industrial discharge, agricultural practices and as well as meteors deposition are washed and dumped in the water bodies [6-8]. The threat of toxic heavy metals in the environment was more serious than those of other pollutants, as a result of their non-biodegradable nature, accumulative properties and long biological half-lives [6]. They are difficult to read out

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completely from the environment once they entered [9]. The impact of heavy metals on aquatic life and fish may pose potential health implication and eating contaminated fish are areas of considerable concern. Because fish was at the top apex of the food chain, ones affected the entire food chain may also be highly risk [10].

In Nigeria, there are limited reports on safety assessments of heavy metals in imported frozen fish. Looking in to the fact that, some of these fish might be brought from heavily contaminated waters with heavy metals. Several studies shows that wild captured marine fish bio-accumulates heavy metals to a greater extend that remain an area of concern [11-15]. These wild captured fish are supplied in frozen form, deep frozen method can only preserved it from decomposition by slowing down some biochemical activities, but do not have any impact on the presence of heavy metals contaminants [16]. Therefore, this research aimed at the assessments of heavy metals safety levels in *Trachurus Murphyi* fish species imported and sold in Zaria Metropolis, Nigeria and determined the associated health risk as a result of the fish consumptions.

## 2. Experimental

### Quality assurance

The reagents used in this work were grades of chemicals from Sigma-Aldrich Company. Double distilled and de-ionized water were used throughout the experimentation except where indicated otherwise. Dissecting surgical blades, plastic containers and trays were washed and rinsed with distilled water. Glass wares were soaked in 10% HNO<sub>3</sub> (for 24hours) and rinsed with distilled water. Preparations of all the standard solutions were performed in a clean laboratory environment. All the samples were digested along with the blanks. Quantification of metallic content of the digested samples and the blanks was carried out with the aid of Varian AA240 Fast Sequential Flame Atomic Absorption Spectrophotometer (AAS) and vapor generation accessory (Varian VGA 77) with closed end cell was used for Hg determination in MULTI-USER SCIENCE RESEARCH LABORATORY (MSRL), Ahmadu Bello University Zaria.

To ensure that the Varian AA240 Fast Sequential Atomic Absorption Spectrometer remained calibrated at the course of the experimentation, the standards were analyzed after every ten runs. In the absence of reference standard materials, the nitrate salts of the metals were used to prepare multi-element standard solution (MESS) for spiking recoveries in the validation of digestion method. The analyzed samples were spiked and run in AAS again and the concentrations of the metal contents were determined from the calibration curve. The amounts of spiked metals recovered were used to calculate the percentage recoveries (%R) following equation 1 below.

$$\%R = \frac{C1-C2}{C3} \times 100 \quad (1)$$

Where, C1 is the spiked sample result, C2 is the un-spiked

sample result and C3 is the concentration of the MESS.

Method validation by conducting limit of detection (LOD), limit of quantification (LOQ) and precision relative standard deviation (RSD<sub>r</sub>) for repeatability within laboratory was also evaluated considering six level of standard solutions (N=6) with concentration values between 0.1 to 2.50mgkg<sup>-1</sup> were prepared. Determination of metals concentrations were carried out in triplicates per sample of fish tissues. Dilution factors of the collected data were corrected by calculations and the values were presented in the units of mgkg<sup>-1</sup>.

### Fish sampling

Twelve (12) different batches of *Trachurus Murphyi* species were purchased from frozen fish depot at Sabon-Gari in Zaria metropolis, considering six (6) batches, each obtained from two fishing origins (zones). Indicated on the cartons leaflets, batches of fish from Russia (RS) were labeled 1 - 6 and those from Europe (EU) were labeled 6 - 12. The samples were collected in clean polythene bags, labeled and transported immediately to the laboratory. In the laboratory the samples were stored in refrigerator to remain freeze prior to experiment. Fish destination and other some general information was obtained from the cartons information leaflets. The incremental number of fish drawn per carton was based on the recommendation by FAO [17]. Since the cartons net weight was found to be less than 50kg, thereby three (3) incremental fish were drawn randomly and composites from each batch of the carton, because some organs are tiny.

### Fish pre-treatment and dissection

Frozen fish samples were thawed, washed with distilled water and then allowed to attain room temperature in desiccators before dissection. The skin and muscle were removed following *Tru-cut* method by Baker *et al.* [18]: few scales were removed from the dorsal region on the first side of the fish just below the dorsal fin using a sterilized notched needle. The outer barrel was inserted to a depth of about 1cm into the fish muscle tissue beneath the scale at an oblique angle (to minimize penetration depth). The 2cm long notched needle (inner barrel) was then extended into the flesh. The containment cover (i.e. sharp outer barrel) slides over the extended needle to cut the tissue and capture it within the notch. The needle was also withdraw, the barrel opened and tissue slug remove with stainless steel tweezers (which were washed between samples) and placed in a labeled plastic Petri dish. While at the other side, few scales were removed and the skin was cut-off firmly making sure no part of the muscle was attached and placed in its labeled drying dish.

The gills, brain, liver, intestine, kidney and bone tissues were dissected based on the modified NIVA method by Rosseland *et al.* [19]. Dissections were done on plane plastic tray in to separate sample tissues/organs: The operculum gill cover was lift up and cut to expose the entire gill ash, filament and the rake. The abdominal wall was cut through the tail using tweezers and laid the fish on its right side with head to left. The bile bladder was removed from the liver and

then whole pooled liver was also freed onto the abdominal wall and cut out. The whole intestine was cut out and placed in to its sample container. The roof of the head was cut horizontally from the nostrils through the end of the skull in order to exposed the brain and fetch out and cut accordingly. Bones were de-fleshed and cut out entirely from the head to the tail. The organs were dried to constant weight at 80°C on plastic Petri dish and cooled in desiccators and powdered in porcelain mortar and pestle.

### Digestion

The digestion of the sample organs were based on the Microwave-assisted wet digestion method as described by Taghipour and Aziz [20]: 1g constant weight samples tissues/organs were placed in polytetrafluoroethylene (PTFE) tube for microwave. Digestion reagents (mixtures of 6ml ultra-pure Nitric acid, 65% and 2ml hydrogen peroxide, 35% in a ratio of 3:1) were added and placed in a microwave oven for 2minutes and then cooled to temperature of 25°C inside oven. The cleared solution were also diluted with de-ionized water to 50ml for skin, muscles, gills, liver, intestine, kidneys, brain and bones and then filtered using Whatman filter paper (90mm). The levels of Cd, Pb, Hg, Fe and Ni in the samples were determined with AA 240 Fast Sequential Atomic Absorption Spectrophotometer (AAS). In contrast to the original method by Taghipour and Aziz [20], modification was made based on the adjustment of heating duration in microwave oven. In their work they used 20minutes period of digestion, but in this work, attempting to digest fish tissues samples beyond 2minutes lead the sample inside polytetrafluoroethylene (PTFE) tube to burn leaving behind carbon residue. Hence, our modification of this method came in digestion duration (2mins instead of 10mins).

**Table 1.** Microwave operation

Operation	Output
Machine model number	MW028A-MG720
Power (output)	700W
Turntable Diameter	255m
Function/power operation	High (100%)
Digestion time	2minutes

### Risk assessment

Risk assessment in this study was evaluated by considering only the muscles tissues of *Trachurus Murphyi*, because it is the edible part of fish consumed by the population of fish consumers in Zaria metropolis, Nigeria. The evaluation includes the Heath Quotient (HQ), Daily Intake Metal (DIM) and Health Risk Index (HRI) according to methods by Sajjad *et al.* and Okunola *et al.* [21, 22].

### Hazard Quotient (HQ)

Hazard Quotient (HQ) characterized the risk of human health contamination with the intake of heavy metal pollutants in fish. It is a determined ratio to the metal reference dose (RD) that can entails an estimate of metal

hazard on the population in the latter life with fish consumption. If the HQ value is less than one (1) then the metal will pose no risk on the population due to fish consumption. However, if HQ value is greater than one (1) then the population would experience risk of hazardous metals. The equation (2) below was used.

$$HQ = \frac{[W_{fish}] \times [M_{fish}]}{R_fD \times B_o} \quad (2)$$

Where,  $W_{fish}$  is the dry weight of edible fish consumed per day ( $gd^{-1}$ ),  $M_{fish}$  is the concentration of metal in the fish ( $mgkg^{-1}$ ),  $R_fD$  is the metal reference dose ( $mgkg^{-1}d^{-1}$ );  $1.0 \times 10^{-3}$ ,  $3.5 \times 10^{-3}$ ,  $1.0 \times 10^{-4}$ ,  $7.0 \times 10^{-1}$ , and  $2.0 \times 10^{-2} mgkg^{-1}d^{-1}$  for cadmium, lead, mercury, iron and nickel respectively [23] and  $B_o$  is the average body weight (kg).

### Daily Intake of Metals (DIM)

The daily intake of metals (DIM) was calculated to estimate the daily loading of metals into the body system (via the consumption of fish muscles recommended for daily intake as specified in this study) of a specified body weight of a consumer. The daily intake of metals (DIM) was determined by the equation below:

$$DIM = \frac{C_{metal} \times D_{fish} \times C_{factor}}{B_o} \quad (3)$$

Where,  $C_{metal}$  is the concentration of heavy metals in the fish ( $mgkg^{-1}$ ),  $D_{fish}$  is the daily nutritional intake of fish ( $gday^{-1}$ ),  $C_{factor}$  is the factor for conversion of fresh fish to dry constant weight. For *Trachurus Murphyi* fish Species in this study,  $C_{factor}$  was considered as 0.299 as computed by the equation below as outline by USEPA [24].

In this study, the daily intake of fish for nutritional requirement was 100g intake rate for adults with average body weight of 70kg (aged 19years and above), 80g intake rate for children with average body weight of 48kg (aged range from 7- 18years) and 60g intake rate for individuals with body weight of 19kg (aged range 6years and below) using the method by Portier *et al.* [25] as recommended by USEPA [26].

$$C_{factor} = IR_{ww} - IR_{dw} \quad (4)$$

$$IR_{ww} = IR_{dw} \left[ \frac{100-W}{100} \right] \quad (5)$$

Where,  $IR_{dw}$  is the dry weight intake rate;  $IR_{ww}$  is the wet weight intake rate and  $W$  is the percent water content of the raw muscles (in this study was 70.10%).

### Health Risk Index (HRI)

The health risk index (HRI) for the populations through the consumption of contaminated fish were assessed based on the daily intake of metals (DIM) relative to reference oral dose ( $R_fD$ ) for each metal. This is an index justifying individual's risk of heavy metals exposure. The HRI value of less than one (1) implies safe exposure from such heavy metal and considered acceptable, otherwise the fish may pose heavy metals risk. The following formula was used for the calculation of HRI.

$$HRI = \frac{DIM}{R_fD} \quad (6)$$

**Table 2.** Quality assurance for mean concentration of metals in the tissues of *Trachurus Murphyi* species

Heavy metal	Tissues	Concentrations of the sample (mgL <sup>-1</sup> )	Concentrations of the spiked sample (mgL <sup>-1</sup> )	%Recovery±SD	LOD (mgkg <sup>-1</sup> ) N=6	LOQ (mgkg <sup>-1</sup> ) N=6	%RSD <sub>r</sub>
Cd	Skin	0.040	1.127	108.7±0.32	0.061	0.262	0.29
Pb	Muscle	0.021	1.025	100.4±0.20	0.004	0.012	1.92
Hg	Gills	2.509	3.501	99.2±0.45	4.86E-4	0.002	4.54
Fe	Liver	12.828	13.900	107.2±0.15	6.78E-5	3.21E-4	13.99
Ni	Intestine	0.055	1.051	99.6±0.10	0.012	0.050	1.00
Cd	Kidneys	0.291	1.309	101.8±0.31	0.024	0.070	0.45
Pb	Brain	0.078	1.069	99.1±0.25	0.005	0.011	2.52
Hg	Bones	1.494	2.488	99.4±0.35	9.23E-4	0.003	3.52

Average of eight observations from three replicate analyses of each of analyzed fish tissues was spiked

### Statistical analysis

The data were expressed as the metal concentration in the tissues across the batches with the average means ± standard deviation. To show whether there is significant difference between the batches, Post-Hoc analysis of variance (Duncan) was used and the Pearson correlation (r) was used to establish the degree of relationship among the tissues based on the analyzed metals across the fishing areas using a statistical software package (IBM SPSS version 20).

## 3. Results and Discussion

### Quality Assurance

As shown in Table 2, the results of validation parameters for the analytical procedures including recoveries of the spiked fish tissues obtained for the investigated metals (Cd, Pb, Hg, Fe and Ni), which varied between the ranges of 99.10% to 108.70%. Acceptable recoveries were obtained in all cases, which show that the digestion method used for fish samples tissues and the AAS analysis were reliable. Also, comparison of the recoveries data in this study showed that the values are within the range of 90 – 120% and these were in compliance with the Standard Operating Procedure (SOP) [27, 28].

The evaluated results for limit of detection (LOD), limit of quantification (LOQ) and the precision relative standard deviation (RSD<sub>r</sub>) for within laboratory repeatability validates the methods of experimental analysis used. The results in Table 2 with respects to entire metals shows the LOD values of specification of not more than one tenth and for LOQ values of not more than one fifth as specified by the USEPA regulation [28]. However, the authors determined LOD and LOQ as  $\bar{x} \text{ blank} + 3S_{\text{blank}}$  and  $\bar{x} \text{ blank} + 10S_{\text{blank}}$  respectively, based on the standard deviation of the blanks. Where  $\bar{x}$  blank was the mean of the blank aqueous solution and Sblank as the blank standard deviation. The limits presented here were assessed experimentally by standard addition assessment with fish samples, providing more realistic limits for the method. While the precision taken for %RSD<sub>r</sub> was evaluated as  $(S/\bar{x}) \times 100$ .

### Metal Concentrations

The statistical results of heavy metals; cadmium, lead, mercury, iron and nickel (Cd, Pb, Hg, Fe and Ni) concentrations (mgkg<sup>-1</sup>) in the tissues/organs (skin, muscle, gills, liver, intestine, kidneys, brain and bones) of *Trachurus Murphyi* across two fishing origins including the mean, standard deviation and Post-Hoc test analysis are summarized in Tables 3 to 7. Analysis of variation between fish samples collected from the two fishing origins; Russia (RS) and Europe (EU) and the sample batches within the same zone showed significant differences (P<0.05). The results of correlation analysis among the tissues/organs of the fishing zones across the studied metals are shown in Tables 8 to 17.

### Cadmium concentrations

The results in Table 3 presented the concentrations of cadmium accumulations in the tissues of *Trachurus Murphyi* fish species across the batches of RS and EU fishing origins. The mean concentrations of cadmium accumulations in the tissues of RS fish species were ranged between the values of 1.400mgkg<sup>-1</sup> to 36.650mgkg<sup>-1</sup> shown for the muscles and liver tissues respectively. While for EU fish species, the accumulations of cadmium recorded across the batches were ranged between the values of 2.175mgkg<sup>-1</sup> to 37.867mgkg<sup>-1</sup> shown for the muscles and liver tissues respectively. The results of statistical analysis shows that, significant differences (P<0.05) for cadmium accumulation were recorded among batches of RS fish species and hence, these are shown in the batches of skin, muscles, gills and brain tissues respectively. However, the non-significance differences (p<0.05) recorded among RS batches were shown; for the liver tissues between batch 3, 5 and 6; for the intestine tissues between batch 1, 2 and 4, 2 and 6, 3 and 5, 4 and 6; for the kidneys tissues between batch 1 and 4, 2 and 6, 3 and 5; and for the bones tissues between batch 3 and 4, 4 and 6. In contrast, significance differences were not shown among the batches of EU fish species and hence, the non-significance differences (p<0.05) recorded were shown: for the skin tissues between batch 3 and 4, 5 and 6; for the muscles tissues between batch 5 and 6 only; for the gills tissues between batch 1 and 3 only; for the liver tissues between batch 1 and 4, 2, 3 and 6, 3 and 6; for the intestine

tissues between batch 2, 3 and 5, 2 and 6, 4 and 5; for the kidneys tissues between batch 1, 3 and 6, 2 and 5, 3 and 4, 4 and 5; for the brain tissues between batch 1 and 4 only; and for the bones tissues between batch 2 and 3, 3 and 6. The significant differences shown among the batches of RS tissues (skin, muscle, gills and brain), gives an indication that, contaminations with cadmium are due to the impacts of sources from non-related activities. While, the non-significant differences shown among other batches

(liver, intestine, kidneys and bones) imply that, contamination with cadmium are due to impact of related sources of activities leading to homogeneous contaminations cross the batches. With respects to EU fish species, since the entire batches shows non-significance differences, therefore contaminations with cadmium are strongly from the impact of related sources of activities and hence the entire batches were homogeneously contaminated.

**Table 3.** Cadmium concentrations in the tissues of *Trachurus Murphyi* (mgkg<sup>-1</sup>) wet weight

Tissue	Batch 1	Batch 2	Batch 3	Batch 4	Batch 5	Batch 6	Mean±SD	FAO/WHO[17]
Skin(RS)	2.000 <sup>a</sup>	1.050 <sup>b</sup>	1.350 <sup>c</sup>	1.450 <sup>d</sup>	1.950 <sup>e</sup>	1.750 <sup>f</sup>	1.592±0.065	0.100
Skin(EU)	1.600 <sup>a</sup>	2.300 <sup>b</sup>	2.550 <sup>c</sup>	2.500 <sup>c</sup>	2.150 <sup>d</sup>	2.750 <sup>d</sup>	2.308±0.020	
Muscle(RS)	1.750 <sup>a</sup>	1.050 <sup>b</sup>	1.200 <sup>c</sup>	1.400 <sup>d</sup>	1.600 <sup>e</sup>	1.400 <sup>f</sup>	1.400±0.025	
Muscle(EU)	1.850 <sup>a</sup>	1.750 <sup>b</sup>	2.100 <sup>c</sup>	2.350 <sup>d</sup>	2.100 <sup>e</sup>	2.900 <sup>e</sup>	2.175±0.015	
Gills(RS)	1.500 <sup>a</sup>	1.100 <sup>b</sup>	1.300 <sup>c</sup>	1.350 <sup>d</sup>	2.300 <sup>e</sup>	2.400 <sup>f</sup>	1.658±0.010	
Gills(EU)	3.335 <sup>a</sup>	1.800 <sup>b</sup>	1.900 <sup>a</sup>	6.550 <sup>c</sup>	1.950 <sup>d</sup>	2.100 <sup>e</sup>	2.939±0.005	
Liver(RS)	1.250 <sup>a</sup>	38.100 <sup>b</sup>	56.300 <sup>c</sup>	43.100 <sup>d</sup>	40.000 <sup>c</sup>	41.150 <sup>c</sup>	36.650±0.035	
Liver(EU)	39.700 <sup>a</sup>	67.550 <sup>b</sup>	26.050 <sup>bc</sup>	2.200 <sup>a</sup>	55.350 <sup>d</sup>	36.350 <sup>cb</sup>	37.867±0.030	
Intestine(RS)	8.550 <sup>a</sup>	4.250 <sup>ab</sup>	8.700 <sup>c</sup>	10.150 <sup>ad</sup>	7.100 <sup>c</sup>	6.200 <sup>bd</sup>	7.492±0.015	
Intestine(EU)	3.900 <sup>a</sup>	7.800 <sup>b</sup>	4.350 <sup>bc</sup>	27.200 <sup>d</sup>	7.200 <sup>bd</sup>	6.100 <sup>c</sup>	9.425±0.015	
Kidneys(RS)	14.550 <sup>a</sup>	17.800 <sup>b</sup>	17.050 <sup>c</sup>	6.800 <sup>a</sup>	10.950 <sup>c</sup>	7.800 <sup>b</sup>	12.492±0.015	
Kidneys(EU)	12.900 <sup>a</sup>	10.700 <sup>b</sup>	5.450 <sup>ac</sup>	8.750 <sup>dc</sup>	7.700 <sup>b</sup>	10.200 <sup>ad</sup>	9.283±0.005	
Brain(RS)	1.500 <sup>a</sup>	1.050 <sup>b</sup>	1.800 <sup>c</sup>	1.750 <sup>d</sup>	1.850 <sup>e</sup>	2.050 <sup>f</sup>	1.667±0.020	
Brain(EU)	9.350 <sup>a</sup>	1.650 <sup>b</sup>	1.650 <sup>c</sup>	4.650 <sup>ac</sup>	1.900 <sup>d</sup>	1.850 <sup>e</sup>	3.508±0.035	
Bones(RS)	1.600 <sup>a</sup>	1.250 <sup>b</sup>	2.900 <sup>c</sup>	2.400 <sup>c</sup>	2.350 <sup>d</sup>	2.800 <sup>c</sup>	2.217±0.035	
Bones(EU)	1.800 <sup>a</sup>	2.750 <sup>b</sup>	2.150 <sup>bc</sup>	2.800 <sup>d</sup>	3.000 <sup>e</sup>	2.600 <sup>c</sup>	2.517±0.010	

Values in each row marked by the same superscript letter are not significantly different at P<0.05

**Table 4.** Lead concentrations in tissues of *Trachurus Murphyi* (mgkg<sup>-1</sup>) wet weight

Tissue	Batch1	Batch 2	Batch 3	Batch 4	Batch 5	Batch 6	Mean±SD	FAO/WHO[17]
Skin(RS)	9.400 <sup>a</sup>	7.250 <sup>ab</sup>	9.200 <sup>ab</sup>	10.600 <sup>c</sup>	12.450 <sup>d</sup>	15.600 <sup>e</sup>	10.750±0.015	0.400
Skin(EU)	14.900 <sup>a</sup>	19.950 <sup>b</sup>	19.700 <sup>a</sup>	18.500 <sup>bc</sup>	21.900 <sup>bc</sup>	26.400 <sup>d</sup>	20.225±0.035	
Muscle(RS)	0.300 <sup>a</sup>	1.200 <sup>b</sup>	9.250 <sup>c</sup>	8.900 <sup>c</sup>	12.200 <sup>d</sup>	13.400 <sup>e</sup>	7.542±0.005	
Muscle(EU)	16.150 <sup>a</sup>	15.800 <sup>b</sup>	19.400 <sup>a</sup>	18.250 <sup>bc</sup>	19.500 <sup>bc</sup>	19.600 <sup>d</sup>	18.117±0.020	
Gills(RS)	1.050 <sup>a</sup>	4.900 <sup>b</sup>	8.050 <sup>c</sup>	10.050 <sup>d</sup>	12.250 <sup>e</sup>	14.700 <sup>f</sup>	8.500±0.025	
Gills(EU)	16.800 <sup>a</sup>	17.600 <sup>b</sup>	17.250 <sup>a</sup>	17.050 <sup>b</sup>	18.850 <sup>c</sup>	17.300 <sup>d</sup>	17.475±0.010	
Liver(RS)	2.900 <sup>a</sup>	6.500 <sup>b</sup>	8.700 <sup>c</sup>	11.900 <sup>d</sup>	11.300 <sup>d</sup>	17.850 <sup>e</sup>	9.858±0.005	
Liver(EU)	18.050 <sup>a</sup>	17.600 <sup>ab</sup>	19.900 <sup>c</sup>	16.550 <sup>abd</sup>	22.600 <sup>de</sup>	16.500 <sup>e</sup>	18.533±0.045	
Intestine(RS)	2.200 <sup>a</sup>	6.900 <sup>b</sup>	7.950 <sup>c</sup>	12.750 <sup>d</sup>	13.150 <sup>e</sup>	18.150 <sup>f</sup>	10.183±0.040	
Intestine(EU)	16.500 <sup>a</sup>	19.350 <sup>b</sup>	20.450 <sup>ab</sup>	18.100 <sup>ac</sup>	19.000 <sup>cd</sup>	18.200 <sup>d</sup>	18.600±0.040	
Kidneys(RS)	4.200 <sup>a</sup>	6.950 <sup>b</sup>	9.650 <sup>c</sup>	12.650 <sup>d</sup>	14.200 <sup>e</sup>	17.300 <sup>f</sup>	10.825±0.020	
Kidneys(EU)	19.050 <sup>a</sup>	20.750 <sup>ab</sup>	19.100 <sup>a</sup>	17.300 <sup>a</sup>	19.450 <sup>b</sup>	16.350 <sup>b</sup>	18.667±0.030	
Brain(RS)	3.900 <sup>a</sup>	5.900 <sup>b</sup>	8.450 <sup>c</sup>	10.400 <sup>d</sup>	12.100 <sup>e</sup>	15.350 <sup>f</sup>	9.350±0.010	
Brain(EU)	19.500 <sup>a</sup>	17.750 <sup>ab</sup>	18.400 <sup>bc</sup>	17.000 <sup>ac</sup>	20.250 <sup>a</sup>	13.000 <sup>d</sup>	17.650±0.015	
Bones(RS)	3.450 <sup>a</sup>	6.800 <sup>b</sup>	7.550 <sup>c</sup>	11.200 <sup>d</sup>	14.100 <sup>e</sup>	15.250 <sup>f</sup>	9.725±0.030	
Bones(EU)	17.600 <sup>a</sup>	17.150 <sup>ab</sup>	17.650 <sup>ab</sup>	17.150 <sup>c</sup>	20.300 <sup>e</sup>	13.350 <sup>f</sup>	17.200±0.005	

Values in each row marked by the same superscript letter are not significantly different at P<0.05

**Table 5.** Mercury concentrations in the tissues of *Trachurus Murphyi* (mgkg<sup>-1</sup>) wet weight

Tissue	Batch 1	Batch 2	Batch 3	Batch 4	Batch 5	Batch 6	Mean±SD	FAO/WHO[17]
Skin(RS)	160.30 <sup>a</sup>	73.700 <sup>ab</sup>	101.55 <sup>bc</sup>	109.60 <sup>cd</sup>	113.50 <sup>de</sup>	120.65 <sup>ac</sup>	113.22±0.005	0.500
Skin(EU)	94.300 <sup>a</sup>	93.300 <sup>b</sup>	57.350 <sup>ac</sup>	82.200 <sup>b</sup>	85.800 <sup>cbd</sup>	86.400 <sup>ad</sup>	83.225±0.010	
Muscle(RS)	117.85 <sup>a</sup>	66.500 <sup>ab</sup>	103.15 <sup>c</sup>	99.550 <sup>bd</sup>	115.50 <sup>e</sup>	115.15 <sup>ad</sup>	102.95±0.020	
Muscle(EU)	92.850 <sup>a</sup>	88.450 <sup>b</sup>	66.050 <sup>ac</sup>	79.350 <sup>cd</sup>	81.700 <sup>bd</sup>	78.700 <sup>ab</sup>	81.183±0.005	
Gills(RS)	125.45 <sup>a</sup>	88.550 <sup>ab</sup>	102.95 <sup>bc</sup>	103.25 <sup>cd</sup>	110.40 <sup>d</sup>	112.65 <sup>a</sup>	107.21±0.005	
Gills(EU)	84.200 <sup>a</sup>	88.000 <sup>b</sup>	65.050 <sup>ac</sup>	71.450 <sup>bd</sup>	80.600 <sup>ac</sup>	83.000 <sup>bd</sup>	78.717±0.030	
Liver(RS)	126.50 <sup>a</sup>	114.85 <sup>b</sup>	102.35 <sup>ac</sup>	107.10 <sup>b</sup>	114.35 <sup>c</sup>	126.10 <sup>a</sup>	115.21±0.040	
Liver(EU)	89.400 <sup>a</sup>	96.550 <sup>b</sup>	66.950 <sup>ac</sup>	78.450 <sup>cd</sup>	88.700 <sup>e</sup>	79.150 <sup>ad</sup>	83.200±0.035	
Intestine(RS)	101.65 <sup>a</sup>	110.90 <sup>ab</sup>	96.60 <sup>abc</sup>	110.95 <sup>cd</sup>	115.55 <sup>de</sup>	124.00 <sup>e</sup>	109.94±0.045	
Intestine(EU)	86.650 <sup>a</sup>	99.800 <sup>b</sup>	81.100 <sup>bc</sup>	87.100 <sup>ad</sup>	80.250 <sup>ad</sup>	84.000 <sup>bc</sup>	86.483±0.025	
Kidneys(RS)	91.500 <sup>a</sup>	111.15 <sup>b</sup>	102.10 <sup>b</sup>	113.50 <sup>c</sup>	112.05 <sup>c</sup>	126.80 <sup>d</sup>	109.51±0.035	
Kidneys(EU)	85.450 <sup>a</sup>	62.950 <sup>ab</sup>	68.950 <sup>bc</sup>	84.050 <sup>d</sup>	84.400 <sup>e</sup>	83.250 <sup>ac</sup>	78.175±0.015	
Brain(RS)	63.400 <sup>a</sup>	99.550 <sup>b</sup>	101.65 <sup>c</sup>	105.50 <sup>d</sup>	118.35 <sup>e</sup>	137.10 <sup>f</sup>	104.26±0.010	
Brain(EU)	86.450 <sup>a</sup>	55.150 <sup>ab</sup>	80.650 <sup>bc</sup>	85.150 <sup>ac</sup>	92.400 <sup>d</sup>	87.550 <sup>d</sup>	81.225±0.015	
Bones(RS)	74.700 <sup>a</sup>	95.150 <sup>b</sup>	99.800 <sup>c</sup>	116.90 <sup>d</sup>	116.15 <sup>d</sup>	137.10 <sup>e</sup>	106.63±0.020	
Bones(EU)	87.650 <sup>a</sup>	53.000 <sup>ab</sup>	75.500 <sup>bc</sup>	92.750 <sup>ad</sup>	88.250 <sup>e</sup>	81.450 <sup>cd</sup>	79.767±0.030	

Values in each row marked by the same superscript letter are not significantly different at P<0.05

**Table 6.** Iron concentrations in the tissues of *Trachurus Murphyi* (mgkg<sup>-1</sup>) wet weight

Tissue	Batch 1	Batch 2	Batch 3	Batch 4	Batch 5	Batch 6	Mean±SD	FAO/WHO[45]
Skin(RS)	86.950 <sup>a</sup>	49.050 <sup>ab</sup>	75.250 <sup>c</sup>	58.350 <sup>ab</sup>	1103.6 <sup>d</sup>	117.15 <sup>f</sup>	248.39±0.020	0.800
Skin(EU)	36.200 <sup>a</sup>	37.650 <sup>b</sup>	47.550 <sup>ac</sup>	35.950 <sup>b</sup>	257.50 <sup>cd</sup>	25.800 <sup>ad</sup>	73.442±0.045	
Muscle(RS)	60.250 <sup>a</sup>	43.600 <sup>b</sup>	51.250 <sup>bc</sup>	42.950 <sup>c</sup>	54.400 <sup>d</sup>	38.050 <sup>a</sup>	48.417±0.040	
Muscle(EU)	22.250 <sup>a</sup>	10.900 <sup>b</sup>	0.000 <sup>c</sup>	17.000 <sup>cd</sup>	33.900 <sup>d</sup>	22.200 <sup>d</sup>	17.708±0.045	
Gills(RS)	374.15 <sup>a</sup>	595.60 <sup>b</sup>	379.35 <sup>bc</sup>	394.65 <sup>cd</sup>	521.15 <sup>e</sup>	408.10 <sup>d</sup>	445.50±0.030	
Gills(EU)	443.85 <sup>a</sup>	318.95 <sup>b</sup>	221.20 <sup>bc</sup>	137.35 <sup>a</sup>	516.70 <sup>ad</sup>	342.30 <sup>d</sup>	330.06±0.020	
Liver(RS)	641.40 <sup>a</sup>	655.60 <sup>b</sup>	775.40 <sup>bc</sup>	730.00 <sup>d</sup>	705.60 <sup>d</sup>	636.75 <sup>ac</sup>	690.79±0.025	
Liver(EU)	635.55 <sup>a</sup>	641.90 <sup>b</sup>	315.70 <sup>bc</sup>	300.05 <sup>ad</sup>	363.20 <sup>ad</sup>	334.65 <sup>bc</sup>	431.84±0.005	
Intestine(RS)	112.85 <sup>a</sup>	164.00 <sup>b</sup>	163.10 <sup>bc</sup>	242.25 <sup>bd</sup>	166.40 <sup>d</sup>	133.30 <sup>bd</sup>	163.65±0.010	
Intestine(EU)	70.650 <sup>a</sup>	84.650 <sup>ab</sup>	22.90 <sup>ac</sup>	368.60 <sup>d</sup>	79.150 <sup>bd</sup>	75.100 <sup>cd</sup>	116.84±0.020	
Kidneys(RS)	1205.45 <sup>a</sup>	1192.7 <sup>b</sup>	1108.1 <sup>c</sup>	380.40 <sup>ac</sup>	631.50 <sup>cb</sup>	390.20 <sup>ab</sup>	818.05±0.025	
Kidneys(EU)	325.15 <sup>a</sup>	320.85 <sup>b</sup>	58.45 <sup>ab</sup>	520.80 <sup>a</sup>	165.85 <sup>bc</sup>	514.50 <sup>c</sup>	317.60±0.005	
Brain(RS)	114.70 <sup>a</sup>	41.750 <sup>ab</sup>	79.800 <sup>bc</sup>	102.10 <sup>d</sup>	105.45 <sup>e</sup>	83.100 <sup>ac</sup>	87.817±0.010	
Brain(EU)	349.75 <sup>a</sup>	31.850 <sup>ab</sup>	324.10 <sup>c</sup>	43.650 <sup>cd</sup>	70.950 <sup>cd</sup>	33.650 <sup>ab</sup>	142.33±0.010	
Bones(RS)	58.000 <sup>a</sup>	36.400 <sup>a</sup>	76.250 <sup>b</sup>	95.100 <sup>bc</sup>	68.050 <sup>bd</sup>	78.950 <sup>cd</sup>	68.792±0.015	
Bones(EU)	26.950 <sup>a</sup>	23.500 <sup>ab</sup>	28.900 <sup>bc</sup>	68.050 <sup>c</sup>	34.250 <sup>c</sup>	40.400 <sup>e</sup>	37.008±0.020	

Values in each row marked by the same superscript letter are not significantly different at P<0.05

The result of cadmium correlation analysis for RS tissues presented in Table 8 shows that, significant (p<0.05) positive correlations were recorded between: gills versus brain tissues; liver versus bones tissues; and kidneys versus brain tissues. Whereas, significant (p<0.05) negative correlations were shown between: muscles versus liver, intestine and bones tissues; and kidneys versus bones tissues. While, the correlation result for EU tissues presented in Table 9 shows that, significant (p<0.05) positive relations were shown between: skin versus muscles tissues; gills versus intestine and bones tissues; and kidneys versus brain tissues. Hence, the significant (p<0.05) negative correlations were shown between: skin versus kidneys and brain tissues; gills versus liver tissues; liver versus intestine tissues; and brain versus bones tissues. Variations in the cadmium accumulation were shown among tissues of both RS and EU fish species. These

entails that positive correlations among some tissues implies, the uptake routes of cadmium were from similar sources of contaminations or the accumulation pattern portrayed via similar mechanisms due to physico-chemical condition of the surrounding fish habitat. While, negative correlations among the other tissues indicate that, the uptakes of cadmium are from non-related contaminations sources. In lined with the results of statistical significance, the results of correlation analysis also profound cadmium contaminations (with respects to both fishing origins) are strongly from the impacts of non-related sources. This is because, both revealed they are variations in the routes that lead to cadmium contaminations shown among batches as well as dissimilarities of tissues uptake.

The results in Table 4 presented both RS and EU tissues shows cadmium accumulations were highly shown in the

liver tissues and hence, lower accumulations were in the muscles tissues. This pattern of tissues accumulations was found in coinciding sequence with the reported studies by Canli *et al.* [31], Asegbeloyin *et al.* [32], Muniyan and Ambedkar [33], Shahat *et al.* [34] and Iopa and Adhikari [35] who profound in their studies that high metals accumulation in fish are shown in the liver tissues and least were shown in the muscles tissues. The higher concentration of cadmium shown in the liver relative to other tissues can be attributed to the high coordination of cadmium with metallo-thionein protein [4, 29]. In addition, the liver is the principal organ responsible for the detoxification, transportation, and storage of toxic substances and it is an active site of pathological effects induced by contamination [30]. However, the lower concentrations of cadmium shown in the muscles tissues were as results of being an inactive tissue. This was in lined with the reports shown by many authors that shows muscles tissues are inactive and hence it due accumulate less cadmium [38-40]. The rates of cadmium transport to the muscles tissues are govern by the accumulative nature of the liver. The higher accumulative is the cadmium in the fish liver, the lower chance of cadmium mobility to the inactive tissues such as muscles and bones tissues [41, 42].

With regards to fish from both origins, the cadmium levels shown in the intestine and kidneys tissues were higher than the results obtained for skin, gills, brain and bones. This is because these tissues are involved in the process of metabolism (that is in the absorption, storage and excretion of ingested feed during the process of digestion by the fish). Ahmad and Bibi [36] studied that, metals accumulations in the storage tissues were due to biding with the mucosal protein present in the tissues. Fish studies by Paul *et al.* [37] also show that, absorption of cadmium from the mouth or gills via gastrointestinal tract was mainly accumulates in the liver and kidneys. The mean concentration of cadmium shown with the entire tissues of *Trachurus Murphyi* across the batches of both fishing zones were more pronounced and were above the levels reported in many fish species that was studied based on wild captured [43, 44]. The high levels of cadmium showed across the entire tissues signals that, sources of cadmium were from the contribution of both natural and anthropogenic activities. Furthermore, Table 3 mean concentrations of cadmium shows the entire tissues across the batches of both fishing zones were more pronounced and found above the recommended safety limit specified by FAO [17].

**Table 7.** Nickel concentrations in the tissues of *Trachurus Murphyi* (mgkg<sup>-1</sup>) wet weight

Tissue	Batch 1	Batch 2	Batch 3	Batch 4	Batch 5	Batch 6	Mean±SD	FAO/WHO[46]
Skin(RS)	0.950 <sup>a</sup>	0.250 <sup>b</sup>	1.050 <sup>c</sup>	0.300 <sup>d</sup>	1.250 <sup>e</sup>	2.300 <sup>f</sup>	1.017±0.005	0.200
Skin(EU)	3.400 <sup>a</sup>	4.450 <sup>b</sup>	4.200 <sup>bc</sup>	3.500 <sup>c</sup>	1.750 <sup>b</sup>	1.200 <sup>d</sup>	3.083±0.010	
Muscle(RS)	0.950 <sup>a</sup>	0.300 <sup>b</sup>	0.150 <sup>c</sup>	1.200 <sup>d</sup>	1.150 <sup>e</sup>	0.900 <sup>f</sup>	0.775±0.005	
Muscle(EU)	3.850 <sup>a</sup>	4.100 <sup>b</sup>	3.550 <sup>bc</sup>	3.500 <sup>bc</sup>	0.350 <sup>d</sup>	2.600 <sup>b</sup>	2.992±0.015	
Gills(RS)	0.900 <sup>a</sup>	0.500 <sup>b</sup>	1.020 <sup>c</sup>	2.100 <sup>d</sup>	0.850 <sup>e</sup>	0.700 <sup>f</sup>	1.012±0.020	
Gills(EU)	4.300 <sup>a</sup>	3.100 <sup>b</sup>	5.550 <sup>bc</sup>	3.150 <sup>bc</sup>	0.150 <sup>c</sup>	1.400 <sup>d</sup>	2.942±0.005	
Liver(RS)	1.350 <sup>a</sup>	0.400 <sup>b</sup>	1.200 <sup>c</sup>	2.300 <sup>d</sup>	0.300 <sup>e</sup>	1.450 <sup>f</sup>	1.167±0.010	
Liver(EU)	3.550 <sup>a</sup>	4.000 <sup>b</sup>	4.050 <sup>c</sup>	2.400 <sup>a</sup>	1.150 <sup>bc</sup>	0.500 <sup>d</sup>	2.608±0.025	
Intestine(RS)	2.750 <sup>a</sup>	0.150 <sup>b</sup>	2.750 <sup>a</sup>	4.600 <sup>e</sup>	1.150 <sup>d</sup>	2.100 <sup>e</sup>	2.250±0.010	
Intestine(EU)	2.450 <sup>a</sup>	3.850 <sup>ab</sup>	3.600 <sup>bc</sup>	0.500 <sup>d</sup>	2.100 <sup>e</sup>	2.500 <sup>b</sup>	2.500±0.010	
Kidneys(RS)	5.300 <sup>a</sup>	6.450 <sup>b</sup>	6.750 <sup>c</sup>	1.350 <sup>a</sup>	2.550 <sup>cb</sup>	0.250 <sup>c</sup>	3.775±0.010	
Kidneys(EU)	4.200 <sup>a</sup>	4.850 <sup>b</sup>	6.400 <sup>abc</sup>	2.200 <sup>c</sup>	4.250 <sup>c</sup>	3.350 <sup>b</sup>	4.208±0.005	
Brain(RS)	0.550 <sup>a</sup>	0.750 <sup>b</sup>	0.850 <sup>c</sup>	1.750 <sup>d</sup>	0.650 <sup>e</sup>	2.650 <sup>d</sup>	1.200±0.010	
Brain(EU)	5.000 <sup>a</sup>	4.150 <sup>b</sup>	4.200 <sup>bc</sup>	1.600 <sup>a</sup>	2.650 <sup>b</sup>	3.750 <sup>c</sup>	3.558±0.005	
Bones(RS)	0.100 <sup>a</sup>	0.400 <sup>b</sup>	0.300 <sup>c</sup>	0.000 <sup>d</sup>	0.150 <sup>e</sup>	1.150 <sup>f</sup>	0.350±0.020	
Bones(EU)	4.300 <sup>a</sup>	4.650 <sup>b</sup>	4.400 <sup>c</sup>	0.650 <sup>ac</sup>	1.300 <sup>b</sup>	2.300 <sup>c</sup>	2.933±0.015	

Values in each row marked by the same superscript letter are not significantly different at P<0.05

**Table 8.** Correlation of cadmium concentration (mgL<sup>-1</sup>) in tissues of *Trachurus Murphyi* from RS fishing area

Parameter	Skin	Muscle	Gills	Liver	Intestine	Kidney	Brain	Bones
Skin	1							
Muscle	0.165	1						
Gills	0.305	-0.072	1					
Liver	-0.055	-0.668**	-0.050	1				
Intestine	-0.354	-0.484*	-0.097	0.153	1			
Kidney	-0.115	0.092	0.109	-0.243	0.399	1		
Brain	0.337	0.437	0.677**	-0.341	-0.229	0.571*	1	
Bones	0.075	-0.016	-0.204	0.627**	-0.079	-0.573*	-0.329	1

\*\*Correlation is significant at the 0.01 level (2-tailed)

\*Correlation is significant at the 0.05 level (2-tailed)

### Lead concentrations

Results in Table 4 presented the lead accumulations in the tissues of RS and EU fish species. The concentrations of lead in the tissues of RS fish species were ranged between the values of  $7.542\text{mgkg}^{-1}$  to  $10.825\text{mgkg}^{-1}$  shown for the muscles and kidneys tissues respectively. While in EU fish species, lead accumulations in the tissues were ranged between the values of  $17.200\text{mgkg}^{-1}$  to  $20.225\text{mgkg}^{-1}$  shown for in the bones and skin tissues respectively. The results of statistical analysis show that significant differences ( $p < 0.05$ ) for lead accumulation were recorded among batches of RS fish species and these are shown in the batches of gills, intestine, kidneys, bones and brain tissues respectively. However, non-significance differences ( $p < 0.05$ ) among batches of RS fish species were shown: for the skin tissues between batch 1, 2 and 3, 2 and 3; for the muscles tissues between batch 3 and 4; and for the liver tissues between batch 4 and 5. Whereas for the EU batches, the significant differences ( $p < 0.05$ ) for lead accumulations were shown in

the batches of brain tissues only. While the non-significant difference ( $p < 0.05$ ) among batches were shown: for the skin tissues between batch 1 and 3, 2, 4 and 5, 4 and 5; for the muscles tissues between batch 1 and 3, 2, 4 and 5, 4 and 5; for the gills tissues between batch 1 and 3, 2 and 4; for the liver tissues between batch 1, 2 and 4, 2 and 4, 4 and 5, 5 and 6; for the intestine tissues between batch 1, 3 and 4, 2 and 3, 4 and 5, 5 and 6; for the kidneys tissues between batch 1, 2, 3 and 4, 2, 5 and 6; and for the brain tissues between batch 1, 2, 4 and 5, 2 and 3, 3 and 4. The results recorded that, significant differences are predominantly shown among the batches of RS tissues (gills, intestine, kidneys, bones and brain) and hence, these imply that, there is impacts of different sources of activities that govern the contaminations of lead. On the other hand, the EU tissues recorded non-significant differences are dominantly shown across the entire batches and these also imply that, there is homogeneity in the sources that leads to lead contaminations across the batches.

**Table 9.** Correlation of cadmium concentration ( $\text{mgL}^{-1}$ ) in tissues of *Trachurus Murphyi* from EU fishing area

Parameter	Skin	Muscle	Gills	Liver	Intestine	Kidney	Brain	Bones
Skin	1							
Muscle	0.691**	1						
Gills	-0.014	0.134	1					
Liver	-0.316	-0.439	-0.779**	1				
Intestine	0.278	0.202	0.899**	-0.660**	1			
Kidney	-0.560*	-0.146	0.113	0.284	-0.104	1		
Brain	-0.769**	-0.284	0.470*	-0.252	0.070	0.652**	1	
Bones	0.459	0.240	0.064	0.155	0.446	-0.283	-0.634**	1

\*\*Correlation is significant at the 0.01 level (2-tailed)

\*Correlation is significant at the 0.05 level (2-tailed)

**Table 10.** Correlation of lead concentration ( $\text{mgL}^{-1}$ ) in tissues of *Trachurus Murphyi* from RS fishing area

Parameter	Skin	Muscle	Gills	Liver	Intestine	Kidney	Brain	Bones
Skin	1							
Muscle	0.809**	1						
Gills	0.818**	0.956**	1					
Liver	0.852**	0.885**	0.962**	1				
Intestine	0.837**	0.887**	0.978**	0.988**	1			
Kidney	0.864**	0.943**	0.994**	0.975**	0.988**	1		
Brain	0.891**	0.943**	0.988**	0.978**	0.983**	0.996**	1	
Bones	0.837**	0.907**	0.979**	0.936**	0.974**	0.983**	0.974**	1

\*\*Correlation is significant at the 0.01 level (2-tailed)

\*Correlation is significant at the 0.05 level (2-tailed)

**Table 11.** Correlation of lead concentration ( $\text{mgL}^{-1}$ ) in tissues of *Trachurus Murphyi* from EU fishing area

Parameter	Skin	Muscle	Gills	Liver	Intestine	Kidney	Brain	Bones
Skin	1							
Muscle	0.656**	1						
Gills	0.404	0.366	1					
Liver	-0.005	0.354	0.776**	1				
Intestine	0.356	0.400	0.393	0.398	1			
Kidney	-0.446	-0.551*	0.340	0.488*	0.328	1		
Brain	-0.674**	-0.177	0.333	0.733**	0.003	0.628**	1	
Bones	-0.491*	-0.079	0.560*	0.803**	0.185	0.661**	0.941**	1

\*\*Correlation is significant at the 0.01 level (2-tailed)

\*Correlation is significant at the 0.05 level (2-tailed)

**Table 12.** Correlation of mercury concentration ( $\text{mgL}^{-1}$ ) in tissues of *Trachurus Murphyi* from RS fishing area

Parameter	Skin	Muscle	Gills	Liver	Intestine	Kidney	Brain	Bones
Skin	1							
Muscle	0.823**	1						
Gills	0.981**	0.902**	1					
Liver	0.584*	0.348	0.593**	1				
Intestine	-0.123	0.082	-0.036	0.456	1			
Kidney	-0.424	-0.086	-0.330	0.078	0.882**	1		
Brain	-0.433	0.052	-0.292	-0.087	0.760**	0.936**	1	
Bones	-0.256	0.191	-0.138	-0.027	0.794**	0.939**	0.959**	1

\*\*Correlation is significant at the 0.01 level (2-tailed) \*Correlation is significant at the 0.05 level (2-tailed)

**Table 13.** Correlation of mercury concentration ( $\text{mgL}^{-1}$ ) in tissues of *Trachurus Murphyi* from EU fishing area

Parameter	Skin	Muscle	Gills	Liver	Intestine	Kidney	Brain	Bones
Skin	1							
Muscle	0.944**	1						
Gills	0.909**	0.851**	1					
Liver	0.572*	0.557*	0.387	1				
Intestine	0.519*	0.555*	0.555*	-0.363	1			
Kidney	0.299	0.209	0.037	0.859**	-0.581*	1		
Brain	-0.156	-0.223	-0.321	0.666**	-0.902**	0.876**	1	
Bones	-0.055	-0.095	-0.359	0.671**	-0.729**	0.914**	0.923**	1

\*\*Correlation is significant at the 0.01 level (2-tailed) \*Correlation is significant at the 0.05 level (2-tailed)

**Table 14.** Correlation of iron concentration ( $\text{mgL}^{-1}$ ) in tissues of *Trachurus Murphyi* from RS fishing area

Parameter	Skin	Muscle	Gills	Liver	Intestine	Kidney	Brain	Bones
Skin	1							
Muscle	0.085	1						
Gills	-0.283	-0.170	1					
Liver	-0.297	0.112	-0.242	1				
Intestine	-0.543*	-0.385	0.059	0.588*	1			
Kidney	-0.408	0.567*	0.182	-0.042	-0.478*	1		
Brain	0.461	0.545*	-0.626**	0.094	-0.011	-0.321	1	
Bones	0.253	-0.249	-0.699**	0.493*	0.516*	-0.746**	0.548*	1

\*\*Correlation is significant at the 0.01 level (2-tailed) \*Correlation is significant at the 0.05 level (2-tailed)

**Table 15.** Correlation of iron concentration ( $\text{mgL}^{-1}$ ) in tissues of *Trachurus Murphyi* from EU fishing area

Parameter	Skin	Muscle	Gills	Liver	Intestine	Kidney	Brain	Bones
Skin	1							
Muscle	0.638**	1						
Gills	0.635**	0.714**	1					
Liver	-0.207	0.010	0.399	1				
Intestine	-0.158	0.099	-0.580*	-0.319	1			
Kidney	-0.463	0.272	-0.280	-0.021	0.626**	1		
Brain	-0.186	-0.355	0.089	0.215	-0.425	-0.547*	1	
Bones	-0.029	0.198	-0.527*	-0.633**	0.911**	0.590**	-0.444	1

\*\*Correlation is significant at the 0.01 level (2-tailed) \*Correlation is significant at the 0.05 level (2-tailed)

**Table 16.** Correlation of nickel concentration ( $\text{mgL}^{-1}$ ) in tissues of *Trachurus Murphyi* from RS fishing area

Parameter	Skin	Muscle	Gills	Liver	Intestine	Kidney	Brain	Bones
Skin	1							
Muscle	0.215	1						
Gills	-0.374	0.435	1					
Liver	-0.030	0.369	0.785**	1				
Intestine	-0.275	0.372	0.906**	0.848**	1			
Kidney	-0.483*	-0.784**	-0.343	-0.419	-0.182	1		
Brain	0.534*	0.302	0.219	0.545*	0.121	-0.781**	1	
Bones	0.699**	-0.197	-0.452	-0.005	-0.467	-0.338	0.714**	1

\*\*Correlation is significant at the 0.01 level (2-tailed) \*Correlation is significant at the 0.05 level (2-tailed)

**Table 17.** Correlation of nickel concentration ( $\text{mgL}^{-1}$ ) in tissues of *Trachurus Murphyi* from EU fishing area

Parameter	Skin	Muscle	Gills	Liver	Intestine	Kidney	Brain	Bones
Skin	1							
Muscle	0.745**	1						
Gills	0.791**	0.796**	1					
Liver	0.949**	0.722**	0.839**	1				
Intestine	0.348	0.245	0.298	0.469*	1			
Kidney	0.386	0.078	0.478*	0.541*	0.782**	1		
Brain	0.242	0.421	0.462	0.473*	0.741**	0.602**	1	
Bones	0.583*	0.599**	0.629**	0.740**	0.842**	0.697**	0.897**	1

\*\*Correlation is significant at the 0.01 level (2-tailed) \*Correlation is significant at the 0.05 level (2-tailed)

The results of lead correlation analysis recorded for RS tissues in Table 9 shows that, significant ( $p < 0.01$ ) correlations were shown positively across the entire tissues. Hence, significance negative correlations were not shown. While in Table 10, the EU tissues show significant ( $p < 0.05$ ) correlations were shown positively between: skin versus muscles; gills versus liver and brain; liver versus kidneys, brain and bones; intestine versus brain; kidneys versus brain and bones; and brain versus bones tissues. However, the significant ( $p < 0.05$ ) negative correlations among tissues were shown between: skin versus liver, brain and bones; and muscles versus kidneys tissues. The existence of strong positive correlations shown among RS tissues indicates that lead contamination with respects to entire tissues are due to impact of similar sources of contamination or the uptake routes among the entire tissues portrayed by similar mechanisms. With referenced to results of statistical significance, lead accumulations across the batches of RS fish species were from different sources of non-related activities. However, the correlations results in other hand show that, the uptakes of lead are homogeneously shown across the entire tissues. While for EU tissues, the significant negative correlations recorded among other tissues entail that; contaminations with lead are related to contributions from different sources of activities. Therefore, the results obtained were in accordance with the results of statistical test of significance, since both agreed on the facts that lead contamination were due to impacts of different sources of activities taking place in the fishing zone.

With regards to results presented in Table 4, lead accumulations recorded for the tissues of RS and EU fish species were highly shown in the kidneys ( $10.825\text{mgkg}^{-1}$ ) and skin ( $20.225\text{mgkg}^{-1}$ ) tissues respectively. While, low lead accumulations were shown in the muscles ( $7.542\text{mgkg}^{-1}$ ) and bones ( $17.200\text{mgkg}^{-1}$ ) tissues of RS and EU fish species respectively. Kidney is an organ next to liver that plays a vital role in the process of digestion. Detoxifications of metals by the liver tissues mostly get accumulated in the kidneys [47]. The higher concentrations of lead shown in the kidneys than the liver tissues of RS fish species indicate that, liver under goes more detoxification process. This fact was in accordance with the study reported by Yilmaz *et al.* [14].

However, the low accumulation of lead shown with the muscles tissues was in agreement with studies of many authors, who reported that muscle tissues is not an active organ and of such cannot accumulates lead as it does with the

active tissues [22, 48, 49]. The in active part of muscles tissues was as a results low morphological mechanism of lead transport. Another fact is that fish muscles tissues due not taking part in the digestion process. In fish, intestine, liver and kidneys are regarded as organs that are involve in the digestion process and hence, these parts are referred to as target active organs where bioaccumulation of lead can takes place in the presence of mucus and metallo-thionien proteins. In other point of view, skin and gills tissues were shown to accumulate high level of lead. This is because these parts were in direct contact with the lead metal present in the water body. This fact also was in lined with the fish studies as found by many authors, who noticed that, accumulations of lead in the skin were due to direct exposure with the water body [50-52]. Therefore, level of lead concentration in the surrounding fish habitat. Fish bones were not an active tissue and hence cannot accumulate lead to a greater extent as does in the active tissues. Generally, the concentrations of lead shown with the entire tissues of the this study were higher than the results presented in studies on *Trachurus Murphyi* species by Ikem *et al.* [53] and Ogundiran *et al.* [54]. Also in this study, the lead concentrations were higher than the safety levels recommended by FAO [17].

### Mercury concentrations

The results presented in Table 5 shows that, mercury accumulations among the tissues of RS fish species were range between the values of  $102.95\text{mgkg}^{-1}$  to  $115.21\text{mgkg}^{-1}$  shown for the muscles and liver tissues respectively. While accumulation in the tissues of EU fish species were range between the values of  $78.717\text{mgkg}^{-1}$  to  $86.483\text{mgkg}^{-1}$  shown for the gills and intestine tissues respectively. Results of statistical analysis shows that significance differences ( $p < 0.05$ ) with regards to accumulation of mercury among the batches of RS fish species are shown in the brain tissues only. However, non-significance differences ( $p < 0.05$ ) recorded among the batches of RS tissues were shown: for the skin tissues between batch 1, 2 and 6, 2 and 3, 3 and 4, 4 and 5, 5 and 6; for the muscles tissues between batch 1, 2 and 6, 2 and 4, and 6; for the gills tissues between batch 1, 2 and 6, 2 and 3, 3 and 4, 4 and 5; for the liver tissues between batch 1, 3 and 6, 2 and 4, 3 and 5, 4 and 6; for the intestine tissues between batch 1, 2 and 3, 2 and 3, 3 and 4, 4 and 5, 5 and 8; for the kidneys tissues between batch 2 and 3, 4 and 5; and for the bones tissues between batch 4 and 5 only. While for EU

tissues, significance differences were not shown in any of the batches. Hence, non-significance recorded among the batches of EU tissues were shown; for the skin tissues between batch 1, 3 and 6, 2, 4 and 5, 3 and 5, 5 and 6; for the muscles tissues between batch 1, 3 and 6, 2, 5 and 6; for the gills tissues between batch 1, 2 and 6, 2, 4 and 6, 3 and 5, 4 and 6; for the liver tissues between batch 1, 3 and 6, 2, 3 and 6, 3 and 4, 4 and 6; in the intestine tissues between batch 1, 4 and 5, 2, 3 and 6, 3 and 6, 4 and 5; for the kidneys tissues between batch 1, 2 and 6, 2 and 3, 3 and 6; for the brain tissues between batch 1, 2 and 4, 2 and 3, 3 and 4, 5 and 6; and for the bones tissues between batch 1, 2 and 4, 2 and 3, 3 and 6, 4 and 6. Except with the batches of brain tissues for RS fish species, the entire batches (with respects to both RS and EU fish species) show non-significant differences and hence, these indicate that, contaminations with mercury across the entire batches were due to impacts of similar sources of activities.

The results in Table 12 show that, the correlation of mercury concentrations in the tissues of RS fish species recorded significant ( $p < 0.05$ ) positive correlation were shown across the entire tissues. While in Table 13, the EU tissues also show significant ( $p < 0.05$ ) positive correlation were shown across the entire tissues. However, significant negative correlations were not shown in any of the RS and EU tissues. Therefore, the results of correlation analysis obtained, (with regards to both RS and EU tissues) were in agreement with the results of statistical analysis, in which both analyses reported that mercury contaminations were as a result of similar sources of contaminations. These justifies that the route of tissues uptake and accumulations of mercury across the entire batches are homogeneously contaminated or the uptake mechanisms were governed by similar physico-chemical factors.

The concentrations of mercury recorded for the tissues of RS fish species indicate that, higher accumulations were shown in the liver tissues ( $115.21 \text{mgkg}^{-1}$ ) and lowest accumulations were shown in the muscles tissues ( $102.95 \text{mgkg}^{-1}$ ). While for the EU tissues, intestine exhibit higher concentrations ( $86.483 \text{mgkg}^{-1}$ ) and lowest accumulations were shown in the gills tissues ( $78.717 \text{mgkg}^{-1}$ ). The routes by which mercury gets into the fish system are through ingestion by mouth or absorption via skin or gills from the surrounding water. Fish ingest contaminated mercuric food, flown through gastrointestinal tract, absorbed and distributed by the liver [55]. The high levels of mercury shown in the liver tissues of RS fish species were attributed to the high coordination of mercury with the metallo-thionein protein present in the liver. In addition, the liver is the principal organ responsible for the detoxification, transportation, and storage of toxic substances and it the active site of pathological effects induced by contamination [4]. However, lower levels of mercury in the muscles of RS fish species were as a result of its role as inactive tissues. This is because the more mercury became accumulated in the digestive tissues the low its mobility would be to the inactive tissues (such as muscles,

bones and brain). While for EU tissues, intestines were shown to accumulate mercury highly, due to its role in the digestion process. This fact was in agreement with the study by Zhen-bin [56] who reported that, the most active sites in fish heavy metals bio-accumulate includes the intestine tissues because of its role in the digestion process. The entry of mercury through the ingestion path by mouth, flows to the stomach and intestine where absorption can take place, then processed and distributes by the liver and exit through kidneys. These parts are referred to as target or active organs where mercury bioaccumulation is very high due to presence of binding mucus and metallo-thionein protein [56]. In contrast, gills tissues of EU fish species shows low accumulation of mercury. The fact that many studies reports that, gills tissues are good indicators of the mercury levels that were present in water body of the fish habitat [36, 57]. Hence low levels of mercury shown in the gills tissues, predicted mercury contaminations in EU fish origins are largely from the feeding sources of the fish. This was also found in line with the work of Kannan *et al.* and Etornyo *et al.* [59, 60]. However, the concentrations of mercury across other tissues were fairly uniform. This also shows that, contaminations with mercury were due to contribution impact of both natural and anthropogenic activities. Also, mean concentrations of mercury with respect to entire tissues of both RS and EU fish species were above the levels reported in the studies by Tulay *et al.*, Haluk *et al.* and Ekeanyanwu *et al.* [61-63] and as well as the safety limits recommended by FAO and FAO/WHO [17, 45].

### Iron concentrations

The results presented in Table 6 shows that, the iron concentrations recorded in the tissues of RS fish species were ranged between the values of  $48.417 \text{mgkg}^{-1}$  to  $818.05 \text{mgkg}^{-1}$  shown for the muscles and kidneys tissues respectively. While for EU fish species, the iron concentrations recorded were ranged between the values of  $17.708 \text{mgkg}^{-1}$  to  $431.84 \text{mgkg}^{-1}$  shown for the muscles and liver tissues respectively. The results of iron statistical analysis shows that significant differences ( $p < 0.05$ ) were shown among the batches of skin tissues only for RS fish species. The non-significance differences ( $p < 0.05$ ) recorded among the batches of RS fish species were shown: for the muscles tissues between batch 1 and 6, 2 and 3, 3 and 4; for the gills tissues between batch 2 and 3, 3 and 4, 4 and 6; for the liver tissues between batch 1 and 6, 2 and 3, 3 and 6, 4 and 5; for the intestine tissues between batch 2, 3, 4 and 5, 4, 5 and 6; for the kidneys tissues between batch 1, 4 and 6, 2, 5 and 6, 3, 4 and 5; for the brain tissues between batch 1, 2 and 6, 2 and 3, 3 and 6; for the bones tissues between batch 1, and 2, 3, 4 and 6, 5 and 6. While for EU fish species, significant differences was not shown with any of the batches and hence the non-significance differences ( $p < 0.05$ ) recorded among the batches were shown: for the skin tissues between batch 1, 3 and 6, 2 and 4, 3 and 5, 5 and 6; for the muscles tissues between batch 3 and 4, 4, 5 and 6; for the gills tissues between batch 1, 4 and 5, 2 and 3, 5 and 6; for the liver

tissues between batch 1, 4 and 5, 2, 3 and 6, 3 and 6, 4 and 5; in the intestine tissues between batch 1, 2 and 3, 2 and 5, 3 and 6, 4, 5 and 6; for the kidneys tissues between batch 1, 3 and 4, 2, 3 and 5, 5 and 6; in the brain tissues between batch 1, 2 and 6, 3, 4 and 5, 4 and 5; and in the bones tissues between batch 1 and 2, 2 and 3, 4, 5 and 6. The significant difference existing between the batches of skin tissues RS fish species were due to differences in the sources of iron contamination. Although, the rest of the batches shows non-significant differences in the accumulation of iron and this implies that the sources of iron are dominantly from similar sources of activities. Whereas for EU tissues, the entire batches presented non-significant differences in the accumulation of iron and these were as a results of homogeneous contaminations due to impacts of related sources of activities.

The results presented in Table 14 shows that, the correlations of iron concentrations in the tissues of RS fish species recorded significant ( $p < 0.05$ ) positively correlation were shown between muscles versus kidneys and brain tissues; liver versus bones tissues; intestine versus bones tissues; and brain versus bones tissues. However, significant ( $p < 0.05$ ) negative correlations were recorded between skin versus intestine tissues; gills versus brain and bones tissues; liver versus kidneys tissues; intestine versus kidney and brain tissues; and kidneys versus bones tissues. While the results of correlation analysis for EU tissues in Table 15 shows that significant ( $p < 0.05$ ) relations were recorded positively between skin versus muscles and gills tissues; muscles versus gills tissues; intestine versus bones tissues; and kidneys versus bones tissues. Also significant ( $p < 0.05$ ) negative correlation were shown between skin versus bones tissues; muscles versus liver tissues; gills versus intestine and bone tissues; liver versus kidneys and bones tissues; and kidneys versus brain tissues. Positive correlations among tissues (with respects to both fishing zones) indicate that the intakes of iron were from similar sources of contamination or the uptake pattern portray via similar mechanism due to physico-chemical condition of the fish habitat. While, negative correlation is an indication of the inverse conditions as with the case of positive correlations above.

Iron concentrations in Table 16 recorded that highly accumulation were shown in the kidneys ( $818.05\text{mgkg}^{-1}$ ) and liver ( $431.84\text{mgkg}^{-1}$ ) tissues for RS and EU fish species respectively. While, lower accumulations were shown in the muscles tissues of both RS ( $48.417\text{mgkg}^{-1}$ ) and EU ( $17.708\text{mgkg}^{-1}$ ) fish species. Liver as the principal organ for storage also stores iron to considerable higher levels in fish tissues. The higher level of iron in the liver relative to other tissues may be attributed to the high coordination of iron with the metallo-thionein protein [64]. Liver as the principal center for storage and distribution, it is also a major active organ of metals detoxification [4, 65]. A kidney is the receiving organ next to liver after metals detoxification. Hence, higher iron concentrations in the fish kidneys tissues were due to excess detoxification by the liver tissues. However, certain high concentrations of iron were shown to

accumulate in the gills and intestine tissues of the fish. The concentration of iron in the gills would fairly entail the level of iron present in the surrounding water of the fish. Because the gills tissues are directly in contact with the iron in the water body. Generally, the level of iron shown in the entire tissues with regards to both RS and EU fish species were higher than the safety limits recommended by FAO/WHO [45].

#### Nickel concentrations

Recorded in Table 7, the concentrations of nickel shown for the tissues of RS fish species were ranged between the values of  $0.350\text{mgkg}^{-1}$  to  $3.775\text{mgkg}^{-1}$  shown for the bones and kidneys tissues respective. While for EU fish species, the accumulations of nickel were ranged between the values of  $2.500\text{mgkg}^{-1}$  to  $4.208\text{mgkg}^{-1}$  shown for the intestine and kidneys tissues respectively. The results of statistical analysis shows that, significant differences ( $p < 0.05$ ) in the nickel accumulations were recorded among the batches of RS fish species and these are shown in the batches of skin, muscles, gills, liver and bones tissues. Hence, the non-significance differences ( $p < 0.05$ ) recorded among the batches of RS tissues were shown: for the intestine tissues between batch 1 and 3; for the kidneys tissues between batch 1 and 4, 2 and 5, 3, 5 and 6; and in the brain tissues between batch 4 and 6. While, for EU tissues, significant differences were not shown in any of the batches and hence, the non-significant differences ( $p < 0.05$ ) recorded among the batches were shown: for the skin between batch 1 and 3, 2 and 5, for the muscles between batch 1 and 3, 2 and 6, 3 and 4; for the gills between batch 1 and 4, 2 and 3, 3, 4 and 5, in the liver between batch 1 and 4, 2 and 5, 3 and 5; for the intestine between batch 1 and 2, 2, 3 and 6; for the kidneys between batch 1 and 3, 2, 3 and 6, 3, 4 and 5; for the brain between batch 1 and 4, 2, 3 and 5, 3 and 6; and for the bones between batch 1 and 4, 2 and 5, 3, 4 and 6. The results recorded that, significant differences are dominantly shown among the batches of RS tissues (skin, muscles, gills, liver and bones) and hence, these imply that, there is impacts of different sources of activities that govern the contaminations of lead. On the other hand, the EU tissues recorded non-significant differences are dominantly shown across the entire batches and these also imply that, there is homogeneity in the sources that leads to lead contaminations across the batches.

The result of correlations analysis presented in Table 17 for RS tissues shows that, significant ( $p < 0.05$ ) positive relations were recorded between: skin versus brain and bones; gills versus liver and intestine; liver versus intestine; and brain versus bones. While, significant ( $p < 0.05$ ) negative correlations shown among tissues were between: skin versus liver and kidneys tissues; muscles versus kidneys tissues; and kidneys versus brain tissues. Also, the correlations of EU tissues presented in Table 18 shows that, significant ( $p < 0.05$ ) positive relations recorded among tissues were between: skin versus muscles, gills, liver and bones tissues; muscles versus gills, liver and bones tissues; gills versus liver, kidneys and bones tissues; liver versus intestine, kidneys, brain and bones

tissues; intestine versus kidneys, brain and bones tissues; kidneys versus brain and bones tissues; and brain versus bones tissues. However, significant inverse correlations were not recorded in the EU tissues. Differences in the nickel accumulation were shown among the tissues of RS fish species. These entails that positive correlations among some tissues implies, the uptake routes of nickel were from similar sources of contaminations or the accumulation pattern portrayed via similar mechanisms due to physico-chemical condition of the surrounding fish habitat. While, negative correlations among the other tissues indicate that, the uptakes of nickel are from non-related contaminations sources. In lined with the results of statistical significance, the results of correlation analysis also profound nickel contaminations are strongly from the impacts of non-related sources. While the correlation results for EU tissues shows the entire tissues shows significant positive correlations and these indicates that nickel uptake among tissues were dominantly from similar sources of contaminations. Therefore, the results obtained were in accordance with the results of statistical test of significance, since both agreed on the facts that nickel contamination were due to impacts of similar sources of activities taking place in the fishing zone.

The concentrations presented in Table 16 shows that, nickel accumulation were highly shown in the kidneys tissues as with regards to both RS ( $3.775\text{mgkg}^{-1}$ ) and EU ( $4.208\text{mgkg}^{-1}$ ) fish species. While the lowest levels of nickel accumulations were shown in the bones ( $0.350\text{mgkg}^{-1}$ ) and intestine ( $2.500\text{mgkg}^{-1}$ ) tissues of RS and EU fish species respectively. Kidney is the major gateway of heavy metal

detoxification in the body system of fish. In kidney tissues, despite its role as outlet of metals, considerable amounts of heavy metals were accumulated due to high coordination with metallo-thionein protein present [66]. The high concentrations of nickel accumulation shown in the kidneys relative to other tissues (of both RS and EU fish species) however, this study also compromised with the studies by Sahar *et al.* [66] who reported that nickel analysis in *P. Indicus* and *P. Argenteus* fish species were highly accumulated in the kidney tissues and Shaikh, [67] also reported *Cirrhina Mrigala* fish species show nickel accumulations were highly shown in the kidney tissues. In considering the other tissues, certain high levels of nickel were accumulated in the skin, gills, liver and brain tissues (with respects to both groups of fish species). The gills perform the function of respiration and are directly in contact with the nickel present in the water body. Thus, concentrations of nickel shown with the gills tissues can also reflects the concentration of nickel present in the water body where the fish lives [4, 66].

In contrast, EU tissues show lowest nickel accumulations were shown in the intestine tissues. This would be as a result of high detoxification process that took place within the intestine. While, lower nickel concentrations in the bones were due to the facts bones are not an active tissues and hence this was agree with the findings of many authors [67, 68]. The mean concentrations of nickel shown with the entire tissues with respects to both RS and EU fish species were above the safety limit stated by WHO [46].

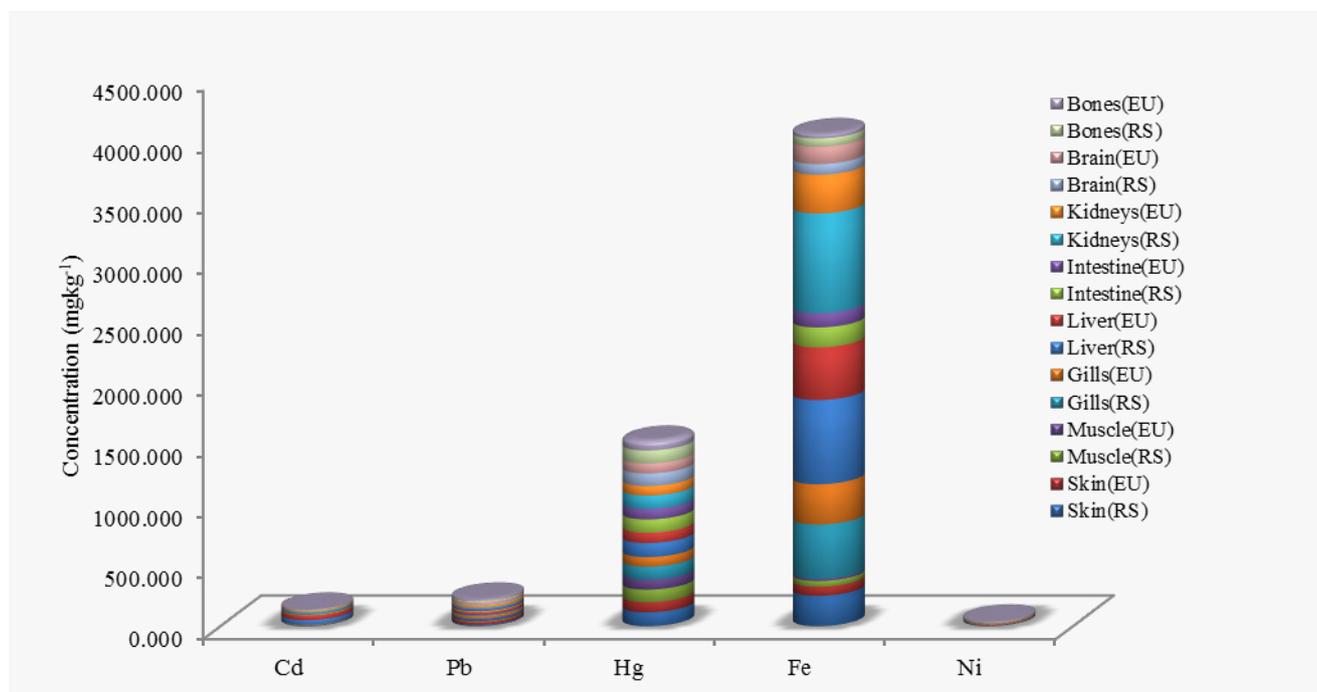


Figure 1. Mean concentrations of metals in tissues of *Trachurus Murphyi* species

**Table 18.** Individuals HQ, DIM and HRI response for heavy metals in muscle tissues of *Trachurus Murphyi* species

Heavy metal	Range (for 12 batches)	Mean±SD	Individuals (Category)	HQ	DIM	HRI
Cd	1.050 - 2.900	1.788±0.020	Adults (19yrs and above)	2.554E+0	7.595E-4	0.759E+0
			Children (7– 18yrs)	3.179E+0	9.504E-4	0.950E+0
			Children (1 – 6yrs)	9.442E+0	1.688E-2	1.688E+1
Pb	0.300-19.600	12.830±0.020	Adults (19yrs and above)	5.237E+0	5.480E-3	1.566E+0
			Children (6 – 18yrs)	6.517E+0	6.820E-3	1.989E+0
			Children (1 – 6yrs)	1.158E+2	1.211E-1	3.461E+1
Hg	66.50-117.85	92.067±0.015	Adults (19yrs and above)	1.315E+3	3.932E-2	3.933E+2
			Children (7– 18yrs)	1.636E+3	4.893E-2	4.894E+2
			Children (1 – 6yrs)	2.907E+4	8.693E-1	8.693E+3
Fe	10.90-60.250	33.063±0.043	Adults (19yrs and above)	0.675E-1	1.412E-2	0.202E-1
			Children (7– 18yrs)	0.840E-1	1.757E-2	0.251E-1
			Children (1 – 6yrs)	1.492E+0	3.122E-1	0.446E-0
Ni	0.300 - 4.100	1.884±0.010	Adults (19yrs and above)	0.135E+0	8.047E-4	0.402E-1
			Children (7– 18yrs)	0.168E+0	9.993E-4	0.499E-1
			Children (1 – 6yrs)	2.995E+0	1.779E-2	0.889E+0

yrs = age of years

### Profile of Studied metals

The profile of heavy metals; cadmium, lead, mercury, iron and nickel (Cd, Pb, Hg, Fe and Ni) in the analyzed tissues of *Trachurus Murphyi* fish species were summarized in Figure 1. The trends in the accumulation pattern of the studied metals with respects to the entire the tissues were in this order: **Fe > Hg > Pb > Cd > Ni**. The concentrations of iron shown with the entire tissues were extremely higher, compared to the concentrations of the other analyzed metals. These high levels of iron were due to contributions from both natural and anthropogenic activities which has high impacts on the water body. Natural activities such as meteor rays deposition, is an example of frequent occurrence in the Mediterranean and Russian ocean that can contributes to level of iron concentrations. Iron is an essential trace element required by all forms of life. The effects of toxic doses of iron in animal studies are characterized by initial depression, coma, convulsion, respiratory failure and cardiac arrest. Post-mortem examination reveals adverse effects on the gastrointestinal track with excess iron intake may result in siderosis (deposition of iron in tissue) in liver, pancreas, adrenals, thyroid, pituitary and heart [69]. These are characterized by initial depression, coma, convulsion, respiratory failure and cardiac arrest.

Mercury is the second metal that was shown to accumulate highly in the tissues of *Trachurus Murphyi* fish species. Mercury, as a non-essential element, is not expected to have its uptake/elimination actively regulated and subsequently its tissue concentrations can vary in a wide range, reflecting exposure to environmental levels and feeding behavior [70]. Hence, mercury body burdens in bio-indicator species provide sensitive indications of aquatic pollution as well as the potential impact in human health [59, 71]. Mercury together with its counter cadmium and lead were regarded as bed rock for most form of cancer because of their mutagenic

properties [72]. Excess nickel exposures at high concentrations were associated with lung and nasal cancers [73]. High levels of heavy metals in the tissues of fish were studied with regards to selected batches and the fishing areas, which imply that natural and anthropogenic human activities were reached in their destinations. Occurrences nearby water bodies such as volcanoes, deposition of meteor rays, weathering and erosion, as well as human activities such as industrial, agricultural, domestic, mining, testing of hazardous substances from military facilities, vessels spills, chemical destruction in Mediterranean seas, oceanic surge may render high concentration of these heavy metals in the water body. In this study, the levels of the studied heavy metal varied significantly in different tissues of the *Trachurus Murphyi* fish and their concentration in all the tissues were considerably higher than the safety limits recommended by WHO [45] irrespective of the batch and fishing area.

### Risk Assessment

The risk assessment of heavy metals; cadmium, lead, mercury, iron and nickel (Cd, Pb, Hg, Fe and Ni) concentrations ( $\text{mgkg}^{-1}$ ) in the muscle tissues of *Trachurus Murphyi* across the batches of both RS and EU fish species; including ranges, average means, standard deviations, individuals variations (first category; adult of aged 19years and above, second category; children of aged 7 – 18years and last category; children of aged 1 - 6years), health quotient (HQ), daily intake of metal (DIM) and health risk index (HRI) are summarized in Tables 18.

The mean concentration of cadmium in the muscles tissues of *Trachurus Murphyi* across the twelve (12) batches (of both RS and EU fishing zones) was given as  $1.788\text{mgkg}^{-1}$ . The assessments of individual's variation were based on average body weight relative to ranges of age groups. The results of HQ analysis for the entire categories show a value

of greater than one (1) with each of them. These imply that, the entire population would experience high dose of cadmium. The results of DIM shown for individual's daily loading of cadmium were given as  $7.595 \times 10^{-4}$ ,  $9.504 \times 10^{-4}$  and  $1.688 \times 10^{-2}$ . These correspond to HRI with ratio values of less than one (1) shown for the first and second categories respectively. While a ratio values of greater than one (1) is shown with the last category. These indicate that only the last category would expose to hazard of cadmium with the consumption of fish muscles as specified in this study.

The mean concentration of lead in the muscles tissues of *Trachurus Murphyi* across twelve (12) batches was given as  $12.830 \text{mgkg}^{-1}$ . The results of HQ analysis recorded that a ratio values of greater than one (1) were shown with the entire categories. These imply that, the population of individuals would expose to health hazard of lead metal based on consumption of fish muscles as recommended in this study. The results of DIM shown for individual's daily loading of lead were given as  $5.480 \times 10^{-3}$ ,  $6.820 \times 10^{-3}$  and  $8.693 \times 10^{-1}$ . These correspond to HRI with ratio values of greater than one (1), shown with each of the individual categories. This indicates that the categories of the entire individuals would expose to hazard of lead metals in the later live based on the consumption of *Trachurus Murphyi* fish muscles as recommended in this study.

The mean concentration of mercury was found to be  $92.067 \text{mgkg}^{-1}$  across the batches of both RS and EU fishing zones. The result of HQ analysis shows that ratio values of greater than one (1) were shown with each of the individual's categories. The results of DIM recorded for individuals daily loading of mercury were given as  $3.932 \times 10^{-2}$ ,  $4.893 \times 10^{-2}$  and  $8.693 \times 10^{-1}$ . These correspond to HRI with ratio values of greater than one (1), shown with each of the individual's categories. Since the results of HQ and HRI presented the ratio values were shown above one (1), hence, this indicates that the entire categories of individuals would expose to hazard of mercury metals in the latter live with the consumptions of *Trachurus Murphyi* fish muscles as recommended in this study.

The mean concentration of iron in the muscles tissues of *Trachurus Murphyi* across twelve (12) batches was given as  $33.063 \text{mgkg}^{-1}$ . The results of HQ analysis show that a ratio value of less than one (1) was shown with each of the first and second categories. While a ratio values of greater than one (1) was shown for the last category. These imply that, the population of first and second category and would not experience high dose of iron concentrations with the consumption of fish muscles. Whereas, the populations of last category, would expose to high dose of iron, based on the consumption of fish. The results of DIM shown for individual's daily loading of cadmium were given as  $1.412 \times 10^{-2}$ ,  $1.757 \times 10^{-2}$  and  $3.122 \times 10^{-4}$ . These correspond to HRI values of less than one (1) shown with the entire categories and hence the entire individual's population would benefit with iron nutrient with the consumption of fish as recommended for daily intake in this study.

The mean concentration of nickel in the muscles tissues of *Trachurus Murphyi* fish species across the twelve (12) batches was given as  $1.884 \text{mgkg}^{-1}$ . The results of HQ analysis show that a ratio value of less than one (1) was recorded for the first and second categories. The last category shows a ratio value of greater than one (1). These imply that, only the population of last category would experience high exposure of nickel with consumption as specified in this study. The results of DIM recorded for individual's daily loading of nickel were given as  $8.047 \times 10^{-4}$ ,  $9.993 \times 10^{-4}$  and  $1.779 \times 10^{-2}$ . These correspond to HRI with the ratio values of less than one (1) shown with the entire categories. These also indicate that the entire categories would not expose to high dose of nickel.

Finally, the health quotient (HQ) results shown in Table 18 revealed that the population of fish consumers in Zaria metropolis would expose to high loading dose of heavy metals; cadmium, lead and mercury at all levels with the consumption of *Trachurus Murphyi* fish muscles. In contrast, only the groups of children would experience the impact of iron and nickel. Whereas, for individual health risk consideration, the index results proved that, the ratios of heavy metals; lead and mercury were above the value of one (1) and these justify an index that in the latter live these metal would pose a serious health hazards.

## 4. Conclusions

The health status of human with respect to contamination by heavy metals (Cd, Pb, Hg, Fe and Ni) in the imported frozen fish *Trachurus Murphyi* species sold in Zaria metropolis, Nigeria was evaluated in this study. Based on the analysis obtained, the risk for human exposure to heavy metal contamination through fish consumption was significant. Since the levels of the studied heavy metals in all the analyzed tissues were above their corresponding permissible limits recommended by FAO [17, FAO/WHO [45] and WHO [46]. The population health risk from consumption of fish muscle tissues also, shows a chance of exposure were higher for lead and mercury than cadmium and less with iron and nickel. However, individuals consuming fish livers, intestine and kidneys may face considerable risk from ingestion of toxic metals at unacceptable concentrations. Finally, this work may provide valuable database for continuing research on the import of frozen fish in Nigeria.

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