LEVELS OF LEAD, CADMIUM, AND MERCURY IN CAGAYAN RIVER ALONG BUNTUN, CAGGAY, AND CATAGGAMAN NUEVO

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ABSTRACT

In this research study, levels of Lead, Cadmium, and Mercury in Cagayan River along Buntun, Caggay, and Cataggaman Nuevo were measured in MTEC Water Treatment Technologies using Inductive Coupled Plasma. The researchers compared the results to the standard level set by the Department of Environment and Natural Resources. The levels of Lead and Mercury are ≤0.010, and the levels of Cadmium ranged from 0.011-0.015. The statistical analysis showed that the levels of Lead and Mercury in the Cagayan River is below the standard value (p-values are greater than α= 0.05). While, the levels of Cadmium is above the standard level (p-value= 0.027< α=0.05). Continuous monitoring of water bodies in the area of collection is needed to minimize its health associated risks. And also, the levels of Lead, Cadmium, and Mercury do not affect the temperature and pH of the water in the river.

Key words: *Heavy metals, cadmium, lead, mercury, inductive-coupled plasma, Cagayan river, standard level*

INTRODUCTION

Water is one of the most important substances on earth. Animals, plants and all human beings must have water to survive particularly the used for drinking, food preparation, cleaning, and, not least, hygiene (Department of Health [DOH], 2017). 97 percent of water is ocean and the remaining 3% is fresh water. Of this, 69% resides in glaciers, 30% in underground, and less than 1% is located in rivers. 165 major rivers form the percentage of the river.

Philippines has 19 considered major river basins and Cagayan River is the longest among these. It is also known as the Rio Grande de Cagayan. The river's headwaters are in the Caraballo Mountains of the Central Luzon at an elevation of approximately 1,524 meters. The river flows north for some 505 kilometers to its mouth at the Babuyan Channel near the town of Aparri, Cagayan. It is categorized under type III climate zone which regarded as a no pronounced maximum rain period and a short dry period. It is comparatively dry from November to May and wet for the rest of the year (Department of Science and Technology-Philippine Atmospheric Geophysical and Astronomical Services Administration, 2013). Cagayan River is used for transport, recreation and tourism but it is mainly used for irrigation. It also supports the lives of numerous endemic and endangered species, like the Luzon bleeding-heart pigeon (*Gallicolumba luzonica*), Philippine eagle

(*Pithecophaga jefferyi*) and a rare riverine fish, locally called Ludong (*Cestreaus plicatilis*). However, these habitats are now threatened with pollution (Water Environment Partnership in Asia (WEPA), 2017).

Water pollution is a major problem in the Philippines. Fifty rivers in the country are considered dead and are unable to support life because of water pollution. Population, urbanization, agriculture and industrialization have all reduced quality of water in the Philippines.Pollution can cause disease outbreak and associated health risks (Tchounwou, 2014).

Pollutants are the major cause of water quality degradation that affects the ecosystem all over the world. Heavy metals that were caused by industrialization and agriculture is one of the pollutants in the river (Ojekunle, 2016). Heavy metal is defined as environmentally stable element of high specific gravity and atomic weight (Tchounwou, 2014). Manila Bay, a famous tourist spot, is contaminated with heavy metals such as Lead, Cadmium, Zinc, and Copper, which really contributed to the pollutants of the bay. These heavy metals were the effects of the modernization besides the bay (Prudente, 2014).

Heavy metals can contaminate the environment through industrial effluents; organic wastes refuse burning, transport and power generation. They can be carried to places by the wind, whether they are in gaseous form or as particulates. Another means of dispersal is the drainage of water from catchment areas which have been contaminated by waste from mining and smelting units. Heavy metal concentration in water occurs as either a natural phenomenon or through anthropogenic activities (Addleman, 2009).

This study assessed the levels of heavy metals specifically Lead, Cadmium, and Mercury in Cagayan River along Buntun, Caggay, and Cataggaman Nuevo.

Research Questions

This study aimed to identify the levels of lead, cadmium, and mercury in the Cagayan River within Tuguegarao City and its preventive measures.

Specifically, the researchers answered the following questions:

1. What is the physical profile of Cagayan River along Buntun, Caggay, and Cataggaman Nuevo in terms of:
   a. Temperature
   b. pH
2. What are the levels of Lead, Cadmium, and Mercury in Cagayan River along Buntun, Caggay, and Cataggaman Nuevo during:
   a. Dry season

b. Wet season

3. Is there a significant difference in the levels of Lead, Cadmium, and Mercury during dry and wet seasons?
4. Is there a significant difference in the levels of Lead, Cadmium, and Mercury and the physical profile of Cagayan River among the different locations of sample collection i.e. Buntun, Caggay, and Cataggaman Nuevo?

Hypotheses

- There is no significant difference on the levels of Lead, Cadmium, and Mercury during dry and wet seasons.

- There is no significant difference in the levels of Lead, Cadmium, and Mercury and the physical profile of Cagayan River among the different locations of sample collection.

Significance of the Study

This study assessed the levels of heavy metals specifically Lead, Cadmium, and Mercury in Cagayan River along Buntun, Caggay, and Cataggaman Nuevo. This information can give awareness to Cagayanos about the condition of the Cagayan River which serves as a source of living.

This research is also beneficial to the government, by providing them information that can help them to choose the right programs and courses of action that will lessen the contamination of the Cagayan River, particularly in areas of Buntun, Caggay, and Cataggaman Nuevo.

Literature Review

Cagayan River Profile

Cagayan River also known as Rio Grande de Cagayan is the second longest river in the Philippines. A drainage area of 27, 300 square kilometer in the province of Apayao, Aurora, Cagayan, Ifugao, Isabela, Kalinga, Mountain Province, Nueva Vizcaya, and Quirino and 53, 943 million cubic meters is the estimated annual discharge with 47, 875 million cubic meters that of reserve ground water. Its tributaries include the Elogan River which drains a major portion of the eastern watershed, the southern portion drains at the Magat River that originates in the Nueva Vizcaya municipality of Aritao and Chico River that is encompassing the regions of Cordillera and Cagayan Valley which drains most of the north-western area. Babuyan channel, Cagayan, and empties at the Aparri Estuary is the location of the mouth of the Cagayan River. Its temperature has a tropically different and the

northern portion has a high precipitation due to the exposure of the northeastern air movement (Aromin & Cimatu, 2017).

Before it drains into the Babuyan Channel, the water supply of the region was provided by Cagayan River for agriculture, industry, domestic and municipal needs. And during the first search for Cleanest Bodies of Water (River Category) of the Clean and Green Program the Cagayan River was adjudged as “The Cleanest River in the Country” (Villena & Rimando, 2005).

Biodiversity in Cagayan River

Cagayan River is the longest river in the country and one of the biggest resources of the region II primarily for the irrigation system and also other purposes such as tourism, transport, and fisheries industry. Given its wide area, it is one of the habitats and the major sources of freshwater fish including endemic species such as Ludong, and indigenous species of shellfish and fish like mollusks (unnok; dalilea spp. ,balinggasa; anodonta woodiana, tulya; corbicula manilensis, and kabibe; batissa violacea) and crustacean (aramang; palaemon andacetes spp.), (R.B. Villena and M.A. Rimando, 2005). In addition, the historical information stated that some freshwater crocodiles and water and migratory birds were also resided (Weerd and Ploeg, 2004). Boxed turtles were recorded as the only reptile species and Cantor’s Giant Soft Shelled Turtle (Pelochelys cantorii) was recorded as the only endangered species documented. At present, Cagayan River is the major inland fishing ground for both full-time and part-time fisher folks on nearby areas.

However, declining quality of the river has been noted due to deterioration and contamination caused by sewage disposal from residential and industrial areas and mine tailings, heavy use of chemicals in farming and high rate of erosion. Therefore, improvement of the deteriorating water quality in the Cagayan Riverine Zone must be the main concern.

Elements Affecting Biodiversity in Cagayan River

Humans are the number one cause of the alteration of the biodiversity of the River. The continuous use of inorganic fertilizers, the modernization such as the building of bridges, and the lack of discipline of humans lead to the destruction of the biodiversity in the river. Humans made the river as their trash bins. The trash that has been accumulated in the river alters the biodiversity. Inorganic fertilizers contain toxins that harm the living organism in the River. It contains heavy metals that can accumulate in the water of the river. The river serves as the catch basin and the source of the water in the different irrigations. Repainting of bridges may also affect the biodiversity in the river. Paints contain lead which is dangerous to the aquatic organisms. Lead poisoning of fishes is very common nowadays. Because of these heavy metals is one of the elements that affect the biodiversity of the river. Heavy metals are identified as a group of elements specifically metalloids and

transition metals that have a high atomic weight and a density at least 5 times greater than water. Their multiple domestic, industrial, agricultural, medical and technological applications lead to the arising concerns to human health and the environment (Weerd & Ploeg, 2004).

Environmental Effects of Heavy Metals

Cadmium can contaminate the plants and rivers which will become a source of cadmium contamination to humans. The river serves as the catch basin of water from irrigations and farms which makes it prone to contamination. Cadmium is known to be a component of fertilizers used by farmers to treat their plants. The cadmium contaminates the water of the farm which will then contaminate the river. The amount of the cadmium has a significant positive correlation with the cadmium loading of the cropping system. Places with few plantations have the lowest recorded level of cadmium contamination (Bandara, Jayasooriya, Rajapaksha, and Wijewardena, 2010).

Ionic mechanism and oxidative stress of lead metal can cause toxicity to living cells. Lead can destroy the ecosystem under the river. One of the major causes of the mass death of fishes is lead. Over accumulation of lead in the fishes can destroy their intestines which will cause their death (Anbalagan, Beeregowda, Jaishankar, Mathew, & Tseten, 2014).

Rivers and other water bodies are common sources of mercury because it serves as the basin of wastes of the residents near the rivers. It indirectly affects man since the river water is used to irrigate a nearby farmland (Awofolu, Fatoki, Mbolekwa, & Mtshemla, 2005). Sediments such as fish and other aquatic organisms are the main sources of mercury as they are considered the major metabolic pathway of human consumption of mercury (Hosseini, Nabavi & Parsa 2013).

National Standard on the Levels of Cadmium, Lead, and Mercury and on the Physical Profile of the River

According to Department of Environment and Natural Resources (DENR) Administrative Order No. 34 Series of 1990 under Section 68 under the Water Usage and Classification – The quality of Philippine water shall be maintained in a safe and satisfactory condition according to their best usages.

Classifications have been listed and enumerated according to 8 classes namely: Class AA *(Public Water Supply Class I),* Class A *(Public Water Supply Class II)*, Class B *(Recreational Water Class I),* Class C *(Fishery Water, Recreational Water Class II, and Industrial Water Supply Class I)*, and Class D *(Industrial Water Supply Class II).* Class SA, Class SB *(Recreational Water Class I, and Fishery Water Class),* and Class SC *(Recreational Water Class II, Fishery*

Water Class II). Classifications were arranged in accordance to the protection necessary to each class; Class AA and SA according to the memorandum requires a strict compliance and monitoring to water surface and marine lives, respectively; and Class D and SD have a lax compliance and monitoring to water surface and marine lives, respectively. These criteria have been used in assurance to conditions suitable and necessary to water bodies.

The Cagayan River is classified under Class C of Section 68 of the memorandum. A pH ranging from 6.5-8.5 has been listed according to Table 1 of the memorandum: *Water Quality Criteria for Conventional and Other Pollutants Contributing to Aesthetics and Oxygen Demand for Fresh Waters.* A 3-degree rise in temperature of the water bodies are monitored as it can cause substantial and direct damage to aquatic life or vegetation therein, states by Section 69 Water Quality Criteria of the memorandum.

Heavy metals including Lead, Cadmium, and Mercury must be maintained for national standard levels set by the DENR **(**Department of Environment and Natural Resources) according to their parameter and classes of classifications. Lead under Class C of the national standard must be within 0.05. Cadmium is expected at 0.01 of the standard levels, and Mercury to be considered standard shall be maintained at 0.002.

Research Simulacrum

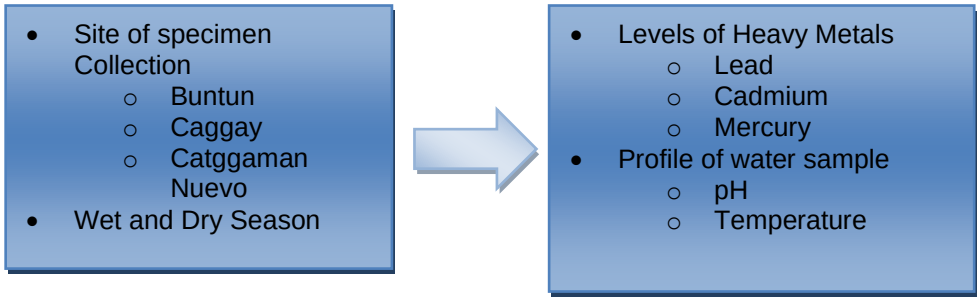


Figure1. *Research Simulacrum*

The figure shows the whole concept of the study in which the researchers determined the physical profile of Cagayan River in terms of temperature and pH. The researchers also assessed the levels of Lead, Cadmium, and Mercury in Cagayan River along Buntun, Caggay and Cataggaman Nuevo during wet and dry season. The samples were sent and analyzed by a water analyst at MTEC Water Treatment Technologies Incorporated in Cabuyao, Laguna.

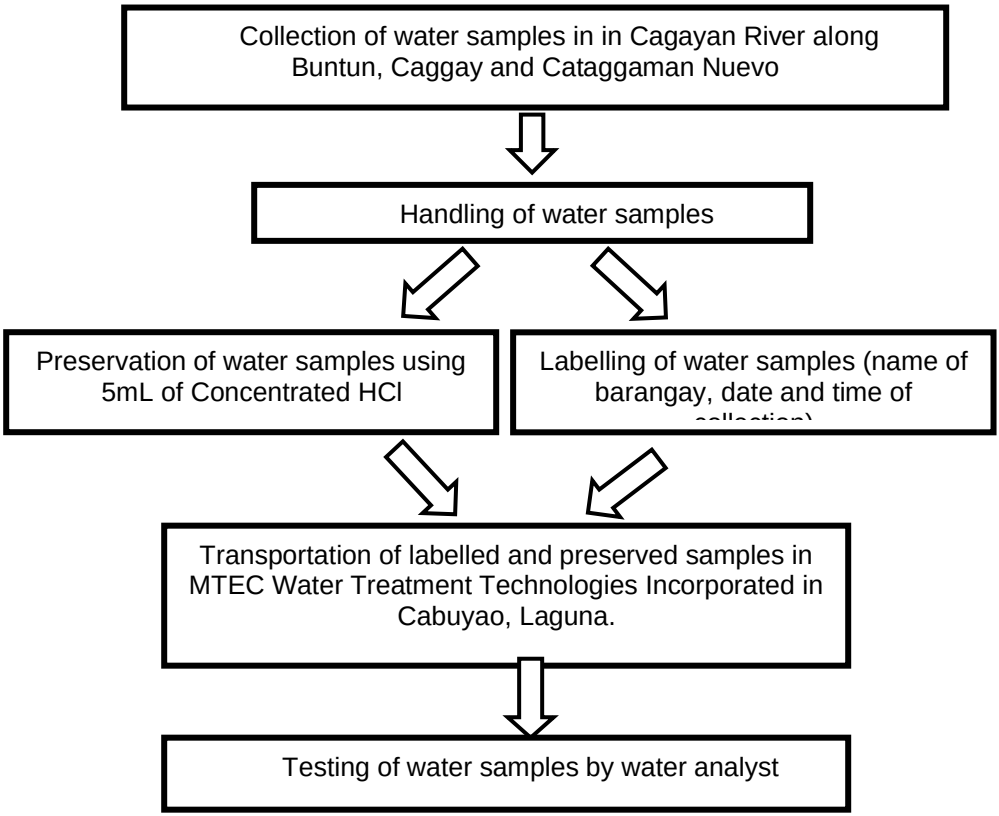
METHODS

Research Design

This research study utilized descriptive correlational approach in which the water from Cagayan River within Tuguegarao City, specifically Buntun, Caggay, and Cataggaman Nuevo was tested for the concentration of Lead, Cadmium, and Mercury. Further, no controls were utilized.

Locale of the Study

The study was conducted in Buntun, Caggay and Cataggaman Nuevo, Tuguegarao City. Inductive Coupled Plasma was used to identify the levels of Cadmium, Lead, and Mercury which was conducted at MTEC Water Treatment Technologies Incorporated in Cabuyao, Laguna. The collection and the measuring of temperature and pH were performed by water analyst from Metropolitan Water District.



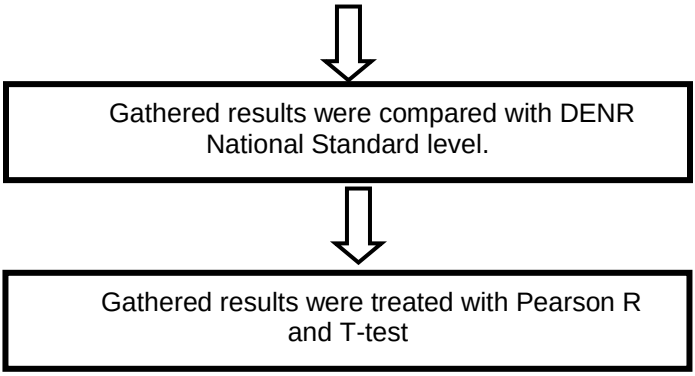


Figure2. *Methodological Workflow*

Data Gathering Procedure

1. Collection and Handling of water samples

The procedure followed by the researchers was based on Standard Operating Procedure of MTEC Water Treatment Technologies and Department of Environment and Natural Resources (2017).

1.1. The polyethylene containers or bottles of distilled water were used for collecting and storing of water samples.
1.2. The containers were washed with 16% hydrochloric acid and rinsed several times with distilled water.
1.3. The researchers collected a total of eighteen (18) samples with three trials and three replicates for both wet and dry season, samples must be 1 liter.
1.4. The collection of water samples from the river must at the center of the flow, 0.5 cm of the surface between 9:00 am to 4:00 pm.
1.5. 5 ml of concentrated hydrochloric acid was added to the water sample to preserve the water during transportation and to ensure the availability of the chemical components.
1.6. The containers must be sealed or capped to avoid leakage and contamination.
1.7. The containers were labelled with name of the barangay and date & time of collection.
1.8. The water samples were placed in a box of ice and the box were properly sealed.
1.9. The water samples were transported within 24 hours from Tuguegarao City to MTEC Water Treatment Technologies, Inc. in Cabuyao, Laguna.

2. Testing of the water samples

The samples were sent to MTEC Water Treatment Technologies, Inc. in Cabuyao, Laguna for the measurement of the concentration of Cadmium, Lead, and Mercury.

Data Analysis

The researchers utilized pearson-R and T-test to compare the gathered concentrations of Lead, Cadmium, and Mercury during the dry and wet seasons to the standard levels given by the Environmental Management Bureau (EMB) under the Department of Environment and National Resources (DENR).

Waste Disposal Management

The MTEC, Water Technologies, in Cabuyao, Laguna was responsible in the disposal of the tested water samples and its containers and the chemical used for the testing.

Ethical Considerations

Prior to the experimentation, the researchers sought permission from the University through the Associate Dean, Academic Dean, University Ethics Board, Vice President for Academics and President of University for the conduct of study. The researchers were given an ethical clearance number 51528.

The researchers also sought for the permission of the Barangay Captains of Buntun, Caggay, and Cataggaman Nuevo for the conduct of the study in their barangay with the assurance of the safety of the residents.

The researchers sought assistance from a licensed water analyst in collecting and measuring the temperature and pH of the samples during the wet and dry season.

Lastly, the researchers sought the assistance of MTEC Water Treatment Technologies Incorporated for the testing of the levels of Cadmium, Lead, and Mercury.

RESULTS

Table 1.1. *Temperature of Cagayan River along Buntun, Caggay, and Cataggaman Nuevo*

	R1	R2	R3	Mean	Difference in Mean temperature (Wet and Dry Season)
Wet Season					
Buntun	25.83	24.30	25.4	25.18	
Caggay	28.60	26.10	26.80	27.17	
Cataggaman Nuevo	29.06	24.60	24.7	26.12	
Dry Season					
Buntun	29.9	31.77	32.8	31.49	6.31*
Caggay	26.1	31.3	35.4	30.93	3.76*
Cataggaman Nuevo	29.5	33.8	37.6	36.63	10.51*

**Increase of temperature is above the standard value of 3^oC*

Table 1 shows that the increase in temperature in Cataggaman Nuevo during the dry season is above 3^0C.Which is above the standard value for the rise of temperature in the river.

Table 1.2. *Temperature of Cagayan River along Buntun, Caggay, and Cataggaman Nuevo*

	R1	R2	R3	Mean	Qualitative Description
Wet Season					
Buntun	8.18	7.99	8.5	8.22	Within standard value
Caggay	9.93	8.39	9.4	9.24	Above standard value; acidic
Cataggaman Nuevo	8.14	8.5	8.8	8.48	Within standard value
Dry Season					
Buntun	7.89	8.28	8.5	8.22	Within standard value
Caggay	7.99	8.4	7.8	8.06	Within standard value
Cataggaman Nuevo	8.17	8.2	8.0	8.12	Within standard value

**Standard value of pH- 6.5- 8.5*

The pH value of the water measured is within the standard value given by the DENR except for the first and second sampling in Caggay which measured 9.93 and 9.4, respectively. There was also an increased in the pH of the third sampling in Cataggaman Nuevo.

Table 2.1. *Lead Levels in Water Sample during Dry Season*.

	R1	R2	R3	Mean	Qualitative Description
Wet Season					
Buntun	<0.010	<0.010	<0.010	<0.010	Within standard value
Caggay	<0.010	<0.010	<0.010	<0.010	Within standard value
Cataggaman Nuevo	<0.010	<0.010	<0.010	<0.010	Within standard value
Dry Season					
Buntun	<0.010	<0.010	<0.09	<0.010	Within standard value
Caggay	<0.010	<0.010	<0.010	<0.010	Within standard value
Cataggaman Nuevo	<0.010	<0.010	<0.010	<0.010	Within standard value

Standard value Lead- 0.005 mg/L

Table 2.1. shows that the levels of Lead in Cagayan River along Buntun, Caggay, and Cataggaman Nuevo during the wet and dry seasons are within the normal values given by the Department of Environment and Natural Resources.

Table 2.2. *Cadmium Levels in Water Sample during Dry Season*.

	R1	R2	R3	Mean	Qualitative Description
Wet Season					
Buntun	<0.001	0.012	0.013	0.009	Within standard value
Caggay	<0.001	0.011	0.011	0.008	Within standard value
Cataggaman Nuevo	<0.001	<0.001	<0.001	<0.001	Within standard value
Dry Season					
Buntun	0.013	0.015	0.014	0.014	Above standard value
Caggay	0.012	0.014	0.013	0.013	Above standard value
Cataggaman	<0.001	0.011	0.011	0.008	Within standard

Nuevo					value

Standard value Cadmipum-0.01 mg/L

Table 2.2 shows that the Cadmium levels in Cagayan River along Buntun and Caggay are slightly higher than the recommended values given by the DENR; however, this is only true during dry season.

Table 2.2. *Mercury Levels in Water Sample during Dry Season*.

	R1	**R2**	**R3**	**Mean**	**Qualitative Description**
Wet Season					
Buntun	<0.010	<0.010	<0.010	<0.010	Within standard value
Caggay	<0.010	<0.010	<0.010	<0.010	Within standard value; acidic
Cataggaman Nuevo	<0.010	<0.010	<0.010	<0.010	Within standard value
Dry Season					
Buntun	<0.010	<0.010	<0.010	<0.010	Within standard value
Caggay	<0.010	<0.010	<0.010	<0.010	Within standard value
Cataggaman Nuevo	<0.010	<0.010	<0.010	<0.010	Within standard value

Standard value Mercury- 0.002 mg/L

Table 2.1. shows that the levels of Lead in Cagayan River along Buntun, Caggay, and Cataggaman Nuevo during the wet and dry seasons are within the normal values given by the Department of Environment and Natural Resources.

Table 3. Test of *Significant Difference in the Levels of Lead, Cadmium, and Mercury in the Water Samples between Wet and Dry Seasons*

Heavy Metals	**t-value**	**p-value**	**Decision**
Lead	1.000	0.332	Accept H_{o1}
Cadmium	2.431	0.027	Reject H_{o1}
Mercury	2.000	0.063	Accept H_{o1}

at 0.05 Levels of Significance

Table 3 shows that the levels of Lead and Mercury are still comparable during the wet and dry season. Whereas, the levels of Cadmium is higher during the dry season.

Table 4.1. *Test of Significant Difference in the Physical Profile of the Cagayan River according to site of sample collection*

Category	F-value	p-value	Decision
Temperature during Dry Season	.454	.655	Accept H_o
Temperature during Wet Season	1.017	.417	Accept H_o
pH during Dry Season	.292	.757	Accept H_o
pH during Wet Season	3.194	.114	Accept H_o

Table 4.1 shows no significant difference in the temperature and pH of the river during the dry and wet seasons.

Table 4.2. *Test of Significant Difference in the Levels of Cadmium in the Cagayan River according to site of sample collection*

Category	F-value	p-value	Decision
Wet Season	2.011	.215	Accept H_o
Dry Season	2.748	.142	Accept H_o

The table above shows that the Cadmium levels of the Cagayan River are significantly the same in the different sites where samples were collected. It should be noted that the Lead and Mercury levels cannot be tested for significant difference due to the same values obtained during testing.

DISCUSSION

The primary purpose of this research study was to determine the concentration of Cadmium, Lead, and Mercury. Water samples were collected along the rivers of Buntun, Caggay, and Cataggaman Nuevo in Tuguegarao City, Cagayan and were submitted in MTEC Cabuyao, Laguna for testing in terms of heavy metals concentration during wet and dry season.

The temperature in Cagayan River along Buntun, Caggay, and Cataggaman Nuevo is within the standard level of heavy metals under the DENR Memorandum Number 35, series of 1990. During the dry season, the temperature in Cataggaman Nuevo increases above the standard 3^0C rise. Temperature usually increases in the dry season due to the slow moving flow of water which prolongs the exposure of the water to the sun (Rimando and Villena, 2017)

According to the Department of Environment and Natural Resources, the pH of the water in Class C rivers must be between 6.5-8.5 (DENR Memorandum Number 35 Series of 1990, 2017). The pH value of the water in Cagayan River along Buntun, Caggay, and Cataggaman Nuevo is within the standard value except for the first and second sampling in Caggay which measured 9.93 and 9.4. The alkalinity of water is due to the presence of dissolved salt, carbonates,

bicarbonates, minerals and hydroxide compounds (Oram, 2010). While acidity of water may be caused by the fuel emission of carbon dioxide from agricultural and industrial wastes (He, Islam, Mahmood & Yang, 2007).

The amount of lead present in the water measured within the standard value of 0.05mg/L except for the third sampling in Buntun which measured <0.09 mg/L. In surface and ground waters natural levels of lead are very low (<5 ppb). The little amount of Lead is caused mostly by modernization, such as construction of bridges. Lead coming from fertilizers can also affect the water in the river since river serves as the catch basin (Basu & Dejbler 2013).

The level of mercury in Cagayan River along Buntun, Caggay, and Cataggaman Nuevo is within the standard value of 0.002 mg/L. An increase in the levels of Mercury in the river may be due to the industrial waste but because of the ordinances that prohibit the use of mercury, the levels of mercury in the river may not exceed the standard value.

The measured amount of Cadmium in the three barangays exceeds the standard value of 0.01mg/L. Levels of Cadmium concentrations in river water are typically <0.5 mg/L but because of its use in fertilizers the levels of Cadmium in the river begin to exceed the set standard value (Åkesson et al., 2009). During the collection of water samples, it was observed that the Cagayan River serves as the catch basin for the waste coming from the residential area through the different canals; besides, the river surrounded with different farm plants. These two conditions contributed to the levels of cadmium in this area. The increased level of Cadmium in the Cagayan River is alarming because of its effect to the health.

Cadmium alters the cells. It induces apoptosis resistance to the altered cell causing lung cancer. It also increases punctual cells which is an indication of autophagy or apoptosis deficiency to the altered cells. Increased cadmium exposure to normal cells will result in tumor (Budhrajaet al., 2014).

CONCLUSION

The researchers concluded that the levels of Lead and Mercury along Buntun, Caggay, and Cataggaman Nuevo are within the standard level.

The levels of Cadmium along Buntun, Caggay, and Cataggaman Nuevo are higher than the standard level. The levels of heavy metals such as Lead, Cadmium, and Mercury do not affect the temperature and pH of the water in the river.

RECOMMENDATION

Based on the findings of the study, there is a need to:

- Measure the levels of Lead, Cadmium, and Mercury in Cagayan River along other locations.
- Trace the possible sources of heavy metals.
- Determine the levels of Cadmium in the aquatic lives and the plants within or near the river.
- Identify other possible heavy metals present in the river.
- Initiate preventive measures that will lessen the levels of heavy metals in the Cagayan River by the Department of Environement an Natural resources.

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