Heavy Metal Concentration in Tissues of Selected Common Fish Species from the Upper Ramu River in Northern Papua New Guinea

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***Abstract*- The Ramu River, one of the largest rivers in Papua New Guinea is under threat of contamination due to diversified industries situated within its catchment and drainage areas. In this study, three common fish species namely *Neoarius leptaspis, Prochilodus argentus* and *Bardodes gonionotus* caught upstream and downstream of the Ramu Nickel Mine in the Upper Ramu River basin. They have been evaluated for heavy metal content in their tissues to determine the local ecological quality. Fishes were collected by setting gill nets of various sizes overnight at one impact site and one control site. Liver and muscle tissues of fish were analysed by using Inductively Coupled Plasma – Mass Spectroscopy (ICP-MS) for all metals except Mercury (Hg) by Flow Injection Mercury Systems (FIMS) at the Australian Laboratory Services' Environmental Laboratory in Brisbane. Results indicated that concentration of all metals in muscle tissues of fish from both sites were below the FSANZ and WHO/FAO recommended standards. Five metals (Cd, Hg, Cr, Cu, and Zn) have recorded elevated levels in their liver tissues but were observed to be physiologically regulated and do not show in the muscles. Almost all the heavy metals have sources further upstream of the current study site implicating that heavy metal input into the Ramu River from Ramu Nickel Mine is minimal. The ecological quality of the aquatic environment under study maintained its natural feature while its fish resources pose no immediate environmental health danger to people depending on it. Further studies and continuous monitoring is required to determine the impact of mining activities on the current site of study because of the ongoing mining activities nearer to the area.**

***Index Terms*- Heavy Metals, Ecological Quality, Pollution, Environmental Health**

I. INTRODUCTION

Metals enter aquatic systems via natural and anthropogenic sources. Main routes include industrial, agricultural and municipal wastes discharged into water bodies by humans as well as weathering and erosion of metal rich earthy materials. Some of these metals are required for metabolic functions in humans and aquatic organisms in minimum quantities while others are not needed [1]. However, presence of elevated levels of both essential and non-essential metals can put stress on the aquatic ecology and the organisms depending on it. Presence of metals in freshwater ecosystems is a major environmental concern that has attracted widespread scientific attention. This is attributed to heavy metals' ability to persist in the environment, accumulate and magnify up the food chain and its toxicity on animals including people [1-3]. Heavy metals are naturally present in the environment. Some are essential for normal function in humans and animals [1] such as, copper, iron, manganese and zinc; whereas other metals such as mercury, cadmium and lead are not required even in small amounts by any organism [2]. Almost all metals, including the essential ones, are toxic to animals and humans if levels exceed certain thresholds [2-3].The toxicity of metals varies substantially and is largely due to their ability to interfere with enzyme-mediated processes and disruption of cellular structure [2]. Health effects in humans contaminated by elevated metal levels include neurological disorders, bone deterioration, cancer and immune system disorders [4]. From a human health perspective, the primary contaminants of concern are mercury, arsenic, lead and cadmium [5]. The major route of exposure of these metals to humans is either through direct ingestion of food, particularly seafood or indirectly through exposure to higher levels of metals [5].The potential for heavy metal contamination to affect human health negatively has been revealed through many studies conducted on heavy metal levels in fish and shellfish species, particularly in regions heavily impacted by anthropogenic inputs [6-8].

Levels of contaminants in fish are of interest not only because of the potential effects on the fish themselves, but also because of the effects on organisms that consume them, such as higher order predators and humans [9]. Guidelines on the maximum permitted levels of metals in seafood have been introduced in many parts of the world for the safe consumption of fish species [10]. Studies and monitoring programs examining heavy metal levels in fish are becoming more and more evident.

The Ramu River is one of the biggest rivers in Papua New Guinea (PNG) that has its headwaters in the Yonki district of Eastern Highlands province and flows towards the Northeast through Madang Province and discharged into the sea at the Broken Water Bay in the latter province. The river needs significant freshwater aquatic ecological attention due to the location of many important diversified industries situated within the catchment and drainage system of the Ramu River. The industries include Agro Industries (Ramu Agro-Industries), Hydro-Electricity Plant (Yonki Dam & Hydro-electricity), mining (Kainantu Gold Mine, Ramu Nickel Mine and Yandera Copper-Molybdenum Project) and Petroleum (Ramu Oil and Gas Prospect). Some of these industries are operational while others are in the developmental or exploration stages. All these industrial activities undoubtedly pose some level of environmental threat to the aquatic ecological quality of the Ramu River as their toxic waste can find their way into the river and subsequently contaminate it. Many people along the length of the Ramu River depend on this river for food, recreation, health and hygiene, transport and other livelihood sustaining activities. At the same time, the river support an array of aquatic life, some of them are important resources such as fish. The river is home to many fish species, both introduced and native species.

Considering the numerous aquatic resources presented as the biota of the river, it is quite clear to understand that people along the river use them as sustainable food source as well as selling excess catches to earn some money for other uses which adequately support them as their source of income.

All these will change as waste from the industries starts to pollute the river. Some sensitive species may migrate; others may diminish in population and biomass, while some can die out. Most importantly some polluted fishes can be consumed by the people who live along the river can experience serious health issues.

Therefore, in the present study, selected common fish species were investigated for the level of heavy metals in their muscle and liver tissues in order to determine the ecological quality and environmental health status of the selected study locality.

II. METHODOLOGY

2.1. Study Site

Study site of this research is located upstream and downstream of the Ramu Nickel-Cobalt Mine which is currently operational in open pit mining of laterite ores as shown in Figure 1 below. The GPS coordinates for both sites were taken for both upstream and downstream sites. The coordinates for upstream is 5°31’59.4”S, 145°13’26.3”E and downstream is 5°37’34.2”S, 145°17’34.2”E. The sites were selected as well as other industrial activities upstream to cover the impacts of Ramu Nickel Mine

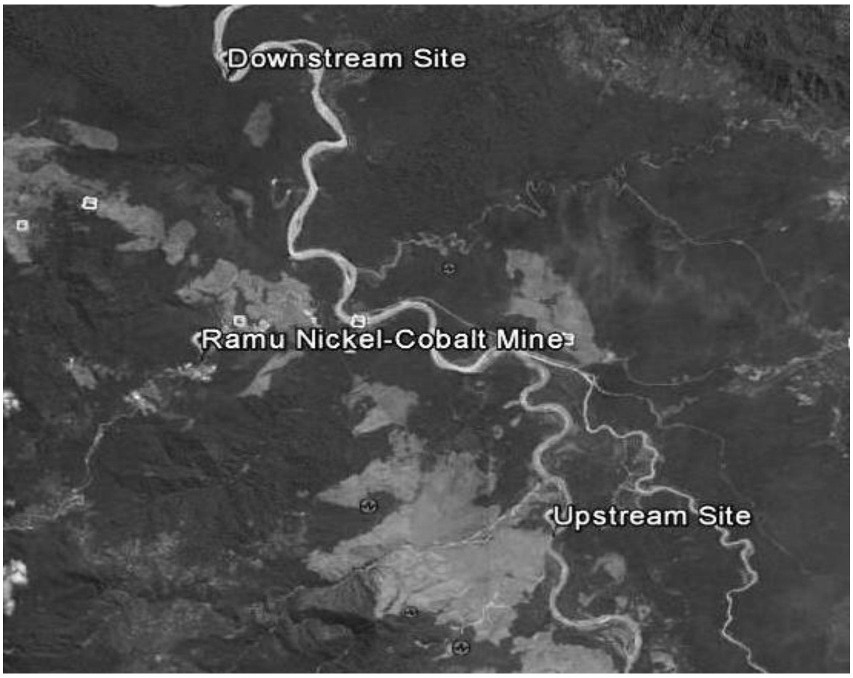

Figure 1: Location of the Study Site

2.2. Sampling

Two sampling sites were selected upstream and downstream of the Ramu Nickel mine respectively to determine level of heavy metals in fish tissues. Three fish species were selected on the basis of their catch abundance in both sites. Gill nets of various sizes (1"- 6") were used in the selected sites to catch fish. The nets were set out overnight and retrieved in the morning of the next day. Fish species were identified and tissues (lungs and muscle) were obtained by dissection of selected specimen from three species; *Neoarius leptaspis, Barbodes gonionotus* and *Prochilodus argentus* which are caught in abundance in both locations.

2.3. Instrumental Analysis

The tissue samples were analysed by Australian Laboratory Services (ALS) for total silver, arsenic, cadmium, cobalt, chromium, copper, nickel, lead, antimony, selenium and zinc utilizing Inductively Coupled Plasma – Mass Spectroscopy (ICP-MS) on a wet weight basis. Total recoverable mercury was analysed by Flow Injection Mercury Systems (FIMS).

III. RESULTS AND DISCUSSION

3.1. Summary Result

The results showed that the concentration in mg/kg of heavy metals was found in muscle and liver tissues of the 3 common fish species under study; *Neoarius leptaspis*, *Prochilodus argentus*, and *Barbodes gonionotus* from the upper Ramu River area. These results are viewed against the WHO and FAO Food Standards of Heavy metal concentrations to measure the level of heavy metals in the fish species and show how safe they are for consumption as well as the determining the aquatic ecological quality of the riverine ecosystem in the study locality.

Table 1: Concentration of heavy metals in tissues of selected fish species.

	Downstream of Ramu Nickel Mine						Upstream of Ramu Nickel Mine						
	N.leptaspis		*P.agentus*		*B.gonionotus*		*N.leptaspis*		*P.agentus*		*B.gonionotus*		WHO/FAO
Metal	Muscle	Liver	Muscle	Liver	Muscle	Liver	Muscle	Liver	Muscle	Liver	Muscle	Liver	
Sb	<0.1	<0.1	<0.1	<0.3	<0.1	<0.3	<0.1	<0.1	<0.1	<0.1	<0.1	<0.3	1
As	<0.05	0.15	0.12	<0.13	0.005	0.68	<0.05	0.13	0.08	0.1	<0.05	0.53	1.4
Cd	<0.01	1.04	<0.01	0.42	<0.01	0.81	<0.01	**2.08**	<0.01	0.35	<0.01	**2.3**	2
Cr	<0.05	0.06	0.19	0.34	0.46	**1.16**	0.15	<0.06	<0.05	0.15	<0.05	0.82	1
Co	<0.05	0.08	<0.05	<0.13	0.12	2.36	<0.05	0.12	<0.05	<0.06	<0.05	0.82	
Cu	0.1	3	0.2	**89.2**	0.1	**95.9**	0.1	3.7	0.1	24.3	0.1	**556**	30
Pb	<0.05	0.09	<0.05	<0.13	0.08	0.59	<0.05	0.05	<0.05	<0.05	<0.05	0.24	6
Ni	<0.05	<0.06	<0.05	<0.13	<0.05	0.52	<0.05	<0.06	<0.05	<0.06	<0.05	<0.13	10
Se	0.11	3.37	0.78	5.78	0.42	3.26	0.25	4.73	0.59	2.53	0.38	4.29	
Ag	<0.1	<0.1	<0.1	<0.3	<0.1	1.1	<0.1	<0.1	<0.1	<0.1	<0.1	3.5	
Zn	5.9	**652**	3.5	**100**	3.9	**120**	8	**328**	3.9	42.4	2.9	72.6	100
Hg	<0.1	**2.1**	<0.1	**0.2**	<0.1	**1.2**	<0.1	**2.7**	<0.1	**0.2**	<0.1	**0.5**	0.1

A general trend observed in this study is the significant variations in the level of heavy metals in liver and muscle tissues (**Table 1**). Levels of heavy metals in the liver are higher than that of muscles irrespective of the type of fish and site of catch. The higher level of trace metals in the liver relative to muscles may be attributed to the high coordination of metallothionein protein with the metals [11].This group of protein have the capacity to bind both essential (such as zinc, copper, selenium) and non-essential (such as cadmium, mercury, silver, arsenic) heavy metals [12]. 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 [13].

3.2 Metals in Muscle

Concentrations of metals Sb, As, Co, Pb, Ni, Se and Ag in both liver and muscle tissues fall below the WHO/FAO food quality guidelines for all species caught on both upstream and downstream of the Ramu Nickel mine. Those metals are present in the riverine system at insignificant quantities.Thus, the upper Ramu River area is under no immediate threat of contamination by the aforementioned metals which mean that the health risk of consuming fish contaminated by these metals is negligible. Attempts to obtain the WHO/FAO standards for metals Co, Se and Ag failed so these metals were not discussed.

3.3 Metals in Liver

Graphs in the ensuing pages show the metals in liver tissues that exceeded WHO/FAO food quality guideline standards. Metals such as Cd, Cr, Cu, Zn and Hg were discovered to be accumulated to various levels in the liver tissues of all fish species studied at both sites whereas in muscles, the metal levels were lower than that of WHO/FAO guideline standards (**Table 1**).

Cadmium (Cd)

Cadmium is widely known to be a highly toxic non–essential heavy metal and it does not have a role in biological processes in living organisms. Cadmium could be readily bio accumulated in lower portion of food chain and bio-concentrated in multiple organs of fish [14].Thus, even at its low concentration, cadmium could be harmful to living organisms [15].

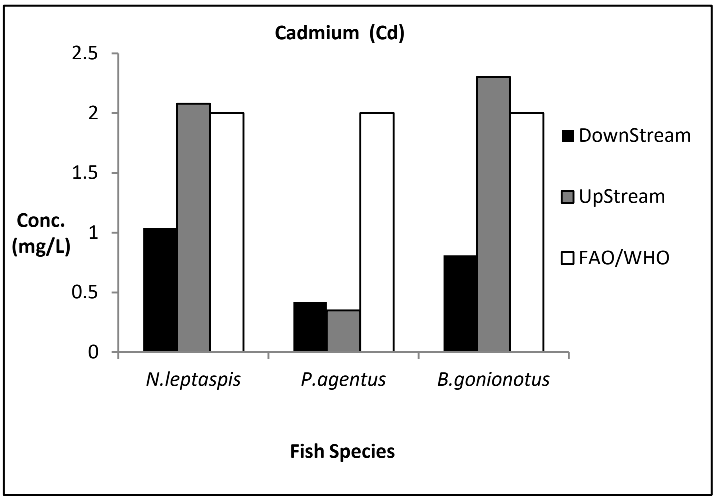

Figure 2: Level of Cadmium in Liver Tissues.

In the current study, Cd levels in the muscle tissues of all species from all sites fall below the detectable limit (<0. 01 mg/kg). Although Cd seemed to accumulate in the liver tissues of fish irrespective of species and locality of catch, they are below the WHO/FAO standard concentration of 2 mg/kg except *Neoarius leptaspis* and *Barbodes gonionotus* caught from the upstream site which accumulate Cd in its liver tissues that exceeded the standard values. Liver tissues of these fishes contain 2.08 mg/kg and 2.3 mg/kg respectively (**Figure 2**), exceeding the food quality standard of 2 mg/kg. Since their site of catch is upstream of the Ramu Nickel Mine, it is believed that presence of Cd in the Upper Ramu River is caused by other industrial activities further upstream. Consumption of these fishes with liver can compromise the health of humans. It also means that fish in the study area can develop ailments related to high Cd levels.

Mercury (Hg)

Mercury is also a non-essential heavy metal and its concentration in liver tissues of fish samples from both sites of the current study presented some serious concerns. All the fishes accumulate Hg in their liver that exceeded the WHO/FAO standard of 0.1 mg/kg (**Figure 3**). *Neoarius leptaspis* recorded the highest concentration in its liver with 2.7 mg/kg from upstream samples and 2.1mg/kg from the downstream catches. *Barbodes gonionotus* followed by 1.2mg/kg and 0.5kg for downstream and upstream respectively. Slightly lower concentrations were observed in *Prochilodus argentus* with 0.2 mg/kg apiece for both sites.

However, all these concentrations are above the WHO/FAO standard and can potentially cause adversities in the riverine ecology as well as cause toxicity ailment to people upon consumption. Mercury is recognized as a highly toxic metal and stringently regulated by waste discharge regulators [14].

Movement of Hg (II) into aquatic ecosystem and its bioaccumulation as methyl mercury in higher trophic levels are strongly influenced by the uptake of bioavailable forms of Hg (II). Fish obtained methylated mercury through dietary uptake, which could be influenced by size, diet, ecological and environmental factors [16].

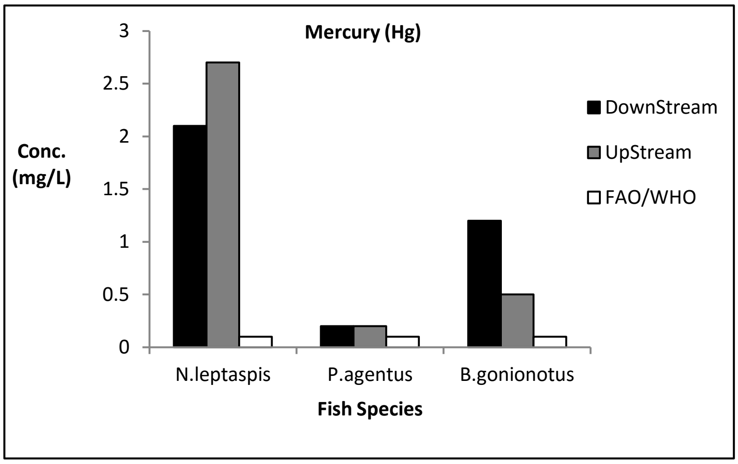

Figure 3: Level of Mercury in Liver Tissues

Chromium (Cr)

Chromium is considered as a heavy metal and pollutant as well as an essential micronutrient.

Chromium exists in different oxidation states which have distinct biological effects [17]. Trivalent form of Chromium is 500 to 1000 times less active against living cells because of its poor uptake [18].

Hexavalent chromium (Cr^{6+}) is a well known carcinogen metal form for animals and human beings. Cr (VI) compounds readily penetrate into cell membranes via anion transport systems. It was clear from previous studies that Cr (VI) itself was highly active and carcinogen should it arrive as Cr (VI) inclusion to the target [19-20].

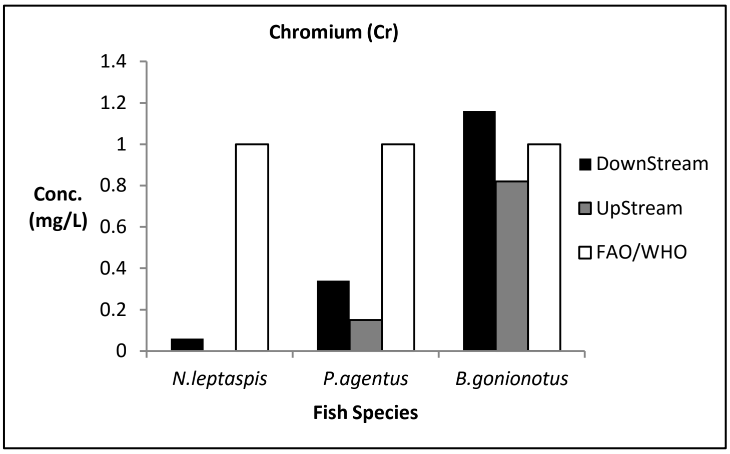

Figure 4: Level of Chromium in Liver Tissues.

In this study, concentration of Cr in liver and muscle tissues of all species from both sites are below the WHO/FAO standard of 1mg/kg except the liver of *B.gonionotus* from the downstream site which contained 1.16mg/kg in concentration. This amount exceeds the recommended value. Another significant observation is the concentration of Cr in liver (0.82mg/kg) of the same species from the upstream site.

Increased concentration from the downstream site can be attributed to the input from the Ramu Laterite Nickel deposit which host Chromite mineral in the ore matrix.

The presence of Chromium in fish tissue is contributed by both natural processes and mine induced means. Surface runoff from both untouched and mined areas can transport laterite soil containing quantifiable Chromite into waterways that are discharged into the Ramu River by its tributaries that are within close proximity to the laterite deposit and mine area. Thus, Cr is accumulated in fish via ingestion where the Cr is available in the feeding cycle of *B.gonionotus*. However, liver of the same species from the upstream site do contain significant amount of Cr. This can mean that *B.gonionotus* has the affinity to bioconcentrate Cr. Additionally, it can also mean that this species is highly mobile and can travel great distances to feed in comparison to other species which are observed to contain relatively low amounts of Cr for both sites.

Copper (Cu)

Copper is an essential metal in fish and is regulated in the muscle tissue [16]. In the current study, Cu concentrations in muscle tissues of all fish from both sites are below the WHO/FAO recommended value of 30 mg/kg. However, concentrations in the liver tissues of *P.argentus* and *B. Gonionotus* exceeded the standard value. Liver of *B.gonionotus* from the upstream site shows a concentration of 556 mg/kg while same tissue from the downstream site has a value of 95.9 mg/kg; both of them exceeded the WHO/FAO recommended concentration. Liver of *P.argentus* from downstream site has liver concentration of 89.2 mg/kg which also exceeded the standard.

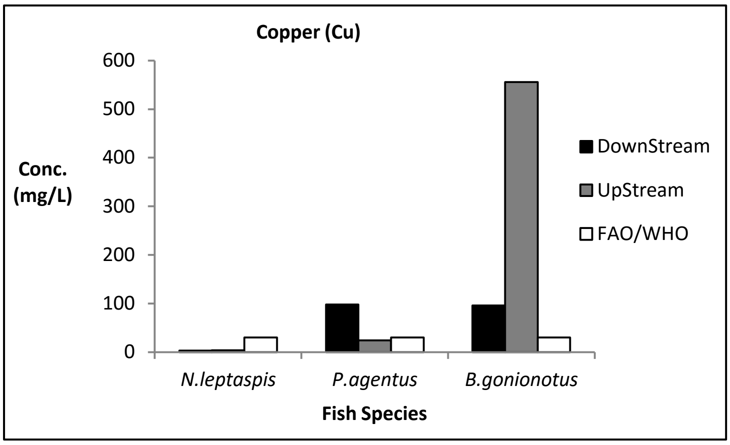

Figure 5: Level of Copper in Liver Tissues.

A trend observed for Cu concentrations showed that muscle tissues contain relatively low levels than livers of all common fish species from all sites (**Table 1**). This observation indicates that fish from the Upper Ramu River are able to physiologically regulate Cu in their bodies. More metals are found in the liver because this organ is responsible for detoxification of toxic substances including heavy metals [12], protecting the muscles from accumulating Cu in high concentrations which can cause toxicity ailments in fish. Thus, this scenario concur with established literature that fish are generally able to regulate essential metals to maintain optimum levels and prevent toxicity [1].

Another aspect of concern is the concentration of Cu in liver respective to the location of catch. *B.gonionotus* of the upstream site has the highest liver concentration of Cu with 556 mg/kg compared to 95.9 mg/kg for the same species downstream and 89.2 mg/kg in liver of *P.argentus* from downstream. All other fishes from both sites also show the same trend; Cu concentration is high in liver of fish from upstream and low in downstream as shown in **Table 1**. Therefore, it is obvious that the source of Cu is upstream of the Ramu Nickel Mine. Possible sources include the Yandera Cu-Mo deposit and the Kainantu Gold Mine or it can come from Cu based agrochemicals used by Ramu Agri-Industries.

Zinc (Zn)

Zinc is also an essential element in food and environment [21]. Possibility exists both for a deficiency and for an excess of this metal. For this reason it is important that regulatory criteria for zinc, while protecting against toxicity, are not set so low as to drive zinc levels into the deficiency area.

In the current study WHO/FAO standard (100 mg/kg) for Zn in food is used on fish tissues. Concentration of Zn in liver of *N.leptaspis* from both upstream and downstream of the Ramu Nickel Mine exceeds the standard concentration of 100mg/kg.

Fish liver tissue concentration is 652 mg/kg for downstream site and 328 for upstream site. The other two fish species; *P.argentus* and *B.gonionotus* have Zn concentrations in their liver that exceeded the standard value at the downstream site. The concentration in their liver tissue is 120 mg/kg and 328 mg/kg respectively.

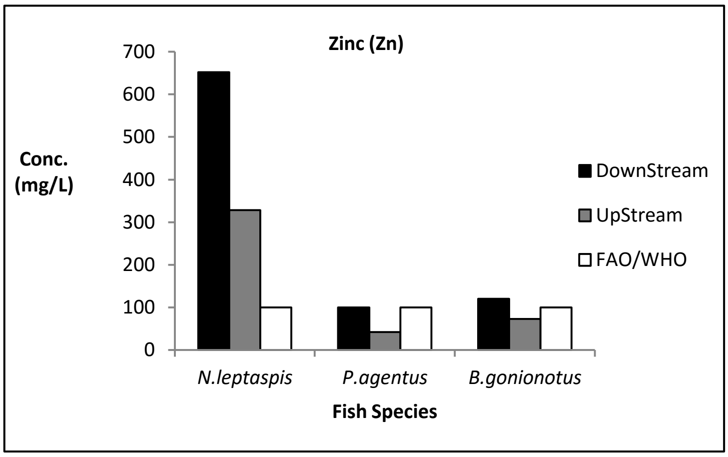

Figure 6: Level of Zinc in Liver Tissues.

The assimilation of Zn in fish provides the indication that the Ramu Nickel Mine partly contributes some Zn into the main Ramu River system. This is supported by the fact that all fish species from the downstream site showed elevated levels of Zn in their liver tissues. Only one species from the upstream site has high Zn concentration in its liver which means that there is also a potential source further upstream.

3.4 Impact of Ramu Nickel Mine on Fish

The source of metals in fish seems to be natural as all metals (Cd, Hg, Cu, Zn) were detected in elevated levels in both upstream and downstream sites except Cr which concentrations is believed to be affected by the mining activity as well as natural factors. This is because it was found to be higher in liver tissues of two fish species from the downstream sites. The mine is extracting laterite ores which is rich in Cr, a metal that is separated as Chromites at the mine site. Erosion of disturbed soil at the mine can carry Cr rich soil into the tributaries of Ramu River and eventually ingested by fish as indicated here.

However, natural erosion before mining can also play a part in the distribution of Cr in the riverine ecosystem so it is too early to ascertain that the high level of Cr in fish is attributable to the mining activities as yet.

IV. CONCLUSION

The level of metals in fish tissues indicate that five metals (Cd, Hg, Cr, Cu, Zn) are present in significant quantities in the riverine ecosystem of the Upper Ramu River area as indicated by their elevated presence in the liver of fish from both upstream and downstream of the Ramu Nickel Mine but those heavy metals cause no environmental health risk to people or other animals living off the river as demonstrated by the lower levels in muscle tissues which is the dominant food source. Avoiding the consumption of liver and gills of fish is recommended for people to evade toxicity induced ailments.

Fish species living in the study area seemed to adapt to their environment very well especially developing advanced excretion or physiological heavy metal regulation traits as high level of metals in the liver is not being found in the muscle tissues.

At this stage, it is conclusive to state that the presence of metals in the study area and its fisheries resource is largely controlled by natural sources.
The mining has minimal to no impact on the metal levels in fish as well as the aquatic ecology. Thus, at present the environmental health risk is low and ecological quality remains within acceptable natural states. However, continuous monitoring is required as heavy metal pollution from the Ramu Mine is possible as the mining activities continue into the years to come.

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