

Health Risk Index of Marinduque Island Population Based on Vegetables with Elevated Metals Concentration

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Highlights

- Daily Intake of Metal (DIM)
- Health Risk Index (HRI)
- Health Vulnerability Indicator
- Marinduque Island Province, Philippines
- Vegetables Quality

Abstract

This study focuses on health risk index for Marinduque province based on the concentration of inorganic metals found in vegetables produced in its six (6) municipalities. Four (4) vegetable types were analyzed using Inductively Coupled Plasma – Optical Emission Spectrometry (ICP-OES). Selection of vegetables was based on the Philippine Statistics Authority and Municipal Agricultural Data recorded as among the most produced and consumed vegetables in Marinduque. The health risk index (HRI) was calculated to understand the susceptibility of the population to mining hazards using daily intake of metals (DIM) and reference oral dose (R_fD) approach. The $HRI > 1$ for indicates adverse effects on health. The higher HRI denotes higher health risk. Based on the results of this study Cd, Cu, and Mg have $HRI > 1$ for all vegetable types. The municipality of Buenavista and Gasan have the highest level of HRI among the six municipalities of Marinduque.

Key Words: Daily Intake of Metal; Health Risk Index, Metal Concentration, Marinduque Philippines, Vegetables Quality

50 **1. Introduction**

51
52 Agriculture is the main source of livelihood for 25-30% of the labor force in the Philippines (FAO of the
53 UN, 2019). Nine percent (9%) of the national population consumed five (5) servings of vegetables and
54 fruits everyday (Gonzales et al., 2016). The average annual per capita consumption of Filipinos for
55 eggplant, okra, tomato, saba and papaya is 4.26, 1.23, 2.97, 7.60 and 2.06 kg (PSA, 2017a) respectively.
56 MIMAROPA Region, where Marinduque is located, recorded an annual per capita consumption of saba of
57 about 13.52 kg (PSA, 2017a). Marinduque consumes 4.94, 106.12, 1.24 kg per capita per annum of
58 amplaya, rice and corn (PSA, 2017b), respectively.

59
60 The desire for rapid economic and industrial development with lack of local human resource competency
61 and information tools lead to public health risk and environmental quality problems. Heavy metal
62 contamination in agricultural lands caused by anthropogenic activities has become a predominant concern
63 for developing and developed countries alike (Lui et al., 2011). The soil uptakes both toxic and essential
64 elements, the water carries impurities present in the environment, and the air contains pollution from
65 atmospheric deposition. Different environmental media contributes to the accumulation of heavy metals or
66 toxins in the edible and non-edible parts of vegetables and other crops (Liang et al., 2018).

67
68 In 1993 and 1996, mine disasters happened in the province of Marinduque which unleashed wastes to
69 Mogpog and Boac river on 6 December 1993 and 24 March 1996 (Magalang,2005), respectively. Three
70 municipalities in Marinduque island province namely Boac, Mogpog, and Santa Cruz were directly affected
71 by the disaster. The incident was considered as Philippine's worst mining disaster (Cinco, 2013) leading to
72 the closure of mining operation in the island in 1997. The mining operations in the Marinduque have
73 abandoned two (2) open mine pits. In 2018, a new research study was comisioned by the DOST-PCHRD
74 (Department of Science and Technology – Philippine Council for Health Research and Development) that
75 conducted sediment quality assessment. This study recorded the concentration of copper and manganese of
76 Boac and Mogpog River sediments was 5 and 158 times higher (Senoro et al., 2019) than the concentration
77 recorded by the study of David, (2002) conducted in 1998. Other metals found on sediments were
78 chromium, iron, lead, cadmium, zinc, nickel and among other metals found in traces (Senoro et al., 2019).
79 Metals concentration has been seen in various plants (Nworie et al., 2019; Tangahu etl., 2011; Navarete et
80 al., 2017) illustrating plants uptake by which perceived to be a pathway to human ingestion. Hence, this
81 paper focuses on the metals concentration assessment in vegetables produced and consumed in Marinduque.

82
83 Marinduque Island Province, which is under the MIMAROPA Region, has about 234,521 population with
84 growth rate of 0.55% [PSA, 2015], is composed of 218 Barangays in six (6) municipalities. Based on the
85 population growth rate, it is expected that it has population of about 240,970 in 2019. According to the
86 Provincial Health Officer, they have recorded increased mental health problems, skin diseases, hypertension
87 cases and significant number of cancer patients.

88 89 *1.1 Health Risk Assessment*

90
91 The Health Risk Assessment (HRA) is typically carried out using the technique of determining health risk
92 index (HRI) and daily intake of metal (DIM). Excessive heavy metal content in food may cause number of
93 diseases. The Health Risk Index (HRI) greater than 1 ($HRI > 1$) is considered to be unsafe for human health.
94 HRI (Eq. 1) determination has been used in various studies (Mahmood and Malik, 2014; Khan et. al, 2014;
95 Okereke et al., 2016; Paula et al., 2016] to define health risks to human brought about by the presence of
96 metals in edible stuffs and in the environment. The DIM (Eq. 2) defines the probable daily ingestion of
97 metal by human. It is assessed to approximate the average daily loading of metal into the human body.

$$HRI = \frac{DIM}{R_f D} \quad (1)$$

where, $R_f D$ represents reference oral dose, details are shown in Table 1.

$$DIM = (C_{\text{metal}} \times C_{\text{factor}} \times D_{\text{food intake}}) / B_{\text{average weight}} \quad (2)$$

where, C_{metal} denotes metal concentration in plant (mg/kg), C_{factor} is 0.085 which is the conversion factor for fresh vegetable weight to dry weight (Rattan, 2005). $D_{\text{food intake}}$ denotes daily intake of vegetable (g/person/day), and $B_{\text{average weight}}$ denotes average body mass of the consumers. The average body weight of the consumers is 56.4 kg (World Data, N.D.). However, the average weight of Philippine men and women is 61.3 and 54.3 kg [FNRI-DOST, 2014], respectively.

Table 1. Reference Oral Dose for metals.

| Element | Reference Oral Dose (mg/kg/day) | Reference |
|---------|---------------------------------|--------------------------------|
| Cd | 0.001 | US EPA, 2015 |
| Co | 0.043 | Food and Nutrition Board, 2004 |
| Cr | 1.500 | US EPA IRIS, 2011 |
| Fe | 0.700 | US EPA, 2015 |
| Mn | 0.140 | FAO/WHO, 2011 |
| Ni | 0.020 | US EPA IRIS, 2011 |
| Pb | 0.004 | US EPA, 2015 |
| Zn | 0.300 | FAO/WHO, 2011 |
| Cu | 0.040 | US EPA, 2015 |
| Mg | 0.950 | US EPA IRIS, 2011 |

At concentrations above the tolerable intake level cause adverse health effects in target internal organ/s, bone defects, skin problems, and cancer at acute or chronic exposure (Jarup, 2013). Aside from skin penetration through air exposure, food ingestion is the most common pathway of toxic substances or metals/elements to enter the human system. Hence, the objective this study was to determine the metals concentration in vegetable products in the six municipalities of Marinduque Island province. Further, to assess the health risks due to eating identified vegetables with elevated metals concentration.

2. Methods

2.1. Collection of Vegetables

The vegetable samples were collected from the six municipalities of Marinduque namely Boac, Mogpog, Gasan, Buenavista, Torrijos, and Santa Cruz. These municipalities are labelled as B, M, G, BV, T and S, respectively, in all samples. Four (4) types of vegetable samples were collected in the public markets of the respective municipalities recording the name of barangay where these vegetables were harvested. Other samples were given by households from their backyard garden. The vegetables chosen were among the top produced and consumed vegetables in Marinduque according to the Municipal Agricultural Data namely string beans, sweet potato tops, bitter melon, and eggplant *i.e.* *Vigna unguiculata*, *Ipomoea Batatas*, *Momordica charantia*, and *Solanum melongena*, respectively. About ¼ kg to ½ kg per sample were bought

130 per municipality and placed inside ziplocks. Ice packs and ice were placed together with the samples inside
131 the coolers during transport from Marinduque to Manila. Total number of vegetables collected were 24 all
132 over the island.

133 134 2.2. *Preparation and Analysis*

135 136 2.2.1. *Drying and Grinding*

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138 The vegetable samples collected were cut and sliced into thin sections, ensured that the knives and chopping
139 board were cleaned with unscented soap, tap water, rinsed with 1.5% v/v nitric acid, and rinsed with type I
140 water every after use to ensure no cross-contamination. The sliced samples were layed onto a parchment
141 paper placed on a drying tray. Samples were dried at 105°C oven for 3-5 hours depending on their moisture
142 content until complete dryness is obtained. Drying at 65°C is also acceptable but with longer drying hours.
143 Drying vegetables in Marinduque was also permitted and carried out. Dried samples were powdered using
144 a high-speed blender until fine powder was formed. The blender was cleaned with unscented soap, tap
145 water, rinsed with 1.5% v/v nitric acid and type I water every after grinding. Ground samples were stored
146 in properly labelled PE bags at room temperature until digestion.

147 148 2.2.2. *Heavy Metal Analysis*

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150 EPA Method 200.3 (USEPA, 1991) for digestion of samples was employed. Digested samples were
151 subjected to the Inductively Coupled Plasma - Optical Emission Spectrometry (ICP-OES) Perkin Elmer,
152 Optima 8000 instrument for the determination of heavy metal concentrations. The ICP multi-element
153 standard solution IV in diluted HNO₃ was used. Calibration curves attained with r² range of 0.995 – 0.999
154 for a total of 72 specimen (triplicate per sample). The focus of metals analysis was, and its Limits of
155 Detection (LOD), for Cd, Cr, Fe, Mn, Ni, Pb, Zn, Cu, and Mg were 0.0037, 0.0005, 0.0004, 0.0003, 0.0006,
156 0.0122, 0.0009, 0.0032, and 0.01434, respectively. The location of the ICP-OES analysis was the the Wet
157 laboratory of Yuchengco Innovation Center, Mapúa University in Intramuros, Manila, Philippines.

158 159 2.3. *Health Risk Index (HRI)*

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161 The HRI and DIM were calculated using Eq. (1) and (2) elaborated above. HRI was computed as the ratio
162 of estimated exposure of vegetable samples and oral reference dose (R_fD) (Ramteke, 2016). The DIM tells
163 the probable ingestion rate of metal. It was assessed to approximate the average metal loading into the
164 human body daily. The DIM and the HRI were computed simultaneously. The results of metals
165 concentration/s in vegetables from ICP-OES were used as inputs to Eq. (1) and (2); then compared with
166 the limits set by the Philippine government or the World Health Organization. This is to determine health
167 risk status in Marinduque island.

168 169 **3. Results and discussion**

170 171 3.1. *Health Risk Assessment*

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173 Based from the Philippine Statistics Authority, the estimated vegetable intake is 2.55, 2.84, 2.71, 5.11
174 kg/person-year for string beans, sweet potato tops, bitter melon, and eggplant, respectively. The average
175 body weight of a person residing in the Philippines is 60.6kg for the male and 52.2kg for the female yielding
176 an average of 56.4 kg for a regular adult. Table 2 illustrates the potential daily intake of concerned metals
177 per identified vegetable type for every municipality.

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Table 2. Calculated daily intake of metals per vegetable type per municipality, mg/g/day/person in Marinduque

| Vegetable Type | Mun. | Cd | Cr | Fe | Mn | Ni | Pb | Zn | Cu | Mg |
|-------------------|------|--------|--------|--------|--------|--------|--------|--------|--------|-------|
| String Beans | M | 0.0019 | 0.0003 | 0.6474 | 0.1759 | 0.0003 | 0.0064 | 1.6412 | 0.1484 | 25.16 |
| | B | 0.0019 | 0.0003 | 0.5092 | 0.2813 | 0.0003 | 0.0064 | 1.7252 | 0.1919 | 27.29 |
| | BV | 0.0482 | 0.0733 | 0.6946 | 0.3320 | 0.1212 | 0.0630 | 2.0125 | 0.1200 | 21.05 |
| | G | 0.0476 | 0.0732 | 0.7031 | 0.3761 | 0.1471 | 0.0606 | 2.3103 | 0.1545 | 24.99 |
| | T | 0.0019 | 0.0089 | 0.7216 | 0.3662 | 0.0108 | 0.0029 | 2.3571 | 0.1353 | 29.85 |
| | S | 0.0019 | 0.0059 | 0.5762 | 0.2401 | 0.0099 | 0.0122 | 2.8987 | 0.1810 | 21.26 |
| Sweet Potato Tops | M | 0.0021 | 0.0003 | 3.5939 | 0.3434 | 0.0003 | 0.0071 | 1.5498 | 0.2928 | 25.74 |
| | B | 0.0021 | 0.0003 | 1.3136 | 0.3304 | 0.0003 | 0.0071 | 1.2620 | 0.2388 | 20.82 |
| | BV | 0.0526 | 0.0844 | 4.1113 | 0.6190 | 0.0641 | 0.0654 | 1.4509 | 0.1554 | 25.31 |
| | G | 0.0531 | 0.1092 | 7.3021 | 0.5026 | 0.1291 | 0.0771 | 1.8501 | 0.1998 | 32.23 |
| | T | 0.0021 | 0.0095 | 1.6411 | 0.4656 | 0.0052 | 0.0034 | 1.7941 | 0.1303 | 33.30 |
| | S | 0.0021 | 0.0096 | 0.8299 | 0.3370 | 0.0035 | 0.0272 | 1.8507 | 0.1455 | 26.69 |
| Bitter Melon | M | 0.0020 | 0.0003 | 0.4216 | 0.1309 | 0.0003 | 0.0068 | 2.1273 | 0.1838 | 25.97 |
| | B | 0.0020 | 0.0003 | 0.4666 | 0.1708 | 0.0003 | 0.0068 | 1.8503 | 0.1482 | 21.00 |
| | BV | 0.0505 | 0.0772 | 0.4797 | 0.1843 | 0.0625 | 0.0622 | 2.0965 | 0.1827 | 20.88 |
| | G | 0.0508 | 0.0779 | 0.5340 | 0.4198 | 0.0873 | 0.0642 | 1.6943 | 0.1105 | 28.05 |
| | T | 0.0020 | 0.0082 | 0.5260 | 0.1877 | 0.0066 | 0.0041 | 1.6573 | 0.0969 | 24.79 |
| | S | 0.0020 | 0.0060 | 0.4508 | 0.1259 | 0.0149 | 0.0028 | 1.5491 | 0.0889 | 18.61 |
| Eggplant | M | 0.0039 | 0.0006 | 0.4120 | 0.0392 | 0.0006 | 0.0129 | 2.3222 | 0.3758 | 34.98 |
| | B | 0.0039 | 0.0006 | 0.0675 | 0.1532 | 0.0006 | 0.0129 | 0.0009 | 0.2200 | 42.39 |
| | BV | 0.0956 | 0.1436 | 0.6419 | 0.4265 | 0.1094 | 0.1173 | 1.6451 | 0.2640 | 37.02 |
| | G | 0.0986 | 0.1458 | 0.8343 | 0.4096 | 0.1369 | 0.1378 | 3.1579 | 0.3138 | 51.16 |
| | T | 0.0039 | 0.0155 | 0.7284 | 0.5048 | 0.0071 | 0.0155 | 3.3236 | 0.3561 | 50.26 |
| | S | 0.0039 | 0.0179 | 0.5243 | 0.4203 | 0.0459 | 0.0033 | 1.9297 | 0.3200 | 31.48 |

M = Mogpog, B = Boac, BV = Buenavista, G = Gasan, T = Torrijos, S = Sta. Cruz.

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The calculated HRI values in the four types of vegetable samples, when ingested by human, are shown in Table 3. An HRI > 1 for any metal in the food (vegetables in this case) indicates that the consumer population are at risk. The Cd, Cu, and Mg have HRI > 1 for all vegetable types in all municipalities with HRI ranging from 1.92 – 98.58, 2.22 – 9.39, and 19.59 – 53.85, respectively. The municipality of Gasan and Buenavista have consistently high HRI for all the vegetable types except for Cr which has HRI < 1 for all vegetable types in all municipalities. The most alarming is the very high HRI for cadmium for Buenavista and Gasan reaching up to a range of 47.6 – 98.6. Cadmium and chromium (depending on its species in the environment) are considered not degradable (NCM, 2003) and permanent environmental and health risk (Rosales et al., 2016). Chromium has several oxidation states, the most common forms that are found in the environment are Cr(0), Cr(III) and Cr(VI). The Cr(0) is produced by the steel industry, Cr(III) is less toxic and precipitate at neutral to basic pH; will solubilize at acidic pH. The Cr(VI) is highly mobile, toxic in the

194 environment, and highly soluble at all pH levels (Oliviera, 2012). Hence, Cr(III) is attributed to the form of
 195 Cr in the study area based on the recorded concentration of Cr in vegetables and in the environment.
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Table 3. Health risk index of metals (HRI >1 in bold text are at high risk).

| Vegetable Type | Mun. | Cd | Cr | Fe | Mn | Ni | Pb | Zn | Cu | Mg |
|----------------------|------|--------------|--------|--------------|-------------|-------------|--------------|--------------|-------------|--------------|
| String Beans | M | 1.92 | 0.0002 | 0.92 | 1.26 | 0.01 | 1.60 | 5.47 | 3.71 | 26.48 |
| | B | 1.92 | 0.0002 | 0.73 | 2.01 | 0.01 | 1.60 | 5.75 | 4.80 | 28.72 |
| | BV | 48.20 | 0.0489 | 0.99 | 2.37 | 6.06 | 15.75 | 6.71 | 3.00 | 22.16 |
| | G | 47.61 | 0.0488 | 1.00 | 2.69 | 7.35 | 15.15 | 7.70 | 3.86 | 26.31 |
| | T | 1.92 | 0.0059 | 1.03 | 2.62 | 0.54 | 0.72 | 7.86 | 3.38 | 31.43 |
| | S | 1.92 | 0.0039 | 0.82 | 1.71 | 0.49 | 3.06 | 9.66 | 4.52 | 22.38 |
| Sweet Potato Tops | M | 2.14 | 0.0002 | 5.13 | 2.45 | 0.02 | 1.79 | 5.17 | 7.32 | 27.09 |
| | B | 2.14 | 0.0002 | 1.88 | 2.36 | 0.02 | 1.79 | 4.21 | 5.97 | 21.91 |
| | BV | 52.63 | 0.0562 | 5.87 | 4.42 | 3.21 | 16.35 | 4.84 | 3.89 | 26.65 |
| | G | 53.08 | 0.0728 | 10.43 | 3.59 | 6.46 | 19.27 | 6.17 | 4.99 | 33.92 |
| | T | 2.14 | 0.0063 | 2.34 | 3.33 | 0.26 | 0.85 | 5.98 | 3.26 | 35.05 |
| | S | 2.14 | 0.0064 | 1.19 | 2.41 | 0.18 | 6.81 | 6.17 | 3.64 | 28.09 |
| Bitter Melon | M | 2.04 | 0.0002 | 0.60 | 0.93 | 0.02 | 1.70 | 7.09 | 4.60 | 27.34 |
| | B | 2.04 | 0.0002 | 0.67 | 1.22 | 0.02 | 1.70 | 6.17 | 3.71 | 22.11 |
| | BV | 50.47 | 0.0514 | 0.69 | 1.32 | 3.13 | 15.55 | 6.99 | 4.57 | 21.97 |
| | G | 50.78 | 0.0519 | 0.76 | 3.00 | 4.37 | 16.05 | 5.65 | 2.76 | 29.52 |
| | T | 2.04 | 0.0054 | 0.75 | 1.34 | 0.33 | 1.02 | 5.52 | 2.42 | 26.10 |
| | S | 2.04 | 0.0040 | 0.64 | 0.90 | 0.74 | 0.69 | 5.16 | 2.22 | 19.59 |
| Eggplant | M | 3.85 | 0.0004 | 0.59 | 0.28 | 0.03 | 3.21 | 7.74 | 9.39 | 36.82 |
| | B | 3.85 | 0.0004 | 0.10 | 1.09 | 0.03 | 3.21 | 0.00 | 5.50 | 44.62 |
| | BV | 95.57 | 0.0957 | 0.92 | 3.05 | 5.47 | 29.32 | 5.48 | 6.60 | 38.97 |
| | G | 98.58 | 0.0972 | 1.19 | 2.93 | 6.85 | 34.44 | 10.53 | 7.85 | 53.85 |
| | T | 3.85 | 0.0103 | 1.04 | 3.61 | 0.35 | 3.89 | 11.08 | 8.90 | 52.91 |
| | S | 3.85 | 0.0119 | 0.75 | 3.00 | 2.29 | 0.82 | 6.43 | 8.00 | 33.14 |

M = Mogpog, B = Boac, BV = Buenavista, G = Gasan, T = Torrijos, S = Sta. Cruz.

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 198 **4. Conclusions**
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200 Some vegetables in the province of Marinduque recorded elevated concentration of heavy metals. Elevated
 201 cadmium, copper and magnesium concentration was recorded in four targeted vegetable types in six
 202 municipalities resulting to HRI of greater than 1. Elevated manganese concentration in string beans and
 203 sweet potato tops was also recorded in six municipalities. The municipality of Mogpog has lesser health
 204 risk for bitter melon (amplaya) and eggplant. Further, Sta. Cruz municipality has lesser health risk for bitter
 205 melon, too. The majority of the HRI computed for each vegetable types in every municipality are greater
 206 than 1 posing health risk to the residents of the province. The highest value of HRI was Cd in eggplant
 207 with HRI equal to 98.58 produced in Gasan followed by 95.57 produced in Buenavista. The lowest levels

208 of HRI is found in string beans. The municipality of Buenavista and Gasan have the highest HRI level
209 which corresponds to highest health risk among the six municipalities of Marinduque.

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