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COMPARATIVE EVALUATION OF SEASONAL DYNAMICS OF HEAVY METALS IN *Nematopalaemon hastatus* AND *Farfantepenaeus notialis* IN A TROPICAL BRACKISH ENVIRONMENT

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Abstract
The distribution and concentration of Cadmium, Copper, Iron, Lead, Manganese and Zinc in the cephalothorax and abdomen of two species of shrimps (Nematopalaemon hastatus and Farfantepenaeus notialis) in coastal waters of Ondo State, Nigeria were investigated from April, 2014 to March, 2016 using Atomic Absorption Spectrophotometer. The metal bioaccumulation level in the shrimps was in the order; Zn > Fe > Mn > Cu > Pb > Cd. The highest concentration of Zn (7.519±0.259mg/kg) was recorded in abdomen of N. hastatus in the wet season, while the lowest value 5.485±0.494mg/kg was recorded in the abdomen of F. notialis in the wet season. The highest level of Fe (6.539±0.372mg/kg) was recorded in cephalothorax of F. notialis in dry season. Mn ranged between 4.305±0.616mg/kg in abdomen of F. notialis in the wet season and 2.388±0.356mg/kg in cephalothorax of F. notialis in dry season. The result showed that Zn and Mn exhibited seasonal variation (P<0.05) in N. hastatus while Fe and Mn showed seasonal variation (P<0.05) in F. notialis. The study also revealed that the value of Fe and Mn in the shrimps exceeded the 0.5mg/kg limit recommended by FEPA. Therefore, evaluation of metal pollution of the study area is suggested to minimize the health risk of the population that depends on it for water and fish supply.

Key Words: *Coastal Water, Cephalothorax, Abdomen, Heavy metal pollution, Shrimps*

Introduction

Over the recent decades, there has been growing concern in increasing levels of heavy metal in the marine environment and this has attention of scientist to extensive research on measurement of contamination levels in public food supply, particularly fish (El-Moselhy *et al.,* 2014). The investigation of heavy metals concentration in shrimps is quite essential with respect to human consumption and in management of aquatic resources; resulting from heavy metals released into aquatic ecosystems by anthropogenic activities. The contaminated aquatic organisms may eventually end up in the food chain threatening the health of human who consume such sea foods, resulting in deleterious effect like renal failure, liver damage, cardiovascular diseases, etc. (Bawuro *et al.,* 2018).

The coastal territory of Ondo State, Nigeria is situated between the Lagos and Delta coasts. The zone has expanding home-grown and business exercises while the significant methods for transportation in the region is speedboat with softly substantial traffic of products and people. (Ajibare *et al*., 2017). Heavy metals have been reported to be predominant in used engine oils, fuels (lead) as well as in brake linings (copper), safety fences (zinc), etc. used in vehicles and motorboats (Elnabris *et al.,* 2013). These metals are released into the environment and are sources of pollution in the water system. Sewage from residence within the coast remains a significant wellsprings of contamination in coasts (Bawuro *et al.,* 2018). Agricultural activities have been accounted for to be significant source of contamination in the coastal regions.

According to Ajibare *et al.* (2017), *Nematopalaemon hastatus* as well as *Farfantepenaeus notialis* are prominent in coastal waters and estuaries while Chindah *et al.* (2004) reported that *F. notialis* have the propensity to bioaccumulate certain heavy metals in the environment than others and further suggested that the organism can be used as possible bioindicator for heavy metal pollution monitoring programme in the Niger Delta region. Olawusi-Peters *et al*. (2014) reported low condition factor for shrimps and ascribed it to pollution activities that occur in the area and therefore called for study of pollution status and/or anthropogenic activities to further analyse its sustainability of biodiversity.

The coastal waters of Ondo State in the southern Nigeria empties to the Atlantic Ocean and to some other parts of the country and it is known for sea foods (Ajibare *et al.*, 2017) which means that its contamination might have local and international health and ecological consequences. Thus, there is need to assess the level of contamination caused by human activities in Nigerian coastal waters and also examine the effect of these contamination to the aquatic life and ecosystem in general. The objective of this study was therefore to determine the level of Pb, Zn, Fe, Cd, Cu and Mn in the cephalothorax and abdomen of *N. hastatus* and *F. notialis* with the aim of ensuring the safety of the ecosystem and ascertain that accurate data can be obtained for the national inventory and for other programs to prevent, remediate and manage contaminations caused by human activities in the coastal waters of Ondo State.

Materials and Methods

Study Area

This research was conducted in the coastal area of Ondo State between April, 2014 and March, 2016. The study location is situated at the extreme southern part of Ondo State in Ilaje Local Government Area (Figure 1). Ilaje coastline in Nigeria (about 78km) with long history in fishing tracing all the way back to the pre-provincial days (Ajibare *et al.*, 2017). The zone is situated inside the central evergreen marsh backwoods with two significant seasons (Olawusi-Peters *et al*., 2014) The climate encounters reliably high temperatures (about 32°C) lasting through the year. Since temperature changes only marginally, rainfall distribution, over space and time, becomes the sole most vital factor (Ajibare *et al.*, 2017). For the research, Ayetoro (06°06' N 04°46' E), Idiogba (06°05' N; 04°47' E), Bijimi (06°04' N; 04°49' E), and Asumogha (06°03' N; 04°39' E) were deliberately chosen dependent on data for broad shrimp fishing exercises in the settlements. This zone is noted for ocean food varieties which are devoured inside and outside the state.

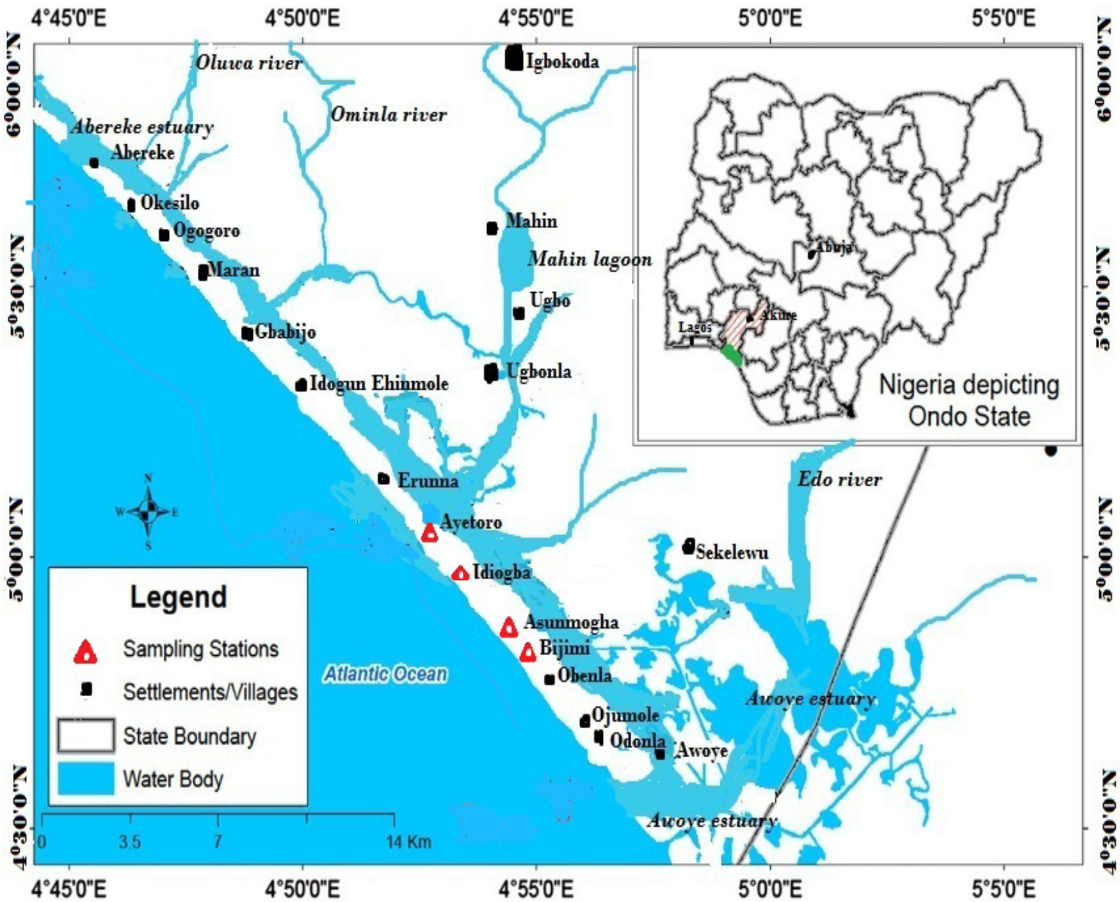

Fig. 1: Map of the Coastal Waters of Ondo State

Collection and Identification of Shrimps

Specimens were gathered month to month starting April, 2014 and ending in March, 2016 with the help of local fishermen. They were quickly preserved in ice and moved to the laboratory and were frozen at - 40°C prior to being utilized for the research. The shrimps were arranged and distinguished to species level utilizing the FAO Species Identification Sheets, (Volume VI) (FAO, 1981) and Powell (1982).

Determination of Heavy Metals

In the laboratory, the frozen shrimp samples were allowed to defrost slowly at room temperature. The shrimps were thereafter dissected with clean stainless steel instruments, separating the cephalothorax from the abdomen. The tissues (cephalothorax and abdomen) were oven dried at 60^{o}C for 48hours. Individual whole specimen was crushed to a uniform particle size and 0.2g of crushed weight was put in a 50ml digestion tube and 2.5ml of H_2SO_4/selenium mixture was added and placed in an aluminium block on a hot plate. This was heated 200°C until solution fumed. Each cylinder was taken out from the hot plate and permitted to cool for 10 minutes. 1ml of 30% H_2O_2 was added to each cylinder. After response died down, each cylinder was followed with an extra 2ml H_2O_2. Each cylinder was supplanted on hot plate and heated to 330°C until clear (typically for 2hrs). The yellow tint of the solution vanished as the digest was finished. The solution was poured into a centrifuge tube and made up to 30ml imprint with refined water. This was centrifuged at 3000rpm for 10mins. The supernatant was tapped into test vials for analysis. The heavy metal analysis was done according to standard methods for heavy metal determination (Novozamsky *et al.,* 1983) using ACCUSYS 211 Atomic Absorption Spectrophotometer. Levels of metals (Cd, Pb, Mn, Cu, Zn and Fe) were expressed in mg/kg dry weight.

Statistical Analysis

Multi-Variate Analysis of Variance (MANOVA) and Duncan multiple range test was used to evaluate the significant difference in the concentration of different parameters with respect to different sites. A probability level of less than 0.05 was considered significant. Descriptive analysis was also used to present tables and figures.

Results

Figure 1 shows that mean concentration of cadmium ranged from 0.001±0.000mg/kg in Ayetoro to 0.002mg/kg in Idiogba for both wet and dry seasons and there was no significant difference between the seasons in all the sampling stations. The concentration of lead in the abdomen of *N. hastatus* ranged from 0.021±0.002mg/kg in the wet season at Bijimi to 0.029±0.028mg/kg at Idiogba in both wet and dry seasons. Pb in the cephalothorax of *N. hastatus* varied from 0.015±0.003mg/kg at Asumogha, Ayetoro and Idiogba to 0.017±0.003mg/kg in Bijimi in the wet season while it ranged from 0.015±0.002mg/kg (Ayetoro) to 0.016±0.003mg/kg (Bijimi and Idiogba) in the dry season (Figure 2a). Pb in the abdomen of *F. notialis* varied from 0.018±0.004mg/kg (Idiogba) to 0.021±0.004mg/kg (Bijimi) in the wet seasons and from 0.016±0.005mg/kg (Ayetoro) to 0.021±0.003 (Asumogha) in the dry season as shown in figure 2b. The mean concentration of lead in the cephalothorax of *F. notialis* ranged from 0.016±0.006mg/kg in Idiogba to 0.020±0.030mg/kg in Asumogha in wet season and from 0.017±0.002mg/kg (Idiogba) to 0.022±0.029mg/kg (Asumogha) in the summer. The figure further reveals that the concentration of Pb in the two species shows no significant difference (P>0.05) between the wet and dry seasons in the four stations.

The concentration of manganese in the abdomen of *N. hastatus* collected from the study area ranged from 2.683±0.391mg/kg at Asumogha in the wet season to 3.312±0.303mg/kg at Idiogba also in the wet season. The concentration of Mn in the cephalothorax of *N. hastatus* ranged from 3.212±0.580mg/kg at Ayetoro to 3.550±0.542mg/kg at Bijimi in the rainy season and from 2.803±0.640mg/kg (Ayetoro) to 3.292±0.510mg/kg (Bijimi) in the dry season as shown in figure 3a. The concentration recorded in the dry season exhibited significant difference with the concentration of the wet seasons except in Idiogba where there was no significant difference at P>0.05. The concentration of Mn as recorded in abdomen of *F. notialis* in wet season ranged from 3.516±0.483mg/kg in Asumogha to 4.305±0.616mg/kg in Ayetoro, and from 3.425±0.434mg/kg (Asumogha) to 3.908±0.386mg/kg (Ayetoro) in the summer and there was important variance (P<0.05) amongst the two seasons. Mn recorded in the cephalothorax of *F. notialis* ranged from 2.822±0.564mg/kg in Asumogha to 3.083±0.391mg/kg in Ayetoro for wet season and from 2.388±0.356mg/kg (Asumogha) to 2.739±0.356mg/kg (Ayetoro) in dry season as shown in figure 3b and shows that the wet season was significantly different from the dry season (P<0.05) in all the four stations.

Concentrations of Heavy Metals

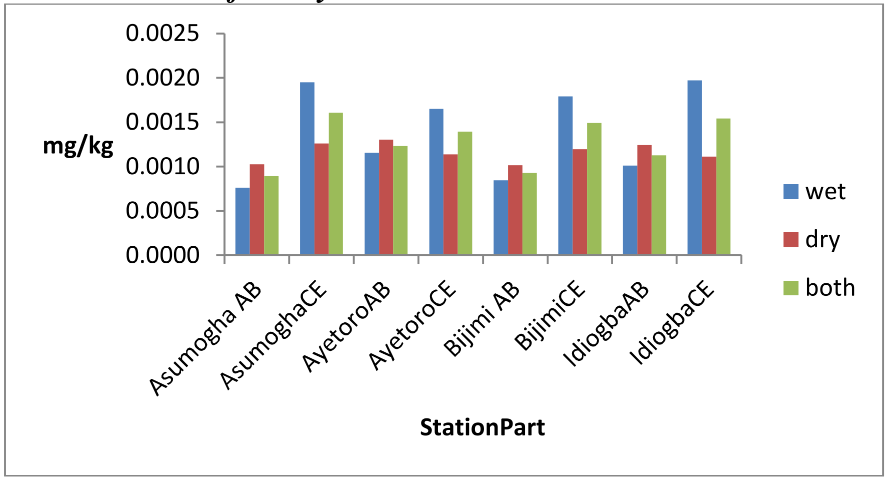

Fig. 1a: Concentration of Cd in *N. hastatus*

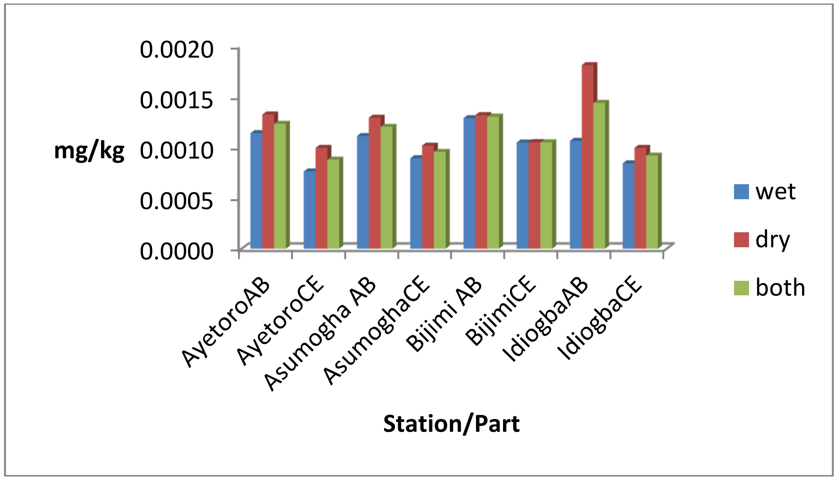

Fig. 1b: Concentration of Cd in *F. notialis*

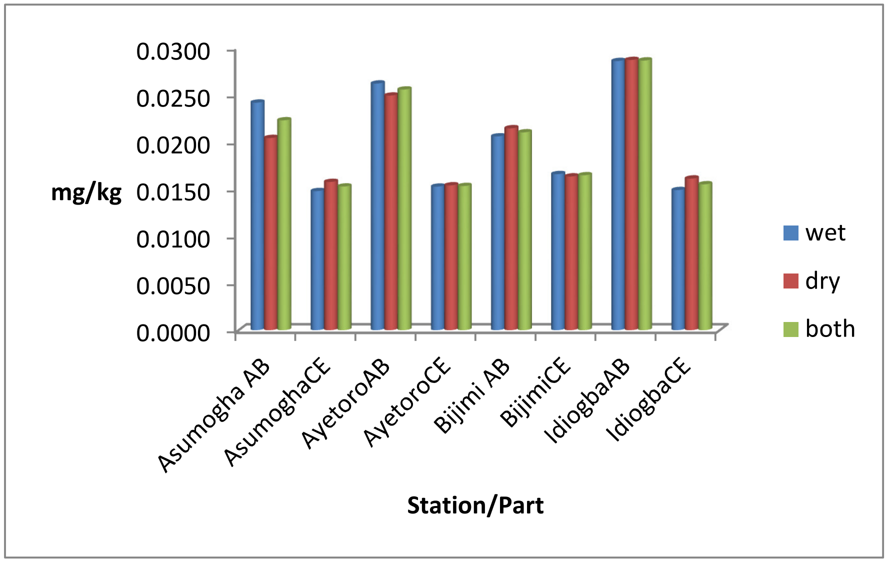

Fig. 2a: Concentration of Pb in *N. hastatus*

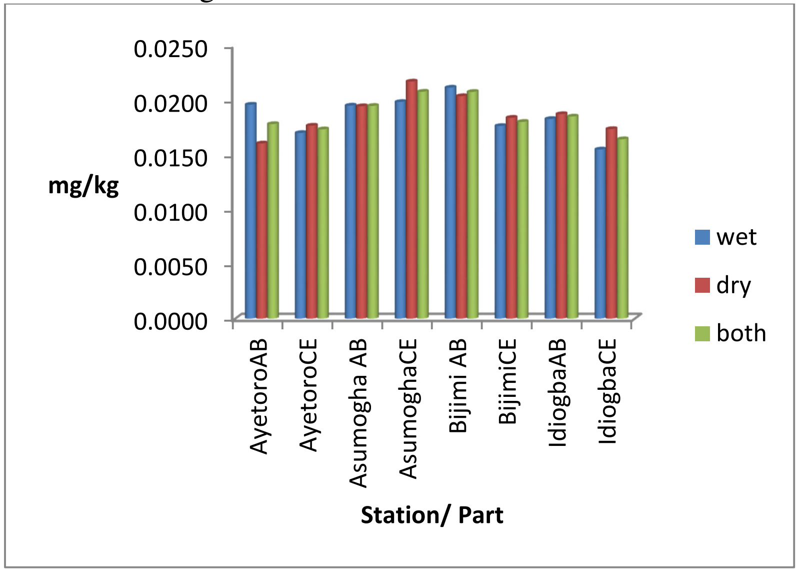

Fig. 2b: Concentration of Pb in *F. notialis*

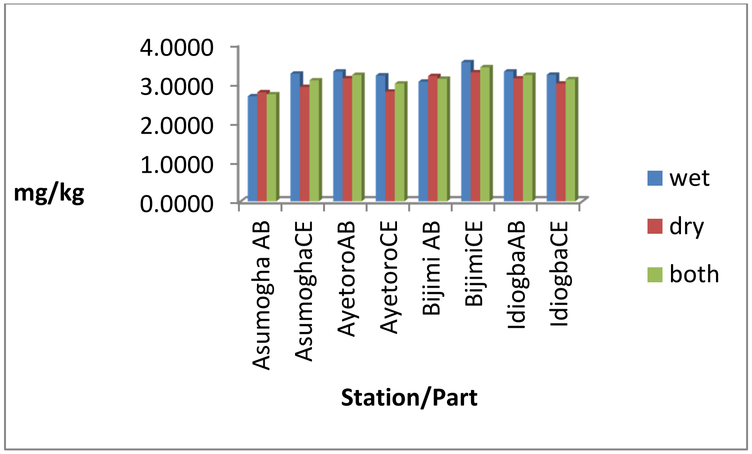

Fig. 3a: Concentration of Mn in *N. hastatus*

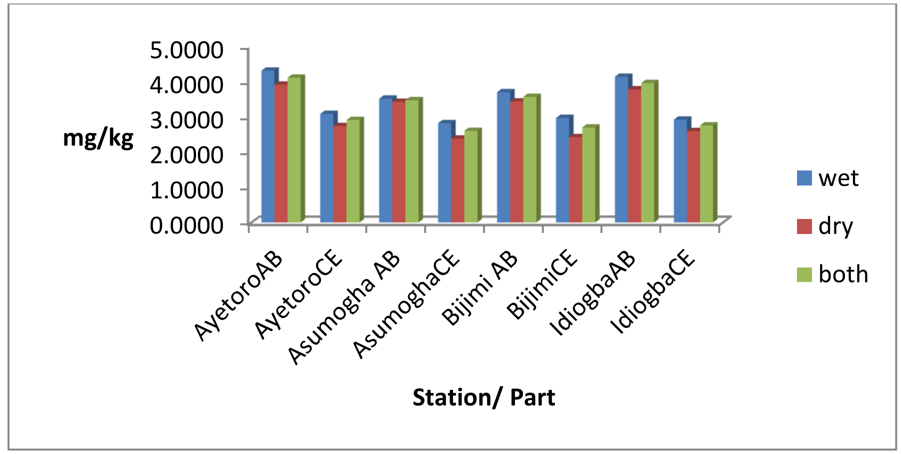

Fig. 3b: Concentration of Mn in *F. notialis*

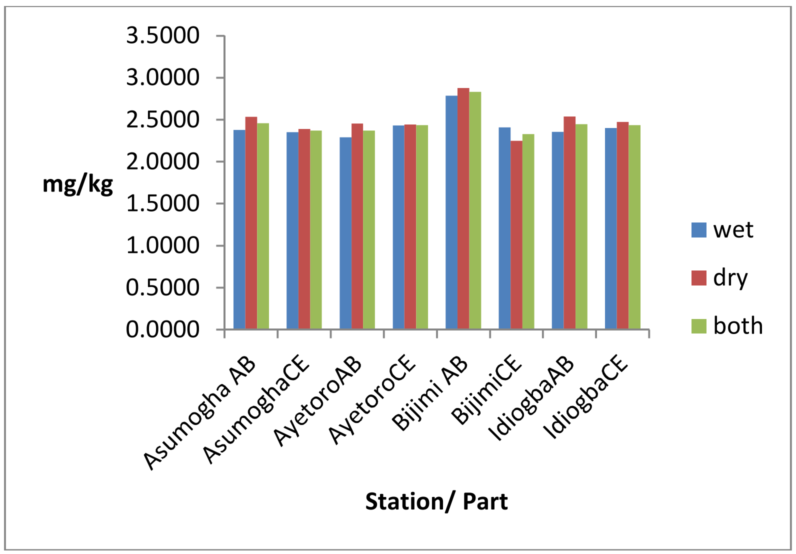

Fig. 4a: Concentration of Cu in *N. hastatus*

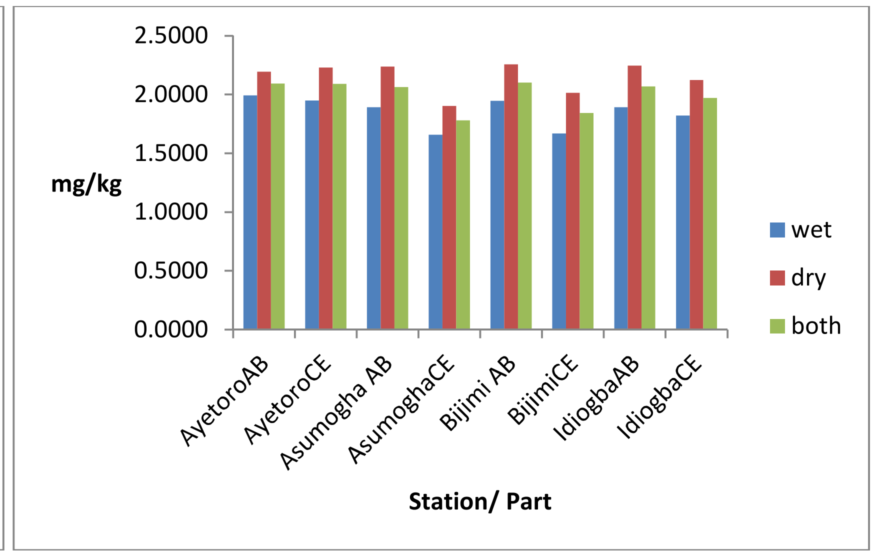

Fig. 4b: Concentration of Cu in *F. notialis*

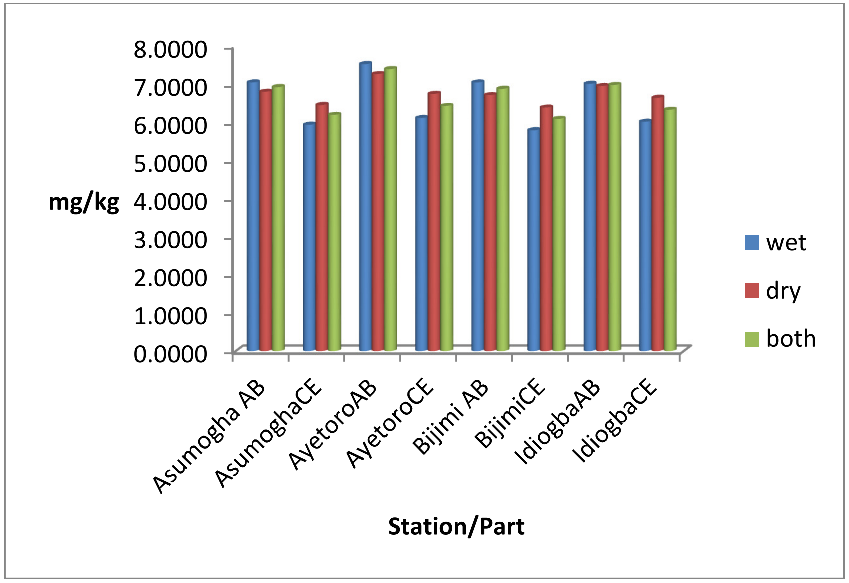

Fig. 5a: Concentration of Zn in *N. hastatus*

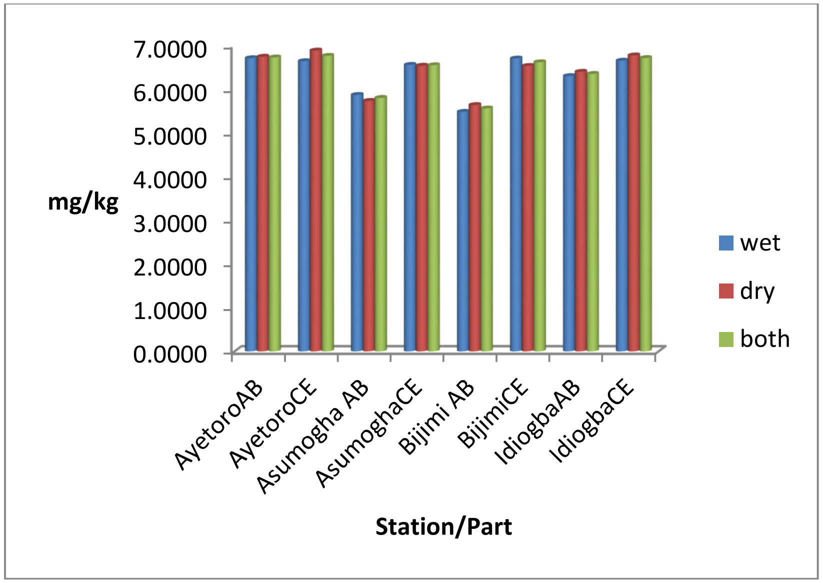

Fig. 5b: Concentration of Zn in *F. notialis*

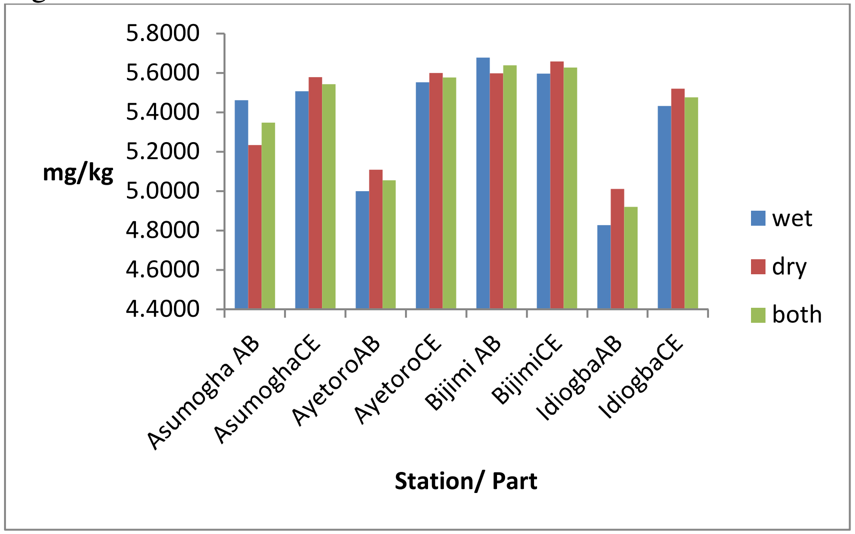

Fig. 6a: Concentration of Fe in *N. hastatus*

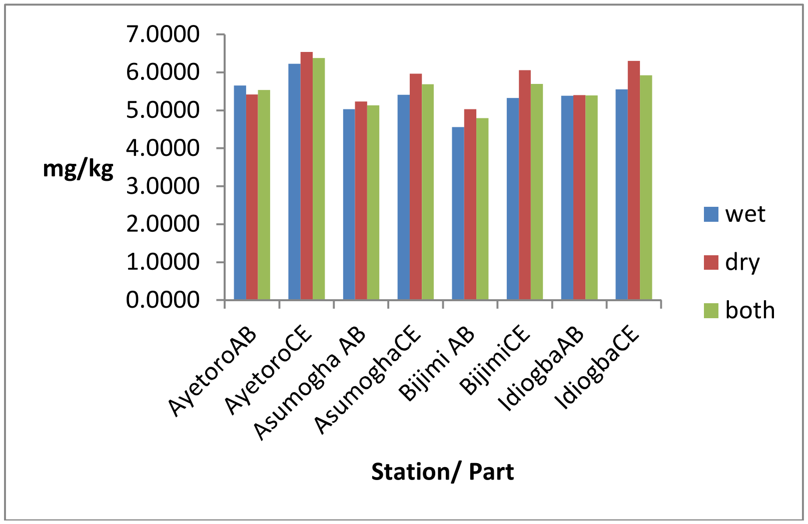

Fig. 6b: Concentration of Fe in *F. notialis*

Figure 4a shows that the mean concentration of copper in the abdomen of *N. hastatus* ranged from 2.292±0.397mg/kg at Ayetoro to 2.788±0.391mg/kg in the wet season and ranged from 2.453±0.442mg/kg (Ayetoro) to 2.878±0.358mg/kg (Bijimi) in the summer and there was no important variance among the wet and dry seasons. The concentration of Cu in the cephalothorax of *N. hastatus* ranged from 2.351±0.175mg/kg at Asumogha to 2.432±0.157mg/kg at Ayetoro in the wet season and from 2.250±0.249mg/kg (Bijimi) to 2.47±0.247mg/kg (Idiogba) in the dry season. The mean concentration of copper in the abdomen of *F. notialis* ranged from 1.892±0.449mg/kg in Idiogba to 1.993±0.390mg/kg (Ayetoro) in the rainy season and from 2.194±0.426mg/kg (Ayetoro) to 2.244±0.497mg/kg (Idiogba) in the dry season. Also, the concentration of Cu in the cephalothorax of *F. notialis* in the wet season varied from 1.658±0.288mg/kg in Asumogha to 1.949±0.350mg/kg in Ayetoro while it ranged from 1.903±0.341mg/kg (Asumogha) to 2.231±0.281mg/kg (Ayetoro) in the dry season (Figure 4b). The figure also reveals that the concentration of copper in *F. notialis* exhibited seasonal variation ($P<0.05$) in all the stations.

Concentration of Zn in the abdomen of *N. hastatus* ranged from 7.002±0.555mg/kg at Idiogba to 7.519±0.259mg/kg at Ayetoro in the rainy season whereas it varied from 6.706±0.418mg/kg (Bijimi) to 7.258±0.338mg/kg (Ayetoro) in dry season (Figure 5a). The figure also shows that there was significant difference between the wet and dry seasons. Zn in the cephalothorax of *N. hastatus* ranged from 5.786±0.639mg/kg (Bijimi) to 6.106±0.700mg/kg (Ayetoro) in the wet season and from 6.381±0.546mg/kg (Bijimi) to 6.739±0.475mg/kg (Ayetoro) in the dry season as shown in figure 5a. The figure shows that both the dry and wet seasons were statistically different from each other in all sampling stations at $P<0.05$. Zn in the abdomen of *F. notialis* ranged from 5.485±0.494mg/kg (Bijimi) to 6.714±0.562mg/kg (Ayetoro) in the wet season and from 5.642±0.454mg/kg (Bijimi) to 6.747±0.478mg/kg (Ayetoro) in the dry season as shown in figure 5b. The figure further reveals that there was no significant difference ($P>0.05$) between the dry and wet seasons in the four stations. Zinc in the cephalothorax of *F. notialis* ranged from 6.562±0.501mg/kg (Asumogha) to 6.706±0.498mg/kg (Bijimi) for the wet season and from 6.536±0.280mg/kg (Bijimi) to 6.889±0.389mg/kg (Ayetoro) in the dry season. The figure further shows that there was significant difference between the wet and dry season except in Ayetoro.

Figure 6a shows that the concentration of Fe in the abdomen of *N. hastatus* varied from 4.828±0.771mg/kg (Idiogba) to 5.678±0.488mg/kg (Bijimi) in the rainy season and between 5.011±0.182mg/kg (Idiogba) and 5.597±0.699mg/kg (Bijimi) in the summer. No significant difference amongst the wet and dry seasons was observed in the four stations. The mean concentration of iron in the cephalothorax of *N. hastatus* in the rainy season varied from 5.433±0.315mg/kg at Idiogba to 5.596±0.272mg/kg at Bijimi and from 5.519±0.312mg/kg (Idiogba) to 5.658±0.399mg/kg (Bijimi) in the summer. The figure also reveals that there was no significant difference between the seasons in the four stations.

The concentration of Iron in the abdomen of *F. notialis* in the rainy season varied from 4.555±0.879mg/kg (Bijimi) to 5.649±0.416mg/kg in

Ayetoro while it ranged from 5.031±0.631mg/kg (Bijimi) to 5.417±0.527 (Ayetoro) in the summer. There was significant difference ($P<0.05$) between the seasons in Ayetoro and Bijimi while there was no significant difference ($P>0.05$) between the seasons in Asumogha and Idiogba. Iron in the cephalothorax of *F. notialis* in the rainy season varied from 5.326±0.896mg/kg to 6.221±0.436mg/kg (Ayetoro) and from 5.967±0.741mg/kg (Asumogha) to 6.539±0.372mg/kg (Ayetoro) in the dry season (Figure 6b). The figure shows that there was significant difference ($P<0.05$) between the seasons in the four stations.

Discussion

This study revealed that all the heavy metals analysed were found in the shrimps. The metal bioaccumulation in the shrimps were in the decreasing order of Zn > Fe > Mn > Cu > Pb > Cd which was similar to the trend observed by Adedeji and Okocha (2011) in *M. macrobrachion* and *M. vollenhovenii* from Epe lagoon and Asejire River. Amongst all the metals analyzed, Mn, Zn and Fe were observed to have the highest concentrations in both shrimp samples. Moreover, the level of heavy metals recorded in shrimp samples in this study were generally low when compared to the WHO (2003) and FEPA (2003) recommendations. Also, in comparison with concentration levels in other areas, Adedeji and Okocha (2011) recorded higher metal levels in *M. macrobrachion* and *M. vollenhovenii* from Epe lagoon and Asejire river in southwest Nigeria, while Abulude *et al*. (2006), from coastal waters of Ondo state, Nigeria; Banjo *et al*. (2010) from different markets in southwest, Nigeria and Jimoh *et al.* (2011) from Epe lagoon recorded similarly higher concentrations.

The range of concentrations of Zn in the shrimps' parts were within the FAO guideline of 30 mg/kg and is low when compared to the zinc level reported by Adedeji and Okocha (2011), Jimoh *et al.* (2011) and Edward *et al.* (2013) in Epe Lagoon and Odo Ayo river respectively. The possible explanation for this could be the difference in fish species, sizes, ages, sampling stations and periods. Adeyeye (1996) reported that differences in metal concentrations in fish were a function of species, while Idodo-Umeh, (2002) reported that bigger fishes tend to accumulate higher concentrations of metals than smaller ones. Zinc exhibited seasonal variation in *N. hastatus* while there was no seasonal variation in *F. notialis* and this may be associated with the fact that Zinc are essential elements that are carefully regulated by physiological mechanisms in most organisms (Olusola and Festus, 2015).

Fe had the second highest concentrations in the two species examined in this study irrespective of the season, body part and station. Also, the results show that the concentration of Fe in the cephalothorax at individual station was numerically higher than the concentration in the corresponding abdomen and this is similar to the reports of Khalil and Faragallah, (2008) who observed higher concentration in the gill and argued that metals get adsorbed onto the gills surface as the first target or point of contact for pollutants in water. Khalil and Faragallah (2008) stated that high concentration of the metals is bioaccumulated in the gills due to element complexion with the mucus coverings in the gills which cannot be completely removed from the gill lamellae before analysis. In this study, the observed mean value of Fe in the shrimps' parts far exceeded the WHO/FEPA recommended limits of 0.5mg/kg in fish foods. Though Fe is an essential heavy metal, it has the tendency to become toxic to living organisms, even when exposure is low (Elnabris *et al.,* 2013).

The concentration of manganese (Mn) in the abdomen of *F. notialis* was higher than the levels in the cephalothorax but did not show a definite pattern in *N. hastatus*. This may be due to the different ways by which Mn is being bio-accumulated by aquatic organisms. Edward *et al.* (2013) stated that Mn is taken directly through the gills or indirectly from food and ingested sediment via gut and that under conditions of metal contamination, metals tend to deposit in the same organs where they may exert toxic effects. Mn levels in the abdomen and cephalothorax of the two studied species exceeded the 0.5mg/kg limit recommended by FEPA (2003) but was lower than the concentration observed by Adedeji and Okocha (2011) and Zodape (2014) in prawns and shrimps species collected from both Epe lagoon and Asejire River, Nigeria and Kolaba market of Mumbai (west coast) India respectively.

Seasonal variation was observed in the concentration of Fe and Mn and their concentrations were higher than the FAO/WHO permissible limit in both species across the study area. This may be due to anthropogenic activities such as washing, swimming, bathing, transportation and waste disposal which continuously increase the amount of heavy metals in the water body as supported by Giguere *et al.* (2004) who stated that metal accumulation in fish depends on pollution, and may differ for various species living in the same water body. El-Moselhy *et al.* (2014) reported that another principal factor that might explain the relatively high concentrations of Fe and Mn are also found in agricultural products such as fertilizers which may eventually accumulate in agricultural soils and become exposed to water bodies and the organisms present in them through run-offs especially during the rainy season. Also, Sivaperrumal *et al.* (2007) reported that Mn does not accumulate with age, thus increase in population, urbanization, industrialization and agriculture practices may be said to be responsible.

Copper (Cu) is one of the metals, which are essential to human health. The concentration of Cu exhibited seasonal variation in *F. notialis* but the seasons were not significantly different in *N. hastatus.* This supports the opinion of Sivaperrumal *et al.* (2007) who stated that the presence of Cu in the aquatic environment may be due to accumulation of domestic and agricultural wastes. Since the Cu recorded in the two shrimp species (irrespective of the part) was slightly below the FAO (1983) guideline of 3mg/kg, it can be said that fish captured from the study area will bioaccumulate Cu with time.

Pb is classified as one of the most toxic heavy metals. The biological effects of sub lethal concentrations of Pb include delayed embryonic development, suppressed reproduction and inhalation of growth, increased mucous formation, neurological problems, enzyme inhalation and kidney dysfunction (Akan *et al.,* 2012). The mean concentrations of Lead (Pb) in the two studied species exhibited no significant difference (P>0.05) between the abdomen and cephalothorax across the four stations although the concentration of Pb in the abdomen of *N. hastatus* was higher than the concentration in the cephalothorax in all the four stations and there was no seasonal variation in both species. The concentration of Pb observed in the shrimps from this study was within the permissible limit of 2mg/kg of FAO (1983).

Cadmium (Cd) like any other substance could be absorbed via the gills and has been known to cause damage to fish gills (Giguere *et al.,* 2004). Cd levels recorded in shrimps' samples from the study site were low when compared to WHO (2003) and FEPA (2003) maximum permissible limit of 0.5mg/kg in fish food. Also, the non-seasonal variation observed in both species as well as the concentrations of Cd in all the shrimp samples was low in comparison to the report of Idodo-Umeh (2002) in fishes of Olomoro water bodies.

Conclusion

It is concluded based on the findings of this study that the six investigated metals (Pb, Cd, Cu, Fe, Mn and Zn) were found to be bio-concentrated in the shrimps. The trend of the examined metals in shrimps shows that their bio-accumulation depends on the extent of pollution and/or the introduction of contaminants into the environment which may be influenced by changes brought by seasonal dynamics. Though, metal toxicity arising from the direct consumption of shrimp in the area has not been reported. This work has provided some data and information that may be useful for such studies and policy formulation.

There is need for Regular public health checks on the level of heavy metals among the community that border the study area. Thus, it is suggested that more species be sought for proper evaluation. This will demonstrate the extent of the trophic transfer of the metals and organics in the area. Also, continued contaminants monitoring of the Ondo Coastal regions are essential if relevant agencies are to ascertain the quality of habitats for resident and migratory wildlife. Consequently, a contaminant monitoring programme should be established not only for heavy metals.

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