

Assessment of Arsenic and Selenium in Water, Sediment and Fish (*Sarotherodon melanotheron*) from Ekerekana Creek, Okrika, Rivers State, Nigeria

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Abstract

The concentrations of arsenic and selenium in water, sediment and fish (*Sarotherodon melanotheron*) from Ekerekana Creek, Okrika, Rivers State were assessed following a standard procedure of using Atomic Absorption Spectrophotometry (AAS). The results showed that the mean concentrations for arsenic in water ranged from 0.224 ± 0.285 mg/L to 0.268 ± 0.936 mg/L and not significantly different between stations or months ($p > 0.05$); Arsenic levels in sediment ranged from 0.485 ± 0.253 mg/kg to 5.918 ± 0.654 mg/kg, while the concentrations in *Sarotherodon melanotheron* ranged from 0.193 mg/kg to 10.930 mg/kg. Similarly, mean concentration of Selenium in water ranged from 0.358 ± 0.425 mg/L to 0.840 ± 0.357 mg/L and showed significant difference ($p < 0.05$), the mean concentrations of selenium in sediments ranged from 0.251 ± 0.017 mg/kg to 2.376 ± 0.202 mg/kg and selenium concentration in fish (*Sarotherodon melanotheron*) ranged from 1.27 mg/kg to 5.63 mg/kg. The above results showed that the arsenic concentrations in sediments were below Department of Petroleum Resources (DPR) target value of 29 mg/kg and the Canadian Sediment Quality Guideline (ISQG 17.0 mg/kg). The concentrations of arsenic and selenium in sediments were higher than concentrations in water, while the concentrations of arsenic and selenium in the muscles of *Sarotherodon melanotheron* were higher than concentrations in sediments. The muscles of *Sarotherodon melanotheron* are edible part and the concentrations of heavy metals (arsenic and selenium) observed in this study could potentially be of concern for human health. A monitoring programme for heavy metal concentrations in Ekerekana Creek of Upper Bonny Estuary is recommended.

Keywords Heavy metals, *Sarotherodon melanotheron*, Concentration, Bioaccumulation, Upper Bonny Estuary.

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I. Introduction

Environmental pollution remains the world's greatest problem facing humanity and the leading environmental causes of morbidity and mortality. Man's activities through urbanization, industrialization, waste disposal, mining, and exploration are at the forefront of global environmental pollution¹. Both developed and developing nations share this burden together. Despite the global attention towards pollution, the impacts of heavy metal pollutants are still being felt due to their severe long term persistence and consequences. Heavy metal levels have increased in the Niger Delta Estuaries over the past decades, due to domestic, industrial, mining and Agricultural activities². Wastes from these activities discharged and released into the aquatic environment may cause extensive ecological differences, due to their level of toxicity, persistence and accumulative behaviour in the organisms that dwell in the marine or estuarine ecosystem³. Most heavy metals are the natural constituent of the earth's crust from where they are taken up by organisms and transferred into food chain. A good number of heavy metals are present in trace amount, but high level of accumulation may affect organisms through the food chain and pose risk to consumers of sea food when concentration exceed the permissible limits⁴. The presence of heavy metals such as Selenium (Se), Arsenic (As), mercury ((Hg), Copper (Cu), Lead (Pb) and other heavy metals in aquatic environment are evidence of contamination or pollution depending on the level of concentration. The ultimate recipient of all forms of pollutants is natural water bodies. Many industries were, and are still being sited near these bodies of water presumably to facilitate easy discharge of effluents and other wastes into them⁵. Disposal of various waste materials into rivers, estuaries, and marine waters therefore is not a modern phenomenon since this practice has been used as a preferred disposal option virtually since the beginning of human civilization⁶. Ekerekana Creek, in Okrika, Niger Delta, through which

effluent waste water from Port Harcourt Refining Company (PHRC) passes to facilitate its discharge into Bonny River, has been a recipient of such wastewater since the company came on stream in 1962⁷. The discharge of wastes into the natural water will cause pollution of these systems to the detriment of their *flora* and *fauna* and subsequent downstream user of the systems⁵. Water quality is a major problem in the 21st century and Niger Delta estuaries are not exception. Industries are the major sources of pollution to the environment as various pollutants are released into the surrounding environment directly and indirectly, making the water unsuitable for drinking, domestic use, recreational and agricultural purposes⁸. In the Niger Delta, the problem of water and sediment pollution has been of concern to all stakeholders, following the rate and extent of degradation of the environment and water bodies by human activities, particularly from industrial and domestic sources. Wastes from these activities discharged and released into the aquatic environment, may cause extensive ecological differences due to their level of toxicity, persistence and accumulative behaviour in the organisms that dwell in estuarine ecosystem⁹. The metal concentrations varies with species of fish, this may be related to their feeding habits and bioaccumulation capacity¹⁰. It is important to monitor the level of metals such as arsenic (As), selenium (Se) and other heavy metals in water, sediment and fish from Niger Delta Estuaries in order to compare them with regulatory standards and guidelines for public health purpose⁹. Fish has various points of entries for heavy metals; skin, oral consumption of water, assimilation through the gills, food and non-food particles. Once absorbed, they are then carried in the blood stream to a storage organ such as the liver for transformation and storage. The fish are widely used to evaluate the health of aquatic systems because pollutants build up in the food chain, and are responsible for adverse effects and death in aquatic system^{11,12}. Upper creeks of Bonny Estuary have been exposed to pollution pressures derived both from anthropogenic activities and natural sources. The communities that are situated along the shore of the river which include the Okochiri, Ekerekana as well as other nearby communities consume fish from Ekerekana Creek as source of nutrients. Fish is known worldwide as a very important component of human diet because of its high nutritive value and significance in improving human health⁵. The concentration and accumulation of arsenic and selenium in fish poses a great health risk to the communities situated along Ekerekana Creek. The aim of this research is to investigate the levels of arsenic and selenium in water, sediment and fish from Ekerekana Creek and to compare them with regulatory/safe limits.

II. Materials And Methods

Study area

The sampling stations were established along Ekerekana creek in Okrika Local Government Area Rivers State, Nigeria. The creek is brackish as evidenced in their vegetation. The main source of the river is the Bonny River. People who live in communities around the river are mostly fisher men and farmers. The Water from the river is also used for irrigation by farmers who farm along the bank of the river. The river serves as a means of livelihood for people who fish and those who depend on the river for growing their crops especially in dry seasons. Anthropogenic activities along the creeks include; sand mining, fishing, navigation, farming, bathing and recreational activities. A major industrial outfit which is situated in the station one (Ekerekana) is the Port Harcourt Refinery company (PHRC), a subsidiary of Nigeria National Petroleum Corporation (NNPC) which generates several volumes of effluents that is channelled into creek through a drainage channel.

Sampling sites

The four (4) sampling stations selected for this study as indicated in the map (Fig. 1) are described below:

- ✓ Station 1- point of discharge of refinery effluent into the Ekerekana Creek.
- ✓ Station 2- Boundary between Ekerekana and Okari Creeks.
- ✓ Station 3- Refinery jetty where pipelines are situated and petroleum products are loaded.
- ✓ Station 4- upstream of refinery jetty (presumed control station).

All sampling points were geo-located with GPS and plotted on a map.

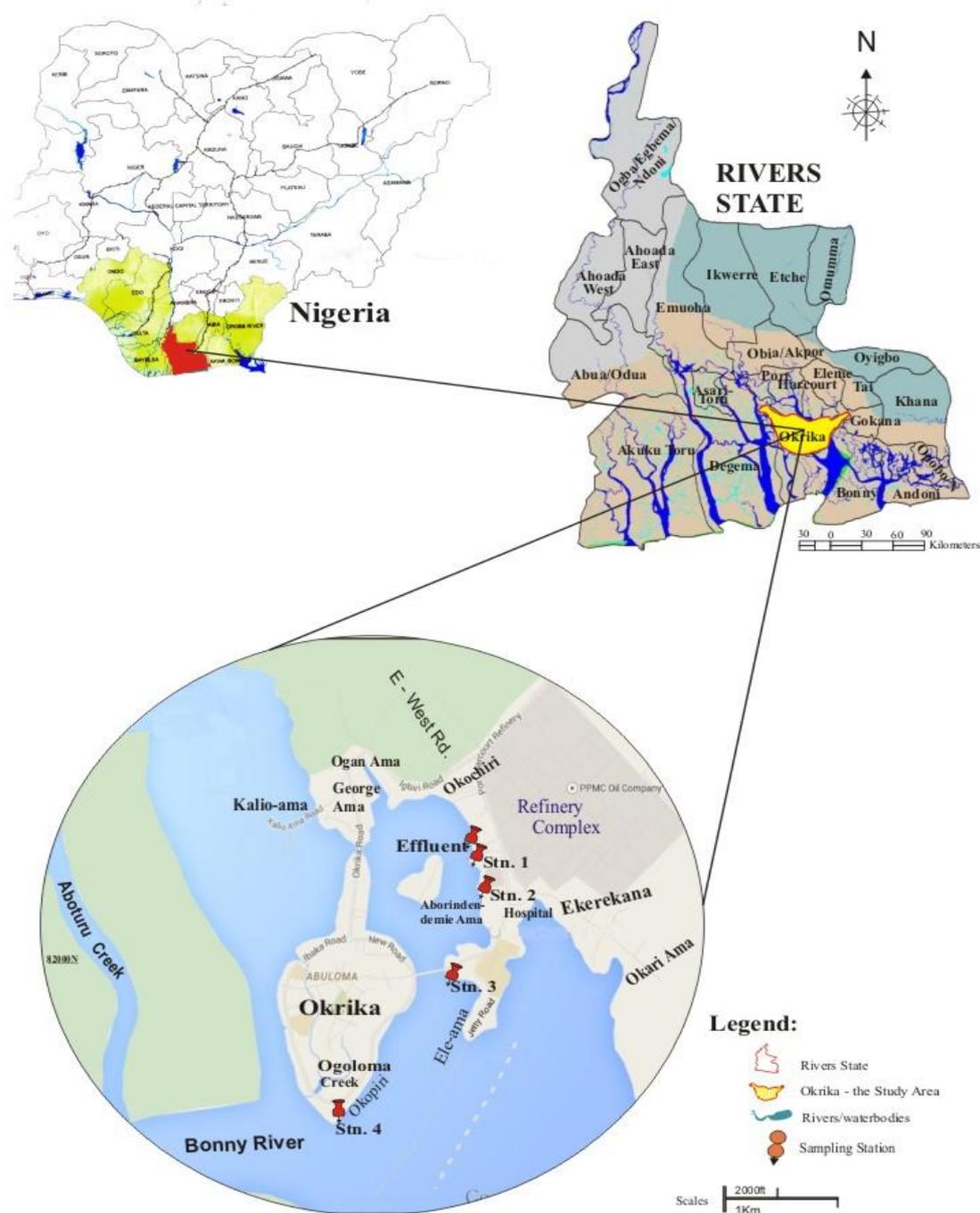


Fig. 1: Map Showing the Sample Sites in the Study Area

Sample collection and Analysis

Water, sediment and fish samples were collected from the four designated stations as indicated in the map. The samples were collected for four (4) months; September and October 2019, representing raining season, while December 2019 and January 2020 represent dry season.

Water Samples

Water samples were collected at low tide at about 30cm depth; Samples for heavy metal analyses were placed in 250 ml plastic containers and fixed with 2 drops of concentrated HNO_3 to preserve the metals. The water samples were stored in an ice box and taken to the laboratory within six hours for further treatment and analysis. The concentrations of arsenic and selenium in water samples were determined by anatomic absorption spectrophotometry (AAS) and the metal concentrations were recorded in mg/l.

Sediment Samples

Sediment samples were collected from the sub-tidal at low tide using a stainless Eckman grab sampler at each location¹³. All sediment samples for metal analysis were preserved in ice chest before transferring to the laboratory for further treatment and analysis.

In the laboratory, Sediment samples collected was air dried at room temperature for two to three weeks until the weight was constant. After drying the sediment sample's visible remains of organisms and debris were removed; sediment samples were crushed in a mortar and sieve using a 200µm sieve to normalize particle size. Samples were processed for analysis of arsenal by Atomic Absorption Spectrophotometry.

Fish Samples

The services of fisher men were employed in the collection of fish samples. The fish samples were collected from the designated stations; the fish samples were labelled, preserved in ice box before taking it to the laboratory where they were preserved in the freezer prior to treatment and analysis.

In the laboratory the whole biota (fish) was properly cleaned with distilled water to remove debris and all external adherents. The samples were thawed on a clean plastic sheet; gills and muscles were excised using a dissecting kit. The excised gills and tissues were dried in oven four to seven days. After drying, the samples were crushed in mortar and processed for analysis of arsenal by Atomic Absorption Spectrophotometry (AAS).

Statistical Analysis

Analysis of variance (One-way ANOVA) was used in analyzing data for heavy metals in water (while Turkey multiple comparison was used to compare the significant differences of the mean values among the sampled stations or months). For the data on metals in sediment and fish tissues of *Sarotherodon maletheron* which had replicate determinations for each station/month, Two-way full crossed analysis of variance(ANOVA) was applied with Tukey tests for multiple comparisons (where ANOVA gave significant differences).

III. Results

Arsenic and Selenium in Water

The concentration of Arsenic varied from <0.001 mg/L to 2.832 mg/L (Fig. 2).. The maximum value was observed in station 1 in September. Minimum value was observed in station 1 in January. Mean value per station ranged from 0.224 ± 0.285 mg/L to 0.268 ± 0.936 mg/L. Analysis of variance ANOVA showed that there is no significant differences between stations and months ($p > 0.05$).

Selenium concentration in water varied from <0.001 mg/L to 2.222 mg/L (Fig. 3). The maximum value of selenium concentration (2.222 mg/L) was observed in station 4, in January while the minimum value of (<0.001 mg/L) was observed in station 1, 2 and 4 in September, October and December. Mean value for stations ranged from 0.358 ± 0.425 mg/L to 0.840 ± 0.357 mg/L in station 3. Analysis of variance ANOVA show that there is no significant differences between stations, but showed significant difference in the mean concentrations of Selenium between months($p < 0.05$).Tukey test gave the inference January>October=September>December.

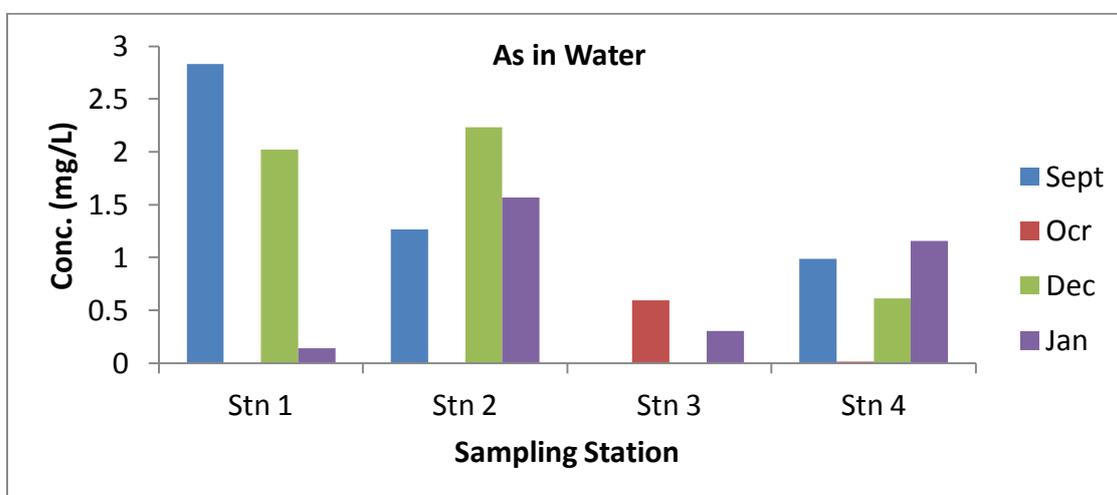


Fig 2: Spatial Variation in the Concentrations of Arsenic from September 2019 to January 2020

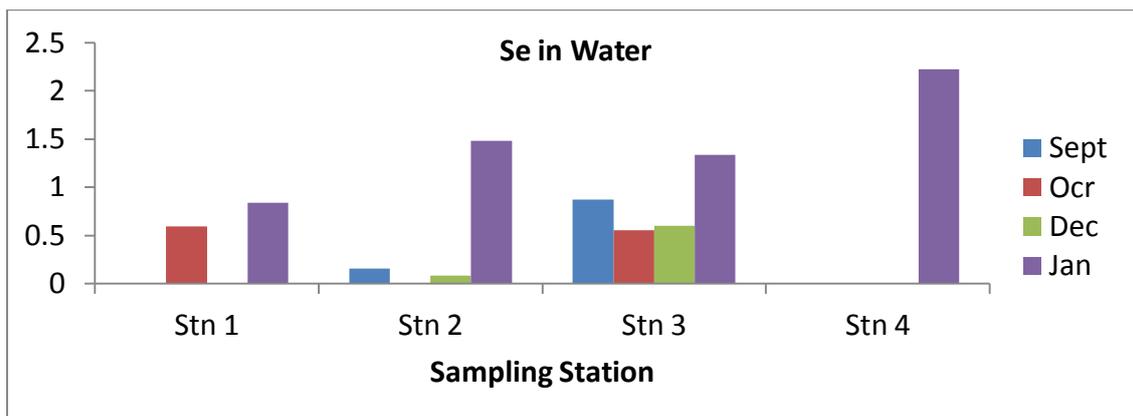


Fig 3: Variation in the Concentrations of Selenium in water from September 2019 to January 2020

Arsenic and Selenium in Sediment

Spatial variations in the concentrations of arsenic in sediment for the four months are presented in Figures 4 to 7. In September, the concentration of arsenic in sediments ranged from 0.123 mg/kg to 3.351 mg/kg. The maximum value (3.351 mg/kg) was observed in station 4, while the minimum value (0.123 mg/kg) was observed in station 3. The mean concentration of Arsenic in sediments in stations varied within the range of 0.485 ± 0.253 mg/kg to 2.626 ± 0.777 mg/kg in September. In October, the concentration of Arsenic in sediments varied from 1.807 mg/kg to 5.912 mg/kg. The maximum concentration (5.912 mg/kg) of Arsenic in sediments was observed in station 3, while the minimum value (1.807 mg/kg) was observed in station 4. The mean concentration of Arsenic in sediments within stations varied within 2.480 ± 0.507 mg/kg to 5.456 ± 0.263 mg/kg. In the month of December, the concentration of Arsenic in sediments varied in stations from 0.175 mg/kg to 2.351 mg/kg. The maximum value (2.351 mg/kg) was observed in station 2, while the minimum concentration (0.175 mg/kg) was observed in station 1. The mean concentration varied in stations from 0.883 ± 0.372 mg/kg to 1.901 ± 0.161 mg/kg. In January, concentration of arsenic in sediments varied from 0.614 ± 0.138 mg/kg to 6.912 ± 0.654 mg/kg. The maximum concentration (6.912 mg/kg) was observed in station 3, while the minimum concentration (0.614 mg/kg) was observed in station 4. The mean concentrations of Arsenic varied in stations from 0.871 ± 0.138 mg/kg to 5.918 ± 0.654 mg/kg. ANOVA shows significant differences in the mean concentration of Arsenic between months ($p < 0.05$). Tukey test did not show seasonal trends in the differences between months as in the rainy season month of October and dry season month of January, but these were significantly different from the month of September and December.

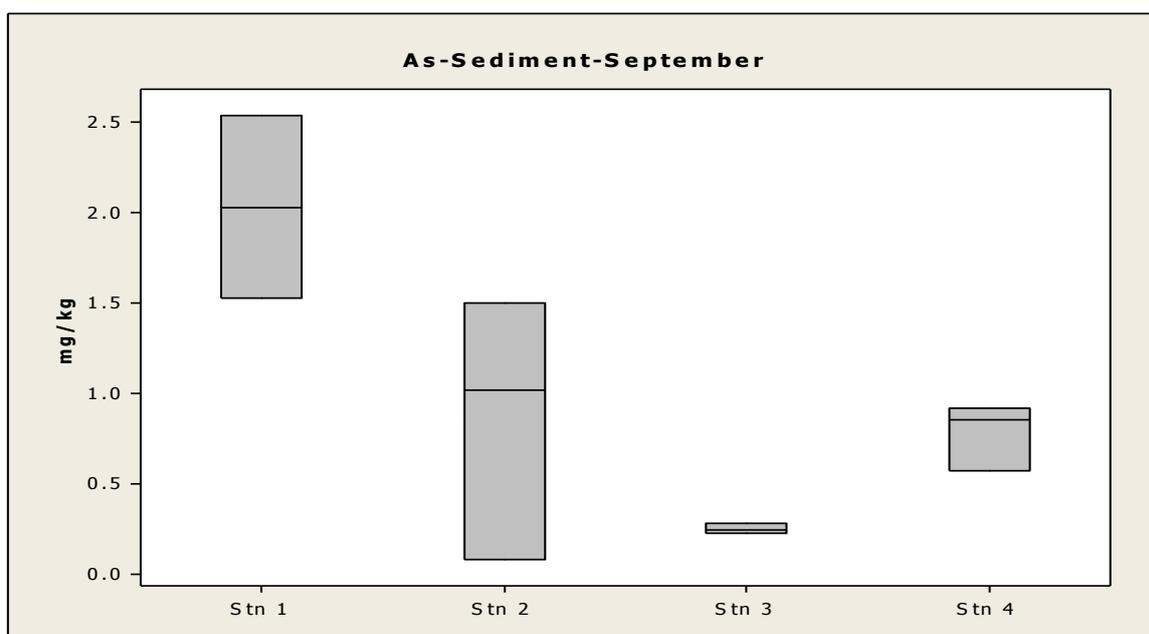


Fig 4: Spatial differences in the Concentration of Arsenic in Sediment Samples from the Study Area in September 2019.

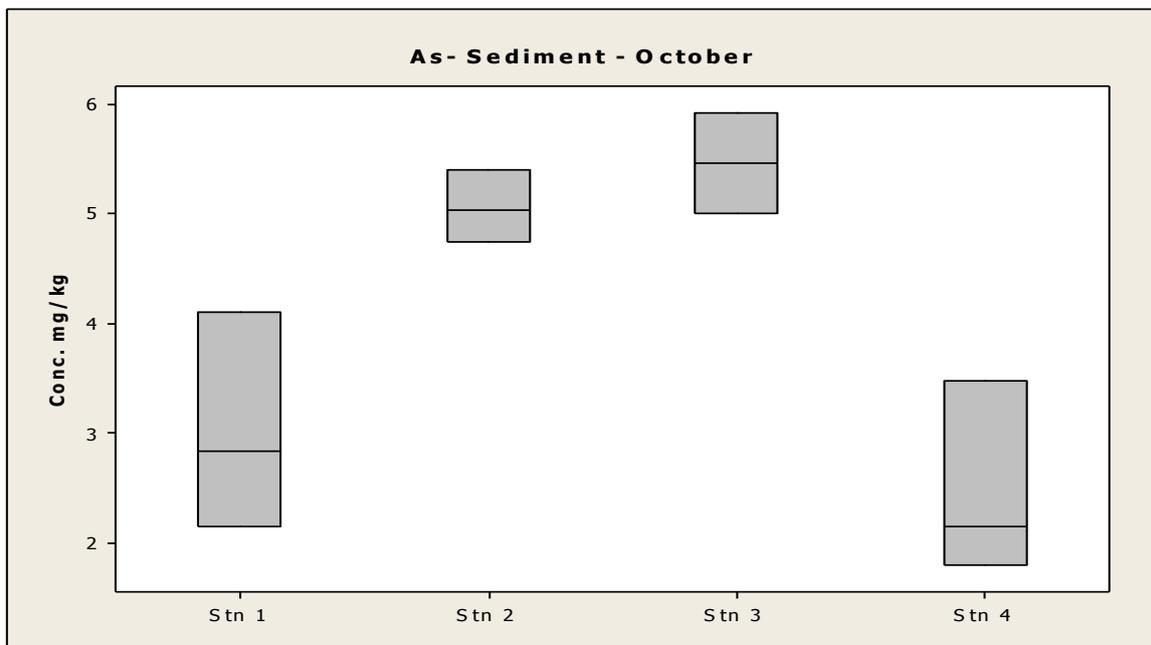


Fig 5: Spatial differences in the Concentration of Arsenic in Sediment Samples from the Study Area in October 2019.

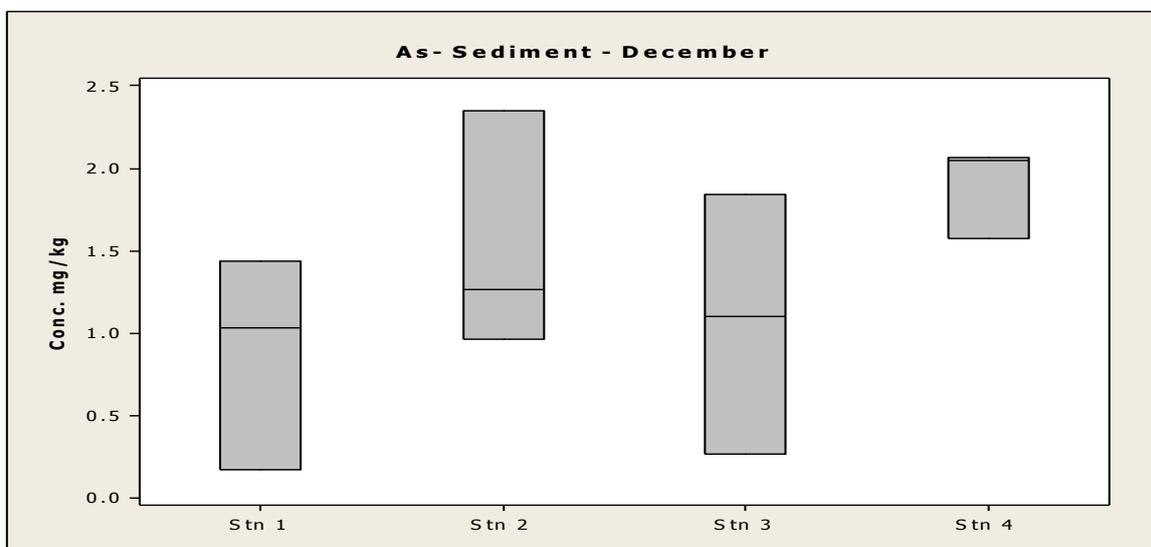


Fig 6: Spatial differences in the Concentration of Arsenic in Sediment Samples from the Study Area in December 2019.

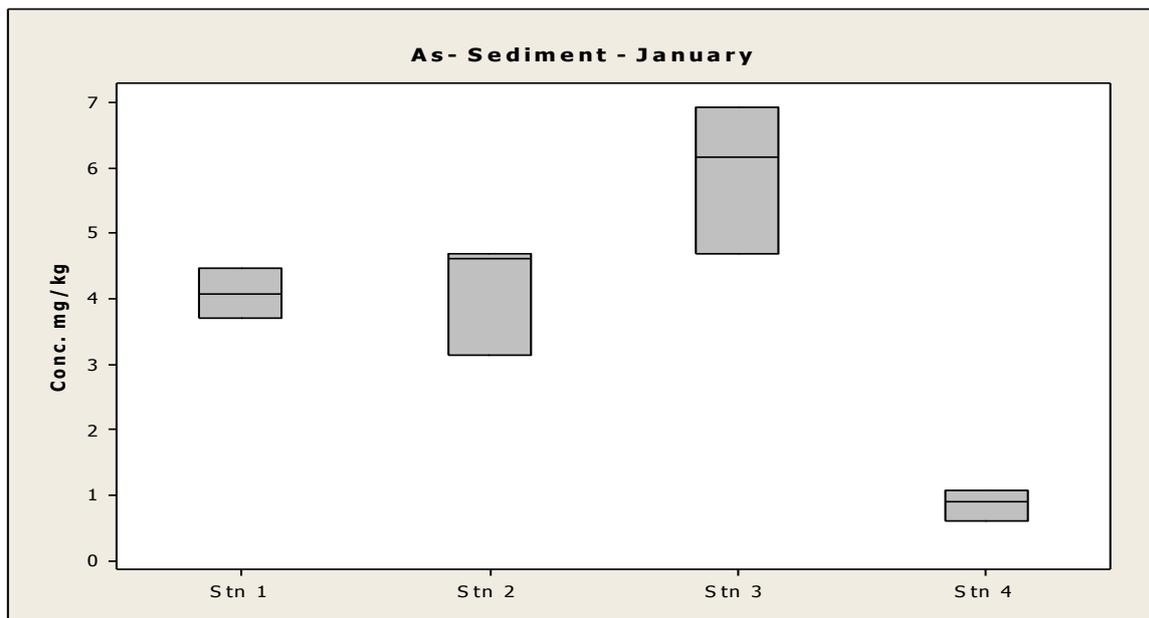


Fig 7: Spatial differences in the Concentration of Arsenic in Sediment Samples from the Study Area January 2020.

The concentrations of selenium in sediments are presented in Figures 8 to 11. The values varied from 0.085 mg/kg to 2.533 mg/kg in September. Maximum concentration (2.533 mg/kg) was observed in station 1, while the minimum value (0.085 mg/kg) was observed in station 3. The mean concentration varied from 0.251 ± 0.017 mg/kg to 2.028 ± 0.291 mg/kg. In October, Selenium concentration varied from 0.311 mg/kg to 2.665 mg/kg. Maximum concentration (2.66 mg/kg) was observed in station 4, while the minimum value (0.311 mg/kg) was observed in station 1. The mean concentration in stations varied from 0.380 ± 0.041 mg/kg to 2.376 ± 0.202 mg/kg. In December, selenium concentrations varied from 0.198 mg/kg to 2.100 mg/kg. Maximum concentration (2.100 mg/kg) was observed in station 4, while the minimum value (0.198 mg/kg) was observed in station 1. The mean concentration in stations varied from 0.756 ± 0.318 mg/kg to 1.827 ± 0.146 mg/kg. In January, selenium concentrations varied from 0.311 mg/kg to 2.439 mg/kg. Maximum value (2.439 mg/kg) was observed in station 2 while minimum value (0.311 mg/kg) was observed in station 4, the mean concentrations varied from 0.879 ± 0.288 mg/kg to 2.291 ± 0.104 mg/kg. ANOVA for Selenium concentration showed significant difference between months and between months and stations ($p < 0.05$). Tukey test did not show seasonal trends in the differences between months as in the rainy season month of October and dry season of January, but these were significantly different from the month of September and January.

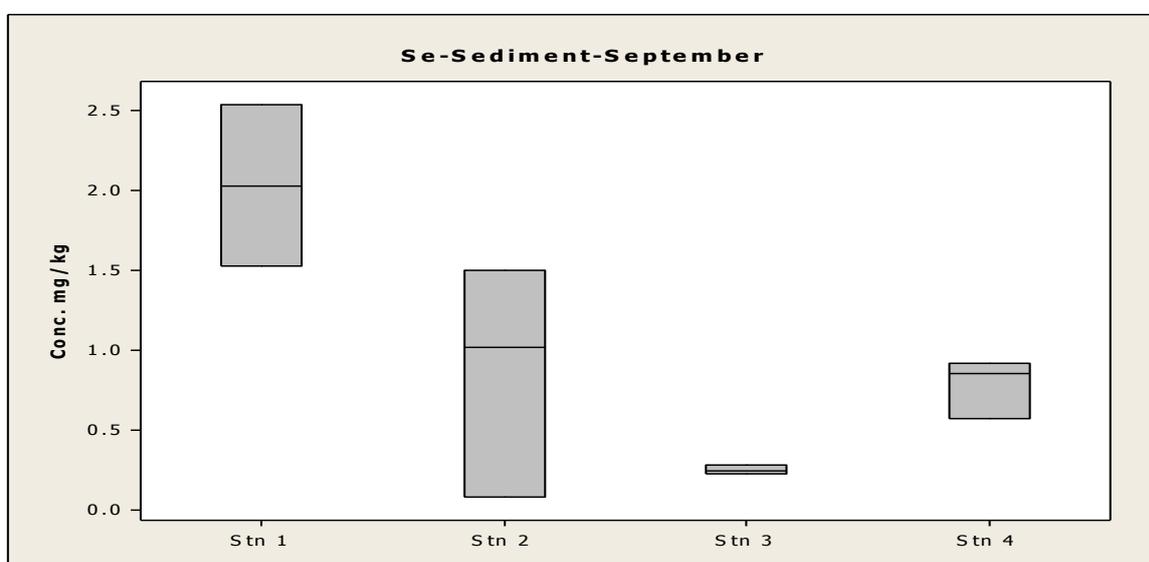


Fig 8: Spatial differences in the Concentration of Selenium in Sediment Samples from the Study Area in September 2019

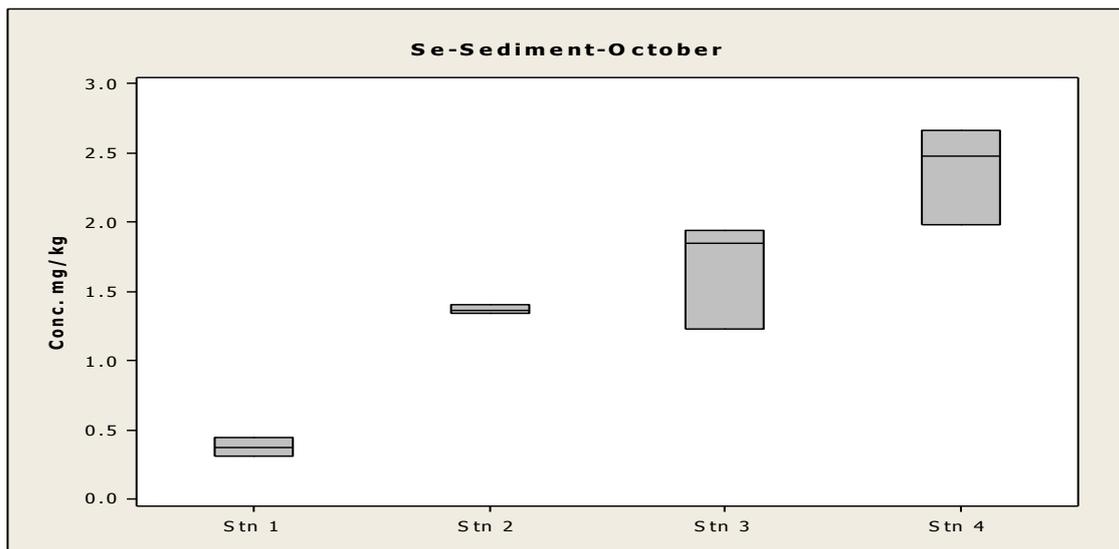


Fig 9: Spatial differences in the Concentration of Selenium in Sediment Samples from the Study Area in October 2019

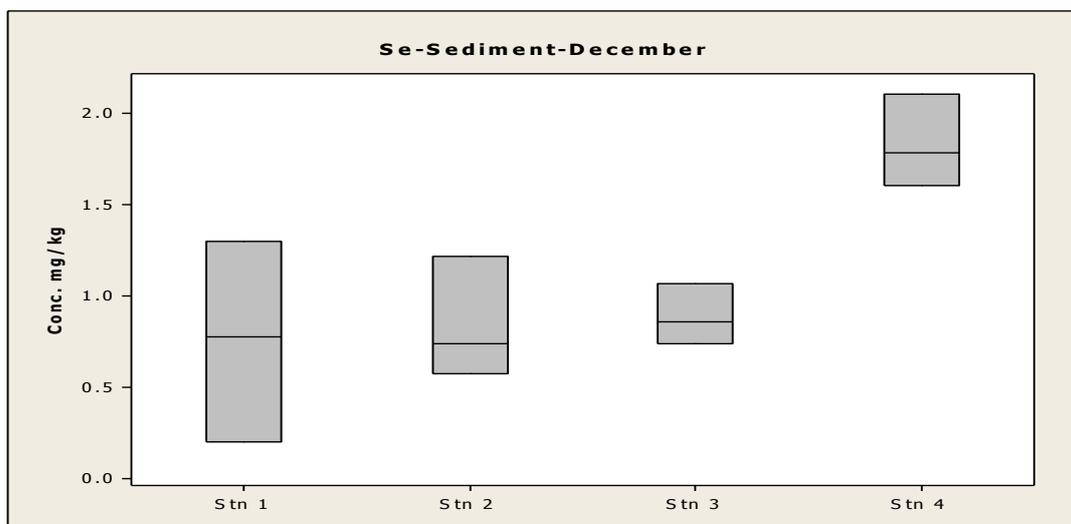


Fig 10: Spatial differences in the Concentration of Selenium in Sediment Samples from the Study Area in December 2019.

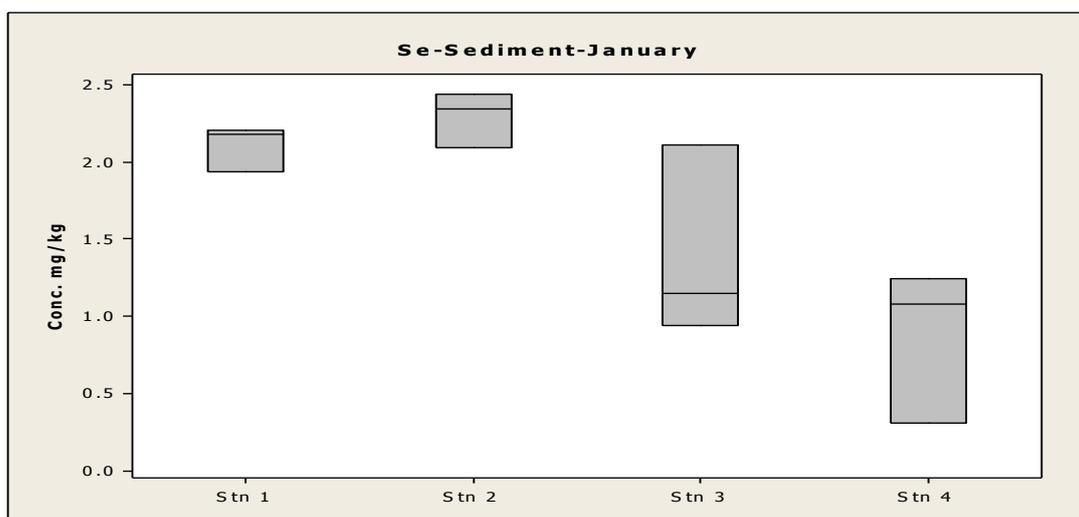


Fig 11: Spatial differences in the Concentration of Selenium in Sediment Samples from the Study Area in January 2020.

Arsenic in Fish ((*Sarotherodon melanotheron*))

Concentration of arsenic in Tilapia (*Sarotherodon melanotheron*) ranged from 0.193 mg/kg to 4.912 mg/kg in September (Fig. 12). The maximum concentration value (4.912 g/kg) was observed in station 4, while the minimum value of 0.193 mg/kg was observed in station 1. The mean concentration in stations varied from 1.526 ± 0.637 mg/kg to 3.386 ± 0.765 mg/kg. In October, concentrations varied from 0.579 mg/kg to 6.930mg/kg (Fig. 13); maximum concentration (6.930 mg/kg) was observed in station 2, while minimum concentration of 0.579 mg/kg was observed in station 3. The mean concentrations in stations varied within 1.673 ± 0.548 mg/kg and 6.146 ± 0.274 mg/kg. In December (Fig. 14) the levels ranged from 3.193 mg/kg to 10.930 mg/kg, with value (10.930 mg/kg) observed in station 4, while minimum value (3.193 mg/kg) was observed in station 1. The mean concentration within stations ranged from 3.673 ± 0.264 mg/kg to $9.649 \pm$ mg/kg. In January (Fig. 15), arsenic concentration values ranged from 2.877 mg/kg to 8.807mg/kg. Maximum concentration was observed in station 1, while minimum concentration was observed in station 3. Mean concentration of Arsenic in fish (*Sarotherodon melanotheron*) varied in stations within the range of 3.076 ± 0.165 mg/kg to 8.708 ± 0.083 mg/kg. ANOVA showed significant differences in the mean concentration of Arsenic in *Sarotherodon melanotheron* between stations, months and between stations and months ($p < 0.05$). Tukey test show seasonal trends in the differences between months as in the rainy season month of September and dry season month of January. But does not show significant difference in stations but the value of Station 1 was significantly different from station 4.

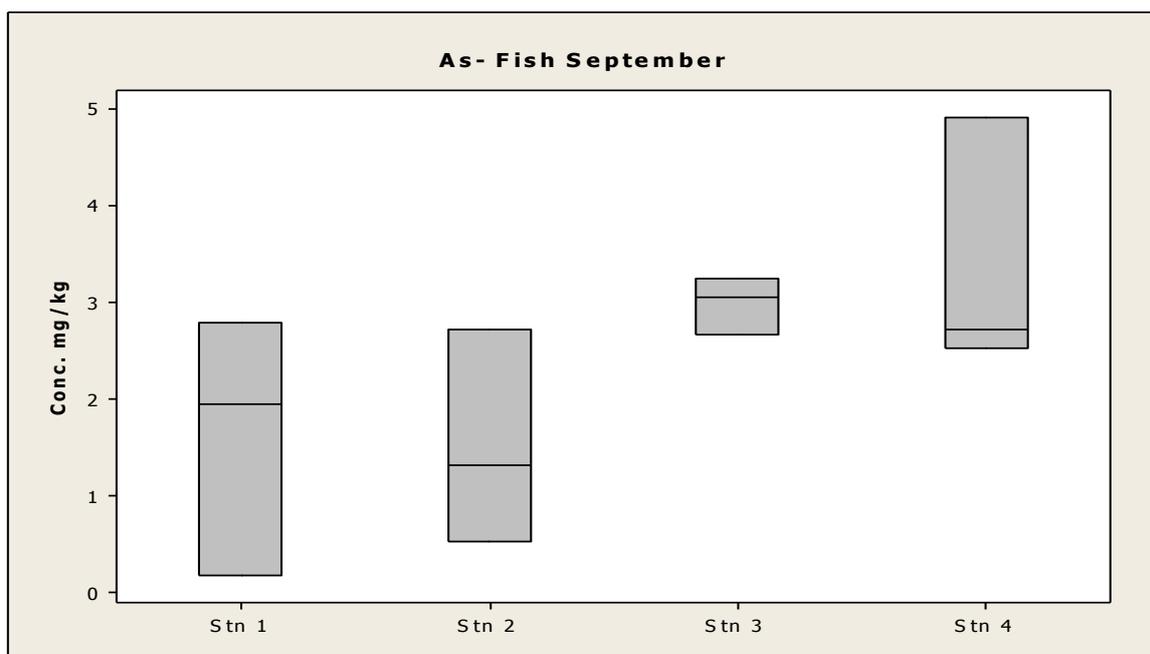


Fig 12: Spatial differences in the Concentration of Arsenic in Flesh of Tilapia (*Sarotherodon melanotheron*) from the Study Area in September 2019

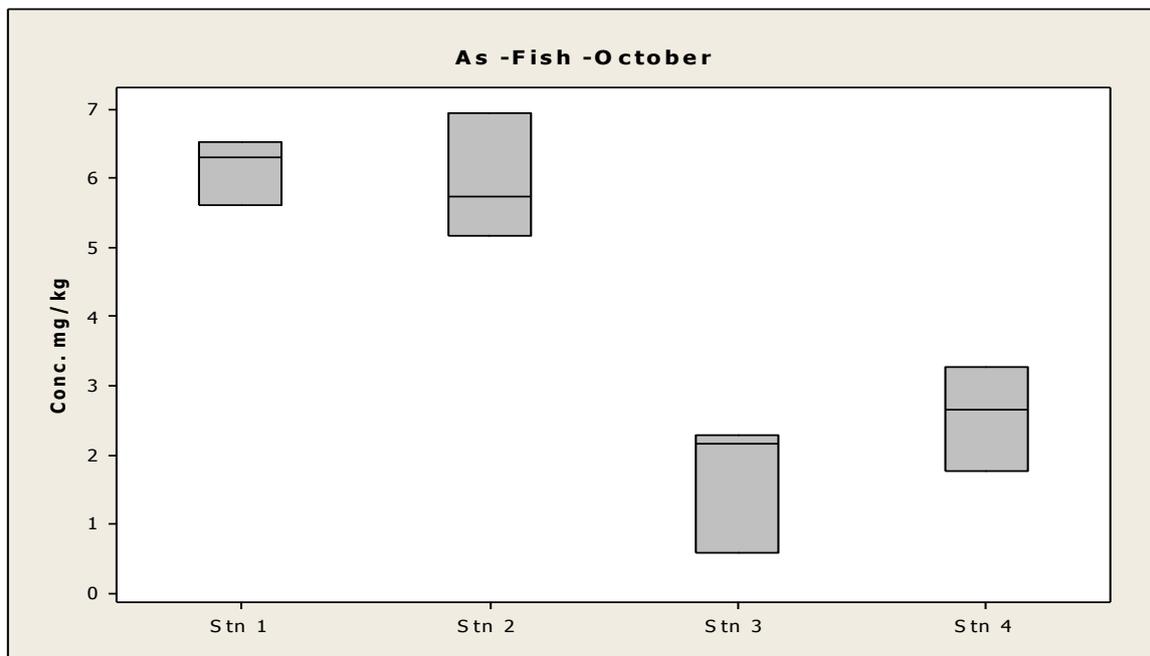


Fig 13: Spatial differences in the Concentration of Arsenic in Flesh of Tilapia. (*Sarotherodon melanotheron*) from the Study Area in October 2019

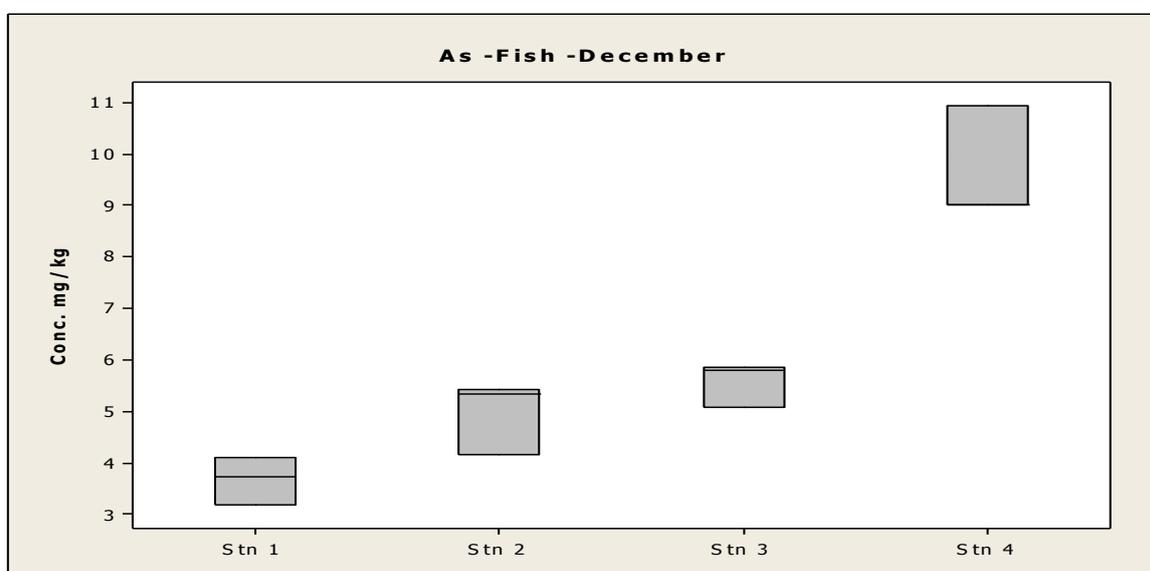


Fig 14: Spatial differences in the Concentration of Arsenic in Flesh of Tilapia. (*Sarotherodon melanotheron*) from the Study Area in December 2019

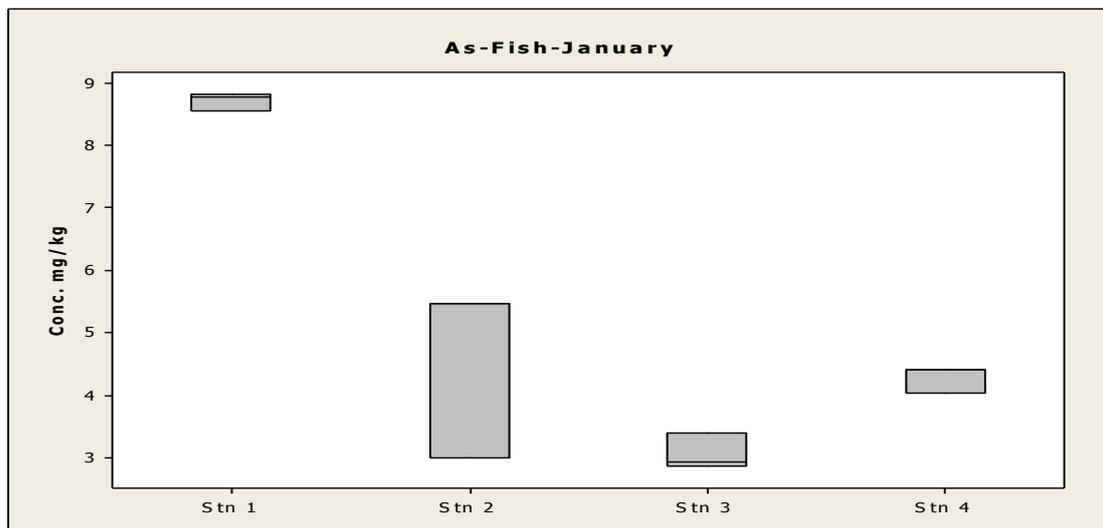


Fig 15: Spatial differences in the Concentration of Arsenic in Flesh of Tilpia. (*Sarotherodon melanotheron*) from the Study Area in January 2020

Selenium in Fish (*Sarotherodon melanotheron*)

The Concentrations of selenium in flesh of Tilapia (*Sarotherodon melanotheron*) are presented in Figures 16 to 19. The values ranged from 1.27 mg/kg to 4.29 mg/kg in September; maximum concentration (4.29 mg/kg) was observed in station 3, while minimum value was observed in station 4. The mean concentration of selenium in Tilapia in stations varied from 1.55 ± 0.28 mg/kg to 3.95 ± 0.18 mg/kg. In October, Concentration value of selenium ranged from 1.73 mg/kg to 3.37 mg/kg. Maximum concentration (3.37mg/kg) was observed in station 4, while minimum concentration value (1.73 mg/kg) was observed in station 2. The mean concentration in stations varied from 1.87 ± 0.12 mg/kg to 3.02 ± 0.33 mg/kg. The concentration of Selenium in flesh of Tilapia (*Sarotherodon melanotheron*) ranged between 1.75 mg/kg to 4.59 mg/kg in December. Maximum concentration (4.59 mg/kg) was observed in station 4, while minimum concentration (1.75 mg/kg) was recorded in station 1. The mean concentration in stations varied from 2.00 ± 0.20 mg/kg to 4.30 ± 0.14 mg/kg. In January, the concentration value of selenium in Tilapia ranged from 2.48 mg/kg to 5.63mg/kg. Maximum concentration (5.63 mg/kg) was recorded in station 1, while minimum value(2.48 mg/kg) was observed in station 4 . Mean concentration varied in stations from 2.75 ± 0.18 mg/kg to 4.84 mg/kg. ANOVA for Selenium concentration in *Sarotherodon melanotheron* showed significant difference in concentration among the months and between months and stations ($p < 0.05$). Tukey test showed seasonality trend in mean concentration of selenium between rainy season month of September and dry season month of January.

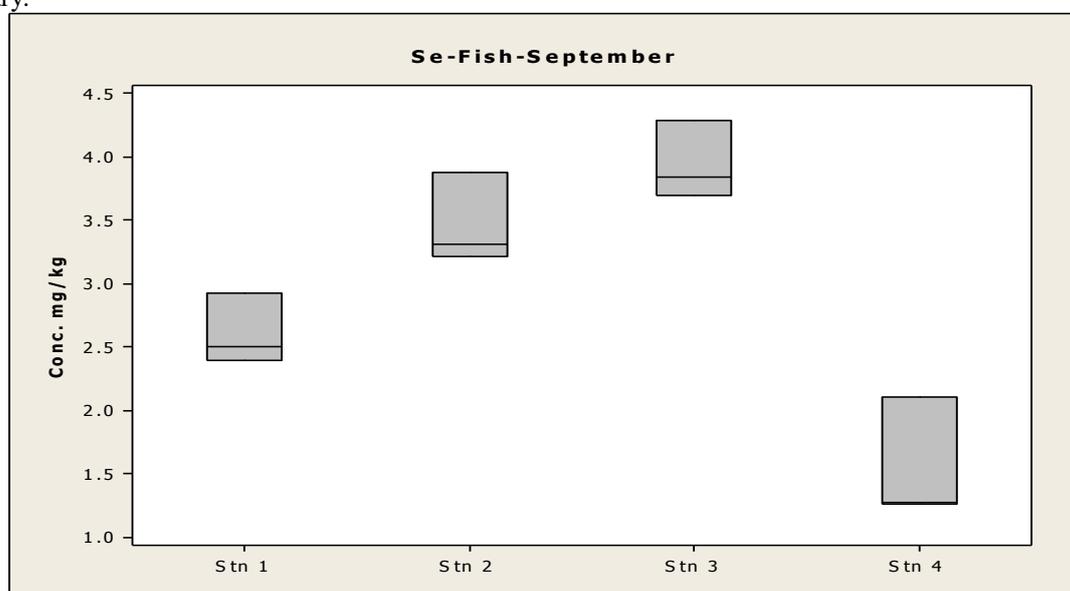


Fig 16: Spatial differences in the Concentration of Selenium in Flesh of Tilapia (*Sarotherodon melanotheron*) from the Study Area in September 2019

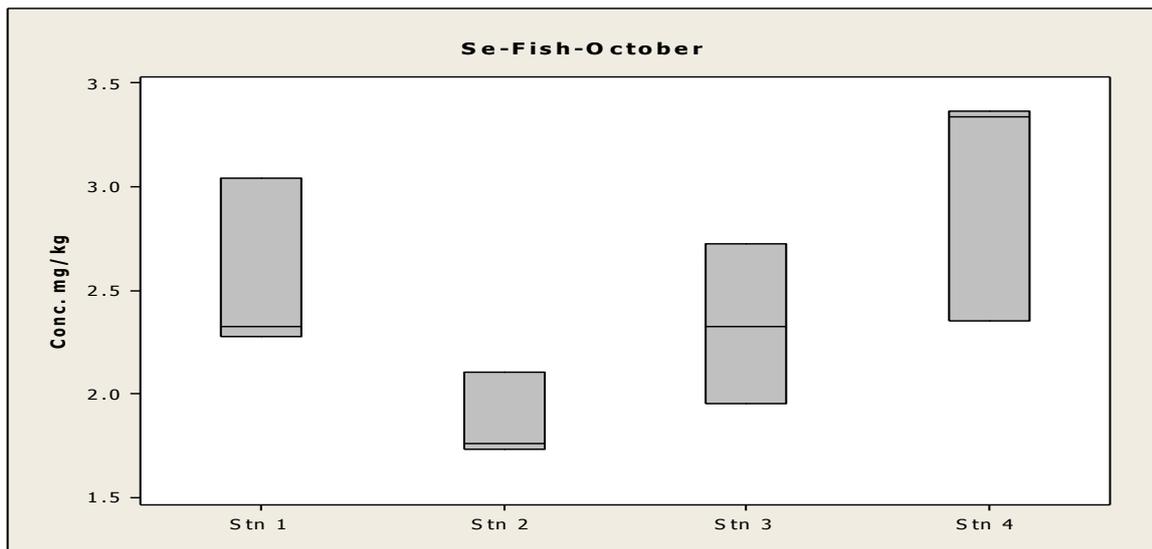


Fig 17: Spatial differences in the Concentration of Selenium in Flesh of Tilapia (*Sarotherodon melanotheron*) from the Study Area in October 2019

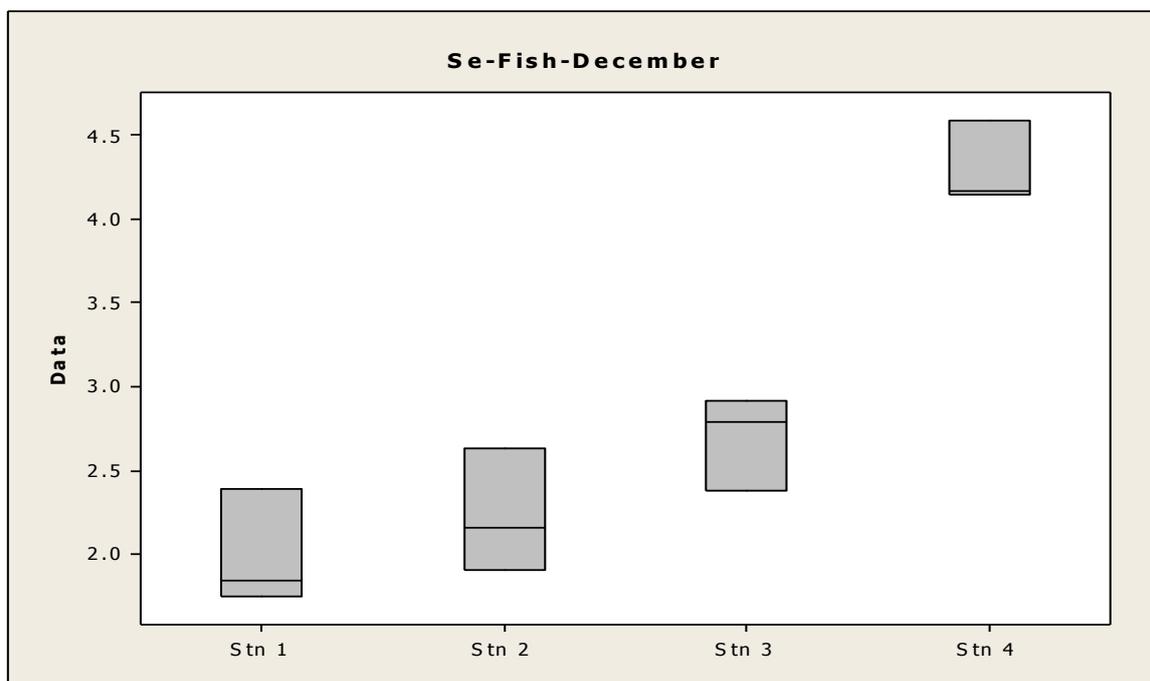


Fig 17: Spatial differences in the Concentration of Selenium in Flesh of Tilapia (*Sarotherodon melanotheron*) from the Study Area December 2019

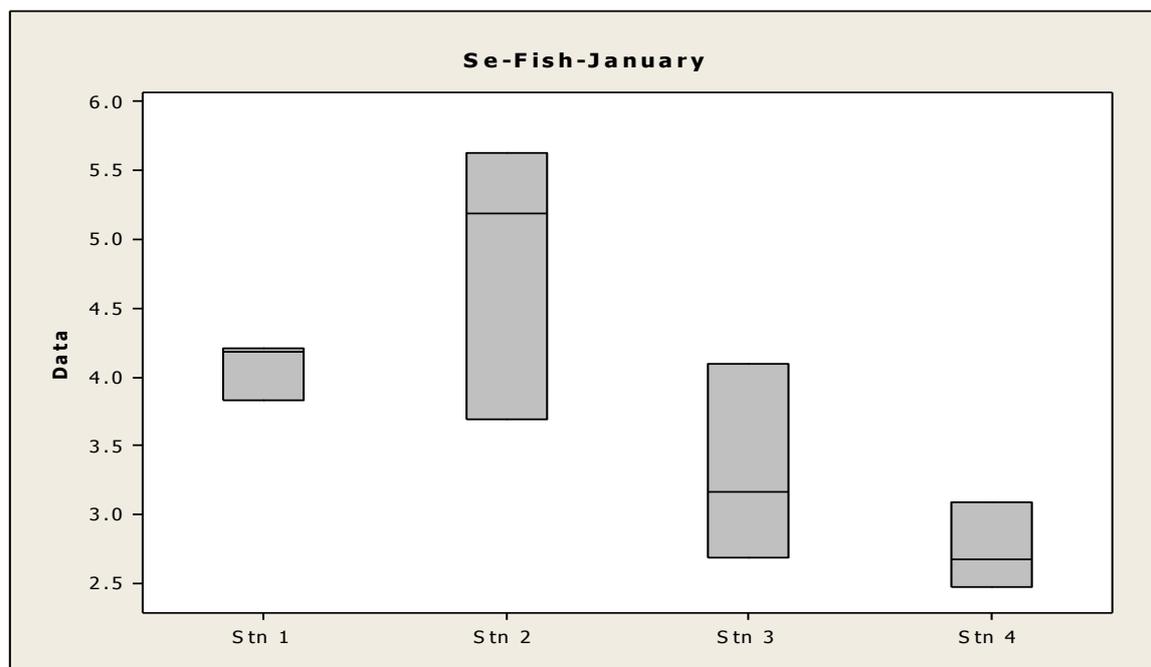


Fig 19: Spatial differences in the Concentration of Selenium in Flesh of Tilapia (*Sarotherodon melanotheron*) from the Study Area in January 2020

IV. Discussion

The concentration of heavy metals in water showed that arsenic had the highest concentration of 2.832mg/L which was observed in station 1 in the month of September compared to selenium with 2.222mg/L as its highest value in station 4 in the month of January. The concentration of arsenic was not significantly different between stations and months ($p > 0.05$), the concentration of selenium was also not significantly different between the stations, but showed significant difference between the months ($p < 0.05$). The general higher values of arsenic and selenium in comparison with the values in effluents indicates that autochthonous processes of remobilization of sediments and other anthropogenic sources contributed to the levels in water.

Concentration of arsenic varied in stations and months, the highest concentration of arsenic (6.92mg/kg) was observed in the month of January in station 4 while the lowest concentration (0.12mg/kg) of arsenic in sediment was observed in September station 3. This suggests seasonal variation in concentration of arsenic in sediment and it showed significant differences in the mean concentrations between months ($p < 0.05$). However, Tukey test did not show clear-cut seasonal trends in the differences between months as in the rainy season month of October and dry season of January, but these were significantly different from the month of September and December. The observed concentrations of arsenic in sediments were lower than Department of Petroleum Resources (DPR) target value of 29mg/kg as well as the Canadian Sediment Quality Guideline (ISQG) of 7.4mg/kg; Portable Effect Level (PEL 17.0 mg/kg) (14).

There were varying concentrations of selenium in sediments from the 4 sampled stations. The concentrations were higher than the concentration in water. Although, there was no significant difference in the mean concentration of selenium between the stations, significant differences existed between months and interaction of months and stations ($p < 0.05$). Concentration of selenium varied in stations and months, the highest concentration (2.533mg/kg) of selenium was observed in station 1, in October while the lowest value of selenium concentration was observed in station 2 in September. No DPR target value is available for selenium neither is there any guideline level from CCME (14).

There were varying patterns of arsenic concentration in the tissues of the Tilapia (*Sarotherodon melanotheron*) from the 4 sampled stations. Result of this research showed that the concentration of arsenic in tissues of *Sarotherodon melanotheron* ranging from 0.192985mg/L to 10.92994mg/L with significant difference between stations ($p < 0.05$). However, Tukey test indicated that the concentrations were significantly higher in the stations (station 4 and 3) that are farthest from the effluent. The high concentration of arsenic in the muscles of *Sarotherodon melanotheron* in this study is in agreement with Anaero-Nweke *et al.* (15) who also recorded high concentration of metals in muscles of fish but not in agreement with El-Moselhy *et al.* (1) and Ugbomeh and Akani (16) that observed lower concentration of metals in muscles. This indicated that the affinity for the accumulation of heavy metals varies from fish to fish and from organ to organ. The muscles of fish is edible part, and the concentration of the heavy metal (arsenic) observed in this study could potentially be of concern for human health.

There were higher concentrations of Selenium in Tilapia *Sarotherodon melanotheron* than water. This is an indication that heavy metals tend to accumulate and concentrate on the aquatic biota than the aquatic environment. The concentration ranged from 2.48mg/kg to 5.63mg/kg and showed significant differences in concentration among the months and between months and stations ($p>0.05$)

V. Conclusion

There were seasonal variations in heavy metal (arsenic) concentrations. Though, arsenic concentrations in sediments were below the Department of Petroleum Resources (DPR) target value and the Canadian Sediment Quality Guideline, the observed concentrations of arsenic and selenium in sediments were higher than the concentrations in water. However, there were varied concentrations of arsenic and selenium in the muscles of *Sarotherodon melanotheron*. In some cases, the concentrations were higher in stations that are farthest from the point of discharge (effluent); this could be attributed to unidentified sources and biogeochemical processes within the system.

The observed concentrations of arsenic and selenium in *Sarotherodon melanotheron* were higher than the concentrations in water and sediments due to bioaccumulation of heavy metals (arsenic, selenium) in biota. Muscles of *Sarotherodon melanotheron* are edible part and the concentrations of heavy metals (arsenic and selenium) observed in this study could potentially be of concern for human health. Hence, there is need for constant monitoring of heavy metal concentrations in Ekerekana Creek of the Upper Bonny Estuary.

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