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Determination of Geographical and Seasonal Variations of Heavy Metals in Swordfish (*Xiphias gladius*) and Yellowfin Tuna (*Thunnus albacares*)

Digoarachchi, D.A.S.U.[1], Walpita C.N.[2], Sandamali J.D.[3]
[1,2,3] Department of Livestock Production, Sabaragamuwa University of Sri Lanka

ABSTRACT: Bioaccumulation of heavy metals in marine fish is a major concern worldwide and it leads to some economical burdens for the tropical fish exporters. However, data is yet to be unavailable in Sri Lanka in this regard. Hence this study was conducted to explore geographical variation of heavy metal accumulation in the Swordfish (*Xiphias gladius*) and Yellowfin tuna (*Thunnus albacares*) as they play a predominant role in the export fish market in Sri Lanka. Secondary heavy metal analysis data of Mercury, Led, Arsenic and Cadmium were collected from two major fish export companies in Sri Lanka during the period of 2015 to 2017. Particular points were traced with the aid of log sheets and heavy metal variations were mapped against fish species and heavy metal concentrations by using QGIS software. Heavy metal analysis data were concentrated between 64 to 70 longitudes and 0 to 8 latitudes, 76 to 80 longitudes and 0 to 8 latitudes, 88 to 92 longitudes and 14 to18 latitudes and 82 to 90 longitudes and 6 to 14 latitudes. Heavy metals such as Cd, As and Pb were not detected in the fish samples. Mercury was the major heavy metal accumulated among the tested and high concentration was detected in Swordfish. Mercury accumulation was highest in longitudes between 64 to 70 and latitudes of 0 to 8.This shows serious health issues that are challenging to humans and provides clear evidence on marine pollution. It is recommended to prevent fishing in these particular areas. Proactive measures and techniques need to be applied to fix safe limits of heavy metals in these natural aquatic ecosystems and protect the organisms in the environment.

KEY WORDS: Fishing Industry, Geographical Mapping, Heavy Metal Accumulation, Swordfish, Yellowfin Tuna.

I. INTRODUCTION

Fishing industry is a growing sector in the world as fish is a main source of protein by a large section of the population. It plays an important role in the Sri Lankan economy and it comprises three major sub sectors named marine, inland and aquaculture. A significant part is played by the marine sub sector providing 49% share for the total fish production of the country. The annual marine fish production of Sri Lanka in 2019 was 415,490 Mt and the country exported 53,483 Mt fish and fishery products by earning 299 USD $ Million to the country [1]. As an island, Sri Lanka has a good potential for the fishery industry with Exclusive Economic Zone (EEZ) area of 517,000 Km^2. Currently 65% of oceanic area is free for fishing with the Northern and Eastern part of the country. Yellowfin tuna (*Thunnus albacares*), Big eye tuna (*Thunnus obesus*) and Swordfish (*Xiphias gladius*) are the dominant finfish species of the seafood export industry in Sri Lanka. The annual estimated catch of Yellow fin tuna and Swordfish was 28,775 Mt and 4,363 Mt respectively [2]. According to the Export Development Board, Sri Lanka has been exporting fish and fishery products to the European Union, United Kingdom (UK) and Asian markets including Japan. The UK is the main market for Sri Lankan seafood followed by France, Italy, Netherlands and Germany [3].

Seafood should strictly comply with the quality parameters of the export fish market instead of preventing rejections and to obtain a higher foreign exchange. One of the major incidents was the European Union against Sri Lanka and it affected Sri Lanka during the period of October 2014 to April 2016. In 2015, Sri Lanka exported 17,461Mt of fish and fishery products and it showed a 34% decline than previous year and export earning showed 29% drawback [1]. This significant loss of export earnings was due to less attention on illegal, unreported and unregulated (IUU) fishing. Hence, this is significant to prevent these types of issues in order to compete in the export fish market.

Heavy metal accumulation is a similar type of problem and has been a major concern. Many aquatic ecosystems in the world including Sri Lanka are contaminated with biological or chemical means with the rapid urbanization, population growth and industrialization. Thus, it has become a huge challenge of producing uncontaminated aquatic products. Apart from that deforestation, domestic and farming sewage discharges, mining activities and some agricultural and industrial activities accounted for heavy metal accumulation in aquatic ecosystems [4]. Some metals like Copper, Zinc, Iron are essential for normal fish metabolism and nonessential metals such as Led (Pb), Mercury (Hg), Arsenic (As) and Cadmium (Cd) are considered as most toxic heavy metals that can be accumulated in fish body [5]. The main export marine fish types such as Yellowfin tuna, Swordfish and the Red snapper were found to be contaminated with heavy metals such as Hg, Cd and Pb. The higher level of Hg was common in Swordfish [6]. Furthermore, marine fish species such as Marlin, Sharks contained higher concentrations of Hg. Therefore, Hg has been a burning issue in the fish export industry during the last few years [6]. Alerts and border rejections in the European Union have reported about 114 chemical hazards in fish and fishery products in 2016 and heavy metal contaminations were very often. The main contaminant was Hg with 61 notifications and both As and Hg were found in sharks. According to the RASFF portal there was at least one notification from top countries that export fishes such as Indonesia, China, India, Canada, and Norway from 2013 to 2017.This provides evidence that heavy metal accumulation is an alarming problem of the fishing industry. Furthermore, RASFF portal has reported 19 notifications of serious heavy metal cases from 2013 to 2017 in Sri Lanka fish exports [7]. Such rejections would tarnish the image and it is more than an economic loss.

Bio accumulation refers to uptake and deposition of contaminants by organisms from their ambient environment from the ambient medium by the air, soil, sediments or water [8]. Metal accumulation ability of fishes may be affected by many factors such as ecological needs, swimming patterns, metabolic activities and living environments [9]. High levels of metals in water are often associated with depositions in skin or gills of the pelagic fishes and sediment associated metals deposit in benthic fishes. Sediments associated with metal are a long term source of contaminants because it can be released into water from time to time [10]. Numerous factors are affected by bioaccumulation of Hg in the fish body. Generally those included the species, size, migration biology, feeding habits and the origin [11]. Considering the feeding habits planktons are in the primary production stage and base of the food chain. Those are mainly phytoplankton and zooplankton. The methyl mercury is reactive with the cellular components and retained in the microbes and the planktons. Thus the bioaccumulation of methyl mercury is dominated in the food chain [12]. Planktons are an important source of Hg in the sea. The plankton concentration and distribution of the sea depend on the nutrition, chemical and physical condition of water and other plankton density. And it may change according to the season and water currents. It is important to consider the planktonic population and species in the selected stations to get an idea about the Hg concentration [13].

Ingestion of these heavy metals causes bad health injuries and those are neurotoxic, carcinogenic, mutagenic or teratogenic. Can cause vomiting, convulsions, paralysis, ataxia, hemoglobinuria, gastrointestinal disorder, diarrhea, stomatitis, tremor, depression and pneumonia, increases in blood pressure or heart rate, skin rashes, and eye irritations [14]. Countries which are having high seafood consumption rate need to maintain their own standard levels of heavy metals and any seafood that does not comply with the standards stipulated should be rejected at the initial production. Since this is a global problem which is arising, it requires techniques and proactive measures to tackle this scourge. Geographical mapping is such intervention that can prohibit the fishing of those identified areas.

II. METHODOLOGY

A. Experimental site and data collection

Secondary data of Pb, As, Hg and Cd analysis were collected from two dominant fish exporting companies in Sri Lanka. Both export companies follow the procedure for heavy metal analysis for fish which are intended to export in order to achieve export quality standards. Testing was done once per three months. Secondary data recorded by the company was used during the period of January 2015 to August 2017.

Hg accumulated in Swordfish (n=302) were collected during the period of June to September 2017. Heavy metals accumulated in yellowfin tuna (n=33) were collected during January 2015 to August 2017. According to the secondary data collected, way points were traced.

B. Trace the way points

Particular fish receiving place and date was traced. Based on that information, particular catching vessels were traced from the updated vessel list. Log sheet of each vessel was found. Log sheets composed with details including type of catch, amount of catch, date of catching and their way points which could be utilized for the study.

C. Mapping

QGIS software was used for mapping of Hg values based on way points.

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D. Mercury level proportionate to harvest

Fish capture area was mainly dispersed in four areas; longitudes between 64 to 70 and latitudes of 0 to 8 (station 1), longitudes between 76 to 80 and latitudes of 0 to 8 (station 2), longitude between 88 to 92 and latitudes of 14 to18 (station 3) and 82 to 90 longitudes and 6 to 14 latitudes (station 4).

Mercury levels were calculated proportionately to the harvest in selected four areas as Hg is the predominant heavy metal accumulation in fish bodies. Maps were generated according to the Hg levels and different levels were indicated by different colors; 1.00ppm to 2.00ppm (Red), 0.80ppm to 1.00 (Yellow), 0.60ppm to 0.80ppm (Green) and 0.00ppm to 0.60ppm (Blue).

$$\text{Hg concentration in particular area} = \frac{\text{Harvest} > \text{Mercury concentration} \times 100}{\text{Total harvest of the area}}$$

III. RESULTS AND DISCUSSION

Heavy metals such as Cd, As and Pb were not detected in the fish samples and Hg was the only heavy metal detected among the tested.

Standard limits for the Hg and the other heavy metals are stipulated in different regulatory bodies. According to the Food and Agriculture Organization (FAO) regulations, the average Hg content shall not exceed 0.5ppm (0.5mg/kg of fresh weight) of the edible parts of the fish products. However, these values can be increased up to 1ppm (1 mg/kg of fresh weight) for the edible parts of some species such as *Xiphias gladius* (Swordfish) and *Thunnus spp.* [15 FAO, 2003]. Table 1 shows some standard limits stipulated by different regulatory bodies.

Table 1: Heavy Metal Regulations for Fishery Products

Regulation body	Hg (ppm)	As (ppm)	Pb (ppm)	Cd (ppm)
EU Regulations	≤ 1	≤ 1	≤ 0.2	≤ 0.1
Codex General Standards	≤0.5		≤ 0.3	
Australian Standards	≤ 0.5	≤ 2	≤ 0.5	
FDA Standards	≤ 0.5			

According to the literature, authors have shown two modes which cause Hg contamination of the Ocean. Those are natural emission and anthropogenic activities. Natural Hg emission processes mainly included volcanic and geothermal processes, biomass burning and erosion of Hg containing minerals [16]. Anthropogenic activities included burning of fossil fuel, coal burning, small scale gold mining, metal production activities, cement production, waste incineration and production of chlorine and caustic soda. Other secondary sources include batteries, paints, switches, electrical and electronic devices, thermometers, blood-pressure gauges, fluorescent and energy-saving lamps, dental amalgam, pesticides, fungicides, medicines and cosmetics.

Marine pollution is influenced by few major factors. About 44% of pollutants are from the land through runoff and discharges of wastewater, 33% from the airborne emission from the land and 12% of pollution due to maritime activities and shipping activities. Accidental spill of oils and balance water also cause to marine water pollution. Dumping of garbage to the offshore and drilling and mining activities took the contribution by 10% and 1% respectively [17]. Generally pollutants are transported by inland water sources to the sea. This water is contaminated with industrial runoff and agricultural runoff. [18]. Deep sea mining is also another source of pollution in marine water. It may produce metal ions in the deep sea [10]. Environment pollution of heavy metals are dominant in mining operation sites and it produce acid mine drainage to release metals. These metals are leached in to the down streams to pollute aquatic resources [19]. Some studies have discovered that marine sediments of industrialized coastal areas are highly contaminated with the heavy metals than other areas.

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A. Mercury Levels of Swordfish

Figure 1 illustrates Hg distribution in different fish harvesting areas in the Indian Ocean.

Mercury accumulation in Swordfish was basically distributed within the longitudes of 76 to 80 and the latitudes of 0 to 8 (Station 2). The highest capture was recorded in that particular area and Hg value exceeded the maximum accumulation level of 0.5mg/kg of fresh weight. Longitudes of 76 to 80 and 0 to 4 latitudes contained highest Hg concentration exceeding the Hg value than 1ppm and also represented Hg within the levels of 0.8 to 1ppm. There is a potential risk in that area to accumulate the Hg level which exceeds the concentration of 1ppm. The capture belongs to the area between 64 to 68 longitudes and 0 to 8 latitudes (Station 1) were found to be contaminated with Hg and the concentration level exceeded the level of 1ppm.

Some way points were clustered in the area within the 88 to 92 longitude and 14 to 18 latitudes (Station 3). It has a proportionally high number of Hg levels within the range of 0.8 to 1ppm and 0.6 to 0.8 respectively.

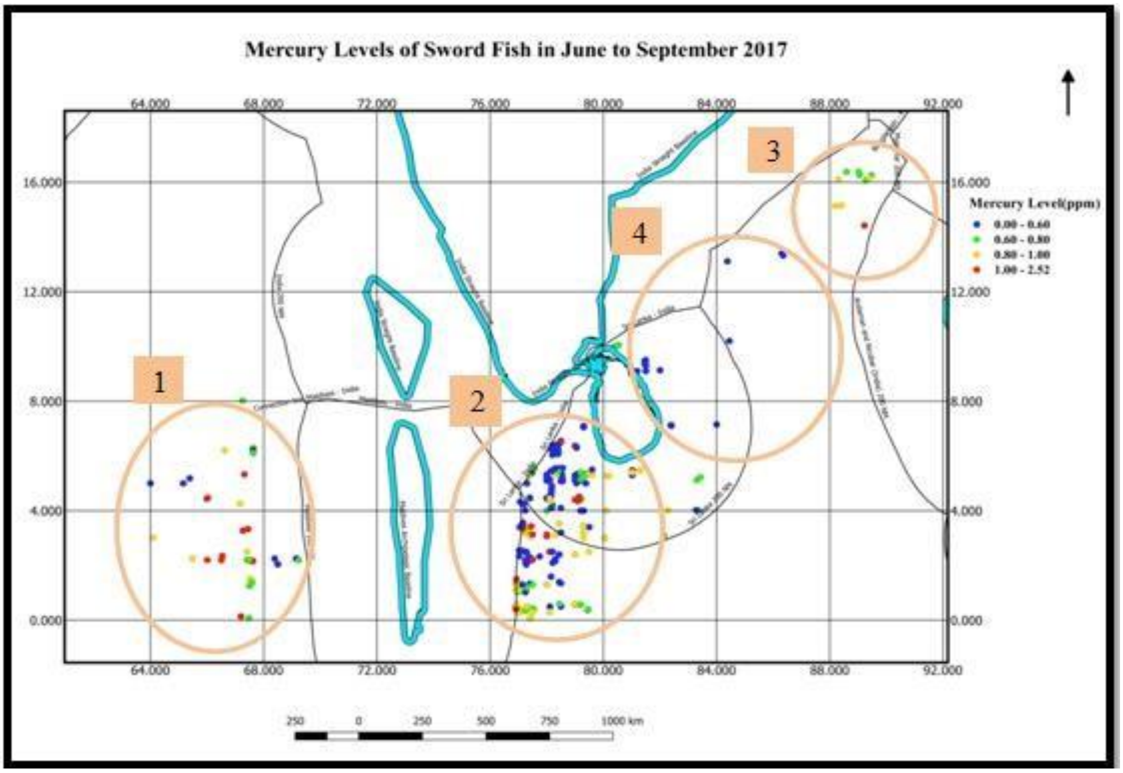

Figure 1: Mercury Levels of Swordfish

Calculating the Hg level based on harvest, station 1 represented 40.33% of Hg accumulation exceeding the concentration of 1ppm and 81.93% exceeding the 0.8ppm.

In station 2, 9.73% of Hg exceeded the concentration of 1ppm and 29.55% exceeded the 0.8ppm.Considering station 3 the Hg value exceeded the 1ppm and 0.8ppm were 10.81% and 72.97% respectively.

B. Mercury Levels of Yellowfin Tuna

Figure 2 indicates Hg levels distributed within the Sri Lankan Exclusive Economic Zone and the deep sea. Basically Hg values were distributed within the area that coordinates of 76 to 80 longitudes and 0 to 8 latitudes. The data were distributed in the North East of Sri Lanka in the coordinates of 82 to 90 longitudes and 6 to 14 latitudes. In station 1, data were within the values of 0.6 to 0.8ppm.The Hg value exceeded 0.6ppm was 20.81%.

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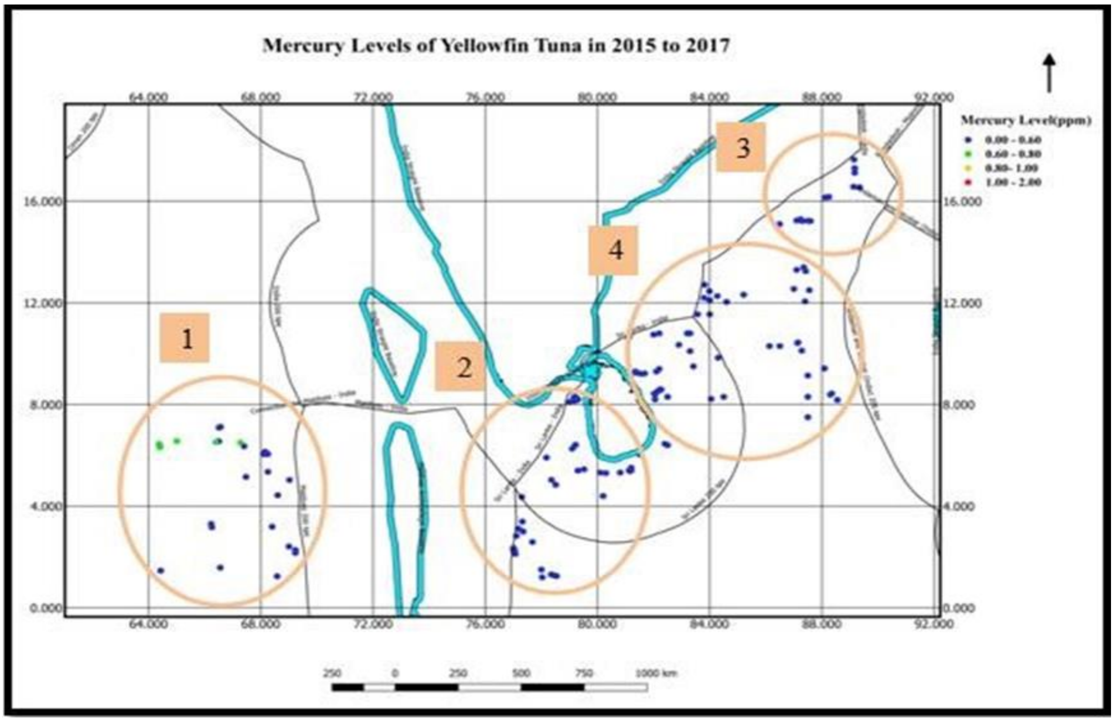

Figure 2: Mercury Levels of Yellowfin tuna in 2015 to 2017

Station 1 and 2 located in the south Asian part of the Indian Ocean and station 3 contributes to the Eastern part of Asia. The data that has exceeded the Hg critical limits was found in the Southern part of the Indian Ocean. According to the United Nations Environment Programme, India has been recognized as one of the largest Hg missioners and several numbers of power plants are driven by the coal burning and the mining industries are mainly located there. Hence, the emission of Hg into the aquatic environment is possible in the area around [20].

Considering Sri Lanka itself, most of the industries are gathered to Western provinces and as a value it is about 72.4%. Population density is highest in Colombo Metropolitan Region (CMR) [21]. A research which conducted to find out the heavy metal concentration of road deposited sediments, found those sediments contaminated with heavy metals such as Fe, Cu, Pb, Cr, Zn and Mn and these contaminations were high in closest to the industrial areas around CMR. This was mainly due to unfriendly environment practices such as poor waste management systems, unorganized drainage network and release of untreated industrial waste into the surrounding environment [22]. Many rivers including Kelani, Kalu, Benthara, Gin and Nilwala have fallen to the ocean near to station 2. These rivers are found to be contaminated with heavy metals and this may lead to increased contamination near station 2 [23].

Sediments obtained near the coast were found to have higher contamination of Hg and among the tidal flats of Mannar and the resulting Hg content was 8 ppm. Marshland peaty sediments contained a maximum of 95 ppm Hg. Oil spills, dumping waste from ships, coral and sand mining and industrial activities caused marine pollution in Sri Lanka [24]. It has found that some areas where ships are highly moving are caused by marine pollution due to illegal discharge of dirty oil and grease into the sea. These discharges are contaminated with Hg, Cr and Pb like trace minerals leading to marine pollution [25]. Colombo fort is one of busiest around the world and ranked among the top 35 ports. In 2015, 4,197 ships arrived at Colombo fort with 161, 410 thousand tons. This may lead to some contamination in the surrounding area of station 2.

Station 1 showed higher concentration among the stations. But there is no clear literature evidence related to that area. It has found that Hg is transported into the remote areas of the sea via the upwelling of water currents [26] and it can be a potential reason to increase the Hg level at station 1. Agro chemicals are used in large scale in agriculture cultivation areas extensively for a long period and this vast usage causes accumulation of heavy metals in soil properties as well with the surface water it contaminates the water streams [27]. Mahaweli region is a best example for that. Evaluating the heavy metal contamination in inland fish reservoirs found

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that fish were contaminated with Hg in North Central province and this was mainly due to agricultural chemicals that are extensively used in those areas [28].

Finding baseline data for Hg accumulation of fish caught from Bay of Bengal was found that, level of Hg has exceeded the permissible levels. Those fish types included skipjack tuna, big eye thresher shark and swordfish. Swordfish which weighed more than 40 kg were accumulated very high Hg contents in their flesh were 1 µg/g wet weight which was over the upper limit of the CODEX and EU guideline levels [23]. This provides the evidence for the higher Hg contamination in station 3.The Hg concentration distribution in the sea and oceans were different region wise and higher concentration was recorded in the Mediterranean Sea and the Northern part of the Atlantic. Lower concentration was found in the Antarctic [19].The Hg level of the sea is dependent on the ocean water currents and it is important to study the way of water current in the Indian Ocean to get the further description about the results.

C. Mercury Levels of both Swordfish and Yellowfin Tuna

Highest Hg concentrations that exceed the maximum tolerable limits were found in the Swordfish comparatively to Yellowfin tuna as indicated in Figure 3.

The United States Food and Drug Administration (2014) have reported that large predatory fish such as Yellowfin tuna and Swordfish contained higher level of Hg and comparatively Hg concentration was higher in the Swordfish [29]

Tuna and swordfish are considered as highly migratory species in the marine water. Migratory patterns mainly depend on the availability of food. Migratory behavior allows for consumption of various foods which increase the risk of bioaccumulation of heavy metal. Furthermore, Swordfish is a top predator in marine pelagic ecosystems and the bio accumulation of Hg is higher than the other pelagic fishes such as Tuna, Marlin and Sharks. Large body size and migration behavior leads to this higher accumulation of heavy metals in the Swordfish than the other fishes [30]. Mercury concentration in the fish body varied on their habitat and higher concentration showed in mesopelagic species than the epipelagic species [31]. The Swordfish and Yellowfin tuna having habitat of the mesopelagic can easily accumulate mercury in their body tissues. According to the data of global thermohaline circulation station 1 and 2 has both surface and deep water currents and food availability of that area is high. Swordfish live in warm water of the ocean and it can be possible because of having a higher population in station 1 and 2 than station 3 and 4. Swordfish have higher longevity than Yellowfin tuna and it may be a possible reason for higher bioaccumulation of Hg than yellowfin tuna [32].

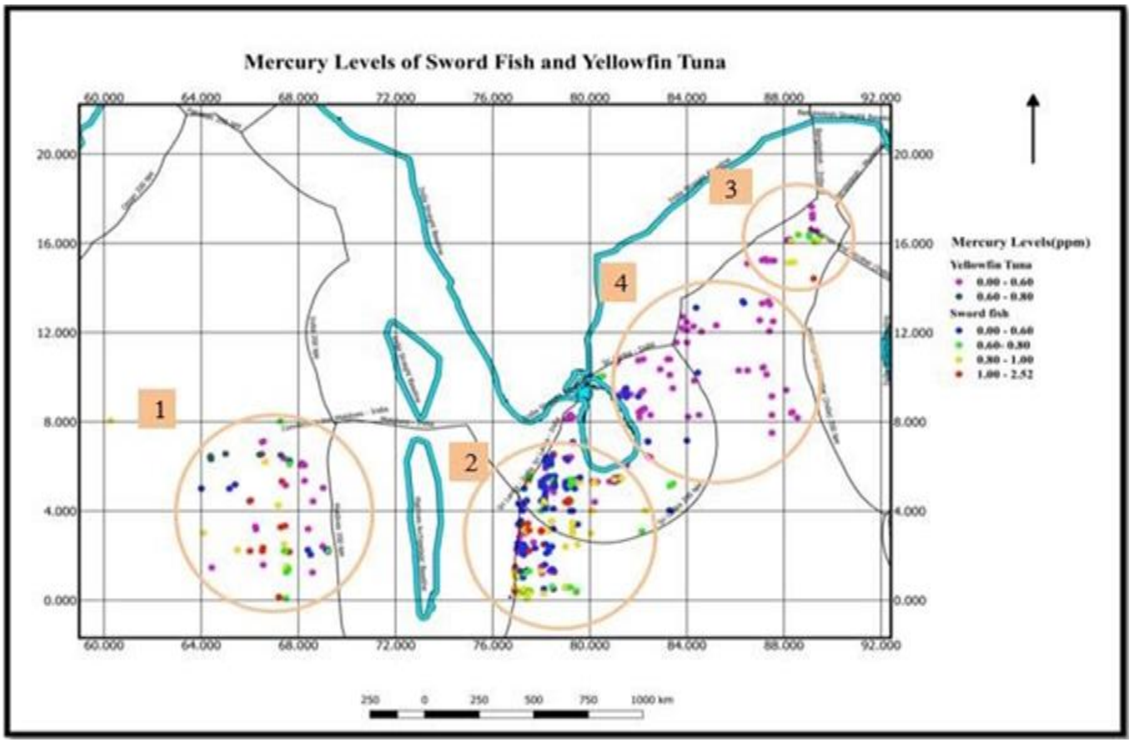

Figure 3: Mercury levels of both Swordfish and Yellowfin tuna

Results were grouped on a monthly basis to find out the seasonal variation of swordfish starting from July to August. Hg levels that exceed 1ppm in swordfish in June, July, August and September month were19.6%, 6.5%, 23.46%, 6.66% at station 2. The levels that exceed 0.8ppm were 77.27%, 9.2%, 54.18%, 36.38%. According to the results, there is no clear pattern of variation among months that was not detected. It requires having long-term results to go for more accurate predictions. Seasonal and geographical variations of heavy metal accumulation depend on many factors including migratory patterns, food availability, water currents, and rainfall patterns etc. Linking all those factors with the results will facilitate predictive modeling of heavy metal accumulation among fish bodies.

IV. CONCLUSION

Mercury was the major heavy metal accumulated among the tested fish species. Bioaccumulation was higher in Swordfish (*Xiphias gladius*) than the Yellowfin tuna (*Thunnus albacares*). Highest accumulations were detected in longitudes between 64 to 70 and latitudes of 0 to 8. The second highest mercury level was detected in the longitudes between 88 to 92 and latitudes of 14 to18. These two areas have shown a potential risk of Hg than the other areas in the Indian Ocean.

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