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Spatial variation of heavy metal accumulation in coastal sea water, east coast of Andhra Pradesh, India

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Abstract
The concentration of heavy metal levels (Cu, Zn, Cd, Pb, Fe, Cr, Mn, Ni and Co) were measured in sea water samples by using Niskin sampler at different depths (10, 20, 30, 40 and 50 mts) and different areas (Visakhapatnam, Rushi konda and Bheemili) with the help of fisherman boats of east coast of Andhra Pradesh, India. Metals are analysed by using ICP-MS (Inductively coupled plasma mass spectrometry) and statistical analysis were done by using one way ANOVA. Concentrations of Cu range in between 8.03±0.09 and 21.21±0.01 ppm at 40 mts depth of Rushi konda coast and Bheemili coast, Zn levels was observed in the range of 22.05±0.83 and 37.05±0.56 ppm, Cd was found in between 0.51±0.16 and 5.36±0.04 ppm, lead was accumulated from 4.63±1.78 to 17.19±0.15 ppm, Fe accumulated from 28.31±1.47 to 40.86±1.05 ppm, Cr was found in between 6.47±0.28 and 16.82±1.16 ppm, Mn was observed in between 3.29±1.01 and 14.67±2.37 ppm, Ni ranges from 6.18±0.54 to 14.38±1.08 ppm and finally Co was accumulated in between 2.71±0.19 and 8.32±2.31 ppm respectively. The mean values of all the metal accumulation in Visakhapatnam coast is Zn>Fe>Cu>Pb>Ni>Cr>Mn>Co>Cd, Rushi konda coast is Fe>Zn>Cr>Cu>Ni>Mn>Co>Pb>Cd and Bheemili coast is Fe>Zn>Cu>Cr>Mn>Ni>Pb>Co>Cd. Based on the order of accumulation the metal content was more and it results pollution was more in the coastal sea water due to release of industrial wastes and anthropogenic activities.

Keywords: Heavy metal; coastal sea water; pollution

1. Introduction

Pollution of the aquatic environment and its effects on the living resources, especially the fishery resources, has assumed considerable interest as well as importance in the recent times. Pollution from metals is a vital problem affecting the estuaries and inshore regions. These pollutants released into air, water or soil is eventually carried into estuarine and coastal water systems. The concentration of metals in the organisms living in such environment is found to be higher than the concentration of pollutants in the surrounding media. Some of these metallic elements have no apparent biological function but affect uptake or excretion. Metallic elements from natural and anthropogenic sources are environmentally ubiquitous, readily dissolved in and are transported by water. They are taken up by aquatic organisms due to bioaccumulation and biomagnification in the food chain as elements or their metabolites, thus causing a concern for the potential concentrations in animals at the top of the food chain (Papagiannis *et al*. 2004) [32]. Non-essential metals (e.g. Pb, Cd and Hg), are held to be the most dangerous, since continuous exposure of marine organisms to their low concentrations may result in bioaccumulation, and subsequent transfer to man through food web. Fishes are widely consumed in many parts of the world by humans due to high protein content, low saturated fat and sufficient omega fatty acids known to support good health (United States Environmental Protection Agency 2004). Since most harvested fishes are located high in aquatic food chains, they accumulate metals from direct absorption from water (minor) and through trophic transfers (major), exposing human beings through food. The accumulation of metals in fish depends on equilibrium between absorption and depuration rates (Hadson 1988; Fowler *et al*. 1993; Mansour and Sidky 2002; Karadede *et al*. 2004) [10, 9, 19, 12], and thus may reflect localized bioavailability of these substances. Coastal area degradation with reference to its stability and also environmental quality are incessantly monitored.

Several causes of coastal destruction have been listed to be the coastal erosion, ill-planned development, land reclamation, uncontrolled marine sand mining, environmental degradation, and coastal resources utilisation (Seang & Latif, 2002) [29].

Industrial activities have been increased natural concentrations causing severe environmental problems. Water plays a significant role in amendable the components of the ecosystem in which the organisms are mostly involved in the mineralization and metabolization of many pollutants. Various studies in the coastal sediments were carried out (Kathiresan *et al*., 1996; Satpathy, 1996; Karthikeyan, 2007; Karthikeyan and Ananthan, 2011) [17, 26, 14, 16]. Marine coastal ecosystems could therefore be endangered by pollutants, such as heavy metals, pesticides and antifoulants that could be easily detected in the water column or in the sediment of harbours and estuaries (Antizar, 2008; Bellas, 2005) [4, 6]. The metal concentration in the marine environment is governed by various physical parameters (Vijayakumaran, 2005; Robin *et al*., 2003). Development of the coastal zones was caused a continuous increase in trace metal pollution of estuarine and marine waters, and high concentration of metals in seawater, sediments, and organisms in coastal areas (OSPAR Commission, 2000) [22]. Toxic elements are very dangerous even at low concentration if consumed over a long period of time. With ever increasing human population and associated rapid industrialization, the problem of environmental pollution have become more critical. Industrial effluents, agricultural run offs, transport, burning of fossil fuels, animal and human excretion, geological weathering, and domestic waste contribute to the heavy metal pollution of aquatic bodies (Ogoyi *et al*. 2011) [21]. Allocation of heavy metals in solution has widely recognized as a major factor in the geochemical behavior, transport and biological effects of these elements in natural waters (Ananthan *et al*., 1992, 2005, 2006; Karthikeyan *et al*., 2004, 2007) [1, 15, 16].

A few studies have been carried out on the concentration of heavy metals in the coast of Andhra Pradesh, India. (Satyanarayana and Prasad, 1985; Subrahmanyam *et al*., 1990; Ramesh *et al*., 2011) [27, 31, 24]. In this view, an attempt has been made to investigate the concentrations of some heavy metals in water samples and find out the pollution level at different stations to determine if industrial effluents extensively contribute to the occurrence of these elements in these areas. This study will help in forecasting and evading the severe damage to marine environment and also regulate toxic waste discharges.

2. Materials and Methods

a. Sample collection and area

The present study area was elected for three regions viz., Visakhapatnam, Rushi konda and Bheemili, East coast of Andhra Pradesh, India, the study area extends off Visakhapatnam to Bheemili. Sampling sites were elected based on propinquity of expected anthropogenic emission sources. Visakhapatnam station, a site near to fishing harbour and port, industrial plants like steel, zinc, fertilizer, oil refineries, and metal alloy, rushi konda station was at nearby Navy office INS Kalinga, which were released effluents directly into the sea water and bheemili station is the site close to pharmaceutical industries along with aquaculture farms. A total number of 15 transects were elected to collect the water samples vertically from three stations and horizontally in five depths of 10, 20, 30, 40 and 50 m using Niskin sampler and each sample was analysed for five determinants, keeping in view of the habitat of most edible fishes. Water samples were collected in pre cleaned, acid washed polypropylene bottles and filtered in Millipore filter paper (mesh size 0.45).

b. Sample digestion process

25ml of sea water was taken in to 100ml teflon vials and digested overnight with 7ml of pure nitric acid (AR grade, specific gravity: 1.38, Qualigens, India) to prevent precipitation of metals and to avoid microbial activity and 3 ml of hydrogen peroxide to stabilize the concentration. The concentration of heavy metals was studied by using Inductively Coupled Plasma Mass Spectrometry (ICP-MS) which is available in the Centre for Bay of Bengal Studies, Andhra University. Statistical analysis has been done by one way ANOVA. The data set was tested for homogeneity of variance and for normal distributes. For all statistical tests, probability of $p<0.05$ was considered significant.

3. Results and Discussion

The mean value of each parameter attributed the heavy metal accumulation in water samples of Andhra Pradesh coast, east coast of India. In the present study, high amount of metal accumulation was in Bheemili followed by Visakhapatnam and Rushi konda. The overall accumulation of metal order in Visakhapatnam region is Zn>Fe>Cu>Pb>Ni>Cr>Mn>Co>Cd, whereas in Rushi konda region accumulation of metal order is Fe>Zn>Cr>Cu>Ni>Mn>Co>Pb>Cd, while in Bheemili region, the metal order is Fe>Zn>Cu>Cr>Mn>Ni>Pb>Co>Cd. In region wise and depth wise, different metal content in sea water of Visakhapatnam, rushi konda and Bheemili region was shown in Figure 2, 3 and 4. More amount of total metal content was observed in 10, 30, 40 and 50 mts depth of Bheemili region whereas in 20 mts depth, total metal accumulation was observed more in Visakhapatnam region (Figure 5).

Copper is a comparatively heavy metal. The density of liquid copper is 8.22 g/cm^3. A very high value of the elements reveals anthropogenic input from the coastal industries. The antifouling paints used for the sea water intake pipeline structure is one of the major sources of higher concentrations of copper. The high concentrations of copper leaching from the ship's antifouling paint have been reported in the Indian Ocean (Danielson, 1980) [8]. In this study, copper accumulation was high in 40 mts depth and less in 50 mts depth of Visakhapatnam and Bheemili region, whereas in rushi konda region copper was more in 10 mts death and less in 40 mts depth (Table 1). In region wise, copper was more accumulated in Bheemili (17.70±0.31 μg/l) region followed by Visakhapatnam (14.89±0.26 μg/l) and rushi konda (11.06±0.41 μg/l) region (Figure 1). In the present study, the mean value of copper was in between 8.03±0.09 μg/l and 21.21±0.01 μg/l which was comparable to that observed in the Visakhapatnam coast (Subrahmanyam and Kumari, 1990) [31].

Zinc is obviously present in sea water. The average concentration of zinc in seawater was 0.65 ppb. Its solubility is depends on the temperature and pH of the water. Zinc was insoluble in water at neutral pH and solubility increases with increasing acidity. Zinc dissolves in water as ZnOH+ (aq) or

Zn2+ (aq). Anionic $ZnCO_3$ has a solubility of 0.21 g/L. Zinc is a relatively nontoxic element. In this study, zinc accumulated more in Visakhapatnam region followed by rushi konda and Bheemili region, while in depth wise, zinc accumulated more in 10 mts depth and low concentration was in 50 mts depth and in region wise, the mean value of zinc accumulated more in Visakhapatnam followed by rushi konda and Bheemili (Figure 1). Zinc ranges from 22.05±0.83 to 37.05±0.56 µg/l and average value was 29.71±0.64 µg/l (Table 1). Excessive intakes of zinc occur only with the inappropriate intake of supplements. In the study area, the zinc concentration is slightly fluctuating in all the stations. High concentration of zinc is may be due to more discharge of zinc containing fertilizers, pesticide through the industries.

Cadmium concentration of the present study is in between 1.81±0.02 and 5.36±0.04 off Visakhapatnam coast, whereas in between 0.51±0.16 and 2.24±0.05 off rushi konda region, while it was in between 1.16±0.41 and 3.64±0.54 of Bheemili region (Table 1), with an average concentrations of 3.29±0.41, 1.48±0.42 and 2.29±0.57 µg/l (Figure 1). High average concentration of cadmium was accumulated in Visakhapatnam region followed by Bheemili and rushi konda, whereas in depth wise, cadmium was high in 10 mts depth and less concentration in 50 mts depth of the given three regions. The concentration of cadmium in seawater ranged between 0.02 and 0.08 µg l-1, with a mean value of 0.05 µg l-1. The average concentration observed in the present study was higher than the values recorded in the Visakhapatnam coast (0.74 µg l-1; Subrahmanyam and Kumari, 1990) [31], coastal and offshore waters of western Bay of Bengal (0.9 and 0.35 µg l-1, respectively; Satyanarayana *et al.*, 1990) [28].

Seawater contains trace amounts of lead (230 ppt). Under normal conditions lead does not react with water. However, when lead comes in contact with moist air reactivity with water increases. A small lead oxide (PbO) layer forms at the surface of the metal. When both oxygen and water are present, metallic lead is converted to lead hydroxide (Pb (OH) 2): 2Pb(s) + O2 (g) + 2H2O (l) > 2Pb (OH) 2(s). Lead and lead compounds are generally toxic pollutants. Lead (II) salts and organic lead compounds are most harmful ecotoxicologically. Lead salts are attributed to water hazard class 2, and consequently are harmful. The same applies to lead compounds such as lead acetate, lead oxide, lead nitrate, and lead carbonate. In the study, lead values were in between 11.06 and 17.19 µg/l and its average is 14.72 of Visakhapatnam region, whereas in rushi konda region, values were range in between 4.63 and 8.06 and an average value is 6.36 µg/l and in Bheemili region, values were in between 7.98 - 12.05 and its mean value is 10.05 µg/l (Figure 1; Table 1).

Table 1: Metal accumulation (ppm) in region wise and depth wise

Spatial	Depth	Cu	Zn	Cd	Pb	Fe	Cr	Mn	Ni	Co
Visakhapatnam	10 mts	15.86±0.64	37.05±0.56	5.36±0.04	13.64±0.73	36.49±1.61	12.64±1.13	4.07±0.03	12.58±1.29	6.63±1.15
	20 mts	15.16±0.27	36.71±1.34	4.87±0.51	16.46±1.12	33.99±0.86	11.18±0.25	5.19±0.17	13.46±1.2	8.32±2.31
	30 mts	14.02±0.17	33.61±0.84	2.25±0.43	17.19±0.15	34.18±2.51	9.78±0.41	5.96±1.02	9.36±0.27	4.17±3.38
	40 mts	17.69±0.15	31.12±0.54	2.16±1.05	15.24±1.14	30.43±0.75	8.16±1.04	8.06±1.19	8.34±1.25	3.36±1.08
	50 mts	11.74±0.08	29.87±0.08	1.81±0.02	11.06±0.87	28.31±1.47	6.47±0.28	7.19±0.25	6.18±0.54	2.89±1.17
Rushi konda	10 mts	15.12±0.06	34.61±1.07	2.24±0.05	5.36±0.28	40.06±1.86	16.82±1.16	8.15±0.31	11.49±2.37	7.63±1.07
	20 mts	12.61±0.63	31.01±0.41	2.16±0.17	7.56±1.02	38.49±2.43	15.71±0.08	3.29±1.01	13.49±1.84	8.96±1.29
	30 mts	9.23±1.05	30.64±0.32	1.83±1.02	8.06±0.73	37.67±0.65	12.19±1.63	7.18±0.54	10.64±0.37	6.61±1.81
	40 mts	8.03±0.09	28.13±0.04	0.67±0.72	6.17±1.41	34.63±1.07	10.92±1.21	6.18±1.27	8.19±1.11	4.70±1.26
	50 mts	10.32±0.20	25.66±1.63	0.51±0.16	4.63±1.78	32.18±1.95	10.05±0.83	9.43±0.49	7.19±1.43	3.96±0.27
Bheemili	10 mts	18.83±0.13	28.64±0.16	3.64±0.54	12.05±1.03	40.86±1.05	14.87±2.05	9.06±0.35	14.38±1.08	8.13±0.59
	20 mts	16.37±0.03	26.32±0.27	2.75±0.18	9.86±0.66	36.32±0.08	13.79±1.19	11.73±1.07	12.11±0.01	6.17±1.74
	30 mts	17.15±1.21	25.53±0.19	2.05±1.05	11.97±1.75	39.64±1.64	15.30±0.24	14.67±2.37	9.42±0.09	5.96±2.61
	40 mts	21.21±0.01	24.71±1.37	1.86±0.67	8.37±2.01	34.18±2.14	10.79±0.86	14.35±0.58	9.06±0.36	3.37±0.67
	50 mts	14.96±0.19	22.05±0.83	1.16±0.41	7.98±0.60	33.09±1.08	10.19±0.05	10.62±1.23	8.24±1.86	2.71±0.19

Values are mean±SE, n=5

The iron accumulation in the present study of Visakhapatnam region was in between 18.31 and 36.49, while in rushi konda region, it was in between 21.18 and 40.06, whereas in Bheemili region it is a range from 16.09 to 36.32 µg/l respectively (Table 1). The average concentration of iron content was observed high in Bheemili followed by rushi konda and Visakhapatnam (Figure 1). This might be due to extensive iron ore handling in the outer Visakhapatnam harbor, its gradual dissipation into the coastal regions, and surface runoff from the river.

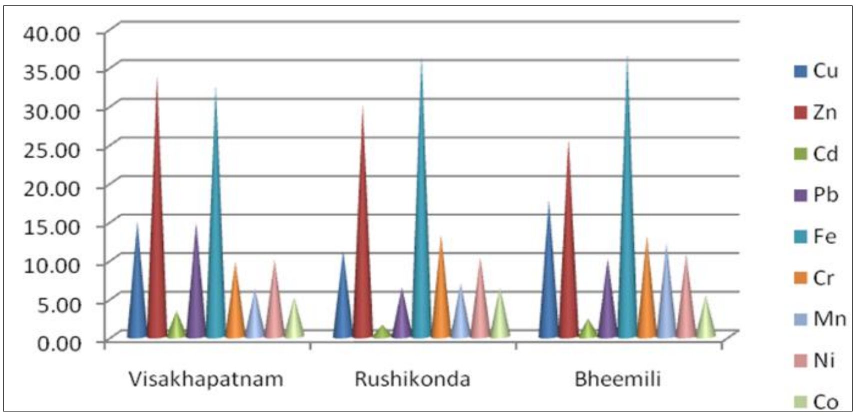

Fig 1: Metal accumulation (ppm) in region wise

Chromium does not occur freely in nature. The main chromium mineral is chromite. As was mentioned earlier, chromium compounds can be found in waters only in trace amounts. Seawater chromium content varies strongly, and is typically from 0.2 to 0.6 ppb. Chromium may be present in domestic waste from various synthetic materials. Through waste incineration it may spread to the environment when protection is insufficient. In the study area, the values were accumulated from 6.47 to 12.64 µg/l of Visakhapatnam region, whereas its range in between 10.05 and 16.82 µg/l of rushi konda region, while it's in between 10.19 and 15.3 µg/l of Bheemili region which has shown in Table 1. The average values of chromium were observed more in rushi konda region followed by Bheemili and Visakhapatnam (Figure 1).

Manganese is the second most profuse cation in sea water next to sodium. It does not appear to take part in the biological cycle; thereby it behaves as a conventional element in sea water. Therefore, its ratio to chlorinity is more or less constant except in nearshore areas where fresh water and other industrial activities modify the relationship between these two ions. In the present study, manganese range in between 4.07 - 8.06 µg/l and it average value is 6.09 µg/l in Visakhapatnam coast, whereas values were in between 3.29 - 9.43 µg/l and average is 6.85 µg/l in rushi konda region, while in Bheemili region values were in between 9.06 - 14.67 µg/l and its average was 12.09 µg/l (Figure 1; Table 1). The present study values were comparable with Solai *et al.*, 2010.

Seawater contains approximately 0.5-2 ppb of nickel. Phosphate fertilizers contain traces of nickel. Nickel is often present in agricultural soils situated near fossil fuel industries. Organic matter often adsorbs nickel, causing coal and oil to contain traces of the element. Nickel compounds may also be found in sludge, and in slags and fly ashes from waste incinerators. In this study, nickel values were in between 6.18 and 13.46 µg/l and its average value is 9.98 µg/l of Visakhapatnam region, and in rushi konda region values in between 7.19 and 13.49 µg/l and average value was 10.20 µg/l, whereas in Bheemili region, it was from 8.24 to 14.38 µg/l and average is 10.64 µg/l. More nickel was observed in Bheemili region due to cultured ponds.

The speciation of several trace metals in seawater is strongly influenced by complication to natural organic ligands, which has important implications for geochemical cycling, biological uptake and toxicity (Bruland *et al*., 1991; Hunter *et al*., 1997; Sunda and Huntsman, 1998) [7, 11, 32]. In open-ocean waters, cobalt has a relatively unique profile. Concentrations are generally low in surface waters (10–40 pM), which increase to a maximum in the upper thermocline (30–100 pM), and then decrease to values in deep waters of 10–30 pM (Martin *et al*., 1993) [20]. Cobalt is a biologically important metal although only few cobalt metalloproteins are known (Kobayashi and Shimizu, 1999) [18]. In this study, more amount of cobalt was accumulated in 20 mts depth and less in 50 mts depth of Visakhapatnam region, whereas in rushi konda region less amount was observed in 50 mts depth followed by 40 and 30 mts depth, while in bheemili region more cobalt content was observed in 10 mts depth and less amount was found in 50 mts depth (Table 1). The average values of cobalt was 5.07 µg/l in Visakhapatnam region, 6.37 µg/l was found in rushi konda region and 5.27 µg/l was observed in Bheemili region (Figure 1) respectively.

In all the transects the heavy metal concentrations were recorded high at 10 m depth followed by 20, 30, 40 and 50 mts depth. While increasing the depth, the metal concentration was deceased. The overall percentage of metal distribution observed in the present study is highest in bheemili region followed by Visakhapatnam and rushi konda region respectively. In all the stations, observations were more or less within the threshold limits for sea water from WHO. But the values are exceptionally high at Visakhapatnam region exceeding the permissible limits where the waters are highly influenced by anthropogenic inputs.

4. Conclusion

This study was carried out to provide information on heavy metal concentrations of sea water at different depths of Visakhapatnam, rushi konda and bheemili region. Moreover, these results can be used to evaluate possible risks associated with increasing the anthropogenic effects. The elevated concentrations of these heavy metals are apparently indicative of sea water pollution by toxicants, leading to bioaccumulation in aquatic organisms, which may surpass to humans causing ailments and deficiencies. Hence intensive studies to control and maintain the sea water parameters for sustainability of the valuable aquatic resources are in need.

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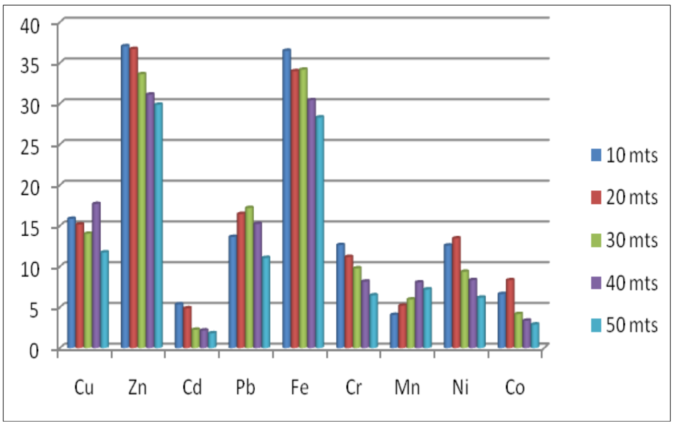

Fig 2: Depth wish metal content in Visakhapatnam region

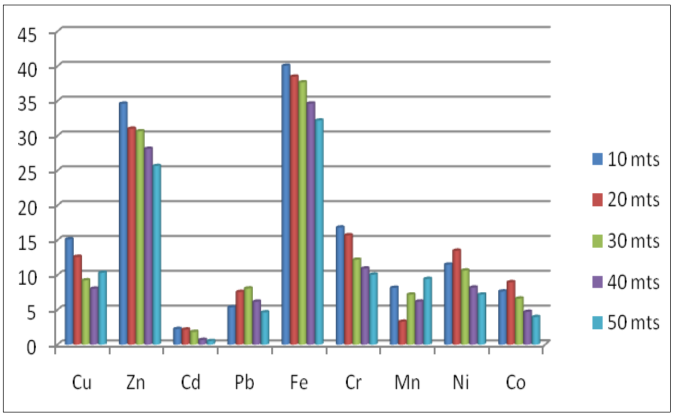

Fig 3: Depth wish metal content in Rushi Konda region

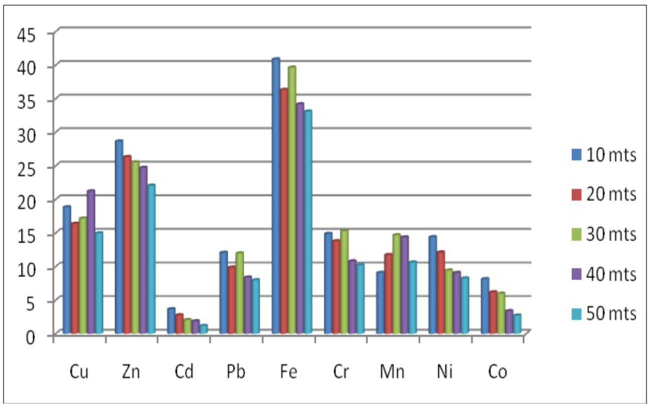

Fig 4: Depth wise metal content in Bheemili region

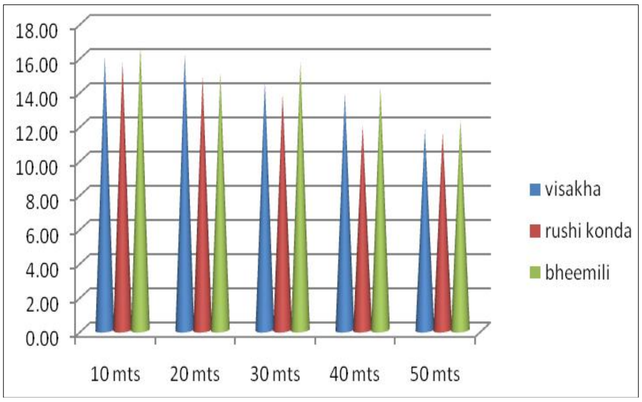

Fig 5: Overall metal content at various depths in region wise

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