Color And Image Analysis Approach in Determination of Soluble Copper in Water Using Tannic Reaction Analysis

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The 1993 and 1996 Marcopper Mining Disaster caused excessive amounts of copper to water sources in Marinduque. Determining this soluble metal in water has been a challenge to Marinduqueños as there is no water testing laboratory in the province. This research aims to develop a system that determines soluble Copper (Cu) in water through color reaction analysis of tannins. Specifically, it aims to develop a system for determining different levels of soluble copper in water and evaluate the developed system through comparative analysis of the results from laboratory. An image processing box was prepared to capture the color reaction of Jatropha Curcas sap to water solutions containing 0.5 to 5 ppm soluble copper. The color reaction was recorded using a Logitech C922 Camera through image acquisition and labeler of MatLab Software. The recorded color reactions were analyzed for their red, green, and blue (RGB) channels using a color detection system of the same software. The RGB color values were then related to the copper concentrations. To assess the reliability of the data generated, it was compared to water samples of unknown soluble contents from various locations in the province. Based on these procedures, it was found that copper produces green and blue colors, especially for the water solutions with 2.0 to 3.5 ppm mg/L of copper. It was concluded that the system can determine the soluble copper in water samples (from 0.5 mg/L to 5mg/L), but calibration must be done using genetic algorithm optimization features and other ways such as machine vision. Relevant to analytic chemistry, it is recommended that titrimetric conditioning and the use of the Sigmoid function be performed to improve color resolution.

Keywords: Color Reaction Analysis; Copper; Image Processing; Tannic Reaction;

I. INTRODUCTION

Marinduque Island is a small island province in the Philippines' Luzon southernmost region. In 2017, the Mines and Geosciences Bureau (MGB) reported that the country's total projected production for metallic minerals was PhP 109.45 billion. Marinduque was thought to have one of the Philippines' largest copper reserves. Mining techniques for copper extraction began in Marinduque between 1969 and 1996. [1]

In 1991, an earth dam was built in the Mogpog River's hilly source area. The Mogpog River's silt from a waste dump for the new San Antonio Mine Pit was intended to be captured by the dam. In 1993, the dam burst, devastating the communities below. Mogpog's town suffered severe devastation, leading to large losses in terms of structural assets, agricultural, and public health. The toxic waste from the dam had a significant negative impact on the 23-kilometer Mogpog River. [1] Three years later, in 1996, one of the worst mining accidents in history occurred when the Tapian Pit burst, leaving tailings in the nearby location polluting the 27-kilometer Boac River on the island of Marinduque. 20,000 people had to be evacuated, and five villages had to be abandoned. [2] The pollution's impact on the river and coastal ecosystems were so severe that the United Nations classified the occurrence as an environmental disaster. [3]

Another affected area is the Boac River which is a prominent river system in the Philippines' province of Marinduque. Before 1996, the river provided a significant source of subsistence food and family income for the residents. The Marcopper catastrophe, on the other hand, occurred on March 24, 1996, releasing millions of tons of poisonous mining wastes into the Boac River. [4]

Heavy metals, which are metallic elements with a relatively high density and are dangerous even in low quantities, may be found in mine waste. Although heavy metals such as copper, cobalt, iron, magnesium, nickel, zinc, and others are needed for many biochemical and physiological processes, they can be potentially detrimental in excessive amounts. Heavy metals discharged with mine tailings can be absorbed by plants and accumulate along the food chain, putting animal and human health at risk. [4]

In the various assessment conducted through the years, it was found that the mining-affected areas are heavily concentrated with Copper and other heavy metals making the affected bodies' water unsafe for drinking and domestic uses. [1] [4] [5] [6] [7]The findings from these studies have been evident in the plants growing on the riverbanks. [8] [7] Even though it has been years since the tragic mining accident happened, huge toxic waste is still prevalent in the mining-affected areas. [5]

Given these findings, water potability along the mining-affected areas has been one of the challenges. [1] [8] [5] In addition, the effects on health due to the increased presence of heavy metals were manifested in the municipalities of Boac, Sta. Cruz and Mogpog. It has been recommended by various agencies that due to the high level of Copper concentration, rehabilitation and testing of sources of water should be addressed. Based on the geographic structure of the province, testing water has been one of its main challenges. The nearest water testing facilities for heavy metals in Marinduque can be found in other provinces or the Metropolitan Manila. [9]

This research aims to develop a system that determines soluble Copper (Cu) in water through color reaction analysis. Specifically, it aims to develop a system for determining different levels of soluble copper in water. Lastly, the research aims to evaluate the developed system through a comparative analysis of the results from a registered laboratory.

II. Determination of Heavy Metals in Water

Throughout the years, there have been various methods used in determining trace metals as to whether it complies with the standard limits for different purposes. Known methodologies are atomic absorption spectrophotometry (AAS) and inductively-coupled plasma-mass spectrometry (IC-PMS). These methods usually involve the mixing of reagents, preparation of standard solutions, and pre-treatment and are dependent on the sample constitution. [10, 11] In the Philippine setting these approaches are the standard determination techniques in laboratories for testing water quality in terms of heavy metal contents. Further, these methods were also found to be useful in determining heavy metals in other matters such as macroinvertebrates and aquatic resources.

Another method that can be used to determine the trace metals in water is observing the gradual reaction of stilbene phosphorescence. This method is applicable in large-scale water testing such as those from reservoirs and water reserves. [12] The aforementioned method relies on the absorption properties of the Intercalated Layered Rare-Earth Hydroxide Tablets. The phosphorescence reaction is observed under the microscope.

It is also possible to identify the heavy metallic elements by observing how hazardous they react with DNA-based fluorescence chemo-sensors. In this type of technology, four fluorescent 2′-deoxyribosides of metal-binding ligands that were discovered and manufactured are utilized to assess the level of contamination in water. Such a methodology relies on the ability of bead-based ODF chemo-sensors to analyze heavy metal contamination in water samples using a quick and low-cost optical technique. [13] Another parallel method that is based on adsorption properties was utilized using functionalized Nylon 66 which was utilized as Photocatalyst. This process involves photodegradation of Eriochrome Black T-Dye in an Aqueous Medium. This technique uses light and temperature as the determining factor of copper in water. [14]

In 2008, it was found that tannins found in the sap of Physic nut (*Jatropha Curcas*) react with heavy metals found in water. The reaction is manifested in the color reaction of tannins in soluble metals such as Copper, Iron, Manganese, Lead, Nickel, and Zinc. [15] [16] A paper strip using this varying amount of the mentioned sap can be used in determining the heavy metals in water. This color reaction was also established as a basis as to whether the water is potable and safe for domestic purposes on the island of Marinduque. [17]

Another approach that utilizes the color reaction analysis was developed using a tannic acid dispenser. Tannic acid is being dispensed using a controller in proportion to the sample water. [18] The color reaction of tannins present in *Jatropha Curcas* to soluble copper is illustrated in Figure 1.

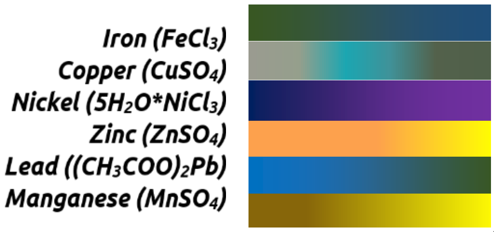


***Figure 1**. Color Reaction of Tannins to Soluble Copper [15]*

Based on these data, it can be deduced that most of the heavy metal detection in water is based on color processing or light reaction properties. In this study, the color produced by the reaction of tannins found in Physic Nut in copper will be used to develop a system that determines copper concentrations in water.

Color Reaction Analysis Using Image Processing

With the premise of this study relying on color reaction produced from the reaction of tannins and soluble metals, analysis of the produced colors through machine vision will be utilized. Compared to visual and instrumental analysis, machine vision systems are better able to measure irregular colors and shapes and check particular portions of the image. [19] Usually, the industry has made considerable use of colorimeters and spectrophotometers to measure color. Machine vision can get around some of the possible drawbacks of utilizing colorimeters and spectrophotometers. When the sample's colors are not homogeneous, one issue could develop. In these circumstances, color values obtained through repeated surface measurements or from several samples within the same batch are averaged. In this type of technology, colors are quantified in terms of both quantity and definition. Based on how the light source, sample plane, and detector are arranged, there are two types of instruments: directed and diffuse. They may have a distinct perspective on the sample and produce various color outcomes. [20]

To address these concerns in the established techniques – machine vision or image processing is usually utilized. A camera that is connected to a computer, controlled lighting (often in the form of a light box), and software that manages camera settings, picture capture, and processing are all required for machine vision (MV). The charge-coupled device (CCD) used as the camera's sensor transforms photons into electrical impulses. Once a picture has been taken, it can either be analyzed right away or saved for further comparisons and analysis. As a result, MV offers record-keeping and documentation. [19]

Color measurement of irregular shapes and hues is possible using MV. Its color(s) can be measured as long as the sample is within the camera's field of vision. It is possible to analyze multiple objects at once. No matter the sample size or shape, since every pixel in the sample image is examined, it is also possible to calculate the "average" color of each object, simulating the results of a colorimeter. [21] Color machine vision systems may detect tiny color differences. The amount of color management required in the system (and thus the cost) is determined by the application. The key to success is analyzing what distinguishes a good color from a bad color and establishing a system that emphasizes this difference. Color vision systems, from simple sorting to complicated print analysis, can enable production verification if the right controls are included in the system. [22]

The advantages provided by machine vision in color applications make it effective in application where color recognition, classification, and pattern formation is necessary. It is widely used in automated systems such as assembly lines reducing human involvement. [23] There is no definite physical link between color recognition characteristics and color categories. As a result, typical mechanism modeling methods do not meet the criteria of color recognition. This gap is being addressed by artificial intelligence. Artificial intelligence methods have progressed significantly in recent years. And one of the most common algorithms is the Artificial Neural Network (ANN). ANN is capable of tackling nonlinear issues. Implicitly, neural networks can provide the variable mapping between inputs and outputs. [24] [25] The Artificial Neural Network (ANN) method is frequently utilized in color recognition.

Depending on the application and technique to be used, artificial neural networks come in handy when it comes to determining heavy metals. A deep Neural network with a visible neural network and infrared spectroscopy of Soil was found to be a robust developed model for estimating metals in soil. [26] As benchmark models, conventional back-propagation neural network (BPNN) and nonlinear autoregressive network with exogenous inputs (NARX) approaches were used in long-term toxic heavy metal prediction. An 18-year data set was used to train, validate, and test these models, which were then illustrated using simulation results from a 2-year data set. [27] Artificial Neural Networks (ANN) and Image Processing were also used in the study of the effects of heavy metals absorption of Pennyroyal Plant leaf discoloration. [28]

Based on these techniques employed in determining heavy metals in various mediums, this research will utilize image processing in determining soluble copper in water.

III. Materials and Methods

To carry out the research objectives, phases of the research were developed as illustrated in the figure 2.

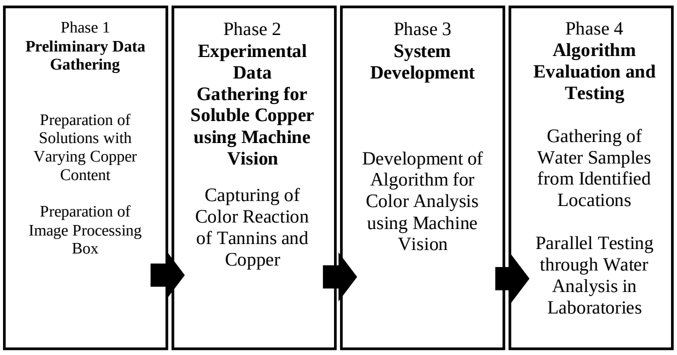


***Figure 2**. Phases of Research Development*

As illustrated in Figure 2, there were four (4) phases employed in this research. The researcher utilized a product innovation model design whereas to develop the expected output – the gathering of raw data must be done. The gathered data will then be used to design the new output. This research design is based on existing practices involving product development. [29] The phases of the research are substantially discussed as follows:

Phase 1: Preliminary Data Gathering Method

The first phase of data gathering is concerned with the initial steps to gather data for the systems development. Preparation of water solutions with soluble copper was done. An increasing number of copper concentrations were computed using the following equations:

$$M = \frac{Mass\ of\ solute}{Molar\ mass\ of\ Solute\ x\ Volume\ Solution}\ ;assuming\ V = 1\ liter \quad \textbf{(1)}$$

$$Mass\ Solute = Molarity\ x\ MM\ Molar\ Mass\ of\ Solute\ x\ Volume\ Solution \quad \textbf{(2)}$$

$$Molar\ Mass\ of\ Copper = 63.55\ g\ Cu^{2+}$$

$$Molar\ Mass\ of\ Copper\ Sulfate\ ; 159.62\frac{g}{mol}$$

The following formulas were based on existing molarity computation using parts per million computations. Soluble copper (Cu^{2+}) present in Copper Sulfate was used to come up with the solutions. Table 1 presents the amount of copper used per solution.

Table 1. *Amount of Copper per Solutions*

Solution No.	Copper Present (in ppm)	Conversion to g/L	Amount in Mole	Copper Sulfate Dissolution
1	0.50	0.00050	7.86782E-06	0.00125586
2	1.00	0.00100	1.57356E-05	0.00251172
3	1.50	0.00150	2.36035E-05	0.00376758
4	2.00	0.00200	3.14713E-05	0.00502345
5	2.50	0.00250	3.93391E-05	0.00627931
6	3.00	0.00300	4.72069E-05	0.00753517
7	3.50	0.00350	5.50747E-05	0.00879103
8	4.00	0.00400	6.29426E-05	0.01004689
9	4.50	0.00450	7.08104E-05	0.01130275
10	5.00	0.00500	7.86782E-05	0.01255862

As presented in Table 1, 10 solutions were prepared and which will serve as the baseline data of color reaction analysis in Machine Vision. For this study, it focused on determining soluble copper in water, thus being limited to at least 0.5 ppm present in water.

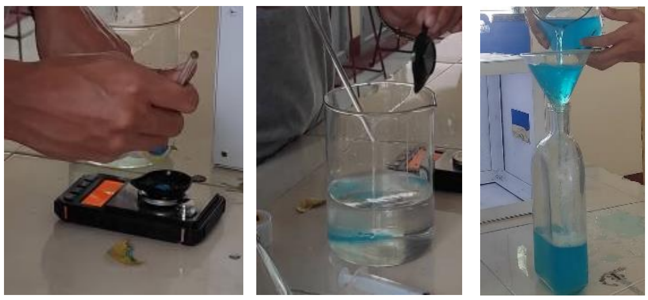
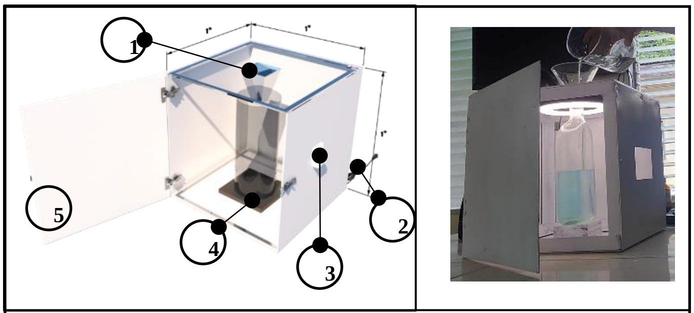


***Figure 3.** Preparation of the Water Solutions*

Reflected in Figure 3 are the preparations of soluble copper. The amount of copper sulfate to be dissolved in purified one (1) liter of water (in the last column of Table 1) was weighed using a digital analytical balance. Since the analytical balance was limited to x10^{-1} calibration, dilution was done as seen in Figure 3.

This was done to lower done larger amount of soluble copper present in water. Purified water was used to eliminate any unwanted minerals and contents. To capture the color reaction made by the reaction of Copper and tannins an image capturing box was prepared.



Parts:
1. Funnel Slot – for insertion of various panels and can be covered if needed.
2. LED Light Cable
3. Camera Slot
4. Removable Platform for Easy adjustment of Subject to be observed
5. Cover

Figure 5. Image Capturing Box for Color Reaction Analysis

As shown in Figure 4, the image capturing box has a cubic meter capacity. It was fabricated using a galvanized iron sheet. It is colored white to provide a better range when the color reaction analysis was made. It has installed LED lights for a better resolution and a single lighting source. A Webcam Slot was also provided for machine vision where a Logitech C922 Web Camera was attached during the data gathering process.

Phase 2: Experimental Data Gathering for Soluble Copper using Image Processing

The second phase of this research concerns the capturing of the color reaction of tannins to soluble metals in water. The prepared solutions in the preceding phase were placed in the image capturing box. *Jatropha Curcas* sap that contains tannins was diluted to 30 % Volume per Volume ($^{v}/_{v}$) Concentration and the recommended minimum amount that reacts to heavy metals in water. [15] [18]

Figure 5 shows the preparation and addition of Tannins to the water solution using a syringe. For the homogeneity of the system, 10 mL of diluted 30% ($^{v}/_{v}$) *Jatropha curcas* was measured using a syringe and was added to each solution. The researcher utilized the Logitech C922 Web Camera for recording the color reaction.

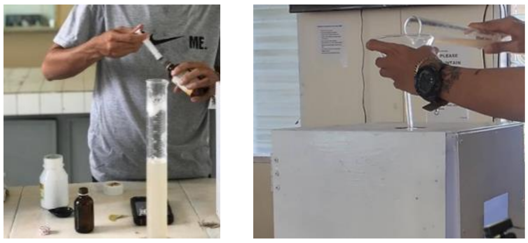


Figure 4. Preparation and Addition of Jatropha Curcas Solution

Phase 3. System Development

The color produced in the image processing box was captured and labeled using the image acquisition feature and Image Labeler of MatLab. After the color reaction was labeled according to the contained dissolved copper and analyzed for color channels (red, green, and blue).

To analyze the color, hue, and saturation of the colors – the color distribution feature of the MatLab application was used. This was done through a channel and coordinate analysis.

Based on this analysis, corresponding values of these colors have been captured and recorded. Further, color detection attributes were correlated to the soluble copper content.

Phase 4. Algorithm Evaluation and Testing

For the last phase of the research, system evaluation and testing were done. This is to compare whether the developed algorithm will yield the same result as the prescribed and standard determination procedures of testing soluble metals in water.

For this phase, gathering water samples from locations where there are heavy copper concentrations. Figure 6 shows the map of the Marinduque province where the samples were located. These areas are said to be heavily concentrated with soluble copper. [4] [6]

Figure 6. *Map of Marinduque where Water Samples were gathered*

As illustrated in Figure 6, there were three (3) locations where water samples were gathered. These locations are along Brgy. Ino-Capayang, Mogpog (Sample 1), Estuary of Brgy. Lupac, Boac (Sample 2), and the stream of Brgy. Canat, Boac, Marinduque (Sample 3). These locations were purposely selected as they are known to be connected to man-made lakes for mining spills and the Boac River where there are high concentrations of mine tailings containing Copper.

The water samples amounting to 3 (three) liters each were gathered one (1) foot above the surface. It was contained in a brown envelope to avoid any photochemical reaction. 1.5 liters of each sample were sent to HydroLab Water Laboratories in Quezon City to determine the amount of soluble copper present while the other 1.5 liters of the samples were tested using the developed system. Once the result has been received, it was then compared to the result that was generated by the system. Comparative analysis of the two (2) sets of data was done.

IV. Results and Discussion

Based on the applied methodology and objectives of this study, the following results are hereby presented.

A. The reaction of Tannins to Varying Soluble Copper Concentrations

The reaction of tannins to varying soluble copper concentrations was observed and recorded using the installed camera. These color reactions are presented in Table 2.

Table 2. *Observed and Actual Color Reaction of Tannins to Soluble Copper*

Amount of Soluble Copper	Recorded Color	Actual Color	Amount of Soluble Copper	Recorded Color	Actual Color
0.5 ppm			3.0 ppm		
1.0 ppm			3.5 ppm		
1.5 ppm			4.0 ppm		
2.0 ppm			4.5 ppm		
2.5 ppm			5.0 ppm		

As illustrated in Table 2, the color reaction produced by tannins to soluble copper varies from light gray to dark green reaction. The said results are replicates of the findings of Marquez, *et al* (2008) as reflected in Figure 1. However, the color reaction presented above was gathered using a real-time image capturing feature using the MatLab application.

The colors were then plotted to a scale in a coordinate system in MatLab for color detection. These coordinates are the values of the corresponding values of the pixel values as shown in Table 3.

As reflected in Table 3, the corresponding Red-Blue-Green Codes of the color reactions were determined using the color analyzer of MatLab. The amount of copper is set and compared to the standard limit issued by the World Health Organization for Drinking water. [30] These values were correlated to the number of copper contents presented in Figure 7.

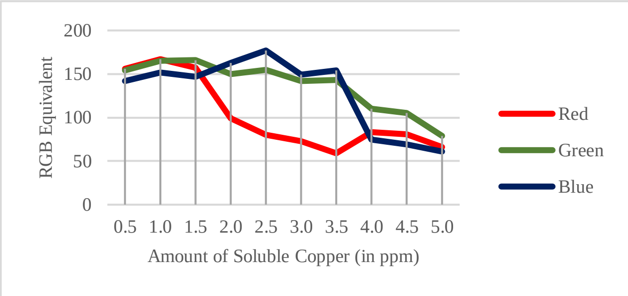


***Figure 7**. Relationship of Soluble Copper Content and RGB Assignment*

Figure 7 depicts the relationship between soluble copper and its equivalent Red-Green-Blue Assignment. It can be observed that as the amount of soluble copper increases the values of RGB decrease as well. This relationship was aligned with the findings of Marquez *et.al.* (2008) [15] where it was established that the higher the concentration of soluble metals, the darker the color reaction. Concerning the RGB values, these support the color scheme that was established that darker colors have lower RGB equivalents. [31] In relevance to the color produced, it can also be absorbed that the highest color values are in the blue and green channels from the solutions with two (2) to 3.5 ppm. This color variation is consonant with the findings of earlier studies [16] that a bluish green to black color reaction is observed for tannins and copper.

***Table 3.** RGB Equivalent of the Color Reactions*

Copper Content (in ppm)	Red Assignments	Green Assignments	Blue Assignments	Comment
0.5	156	154	142	"Less than Standard Limit"
1.0	167	165	152	"Within Standard Limit"
1.5	157	166	147	"Copper Above Standard Limit"
2.0	99	150	163	"Copper Above Standard Limit"
2.5	80	155	177	"Copper Above Standard Limit"
3.0	73	142	149	"Copper Above Standard Limit"
3.5	59	143	154	"Copper Above Standard Limit"
4.0	83	110	75	"Copper Above Standard Limit"
4.5	81	105	69	"Copper Above Standard Limit"
5.0	66	79	61	"Copper Above Standard Limit"

System Development

Once the relationship between the amount of soluble copper and its RGB color reaction was established, a program was developed that will determine the amount of copper in water samples.

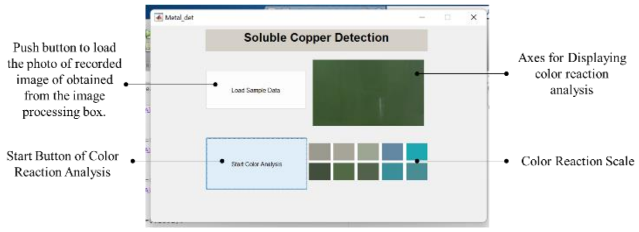


Figure 8. *User-Interface of the Developed Program*

Figure 8 shows the user interface of the MatLab file that was developed. There are two (2) push buttons that will be used by the user. First is the load sample data that loads the image at the axes located on the right side of the panel. The second button is to load the color scale and automatically compare it to the pre-defined baseline data. Thus, after this comparison, a message box will appear that shows the closest amount of soluble copper present in the water sample as illustrated in Figure 9.

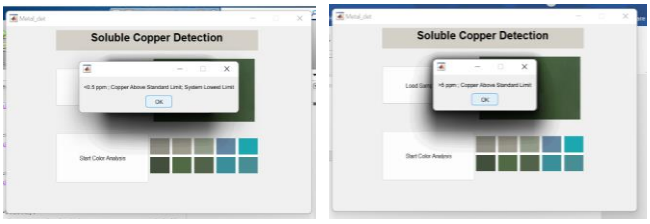


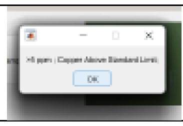
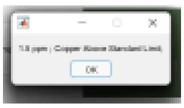
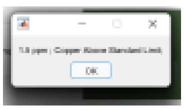
Figure 9. *Copper Content Message Box*

Figure 9 shows the corresponding message boxes that appear when the lowest (0.5 ppm) and highest (5ppm) limits of the system have been reached.

Evaluation and Algorithm Testing

The results of the water tested from various locations are presented in Table 4.

Table 4. *Result of Water Sample Evaluation*

Sample Number	**Location**	**Amount of Soluble Copper – Laboratory Result (in ppm)**	**Color Reaction Produced**	**System Result**	**Standard Limit (WHO Guidelines**	**Comments**
1	Ino-Capayang Man-Made Lake	12.31			1.3 ppm ; 1mg/L	Above Standard Limit
2	Estuary of Brgy. Lupac, Boac, Marinduque	1.330				Above Standard Limit
3	Stream of Brgy. Canat, Boac, Marinduque	1.441				Above Standard Limit

As reflected in Table 4, the copper content of the water samples ranges from 1.441 ppm to 12.31 ppm. All of these samples are above the standard limits for drinking and water uses. The water coming from the Ino-Capayang Lake has the highest copper concentration. The result of the developed system showed higher concentration values compared to the actual copper content from the laboratory results.

This limitation of the developed system to present actual content at lower scales relies on the quality of the image rendered during the observation period of the color reaction. This can be addressed using neural fitting or a convolutional neural network system with optimization through a genetic algorithm.

Given these findings, it can be deduced that the developed system can be able to determine the soluble copper in water samples (from 0.5 mg/L to 5mg/L) but calibration must be done using optimization features of genetic algorithm and the use of other approaches such as machine vision. In the analytic chemistry, aspect, it is suggested that titrimetric conditioning be done and the use of the Sigmoid function be done to improve the color resolution before analysis can be done.

V. Conclusions and Recommendations

Based on the findings of the study, the following conclusion the developed system can detect soluble copper within 0.5 mg/L (0.5 ppm) to 5 mg\L for water samples. This is accomplished through a comparison of color values rendered from baseline data.

It is recommended to use other techniques to optimize the system such as genetic algorithms and neural networks. Lastly, it is also suggested to use other techniques to process images like machine vision.

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