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Management of Acidic Mine Waste Water by Constructed Wetland Treatment Systems: A Bench Scale Study

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Abstract

Constructed wetlands have emerged as a viable option for treatment of acidic mine wastewater. Bench scale experiments were conducted, to evaluate the performance of wetland treatment system, with emergent macrophyte Desmostachya bipinnata and substrate (containing powdered goat manure, wood shavings and soil) for 24, 48, 72, 96 and 168 hours of retention period for different water column heights (100mm, 150mm and 200mm). Within 24 hours of retention period the pH increased from 2.93 to 7.22, alkalinity increased from zero mg/L to 204.30 mg/L as $CaCO_3$, electric conductivity decreased by 27.94 to 35.93 percent, turbidity decreased by 30.14 to 66.65 percent, sulphate reduced by 21.52 to 28.09 percent, acidity reduced by 88.89 to 100 percent, hardness reduction was 18.18 to 26.07 percent. Potential metal removal rates was 95.20 to 95.51 percent for Fe, 88.77 to 91.72 percent for Cu, 76.75 to 78.21 percent Zn, 88.80 to 90.12 Pb, 67.65 to72.33 percent Co, 29.60 to 63.76 percent Ni and 35.90 to 76.44 percent Mn respectively.

Keywords: Acidic Mine Water,Treatment, Constructed Wetland, D. bipinnata.

1. Introduction

The management of acidic mine wastewater from mining and processing of sulphide minerals, and coalmines poses an environmental challenge to the mining industry. Mining of these ores and coals exposes the sulphide/ pyritic and pyrrhotitic minerals to oxygen and water, which coupled with the microbial activity, lead to the formation of waters that are highly enriched with sulphate and a range of heavy metals commonly known as acid mine drainage (AMD). Other pollutants and toxic metals such as arsenic, cyanide may also be present in acidic mine water at elevated concentrations when these sulphide minerals are processed in minerals processing units. Further, mill tailings amount to about 90% of ore is dumped in tailing impoundments and seepage from these impoundments can be source of AMD for decades. Nordstrom and Alpers (1999) estimated that without preventative measures the Richmond Mine, at California's Iron Mountain, would generate AMD, with $pH < 1$ and containing several $g\ L^{-1}$ of dissolved metals, for 3000 years. Kalin (2001) estimated, based on oxidation rates derived from tailings pore water samples, that the site of a small zinc/copper mine in northwest Ontario, Canada, would generate AMD for 1000 to 35,000 years. These may be extreme examples but it is not uncommon for base metal mines and their waste products to generate acid for more than 100 years. Since AMD is self-renewing, an ideal solution to it would also be self-perpetuating(Akcil and Koldas, 2006).

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The most evident consequence of AMD contamination to water bodies is, the precipitation along the streambeds, of brightly colored orange or yellow secondary iron and sulphur minerals, which are referred to as `yellow boy'. If left uncontrolled, it results in contamination of ground and surface waters, as well as of the surrounding land. Because of its leaching capacity, augmented by the activities of the bacteria it attracts, it is self-perpetuating (Kalin et al., 2006). Being associated with both operating and abandoned mining operations, abandoned mine sites may cover very large areas; it is difficult to inhibit the formation of the AMD (Whitehead and Prior, 2005). It has the potential to contaminate ground and Local watercourse thus leading to a serious harmful impact on ecosystem (Grande et al., 2005). An insidious feature of AMD is that its sources may remain active for decades or even centuries after mine closure (Modis et al., 1998).

The treatment of AMD is one of the major concerns for the mining industry. Ironically, even under the best reclamation scenario it requires long-term collection and treatment. There are two distinct strategies for treating AMD. The conventional solution is to collect and chemically treat acidified effluents in a centralized treatment plant by continual addition of alkaline chemicals like lime, hydrated lime, caustic soda, soda ash etc. These chemicals besides being expensive, also, lead to production of voluminous sludge (Cohen, 2006). These active treatment processes are often expensive both in terms of capital and operating costs. Alternatively, effluents can be routed through natural and constructed wetlands within which the biotic and abiotic communities treat acidic mine wastewater. In the past decades, therefore, research efforts have been directed towards wetlands as an alternative passive means of treating AMD. Such a passive treatment is economical, non-polluting, and is not a source of secondary wastes. Moreover, a well-engineered passive treatment system is a closed ecological system and hence is self-renewing. (Kalin, 2004;Matagi et al., 1998).

Wetlands are composite systems where a variety of physical, chemical, microbiological and plant-mediated processes occur that cause significant changes in the water chemistry (Kuyucak, 2002). It was accepted that plants probably remove metal into their biomass and that they provided micro aerobic zones for bacteria that may assist in the removal process (Greenway and Simpson, 1996). Subsequently, much work in the development and engineering of wetlands to treat mine water discharge has been carried out by U.S.Bureau of mines (Hallberg and Johnson, 2005). Engineered wetlands have been successfully applied in many locations to treat AMD. The present study was carried out to examine the feasibility of the passive technology in India. The experiment involved monitoring bench scale artificial wetlands set up in laboratory for several parameters over a period of one-week (retention time). The specific aim was to study the suitability of the organic substrate with the macrophyte *D. bipinnata* to treat acidic mine wastewater so that this technique may be implemented in the field as Indian climate conditions are quite favorable.

2. Methodology

2.1 Generation of Acid Mine Water in the Laboratory

An excavation of size 6.1m x 3.7m x 1.75m was made in the ground. It was then lined with stone masonry and cement plaster with three coats of acid resistant paint

(Epoxy Paint) on the entire excavated area. For the generation of AMD in the laboratory one tone copper ore (Chalcopyrite in powdered form), one tone of lead-zinc ore (Galena+Sphalrite), one tone mill tailings (Copper ore) and 20 Kg. pyrite (powdered) were put into the storage tank and kept moist for six months in this excavated tank. When the pH of the mixture started decreasing additional water was added until the pond was completely filled. The whole process took 1.5 years to reach a pH close to 3.0.

2.2 Construction of Bench Scale Wetland Test Cells

Simultaneously three bench scale test cells of 1m x 1m x 1m were constructed near the storage tank and plastered from both inside and outside with cement mortar. The same acid resistant paint was also applied on the inner surface of all the three test cells. The first cell was constructed on the ground level, second was constructed on a platform of 0.3m above the ground level and third cell was about 0.6m above the ground level (Figure 1). A pipe was connected about 0.25m from the top of the third cell and was connected to the second cell. Similarly from the top of the second cell 0.25m below a pipe was fixed and connected to the first cell. Initial idea was to feed AMD in third cell and than allowing the AMD to flow in the second cell and subsequently in the first cell in a downstream manner.

After the completion of construction work the base of the test cells was filled with sandstone, gravel, pebbles up to the height of 0.15 m. The purpose of putting these stone pieces is to enhance stability of the plants by providing gripping facility to the roots. It was realized that establishing and maintaining the proper environment in the substrate is the key to success for removal of pollutants. Various researchers used different substrate combination for removal of the contaminants(Gibert et al., 2004).

In the present experimental work the substrate used was soil 75 percent, goat manure (in powdered form) 20 percent and wood shavings 5 percent. The thickness of the substrate was 0.50m(Figure 2). On the top of the substrate a thin layer of 10 mm of goat manure was also spread after planting the plants in different test cells obtained from a natural wetland receiving AMD at Khetri Copper Project tailing dam downstream side (India). Downstream the seepage of the tailing dam has created natural wetland and the aquatic plant dominating in the wetlands is *Desmostachya bipinnata* (*D. bipinnata*).

D. bipinnata is tufted, stout, rhizomatous perennials, upto 1.25 meters tall with creeping root-stock. Leaves 20mm-50mm x 3.5mm-7.5mm,acuminate; sheath glabrous, densely flabellate. Inflorescence 0.25m-0.50m long. Flowering season is from July to November. It is commonly found near moist places, particularly in sandy and clay soils. It is available all over the Rajasthan state (India).

2.3 Experimental Procedure

Initially after transplanting the plants in the respective bench scale biological test cells, normal tap water was fed to them. Then thereafter AMD generated in the laboratory pond was fed to them. When the water in the pond reached pH between 3.0-3.5 and plants in all the cells were well grown up and height of *D. bipinnata* ranged upto 1.25 m covered the full area of the cell and then it was decided to conduct the experiments (figure 3 a,b). Water in the tank was well mixed by running the electric monoblock pump for about two hours, taking water from one end of the tank and delivering it to the other end so that the water composition in the tank becomes homogenized. AMD from the generation pond was pumped to a tank placed on an elevated platform (Figure1). From this tank it was designed initially that AMD will be filled in the third cell of the set and than on the next day it will be allowed to flow in the next cell through a pipe fixed for the purpose. However due to blockage of pipes due to litter and roots, pipe flow could not materialize therefore each cell was separately filled with AMD. In the first set of the experiments, water in each test cell was filled up-to 100mm height. One liter of the water sample was taken for the chemical examination before and after 24, 48, 72, 96 and 168 hours. These samples were taken using a small plastic suction pump manually, collecting water from four corners of the cells and also from the center keeping the suction pump approximately in the center of the water column so that a true representation of water quality in the test cell can be obtained by the sample.

In the second set of experiment the height of water column was increased to 150mm and in the third set of experiment the height of water column was increased to 200mm and input water samples and output water samples were taken for analysis similar to the first set.

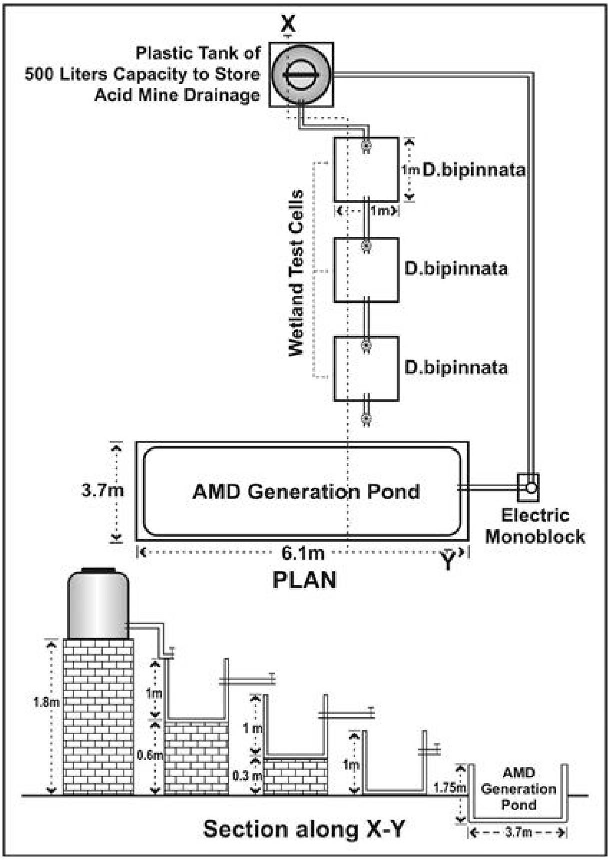

Figure 1. Plan and section of experimental setup

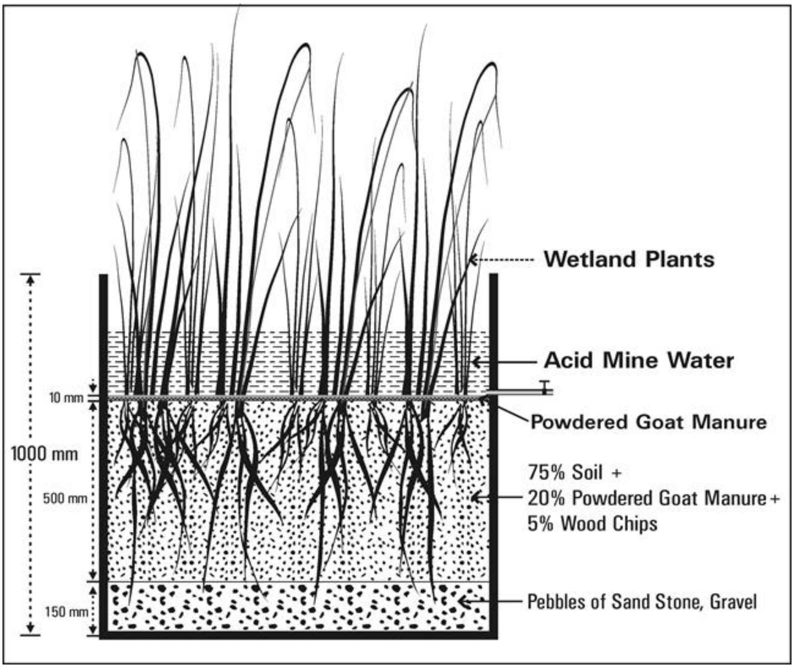

Figure 2. Section of constructed wetland test cells

The following parameters were examined in the laboratory daily - pH, Electric conductivity, Turbidity, Total hardness, Acidity, Alkalinity, Sulphate and for the metal ions examination the 500 ml filtered water sample were preserved using nitric acid as detailed in Standard Methods for the Examination of Water and Waste Water (APHA, 1992). The pH of water samples was determined using pH meter (Make- RI, New Delhi, Model 151R) Buffer solutions of 4 and 7 and 9 were prepared using the standard chemicals supplied by Qualins. The electric conductivity was measured using conductivity meter (Make- M.S. Electronics India Pvt. Ltd., Model – 601E). The turbidity was measured using turbidity meter (Make- RI, New Delhi, Model -211R). Total hardness, acidity and Alkalinity of the water samples were measured using titration method. For the determination of sulphate ions in the water samples turbidimetric method was used. The method uses precipitation of barium sulphate ($BaSO_4$) by the addition of barium chloride ($BaCl_4$) to create turbidity in the sample, which was measured in a turbidity meter (Make- RI, New Delhi, Model -211R). Samples were diluted prior to addition of barium chloride based on the estimates of sulphate concentrations so as to yield acceptable absorbance values. Blanks and standards (Na_2SO_4) were carried through the procedure to generate a standard curve as stipulated in the method. Metal concentrations were qualified by an atomic absorption spectrophotometer (Make- G.B.C Australia, model-AVANTA). All the parameters were determined as detailed in Standard Methods for the examination of Water and Waste Water (APHA, 1992) was used.

Figure 3. Photograph of wetland test cells

3. Results and Discussion

The monitoring of the treated water, collected from three sets of experiments, revealed efficient removal of pollutants, heavy metals and sulphates in the biological test cells after retention period of 24, 48, 72, 96 and 168 hours (Table 1,2 and 3) in different water column heights. This was due to different biological, chemical and physico-chemical processes. The anaerobic sulphate reducing bacteria produced hydrogen sulphide using dissolved organic substances as electron donors and sulphate ions as electron acceptors (Coetser et al., 2006). The alkalinity produced during microbial sulphate reduction as well as the alkalinity produced by the solubilisation of some acid consuming minerals present in wetland gradually increased the pH and stabilized it around neutral point. It was observed that most of pollutant removal took place within 24 hours retention period, although experiments were performed up to 168 hours. The pH values of the influent were3.31, 3.23 and 2.93 for water column height of 100mm, 150mm and 200mm respectively (Table 1,2 and 3). The pH increased up to 7.14,7.24 and 7.22 in 24 hours of retention period. Treated AMD even exhibit initial reduction in conductivity up to35.93 percent in test cells with 100 mm water column but gradually values for the conductivity increased at the outlet up to29.27 percent with 150 mm and 27.94 percent with 200 mm respectively. This increase in conductivity may be due to release of nutrients back into the water as a result of plant decay, increasing the dissolved ion contents. (Mashauri et al. 2000). Reduction in turbidity ranged up to 66.65 percent when height of column was 100mm, reduced to 30.14 percent when height of column was 150 mm and reduced to 33.33 percent with 200 mm. The influent sulphate values ranged from 395, 415 to 445 mg/L in the different water column heights of 100mm 150mm and 200 mm respectively. After 24 hours of treatment sulphates reduced to 310.00mg/L, 303.33mg/L and 320.00 mg/L respectively. The removal rate of sulphates gradually decreased the acidity due to sulphate ions thus making the drainage alkaline. Alkalinity increased from zero to 204.30 mg/L as $CaCO_3$ in retention period of 24 hours with water column height of 100mm; similarly when water column height was raised to 150mm and 200mm, the alkalinity increased up to 83.23 and 60.53 mg/L as $CaCO_3$. Organic substrate with well-grown *D. bipinnata* was highly efficient in up taking heavy metals from AMD. Among heavy metals, iron was the first element to be removed, followed closely by copper, zinc, lead and other heavy metals present in mine water.

Table 1: Water quality before and after different retention period in test cells with water column height 100 mm

Parameters	Input	After 24 hours	After 48 hours	After 72 hours
pH	3.31	7.14	7.43	7.46
Conductivity μS /cm	1515.00	970.67	1013.00	1013.00
Turbidity JTU	20.00	6.67	4.00	3.33
Sulphate mg/L	395.00	310.00	253.33	248.33
Acidity mg/L as $CaCO_3$	125	0.00	0.00	0.00
Alkalinity mg/L as $CaCO_3$	0	204.30	227.00	246.37
Hardness mg/L as $CaCO_3$	591.85	437.58	409.40	395.06
Iron mg/L	17.642	0.792	0.407	0.233
Copper mg/L	14.110	1.250	0.553	0.504
Zinc mg/L	29.100	6.150	3.200	1.662
Lead mg/L	1.700	0.177	0.123	0.093
Cobalt mg/L	0.300	0.083	0.060	0.057
Nickel mg/L	0.379	0.137	0.120	0.110
Manganese mg/L	2.010	0.747	0.513	0.397

Sample could not be taken on day 4 and 7 as water had been absorbed.

Table 2: Water quality before and after different retention period in test cells with water column height 150 mm

Parameters	Input	After 24 hours	After 48 hours	After 72 hours	After 96 hours	After 168 hours
pH	3.23	7.24	7.36	7.46	7.53	7.84
Conductivity μS/cm	1565.00	1107.00	1128.00	1195.33	1196.33	1251.67
Turbidity JTU	21	14.67	12.33	10.00	3.33	2.00
Sulphate mg/L	415	303.33	288.33	280.00	273.33	261.67
Acidity mg/L as $CaCO_3$	156.25	13.88	8.67	3.47	0.00	0.00
Alkalinity mg/L as $CaCO_3$	0	83.23	117.28	132.42	162.68	230.78
Hardness mg/L as $CaCO_3$	623	499.88	471.70	452.42	431.65	424.23
Iron mg/L	17.950	0.812	0.454	0.344	0.216	0.143
Copper mg/L	14.850	1.667	0.773	0.673	0.613	0.419
Zinc mg/L	29.500	6.374	4.034	3.931	2.005	1.096
Lead mg/L	1.720	0.170	0.122	0.108	0.090	0.027
Cobalt mg/L	0.330	0.103	0.090	0.067	0.063	0.043
Nickel mg/L	0.382	0.253	0.160	0.124	0.089	0.070
Manganese mg/L	2.200	1.410	1.298	1.230	1.136	0.897

Table 3: Water quality before and after different retention period in test cells with water column height 200 mm

Parameters	Input	After 24 hours	After 48 hours	After 72 hours	After 96 hours	After 168 hours
pH	2.93	7.22	7.45	7.53	7.77	8.03
Conductivity μS/cm	1589.00	1145.00	1208.67	1246.67	1258.33	1548.33
Turbidity JTU	24	16.00	12.67	8.33	7.00	5.33
Sulphate mg/L	445	320.00	301.67	283.33	276.67	268.33
Acidity mg/L as $CaCO_3$	177.08	20.83	10.42	10.42	10.42	6.93

Alkalinity mg/L as $CaCO_3$	0	60.53	113.50	109.72	139.98	185.38
Hardness mg/L as $CaCO_3$	640	523.62	485.05	477.63	440.57	423.62
Iron mg/L	17.990	0.863	0.489	0.405	0.279	0.206
Copper mg/L	14.900	1.233	0.722	0.657	0.610	0.730
Zinc mg/L	29.500	6.859	3.860	2.786	2.147	1.101
Lead mg/L	1.840	0.206	0.131	0.113	0.061	0.033
Cobalt mg/L	0.340	0.110	0.077	0.077	0.057	0.057
Nickel mg/L	0.402	0.283	0.182	0.132	0.093	0.070
Manganese mg/L	2.220	1.633	1.467	1.387	1.357	1.253

Table 4: Percentage removal after 24 hours of retention in test cells with three different water column height.

Water column height → Parameters ↓	100 mm	150 mm	200 mm
pH	3.31-7.14	3.23-7.24	2.93-7.22
EC	35.93	29.27	27.94
Turbidity	66.65	30.14	33.33
Sulphate	21.52	26.90	28.09
Acidity	100.90	88.89	92.16
Alkalinity mg/L as $CaCO_3$	0-204.30	0-83.23	0-60.53
Hardness	26.07	19.76	18.18
Iron	95.51	95.48	95.20
Copper	91.14	88.77	91.72
Zinc	78.21	78.39	76.75
Lead	89.59	90.12	88.80
Cobalt	72.33	68.79	67.65
Nickel	63.76	33.77	29.60
Manganese	62.83	35.90	76.44

Except pH and Alkalinity all values are in percent.

Table 5: Part of general standards for discharge of environmental pollutants. Part A, effluent, (Schedule-VI, 1993). MOEF, GSR 422(E), 19.05.93. (The standards shall also apply to effluent discharge such as mining, mineral processing activities and sewage. Maiti, 2001).

Parameters	Inland surface water	Public sewers	Land for irrigation	Marine coastal areas
pH value	5.5 to 9.0	5.5 – 9.0	5.5 – 9.0	5.5 – 9.0
Lead (as Pb), mg/L, Max.	0.1	0.1	-	2.0
Cadmium (as Cd), mg/L, Max.	2.0	1.0	-	2.0
Copper (as Cu), mg/L, Max.	3.0	3.0	-	3.0
Zinc (as Zn), mg/L, Max.	5.0	15	-	15
Nickel (as Ni), mg/L, Max.	3.0	3.0	-	3.0
Sulphate (as SO4), mg/L, Max.	1000	1000	1000	-
Manganese (as Mn), mg/L, Max.	2	2	2	2
Iron (as (Fe), mg/L, Max.	3	3	-	3

Copper sulphide is highly insoluble over a broad pH range and sulphide precipitation of copper ions is therefore a rapid and efficient process in acid mine waters. The influent load of iron was 17.642 mg/L, 17.950 mg/L and 17.990 mg/L in the different water columns respectively. Iron concentration reduced to 0.792 mg/L 0.812 mg/L, and 0.863mg/L in 24 hours of retention period in different water column heights. The removal efficiency ranged from 95.20% to 95.51% (Table 4). Copper and zinc influent load was 14.110 mg/L, and 29.100 mg/L in 100mm of water column height, 14.850 mg/L and 29.500 mg/L in 150mm of water column height. Likewise when the height of water was raised to 200mm the copper concentration in influent was 14.900 mg/L and zinc load was 29.500 mg/L. After 24 hours of treatment high reduction was observed. The copper concentration reduced to 1.250 mg/L; 1.667 mg/L and 1.233mg/L and zinc concentration dropped to 6.150 mg/L, 6.374 mg/L and 6.859 mg/L respectively. The removal efficiency of copper ranged up to 88.77% to 91.72 % and zinc ranged up to 76.75 % to 78.39% respectively. Lead and cobalt concentration observed in influent load was 1.700 mg/L and 0.300 mg/L; 1.720` mg/L and 0.330 mg/L; 1.840 mg/L and 0.340 mg/L in different water column heights of 100mm 150mm and 200mm respectively in the retention period of 24 hours. The lead concentration dropped to 0.177 mg/L, 0.170 mg/L and 0.206 mg/L whereas cobalt concentration dropped to 0.083 mg/L, 0.103mg/L and 0.110 mg/L in different water column heights, the percentage of removal efficiency of lead ranged up to 88.80% to 90.12% and cobalt ranged from 67.65% to 72.33% respectively. Nickel and manganese concentration load observed in mine water was 0.379 mg/L and 2.010 mg/L in 100 mm of water column height; 0.382 mg/L and 2.200 mg/L in 150 mm of water column height and 0.402 mg/L and 2.220 mg/L in 200mm of water column height respectively. In 24 hours of retention period, in constructed wetland containing *D. bipinnata* with organic substrate the nickel concentration dropped to 0.137 mg/L, 0.253 mg/L and 0.283 mg/L whereas manganese concentration dropped to 0.747mg/L, 1.410 mg/L and 1.633mg/L with different water column heights. The percentage removal efficiency of nickel was 29.60% to 63.76% and manganese concentration was 35.90% to 76.44%.

Conclusion

The pollutants removal mechanism in wetlands is through physical, chemical and various biological processes which involve sedimentation, settling, filtration, adsorption, precipitation, co-precipitation into insoluble compounds. It is, however, difficult to illustrate what actually occurs or which reactions takes place in the wetland. The entire processes are dependent on each other, thus making the whole process of pollutants removal mechanisms very complex. More or less, the extent to which these reactions occur depends on the composition of the substrate, sediment pH, nature of wastewater and plant species (Sheoran and Sheoran, 2006).

This study revealed that the height of water column in the wetland test cells does not play any significant role. It was also observed that most of the pollutants were removed within 24 hours of retention period itself. The system seems to work efficiently probably also due to high temperatures in arid climate of Rajasthan, India. Further retention period up to 168 hours revealed that no significant changes occurs (proportionate to retention period). An efficient removal of pollutants was achieved and there residual concentration in the wetland test cells were decreased below the relevant permissible level (Table 5) subscribed by Bureau of Indian Standards and Ministry of Environment and Forests (MOEF) after 24 hours of retention period except lead where input concentration was high.

Substrate containing powdered goat manure with wood shavings accelerated sulphate reduction leading to increase in pH and heavy metal removal from the AMD as metal. This addition of biodegradable organic substrate provides carbon for anaerobic alkalinity generating processes. The metal removal is associated with reducing conditions and is accelerated by the addition of readily degradable organic amendments, which feed the microbial processes. Also method, which involves indigenous plant species, needs to be encouraged. Plants seem to remove metals by uptake or oxidative precipitation near the roots. Although plants have been reported to account for only a small percentage of the metal removal capacity of the wetland treatment systems. Adsorption of metals to the organic substrates of the treatment systems can result in metal removal. It might be feasible to stimulate bacterial sulphate reduction in in-situ water filled mines and open pits. Also this was a preliminary study, a detailed study is needed to investigate the criteria for proposed design of constructed wetlands and choice of aquatic plants to treat industrial and other domestic waste water in India. For a developing country like India natural treatment is required by constructed wetlands, which have minimum energy input, relatively simple operational and with minimum maintenance, for which even local people can be easily trained. Thus constructed wetland being a cost effective alternative for mine water treatment can singly or in combination with other treatment systems is highly commendable.

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