

SPATIAL DISTRIBUTION OF HEAVY METALS AROUND THE GOLD MINE ORE TAILINGS OF HATTI, KARNATAKA STATE, INDIA

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Abstract

Mining is an imperative segment of the world economy as it contributes socio-economic status of the nations. However, developing countries like India due to lack of high profile industrial techniques and equipment, eluting effluents from the industrial process may contain various hazardous substances which greatly affect the environmental and human health. The present work is aimed with the distribution of heavy metals in and around Hatti Gold Mine Ore Tailing (H-GOT). The results elicit the mine ore tailings are having high-level contaminants of heavy metal than the crop lands of Hatti (Hs), Kotha (Ks), Chikka Nagur (Cs), Tawag (Ts), Lingsugur (Ls) of Raichur District, Karnataka. It was reported that, Hatti Gold Mine ore tailings hold about 41.31 ± 0.49 mg/kg, 2.1 ± 0.31 mg/kg, 71.96 ± 3.26 mg/kg, 39.56 ± 1.47 mg/kg and 73.4 ± 2.19 mg/kg of Arsenic (As), Cadmium (Cd), Copper (Cu), Lead (Pb) and Zinc (Zn) heavy metals respectively. While the crop lands metal contamination range depends on seasonal variation. In south-west monsoon farming lands metal contamination order is $Hs > Ks > Cs > Ts > Ls$, and it was decreased during Post-monsoon. This is the hallmark of the fetching huge amount of toxic heavy metals from mining center to nearest crop lands. The continuous squeezing of these toxic metals could trigger the bio-magnification in both aquatic and terrestrial ecosystem and it may impact various metabolic disorders.

Keywords: Atomic Absorption Spectra (AAS), Gold Ore Tailing, Heavy metals, Inductive Coupled Plasma- Optical Emission Spectra (ICP-OES), Post-Monsoons, South-West monsoons

1. Introduction

Mining is the foremost supplier of revenue to the nations. Akpalu and Normanyo (2017) rumored that owing to lack of high profile equipment and public capital investment in industrial sectors of the third world countries, they attracted the private/foreign capitalist by compromising with the environmental standards. This insight leads to high-level pollution in mining sites and encompassing atmosphere. In developing countries like India, a substantial quantity of mining resulted waste were dropped publicly open space with none contraceptives (David et al. 2015; Harish and David, 2015) that

results in overburden of the ore tailings (Rao and Reddy 2005), discharge of tailing leachate into in and around the mining sites, additionally to those government owing activities like mineral effluents implicate as fertilizers (Kumar, 2013) are the foremost supply of environmental pollution. Moreover, pollutants carried out by mining activities in developing countries remains controversial, since it may impact hugely on the cultural, physical and socio-economic status of the local people (Verma et al. 2012; Harish and David 2015; Mustak et al. 2015).

In India, Karnataka is one of the major historical backgrounds in mining.

Throughout their mining, once extracting the specified parts from the well-mined ore, the left out solid wastes were omitted without being used. These solid wastes do contain an appreciable amount of micronutrient, heavy metal and other noxious elements (Harish and David 2017). Oftenly, this solid waste fetches its toxicants to adjoining environments through leachate, run-off, atmospheric fallout and wind erosion leads to a potential risk to the environment as well as human health (Armenta et al. 2003; Jung 2008; Sisodia 2013). Several authors reported that gold mine industries may degrade the natural setting, human health

and livelihood (Rao and Reddy 2005; Hazard 2013; Basavarajappa and Manjunatha 2015). Ono et al. (2012), Cobbinal et al. (2013) reported that, gold ore dumps are the major source of xenobiotic contamination through weathering of the heap waste materials, which results the discharge of toxic chemicals such as cadmium, lead, copper, arsenic, selenium, and mercury. However, most of the work was meted out on a spacial distribution of heavy metals and other xenobiotics in effluents, surface water and underground water source (Smith et al. 2000; Caldwell et al. 2006; Mishra et al. 2010; Ato 2010; Corriveau et al. 2011; Bempah et al. 2013) very little work was

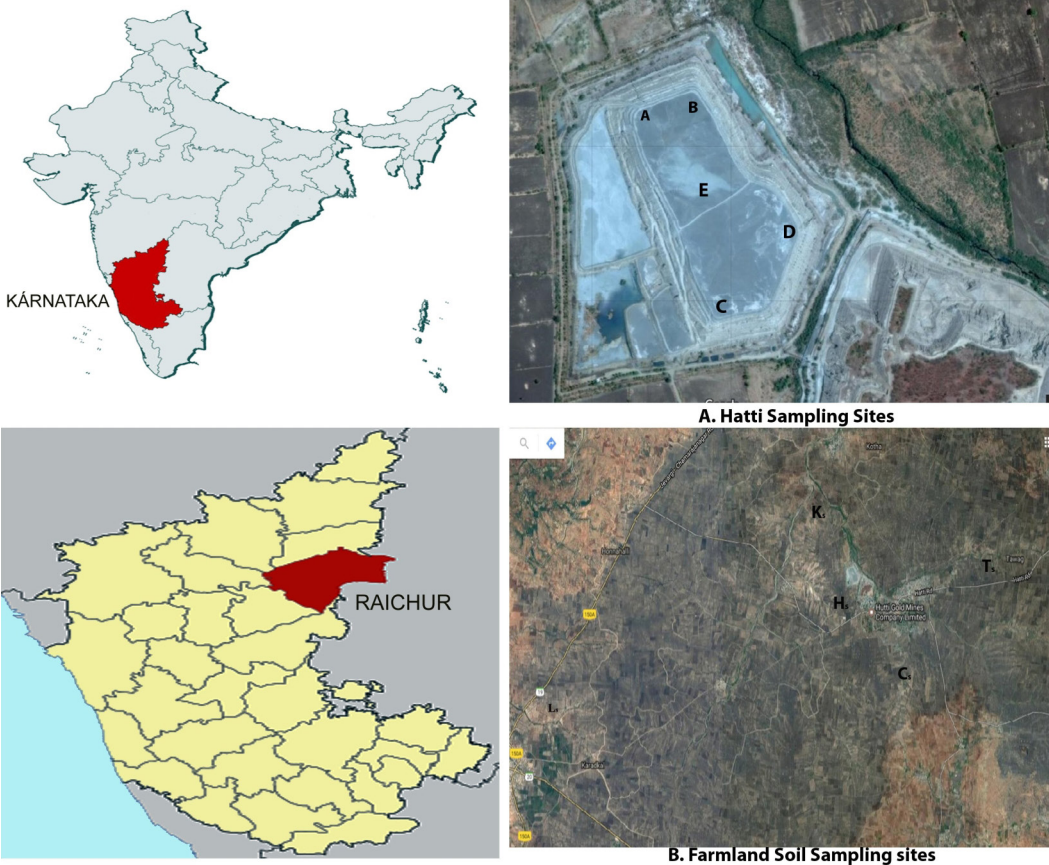


Fig. 1. Sampling sites of Goldmine ore tailing and farmland soil samples a). Sampling sites from the Hatti Gold Mine Ore Tailing (H-GOT) collected from the New Plot of H-GOT. b). Sampling sites in around the location (0.5 km to 5 km) of Hatti Mine Ore tailings, Hatti(Hs), Kotha (Ks), Chikka Nagur (Cs), Tawag (Ts), Lingusugur (Ls) farmland soil. Sources: Figure was collected from Google Map Source

carried out in the field of mine ore tailing and its impact on the agricultural field. Hence, the present study distinguishes from previous reports on Hatti Goldmine, because there are no reports available on seasonal variation of heavy metal in and around of Hatti. However, this probe is to assess the physico-chemical properties of Hatti gold mine ore tailing and farming lands of near villages of Hatti, gives an idea about mining activity and its impacts on heavy metal pollution. This study was carried out in two seasons i.e. south-west monsoon months and post-monsoon months.

2. Materials and Methods

Physiography and Topography of the Hatti Gold Mine

Hatti, the most productive gold mine sites in Karnataka. "Hatti Gold Ore" deposited within the Hutti-Maski metamorphic rock belt, it's a Neo-Archaean greenstone belt and it was dominated by a metavolcanic suite of rocks with subordinate amount of meta-sedimented rocks. Since, Pre-Ashoka period, Hatti village, Raichur District of Karnataka was the historical background in gold mining activity, lies within the magnitude of 16°12'17.5" N latitude and 76°38'45.5" longitude. The region was exposed to least amount of rainfall in the Karnataka state towards the south-west as well as the post-monsoon months. During the south-west monsoon months, the rainfall is about 71% of the annual rainfall and in the post-monsoon months it receives only 35% of the annual rainfall as noticed by Government of India, the ministry of water resources, central ground water board (www.cgwb.gov.in). Fig 1. Shows the Hatti gold mine ore tailing dumping yard, it was distributed in 15 acres, divided into an Old plot and New plot. The old plot is fully dried leachate and it was planted with Nilagiri (*Eucalyptus sp.*) trees. In new plot consists of 5 plots, each plot distributed in 1-2 acre. The present sampling was carried out in Plot-II of the new plot ore tailing and agriculture fields from near villages.

Sample Collection

About five ore tailing sample were collected from the plot-II of new plot from the Hatti Gold Mines ore tailings (H-GOT), depicted in (Fig. 1A) and the farm/cropland (Fig. 1B) soil sampling was carried out in the South-West monsoon months and Post-monsoon months respectively. Triplicate samples were collected in farm land sites with a fine polyether bags from each farming land of Hatti (16°18'12"N; 76°65'46"E), Kotha (16°22'899"N; 76°64'06"E), Chikka Nagur (16°18'12"; 76°67'48"E) Tawag (16°21'22"N; 76°69'89"E) and Lingsugur (16°14'21"N;76°52'60"E) of Raichur District, Karnataka, India. From each sampling point (Fig. 1B) three soil sample were collected in a triangle manner (0-10 cm depth) and mixed equally to form a composite sample.

Soil Pre-Treatment and Physicochemical Analysis of Samples

Prior to analysis, the ore tailing and soil samples were air dried at 30°C and grind in a mortar and passed through a 0.2 mm sieve. The resulted soil samples were used throughout the experiment for physicochemical and metal distribution analysis. The physicochemical properties like pH, water holding capacity (WHC), bulk density (BD), electrical conductivity (EC), Organic carbon (OC) were determined by the method used by Tijjani et al. (2013). Particle size distribution was determined by International standard pipette method described by Olmstead and Alexander (1930). Total elemental analysis of macro and micronutrients were determined by digestion and spectrophotometric methods (Pawluk, 1967).

Acid Digestion and Extraction of Heavy Metals

The total concentration of heavy metal elements in the soil samples were determined by EPA standard method 3050B (Roy-Keith 1998; Szakova 2010) with modifications. About 1 g of air-dried soil samples were

completely decomposed in a digestion vessel with a tri-acids mixture $\text{HNO}_3:\text{H}_2\text{SO}_4:\text{HClO}_4$ (2:1:3). The mixture was then heated in microwave assisted wet digestion system for 30 min at 250°C. After cooling, the digest was quantitatively transferred into a 50 ml Teflon vessel and allowed to evaporation at 160°C. The digest was then dissolved in a 3 ml try acid mixture (2:1:3), transferred into a 25 ml glass tube, filled with deionized water, and kept at laboratory temperature until measurement.

Quantification of Heavy Metals by using AAS and ICP-OES

Heavy metals such as Cd, Cu, Fe, and Zn concentration in aliquots were determined by Lefevre laboratory exercise method, (2010) by using AAS (GBC 902) instrumentation facility provided by Laxmi Narayana biotech laboratory, Raipur, Hubli-Dharwad, India. Standard metal solutions were brought from Merck Pvt. Ltd., Standard and working standard solution was prepared by using double distilled water, and blank was run routinely during the sample analysis to monitor instrument drift and overall quality of the analysis. Values were expressed as mg/kg soil. In order to determine the Arsenic and lead, work was carried out by using PerkinElmer inductively coupled plasma optical emission spectrometry (PE Optima 5300DV ICP-OES), kindly provided by Shiva analytical, Bangalore, India. It is a complete autosampler, polyscience chiller, and PC workstation with Winlab 32 software. PerkinElmer NIST® traceable quality control standards for ICP (N9300141, N9300281 and Spex Certiprep® Standard Lot 36-60 As) were used as the stock standards for preparing working standards. The descriptive statistical analysis was carried out by using SPSS (21.0) software.

3. Results and Discussion

Hatti, Raichur District of Karnataka State is one in every of the socio-economically developed towns. Majority of its development

was due to adopting the corporate social responsibility (CSR) policy by Hatti Gold Mine Company Ltd., for several years. In spite of this development, it also causes serious degradation of natural environment by instigating the metal ions through leachate, fallout, erosion, and runoff (Govil et al. 2008; Dasaram et al. 2011; Basavarajappa and Manjunath 2015). Once extracting the desired metals from the ore, the refused wastes are discharged into tailing pond, it may contain a high concentration of macro and microelements along with some noxious metal ions. Goldmine ore tailings are one of the main sources to introduce significant heavy metal pollutants like Cd, As, Cu, Zn and Pb into encompassing environment (Corriveau et al. 2011; Ono et al. 2012; Chakraborti et al. 2013).

Physico-chemical Properties of Gold Ore Tailings and Farmland Soil Samples

In the present study, physicochemical properties of collected ore tailings and farmland soil samples were determined and presented in (Table 1 and 2). The physical properties of H-GOT shows greenish-gray in color, alkaline and had a high percentage of fine sand with medium electrical conductivity. Whereas chemically, it's having a high concentration of elements such as phosphates, potassium, sulphates, calcium, magnesium, sodium and ferrous with low percent of organic carbon and nitrogen compared to farmland soil samples. Farmland soil samples show dark reddish to dark brown in color, with neutral to slightly alkaline pH, and having optimum concentration of macro and microelements. The physicochemical properties of croplands of Hatti (Hs), Kotha (Ks), Chikka Nagur (Cs), Tawag (Ts) and Lingsugur (Ls) results were emphasising that these are the good nutrient source for the growth and expansion of soil flora and fauna.

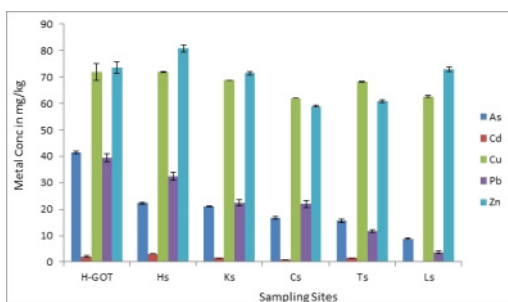


Fig. 2. Heavy metal concentration (mg/kg) in farmland soil samples during south-west monsoon months

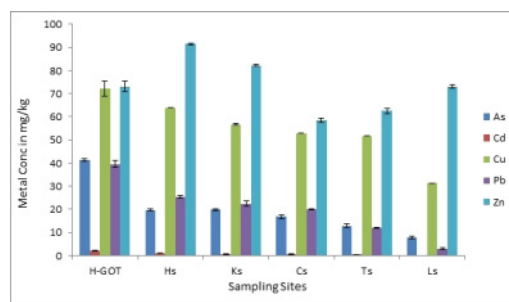


Fig. 3. Heavy metal concentration (mg/kg) in farmland soil samples in post-monsoon months

Heavy Metal Analysis of Gold Ore Tailing and Farmland Samples

Heavy metals from GOT and farmland soil samples were evaluated using Atomic Absorption Spectra (AAS) and Inductively Coupled Plasma-Optical Emission Spectrometry (ICP-OES) instrument and results were presented in Table 3, 4 and 5. Heavy metals of As, Cd, Cu, Pb, and Zn were determined in ore tailing composite and the mean values were 41.31 mg/kg, 2.1 mg/kg, 71.96 mg/kg, 39.56 mg/kg and 73.4 mg/kg

kg respectively, the values were lower than Canadian Council of Ministry of Environment-CME (2009) permissible limits except arsenic. Farmland soil sample shows an increased concentration of heavy metals during the south-west monsoon than post-monsoon season. Increased concentration heavy metal pollution in farmland soil samples was due to, oxidization of gold ore tailings containing metal sulphides on exposure to natural environment leads to production of large quantities of acid mine drainage, which may

Table 1. Physical properties of the gold mine ore tailing and cropland soil samples of in and around H-GOT

Soil samples	Parameters and their Values in Mean \pm SD from the Triplicate soil samples						
	Colour	pH	Moisture (%)	Sand (%)	Silt (%)	Clay (%)	EC (ds/m)
H-GOT	Greenish gray	8.12 \pm 0.15	23.24 \pm 0.02	81.83 \pm 0.42	9.8 \pm 0.05	6.58 \pm 0.04	1.62 \pm 0.02
H _s	Dark Reddish Brown	7.86 \pm 0.03	28.24 \pm 0.03	50.29 \pm 0.03	8.1 \pm 0.06	6.21 \pm 0.02	1.15 \pm 0.05
K _s	Dark Reddish	7.93 \pm 0.03	40.26 \pm 0.04	62.47 \pm 0.05	6.48 \pm 0.05	4.54 \pm 0.02	1.4 \pm 1.1
C _s	Dark Reddish	6.92 \pm 0.04	35.28 \pm 0.08	60.47 \pm 0.02	8.3 \pm 0.08	6.55 \pm 0.04	2.0 \pm 0.03
T _s	Dark Brown	8.12 \pm 0.03	32.05 \pm 2.91	61.75 \pm 2.4	9.02 \pm 0.11	6.8 \pm 0.54	1.94 \pm 0.06
L _s	Dark Brown	6.85 \pm 0.04	32.17 \pm 0.02	55.26 \pm 0.02	6.6 \pm 0.26	5.94 \pm 0.14	1.4 \pm 0.52

Table 2. Chemical Properties of gold mine ore tailings and crop land soil samples of in and around H-GOT

Soil samples	Parameter and their values were in Mean \pm SD from the Triplicate soil samples								
	OC (%)	N (%)	P (mg/kg)	K (%)	S (mg/kg)	Ca (%)	Mg (%)	Na (%)	Fe (%)
H-GOT	0.58 \pm 0.03	0.14 \pm 0.01	421.40 \pm 1.44	0.358 \pm 0.05	103.94 \pm 0.06	4.62 \pm 0.025	1.63 \pm 0.045	0.16 \pm 0.14	3.46 \pm 2.91
H _s	0.50 \pm 0.01	0.33 \pm 0.01	161.21 \pm 0.015	0.143 \pm 0.01	40.05 \pm 0.038	0.3 \pm 0.01	0.89 \pm 0.05	0.17 \pm 0.05	2.33 \pm 0.04
K _s	0.34 \pm 0.01	0.43 \pm 0.02	120.59 \pm 0.015	0.131 \pm 0.03	28.37 \pm 0.016	0.6 \pm 0.025	0.83 \pm 0.25	0.89 \pm 0.49	3.22 \pm 0.026
C _s	0.43 \pm 0.15	0.39 \pm 0.012	175.80 \pm 0.52	0.14 \pm 0.10	32.22 \pm 0.01	0.2 \pm 0.57	0.77 \pm 0.01	0.52 \pm 0.01	2.82 \pm 0.025
T _s	0.62 \pm 0.01	0.27 \pm 0.053	172.23 \pm 0.09	0.152 \pm 0.02	45.72 \pm 0.43	0.34 \pm 0.22	0.92 \pm 0.19	0.21 \pm 0.09	2.98 \pm 0.5
L _s	0.35 \pm 0.01	0.65 \pm 0.25	182.56 \pm 0.045	0.149 \pm 0.01	42.34 \pm 0.02	0.45 \pm 0.35	0.72 \pm 0.15	0.12 \pm 0.015	1.63 \pm 0.01

transfer to the surrounding environment through atmospheric dispersion and through flow of water which leads to release of metals from ore tailing (Rao and Reddy, 2005). This was one of the most important sources of heavy metals pollution in the encompassing environment. The evaluated results were compared with international standards bodies of United State Environmental Protection Agencies-US EPA, (2002), CME, (2009) as referred by He et al. (2015).

Fig. 2 and 3 depict the heavy metal loads in farmland soil samples during south-west monsoon and post-monsoon season. The major factors governing the dynamics of heavy metals in soils are pH, soluble organic matter content, hydrous metal oxide content, clay content, the presence or absence of organic and inorganic ligand and competition from other metal ions. It was reported that heavy metals such as As, Cd, Cu, Pb and Zn load was decreased in post monsoon season than the south-west monsoon. Arsenic load in croplands of H(s), K(s), C(s), T(s), shows more than permissible limits of arsenic (12 mg/kg), except L(s) soil shows below the standard value of CME (2009), but above the standard value of US EPA, (2002) for both season. Cadmium is slow water dissolving and less mobile in soil. The present study results report that, Cd concentration was increased in cropland of Hatti (3.05 mg/kg), Kotha (1.54) and Tawag (1.6 mg/kg) during south-west monsoon months and shows higher than CME (2009) permissible level (1.4 mg/Kg), whereas in post-monsoon months values were lower than permissible level except H(s) and it was absent in L(S) in

both seasons.

Draining of Zinc from the tailing pond to farmland soil shows elevated in H(s) and K(s) was due to airborne, atmospheric fallout. Zn contributed from ore tailings during post-monsoon due to wind erosion, remains shows less than ore tailing concentration. The concentration of Zinc is in the order of Hs>Ls>Ks>Ts>Cs in south-west monsoon where as in post-monsoon it shows in the order of Hs>Ks>Ls>Ts>Cs. Low Zinc permeability in south-west monsoon than post-monsoon in farmland soil was due to pH is nearly neutral and slow permeability was due to the major part of zinc was retained in the ore tailing due to the presence of high quartz and clay with finely grinded ore tailings. Also reported that, the behavior of Zn appears different in sandy rich and neutral loamy soils and these metals are associated with the organic matter hence, leachate may be neutral compared to other metals (Rahman et al., 2012).

Draining of copper from the tailing pond to farmland soil was elevated in both seasons. Present data indicate that, the concentration of Cu contamination in farmland soil shows high in south-west monsoon, low in post-monsoon season. Studies reported that, soil pH from 6-7.4 may increase the availability of Zn and Cu in farmland soil (Roy et al. 2012). Similar observations were found by Katyal and Sharma (1991) for Zinc and Wu et al. (2010) for Cu level. Rahman et al., 2012 also observed the uniform distribution of Cu and Zn in wet and dry season, the reason for this may be due to various anthropogenic activity and agricultural practice, the results

Table 3. Heavy metal content of the Hatti-Gold Mine Ore tailing Soil samples

Parameter	Concentration (mg/kg)	CCME Standard at Industrial Sites ^a (mg/kg)
As	41.31±0.49	12 ^a
Cd	2.1±0.31	22 ^a
Cu	71.96±3.26	91 ^a
Pb	39.56±1.47	600 ^a
Zn	73.4±2.19	360 ^a

^a Canadian Council of Minister of the Environment (CCME) Soil quality guidelines notified concentration of heavy metals at industrial sites

were coincident with his report. Lead (Pb) draining in both seasons is in the order of Hs>Ks>Cs>Ts>Ls and shows less than CME and USEPA standard in Agricultural soil (70 mg/kg). The present assessment noted that, the heavy rain falls during the south-west monsoon months may increase the leachate containing noxious metals of H-GOT towards the lower altitudes. Except, Lingusugur, the soil sample of the Hatti, Kotha, Chikka Nagur and Tawag showed the high content of heavy metals in south-west monsoon months. While in post-monsoon months the screaming of leachate is less as compared to south-west monsoon months due to lowest rainfall (http://cgwb.gov.in?District_Profile/Karnataka).

Atmospheric fallout/wind erosion/runoff/leachates were the major natural transport phenomenon of soil macro and microelements from one location to other locations (Zawadzki and Fabijanczyk 2013). Moreover, mine tailings leachate may contain nutritional elements like organic carbon, nitrogen, sodium, potassium, sulphur, iron, magnesium which are essential for soil microbial and plant growth. While the trace elements on overburden became a major problem, for example, the amendment of metal ions in soil could change the soil geochemistry (Bahuguna et al. 2012) and the trace amount of toxic elements entering the body via various routes can induce physiological and genetic alteration (Duruibe et al. 2007).

In some extents the huge nutritional availability and the high percentage of clay in ore tailings are implicated in compost preparation and also tailings are used in playgrounds preparation, the concrete composition which increases the flexibility of concrete (Skanda Kumar et al. 2014). In developing countries like India these waste are used in different ways in agricultural, concrete, playground preparations, roadside footpath constructions. However, these mining waste (effluents) are deporting the various toxic heavy metals into the surrounding environment and causes the

soil pollution (Govil et al. 2008; Hariprasad et al. 2013; Cobbinal et al. 2013; Chiroma et al. 2014).

There are massive findings on heavy metal pollution and health impacts throughout the country. Rao and Reddy (2005) reported that, silicosis is the most common health issue in gold mine ore tailing encompassing atmosphere was due to the presence of rich fine sandy in gold mine ore tailing. Nazir et al. (2015) reported that, the industrial waste used in irrigated croplands accumulates heavy metal (Ni, Cu, Cd, Cr, and Pb) and their uptake was high in spring season plants and shows some morphometric and physiological dysfunction. Chakraborty et al. (2013), Hazard (2013) were reported that, the arsenic contamination in the historic gold mining area of the Magalur greenstone belt of India was high and it causes arsenicosis in most of the local people. Also, various reports from the Raichur district depict the arsenic contamination and its effect on human health. Arsenic acid, arsenic pentoxide, and sodium arsenate are known human carcinogens (Cancer-causing agents) in adults. However, little childhood specific information is available regarding cancer effects of inorganic arsenic exposure during development (<http://www.epa.gov/teach>).

The present study reported that, the leachate/runoff/erosion/fallout phenomenon may lead to heavy metal soil pollution. In (Table 4) triplicate mean \pm SD values of heavy metals concentration in South-west monsoon months. During this period the ore tailing leachate fetches some amount of heavy metals to the surrounding croplands. While in the post-monsoons season, reduced levels ensured due to lowest rainfall. These results strongly emphasise the impact of mining activities on heavy metal contamination to crop lands in different season, the results were concurrent with Wuana and Okieimen (2011). In the present study, the concentration of heavy metals rising in and around of Hatti Gold mine area especially in rainy seasons. Consequently, the ore tailing leachate streaming across crop

lands towards the lower altitude of Krishna River belt. This may trigger biomagnifications in both aquatic and terrestrial ecosystems.

4. Conclusion

Obviously, mining activities improve the socioeconomic status of the developing countries. However, it causes serious environment degradation due to a substantial amount of mine waste discharged into environment. Augmented level of heavy metals in croplands of Kotha, Tawag, Hatti and Chikka Nagur during south-west months as compared to Post-monsoon months, elicits farther implementation of ore tailings purification and bioleaching techniques in mining program before discharging the mine effluents into Tailing Disposal Units (TDU).

Declaration

The authors declare that they have no competing interests.

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