

Contamination of Heavy Metals and Nutrients in Sediment, Sludge and Sewage of India

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Abstract

The stagnant water bodies in India are sink for contaminant *i.e.* detergent, fertilizer, nutrients, heavy metal, pesticide, microbe, etc. The contamination and sources of elements *i.e.* Al, K, P, S, Cl, As, Ca, Sr, Ba, Ti, V, Cr, Mn, Fe, Ni, Cu, Zn and Pb in the sediment, sludge and sewage materials of the most industrialized cities of central India *i.e.* Raipur, Bhilai and Korba is described. The dominated metals *i.e.* Al, K, Ca, Ti, Fe and Mn in the geowaste materials (n = 20) contributed in the range of 4.8% - 36.3% with mean value of 10.2% ± 2.9%. The Σ_4 concentration of nutrients *i.e.* P, K, S and Cl ranged from 1.2 - 12.5 g/kg with mean value of 7.9 ± 1.3 g/kg. The concentration of other heavy metals (HMs) *i.e.* As, V, Cr, Ni, Cu, Zn and Pb ranged from 12 - 105, 35 - 175, 88 - 392, 14 - 77, 32 - 185, 38 - 626 and 18 - 228 mg/kg with mean value of 644 ± 78, 83 ± 15, 182 ± 41, 44 ± 7, 68 ± 18, 199 ± 71 and 85 ± 25 mg/kg, respectively. The spatial and vertical distribution, enrichment and sources of the elements in the sediments are discussed.

Keywords

Heavy Metal, Nutrients, Contamination, Sources, Sediment

1. Introduction

The sediment consists of soil, pebbles, silt, clay and other materials [1]. The bottom of oceans, rivers, streams,

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lakes, ponds and mudflats is made up of sediment. The problem of sediment contamination is increasing in many areas throughout the world [2]. Sediment is often a rich source of food for the living creatures in water. Many of the sediments in our rivers, lakes, and oceans have been contaminated by pollutants *i.e.* black carbon, heavy metals, polychlorinated biphenyls (PCBs), dioxins and organochlorine pesticides, etc. [3]-[8]. Many heavy metals (HMs) cause damage to the liver, kidneys, blood circulation, nerves, and may also be a trigger of cancer [9] [10]. The HMs contaminations were reported in sediments of major rivers of the world [11]-[20]. The sludge, sewage, litter, marine boat traffic, and runoff waters are all potential sources for the HMs contamination of sediments [21]-[30]. In the present work, the contamination of heavy metals (*i.e.* As, V, Cr, Mn, Fe, Ni, Cu, Zn and Pb) and nutrients (*i.e.* P, K, S and Cl) in the sediment, sludge and sewage of Raipur, central India is described.

2. Materials and Methods

2.1. Study Area

Three most industrial cities of Chhattisgarh state namely: Raipur (21°23'N, 81°63'E), Bilai (21°18'N, 81°28'E) and Korba (22°21'N, 82°40'E) were selected for the proposed investigation. Raipur is the capital city of the Chhattisgarh state, India with population of ≈ 2.0 million. The Raipur city and its neighborhood are now becoming an important regional commercial and industrial destination for the coal, power, steel and aluminum industries. Raipur is one of biggest iron and cement market in the country. Bilai is the second-largest city inclusive of Durg city in Chhattisgarh with population of ≈ 1 million, and is located in the west of Raipur ≈ 22 km away. The town is famous for the operation of one of the largest steel plant in the World (capacity: 3.15 MT/Yr). Korba is another city in Chhattisgarh with ≈ 0.5 million population, famous for power supply and aluminum plant.

2.2. Sample Collection

The samples were collected using a stainless-steel scoop in the summer, 2012 from 20 locations of industrial area of Chhattisgarh, **Figure 1**. The sediment samples were collected from 18 ponds of cities: Bilai, Raipur and Korba. The sludge and sewage were collected from the industrial site (*i.e.* Siltara) of Raipur city. They were kept in glass bottle (250 mL) and dried at 30°C in an oven for overnight. The samples were crushed into fine particles by mortar and sieved out the particles of mesh size < 0.1 mm. The samples were stored in aluminum foil.

2.3. Analysis

A 5.0 g of the sample was extracted with 25 mL distilled water for 12 hr. The extract was decanted out for the pH value measurement. The CHNSO-IRMS Analyzer by SV Instruments Analytica Pvt. Ltd. was used for quantification of the total carbon (TC). The total carbon (TC = BC + OC + CC) in the soil sample was oxidized at 1020°C with O₂ in constant helium flow stream by detecting the resulting CO₂ gas with a thermal conductivity detector. The H₃PO₄ (10 drops) treated soil sample was oxidized with O₂ at 1020°C in a similar way for determination of BC and OC contents. The OC was analyzed by titration method using K₂Cr₂O₇ as oxidant [31]. The CC content in the soil was evaluated by subtracting the BC and OC content to the TC.

The Bruker S2-Picofox TXRF portable spectrometer equipped with poly capillary lens and the X-ray beam was used for the characterization of elements in the sediment samples. A suspended solution was prepared by mixing sample (10 mg) with solution (10 mL) containing triton 1% (w/v) and Ga 10 µg/mL in ultrasonic bath for 15 min. A 10 µL of sample solution was sprayed on the quartz filter by subsequent drying. The X-ray source was focused on the filter for quantification of the elements. The peak area of the signal was computed. Three replicate measurements for each sample were carried out. The content of 18 elements (*i.e.* Cl, S, P, K, Al, Ca, Sr, Ba, As, Ti, V, Cr, Mn, Fe, Ni, Cu, Zn and Pb) in each sample was analyzed.

2.4. Pollution Indices

The enrichment factor (E_f) was used to determine metal contamination in the geowaste materials with respect to the base line concentration. The concentration ratio of an element, X, to a crustal element (e.g. Al) in the soil sample to the $\left\{ \frac{[X]}{[Al]} \right\}$ ratio in the earth's crust was normalised by using following indices [32] [33].

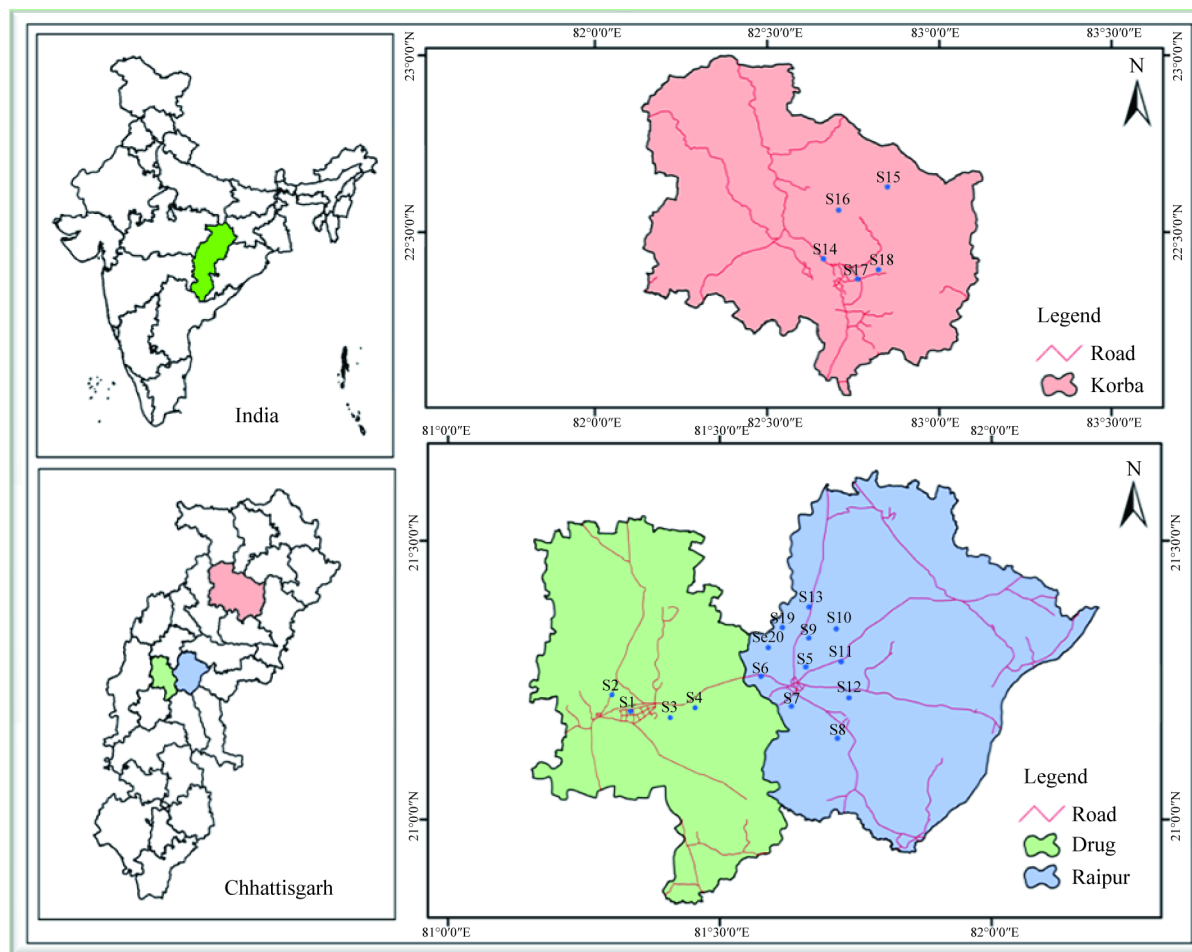


Figure 1. Representation of sampling locations in Raipur, Bilai and Korba city, India.

$$E_f = \left\{ [X_s] / [Al_s] \right\} / \left\{ [X_e] / [Al_e] \right\}$$

where, symbols: X_s , X_e , Al_s and Al_e denote concentration of metal and Al in the soil and earth crust, respectively.

3. Results and Discussion

3.1. pH of Extract

The color of sediments was varied from reddish (R) to black (B) due to deposition of the BC, metal oxides and organic residues. The pH value of the extract ($n = 20$) was ranged from 6.2 - 8.2 with mean value of 7.4 ± 0.3 . The lower pH values of the SeK, SIR and SwR was observed due to presence of higher content of chlorine and sulfur, **Figure 2**.

3.2. Carbon Content

The carbon content of the geowaste materials is presented in **Table 1**. The concentration of BC, OC and CC was ranged from 4.87 - 9.62, 0.07 - 0.32 and 0.05% - 0.17% with mean value of 8.12 ± 0.57 , 0.15 ± 0.02 and $0.11\% \pm 0.01\%$, respectively. They were found to correlate well ($r = 0.72$), indicating origin from the similar sources *i.e.* burning processes. The comparable content of the BC and CC in all types of waste materials were marked, **Figure 3**. The significantly higher content of OC in the sewage was seen due to input by the organic waste, **Figure 3**. The BC content in the sediment of the studied area was observed to be higher than in of other regions, probably due to huge coal burning in this region [34]-[36].

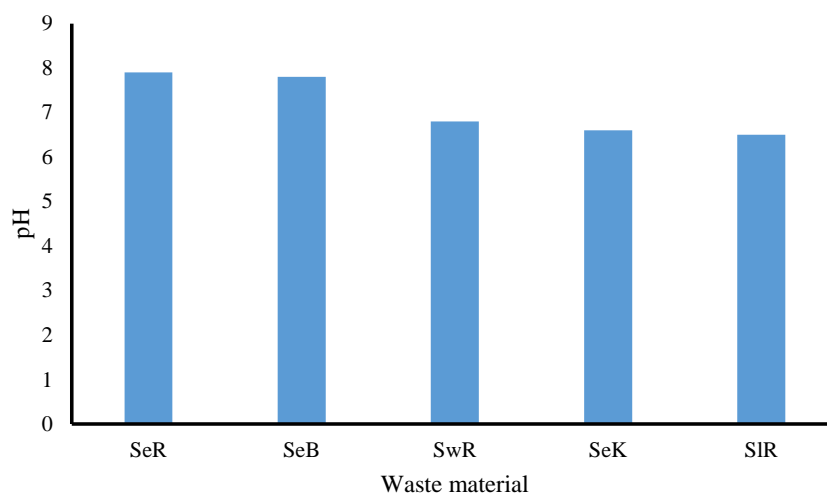


Figure 2. pH variation of sediment, sludge and sewage, Se = Sediment, SI = Sludge, Sw = Sewage, R = Raipur, B = Bhilai, K = Korba.

Table 1. Acidity and carbon content of waste materials.

S. No.	City	Pond	Color	pH	BC, %	OC, %	CC, %
SeB1	Bhilai	Sector-7	B	7.9	9.62	0.14	0.12
SeB2	Bhilai	Sector-11	B	7.8	4.87	0.07	0.05
SeB3	Bhilai	Purena	DB	8.1	7.12	0.10	0.06
SeB4	Bhilai	Bhilai-3	LG	7.6	5.45	0.07	0.07
SeR5	Raipur	Sarora	B	7.8	8.11	0.16	0.11
SeR6	Raipur	Ashi	LB	7.9	8.43	0.12	0.10
SeR7	Raipur	Budheshwar	R	7.8	9.33	0.18	0.13
SeR8	Raipur	Vivekanand	LB	8.0	9.43	0.16	0.13
SeR9	Raipur	Birgoan	R	8.0	8.43	0.14	0.12
SeR10	Raipur	Urkura	LB	8.2	9.01	0.13	0.13
SeR11	Raipur	Pandri	B	7.9	8.89	0.17	0.12
SeR12	Raipur	Raja	R	7.9	8.94	0.16	0.11
SeR13	Raipur	Siltara	R	7.6	9.32	0.17	0.15
SeK14	Korba	Jalgoan	R	6.9	8.76	0.14	0.10
SeK15	Korba	Survari	B	6.7	8.54	0.16	0.12
SeK16	Korba	Dabri	LB	6.2	6.89	0.11	0.05
SeK17	Korba	Risdi	B	6.3	8.56	0.14	0.12
SeK18	Korba	Jhagraha	B	6.7	7.89	0.14	0.09
SiR19	Raipur	Birgoan	B	6.5	8.15	0.17	0.17
SwR20	Raipur	Birgoan	LB	6.8	6.58	0.32	0.11

Se = Sediment, SI = Sludge, Sw = Sewage, B = Black, DB = Deep back, LB = Light black, R = Reddish.

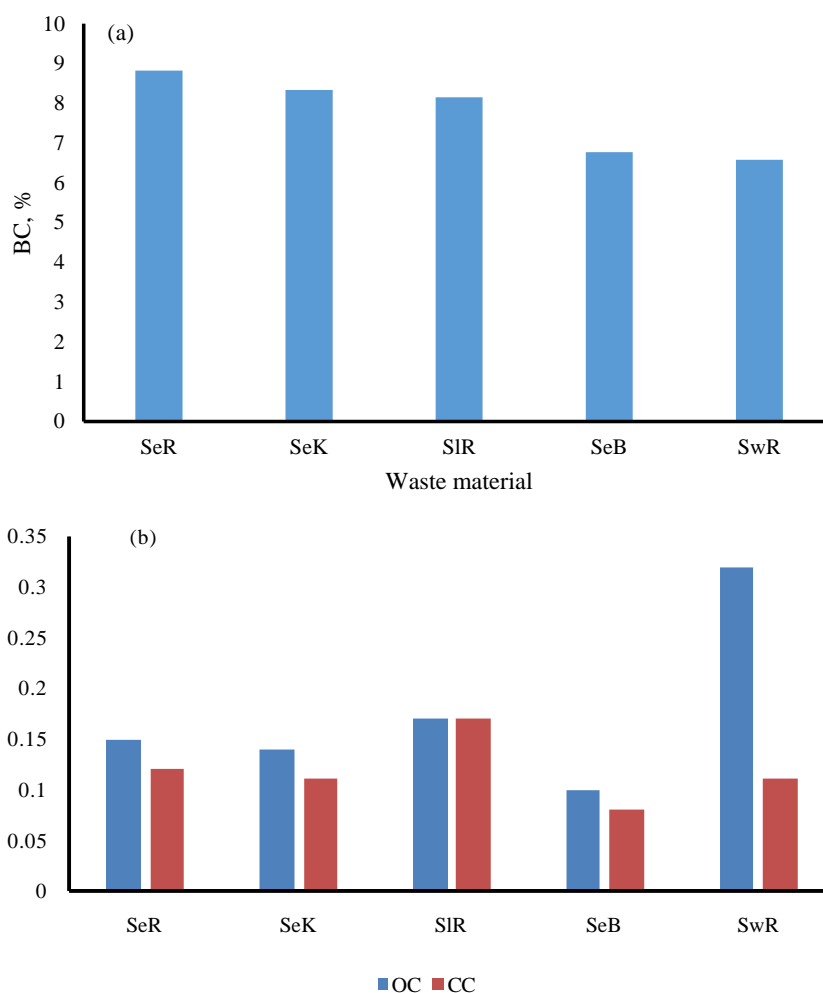


Figure 3. Spatial variations of carbon contents in sediment, sludge and sewage.

3.3. Content of Heavy Metals and Nutrients

The elevated concentration of metals *i.e.* Al, K, Ca, Ti, Fe and Mn in the geowaste materials was observed, ranging ($n = 20$) from 4864 - 18806, 593 - 11179, 579 - 72184, 2135 - 8121, 9737 - 319848 and 127 - 25614 mg/kg with mean value of 11088 ± 1851 , 6874 ± 1304 , 18268 ± 9468 , 4116 ± 671 , 73262 ± 34589 and 2782 ± 2484 mg/kg, respectively, **Table 2**. Among them, the highest Fe content was observed in all geowaste materials due to roasting of iron ores and huge coal burning in the studied areas. The concentration of P, S, Cl, As, Sr, Ba, V, Cr, Ni, Cu, Zn and Pb was marked in the sediments at traces, ranging from 95 - 584, 91 - 1822, 31 - 1117, 12 - 105, 40 - 135, 478 - 1078, 35 - 175, 88 - 392, 14 - 77, 32 - 185, 38 - 626 and 18 - 228 mg/kg with mean value of 232 ± 63 , 604 ± 240 , 231 ± 119 , 39 ± 14 , 79 ± 12 , 644 ± 78 , 83 ± 15 , 182 ± 41 , 44 ± 7 , 68 ± 18 , 199 ± 71 and 85 ± 25 mg/kg, respectively, **Table 2**.

The higher content of elements *i.e.* P, S, Ca, Ba, Cr, Fe, Cu and Pb was observed in the sediment of Bhilai city, due to input by the Steel plant effluents, **Figure 4**. Similarly, higher content of other eight elements *i.e.* Al, K, As, Sr, Ti, V, Ni and Zn was marked in Korba city due to huge coal burning by the Thermal power plants, **Figure 4**.

The content of elements *i.e.* Al, As, Ni, Cu, V, Ti and K was increased as the depth profile of the sediment was increased from 0 to 30 cm, may be due to their less binding with the organic materials, **Figure 5**. The reverse trend was marked with other elements *i.e.* Cl, S, Ca, Ba, Sr, Cr, Mn, Fe, Zn and Pb, may be due to higher adsorption with the organic materials, **Figure 5**.

The content of HMs and nutrient in the sediment, sludge and sewage wastes of Raipur city is presented in

Table 2. (a) Concentration of element in waste materials, mg/kg; (b) Concentration of element in waste materials, mg/kg.

(a)										
Element	SeB1	SeB2	SeB3	SeB4	SeR5	SeR6	SeR7	SeR8	SeR9	SeR10
Al	8756	17562	15960	7687	11147	7475	4864	9935	5691	10815
P	304	261	124	561	147	116	225	372	116	117
S	241	839	723	1750	360	412	1822	100	244	224
Cl	91	222	106	337	119	86	129	124	293	43
K	593	11179	7118	7775	7358	5354	5145	9233	5742	6693
Ca	72184	12831	11836	15412	8601	4566	17131	15052	12051	3426
Ti	3683	4934	4275	3045	3758	4392	2714	3765	3688	4448
V	40	131	110	78	72	66	45	66	72	75
Cr	362	321	184	214	138	100	110	117	392	148
Mn	8029	2595	1977	1309	1304	795	764	742	2522	741
Fe	31985	77874	68606	44268	56025	31841	35325	42657	56612	33595
Ni	14	71	56	39	40	33	35	31	62	49
Cu	93	74	50	87	89	32	46	75	58	34
Zn	273	171	87	150	149	38	77	156	103	50
As	12	24	21	21	22	18	20	19	18	28
Sr	82	78	59	104	48	44	64	58	47	50
Ba	546	821	568	1078	549	478	517	560	580	533
Pb	228	72	43	150	80	21	31	63	23	34
(b)										
Element	SeR11	SeR12	SeR13	SeK14	SeK15	SeK16	SeK17	SeK18	SIR19	SwR20
Al	6551	10563	7859	16099	18806	16808	13805	13621	7962	9790
P	264	164	214	232	99	156	120	95	375	584
S	549	215	1585	91	343	1004	198	236	298	855
Cl	838	95	1117	31	140	181	123	143	95	298
K	2725	7726	5468	7937	6407	9812	9866	10526	911	9904
Ca	12808	5441	24545	25153	7221	4156	579	707	81166	30493
Ti	2135	4168	3762	4725	5934	4618	6109	8121	770	3278
V	35	62	57	87	116	104	131	175	78	61
Cr	88	118	200	106	180	108	110	125	325	191
Mn	2561	586	1816	709	127	248	287	338	4035	1096
Fe	48643	41653	117506	35266	9737	35429	43733	52435	267975	46215
Ni	23	39	52	38	77	53	54	49	15	52
Cu	164	35	57	57	41	49	37	47	57	185
Zn	580	66	170	626	136	182	150	171	211	429
As	17	22	29	100	77	89	92	105	27	28
Sr	135	40	96	95	106	70	78	104	86	133
Ba	1038	493	823	554	530	612	573	810	515	700
Pb	156	18	84	54	70	91	102	123	171	93

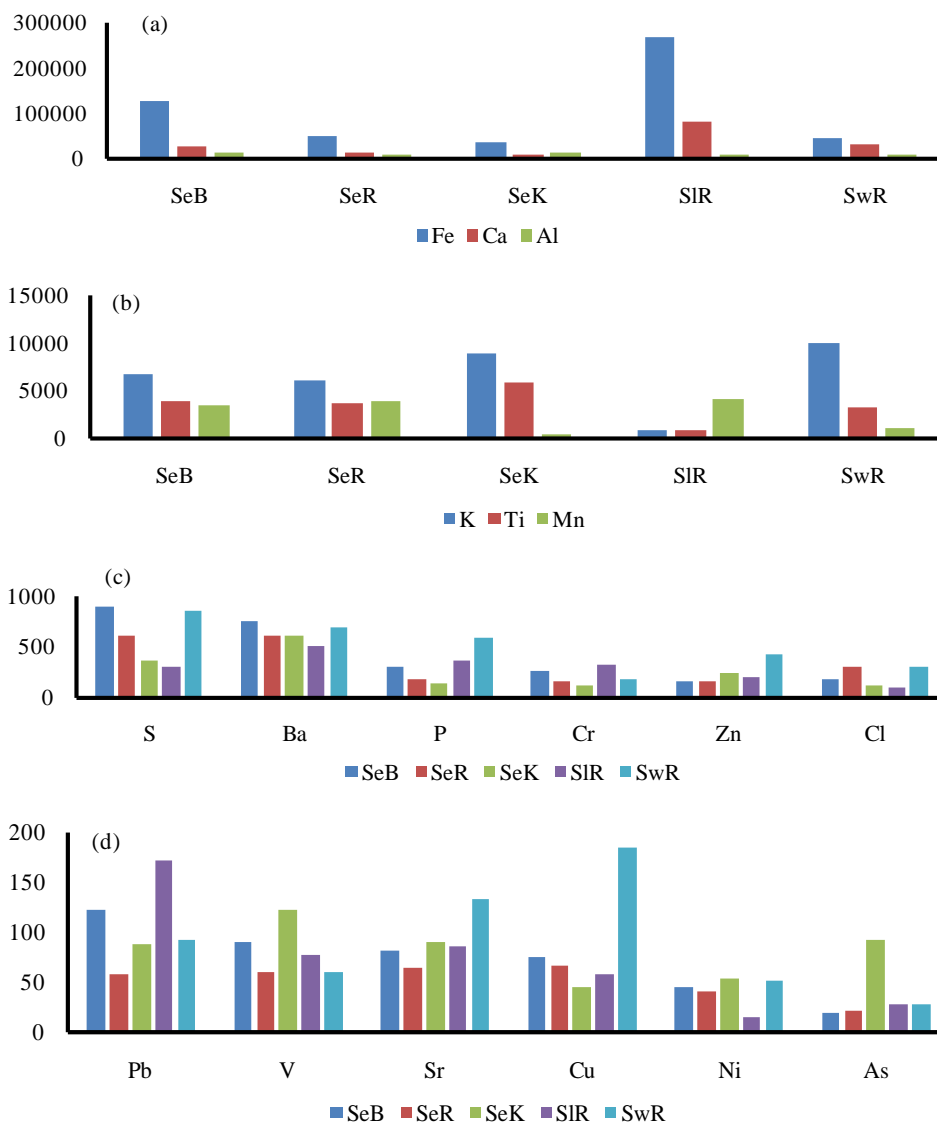


Figure 4. Spatial distribution of elements in the geowaste materials.

Figure 6. Sludge is a solid waste material produced by the iron and thermal power plants in the studied area. Sewage is also a solid waste contributed by domestic households, industrial and agricultural wastes. The highest content of Fe and Ca was marked in the sludge waste. The sewage waste was found to be very rich (1.2%) in nutrient (*i.e.* P + K + Cl + S) levels. However, the higher content of HMs (*i.e.* As + Ti + V + Cr + Mn + Ni + Cu + Zn + Pb) was seen in the sediment. The concentration of HMs *i.e.* Fe, Mn, As and Pb in the sediment of the study area was found to be higher than sediments of other regions due to a huge mineral and coal roasting [11]-[30].

3.4. Enrichment of Elements

The E_f values of the elements are summarized in **Table 3**. The background concentration of Al, Fe, K, Ca, Ti, Mn, P, S, Cl, V, Cr, Zn, Ni, Cu, Pb and As reported was 82000, 39000, 23000, 22000, 4650, 775, 62, 655, 370, 97, 92, 67, 47, 28, 17 and 4.8 mg/kg, respectively [37]. Two metals *i.e.* As and Pb was found to be highly enriched ($E_f > 27$) in all samples. The highest E_f value for elements *i.e.* S, Cl, Cu and Zn was seen in the sewage waste, may be due to contribution by vehicle and cooking effluents. The extremely high E_f value of Fe, Mn, Cr and Ca was observed in the sludge waste due to input of the sponge iron and thermal power plant effluents.

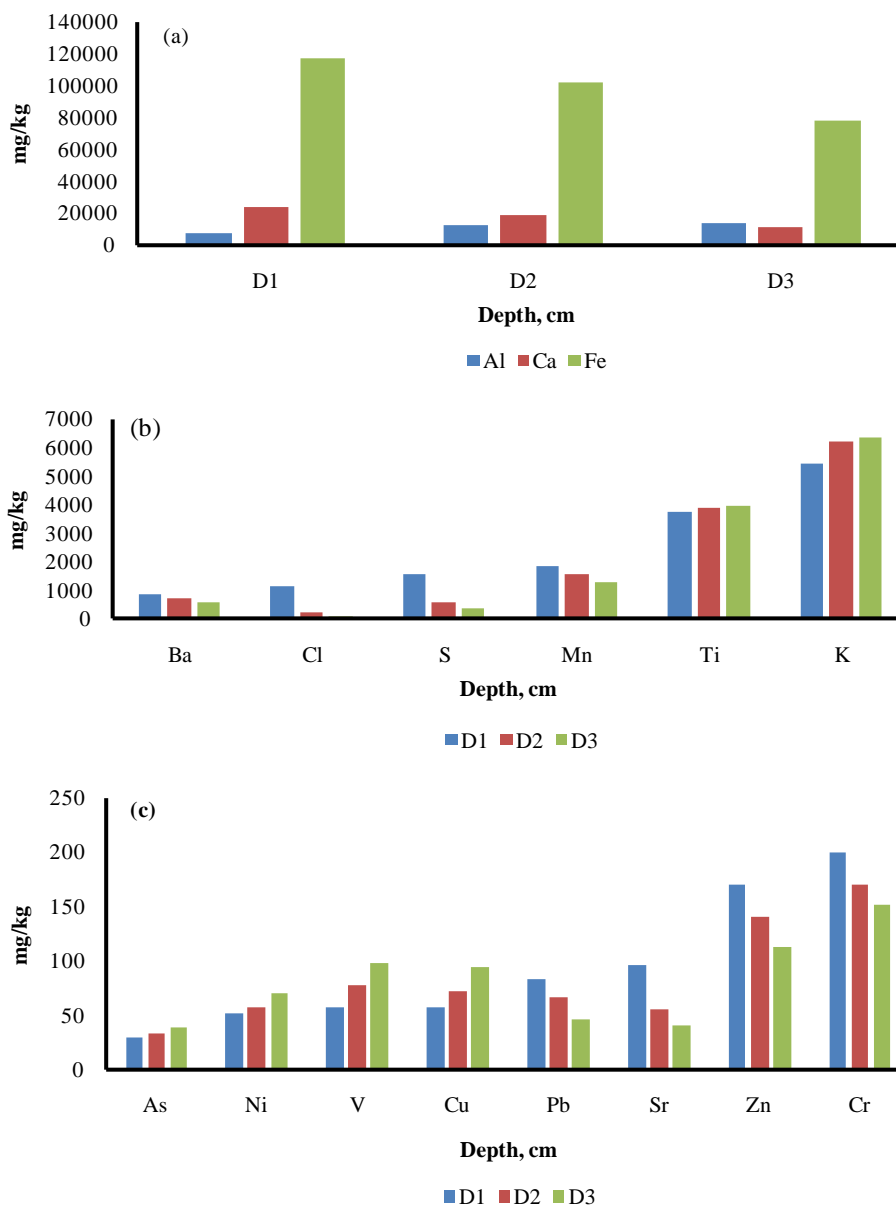


Figure 5. Depth profile studies of elements in Siltara sediment. D1 = 0 - 10 cm, D2 = 10 - 20 cm, D3 = 20 - 30 cm.

3.5. Composition of Geowaste Materials

The elemental and carbon fractions in the sediment, sludge and sewage are shown in **Figure 7**. The fraction of 18 elements (*i.e.* Al, K, P, S, Cl, As, Ca, Sr, Ba, Ti, V, Cr, Mn, Fe, Ni, Cu, Zn and Pb), BC, OC and CC in all sediment and sewage wastes was found comparable. However, two metals *i.e.* Ca and Fe were enriched several folds higher in the sludge due to sponge iron and their effluents.

3.6. Sources of Elements

Cluster analysis was performed on the dataset by Ward's method using Euclidean distance as similarity measure. The variables were interrelated to each other according their maximum similarities. First, the interrelation takes place between two variables which have the most similarity and the next repetition other similar pair clusters were related together. Four clusters or class of sample sites were identified with distinct cluster centers (**Figure 8**).

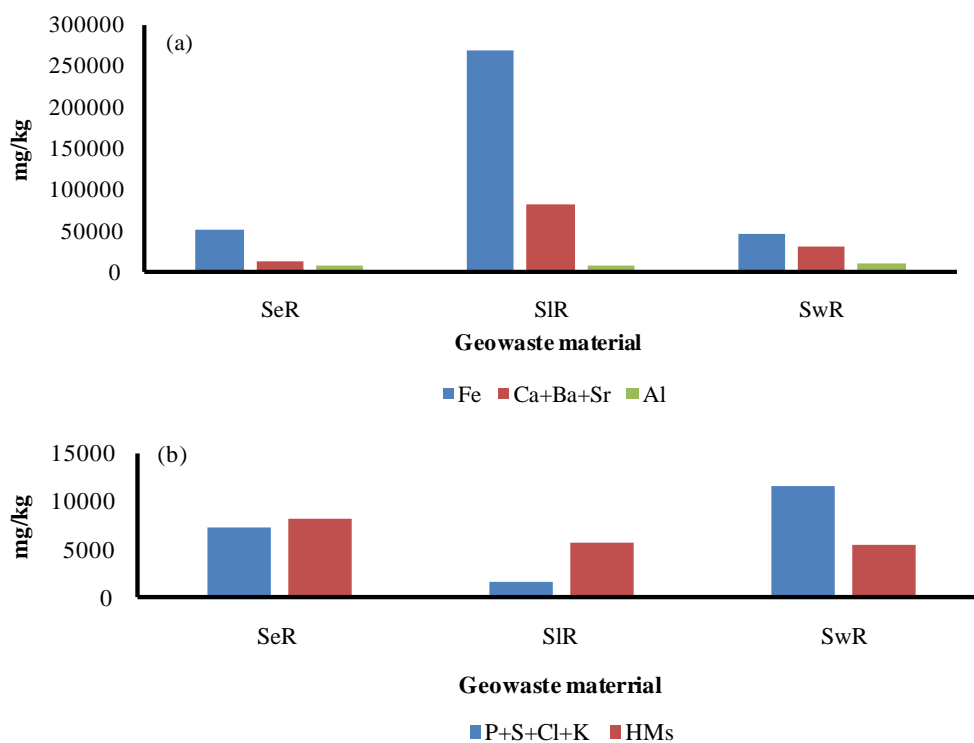


Figure 6. Distribution of HMs and nutrients in sediment, sludge and sewage at Raipur.

Table 3. Enrichment of elements.

Element	SeB	SeR	SeK	SIR	SwR
P	3.7	3	1.0	5.9	4.9
S	87	85	27	37	87
Cl	4.1	8	1.9	2.4	6.1
K	31	2.6	2.1	0.4	2.8
Ca	6.1	5.1	0.6	33	9.9
Ti	20	9.4	8.7	2.1	7.1
V	62	7.4	8.7	9.8	6.3
Cr	1.9	20	8.3	41	20
Mn	22	52	1.7	51	11
Fe	25	15	5.8	81	11
Ni	7.3	8.8	6.4	3.2	9.2
Cu	9.4	24	8.3	21	56
Zn	7.3	28	12.7	32	54
As	34	43	99	57	48
Pb	28	33	30	102	45

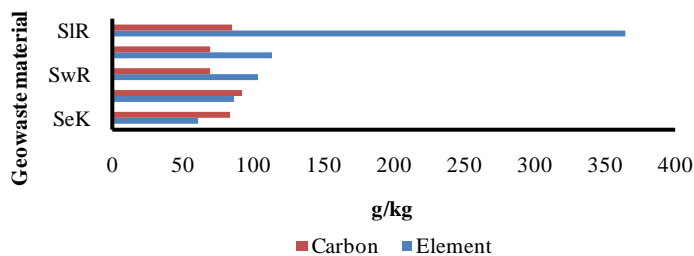


Figure 7. Fraction of element and carbon in sediment, sludge and sewage of Raipur.

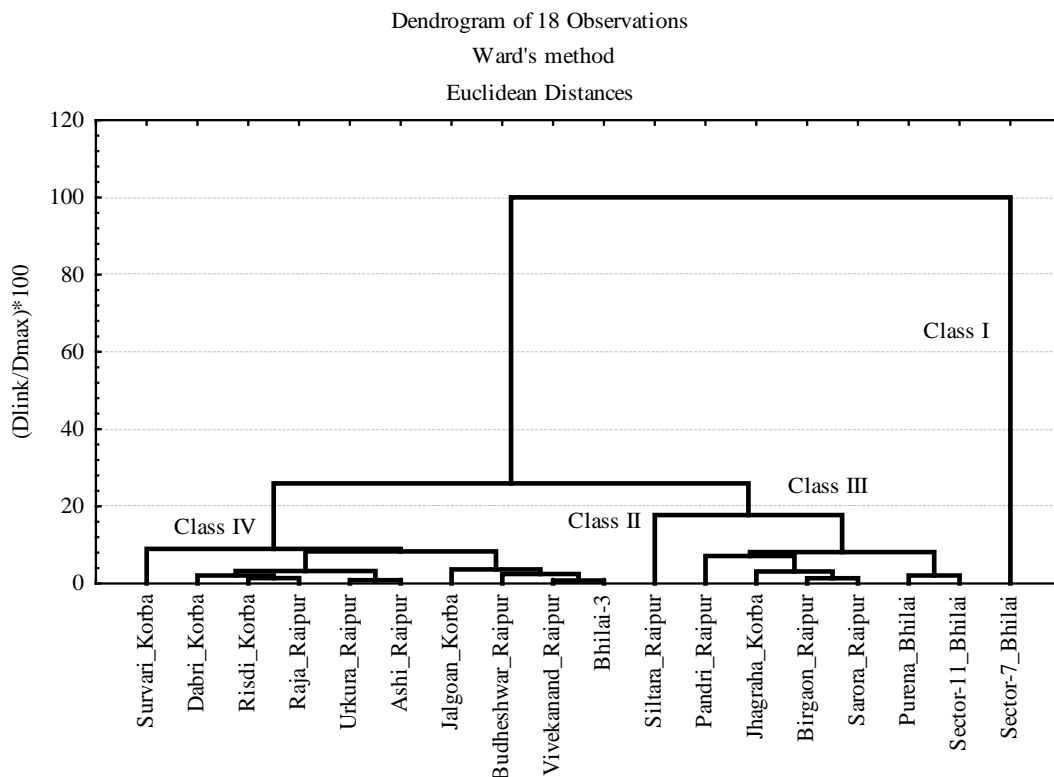


Figure 8. Dendrogram of eighteen sediment observations.

Class-I ($n = 1$) and Class-II ($n = 1$) comprise each one 5.56% of the total samples. Class I has the highest concentration of P, Ca and heavy metals (Cu, Cr, Mn, Fe, Zn and Pb). Class-II has the highest concentrations of OC, CC, S, Cl, Ni, As, Sr and Ba. Class-III ($n = 6$) comprises 33.33% of the total samples and has the highest concentration of Al, Ti and V. Class-IV ($n = 10$) has the highest concentration of K and lower or intermediate concentration of other elements between the class. On the whole, the concentrations of elements exceed highly their respective limit concentrations. The results of k-means clustering of the sediments in all the samples are shown in Table 4.

Using factor analysis (FA), linear correlation between heavy metal concentrations in the sediments was determined. After varimax rotation, heavy metals belonging to a given factor were defined by factor matrix. Metals having strong correlation were grouped into factors and the identification of factors is based on dominant influence [38]. Six factors with eigenvalues higher than 1 [39] were extracted by principal component method. These factors explained 86.56% of the total variance. The variable loadings, eigenvalues, explained variance and cumulative variance are presented in Table 5.

Factor-1 exhibit 34.31% of the total variance with high positive loadings on Mn (0.86), Cu (0.86), Zn (0.80) and Sr (0.81). This factor indicates strong association of Mn, Cu, Zn and Sr in the sediments. Factor-2 exhibit 19% of the total variance with high negative loadings on BC (-0.94), OC (-0.94) and CC (-0.89). This suggests that

Table 4. Results of the k-means hierarchical cluster analysis.

Parameter	Class-I	Class-II	Class-III	Class-IV
pH	7.9	7.6	7.89	7.77
BC	9.6	9.3	7.55	8.33
OC	0.1	0.2	0.13	0.14
CC	0.1	0.2	0.09	0.11
Al	8756	7859	11755	11686
P	304	214	168	216
S	241	1585	492	616
Cl	91	1117	270	99
K	593	5468	7441	7595
Ca	72184	24545	9806	9814
Ti	683	3762	4485	4392
V	40	57	99	83
Cr	362	200	208	131
Mn	8029	1816	5725	631
Fe	319848	117506	60033	35350
Ni	14	52	50	45
Cu	93	57	80	49
Zn	273	170	194	143
As	12	29	21	21
Sr	82	96	79	71
Ba	546	823	728	593
Pb	228	84	66	44

BC, OC and CC proceed from the same source. Factor-3 accounts for 12.14% of the total variance with high positive loadings on Ca (0.85), Cr (0.82) and Fe (0.91). Taking account of some high concentrations in Fe, Ca this factor can be attributed to mixed sources of Fe in the sediments from geogenic and anthropogenic source.

Factor-4 accounts for 9.04% of the total variance with high positive loading on Cl (0.73). High concentrations of Cl in the sediments could come from runoff, wastewater. Factor 5 accounts for 6.53% of the total variance with high negative loadings on Al (-0.73), V (-0.82) and Ti (-0.80). This factor shows the relationship of anthropogenic Ti and V with reference element Al. Factor 6 accounts for 5.55% of the total variance with high negative loading on pH (-0.87).

4. Conclusion

The geowaste materials *i.e.* sediment, sludge and sewage are rich with heavy metal and nutrient contents. The remarkably higher nutrient level is observed in the sewage, found useful as manure for crop productions. Iron is seen to be extremely enriched up to 27% in the sludge waste, and could be a cheaper resource for Fe recovery. The high BC levels in geomaterials may stabilize the heavy metal and nutrient contents. The toxic metals *i.e.* As and Pb were highly enriched ($E_f > 27$) in all geowaste materials. The vertical distribution of As was increased remarkably with increasing depth profile of the sediment.

Table 5. Rotated varimax factor of chemical constituents.

Element	Factor-1	Factor-2	Factor-3	Factor-4	Factor-5	Factor-6
pH	0.01	-0.13	0.06	-0.09	0.20	-0.87
BC	-0.04	-0.94	0.06	-0.20	0.18	-0.08
OC	0.08	-0.94	-0.11	-0.03	0.09	0.02
CC	-0.02	-0.89	0.08	0.18	0.17	-0.12
Al	-0.02	0.34	-0.07	-0.34	-0.73	0.22
P	0.32	0.41	0.18	0.15	0.62	0.23
S	-0.05	0.24	0.01	0.66	0.41	0.43
Cl	0.50	-0.23	0.10	0.73	0.18	0.00
K	-0.26	0.47	-0.50	0.02	-0.48	0.22
Ca	0.18	-0.13	0.85	-0.19	0.34	0.10
Ti	-0.21	0.08	-0.45	0.00	-0.80	0.16
V	-0.12	0.38	-0.17	-0.05	-0.82	0.23
Cr	-0.09	0.27	0.82	0.20	-0.09	-0.32
Mn	0.86	-0.16	0.09	0.02	0.23	-0.23
Fe	0.10	-0.10	0.91	-0.11	0.18	0.07
Ni	-0.29	0.26	-0.09	0.33	-0.75	-0.05
Cu	0.86	0.07	0.16	0.04	0.34	-0.13
Zn	0.80	-0.06	0.04	-0.23	0.16	0.02
As	-0.22	0.01	-0.35	0.63	-0.30	0.00
Sr	0.81	-0.01	0.09	0.20	-0.20	0.39
Ba	0.69	0.37	-0.02	0.55	0.07	0.15
Pb	0.57	0.10	0.61	0.01	0.46	0.06
Eigenvalue	7.55	4.18	2.67	1.99	1.44	1.22
%Variance	34.31	19.00	12.14	9.04	6.53	5.55
CV (%)	34.31	53.31	65.45	74.49	81.02	86.56

Significant loadings > 0.7 (in bold) at $p < 0.05$.

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