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Assessment of Potential Human Health Risks from Exposure to Select Heavy Metals in Road Dust Around Mining Sites in Carrascal, Surigao Del Sur, Philippines

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Abstract Road dust samples around human settlements nearby mining areas in Carrascal, Surigao del Sur, Philippines were investigated to quantify the levels of chromium (Cr), nickel (Ni), manganese (Mn) and zinc (Zn) and assess the potential health risks from selected heavy metals. Metal concentrations of chromium (Cr), nickel (Ni), manganese (Mn) and zinc (Zn) in road dust were analyzed using Energy Dispersive X-ray Fluorescence (EDXRF) spectrometer. Analytical results showed that the average concentrations of Cr, Ni, Mn, and Zn were very high at 15,668, 14,814, 7,054, and 684 mg/kg, respectively. Measured concentrations exceeded standards stipulated in international regulations and guidelines by several orders of magnitude. Mortality and morbidity cases with causes probably related to dust inhalation (e.g., upper respiratory tract infection and pneumonia) noticeably increased during the peak years of mining operations. Non-carcinogenic health risk assessment revealed that children are more prone to develop non-carcinogenic health effects than adults (HI values >1 for Cr, Ni and Mn for children and only Cr for adults), owing to their smaller body weight and activities exposing them to these metals via the ingestion route. Assessment of carcinogenic risk value or the lifetime probability of an individual to develop cancer due to exposure to Ni revealed that such risk is negligible. This study suggests that mining companies and government units should proactively take measures to reduce dust exposure (e.g., dust inhalation) of mine workers and residents in nearby communities. In developing management measures, children should be provided with attention given their greater risk of developing dust-related illnesses and diseases.

Keywords Heavy Metals, Mining, Road Dust, Potential Health Risk, Non-Carcinogenic Risk, Carcinogenic Risk

1. Introduction

In 2010, the municipality of Carrascal, Surigao del Sur, Philippines shifted its course from being an agricultural and fisheries town to embrace mining and exploration. To date, three (3) different mining companies operate in Carrascal, covering a total land area of 9,669 ha (4,799, 321.40, and 4,547.76 ha). Mining abruptly boosted the municipality's economy and became highly favored by the local community, including the indigenous people, as it provided employment opportunities and enhanced social development. Businesses also sprouted in response to the demands of an increasing population and the improved capacity to pay for goods and services. These opportunities notwithstanding, people still recognize the negative impacts of mining operations to the environment.

Mining operations generate large amounts of pollutants, including a significant amount of dust containing metals and potential toxic elements [1]. Many countries reported that road dust contains toxic organic and inorganic contaminants, including heavy metals such as copper, nickel, chromium, and lead, that could exceed allowable levels in the environment [2-5]. Metals in road dust that enter the body through inhalation, ingestion, and dermal contact, could affect human health and could eventually result in respiratory, cardiovascular, and other organ diseases, sometimes even leading to death [2,6].

Many countries have focused on quantifying the concentration, spatial distribution, and potential health risk of heavy metal-loaded dust in urban environments. However, there is very little information on the levels of heavy metals in road dust near mining areas and there is an even greater challenge regarding pollution abatement. In the Philippines, large-scale mining is widespread across and no research has yet been conducted to quantify metal concentrations in road dust from human settlements adjoining mining areas.

The municipality of Carrascal is one of the many areas in the Philippines that has engaged in the mining industry for many years now. Currently, it is facing major challenges in preventing various negative effects of mining operations to the environment, including the release of large quantities of dust into the atmosphere. It is worthy to note that elevated levels (exceeding maximum allowable concentrations) of metallic particles in dust may pose potential health risks to mine workers, nearby communities and even those living afar due to dust dispersion and resuspension. However, people are not aware of the hazards that ingestion and inhalation of and dermal contact with dust may pose.

Thus, this study was conducted to (i) quantify the concentrations of heavy metals, specifically chromium (Cr), manganese (Mn), nickel (Ni) and zinc (Zn), in road dust collected from human settlements near mining areas in Carrascal, Surigao del Sur, Philippines; (ii) compare the current level of road dust metal concentrations in the study site against reported values in road dust in other countries and against regulatory and guideline thresholds published by international agencies; (iii) assess the potential non-carcinogenic and carcinogenic human health risks to both adults and children from exposure to heavy metals (Cr, Mn, Ni, and Zn) in road dust in the study area; (iv) assess the historical mortality and morbidity records of the study area commencing the year before the peak of mining operations until after the peak years of operations; and (v) recommend steps to enhance existing management measures for the prevention or minimization of exposure to metal-loaded dust in areas near mining sites in Carrascal, Surigao del Sur, Philippines.

2. Materials and Methods

2.1. Sampling Location

The study was conducted in the municipality of Carrascal, Surigao del Sur, Philippines. It is situated along the northern coast of Mindanao, with geographic coordinates of 9°22'13" N and 125°56'57" E (Figure 1). It is bounded by the municipality of Claver, Surigao del Norte in the north, Cantilan, Surigao del Sur in the south, the Pacific Ocean in the east, and Agusan del Norte in the west.

2.2. Sample Collection, Preparation and Analysis

Road dust samples were collected from five sampling stations established in areas near mining sites in Carrascal, Surigao del Sur (Figure 1). At each sampling station, three homogenized samples of dust approximately weighing 300g each were collected from roads, pavements, house yards, and leaves, among others, using polyethylene brush and dustpan (cleaned with tap water, followed by distilled water, and dried after every sampling) and were placed in labeled resealable plastic bags. Then, all collected dust samples were air dried using oven before sieving. Dried samples were sieved through 100 µm mesh size metal sieve to remove large particles, then through 75 µm mesh size metal sieve to acquire fine dust samples. The latter mesh size was selected for metal analysis considering that dust particles with sizes below 75 µm can be easily resuspended to the atmosphere, which can then be inhaled directly during breathing [7]. Approximately 2-3 g of dust from each of the 15 sieved samples were taken and placed into XRF cups (labelled with corresponding sample code). One (1) empty XRF cup was placed as sample blank to account for possible unseen dust in the cups. The prepared cups were then placed into the spectrometer as instrument form for heavy metal analysis. The concentration of metals was quantitatively analyzed in Epsilon 4 software following the Omnian (standardless) module. Lead (Pb) was also considered as parameter during the analysis but no concentration found after the analysis.

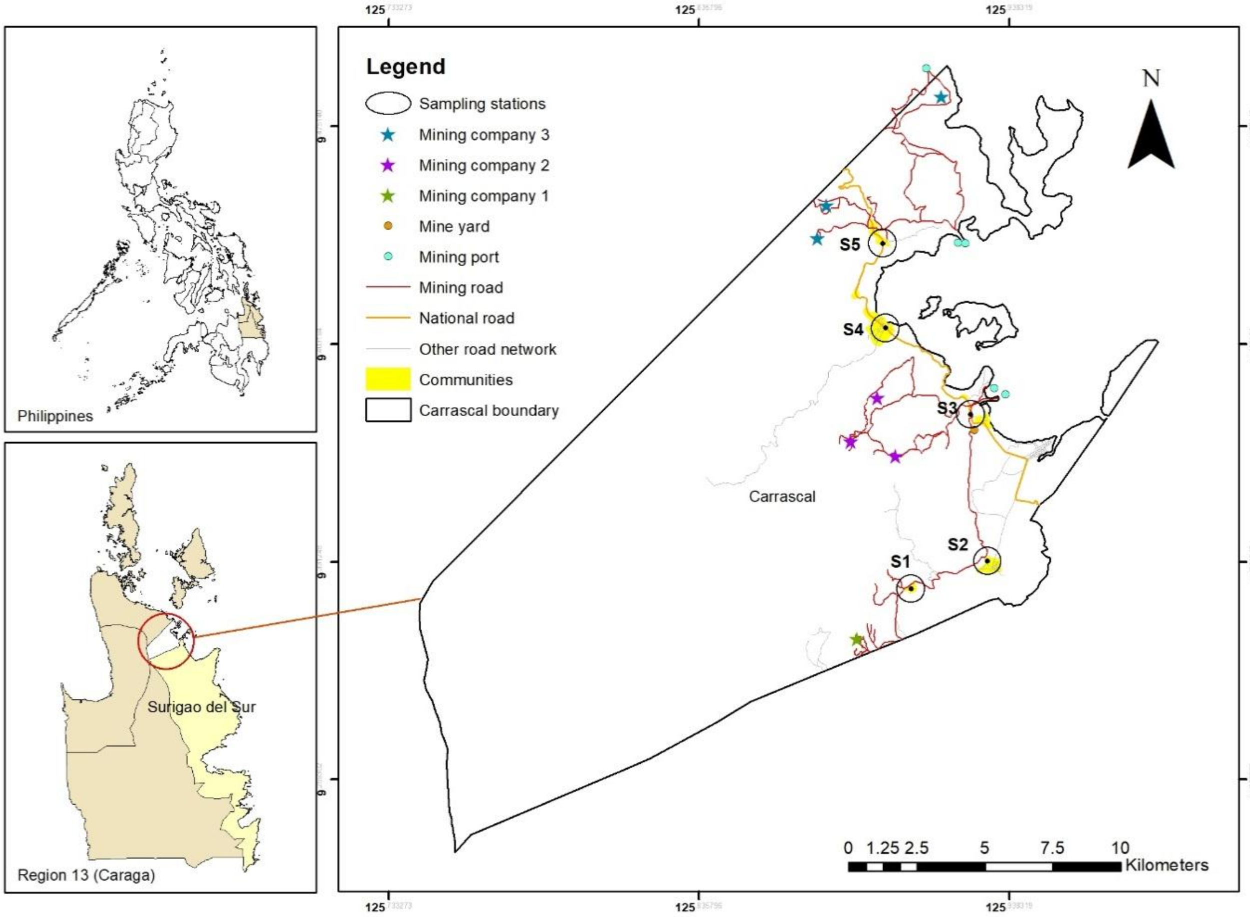

Figure 1. Map showing the sampling sites in Carrascal, Surigao del Sur, Philippines

2.2.1. Potential Health Risk Assessment

In this study, non-carcinogenic and carcinogenic risks from exposure to metal-loaded dust in Carrascal were assessed using the risk assessment model developed by the United States Environmental Protection Agency [8-9].

2.2.2. Non-carcinogenic Risk Assessment

Exposure for non-carcinogenic risk assessment was expressed in terms of daily dose contacted through ingestion, inhalation of dust particles, and skin contact. The calculation for human (children and adult) exposure to heavy metals in road dust for each exposure pathway was based on equations 1-3. ADD stands for Average Daily Dose with a unit of mg/kg/day (9). The parameters used for each equation are highlighted in Table 1:

$$ADD_{ing} = \frac{c \times R_{ing} \times EF \times ED}{BW \times AT} \; X \; 10^{-6} \qquad (1)$$

$$ADD_{inh} = \frac{c \times R_{inh} \times EF \times ED}{PEF \times BW \times AT} \qquad (2)$$

$$ADD_{derm} = \frac{c \times SA \times SL \times ABS \times EF \times ED}{BW \times AT} X \; 10^{-6} \qquad (3)$$

Table 1. Parameters used in health risk assessment.

EXPOSURE VARIABLES	DESCRIPTION	UNIT	VALUE		REFERENCES
			Children	Adults	
C	Concentration of metals in dust	mg/kg			Present study
R_{ing}	Ingestion rate	mg/day	200	100	USEPA (2001)
R_{inh}	Inhalation rate	m^3/day	7.63	20	USEPA (2001)
EF	Exposure frequency	days/year	183	183	Site specific
ED	Exposure duration	years	6	24	USEPA (2001)
BW	Average body weight	kg	15	70	USEPA (2001)
AT	Average time (Non-carcinogenic)	days	365xED	365xED	USEPA (2001)
	Average time (carcinogenic)	days	70x365[a]	70x365	USEPA (2001)
PEF	Particle emission factor	m^3/day	$1.36x10^9$	$1.36x10^9$	USEPA (2001)
SA	Exposed skin area	cm^2	2800	5700	USEPA (2001)
SL	Skin adherence factor	mg/cm^2/day	0.2	0.7	USEPA (2001)
ABS	Dermal absorption factor	unitless	0.001	0.001	US Department of Energy (2004)

The calculated average daily dose (ADD) per exposure pathway of metals was subsequently used to assess the hazard quotient (HQ) and hazard index (HI), which are terms used to express the non-carcinogenic risk of exposure. HQ is used to compare exposure estimates of metals to its corresponding reference dose. Assessment for non-carcinogenic effects, or the potential of human exposure to develop into non-cancer health effects as a result of daily exposure to heavy metal concentrations, was measured using a model developed by USEPA [8], which is expressed by the following equation:

$$HQ = \frac{ADD_i}{R_f D}$$

where, *HQ* is the hazard quotient, ADD_i is daily exposure amount of heavy metals in street dust per exposure pathway, R_fD is the reference dose or the daily intake rate that is estimated to pose no applicable risk of adverse health effect [10] (see Table 2).

Table 2. The reference dose of metals in this study (mg/kg/day) [11]

	Mn	Ni	Cr	Zn
R_fD_{ing}[a]	4.70E-02	2.00E-02	3.00E-03	3.00E-01
R_fD_{inh}[b]	1.43E-05	2.06E-02	2.86E-05	3.00E-01
R_fD_{dermal}[c]	1.84E-03	5.40E-03	6.00E-05	6.00E-02

[a] Reference dose for ingestion

[b] Reference dose for inhalation;

[c] Reference dose for dermal contact

Hazard index (HI) was used to further estimate the total hazard of exposure to concentrations of heavy metals in dust. According to USEPA [8], HI can be calculated using the following equation:

$$HI = \sum HQ$$

where, HI is the summation of HQs.

The interpretation of HI values for non-carcinogenic risk is shown in Table 3.

Table 3. Hazard index value description [8]

HI VALUES	MEANING
HI ≤ 1	Indicates no significant risk of non-carcinogenic effects
HI>1	Indicates a chance that non-carcinogenic effects may occur.

2.2.3. Carcinogenic Risk Assessment

The lifetime average daily dose (LADD) is a dose often presented to describe lifetime probability of an individual in response to effects such as cancer [12]. For Ni, which is a human carcinogen [13], LADD through inhalation exposure was used in assessing cancer risk. Calculation of LADD was done using equation 6. Parameters used in the equation are indicated in Table 1:

$$LADD = \frac{c \times EF}{AT \times PEF} \times \left(\frac{R_{inhchild} \times ED_{child}}{BW_{child}} + \frac{R_{inhadult} \times ED_{adult}}{BW_{adult}} \right) \quad (6)$$

The following equation was used to assess the carcinogenic risk (CR) of metals:

$$\text{CR} = \text{LADD x SF} \quad (7)$$

where, *LADD* is the lifetime daily dose of exposure to metals in road dust, *SF* is the carcinogenic slope factor (SF for inhalation of Ni = 0.84 mg/kg/day [11].

Carcinogenic risk is assessed using the index provided in Table 4.

Table 4. Carcinogenic risk value description [4, 7, 14]

CARCINOGENIC RISK (CR) VALUE	MEANING
$< 10^{-6}$	Risk is negligible
10^{-6} to 10^{-4}	Acceptable or tolerable risk for regulatory purposes and desirable recommendation.
$>10^{-4}$	Likely harmful to humans

It should be noted that carcinogenic risk was assessed through the inhalation exposure model only due to the unavailability of cancer slope factor for other routes of exposure for specific carcinogenic metals.

2.3. Statistical Analysis

Basic descriptive statistical analysis, such as mean, standard deviation, median, maximum, and minimum, was conducted for metal concentrations in road dust collected from five (5) sampling areas near mining sites in Carrascal, Surigao del Sur. T-test for two samples assuming equal variance was done using Microsoft Excel to determine if there is significant difference between metal concentrations in this study and levels reported in other cities. Pearson's correlation was calculated to see the relationship between distance from mining sites to sampling stations (road and direct distance) and metal concentrations measured in this study.

3. Results and Discussions

Metal Concentrations in this Study Compared to Levels Reported in Other Cities

Total Cr, Ni, Mn, and Zn concentrations detected in road dust samples collected around human settlements nearby mining areas in Carrascal, Surigao del Sur are presented in Table 5.

Study results were compared with those reported by other studies, which mostly measured metal concentrations in urban road dust, with traffic and industrial processes as predominant sources (Table 6). Average concentrations of Cr, Mn, Ni, and Zn are reported by these studies ranged from 26 – 218, 258 - 2,256, 4 – 41, and 23 – 496 mg/kg, respectively. In this study, measured average concentrations were significantly higher than these values, with p-values less than 0.05 ($p = 0.002$ for Cr, $p = 0.0001$ for Mn, $p = 9.29E-06$ for Ni, and $p = 0.04$ for Zn).

High metallic concentrations are somehow expected, as the study area is rich in naturally-occurring metals. Aggravating the situation are mining-related activities that could contribute to greater dust dispersion and resuspension. Results suggest that potential health risks to mine workers and locals from their daily exposure to dust could be greater and deserve as much attention as in urban environments.

Table 5. Metal concentration (mg/kg dw) in road dust samples collected from areas near mining sites in Carrascal, Surigao del Sur (n = 15).

METAL	MEAN	STANDARD DEVIATION	STANDARD ERROR	RANGE (MIN.-MAX)
Cr	15,668	4,062	1,049	7,282-21,352
Ni	14,810	2,121	548	10,406-18,986
Mn	7,054	1,225	316	4,158-8,838
Zn	684	201	52	457-1,070

Table 6. Average metal concentration in road dust measured in this study and in other countries.

CITY, COUNTRY	CONCENTRATION (mg/kg)				SOURCE OF POLLUTION	REFERENCES
	Cr	Mn	Ni	Zn		
Carrascal, Surigao del Sur, Philippines	15,668	7,054	14,810	684	Mining related dust	This study
Shijiazhuang, China	132	-	41	496	Urban road dust – traffic and industry related source	Cai and Li (2019)
Dhaka, Bangladesh	144	262	37	239	Urban road dust – traffic and industry related source	Rahman et al. (2019)
Yazd, Iran	-	2,256	4	23	Urban road dust – traffic and industry related source	Esfandiari et al. (2019)
Lublin, E Poland	86	-	17	241	Urban road dust – traffic related source	Zglobicki et al. (2018)
Yuanping, China	149	-	27	348	Urban dust – industry related source	Li and Liao (2018)
Baotou, China	154	504	25	50	Urban dust – industry related source	Han et al. (2017)
Beijing, China	85	-	25	222	Urban road dust – traffic related source	Wei et al. (2015)
Islamabad, Pakistan	125	433	23	166	Urban road dust – traffic related source	Faiz et al. (2012)
Luanda, Angola	26	258	10	317	Urban dust – industry related source	Ferreira-Baptisa and De Miguel (2005)
Palermo, Italy	218	302	14	207	Urban road dust – traffic related source	Varrica et al. (2003)
Ottawa, Canada	43	432	15	113	Urban dust – human settlements-related source	Rasmussen et al. (2001)

Table 7. Correlation between road distance from mining sites to sampling stations and metal concentrations measured near mining sites in Carrascal, Surigao del Sur.

	Mn conc.	Cr conc.	Ni conc.	Zn conc.
Road distance from the mining sites to sampling stations (km)	-0.64	-0.03	-0.37	0.38
Direct distance from the mining sites to sampling stations (km)	-0.58	-0.18	-0.30	0.29

Possible Role of Sampling Site Proximity to Mining Operations and Vehicle Movement on Road Dust Metal Concentrations

In order to see patterns in the potential effect of distance from mining sites and movement of vehicles on road dust concentration of heavy metals, data was assessed against road distance and direct distance to mining sites (Table 7; Figures 2 and 3).

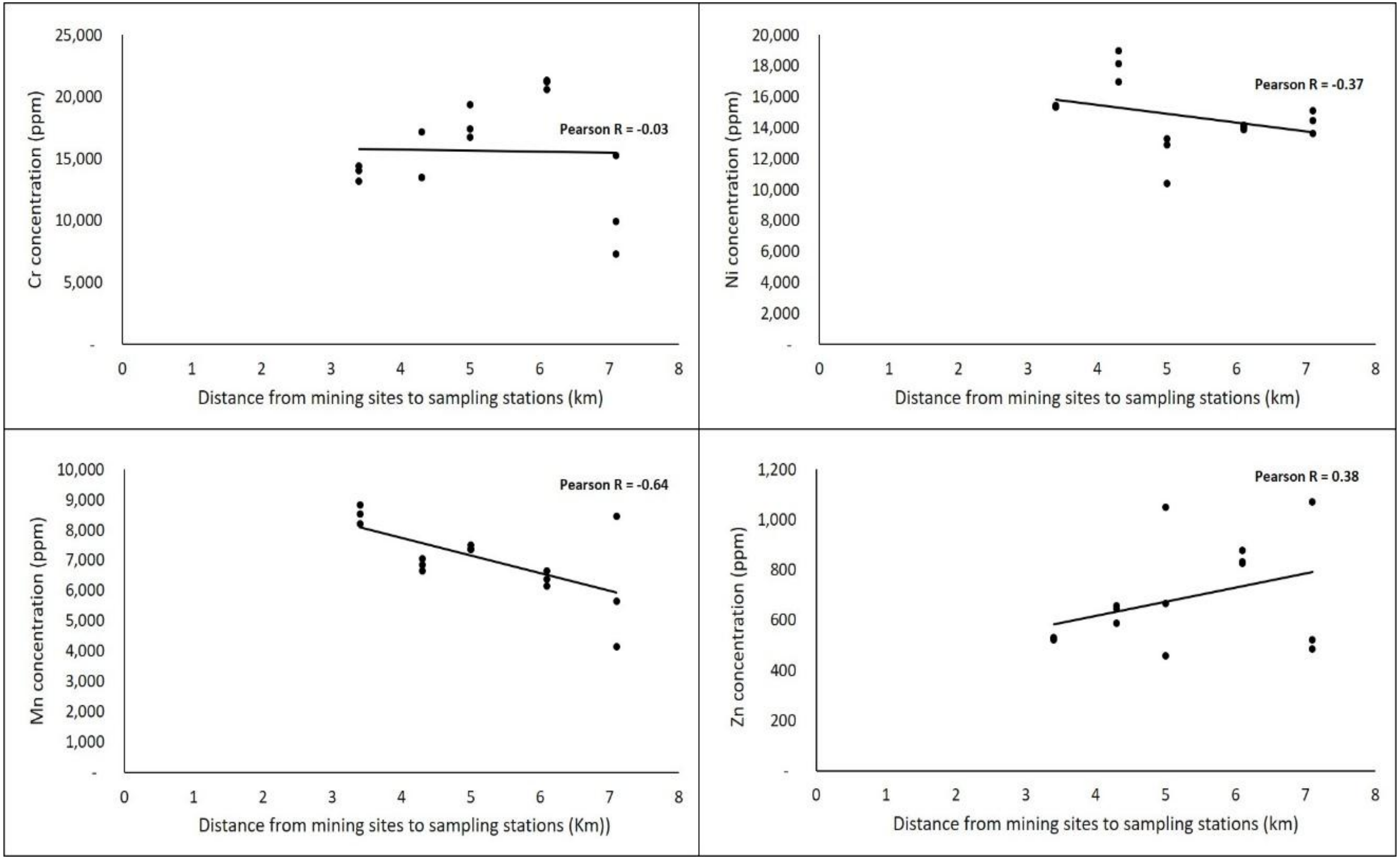

Figure 2. Relationship between road distance from mining sites to sampling stations and metal concentrations measured near mining sites in Carrascal, Surigao del Sur.

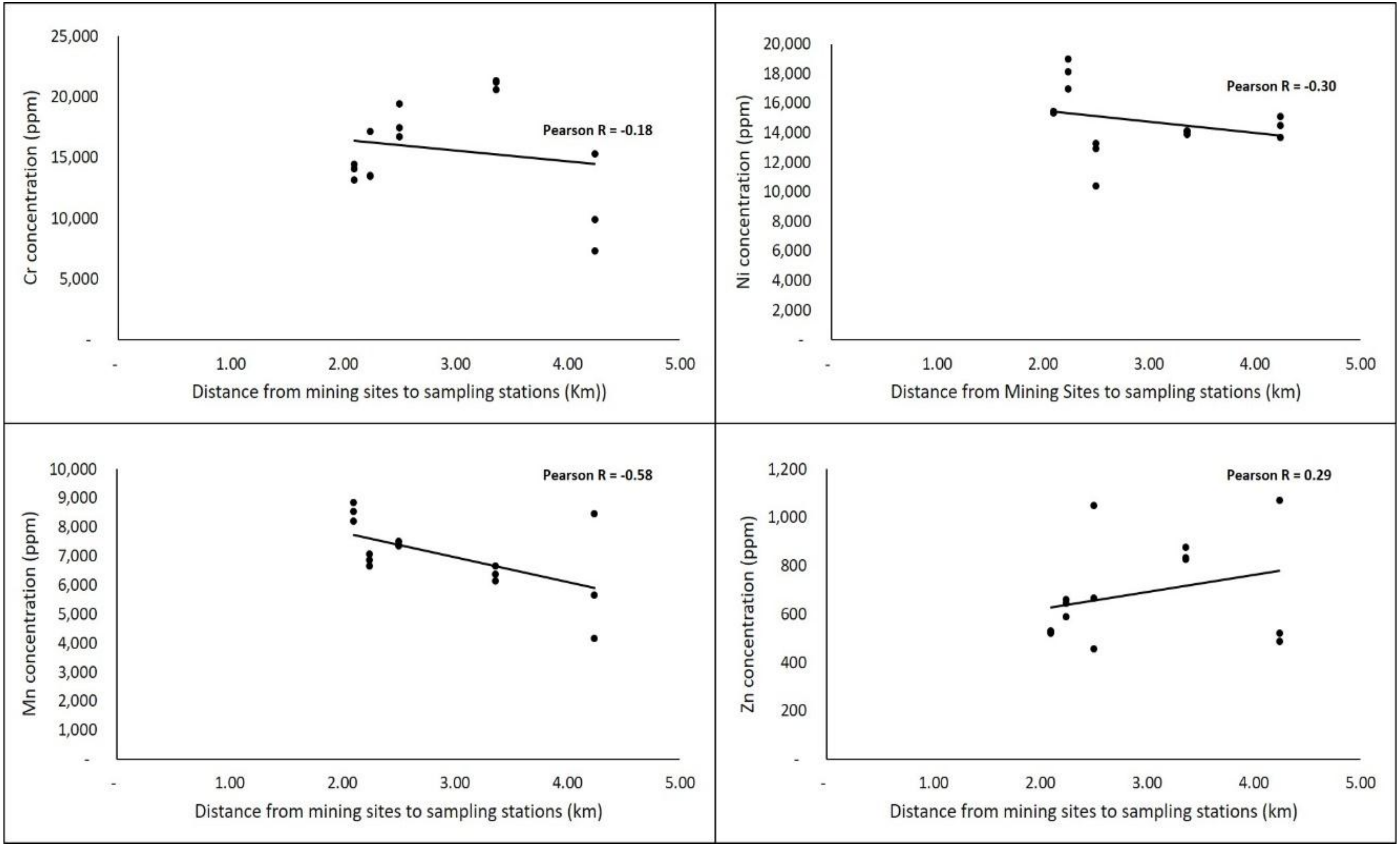

Figure 3. Relationship between direct distance of mining sites to sampling stations and metal concentrations measured near mining sites in Carrascal, Surigao del Sur

Weak negative relationships were overserved between distance from mining areas (for both road and direct distance) and concentrations of metals Cr, Ni, and Mn (R value between metal concentration and road/direct distance of -0.03/-0.18, -0.37/-0.30, and -0.64/-0.58 for Cr, Ni, and Mn, respectively). This concurs with a U.S geological survey that reported that concentrations of metals were noticeably higher near the point source and decreased immediately as distance from source increases [15].

Meanwhile, a reverse trend was observed for Zn (R value between Zn concentration and road/direct distance of 0.38/0.29), suggesting that factors other than proximity may come into play in the increased metal concentration in a certain area. These may include geogenic and anthropogenic influences in the area, such as movement of vehicles [16, 17].

Data were also grouped according to site to identify other potential factors that may affect metal concentrations (Figure 4). However, the distribution of metals based on sampling location was highly variable or there is no observable trend. This further indicates that the distribution of metal concentrations may not be solely determined by its distance to active mining sites. Other factors, such as elevation, rainfall, effect of wind on dust dispersal, natural soil composition in the area, mineral loading and unloading, or movement of vehicles loaded with mining ores, may have potential roles in determining whether an area should be expected to have elevated metal concentrations or not [18, 19].

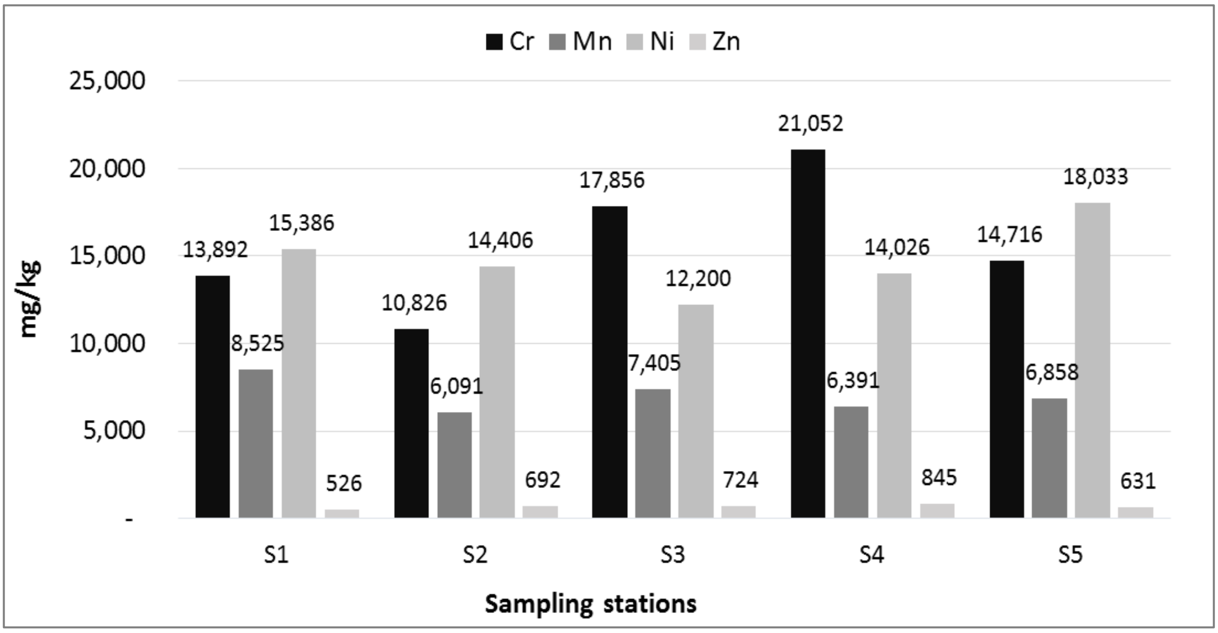

Figure 4. Individual metal concentration per sampling stations near mining sites in Carrascal, Surigao del Sur.

Table 8. Average metal concentration in road dust measured in this study and the national regulations and guidelines applicable to Cr, Ni, Mn. and Zn.

METAL CONC. IN ROAD DUST (This study)	NATIONAL REGULATION AND GUIDELINES		
	Description	**Information**	**Reference**
Nickel (14,810 ppm)	TLV (8-hr TWA)	1.5 mg/m^3 or 0.0015 ppm	ACGIH, 2003
	PEL (8-hr TWA) for general industry	1.0 mg/m^3 or 0.001 ppm	OSHA 2003a 29 CFR 1910.1000, Table Z-1
	REL (10-hr TWA)	0.015 mg/m^3 or 0.000015 ppm	NIOSH 2003a, 2003b
	IDLH	10 mg/m^3 or 0.01 ppm	
	EPA RfD	0.02 ppm/day	IRIS, 2005
Chromium (15,668 ppm)	TLV (8-hour TWA)	0.5 mg/m^3 or 0.0005 ppm	ACGIH, 2007
	REL (8-hour TWA)	0.5 mg/m^3 or 0.0005 ppm	NIOSH, 2005
	IDLH	250 mg/m^3 or 0.25 ppm	
Zinc (684 ppm)	EPA RfD	0.3 ppm/day	IRIS 2003
Manganese (7,054 ppm)	TLV (8-hour TWA)	0.2 mg/m^3 or 0.0002 ppm	ACGIH, 2007
	PEL (8-hour TWA) for general industry (ceiling limit)	5 mg/m^3 or 0.005 ppm	OSHA 2007c 29 CFR 1910.1000, Table Z-2
	REL (10-hour TWA)	1 mg/m^3 or 0.001 ppm	NIOSH, 2005
	STEL (15-minute TWA)	3 mg/m^3 or 0.003ppm	
	IDLH	500 mg/m^3 or 0.5 ppm	
	EPA RfC	5x10^{-5} mg /m^3 or 5x10^{-8} ppm	IRIS, 2011
	EPA RfD	0.14 ppm/day	

ACGIH = American Conference of Governmental and Industrial Hygienists; EPA = Environmental Protection Agency; IDLH = Immediately Dangerous to Life and Health; IRIS = Integrated Risk Information System; NIOSH = National Institute for Occupational Safety and Health; OSHA = Occupational Safety and Health Administration; PEL = Permissible Exposure Limit; REL = Recommended Exposure Limit; RfC = Inhalation Reference Concentration ; RfD = Reference Dose; STEL = Short-Term Exposure; TLV = Threshold Limit Values; TWA = Time-weighted Average.

Comparison of Metal Concentrations in this Study to Regulation and Guidelines

To determine how high the measured metal concentrations in road dust are against current regulations and guidelines set by various agencies and published by the Agency for Toxic Substances a Diseases Registry (ATSDR), a comparison was made and presented in Table 8. The limits provide an estimate of human daily exposure that is likely to have appreciable risk of adverse health effects over a specified period of time [20, 21].

Nickel concentrations in road dust from Carrascal were several magnitudes higher than all limits set by several organizations (i.e., more than a thousand times higher than the immediately dangerous to life and health (IDLH) concentration set by the National Institute for Occupational Safety and Health (NIOSH)). Such road dust concentration is alarming, considering that Ni compounds are identified as group 1 substances (human carcinogens) and Ni (metallic) is identified as group 2B substance (possibly carcinogenic to humans) [22]. Total chromium concentrations in the road dust from Carrascal were also several magnitudes higher than all limits set by several organizations (i.e., more than a thousand times higher than the threshold limit value concentration set by ACGIH). Meanwhile, average Zn concentrations were more than a hundred times higher than the reference daily dose limit set by EPA. Mn concentrations were also several magnitudes higher than all limits set by several organizations (i.e., more than a thousand times higher than the IDLH concentration set by NIOSH).

In general, Cr, Ni, Mn, and Zn concentrations in sample road dust from Carrascal greatly exceeded the recommended levels, indicating potential high risk for developing adverse health effects over a period of time. Therefore, it is urgent for mining companies to develop management controls in order to reduce dust generated due to vehicle and material movement. The local government should likewise develop guidelines for the reduction of dust exposure to residents in nearby human settlements.

Historical Morbidity and Mortality Cases in the Study Area

The morbidity and mortality cases in Carrascal, Surigao del Sur were assessed to determine the annual percentage of possible dust-related cases and the annual percentage rise/drop of these cases.

The Rural Health Unit (RHU) of Carrascal has recorded some leading health problems (morbidity) in the local community that can be associated to human exposure to dust (Table 9).

Based on mineral production records obtained from Mines and Geosciences Bureau-Caraga, 2012, 2013, and 2014 were the peak years of mining operation in the area. From 2010 (the onset of mining activity) to the succeeding years of mining operations, a significant rise in possibly dust-related human health problems was observed (p-value 0.000). Table 9 shows that 2012 had the highest percentage rise (30%) in possibly dust-related morbidity cases. Furthermore, it can be observed that the leading morbidity cause was upper respiratory tract infection (URTI), with the highest number of cases recorded in 2012 (1,643), 2013 (1,632), and 2014 (1,561). In decreasing order, URTI was followed by pneumonia, acute bronchitis, flu, skin diseases, and impetigo. Noticeably, a rise in pneumonia cases from 2012 to 2014 was noted. Mining production in the area has decreased since 2015, along with the number of needed mining workers and subcontractors, which may explain the decline in dust-related morbidity cases after 2014. In addition, the paving of unpaved major roads connected to mining roads by the mining companies after 2014 may have contributed to the reduction of dust generation.

Table 9. Morbidity cases in Carrascal, Surigao del Sur.

MORBIDITY	YEAR							
	2010	2011	2012	2013	2014	2015	2016	2017
Upper Respiratory Tract Infection (URTI)	367	723	1643	1632	1561	734	735	203
Pneumonia	414	445	292	463	652	437	428	110
Acute Bronchitis	263	222	108	146	134	242	212	62
Flu	50	37	0	142	0	130	0	62
Skin Diseases	0	0	0	0	96	0	0	0
Impetigo	0	0	0	0	0	0	101	0
Total possible dust-related cases per year	1,094	1,427	2,043	2,383	2,443	1,543	1,476	437
Total morbidity cases per year	1,607	2,503	2,976	4,115	3,782	2,659	2,365	868
Percentage of possible dust-related cases per year[a]	68%	57%	69%	58%	65%	58%	62%	50%
Percentage rise/drop of possible dust-related cases per year[b]		23%	30%	14%	2%	-58%	-5%	-238

[a] Percentage of possible dust-related cases per year = Total possible dust-related cases per year/ Total morbidity cases per year x 100.

[b] % Decrease = orig number – new number / orig number x 100. % Increase = new number – orig number / orig number * 100

Table 10. Mortality cases in Carrascal, Surigao del Sur

MORTALITY	YEAR				
	2013	2014	2015	2016	2017
Pneumonia	10	15	12	17	15
Chronic Obstructive Pulmonary Disease	6	5	5	5	1
Lung Cancer	1	0	3	0	0
Status Asthmaticus	0	0	4	0	0
Asthma	0	0	0	0	1
Pulmonary Tuberculosis	0	0	0	0	1
Total possible dust-related cases per year	17	20	24	22	18
Total mortality cases per year	36	64	53	43	33
Percentage of dust-related cases per year[a]	46%	31%	45%	51%	55%
Percentage rise/drop of possible dust-related cases per year[b]		15%	17%	-9%	-22%

[a] Percentage of possible dust-related cases per year = Total possible dust-related cases per year/ Total mortality cases per year x 100.

[b] % Decrease = orig number – new number / orig number x 100. % Increase = new number – orig number / orig number * 100

RHU-Carrascal also recorded mortality cases that could be possibly associated with dust exposure (Table 10). Pneumonia was identified as the leading potentially dust-related cause of death from 2013 to 2017 (RHU-Carrascal records, 2013 to 2017). This was followed by chronic obstructive pulmonary disease, lung cancer, status asthmaticus, asthma, and pulmonary tuberculosis. A significant increase (p-value 0.02) in mortality cases was observed in the area from 2013 to 2015. The highest percentage rise in mortality potentially related to dust exposure was recorded in 2015 at 17%.

Potential Non-carcinogenic and Carcinogenic Health Risk Assessment

In order to assess the potential health risks from daily exposure of children and adults to the measured levels of Cr, Ni, Mn, and Zn in road dust near mining areas, the risks of developing non-carcinogenic and carcinogenic health effects were calculated.

Non-carcinogenic Health Risk Assessment

Non-carcinogenic health risk assessment was conducted to determine the possibility of developing non-cancer health effects as a consequence of daily exposure to heavy metals in the study area. The average concentrations of Cr, Ni, Mn, and Zn measured in this study were used to calculate the hazard quotient and hazard index for exposure assessment in children and adults using the formula developed by USEPA [8].

Looking at the HQ values, exposure to Cr in children is highest through ingestion, followed by dermal contact. Meanwhile, exposure to Cr is highest among adults through dermal contact, followed by ingestion (Table 11). Inhalation route of exposure had the lowest HQ values for all metals in both children and adults.

It is notable that metal exposure among children via the ingestion route is higher compared to that of adults (HQ_{ing} values ranging from 0.0152 to 34.9 in children versus 0.00163 to 3.74 in adults). This trend could be due to a more frequent intentional and unintentional hand-to-mouth or object-to-mouth ingestion of soil and dust among children than adults, such as eating of food or putting object dropped on dusty ground into the mouth [7, 23] These activities by children may be more frequent compared to the possible exposure in adults via ingestion, such as not washing hands before meals.

Adults showed higher exposure to Cr through dermal contact (HQ_{derm} ranging from 3.26E-04 to 7.46 in adults versus 2.13E-04 to 4.89 in chidlren). However, exposure to Ni and Zn in adults remained higher via the ingestion route of exposure, while there was not much difference when it comes to exposure to Mn via ingestion and dermal contact. According to USEPA [23], the dose of dermal exposure to metals varies according to the surface area of the skin. Thus, adults may have higher total surface area of skin exposed to suspended dust compared to children. Exposure of both children and adults to most metal via the inhalation route remained the lowest. A similar trend of high HQ values via the ingestion route, followed by dermal contact and inhalation was reported in other countries, such as India [24], Trinidad, West Indies [25], Metropolitan City, Nanjing, SE China [26], Urban Parks of Beijing, China [27], and Luanda, Angola [7].

HQ estimates from the three routes of exposure were added to derive the hazard index or HI value. According to USEPA [8], an HI value of less than or equal to one ($HI \leq 1$) indicates that exposure to metals will not likely result in adverse non-cancer health effects, while an HI value greater than one ($HI > 1$) indicates the possibility for occurrence of non-carcinogenic effects. Calculated HI values in this study are presented in Table 11. The same information is presented in Figure 5 for better visual presentation (red line indicates the border line value of 1).

Table 11. Hazard estimation for children and adults from exposure to heavy metals

ELEMENT	CHILDREN				ADULT			
	HQ_{ing}[a]	HQ_{inh}[b]	HQ_{derm}[c]	HI[d]	HQ_{ing}	HQ_{inh}	HQ_{derm}	HI
Cr	34.9	0.0103	4.89	39.1	3.74	0.0577	7.46	11.3
Ni	4.95	0.000135	0.000213	5.0	0.530	0.0000757	0.0784	0.609
Mn	1.00	0.0925	0.0718	1.17	0.107	0.052	0.110	0.269
Zn	0.0152	4.27E-07	0.0513	0.0155	0.00163	2.40E-07	0.000326	0.00196

[a] HQ_{ing}: Hazard quotient for ingestion

[b] HQ_{inh}: Hazard quotient for inhalation

[c] HQ_{derm}: Hazard quotient for dermal contact

[d] HI: Hazard index

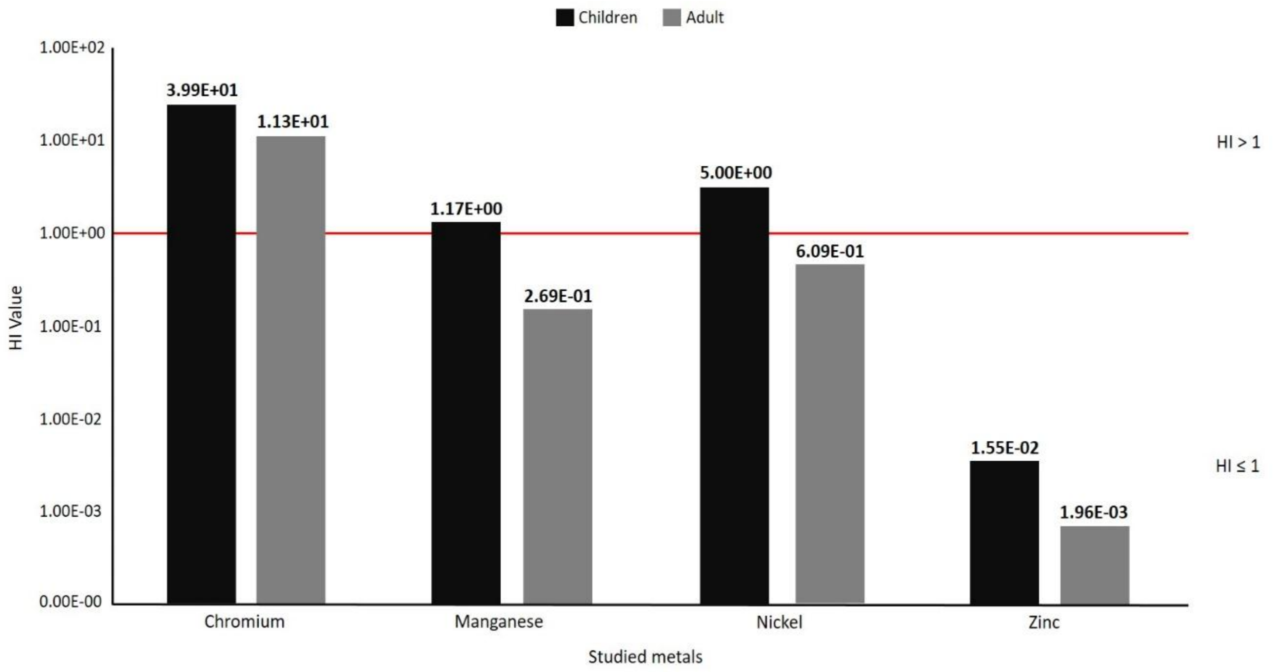

Figure 5. HI values of heavy metals in dust.

Looking at the HI values for children, Cr, Ni, and Mn all produced HI values greater than 1, while only Zn had a value less than 1. For adults, Cr yielded an HI value >1, while the rest of the metals yielded <1 HI values. These results signify greater risk among children compared to adults when exposed to heavy metals in dust of the same concentrations, probably owing to children's lower body mass. Also notable is that both children and adults are at a risk of non-carcinogenic health effects from exposure to Cr, with HI values of more than 1, suggesting that future studies on Cr speciation would be ideal to further assess its risks.

In general, study results indicate that children in Carrascal town could be at a higher risk of potential non-carcinogenic health consequences from daily exposure to metal-loaded dust compared to most adults (depending on occupation). Furthermore, exposure to Cr is likely to result in non-carcinogenic health risks in both children and adults.

Although exposure via ingestion and dermal contact could contribute more to HI values in both children and adults, the role of inhalation route of exposure in increasing potential human health risks from dust should not be downplayed. It should be noted that dust particles may directly enter the lungs and pause greater risks, depending on the nature of heavy metals contained in dust. Adults and older children could be more aware of proper hand and body hygiene, thus they could protect themselves from exposure to metallic concentrations in dust via ingestion and dermal contact routes. However, exposure via inhalation route may be of greater concern, as it is something not addressed or given much attention, even by adults in the study, area as most do not use masks or any other protective devices to prevent inhaling dust particles.

Carcinogenic Health Risk Assessment

Among the four heavy metals investigated in this study, metallic Ni belongs to group 2B, indicating that it is possibly carcinogenic to humans, while Ni compounds belong to group 1, which are carcinogenic to human [28]. Metallic Cr belongs to group 3, which are not classifiable by its carcinogenicity to humans, given that only Cr (VI) is carcinogenic [29]. No carcinogenicity data were available for metals Mn and Zn. Since this study only quantified total concentrations of metals, only the potential carcinogenic risk from exposure to Ni could be assessed.

In this study, the probability of developing cancer or the carcinogenic risks to humans from lifetime exposure to Ni was assessed. The assessment method involved calculating the lifetime average daily dose or LADD of an individual due to exposure to a substance and its corresponding cancer slope factor. In the model developed by USEPA [8], the LADD of exposure was multiplied with the cancer slope factor specific to the metal (0.84 for Ni) in order to determine the cancer risk value or the lifetime probability of an individual to develop cancer. The calculated cancer risk values have corresponding descriptions presented in Table 4. Results of the cancer risk assessment due to inhalation of Ni are presented in Table 12.

Table 12. Carcinogenic risk value of Ni in road dust near mining areas of Carrascal, Surigao del Sur.

ELEMENT	SF_{INH}	$LADD_{INH}$	CARCINOGENIC RISK VALUE	INDICATION
Ni	0.84	7.73E-07	6.49E-07	Cancer risk is negligible.

SF_{INH} = carcinogenic slope factor for inhalation route of exposure

LADD = lifetime average daily dose of exposure to metals in road dust

These results indicate that there is negligible cancer risk associated with concentrations of Ni ($6.49x10^{-7}$) in road dust potentially inhaled by the local community and mine workers, since values fell below 10^{-6}.

Management measures in reducing metal-loaded dust generation in areas near mining sites in Carrascal, Surigao del Sur, Philippines.

Study results showed that road dust generated by mining operations in Carrascal generated a high potential risk of developing adverse health problems over a period of time from daily exposure. Therefore, the mining companies and the national and local government are obligated to perform their duties and responsibilities to improve management control measures and develop guidelines in order to reduce dust generation in the mining area.

Based on the Philippine Mining Act of 1995 (RA 7942) and Clean Air Act of 1999 (RA 7942), protecting the environment from the negative impacts of mining activities must be a shared responsibility of mining companies and the local and national government. Mining companies must always be guided by current best practices in environmental management in order to reduce the impacts of mining while efficiently and effectively protecting the environment. In addition, mining activities must be undertaken with due and equal regard for economic and environmental considerations, as well as for health, safety, social, and cultural concerns. The local government, acting as both a beneficiary and an active participant in mineral resource management, should ensure that relevant laws and requirements on public notices, consultations, and public participation are complied with. The national government, meanwhile, is tasked to impose on mining companies the monitoring and implementation strictures stipulated in national policies and guidelines to ensure sustainable operation.

4. Conclusions

Road dust samples collected around human settlements nearby mining areas in Carrascal, Surigao del Sur were found to contain high levels of Cr, Ni, Mn, and Zn, with average concentration values of 15,668, 14,810, 7,054, and 684 mg/kg, respectively. Analyzed concentrations of Cr, Ni, Mn and Zn were significantly higher than the average metal concentrations in urban road dust in cities around the world, which mostly originated from traffic and industrial activities. Furthermore, measured metal values in the present study greatly exceeded several regulatory and guideline thresholds set by various international agencies.

Non-carcinogenic risks assessment revealed that children could be exposed to heavy metals in dust to a greater extent through the oral route of exposure. Meanwhile, dermal contact with dust could pose greater risk to adults. Furthermore, hazard index (HI) assessment revealed that children are more at risk of developing non-cancer-related health effects from exposure to metals in dust compared to adults, probably owing to their lower body mass (Cr, Ni, and Mn had HI values greater than 1 for children, while only Cr had HI value of 1 for adults). Carcinogenic risk assessment through the inhalation route of exposure showed that concentrations of metals in road dust from Carrascal were not of great concern when it comes to cancer risk in humans. Carcinogenic risk value (or the lifetime probability of an individual to develop cancer) of $6.49x10^{-7}$ for Ni indicated that cancer risk is negligible.

Assessment of morbidity and mortality records of Carrascal, starting from the onset of mining activities toward the peak of mining operations and succeeding years, indicated an increase in the incidence of respiratory system-related cases (e.g., upper respiratory tract infection, pneumonia, acute bronchitis, flu, skin diseases, and impetigo). Furthermore, among probable dust-related causes of mortality, pneumonia was the leading cause, followed by chronic obstructive pulmonary disease, lung cancer, status asthmaticus, asthma, and pulmonary tuberculosis.

Management measures to minimize metal-loaded dust generation and dispersion in Carrascal, such as enhancement of dust suppression techniques for mining roads networks, traffic pattern optimization, and setting up maximum speed limit for all vehicular equipment passing through unpaved roads, among others, must be complied with and strictly implemented by mining companies and the national and local government in order to prevent potential adverse human health impacts on workers and residents.

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