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Research Article

Spatial distribution of agricultural yields with elevated metal concentration of the island exposed to acid mine drainage

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Abstract: An island province in the Philippines exposed to acid mine drainage for about 22-25 years, uncovered new discovery in selected agricultural yields. The acid mine drainage was from two open mine pits of higher elevation flowing to Boac and Mogpog River system. A total of 78 various agricultural yields samples with 234 specimens were collected and analyzed from six municipalities of Marinduque, Philippines in 2019. These agricultural yields were (A) vegetables, (B) root crops, (C) fruits, and (D) rice. Inductively Coupled Plasma – Optical Emission Spectrometry (ICP-OES) Perkin Elmer Optima 8000 with ICP multi-element standard solution IV were used to detect metals concentration in the agricultural samples. Digestion of samples followed the EPA Method 200.3. Results were compared with the WHO/FAO limit followed by the identification of yields and areas that pose risks to public health. Determination of the spatial distribution was by ArcGIS. The six municipalities; i.e., Boac, Buenavista, Gasan, Mogpog, Torrijos and Sta. Cruz, were labelled as B, BV, G, M, T and S, respectively. Record showed that Sample A from G contain higher metal concentration among other yields. Manganese concentration in Samples A, B, and D were found to be higher than WHO/FAO limit. However, copper and zinc concentration in Sample C in all municipalities exceeded the allowable limit. Elevated total chromium concentration was found in Sample D collected from G, T, and S. These results would help relevant government agencies and units design strategies to mitigate the degraded agricultural lands and protect public health.

***Keywords**: acid mine drainage, spatial distribution, vegetables and crops quality*

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Introduction

The Marinduque island province of the Republic of the Philippines exposed to two mining disasters that were considered as among the most disastrous mining disasters in the world. Two abandoned open mine pits in the municipality of Sta. Cruz (i.e., at Tapian and San Antonio) exist in the island after the 1993 and 1996 disastrous events until today. The island province lies within the Pacific tropical storm path with record of at least one extreme typhoon event per year that created flooding in the area of at least once a year. This means that overflow and runoff are eminent every typhoon event. In addition, the island province sits on volcanic and/or sedimentary rocks, which means that subsurface has porous media with the capability to transport contaminants from higher to lower elevation. Marinduque is a small mountainous and agricultural island with land area of about 952.6 square kilometers and with a longest river length of about 35.8 kilometers only. Migration of environmental pollution through land surface and subsurface porous media is potential. Plants, crops, vegetables, and tress uptake metals from soil and water; concentration of which depends on the contamination level of metals in water, the soil properties (Kos et al., 2003; Singh et al., 2011; European Commission, 2013; Balkhair and Ashraf, 2016) and the species of plants (Barman et al., 2000 and Singh et al., 2012). The reported large unutilized economic agricultural and non-agricultural areas in Marinduque has been associated with the environmental pollution condition (Senoro et al., 2020a).

There had been studies about coastal and river system pollution (UNEP, 1996; Plumlee et al., 2000; David, 2002; Plumlee et al., 2014; Senoro et al., 2019), biodiversity (Sosa III et al., 2014), natural resource and its utilization (Ney al., 2019), forest cover and cave stream interconnectivity (Mascareñas et al., 2019), and confirmatory factor analysis (Prasetyo et al., 2020). However, there has been no study yet focusing on soil quality and the metals concentration in agricultural yields as well as its spatial distribution in the island province. Hence, this paper provides information on soil and some agricultural yields' quality in 2019 with respect to metals concentration. In addition, this paper illustrates the spatial distribution of targeted metals in soil and agricultural yields over the island province. This information would provide inputs to the local, regional, and national government to design strategies to mitigate the degraded agricultural lands, enhance existing or develop new guidelines/policies, and protect public health.

Materials and Methods

Thirty (30) soil samples and twenty-four (24) agricultural yield samples were collected, making a total of 162 specimens were prepared and analyzed in six municipalities of Marinduque island province. Location samples were recorded using GARMIN Montana 680 Global Positioning System (GPS).

Samples preparation and analysis

Soil samples were collected using a stainless trough and kept in a ziplock before transporting to the preparation room. The agricultural yield samples were collected from local markets identifying the name of barangay from where the samples came from. Other samples were collected from household backyards. All agricultural yields were placed in a ziplock with complete label and transported to the preparation room. Agricultural yield samples were washed, cut/sliced thinly and dried at 105ºC drying oven for 3-5 hours, depending on their moisture content, until completely dried. Utensils and laboratory equipment used in the preparation were cleaned by soap and water, rinsed with 1.5% v/v nitric acid and type 1 water every use per sample. Dried samples were ground into fine particles, using 700W with 19,000-rpm cyclonic vortex blender (Nutri-blitzer) and kept in a properly labelled disposable petri dish and/or glass vials at room temperature until digestion was performed.

Digestion of soil and agricultural yields followed the EPA Method 3050B (USEPA, 1996) and 200.3 (USEPA, 1991), respectively. The analysis of metal in these samples utilized the Inductively Coupled Plasma – Optical Emission Spectrometry (ICP-OES) Perkin Elmer Optima 8000 instrument with the multi-element standard solution IV following the EPA Method 6010C (USEPA, 2007). Eight metals were targeted in the samples such as cadmium (Cd), chromium (Cr), copper (Cu), iron (Fe), manganese (Mn), nickel (Ni), lead (Pb), and zinc (Zn).

Plotting and mapping

The ArcMap 10.7.1 was employed for the plotting of coordinates of sample locations, spatial distribution of meals concentration in soil and agricultural yields. Barangay name was recorded for samples bought from local market, and coordinates of the barangay hall were recorded and used to plot those samples bought from the local market. These samples locations are shown in Figure 1.

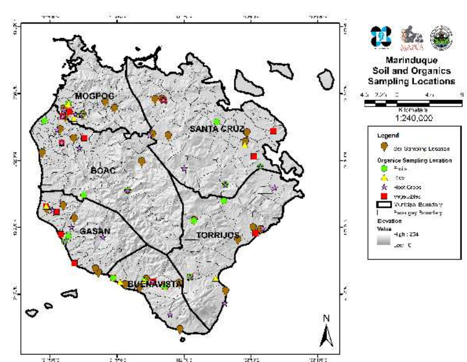

Figure 1. Sampling locations.

In addition, ArcMap 10.7.1 was used to map the spatial distribution of metals concentration in soil and agricultural yields over the island province.

Results and Discussion

The subsequent sections below discuss and illustrate the soil pollution/degradation based on elevated metal concentration, its spatial distribution in the island province, and metals concentration detected in agricultural yields. The metals concentration in agricultural yields was compared with the limit/s set by the international organization intended for human consumption.

Heavy metals in soil

Figure 2a-h illustrates the results of metals concentration in soils and its spatial distribution over the island province. Cadmium was not detected in soil. High concentration of Cr detected in large areas in Gasan; Cu at Boac and Mogpog; Fe all over the island with dominant concentration in Gasan; Mn elevated concentration over all areas with highest at Buenavista; Ni at Gasan; Pb at Mogpog and Buenavista; and Zinc at Mogpog and Boac. This corroborated the result of the study in 2018 by Sanchez et al. (2018) in which it detected heavy metals (i.e., Cr, Cu, Fe, Mn, Pb, and Zn) on free-range domestic chicken feathers. Also, these metals with the inclusion of Cd were detected in soil. Elevated metals concentration in soils tends to create a condition with microbes of high resistance to these heavy metals (Schmidt et al., 2009). Assimilation of metals by high metal resistant microbes, together with the oxidation process of iron sulphides (Udahyabhanu and Prasad, 2010) are not favorable to agricultural use as acid is released from this process into the environment. Soil pH range favorable to agriculture is 6.0-7.0 for optimum nutrients availability. The pH range of soil in Marinduque was 4.3-4.8 (Mante et al., 2019).

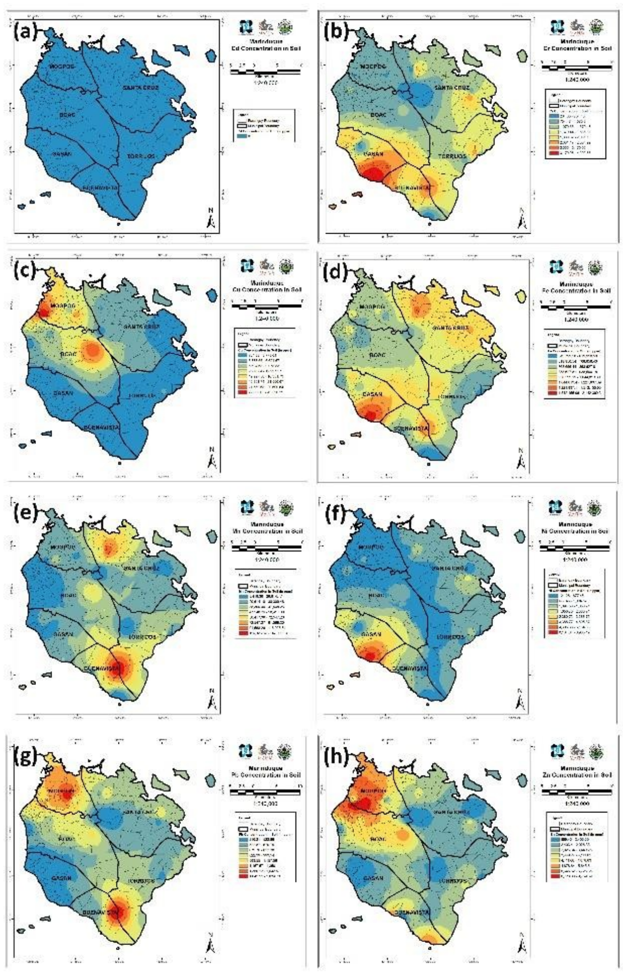

Figure 2 Spatial distribution of metals concentration in soil.

Heavy metals in some agricultural yields

Results of the ICP-OES analysis for some agricultural yields such as vegetables (Sample A), root crops (Sample B), fruits (Sample C) are presented in Figure 3-5. Whereas, result for rice is presented in Table 1. These concentrations were compared with the maximum limit set by the international organizations (FAO/WHO, 2001; FAO/WHO, 2002; FAO/WHO, 2003; FAO/WHO, 2011; FAO/WHO, 2014; ASTDR, 2012). A horizontal red line on every figure illustrates these limits. It was recorded that Mn and Zn concentration range, i.e., 1.86 – 52.84 mg/kg and 0.04 – 275.5 mg/kg, respectively, was high in all types of vegetables across the island province except for eggplant collected from Boac and Mogpog. Concentration of Cu in all types of Sample A (e.g., string beans, sweet potato tops, bitter melon, and eggplant) across the island is within the concentration limit. Concentration of Ni in vegetables cannot be determined if it exceeded the limit or not as there has been no known maximum permissible limit yet that has been set by any international organization. The metals concentration in Sample B such as raddish, turmeric, taro, sweet potato and arrowroot are shown as Figure 4. The sweet potato samples in all six municipalities did not record elevated metals concentration. The Cd, Cr, Cu, Ni and Pb in the root crops were within the set limit except for Cr in arrowroot collected at Buenavista. The elevated Zn concentration (38.04 – 569.35 mg/kg) was dominant Sample B except for sweet potato. Elevated concentration of Fe (438.46 – 509.40mg/kg) and Mn (503.78 - 765.08 mg/kg) w was detected in turmeric samples collected from Torrijos and Sta. Cruz. Among the 72 specimens, only Torrijos and Sta. Cruz had issues on elevated concentration of Mn in root crops. These metals (Cd, Cr, Cu, Mn, Ni, Pb, and Zn) were also found in food/root crops at various countries such as Argentina, Brazil, China, India, Egypt, Pakistan, Spain, and USA (Rai et al., 2019).

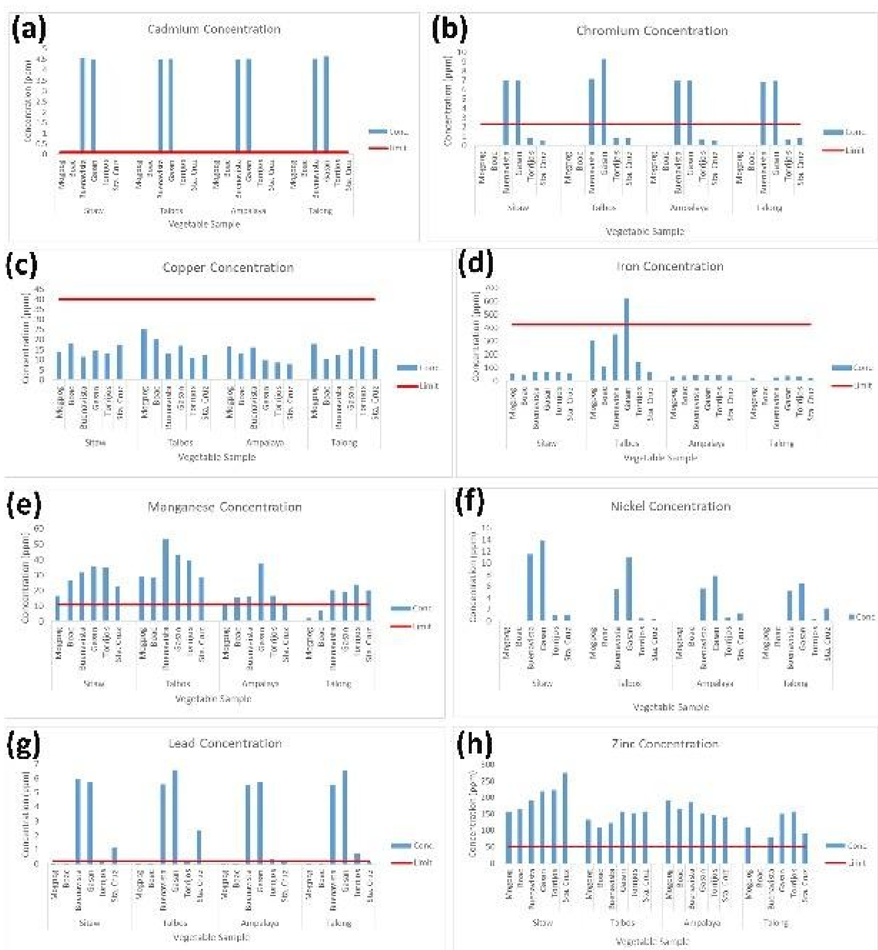

Figure 3 Metals concentration in vegetables across Marinduque.

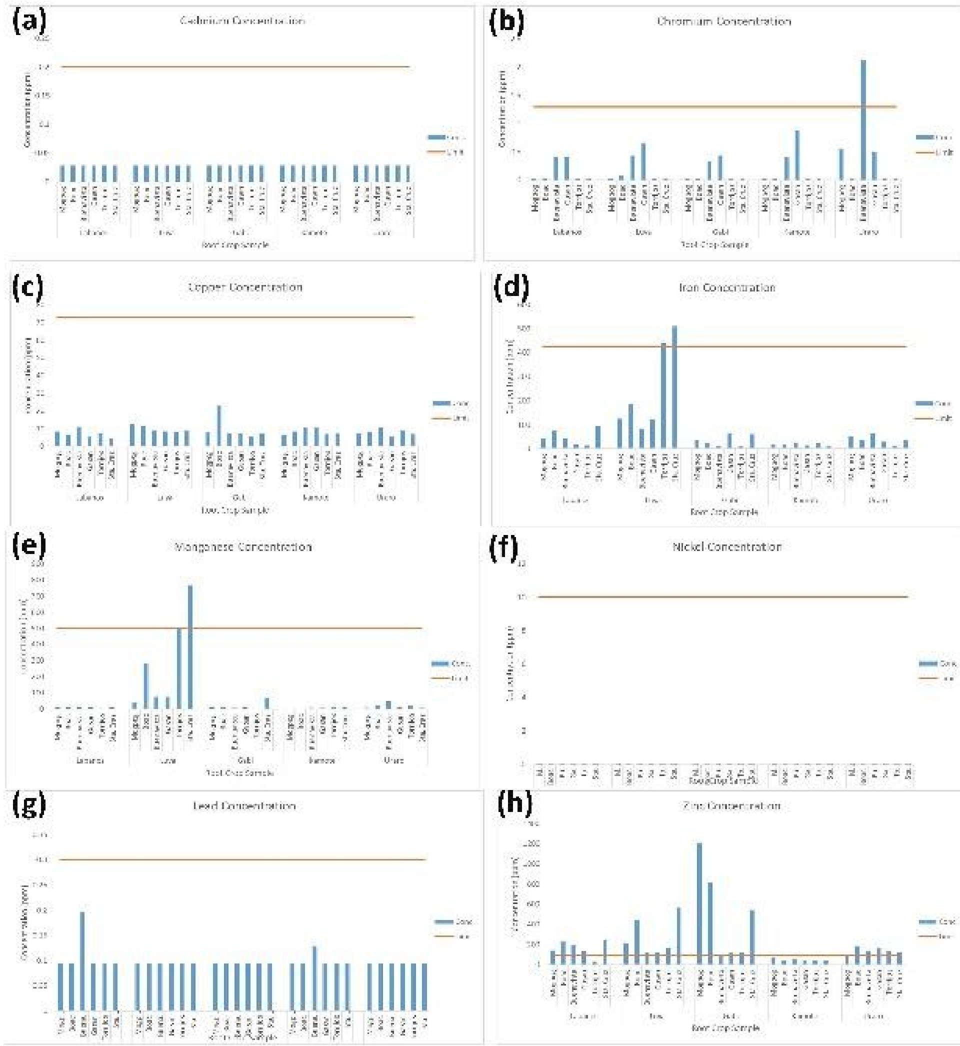

Figure 4. Metals concentration in root crops.

Fruit samples (Samples C) analyzed were tomato, coconut, soursoup and banana. Figure 5 shows the metals concentration in Sample C. Elevated Zn concentration (36.34 – 191.78 mg/kg) was found in all samples across the island. Also, Cu was found in elevated concentration (up to about 17.65 mg/kg) in Sample C except banana collected from Torrijos. The elevated concentrations of Cd, Cr, Cu, Pb and Zn in agricultural yields were considered anthropogenic intrusion (Naveedullah et al., 2013). The elevated concentration of metals in agricultural yields was attributed to the increasing level of metals contamination in soil (Intawongse and Dean, 2007). Analysis of rice/palay (Sample D) samples shown in Table 1 enumerates the symbol B, BV, G, M, T and S. These symbols/letters are names of municipalities such as Boac, Buenavista, Gasan, Mogpog, Torrijos and Sta. Cruz, respectively. Table 1 recorded elevated Mn and Zn concentration in all Sample D throughout the island province. The recorded Mn concentration range of 7.12 – 69.51 mg/kg was very alarming as the maximum limit (in red color fonts) for Mn in rice shall only be 2.0 mg/kg. Concentration of Zn in Sample D range was 69.34 -116.72 mg/kg; however, the maximum limit is only 50 mg/kg. Figures in Table 1 with blue color fonts are metal concentrations that exceed the maximum limit set by the international organization. The highest alarming concentration of Mn with respect to its allowable maximum concentration for human consumption was in rice collected from Sta. Cruz. This Mn concentration (69.51 mg/kg) in rice/palay was the highest among other municipalities. The detected concentration was 4 to 35 times higher than the maximum limit.

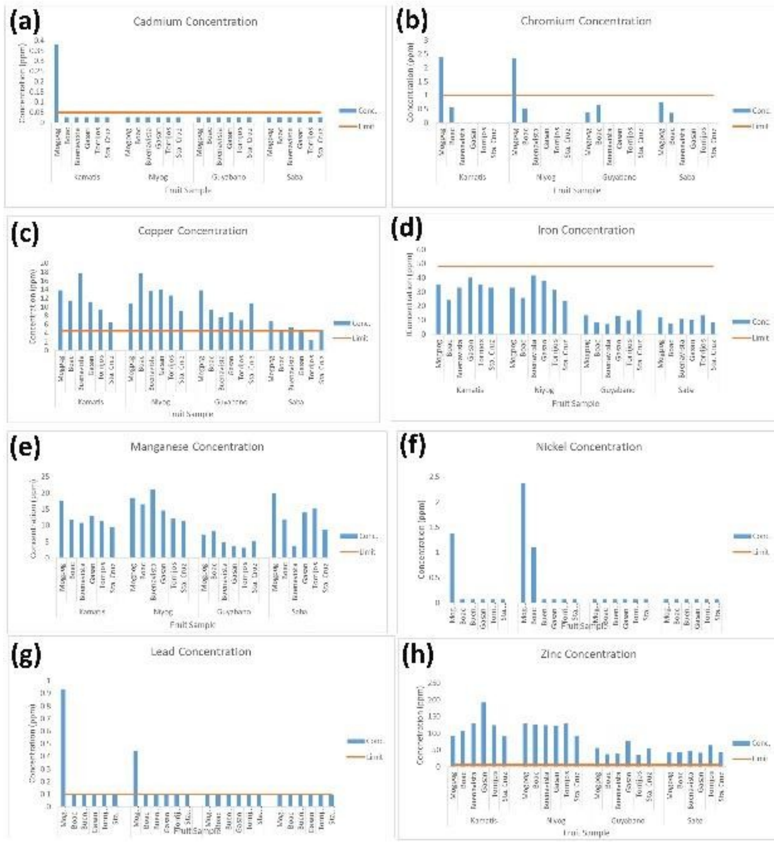

Figure 5. Metals concentrations in fruits.

Table 1 Metal concentration in rice (palay) samples, mg/kg, N=33.

Element	Cd	Cr	Cu	Fe	Mn	Ni	Pb	Zn
Max Limit	**0.2**	**1.0**	**20**	**20**	**2.0**	**10**	**0.2**	**50**
B	0.22	ND	4.88	3.61	16.79	ND	0.22	98.14
BV	ND	ND	5.02	5.92	10.46	ND	ND	88.74
G	ND	1.12	5.48	26.29	17.62	ND	ND	90.58
M	ND	ND	5.94	5.58	7.12	ND	ND	116.72
T	0.07	1.62	4.01	23.46	24.42	ND	0.25	92.98
S	0.03	7.48	5.67	60.55	69.51	0.06	0.10	69.34

ND = negative detection.

Results showed that rice from Sta. Cruz has the highest Mn concentration followed by Torrijos and Gasan. Mogpog has the highest concentration with respect to Zn in rice. The Mn and Zn data corroborates with the increasing mental health issues and cases in Sta. Cruz as reported by the Provincial Health Officer of Marinduque on July 16, 2019 (Senoro et al., 2019b). The major target organ of manganese and zinc toxicity is the central nervous system (WHO, 2001; Cadet e al., 2007; Balachandran et al., 2020) in which symptoms may appear as soon as 1 or 2 months or as 20 years after exposure. The above research work data were attributed to the mine drainage in which the island has experiencing. This leads to land degradation and affects the agricultural land use. This condition was also discussed in the work of Barman et al. (2000); Intangwogse and Dean (2007); Kumar, (2013); Magahud et al. (2015); Marquez et al. (2018). These works were carried out ‘in-vitro’, applied research in Philippines (specifically in Zambales and Negros Occidental) and Vietnam, respectively. Study area in Philippines and Vietnam was carried out in acid mine areas, too.

Conclusion

The Marinduque island province experienced mining disaster in 1993 and 1996. Environmental quality monitoring, with respect to soil and agricultural yields quality, was carried out in 2019. Results showed that elevated Mn concentration in soil was recorded all across municipalities. Other metals with elevated concentration in soil were Ni at Gasan, Pb at Mogpog and Buenavista, Zn at Mogpog and Boac. Soils with elevated metals concentration tend to create acidic environment not favorable for agricultural use. Elevated Mn and Zn concentration was also detected in all vegetables samples across the island, elevated Mn concentration in root crops collected from Sta. Cruz and Torrijos, and rice (palay) in all samples in the whole province with Sta. Cruz having the highest elevated Mn concentration. Elevated Zn concentration was detected in all agricultural yield samples; elevated concentration of Cu was detected in fruits except for banana collected from Torrijos. The elevated concentrations of these metals were attributed to acid mine drainage and as anthropogenic intrusion. Furthermore, additional comprehensive monitoring study for soil and agricultural yields is necessary in the island.

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