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Assessment of Potentially Toxic Cyanide from the Gold and Copper Mine Ore Tailings of Karnataka, India

E. R. Harish
Research Scholar, Department of PG Studies in Zoology
Environmental and Molecular Toxicology Laboratory, Karnatak University, Karnataka, India
M. David
Professor, Department of PG Studies in Zoology,
Environmental and Molecular Toxicology Laboratory, Karnatak University, Karnataka, India

Abstract:
Concerns have raised that, due to its continuous use in the industrial application and through natural resources every inhabitants on the earth are continuously exposed to cyanide in everyday life. Since, the huge amount of cyanide processing ore tailings of Gold and Copper mine industries were dumped into the subsurface environment causes serious hazards to existing livelihood. The study intended with spectrophotometric estimation of cyanide through standard distillation from the gold and copper ore tailings of Karnataka, India. The cyanide content of the samples was determined with a range between 20-260 mg/kg from the different mine ore tailings samples. The highest concentrations of cyanide were observed in the ore tailings of Hatti Gold Mine Ltd. New Plot (HGML-New plot), Hatti than the ore tailings of Kennedy line dump of Kolar and Copper ore tailings of Ingaldhal. Since, simple and complex Cyanide compounds can release the extremely toxic free cyanides (hydrogen cyanide, and cyanide ion); there is a need to continuous monitoring of cyanide levels in the ore tailings of Gold Mine Industries.

Keywords: *Copper Ore tailings, Cyanide, Gold Ore tailings, Spectrophotometric, Standard distillation*

1. Introduction

Gold and Copper mining at Kolar, Hatti and Ingaldhal in the Karnataka State started dates back to the ancient times as the mining activities can be traced as far back as 2,000 years i.e., during Indus valley civilization (Gita, 2013), Pre Harappan period (Bagchi and Ghose, 1980), and Shatavahana periods (Shankar, 1989) respectively. Of these Hatti Gold Mine Industry at Hatti remains the oldest viable mine industry in Karnataka, whereas the Kolar (2000) and Ingaldhal (2005) mining had been closed due to availability of low grade ore. The long history of mining in these areas generated huge amount of cyanide contaminated ore tailings: 50 million tons at Kolar (Rao and Reddy, 2006), 1 billion tons at Hatti (Swaroop et al., 2013), 1.7 million tons at Ingaldhal (Purushotham et al., 2010). Huge amounts of this cyanide contaminated ore tailings constitute a major source of release of many trace elements, including cyanide into the environment and it can contaminate groundwater quality (Gupta and Roy 2012), atmospheric quality (ATSDR 1995), and loss of soil fertility (Faz et al. 2013). In addition to that, sewerage system in township areas, agriculture and earlier mining activities constitute a major source of release of cyanide into the environment. Several authors reported that, Highly toxic compounds of cyanide used by the international mining community to extract gold and other precious metals through milling and heap leaching through which millions of liters of alkaline water containing high concentrations of potentially toxic sodium cyanide, free cyanide and metal cyanide complexes are frequently accessible to aquatic ecosystems and wildlife, many aquatic species and birds were found dead in the immediate vicinity of gold-mining leach extraction and tailing ponds (acute LC < 1mg l^{-}), presumably as a result of contamination of cyanide in the environment (Eisler and Wiemeyer 2004; David et al., 2015).

Cyanide is a highly reactive, unstable hazardous substance, used in different applications due to its outstanding properties in industries like gold, silver and copper mining, electroplating industries (Yu et al., 2010), coal processing (Agarwal, 2005) Petrochemical plants (Behnanfard and Salariad, 2009) agricultural industries and cassava starch industries (Kharmar et al., 2015). As of today, cyanide usage equates to an annual mass of approximately 1.1 million tons (ICMC, 2013). This extremely large amount of cyanide utilized industrially per annum is indicative of the extensive commercial demand. Thus widespread use and high amount of cyanide production have potential for contamination of underground water, animals, food, environment and human lives. In nature, various organism including bacteria, fungi, algae, plants, are able to produce cyanide for defensive (Zagrobelny et al., 2008) as well as

offensive purpose (Gallagher and Manoil, 2001). However, the contamination of the environment with cyanide is mainly due to human activities (Mudder et al., 2001).
Long term deposition of this cyanide containing ore tailing can lead to accumulation, transport and biotoxicity of cyanide compounds. Brasel et al., (2006) reported that, effluents from the mining industry may contain cyanide concentration ranging from 0.3 to 216 ppm of free cyanide along with metal complex which can leads to environmental pollution. There are several reports reported that, sublethal exposures of cyanide may elicit a number of toxic effects including neurotoxicity (Soto-Blanco et al., 2008), testicular toxicity, epidemic spastic paraparesis (Shivanoor and David, 2014), pancreatic diabetes (Soto-Balanco et al., 2002), cardiotoxicity, and lactic acidosis (Bebarta et al., 2012).
Since cyanide compounds can release the extremely toxic free cyanides (HCN, CN^-), the continuous monitoring of cyanides from the ore tailings is important to rapid and precise determination of cyanides in air, waste waters, ground waters, and soils. In this present research work, effort have been made to spectrophotometric determination of the cyanide levels in Gold and Copper mine ore tailings of Karnataka State, India by using standard distillation method.

2. Materials and Methods

2.1. Soil Sampling

The Gold Ore Tailings (GOT) and Copper Ore Tailings (COT) used for this study was collected from New Plot ore tailing pond of Hutti Gold Mine Company Ltd. Hatti, Raichur District, Karnataka; Kennedy Line Dump also known as cyanide mountain, Bharath Gold Mine Ltd., Kolar Gold Fields, Kolar, Karnataka; and the ore tailings of Copper Mine Ore Ltd, Ingaldahal, Chitradurga District, Karnataka. Fig.1 and Table.1 shows the geographical distribution i.e latitude, altitude and elevation of the sampling area. Sampling was carried out from different parts of each ore tailings and samples were collected in a sterilized plastic container and transported to laboratory for further analysis.

2.2. Physico-Chemical Characteristics of the Mine Ore tailings of GOT and COT

The collected ore tailings samples were air dried, crushed to fine particle and sieved using 2 mm sieve before analysis. Particle size was determined by Hydrometer method as described by Bouyoucous (1951) and Agbenin (1995). After, silt and clay determination, the value of sand was obtained by subtracting the values of silt and clay from 100. P^H of the soil was determined using Potentiometric method as described by Brady and Weil (1990) by using Elico Ultra pH Meter (Model LI 120). Organic carbon was determined according to the wet oxidation method of Walkey and Black (1945) and its moisture content was determined by Michael (1984).

2.3. Estimation of Total Cyanide

2.3.1. Reagents Required

5M hydrochloric acid, 1M Sodium hydroxide solution, 20% Copper (II) Sulphate Solution, 5% Tin(II) chloride solution, 0.5% Chloramine T solution, Barbituric acid–pyridine solution, Phosphate Buffer solution (pH-5.4), NaCN solution.

2.3.2. Standard Sodium Cyanide Solution Preparation

Sodium cyanide (95%) (Batch No. V-1058/1; Cas No. 143-33-9) was procured from Loba Cheme Pvt Ltd. Mumbai, Maharashtra, India. A stock solution was prepared by dissolving 18.9 mg sodium cyanide (equivalent to $100mg.L^-$) in 100 ml of 0.1M NaOH solution. Standard solution was prepared by pippet 10 ml of stock solution in 100ml of 0.1M NaoH solution. Working Standard solution was prepared by pipette 0.2, 0.4, 0.6, 0.8, 1 ml respectively. The stock cyanide solution was standardized argentometrically (Attah Daniel et al., 2013).

2.3.3. Distillation Procedure

The standard distillation was performed by Tim Mansfeldt and Heidi Biernath, (2000) method with slight modification according to EPA, (2011). Fig. 2 represents the distillation apparatus used for this study, it occupies space of 1.30m×0.80m×0.20m (width×height×depth) and standing in a hood.
For distillation, 5 g soil with 100 ml water was added to a 500 ml flask. Then the flask is connected to condenser and inlet tube. The absorber is attached to the vacuum followed with addition of 20 ml of 1M sodium hydroxide solution and connected to the condenser. While digestion 10 ml of 20% $CuSO_4$, 2 ml 5% $SnCl_2$, and 20 ml of 5 M HCl were added carefully and refluxed with 100˚C for 1 h. All digestions were performed with three replicates. For Spectrophotometric determination, the absorption solution was transferred into a 100ml volumetric flask and diluted to volume and mixed thoroughly. An aliquot (1 ml) was pipette into a 50ml volumetric flask, to this 4 ml buffer (pH 5.4) and 2 ml 0.5% chloramine-T was added. After 5min, 6ml barbituric acid–pyridine reagent was added to the solution and mixed thoroughly. After 20min, the samples were measured photometrically (Secomam Anthelie Advance V2.5b) at 600nm.

3. Results and Discussion

Samples of ore tailings from the Gold and Copper mine industries were analyzed for their total cyanide content through standard distillation of an acidified air dried ore tailing soil sample followed by spectrophotometric determination using chloramines-T and pyridine-barbituric acid reagent. During distillation free cyanide as well as the weak and strong metal-complexed cyanides were

converted to HCN and it forms a pink red colored complex by the reaction of cyanide with chloramines-T and pyridine barbituric acid. Table.2 shows the some of the important physico-chemical characteristics of the ore tailings and Table.3 shows the total cyanide levels and relative standard deviation of the ore tailings samples. Fig. 3 shows the standard linear curve for Cyanide concentration against absorbance determined by Spectrophotometer, Fig. 4 shows the total cyanide levels of the samples ranged from 220 to 260 mg/kg, 50 to 60 mg/kg at GOT of Hatti and Kolar and 20 to 40 mg/kg CN^- at COT of Ingaldhal respectively. Whereas it's relative standard deviation was highest at Ingaldhal copper mine than other two mine industries. The results of the ore tailing samples showed the Hatti Gold Mine Industries (243.3±20.81 g/kg) having the highest concentration of cyanide than Bharath Gold Mines (56±5.29) and Ingaldhal copper mine ore tailing (31.39±10.26) owned by Hatti Gold Mine Comapany Ltd..
Karnataka has abundant mineral resources in India, after the extraction of mineral and elements from the mine ore, the left out solid wastes are generally dumped without being used. These solid wastes do contain unextracted and other elements, depending upon the composition of ore. Swaroop et al., (2013) reported that, every day each gold mine industries generated about 2000 tons of cyanide containing ore tailing, such huge dump, know left out at gold mine industries. This has not only acquired a vast recleanable land but also causing environmental problems.
Vidyavathi et al., (2008) reported that, the cyanide concentration in gold ore tailing of Hatti and Copper ore tailing of Ingaldhal was 0.07 mg/Kg soil and Non Detectable level respectively. David (2005) reported that soluble cyanide concentration in gold ore tailing pond of Hatti was 0.05 mg/Kg Soil (Not Published). Rao and Reddy, (2006) had reported that the pyrite mine tailings in Kennedy's line dump area of Kolar gold field are potential source of contamination as it received sulfide bearing tailings in slurry form that comprised of spent ore and process water bearing soluble alkali metal cyanide and through lechate/drainage it could release cyanide ions and acid drainage to the subsurface environment. But, there is no one is reported the cyanide level in the ore tailings of KGF till date. In the present study, it has to be reported that, 243±20.81 $mg.kg^{-1}$ CN^-, 56±5.29 $mg.kg^{-1}$ CN^-, 31.3±10.26 $mg.kg^{-1}$ of total cyanide were present at GOT of HGML New plot, GOT of Kennedy Line dump (Cyanide Mountain) and COT of Ingaldhal copper mine industries respectively.
The variation in cyanide levels from the three ore tailings probably due to pH of the tailing slurry, microbial degradation, lechate, volatilization and viability of mining industries. Acidic pH of Kennedy's Line deposit may be due to atmospheric oxidation, whereas the slightly alkalinity of Hatti, and Ingaldhal tailings were due to high concentration of the calcite mineral. Since, the GOT of Hatti tailings were recently dumped the concentration of the cyanide is high, whereas in GOT of KGF and COT tailings dumps were old dump sites, through volatilization, lechate, and microbial degradation some of the soluble, weak cyanides may be degraded (Rao and Reddy, 2006). Zagury et al. (2004) and Rao and Reddy (2006) reported that, the availability and fate of cyanide in aged and freshly deposited gold tailings from two mining sites in Quebec Canada and he conclude that freshly deposited tailings having the high concentration of cyanide than age tailings, the present reports also shows the decreased level of cyanide in aged tailings than fresh tailings dump.
According to National Environmental Board (2004) and Environmental Protection Agency-EPA (2012), the acceptable limit of cyanide level in the agricultural soil is about 40 mg CN^- equivalent/Kg soil, 11 mg.kg CN^- for inhabitant sites and 55.00 $mg.Kg^{-1}$ for other industrial purposes. The maximum allowable CN^-concentrations in uncontaminated soils used as fill material at regulated fill operations was fixed to 40 mg CN^- equivalent/Kg soil according to methods described in 35 IAC 1100.S (Cavanagh, 2006). Anonym, 1998 estimated that, the soil concentration associated with the allowable one-time absorbed dose is the benchmark for the available hazard identification and dose-response information for the available cyanides was 100 mg/kg soil. Ogundola and Liasu, 2007 reported that, at high concentration of cyanide becomes toxic to soil microorganism which, can no longer change cyanide to other chemical forms, also suggested that the ability of cyanide to remain recalcitrant in the environment for long time without being degraded. It is therefore possible to observe the high levels of cyanide in the ore tailings of GOT and COT. The CN^- levels in the ore tailings soil were found to be higher than the recommended levels of 11.00 $mg.Kg^{-1}$ for soil for habitat and agriculture and 55.00mg/kg for other purposes.

4. Conclusion

Cyanide compounds released from the various mining industries, release the extremely toxic free cyanides (hydrogen cyanide, HCN, and cyanide ion, CN^-), there is a need to continuous monitoring of cyanide levels in the ore tailing samples and the introduction of legislation to seriously limit the discharge of cyanide to the environment in most cyanide producing regions of the world, and industries are adopt cyanide recovery and reuse as a alternative.

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ANNEXURE

Sl.No	Area	Location	Lattitude	Longitude	Elevation
1.	Hatti, Raichur (Dt), Karnataka, India	North Eastern Karnataka	16˚20'45" N.	76°64'62" E	1312
2	Kolar Gold Field, Kolar (Dt), Karnataka, India	South Eastern Karnataka	12º 95'77" N.	78º26'53" E.	3195
3.	Ingaldhal, Chitradurga (Dt), Karnatak, India	Central Karnataka	14° 17'97" N,	76°43'99" E.	2411

Table 1: Geography of Mine ore tailings dump (Sampling sites), Karnataka

Parameters	Units	HGML-NP	KGF-CNM	COT-ING
pH		8.116	3.48	8.021
EC	dS/m	1.623	5.356	1.21
OC	%	0.13	0.19	0.16
WHC	%	23.33	24.14	29.38
Sand	%	81.35	69.45	77.38
Slit	%	9.76	28.96	13.58
Clay	%	6.54	0	9.87

Table 2: Physico-chemical Characterstics of Mine Ore tailings of Hatti, Kolar, Copper Mine tailing dumps
EC- Electrical Conductivity; OC- Organic carbon; WHC-Water Holding Capacity

Sample Name	Place of Collection	Total Cyanide (mg/Kg)	Relative Standard Deviation (RSD)
HGML-NP	Hatti New Plot Ore tailing	243.3±20.81	8.55%
KGF-CNM	Kennedey Line dump	56±5.29	9.45%
COT –IG	Ingaldhal Copper Mine ore tailing	31.3±10.26	32.75%

Table 3: Cyanide concentration, Relative Standard deviation (RSD) of the Gold and Copper ore tailings determined by Standard Distillation Method (triplicates)

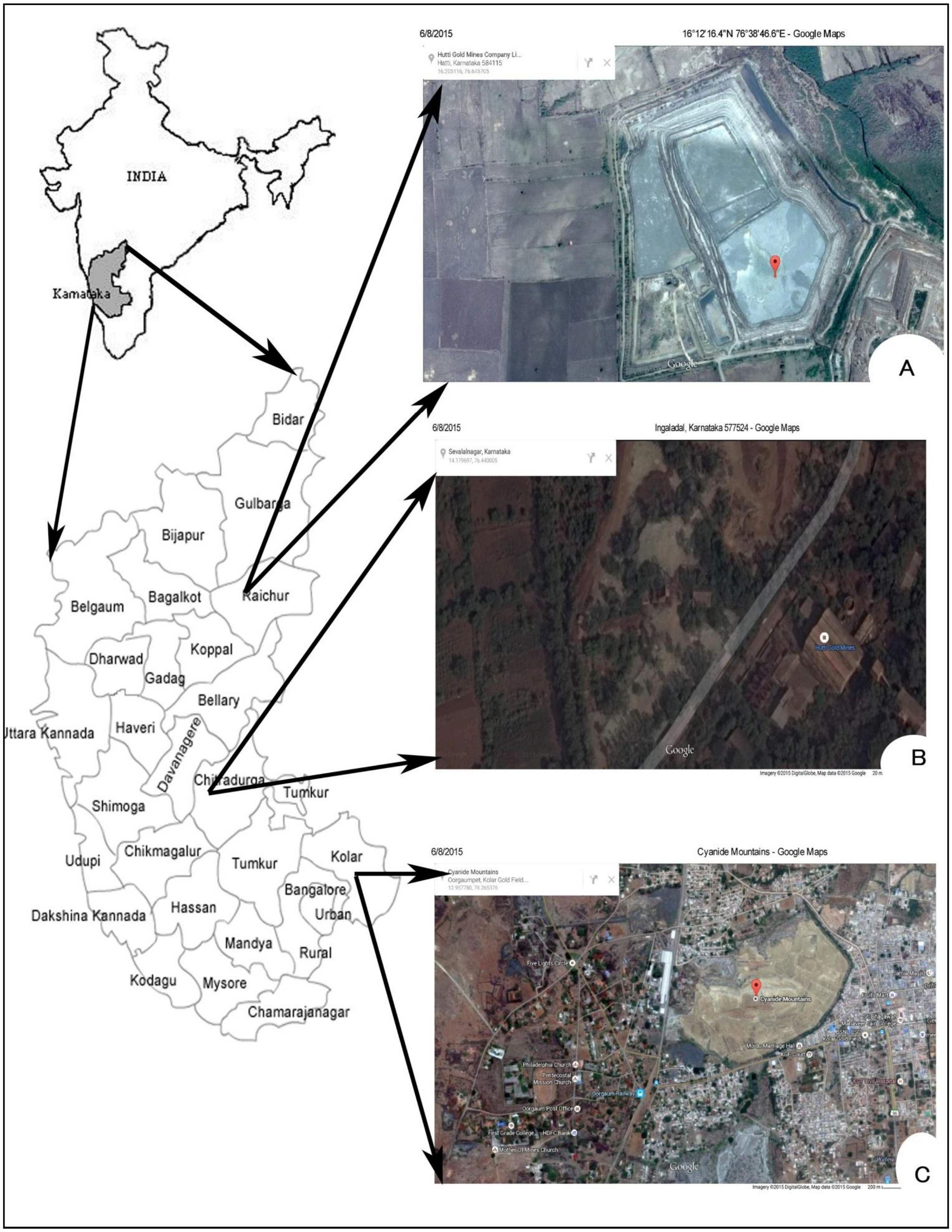

Figure 1: Sampling Sites of A. Ore tailings of HGML New Plot (HGML-NP), Hatii, Raichur; B. Ore tailing of Ingaldhal Copper mine owned by HGML (HGML-COT), Ingaldhal, Chitradurga; C. Ore tailings of Bharath Gold Mine (KGF-CNM, Kolar), Kolar Gold Field.

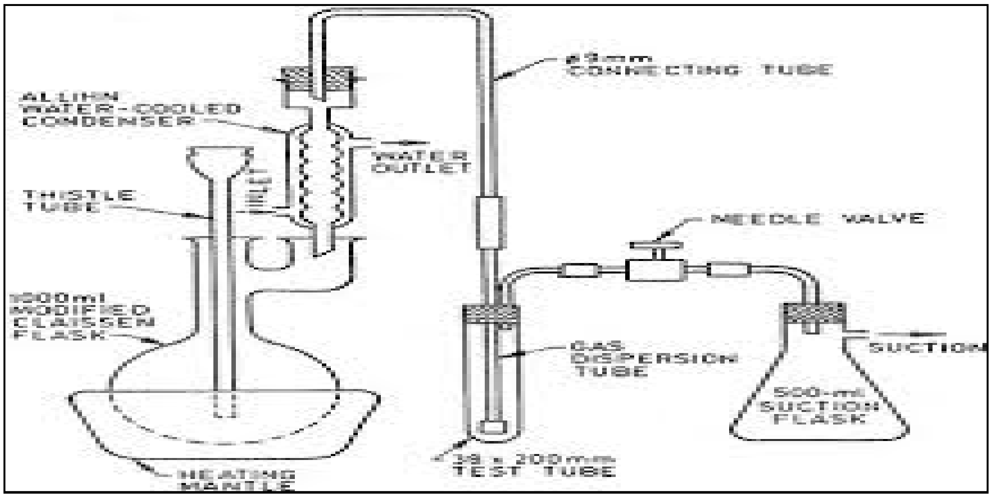

Figure 2: Standard Cyanide Distillation Apparatus

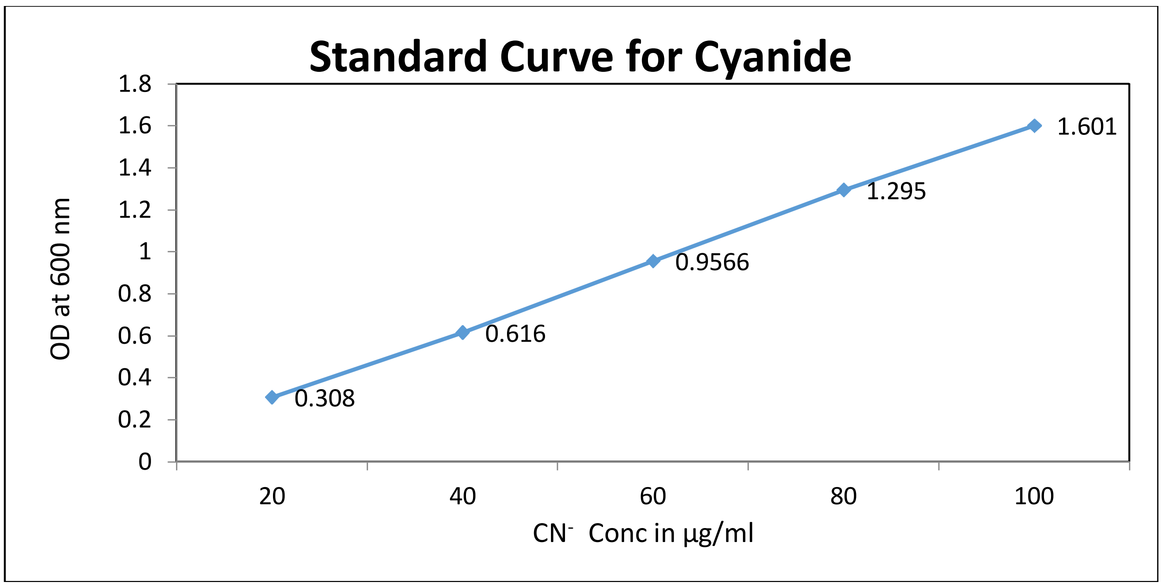

Figure 3: Standard Linear curve for Cyanide concentration against absorbance determined by Spectrophotometer

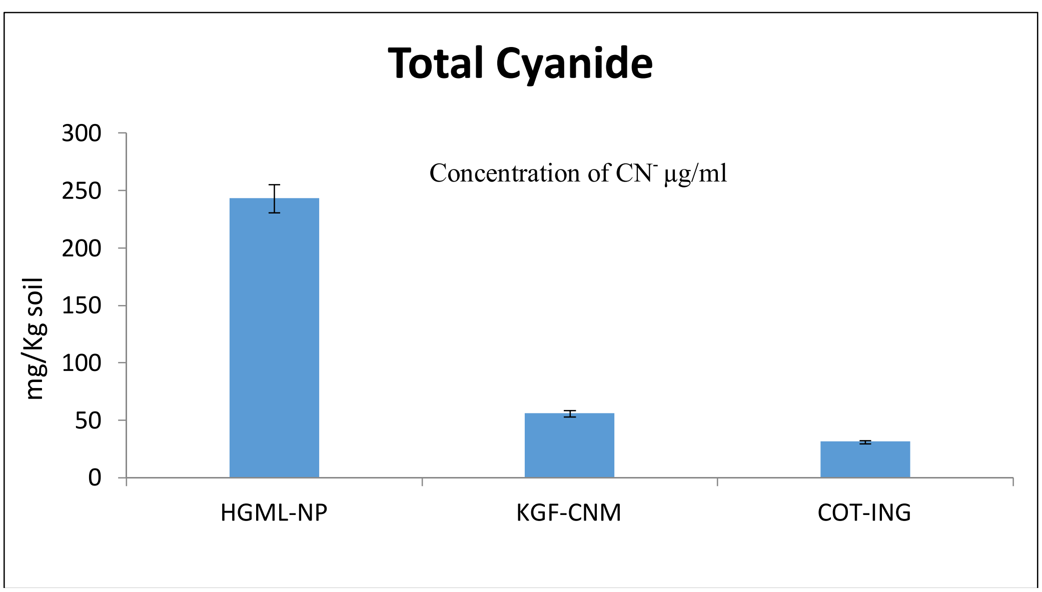

Figure 4: Total Cyanide level at HGML-NP, KGF-CNM, COT-ING Ore tailings, Karnataka, India