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Comparative Assessment of Nutrient Mobilisation and Heavy Metal Indices in Chronosequence of Manganese Mine Spoil

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ABSTRACT

Mine tailings are generated during mineral extraction that greatly influence soil properties and provide a least opportunity for vegetation. In current study, accumulation of available nutrients and their effect on concentration of heavy metals were assessed from a chronosequence of manganese mine spoil (MBO0, MBO2, MBO4, MBO6, MBO8, MBO10) and compared it with native forest soil. In mine spoil, required level of nutrients were less than those in native forest soil. Heavy metal content was found to be high in fresh overburden spoil as compared to native forest soil. Fresh overburden mine spoil id acidic in nature due to high accumulation of Mn and Fe metals. However, in course of time, the acidic nature of mine spoil turns into basic due to the improvement in the soil physico-chemical characters. In chronosequence overburden spoil shows increasing trend in soil organic carbon, electrical conductivity and cation exchange capacity leading towards rapid reclamation.

Key words: Chronosequence, overburden mine spoil, vegetation, electrical conductivity, cation exchange capacity, reclamation

INTRODUCTION

Open cast manganese mining poses severe environmental threat causes global concern from few decades and generate large amount of mine spoil from overburden dumps. Mine spoil predominated by rock fragments from top soil and manganese residues generated during mining process (Mitchell 1959). Moreover, mine spoil majorly affects the physico-chemical properties of soil and act as a negative control to assess fertility status of soil and also causes demolition of natural vegetation in the degraded area. For the assessment of soil quality the selection of vital soil attributes can be used as sensitive indicators of important soil processes and functions. So to understand the quality of the individual mine spoil, it can be compared with nearby forest soil or through periodic observation of changes over time (Larson and Pierce 1994).

Heavy metals presents in mine spoil destroys the exquisite of landscape also interferes soil constituents like soil horizon and structure, microbial interactions, cycling of nutrients that are pivotal for sustentation of healthy ecosystem and consequently results in obliteration of well functioned vegetation and soil profile (Kundu and Ghosh 1997**).** The tailings contains detrimental factors such as uplifting heavy metals, increasing sand content, low moisture content, accelerates compaction of soil, lowering of organic matter. Overburden dumps with acidic nature unleash sulphide materials or salts that creates acid mine drainage (Ghose 2005, Mensah et al. 2015).Therefore, implacable constraints must be there for the implantation of vegetation in mine spoil (Hossner and Hons 1992). The lack of vegetation in mine spoil, responsible for acidification of neighbouring soils by leaching creates high environmental contamination (Ghose 2005, Vega et al. 2004). Restoration of degraded landmass anticipated to be assured sustainable environment while favouring livelihood of landowners. According to Mborah et al. (2016) and Bansah and Addo (2016) different uses of reclaimed land after post mining land provide adequately to enhance land use capacity and built a stable ecosystem. Moreover, during reclamation phase, wastages arise from mines blend with surface soil results in formation of irreversible and persistent metals in reclaimed land. Further, deposition of heavy metals by crossing threshold level poses contamination of soil and diminishes soil fertility, ultimately affecting soil productivity that is detrimental to human health (Tang et al. 2018). The other factors are extraction of metal ores that commonly poses a multiple metal pollution to the environment (Dudka et al. 1997). The recuperation of mine spoils, with relatively high toxic metal levels, acquire major challenge, because these trace elements are persistently conferring high concentrations and hinders the revegetation in disturbed sites (Vega et al. 2004). In addition, heavy metals when present excessively, decreases the absorption of nutrients and water by plants, respiration of roots and also hampers mitotic division in meristematic regions of roots. Factors like age of plant pH, cation exchange capacity and organic matter content also affects metal absorption by plant (Adriano 1986). Restoration is a process where the effect of mining on the environment can be revitalized. There-fore, it is urgent need to develop separate operational management for restoration of disturbed areas and ultimately post-mining land utilization strategies (Chaubey et al. 2012). The objective of the present study was to analyse available nutrient flux and effect on heavy metal concentration in chronosequence manganese mine spoil. The current investigation was focuses on to quantify the heavy metal concentration and relative contamination in different chronosequence manganese mine spoil that helps to convert an unproductive landmass into a productive land.

MATERIALS AND METHODS

Site description

The study was carried at Kanther Manganese mines, Koira which is situated in the revenue district of Sundargarh, Odisha, India. The area is located between 85°20’09.67" east longitude and 21°53’45.34" north latitude. The entire area is maintained by Rungta mines Ltd. includes 73.653 ha lease area that is under observation and is located at 1,940 ft. altitude. A part of the lease area falls under Bhani Pahar reserve forest.

Sampling

Sampling was done in accordance with the general methods for soil microbiological study (Parkinson 1971). Each site was divided into three blocks and during each sampling five soil samples were collected randomly from 0-15 cm soil depth by digging pits of 15 x 15 x 15 cm in each block and referred to as sub-samples, ultimately set up composite sample by blending all together thoroughly and stored in polythene bag and subjected to sieving (2 mm mesh size) for characterization and keep it for future reference after grinding. For the present study, sampling sites were fresh manganese mine spoil (represented as MBO0), two year (MBO2), four year (MBO4), six year (MBO6), eight year (MBO8) and ten year (MBO10) old mine spoil dumps. Forest soil (FS) was taken as undisturbed control.

Physico-chemical characterization

In physico-chemical analysis bulk density was determined by following the TSBF Handbook (Anderson and Ingram 1992). The moisture content and water holding capacity was done according to the methods of Mishra (1968). Measurement of soil pH (1:2.5 ratio of soil: water) was done using digital pH meter to check the acidity or basicity of overburden mine soil. Electrical conductivity was determined by using glass electrode method (Jumkis 1965). Titration method was used for estimation of soil organic carbon (Walkley and Black 1934) and Kjeldahl method for total nitrogen (Jackson 1958). Olsen method was followed for determination of available phosphorous and potassium content in different mine overburden spoil samples (Olsen and Sommers 1982). Exchangeable cations were estimated by ammonium acetate methods (Trivedy and Gudekar 1987).

Estimation of heavy metals

Total dissolved Cd, Cr, Cu, Co, Ni, Pb, Mn, Mo, Fe, Zn were extracted from chronosequence mine spoil using methods suggested by Houba et al. (2000). Metals were extracted by means of acid digestion in a mixture of concentrated nitric, hydrochloric and hydrofluoric acids (1:3:3 v/v) (Marcet et al. 1997) and quantified by inductively coupled plasma-atomic emission spectrometry (ICP-AES).

Statistical Analysis

All the experiments were done in triplicate and the experimental data obtained were analysed for statistical significance of means using the programme SPSS version 16.0 for simple correlations (Norussis 1992). Analysis of variance (ANOVA) was performed to know significant difference. Multiple regression and principal component analysis for different soil profile were done using STRATA 15X 64.

RESULTS AND DISCUSSION

In the present study, degree of variability with respect to different soil biochemical properties and its influence on heavy metal concentrations were analysed from as chronosequence manganese mine spoil. Soil pH is an important index of ecological health of terrestrial surface (Rai et al. 2011). The pH value of chronosequence manganese mine spoil ranges from 5.79 ±0.035 (MBO0) to 7.15±0.05 (MBO10), which is slight acidic to neutral in nature (Table 1). Soil pH of native forest is more than of pH of overburden mine spoil. The elevation of pH in natural forest soil as compare to mine spoil is due to high organic matter (3.469±0.11, $r = 0.975$; $p<0.001$) accumulation that shown positive correlation and assists in shifting of mine soil from acidic to basic nature by modifying soil pH (Table 1). Electrical conductivity increased from fresh mine spoil (0.31±0.1 mS/cm) to native forest soil (0.68±0.013 mS/cm). Lower electrical conductivity is responsible for lower availability of organic carbon and exchangeable cations in the positive change in EC along the chronosequence mine spoil is due to accumulation of organic matter and improvement in availability of more macropores, ultimately increasing water holding capacity. Exchangeable cations assimilate on the soil surface that can able to exchange with other cations in solution. Wilson and Tilman (2002) reported that improvement in soil physico-chemical properties is due to the improvement in exchangeable calcium concentration. In the present study, concentration of Ca (0.0299±0.002 cmol(+)/kg), and Mg (0.0468 ± 0.001 cmol(+)/kg) in mine spoil is lower than native forest soil (Ca and Mg 0.3173±0.006 and 0.2287±0.003 cmol(+)/kg, respectively). Concentration of monovalent cations (K and Na) did not show significant variation, but concentration of divalent cations (Ca and Mg) was significantly higher in overburden spoil and the native forest soil ($\alpha = 0.05$, Table 1). The reason behind this difference was that monovalent cations (K and Na) might have leached from the top soil to a large extent while the concentration of divalent cations (Ca and Mg) are strongly adsorbed to soil particles and elevating the moisture content of the soil. Mining activities such as stripping, stockpiling and reinstatement generally caused physical alterations of the top soil which further leads to nutrient loss (Sheoran et al. 2010).

Table 1. Variations in accumulation of physico-chemical parameters in chronosequence manganese mine spoil

Site	**pH**	**EC (mS/cm)**	**Exchangeable cations (cmol(+)/kg)**			
			Ca	**Mg**	**Na**	**K**
MBO (O)	5.79±0.035	0.31±0.01	0.0299±0.0002	0.0486±0.001	0.0139±0.0002	0.011± 0.0005
MBO(2)	6.09±0.089	0.36±0.001	0.0411± 0.0004	0.0587±0.0014	0.0132±0.0003	0.0075±0.005
MBO(4)	6.28±0.042	0.44±0.044	0.0511± 0.001	0.0648±0.0009	0.0110±0.0001	0.0096±0.001
MBO(6)	6.57±0.043	0.57±0.005	0.0694±0.0004	0.0795±0.0005	0.0092±0.0002	0.0092±0.0001
MBO(8)	6.78±0.05	0.59±0.005	0.0877± 0.002	0.0823±0.0006	0.0074±0.0004	0.0083± 0.0002
MBO(10)	7.15±0.05	0.63±0.026	0.1136± 0.002	0.115±0.003	0.0053±0.0002	0.0078±0.0001
NF	7.5±0.043	0.68±0.013	0.3173±0.006	0.2287±0.0003	0.0325± 0.003	0.0285±0.0004

In the present study, available nitrogen content showed increasing trend from fresh mine spoil (11.831± 0.556 mg/kg) to native forest soil (111.823± 0.699 mg/kg) (Table 2). Nitrogen assimilation was higher in case of native forest soil because soil N_2 is bound to organic matter leading to accumulation of more litter fall and microbial activity. Phosphorus plays an important role in storage of energy, photosynthesis, respiration, cellular division and enlargement and also acts as an important component of nucleic acid. Available phosphorus in the current study is in the range between 9.22±0.086 to 19.734±0.074 mg/kg in the chronosequence mine spoil as compared to 55.648±0.736 mg/kg in native forest soil. Available phosphorus is higher in native soils as ready availability of phosphorous due to high concentration of organic matter (0.950, $p<0.001$). Potassium content was low in mine spoil (2.308± 0.521 to 29.041± 0.061 mg/kg) as compare to native forest soil (45.838±0.638 mg/kg) and showed increasing trend because of aggregation of more organic matter (0.893, $p<0.001$) that helps to regulate more nutrient uptake to plant. Soil organic carbon in mine spoil is significantly very low 0.126% as compare to native forest soil 3.469%. The predominant source of organic carbon in native soil is dead and decaying organic matter, plant debris and residuals, falling leaves and litters. Low level of organic carbon in overburden dump accounts for dysfunctioning of ecosystem (Stark et al. 1997) and leading towards less availability of organic pool (Parkinson 1979). Cation exchange capacity (CEC) showed increasing pattern from mine spoil (4.458±0.048 Cmol(+)/kg) to native forest soil (6.617±0.016 Cmol(+)/kg) because of high available nutrient content (AN-111.82±0.699 mg/kg; AP-55.648±0.736 mg/kg; AK- 45.838±0.638 mg/kg) and organic matter 3.469%. Low cation exchange capacity results into less water holding capacity, reduced organic carbon and available nutrients (Bahrami et al. 2010).

Heavy metal concentration for the chronosequence mine spoil is presented in Table 3. The mean concentration of Zn ranged from 42.322±0.611 to 68.728±0.461 mg/kg, Fe from 65.827±0.424 to 42.551±0.579 mg/kg, Mn from 311.17±1.008 to 98.866±2.12 mg/kg, Cr from 67.731±0.577 to 50.929±0.567 mg/kg, Pb from 25.019±0.064 to 14.041±0.024 mg/kg, Cd from 1.79±0.069 to 0.787±0.034 mg/kg, Cu from 17.713± 0.225 to 35.438±0.956 mg/kg, Co from 13.641±0.383 to 9.187± 0.084 mg/kg, Mo from 42.247±0.356 to 25.533±0.623 mg/kg and Ni from 31.874±0.064 to 21.509± 0.598 mg/kg (significant at $p < 0.05$) as compared to the concentration of native forest soil (50.381±0.549 mg/kg for Zn, 38.158±0.855 mg/kg for Fe, 52.28±0.66 mg/kg for Mn, 48.673±0.582 mg/kg for Cr, 12.43±0.44 mg/kg for Pb, 1.064±0.04 mg/kg for Cd, 29.14±0.578 mg/kg for Cu, 14.457±0.201 mg/kg for Co, 24.613±1.411 mg/kg for Mo and 36.212±0.0850 mg/kg for Ni). While Mn had highest mean concentration (311.17±1.008 mg/kg for mine spoil, 52.28±0.66 mg/kg in native forest soil) followed by Fe (65.827±0.424 mg/kg in mine spoil 38.158±0.855 mg/kg in native soil), Cd was present in lowest concentration (1.79±0.069 mg/kg in mine spoil and 1.064±0.04 mg/ kg in native forest soil). High accumulation of heavy metals such as Mn, Fe (-0.987, -0.990, $r < 0.01$) in mine spoil cause acidification of soil due to change in pH towards acidity (Taha et al. 2017) and also due to leaching of basic cations like Mg^{2+} (Shown negative correlation with Mn (-0.854), Fe (-0.843), $r<0.01$) (Singh et al. 2020). Increment in the concentration of Mn in mine spoil supports the translocation of Fe to surface ($r = 0.983$, $p<0.01$) and elevating concentration of both Mn, Fe in mine spoil is negatively correlated with concentration Zn (-0.538, -0.540, respectively, $r<0.05$). Increased level of heavy metals such as Fe, Mn for prolonged period decrease the availability of Zn for reclamation and growth of crops although favour translocation (Sharon et al. 2014). Liu et al (2020) reported Mn concentration in excess might be the major reason for lowering of concentration Cu (-0.843, $r<0.01$) and Zn (-0.538, $r<0.05$) in soil, supports the trends in current study (Table 4) concentration in soil. But concentration of Cu, Zn, Mn, Fe (29.14±0.578 mg/kg for Cu, 50.381± 0.549 mg/kg for Zn, 29.14±0.578 mg/kg for Mn, 38.158±0.855 mg/kg for Fe) was less in native forest soil (á = 0.05) due to sorption and desorption and availability of organic matter. These metals might be making complex and sequestered thus become inaccessible to plants (Chaubey et al. 2012). Fe, Mn ($r = -0.950, -0.962$, $r<0.01$) are negatively correlated with organic carbon and available nitrogen, phosphorus and potassium ($r = 0.997, 0.906, 0.981,$

Table 2. Change in available nutrients, organic matter and CEC concentration in chronosequence manganese mine spoil

Site	Available nutrients			Org C (%)	CEC (cmol(+)/kg)
	N (mg/kg)	P (mg/kg)	K (mg/kg)		
MBO(0)	11.831± 0.556	9.22±0.086	2.308± 0.521	0.126± 0.002	4.458±0.048
MBO(2)	21.357±0.506	10.714± 0.339	10.571±0.556	0.312±0.014	4.802±0.029
MBO(4)	31.506± 0.516	12.204± 0.1105	16.864± 0.575	0.626±0.112	4.958±0.044
MBO(6)	45.915± 0.014	14.52± 0.203	20.973±0.582	1.247±0.035	5.193±0.057
MBO(8)	62.014±0.703	16.393± 0.113	24.704±0.521	1.687±0.048	5.645±0.05
MBO(10)	72.445± 1.528	19.734±0.472	29.041±0.061	2.048±0.09	5.974±0.034
NF	111.82± 0.699	55.648± 0.736	45.838±0.638	3.469±0.11	6.617±0.016

Table 3. Variations in heavy metal concentration (mg/kg) in different chronosequence manganese mine spoil

Site	**Zn**	**Fe**	**Mn**	**Cr**	**Pb**	**Cd**	**Cu**	**Co**	**Mo**	**Ni**
MBO	42.322± 0.611	65.827± 0.424	311.17± 1.008	67.731± 0.577	25.019±0.064	1.79±0.069	17.713± 0.225	13.641± 0.383	42.247± 0.356	31.874± 0.064
MBO 2	52.86± 0.397	64.406± 0.562	250.73± 1.404	64.368± 0.629	21.016±1.171	1.53± 0.060	19.374± 0.457	12.811± 0.090	40.187± 0.51	30.722± 0.154
MBO4	56.11± 0.804	57.77± 1.05	201.8± 0.754	59.578±	18.834± 0.078	1.19± 0.035	21.742±1.022	11.370± 0.54	35.28± 1.108	28.481± 0.156
MBO6	58.660± 0.608	51.681± 0.599	191± 0.65	57.020± 0.055	16.60± 0.307	0.953±0.342	26.938± 0.854	11.266± 0.62	32.536±0.517	27.363± 0.1505
MBO8	62.81± 1.0210	48.916± 0.486	155.8± 0.55	54.24± 0.133	15.77±0.27	0.839± 0.002	30.876±0.854	10.211± 0.105	29.563±0.469	25.181± 0.121
MBO10	68.728± 0.461	42.551± 0.579	98.866± 2.12	50.929± 0.567	14.041± 0.024	0.787± 0.034	35.438±0.956	9.187± 0.084	25.533±0.623	21.509± 0.598
NF	50.381± 0.549	38.158± 0.855	52.28± 0.66	48.673± 0.582	12.43± 0.44	1.064± 0.04	29.144± 0.578	14.457± 0.201	24.613±1.411	36.212± 0.0850

Table 4. Correlations (r) among elements in chronosequence manganese mine spoil

	Na	Ca	Mg	K	PH	EC	CEC	OC	AN	AP	AK	Zn	Fe	Mn	Cr	Cu
Na	1															
Ca	0.657**	1														
Mg	0.760**	0.720**	1													
K	0.926**	0.719**	0.860**	1												
pH	0.390	0.575**	0.884**	0.577**	1											
EC	0.201	0.427	0.753**	0.419	0.950**	1										
CEC	0.450*	0.616**	0.911**	0.617**	0.990**	0.924**	1									
OC	0.530*	0.622**	0.943**	0.700**	0.976**	0.916**	0.986**	1								
AN	0.513*	0.622**	0.937**	0.681**	0.983**	0.918**	0.992**	0.997**	1							
AP	0.836**	0.737**	0.989**	0.913**	0.824**	0.685**	0.860**	0.906**	0.896**	1						
AK	0.503*	0.621**	0.925**	0.662**	0.980**	0.926**	0.981**	0.981**	0.987**	0.884**	1					
Zn	-0.580**	-0.081	0.036	-0.404	0.484*	0.602**	0.417	0.309	0.345	-0.084	0.377	1				
Fe	-0.311	-0.516*	-0.843**	-0.510*	-0.990**	-0.973**	-0.977**	-0.962**	-0.969**	-0.774**	-0.964**	-0.540*	1			
Mn	-0.344	-0.553**	-0.854**	-0.523*	-0.987**	-0.945**	-0.978**	-0.950**	-0.965**	-0.789**	-0.977**	-0.538*	0.983**	1		
Cr	-0.241	-0.454*	-0.795**	-0.445*	-0.978**	-0.978**	-0.959**	-0.935**	-0.946**	-0.724**	-0.955**	-0.606**	0.988**	0.986**	1	
Cu	-0.138	0.247	0.524*	0.107	0.838**	0.888**	0.808**	0.749**	0.769**	0.417	0.747**	0.803**	-0.883**	-0.843**	-0.891**	1

** significant at 0.01 level;*Significant at 0.05 level; EC- Electrical conductivity, CEC- Cation exchange capacity, OC- Organic carbon, AN- Available nitrogen, AP- Available phosphorus, AK- Available potassium, Zn- Zinc, Fe- Iron, Mn- Manganese, Cr- Chromium, Cu- Copper

$r<0.01$) (Table 4), are positively correlated with organic carbon. This leads towards high EC and CEC that might be responsible to enhance porosity in soil and help in increasing availability of nutrients, more microbial activity, more access of litters from vegetation through time and boost reclamation process (Mukhopadhyay et al. 2016, Swain et al. 2023). There is no significant difference observed in concentration of Cr, Pb, Cd, Co, Mo, Ni in both mine spoil and native forest soil.

The stepwise multiple regression analysis using OC, AN, AP as dependent variables are given in Table 5. A positive relation between OC and PH (95.24%, $p < 0.001$) has been reported by Juwarkar et al. (2016) and Swain et al. (2023). Negative correlation was observed by accumulations of Mn (90.5%, $P<0.001$) with organic matter (Silva et al. 2019, Juwarkar et al. 2016) and additional 2.28% variability in OC was contributed when Fe as second variable was used. The contribution of EC in variability with respect to OC was 83.97%, shows positive correlation reported by Sadhu et al. (2012) and 13.18% variability was contributed by CEC as second variable (Raghunathan et al. 2021).

The step wise multiple regression analysis also explained the positive correlation between AN and pH (96.62%, $p < 0.001$), where as AN negatively regulated by contribution of heavy metals such as Mn (93.06, $p < 0.001$) has been reported by Juwarkar (2009).Contribution of EC in variability with respect to AN was about 84.29% as first variable, and 14.16% was contributed by CEC as second variable (Mukhopadhyay et al. 2016, Swain et al. 2023). Multiple regression analysis for AP as dependent variable was explained with respect to pH was 67.83% and positively related to EC (46.88%, $p < 0.001$). Accumulation of metal Mn is negatively regulated (62.30 %, $p <$

Table 5. Stepwise multiple regression analysis of OC, AP, AN with soil physico-chemical characteristics and heavy metal accumulation in manganese mines spoil

Element	Equations	R^2
OC	-10.937+1.861pH	0.9524
	3.6424-0.0126Mn	0.9033
	-6.06307-0.0019Mn - 0.0936Fe	0.9261
	-2.4789+7.465EC	0.8397
	-6.784+1.481CEC+0.0341EC	0.9761
	0.6371+6.0896Ca	0.3874
	-0.3666+17.814Mg	0.8902
AN	-312.991+55.107pH	0.9662
	119.098-0.3778Mn	0.9306
	184.897-2.0621Fe-0.1425Mn	0.9434
	-62.027+219.7855EC	0.8429
	193.159+45.1200CEC+ 2.847EC	0.9845
	0.613+519.85Mg	0.8778
AP	-123.9391+21.7589pH	0.6783
	7.934+99.835Ca	0.5429
	-5.174+248.917Mg+7.018Ca	0.9798
	-19.942+77.246EC	0.4688
	-117.425-33.542CEC-84.024EC	0.8212
	46.040-0.145Mn	0.6230

0.001) with respect to AP Swain et al. (2023) (Table 5).

Further in order to view the variations among the different soil profiles, principal component analysis was performed to differentiate sites on the basis of accumulation of physico-chemical properties and heavy metal concentrations. Principal component analysis indicates Z1 and Z2 component that signifies maximum variance with respect to physico-chemical characteristics and heavy metal accumulations. The cumulative percentage was quantified to be 96% that can differentiate variation among chronosequence manganese mine spoil into different clusters (Pandey et al. 2016, Kujur and Patel 2012) (Fig. 1).

CONCLUSIONS

Acidic nature of soil in overburden not only affects vegetation but also reduced the availability of nutrients for plant growth. The average value of organic carbon is much higher in natural forest soil than mine spoils indicating negative effect of mining activities on the quality of soil. Cation exchange capacity and concentration of exchangeable cations are also disturbed by mining activity. Acidic soil favours accumulation of metals like Mn, Fe in soil, excess concentration of which causes drastic change in soil properties. Thus, for rehabilitation of mine spoil, quality of soil should be checked periodically by using suitable soil indicators that ultimately assist in development of suitable post-mining strategies to generate successional revegetation in degraded mined land.

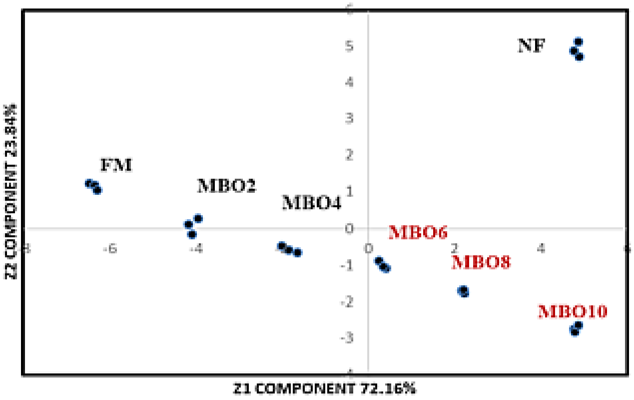

Figure 1. Principal component analysis based on available nutrients and heavy metal concentration level in chronosequence manganese mine spoil

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Authors'contributions: Samapika Dash carried out the field works, collection of samples, executed all the experimental work, analysed the data as a part of project work and drafted the manuscript. Monty Kujur designed the project, supervised and analysed the research work. Both the authors equally contributed to finalise the manuscript before submission.

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