

# Non-carcinogenic health risks of heavy metal in mudfish from Laguna Lake

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## ABSTRACT

This paper examines the potential risks to human health associated with exposure to heavy metal that have bioaccumulated in Mudfish (*Ophicephalus striatus*) from Laguna Lake. Fish samples were collected in eight sampling stations in three major areas of the lake during the dry and wet seasons. Dry season samples were collected from May to June 2010 and wet season samples, from September to November 2010. Coordinates of sampling site locations were recorded using Global Positioning System (GPS) and plotted in Geographic Information System (GIS) digital maps. Heavy metal analyses for cadmium (Cd), lead (Pb), mercury (Hg), arsenic (As), and chromium (Cr) were conducted using Atomic Absorption Spectrophotometer (AAS) and a Mercury Analyzer (Mercur-Duo). Estimates of health risks associated with mudfish consumption were summarized according to non-carcinogenic effects. Non-carcinogenic Hazard Quotient (NHQ) values of five heavy metal showed that lead is the most urgent pollutant of concern in terms of adverse health effects from risks associated with mudfish consumption from all sampling locations in the lake. From the point of view of human health protection and disease prevention, mudfish from Laguna Lake is not fit for long-term human consumption primarily due to lead and mercury contamination.

*Keywords:* Bioaccumulation, health risk assessment, heavy metal, Laguna Lake, mudfish

## INTRODUCTION

Heavy metals from natural sources and anthropogenic activities are continually released into aquatic environment, causing serious threat due to their toxicity, bioaccumulation, long persistence and bio-magnification in the food chain. Fish is considered as one of the most significant indicators in freshwater ecosystems for estimating extent of trace metals pollution (Yousafzai et al., 2010).

With growing urbanization and industrialization, there has been a rapid increase in domestic and industrial wastewater which has intensified environmental pollution in different environmental compartments. The major sources of contamination in surface waters can be traced to industrial discharges, domestic waste disposal and application of agrochemicals on farmlands, among others. Pollutants like heavy metal, after entering into aquatic environment, accumulate in tissues and organs of aquatic organisms (Akan et al., 2009). These contaminants entering the aquatic ecosystem may not directly damage the organisms but they can be deposited into aquatic organisms through the effects of bioaccumulation, biomagnifications and food chain process and eventually threaten the health of humans through fish consumption (Lakshmanan et al., 2009).

Biomagnification of trace elements in living organisms describes the processes and pathways of these potential pollutants from one trophic level to another. Increasing concentration through the food chain causes higher retention time of toxic substances than that of the other normal food components (Sreedhara Nayaka et al., 2009). Fish being situated at high trophic level of the food web may accumulate large amounts of heavy metal from the water and often in concentrations several times higher than in the ambient water (Yousafzai et al., 2010). Some of the metals found in the fish might be essential as they play important role in biological system of the fish as well as in human beings. However, some of them may also be toxic and might cause serious damage in human health even in trace amount at a certain limit or threshold (Hosseinkhezri et al., 2011).

In contrast, fish has been known for its reputation as the established health food for most of the world's

population, particularly in developing countries, as compared to meat, poultry and eggs. The protein content in fish averages from 15 to 20 percent; hence fish provides comparatively cheap and readily available protein sources in complement with long chains of n-3 fatty acids, amino acids, vitamins and minerals that further contribute to healthy nutritional options for a balance dietary intake (Nor Hasyimah et al., 2011). Health risk assessment of heavy metal bioaccumulation in fish therefore is highly important to establish scientific basis for understanding risks versus benefits of fish consumption.

The Philippine Millennium Ecosystem Assessment Sub-global Assessment for Laguna Lake emphasized that the Laguna Lake Basin is a classic model of a multiple resource with multiple users. Its capacity to provide various ecosystem services to various users is continuously being challenged mainly by anthropogenic factors. Deforestation of its watersheds in favor of other uses such as agriculture, industry, and human settlements is expected to cause an imbalance in the lake hydraulic processes. Lake water quality has deteriorated through the years due to various point sources of pollution from industry, agriculture, and domestic sources. Detection of traces of heavy metal like copper, cadmium, chromium, and lead in the water and sediment is a major concern for human health. (Ecosystems and People: The Philippine Millennium Ecosystem Assessment, Sub-global Assessment, Lasco, Espaldon & Tapia, 2005). The main objective of the study is to assess the risks to human health associated with the exposure to heavy metal bioaccumulation of Mudfish (*Ophicephalus striatus*) from Laguna Lake.

## MATERIALS AND METHODS

### Sampling zones and sites

Laguna Lake being the largest inland body of water in the Philippines with surface area of approximately 900 square kilometers was arbitrarily divided into five sampling zones: namely, Northern West Bay, Central West Bay, Central Bay, South Bay, and East Bay. Mudfish samples were collected from each of the five designated sampling zones in the lake. There were two

sampling sites each for Northern West Bay, Central West Bay, and Central Bay; and one sampling site each for South Bay, and East Bay; for a total of eight sampling sites. The summary of sampling zones and sites is shown in Table 1.

The coordinates of the sampling locations of the eight stations in the different zones were recorded using a Global Positioning System (GPS) instrument and plotted in Geographic Information System (GIS) digital maps. The locations and coordinates of the sampling sites are shown in Table 2. This facilitated re-sampling activities and ensured that subsequent samples for the wet season were collected in the area as that of the dry season

samples. A GIS map of Laguna de Bay showing the sampling sites is shown in Figure 1.

### Sampling frequency

There were two batches of mudfish (*Ophicephalus striatus*) samples collected using fish net. The first batch of fish samples was collected in May to June 2010 to represent the dry season conditions in the study area. The second batch was collected during the months of September to November 2010 to represent wet season conditions. Samples were collected for both seasons to determine potential variations in health risk.

**Table 1. Sampling zones and sites**

SAMPLING ZONES	NAME	NUMBER OF SAMPLING SITE/S
1	Northern West Bay	2
2	Central West Bay	2
3	Central Bay	2
4	South Bay	1
5	East Bay	1
	TOTAL	8

**Table 2. Sampling site locations and coordinates**

FISH SAMPLING SITE	LOCATION	COORDINATES
<b>1A</b> (Binangonan)	Northern West Bay	N 14° 28' 57.8'' E 121° 09' 22.6''
<b>1B</b> (Taguig)	Northern West Bay	N 14° 27' 50.6'' E 121° 05' 19.3''
<b>2A</b> (Talim Island)	Central West Bay	N 14° 22' 34.1'' E 121° 12' 03.6''
<b>2B</b> (Sta Rosa)	Central West Bay	N 14° 22' 43.4'' E 121° 04' 30.1''
<b>3A</b> (Jala-Jala)	Central Bay	N 14° 22' 43.9'' E 121° 19' 25.5''
<b>3B</b> (Cardona)	Central Bay	N 14° 28' 13.5'' E 121° 13' 19.4''
<b>4</b> (Calamba)	South Bay	N 14° 11' 41.4'' E 121° 11' 43.5''
<b>5</b> (Pakil)	East Bay	N 14° 22' 12.9'' E 121° 25' 28.8''

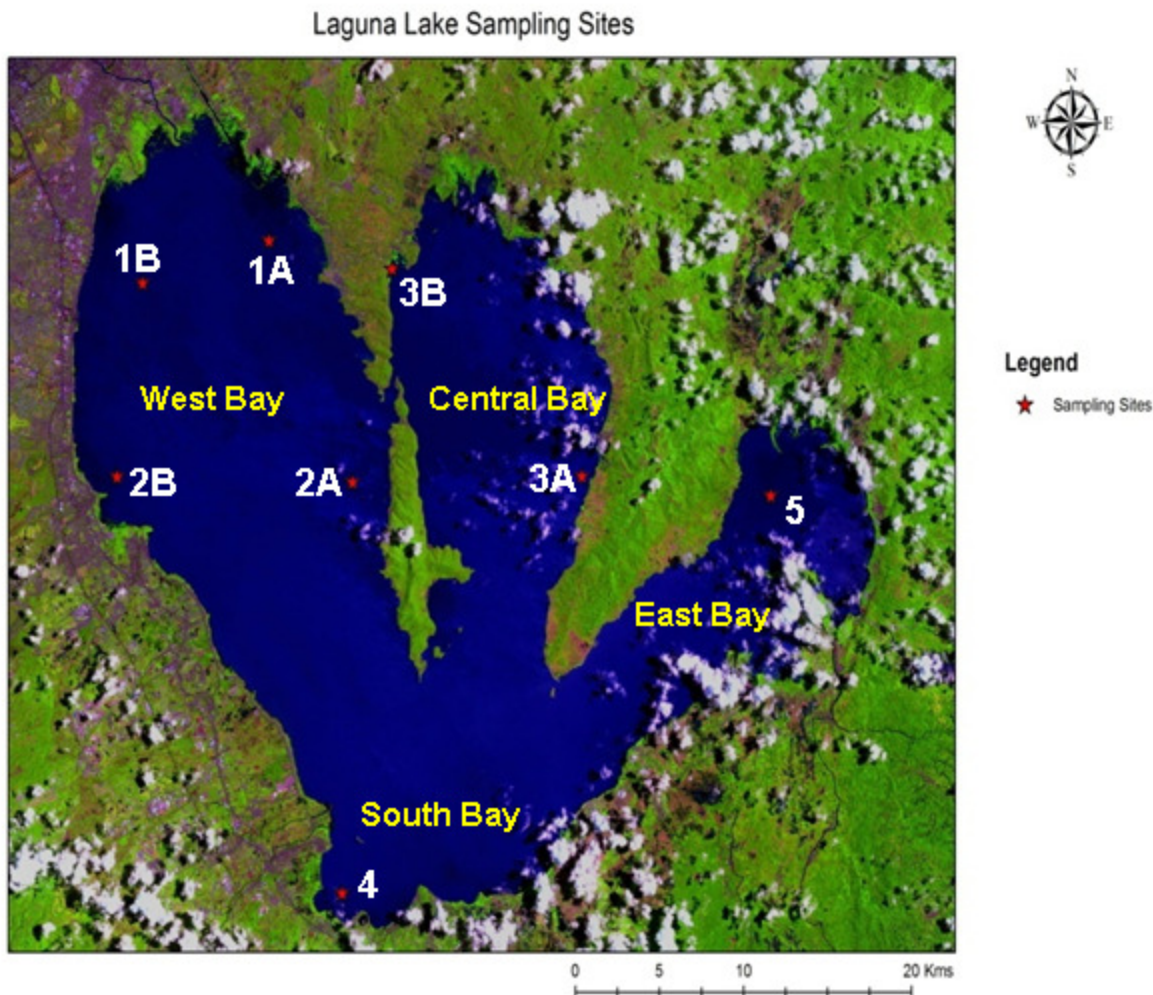


Figure 1. Location of sampling sites (GIS map)

### Heavy metals included in the study

The heavy metals included in the study were cadmium (Cd), lead (Pb), mercury (Hg), arsenic (As), and chromium (Cr). These non-essential metals from the point of view of human health are also known to have the ability to bioaccumulate through the food chain.

### Sample packaging and preservation

Fish samples were individually wrapped in a waterproof plastic sampling bag. The edible portions of the fish samples were processed on-site to avoid puncturing of the packaging material by fish spines. Individual fish samples were sealed in three layers of plastic bags. Each sample was provided with identification tag and sample code. After packaging, samples were kept in

an ice chest (with ice) and immediately brought to the Industrial Technology Development Institute, Department of Science and Technology Laboratory.

### Laboratory procedures and analysis

Samples submitted to the laboratory were stored in freezer until all the samples had been collected to ensure uniform sample preparation. Prior to analyses, samples were thawed then osterized for homogeneity. Replicates were prepared and all quality control parameters were conducted to ensure integrity of the analyses. Cadmium, chromium and lead were analyzed using the AAS (Atomic Absorption Spectrometer). The sample solutions were aspirated into a flame and atomized.

Arsenic analysis involves the generation of arsine gas by reacting the arsenic in the sample with sodium borohydride. Reaction takes place in a hydride generation assembly that is attached to an AAS system. Mercury was analyzed using the Mercur-Duo Mercury Analyzer, a single-beam instrument with a mercury low-pressure lamp as a light source for the excitation of mercury atoms, and a photomultiplier to record the fluorescent or absorption radiation.

## RESULTS AND DISCUSSION

The results on the heavy metal concentrations in the edible portions of collected mudfish are divided into two: (1) Heavy metal levels in mudfish for dry season, and (2) Heavy metal levels in mudfish for wet season.

### Heavy metal levels in mudfish for dry season

Table 3 shows the concentrations of heavy metals (Cd, Cr, Pb, Hg and As) in mudfish from eight sampling stations during the dry season. Cadmium (Cd) concentration ranged from 0.03742 mg/kg in sampling station 3A to 0.0695 mg/kg in station 2B. Chromium (Cr) ranged from 0.02243 mg/kg in station 5 to 0.42589 mg/kg in station 3A. Lead (Pb) ranged from 0.58037 mg/kg in station 2A to 2.80447 mg/kg in station 4. Mercury (Hg) ranged from 0.00314 mg/kg in station 4 to 0.17685 mg/kg in station 3B. Arsenic (As) ranged from 0.00003 mg/kg in station 3B to 0.36976 mg/kg in station 1A. Figure 2 shows the spatial distribution of

heavy metal concentrations in mudfish during the dry season.

### Heavy metal levels in mudfish for wet season

Table 4 shows the concentrations of heavy metals (Cd, Cr, Pb, Hg and As) in mudfish from eight sampling stations during the wet season. Cadmium (Cd) concentration ranged from 0.00241 mg/kg in sampling station 5 to 0.30122 mg/kg in station 1B. Chromium (Cr) ranged from 0.00148 mg/kg in station 3B to 0.29289 mg/kg in station 1B. Lead (Pb) ranged from 0.007 mg/kg in station 2B to 4.41776 mg/kg in station 1A. Mercury (Hg) ranged from 0.01192 mg/kg in station 2B to 0.07668 mg/kg in station 1A. Arsenic (As) ranged from 0.001 mg/kg in stations 2A and 3B, to 0.04696 mg/kg in station 1B. Figure 3 shows the spatial distribution of heavy metal concentrations in mudfish during the wet season.

Analysis of laboratory data for the dry and wet seasons showed that the onset of the rainy season can either increase or decrease the heavy metal concentrations in mudfish depending on where the fish was located in the lake. The positive effect of the rainy season could be due to the dilution of rainwater run-off which was apparent in the South Bay, Central Bay and East Bay. On the other hand, the negative effect of the rainy season could be due to the "flushing-effect" from tributaries and run-off from adjoining areas with significant sources of heavy metal in the environment.

Table 3. Heavy metal concentration, mudfish (mg/kg), dry season

Sampling Site (Dry Season)	Heavy Metal Concentration mg/kg DS				
	Cd	Cr	Pb	Hg	As
1A	0.04243	0.04833	1.09690	0.03574	0.36976
1B	0.06087	0.08773	0.65377	0.17506	0.03194
2A	0.03947	0.06029	0.58037	0.09953	0.15524
2B	0.06950	0.11788	0.85857	0.03432	0.22737
3A	0.03742	0.42589	1.11713	0.10578	0.08388
3B	0.04859	0.03203	0.74580	0.17685	0.00003
4	0.03782	0.03707	2.80447	0.00314	0.11371
5	0.06180	0.02243	1.15327	0.01630	0.21853

Table 4. Heavy metal concentration, mudfish (mg/kg), wet season

Sampling Site (Wet Season)	Heavy Metal Concentration mg/kg WS				
	Cd	Cr	Pb	Hg	As
1A	0.22665	0.06178	4.41776	0.07668	0.03807
1B	0.30122	0.29289	4.10531	0.03514	0.04696
2A	0.01017	0.00314	0.47623	0.02420	0.00100
2B	0.00245	0.00469	0.00700	0.01192	0.00718
3A	0.00386	0.00532	0.01104	0.05341	0.00100
3B	0.01161	0.00148	0.10115	0.03584	0.00100
4	0.00278	0.00311	0.05372	0.03560	0.02558
5	0.00241	0.00301	0.02392	0.01241	0.02755

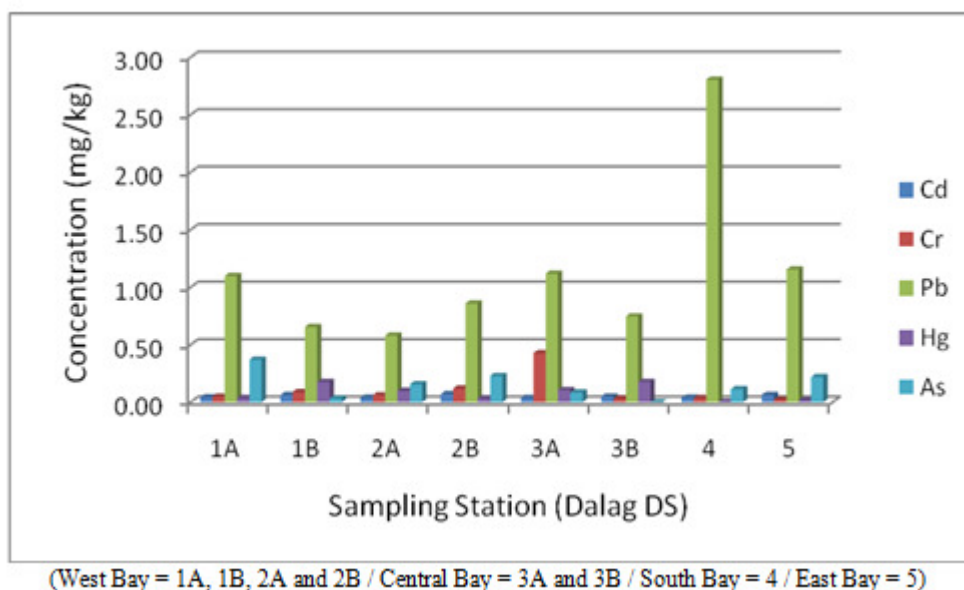


Figure 2. Heavy metal concentrations, mudfish (mg/kg), dry season

This was observed in the West Bay where lead was highest during the wet season.

**Estimate of potential human exposure to heavy metal in mudfish (*Ophicephalus striatus*)**

The basic equation for calculating systemic toxicity (non-carcinogenic hazard) is:

$$\text{Non-carcinogenic Hazard Quotient (NHQ)} = \text{CDI/RfD}$$

Where:

CDI = Chronic daily intake for the toxicant expressed in mg/kg-day

RfD = Chronic (oral) reference dose for the toxicant expressed in mg/kg-day

Chronic oral RfD is defined as an estimate (with uncertainty spanning perhaps an order of magnitude or greater) of a daily oral exposure level for the human population, including sensitive subpopulations, that is

likely to be without an appreciable risk of deleterious effects during a lifetime. Chronic oral RfDs are specifically developed to be protective for long-term exposure to a compound. As a guideline, chronic oral RfDs generally should be used to evaluate the potential non-carcinogenic effects associated with exposure periods greater than 7 years (approximately 10 percent of a human lifetime). Chronic oral reference doses are expressed in units of mg/kg-day. The RfD values of heavy metal in this study were adopted from USEPA. For Arsenic, RfD=0.0003 mg/kg-day, Chromium, RfD=0.003 mg/kg-day, Mercury, 0.0001mg/kg-day, Cadmium, RfD=0.001 mg/kg-day, and Lead, Rfd=0.0000001 mg/kg-day.

Non-Carcinogenic Fish Ingestion Equation: CDI

$$CDI = \frac{C \times EF \times ED \times IRF \times (kg/1000g)}{(365 \text{ days/year}) \times LT \times BW}$$

Where:

CDI = Chronic daily intake for the toxicant expressed in mg/kg-day

C = Concentration of heavy metal in fish (mg/kg)

BW = Body weight (For Filipino adult ~ 65kg)

ED = Exposure duration (30 years)

EF = Exposure frequency (350 days per year)

IRF = Ingestion rate fish (fish consumption) = 102.74 g/day (FAO). This is the estimated average daily per capita consumption of fish in the Philippines from the FAO Fisheries and Aquatic Department.

LT = Lifetime (average), 30 years for non-carcinogenic health effects

The Non-carcinogenic Hazard Quotient (NHQ) is one of the measures of non-carcinogenic health effects of exposure to chemical contaminants. It is the ratio of an exposure level by a contaminant to a reference dose or value selected for the health risk assessment of a particular substance or chemical. If the exposure level is higher than the toxicity value, then there is the potential for risk to the receptor. Computed NHQ value of greater than 1.0 indicates that the exposure to a single chemical or substance will likely result in adverse health effects. The potential health effects are dependent on the type of chemical or substance of concern. NHQ values of 1.0 or below indicate that daily oral exposure level for the human population, including sensitive subpopulations, is likely to be without

an appreciable risk of deleterious effects during a lifetime.

Computed values of Chronic Daily Intake (CDI) and Non-carcinogenic Hazard Quotient (NHQ) of cadmium, chromium, lead, mercury and arsenic in mudfish for all sampling stations during the dry season are summarized in Table 5. NHQ values for cadmium, chromium, and arsenic are less than 1.0 (unit less value) in all sampling stations, showing that the daily oral exposure level for the human population, including sensitive subpopulations, is likely to be without an appreciable risk of deleterious effects during a lifetime. However, NHQ values for mercury have values ranging from 1.36 to 2.41 indicating that long-term mudfish consumption would likely result in adverse health effects. NHQ values of lead in all sampling stations are way above 1.0 (ranging from 8,796 in sampling station 2A to 42,506 in station 4), indicating high risk for adverse human health effects associated with long-term mudfish consumption.

For the wet season, computed values of Non-carcinogenic Hazard Quotient (NHQ) of cadmium, chromium, lead, mercury and arsenic in mudfish for all sampling stations are summarized in Table 6. As with the dry season findings, the NHQ values for cadmium, chromium, and arsenic were less than 1.0 in all sampling stations, showing that the daily oral exposure level for the human population, including sensitive subpopulations, is likely to be without an appreciable risk of deleterious effects during a lifetime. In sampling station 1A NHQ value of mercury is slightly greater than 1.0 (NHQ=1.046), which indicates that the risk value is at the borderline with probable risk for adverse human health effects associated with long-term mudfish consumption. The NHQ values for lead in all sampling stations were way above 1.0 (ranging from 106 in sampling station 2B to 66,958 in station 1A), indicating high risk for adverse human health effects associated with long-term mudfish consumption.

## CONCLUSION AND RECOMMENDATIONS

Results of the study showed that arsenic, cadmium, and chromium do not pose significant non-carcinogenic health effects associated with the consumption of mudfish from Laguna de Bay. However,

concentrations of mercury and lead showed elevated levels that are likely to cause adverse health effects on fish long-term consumers. This study therefore concludes that from the point of view of human health protection and disease prevention, long-term human consumption of mudfish from Laguna de Bay is not safe due to elevated levels of mercury and lead that were found to be above the safe NHQ values.

In light of the above findings, the following recommendations are presented to help policy makers and stakeholders in decision-making as well as in crafting lake management policies and mitigating measures:

1. Urgent measures should be done by concerned authorities to protect health of communities consuming mudfish from the lake especially the children. The immediate goal should be to minimize exposure by minimizing the amount of fish intake and the frequency of consumption.
2. Regular monitoring of heavy metal in fishes should be done at least twice a year (wet and dry seasons) by concerned government agencies at all levels.
3. Regular health advisories regarding quantitative health risks associated with fish consumption should be issued by the Laguna Lake Development Authority or the Regional Office of the Department of Health.
4. Local Government Units, especially the lakeshore communities, should be involved in the heavy metal monitoring in fish and in developing and disseminating advisories and other health-related information to the communities and other stakeholders.
5. Inventory and assessment of potential sources of heavy metal in the lake (e.g., industrial sources) most especially for lead and mercury, should be undertaken.

**Table 5. Summary of NHQ values for dry season**

<b>Summary of NHQ Values for Dry Season</b>					
<b>Sampling Station</b>	<b>Cd</b>	<b>Cr</b>	<b>Pb</b>	<b>Hg</b>	<b>As</b>
<b>1A</b>	0.0643	0.0146	<b>16625</b>	0.4870	0.1864
<b>1B</b>	0.0923	0.0266	<b>9909</b>	<b>2.3880</b>	0.0161
<b>2A</b>	0.0598	0.0183	<b>8796</b>	<b>1.3577</b>	0.0784
<b>2B</b>	0.1053	0.0357	<b>13013</b>	0.4682	0.1149
<b>3A</b>	0.0567	0.1291	<b>16932</b>	<b>1.4429</b>	0.0424
<b>3B</b>	0.0736	0.0097	<b>11304</b>	<b>2.4124</b>	0.0000
<b>4</b>	0.0573	0.0112	<b>42506</b>	0.0428	0.0574
<b>5</b>	0.0937	0.0068	<b>17480</b>	0.2223	0.1104



Table 6. Summary of NHQ values for wet season

Summary of NHQ Values for Wet Season					
Sampling Station	Cd	Cr	Pb	Hg	As
1A	0.3435	0.0187	<b>66958</b>	<b>1.0460</b>	0.0192
1B	0.4565	0.0888	<b>62222</b>	0.4793	0.0237
2A	0.0154	0.0010	<b>7218</b>	0.3301	0.0005
2B	0.0037	0.0014	<b>106</b>	0.1626	0.0036
3A	0.0059	0.0016	<b>167</b>	0.7286	0.0005
3B	0.0176	0.0004	<b>1533</b>	0.4889	0.0005
4	0.0042	0.0009	<b>814</b>	0.4856	0.0129
5	0.0037	0.0009	<b>363</b>	0.1693	0.0139

6. More stringent regulation of effluents from industries around the lake should be enforced.
7. There should be regular monitoring of heavy metal in major rivers and tributaries draining into the lake.

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