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Cyanide and mercury level determination in small scale mining areas in the Philippines

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ABSTRACT

Free cyanide and total mercury concentrations were measured from the 196 water samples collected from the different water sources - rivers, dams, creeks, springs within the vicinity of 40 small scale mining areas in Benguet, Philippines which is the largest mining area in the northern Philippines. Ninety-eight water samples each were subjected for free cyanide and total mercury analysis. While none of the water samples exceeded the maximum limit for free cyanide, there were 90 water samples (92%) had positive readings. The average reading of all the samples of cyanide analysis was 0.047ppm (s.d. 0.0098). While 83% of the water samples were positive with mercury, only twenty-nine percent exceeded the standard. The mean reading of all the positive results of mercury was 0.064ppb (s.d.0.061).The source of mercury and cyanide residues found can be due to historic and current mining operations in the ten municipalities and also to the widespread small-mining operations in the province as a source of livelihood of the people.

KEYWORDS

Cyanide;
Mercury;
Water quality in mining areas;
Small scale mining.

INTRODUCTION

Cyanide and mercury are the two chemicals extensively used in mining to extract microscopic gold particles. These two chemicals pose serious problems to the environment as they are released freely into the streams and rivers as a result of mining[33,34].

In the process of extraction of gold particles, cyanide is used to leach out the microscopic gold particles while mercury is used and placed in pans of gold-rich ore where the element clings to the gold and sinks to the bottom, enabling retrieval of the precious metal. Then, miners apply fire to separate mercury from the gold[7,41].

Other sources of cyanide and mercury in the environment are the discharges from steel and metal factories, and plastic and fertilizer factories, erosion of natural deposits and runoff from landfills and croplands[33]. Natural sources of cyanide are from certain bacteria, fungi, and algae that metabolically produced cyanide. Cyanide may be also found in plants and some foods, such as lima beans and almonds although bulk of cyanide occurrence in environment is mainly due to metal finishing and mining industries[14]. TABLE 1 shows a brief description of the physical properties of cyanide and mercury.

It was estimated that there were about 1.6tonnes

TABLE 1 : Physical properties of cyanide and mercury

Chemical	Physical Characteristics
Cyanide	Under typical conditions in natural waters, cyanide exists predominantly as free cyanide and evaporates[24,40]. Free cyanide is a measure of the cyanide present as HCN or CN^-[20]. Hydrogen cyanide is a colourless liquid with an odour characteristic of bitter almonds and a vapour pressure of 107.6 kPa at 27.2°C. It is completely miscible in water[46]. In general, cyanide concentrations in raw water appear to be low (i.e., <0.1 mg/L)[40]. The half-life of hydrogen cyanide in the atmosphere is about 1 or 3 years[2].
Mercury	The most available information on mercury concentrations in environmental samples and biological specimens refers to total mercury. The maximum allowable concentration (MAC) of total mercury based on US EPA for consumption of water and organisms is 0.050ug/L and 0.00105 ppm for groundwater**.** Inorganic mercury is usually converted to methymercury through a microbial process. As a result, most of the mercury found at the top of the food chain is in the form of methyl mercury. The half-life of methylmercury is 90 years[3,47].

of mercury released for every ton of gold produced in the Philippines, from which higher concentrations are released into the atmosphere and lesser to soils and rivers (Eisler 2004). In Eastern Mindanao, the gold rush started in the 1980s. About 140 tonnes of mercury was released into the environment from 53 mining communities. In the study of Cortes-Maramba et. al., it was noted that about 52kg of mercury per year were used by the small scale gold processors in the southern part of the Philippines.

TABLE 2 shows the fate of cyanide and mercury in soil, water, air, and in biological systems.

In the Philippines and in other countries such as US, Africa, and Thailand, cyanide and mercury are frequently used by mining companies for leaching gold from finely ground ore[10,33].

Mining industry in the philippines

The Philippines is the fifth largest reserve of gold and copper in the world. Mining and quarrying sector which accounts for about 7.3% of the total output of the industry sector and 2.4% of the gross domestic product in 2007. In 2005, the National Statistical Coordinating Body (NSCB) pegged total Gross Value Added (GVA) to be at Php 64B (USD 1.28B) total cost of mining contributions to total exports. There are 7 mining companies registered in the year 2002. But in the year 2005, the number increased to 383 with 2,229 petitions in the country pending approval (MGB, 2002). In the second quarter of 2010, the Department of Environment and Natural Resources (DENR)-Mines and Geosciences Bureau (MGB) reported that mining industries has contributed 14% of the total Gross Domestic Product (GDP) of the country. Large scale companies contributed about 30.5 billion pesos (USD.61B) and small scale companies contributed about 19.3 billion pesos (USD.386B)[15].

Mining as the premier industry in benguet

Benguet has a land area of 2,833.0 sq.km. It has 13 municipalities namely La Trinidad, Sablan, Bokod, Bakun, Tuba, Kabayan, Itogon, Mankayan, Kapangan, Tublay, Atok, Bugias, and Kibungan. Its capital town is La Trinidad. The total population of the province is 330, 129[31].

Benguet is the country's leading gold producer. Approximately 6,227,565 metric tons of primary gold ore and 897,551,435 metric tons of primary copper are in Benguet. Small-scale mining has been the source of livelihood of the rural communities. However, about 66% of the region's total land area is covered by the top existing large mining companies of the country: Philex Mining in Tuba and Lepanto Mining Corporation in Mankayan. The mining firms contribute to about 98% of the province's export revenue[31].

Small-scale mining is a single unit mining operation having an annual production of not more than 50,000 metric tons of ore and satisfying the following requisites- the work is artisanal, either open cast or shallow

TABLE 2 : Environmental fate of cyanide and mercury

Chemical	Environmental Fate
Cyanide	Cyanide in water does not build up in the bodies of fish. There are no reports of cyanide biomagnification or cycling in living organisms, probably owing to its rapid detoxification[23]. Cyanide ions are not strongly adsorbed or retained on soils, and leaching into the surrounding ground water will probably occur. Cyanide seldom remains biologically available in soils because it is either complexed by trace metals, metabolized by various microorganisms, or lost through volatilization[23]. Cyanides usually do not seep into underground water. However, cyanide has been detected in underground waters of a few landfills and industrial waste disposal sites. In this case, cyanide becomes toxic to soil microorganisms. Because these microorganisms can no longer change cyanide to other chemical forms, cyanide is able to pass through soil into underground water[23].
Mercury	Mercury naturally volatilizes from soil and surface waters and enters the atmosphere. Most atmospherically deposited mercury will re-volatilize or adsorb to organic material in the soil. As a result, only a very small amount of mercury is transported to groundwater[1]. Mercury usually settles into rivers, lakes or oceans, where certain microorganisms and abiotic reactions convert it to methyl mercury[1] . Methyl mercury accumulates in biological tissue more quickly than inorganic mercury. Methyl mercury is then biomagnified in predatory fish such as swordfish, tuna, king mackerel and shark as well as in some types of shellfish. Mercury is strongly bound to soil and is attached predominantly to soil organic matter. Therefore, the mobility of mercury and compounds in soil is minimal even in soils contaminated by mercury fungicides. The probability of groundwater contamination with mercury through soil leaching appears unlikely; however, the mobility of mercury in soils may be enhanced by leachates from municipal landfills.

underground mining, without the use of sophisticated mining equipment; minimal investment on infrastructures and processing plant; heavy reliance on manual labor; and owned, managed or controlled by an individual or entity qualified under existing mining laws, rules and regulations (RA 7076). Mining practices of small-scale miners are mostly open-cast or quarrying operations[31].

In Benguet, there are more than 20, 000 small scale miners[9].The Ibaloy and Kankanaey people of Benguet continue to practice traditional small-scale mining till today. Men, women, children and the elderly each have a role to play in the extraction and processing of the ore. They extract only enough gold to meet their basic necessities and receive their share of the gold based on an equitable sharing system. However, as communities are deprived of their land and resources, these traditional small-scale mining methods and positive values are now under threat of vanishing[11].

METHODOLOGY

Study area and sampling

The study area consisted of small scale mining areas in Benguet. There was a total of 196 water samples located near 40 small scale mining. The water samples were taken from the various sources such as spring, river, creek, canal, irrigation, household, dam, and domestic water within the identified farm locations. Two samples/replicates of water samples were taken from each farm. The samples were placed in an icebox, and delivered to the laboratory within 24 hours.

This study was approved by the Research Ethics Board of the University of the Philippines Manila.

Sample analysis and quality control

Atomic Absorption Spectroscopy was used in ana-

TABLE 3 : Sample collection and analytical method

Collection Method Parameters	**Samples of Soil and Water**
Location of samples	Farms in the ten municipalities of the province of Benguet.
Number of samples	196 water samples; 98 samples for cyanide and 98 samples for mercury
Collection method	2 liters of water were place in an icebox and delivered within 24 hours to laboratory for analysis. The water samples were collected using surface grab sampling. The samples were stored in a freezer of less than 20 degree Celsius temperature.
Analyzing method	Atomic Absorption Spectroscopy was used in analyzing cyanide and mercury residue in the water samples. If light of just the right wavelength impinges on a free, ground state atom, the atom may absorb the light as it enters an excited state in a process known as atomic absorption. Atomic absorption measures the amount of light at the resonant wavelength which is absorbed as it passes through a cloud of atoms. As the number of atoms in the light path increases, the amount of light absorbed increases in a predictable way. By measuring the amount of light absorbed, a quantitative determination of the amount of analyte element present can be made. The use of special light sources and careful selection of wavelength allow the specific quantitative determination of individual elements in the presence of others.The atom cloud required for atomic absorption measurements is produced by supplying enough thermal energy to the sample to dissociate the chemical compounds into free atoms. Aspirating a solution of the sample into a flame aligned in the light beam serves this purpose. Under the proper flame conditions, most of the atoms will remain in the ground state form and are capable of absorbing light at the analytical wavelength from a source lamp.
The pH of the samples are checked by the contractor at the time of receipt. He/she shall note if the pH is less than or equal to 2 for metal in a sample receipt log. In case in which the metal samples are not properly preserved, the contractor will adjust the pH, allowing it to reach the equilibrium prior to digestion and shall note it in the Sample Delivery Group (SDG) Narrative. For cyanide samples, the pH should be noted if it is greater than or equal to 12. The pH of a cyanide sample does not need to be adjusted. Though, if it is <12, the Sample Management Office should be informed for further instructions before the analysis[43].	
Analytical Method for Mercury	The analytical method used for total mercury is absorption spectrometry (dithizone calorimetry), neutron activation analysis, and cold vapor atomic absorption spectrometrty. The water samples collected subjected for mercury analysis are brought to the laboratory and filtered using 0.45 μm membrane filter. See Figure 1 for the simplified procedural techniques used in determination of total mercury in water[42].
Analytical Method for Cyanide	The analytical method used for free cyanide determination in water is silver nitrate titration. First, silver ions are added to the solution to form silver cyanide complex. The endpoint of the titration is indicated with the excess silver ions. For accurate determination of cyanide concentration, a normalized silver nitrate solution is dosed with a manual or automatic burette. Potassium iodide or rhodanine can be used for endpoint determination. These indicators change color upon the appearance of free silver ions. The first color change should be noted as the endpoint indication[13].

lyzing cyanide and mercury residue in the water samples. The cyanide and mercury absorb ultraviolet light when they are excited by heat. Each metal has a characteristic wavelength that will be absorbed. The AAS instru-

Sample, 2 L (2 L separatory funnel)

Add 10 ml of 20N H_2SO_4 and mix.

Add 5 ml of 0.5% $KMnO_4$ and mix.

Let stand for 5 min.

Add 20 ml of 10 N NaOH and mix to neutralize

Add 5 ml of 10% $NH_2OH{\cdot}HCL$ solution, mix and allow to stand for 20 min.

Add 5 ml of 10% of EDTA solution and mix.

Add 10 ml of purified 0.01% dithizone-toluene and vigorously shake for 1 min.

Allow to stand for at least 1hr.

Organic phase (10-ml conical centrifuge tube) **Aqueous phase**

(When an emulsion is formed, add 0.5 g of Na_2SO_4 and shake).

Centrifuge 1,200rpm for 3 min.

Organic phase, 7 ml (sample digestion flask)

Evaporate to dryness

Residue

Distilled water, 1 ml

$HNO_3{\cdot}HCLO_4$ (1+1), 2 ml

H_2SO_4, 5 ml

Heat at 200-230°C for 30 min.

Digested sample

Allow to cool.

Top up to 50 ml with distilled water

Test solution, a fixed volume (usually 10 ml)

10% $SNCl_2$ solution, 1 ml

***CVAAS**

*CVAAS stands for Cold Vapor Atomic Absorption Spectrometry

Figure 1 : Schematic diagram of total mercury determination in water

ment looks for a particular metal by focusing a beam of UV light at a specific wavelength through a flame and into a detector. The sample of interest is aspirated into the flame. If that metal is present in the sample, it will absorb some of the light, thus reducing its intensity. The instrument measures the change in intensity. A computer data system converts the change in intensity into an absorbance. As concentration goes up, absorbance goes up. The analyst can construct a calibration curve by running standards of various concentrations on the AAS and observing the absorbances. The computer data system will draw the curve. Then samples can be tested and measured against this curve. See TABLE 3 and Figure 1.

RESULTS

A total of 196 samples of water were collected. The maximum allowable concentration (MAC) for free cyanide, adapted from US Environmental Protection Agency (USEPA) standards is 0.2ppm. The average reading of all the samples of cyanide analysis was 0.047ppm (s.d. 0.0098). (See TABLE 4). While none of the water samples exceeded the maximum allowable limit for free cyanide, there was a total of 90 water samples (92%) that registered positive readings. See TABLES 5 and 6.

TABLE 4 : Overall mercury and cyanide mean reading of all the samples

Chemical	MAC*	# of Samples	Mean Reading of all the Samples (s.d.)
Mercury (ppb)	0.05 ppb[2]	98	0.0505 (s.d.0.0661)
Cyanide (ppm)	0.2 ppm	98	0.0047 (s.d.0.0098)

***Maximum allowable concentration**

The form of mercury considered in this study was total mercury since this form of mercury is the one that is being used for environmental samples and biological specimens. The maximum allowable concentration standard is based on USEPA standard specifically from California Toxic Rule Criteria on Human Health Criteria for Consumption of Water and Organisms. The MAC for total mercury based on USEPA is 0.05ppb. The mean reading of all the results was. 0505 ppb which was above the MAC for total mercury set at. 05 ppb. While 83% of the water samples were positive with mercury, only twenty-nine percent of the total samples of mercury exceeded this standard. The mean reading of all the positive results of mercury analysis was 0.064ppb (s.d.0.061) and the mean reading of the samples exceeded the standard was 0.1186ppb (0.069). See TABLES 5 and 6.

TABLE 5 : Overall mercury and cyanide mean reading of positive results

Chemical	MAC*	# of Samples	# of Positives	%	Mean Reading Positives (s.d.)
Mercury (ppb)	0.05 ppb	98	81	83	0.064 (s.d.0.061)
Cyanide (ppm)	0.2 ppm	98	90	92	0.0054 (s.d.0.010)

***Maximum allowable concentration**

TABLE 6 : Overall mercury and cyanide mean reading of exceeded residues

Chemical	MAC*	# of Samples	# of Exceeded	%	Mean Reading of Exceeded (s.d.)
Mercury (ppb)	0.05 ppb	98	28	29	0.1186 (s.d.0.069)
Cyanide (ppm)	0.2ppm	98	5	5	0.04 (s.d.0.03)

***Maximum allowable concentration**

DISCUSSION

Cyanide and mercury groundwater contamination

There were 98 samples found to be positive with cyanide residues. However, all were within the limits set for cyanide (as free cyanides), based on USEPA standards at 0.2 ppm. Twenty eight percent of the samples subjected for mercury analysis exceeded the USEPA maximum mercury level at 0.05ppb.

The significant amount of cyanide residues found in the areas samples can may be due to the mining operations in the area. Studies showed that groundwater contamination of cyanide can be due to improper waste disposal of mine tailings and undisciplined act of mining companies. Toxic mine tailings are usually impounded in tailings dams. However, when pressure in the tailings dams builds up, especially during times of heavy rainfall, the mining companies drain their tailings dams of water and the tailings eventually find their way out, polluting the water and silting up the rivers and adjacent lands. In addition, the concentration of cyanide in rural areas is dependent on seasonal runoff from mine tailings[23].

The elevated mercury levels found in the araescan be due to past mining and chemical spills, or from improper disposal of materials that contain mercury such as fungicides. In these communities, small-scale mining is the major livelihood of the resident. The small-scale miners use mercury in mining. Thus, the mercury residues found in this study were due to the extensive use of mercury of the small scale miners. Current large-mining operations are also existing in these municipalities. Groundwater generally contains less than 2 μg/L of mercury, although areas near historic mining districts may have locally higher concentrations[27]. However, there is still little evidence to suggest that mercury enters surface or ground waters as a result of acid mine drainage, or leaching from tailings and landfills. Clays and organic matter in soils effectively reduce the quantity of mercury leached from these systems. Soil environments favoring transportation of mercury would be low in pH and contain little clay and organic matter (Irwin et.al.,1997). Therefore, for future researches, it is suggested that soil pH and other parameters should be measured to give clarification on the leaching of mine tailings to groundwater and surface water.

Even low levels of mercury were found in other municipalities, the communities should still be reminded of the danger posed by the mercury given that mercury remains for a long period, averaging a half life of 1,000 days and can be biomagnified from the bottom to the top of the food chain[16]. The concentration of mercury increases as it is passed along the food chain. Therefore, even at very small concentrations of mercury to aquatic ecosystems that are remote from point sources, mercury biomagnification can result in toxic effects in consumers at the top of the food chain, which are the human populations. Humans uptake mercury in the form of methylmercury from fishes that are contaminated with mercury[27].

Methyl mercury accumulates in biological tissue more quickly than inorganic mercury. It is the only form of mercury that accumulates appreciably in fish. Its concentrations can be 10,000 to 100,000 times greater than that of the surrounding water in fish tissue. Methymercury is soluble, mobile, and quickly enters the aquatic food chain. The conversion of inorganic mercury to methyl mercury involves a certain type of bacteria. These are the bacteria that process sulfate (SO_4=) in the environment that take up mercury in its inorganic form and convert it to methylmercury through metabolic processes. These methylmercury-containing bacteria will eventually be consumed by the next higher level in the food chain, or the bacteria may excrete the methylmercury to the water where it can quickly adsorb to plankton. Because animals accumulate methylmercury faster than they eliminate it, animals consume higher concentrations of mercury at each successive level of the food chain. Small environmental concentrations of methy-mercury can thus readily accumulate to potentially harmful concentrations in fish, fish-eating wildlife and people[27].

In Japan, cases of methymercury posioning happened in Minamata Bay was documented and showed severe illnesses and health problems among those who were affected. Adults complained reductions in motor skills and dulled senses of touch, taste, and sight. Unborn children were at greatest risk from low-level exposure to methylmercury[25,27].

Health effect of cyanide and mercury to humans

In one community in the target site, it was shown that residents who inhaled chemical fumes emanating from the mines, suffered from headache, dizziness, cough, chest pain, nasal and eye irritation. Other symptoms reported were skin itchiness, rashes and diarrhea[11]. According to some studies, these symptoms are similar to poisoning symptoms of cyanide and mercury. Exposure to these chemicals can be via inhalation, oral, or dermal exposure. People who are exposed to high levels of cyanide can suffer from ulcerations, brain and heart damage or worst, coma and death. Symptoms of severe cyanide poisoning include breathing difficulties, dizziness,weight loss, blood changes, headaches, and enlargement of the thyroid gland,vomiting, nausea, weakness, confusion, lethargy, cyanosis, weak and ataxic movements, increased respiratory and heart rates, progressing to coma with respiratory depression, seizures, cardiovascular collapse, and death[10,17,33]. In addition, mercury released by mining activities has been proven to damage the kidneys, liver, brain, heart, lungs, colon and immune system. Chronic exposure to mercury may result in fatigue, weight loss,tremors, emotional changes (e.g., mood swings, irritability, nervousness, excessive shyness), insomnia, neuromuscular changes (such as weakness, muscle atrophy, twitching), headaches, disturbances in sensations, changes in nerve responses, performance deficits on tests of cognitive function, rapid breathing and behavioral and personality shifts[17,33]. Mercury is also known to induce hypersensitivity reactions such as contact dermatitis and acrodynia (pink disease).

In a study of Makalinao[27], 26 grade school children from a distant agricultural area in Mabini Bohol died within a few hours from eating some snacks prepared from cassava contaminated with cyanide. In Sibutad, Western Mindanao, a study conducted among the small-scale gold miners showed that gastrointestinal complaints among the respondents was significantly associated with elevated hair methylmercury levels. It was also found that there was an increasing incidence of elevated diastolic blood pressure with elevated hair total mercury levels ($p=0.07$)[12]. In Pantukan, Compostela Valley Province, a study was conducted among the miners engaged in the mercury amalgamation method for gold extraction. It was found in this study that mercury exposure of the miners from artisanal small-scale gold mining resulted in high blood mercury levels, poor memory, anosmia, abnormal gait and balance. In addition, soil and water samples from the river exceeding the existing World Health Organization[48].

Abandoned mercury mines have been one of the major concerns because of their significant long-term environmental problems. In the Philippines, an abandoned mercury mine located southeast of Manila dumped its mine-waste calcines (retorted ore) into nearby Honda Bay. This resulted in adverse health increasing complaints of unusual symptoms (e.g. miscarriages, tooth loss, muscle weakness, paralysis, anemia, tremors, etc.) among the residents living near the bay. Abnormal findings were also resported and these included gingivitis, mercury lines, gum bleeding and pterydium. Neurologic complaints were numbness, weakness, tremors and incoordination. Anemia and elevated liver function tests were also observed in a majority of those examined[29]. In the case of Benguet Corporation in Itogon, it can be suggested that medical assessment should be done among the residents to determine the probable effect of the mine in the area to the health conditions of the people residing the vicinity of the mine.

CONCLUSION

The study showed that 92% of the water samples were positive for free cyanide and 83% were positive for total mercury levels. None of the free cyanide exceeded the MAC while 29 percent exceeded the MAC. This calls for further investigation on cyanide and mercury contamination of the water systems in Benguet which is the largest small scale mining area in the northern Philippines. Mercury, can be biomagnified and therefore can pose serious toxic effects to humans, as the top consumer of the food chain. It is therefore suggested that future investigation should be done on mercury content of the fishes acquired from the different water system of the province. The study has shown the possible association between the prevalence of mercury and cyanide readings with current and past mining activities in the area.

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