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Spatial Changes in Soil Organic Matter and Waste Elements in Contaminated Agricultural Fields at Anthropogenically Transformed Areas within the East Calcutta Wetlands Ecosystem

Shuvadip Adhikari*, Anandamay Barik, Sudin Pal[1], Sanjoy Chakraborty[1] and Subhra Kumar Mukhopadhyay[1]

Department of Zoology, The University of Burdwan, Golapbag, Burdwan, 713104, West Bengal, India

Physical and chemical properties of soil were compared between different agricultural fields of multi-metal waste contaminated East Calcutta Wetlands (ECW) sites and uncontaminated control study sites. Soil organic matter (SOM) contents were significantly different between contaminated ECW and control sites. Agricultural fields at ECW sites were characterized by higher contents of SOM, porosity, moisture, particle size (sandy >63 μm) and metals; while the control sites had higher bulk density, pH and electrical conductivity (EC). Metal accumulation in soils of ECW showed the trend Fe>Zn>Mn>Cr>Pb>Cu; while at control sites like Fe>(Zn=Mn)>Cr>(Pb=Cu). Post-Hoc analyses (with Tukey HSD) clearly pointed out site-wise significant differences (at $p<0.05$) in soil metal contents between ECW and control sites. The results of this study pointed out the importance of chelates like SOM and easily biodegradable organic acids in the waste contaminated soils of ECW to arrest metals in significant amount reducing phytoavailability and, thereby, sustained agricultural practices at the concerned areas without any health hazard for nearly a century.

Key words: East Calcutta Wetlands, metals, phytoavailability, Post-Hoc analyses, soil organic matter

Organic matter present in small amount in soil yet plays important roles in soil dynamics and adsorption of heavy metals with soil and sediments (Qu *et al.* 2019). Soil organic matter (SOM) typically contains 45-50% carbon content is important for soil functioning. Carbon stored in SOM is most dynamic pool of organic carbon (OC). Different physicochemical and biological attributes of soil, influence the bioavailability and mobility of metals (Zwolak *et al.* 2019). Different functional groups present in soil organic matter (SOM) like carbonyl, carboxyl or phenylhydroxyl groups acts as a ligand to bound heavy metals especially monovalent cations. Along with the predominant functional groups on organic surfaces like low-molecular weight organic acids (citric acid, gluconic acid, gallic acid, tartaric acid, series of soluble humic or fulvic acids) or amino acids (like glycine) act as chelating agent which reacts with heavy metals (Moragaspitiya *et al.* 2020). Adsorption by SOM causes immobilization of heavy metals which check the phytoavailability of metals. Therefore, soil acts as a protective barrier against the uptake of heavy metals by plant system (Kwiatkowska-Malina 2018).

East Calcutta Wetlands (ECW), a Ramsar site (no. 1208) is globally appreciated for its traditional management as it is converting waste into wealth for the last century. The ECW comprises an area of 12741 ha which includes 52.32% cropland area. Sustainable agricultural practice in ECW area is a yearlong practice emphasized on garbage farming and wastewater irrigation. There is a 7.39 per cent increase in cropland area in ECW area over 2002 to 2010 (Sarkar and Parihar 2014) due to a marked decrease in wetland area (Li *et al.* 2016). Use of sludge from wastewater carrying canals in agricultural field is also common in ECW area. It is recorded that about 24 species of vegetables or crop plants are cultivated in sludge and solid wastes filled ECW areas. Daily 150 metric tonnes (t) of vegetables are produced from ECW area. Waste elements including different heavy metals released to the environment for varied

*Corresponding author (Email: shuvaadhikari90@gmail.com)
[1]Ecotoxicology Project Laboratory, Government College of Engineering and Leather Technology, Block LB, Sector III, Salt Lake, Kolkata 700106, West Bengal, India

anthropogenic activities and dumping of solid wastes at the wetland areas heavily contaminated the soils of ECW ecosystem. The ECW area receives 2000-3000 t of solid wastes in different forms and 1.1 million m^3 composite industrial effluents along with municipal wastewater daily. Municipal and industrial wastewaters and solid wastes are the sources of heavy metals in ECW ecosystem. Highly contaminated anthropogenically transformed soils of ECW sustainably yield agricultural crops and vegetables for the last century for human consumption in and around Kolkata metropolitan. Hence, Kolkata has been referred to as 'ecologically subsidized city'. The possibilities of such sustainable production over a long time are attained as the elements are partitioned and ameliorated in natural way.

Heavy metals are ubiquitous in all types of environments including both polluted and unpolluted soils (Masindi and Muedi 2018). Different elements, including heavy metals, occur in soils due to natural soil forming processes from parent materials and also due to varied anthropogenic activities (Srinivasarao *et al.* 2014). Metals like copper (Cu), iron (Fe), manganese (Mn) and zinc (Zn) are known essential nutrients (metallic trace elements) for plant growth, however, chromium (Cr) and lead (Pb) are considered to be phytotoxic. Therefore, higher the presence of metals in agricultural fields, greater is the concern for potential hazardous effects of such elements on human health (Salman *et al.* 2019). Thereby, our work is designed to explore three main questions: 1) is there any possible affinity of SOM for tacit arresting of waste elements? 2) is such interaction instrumental in reducing phytoavailability of unwanted elemental loads? and 3) how are resource-recovery agricultural practices sustainably thriving at contaminated ECW soils?

Materials and Methods

Study sites

The ECW (22°33′–22°40′ N, 88°25′–88°35′ E) located on the eastern fringe of Kolkata metropolitan. The ECW ecosystem received solid wastes, municipal and industrial wastewater from Kolkata (Fig. 1). Agriculture and floriculture are common in ECW ecosystem. Solid wastes and sludge from wastewater carrying canals was frequently used as agricultural land-fills. Wastewater is also used for irrigation that altered the properties of soil therefore, the soil in ECW agricultural fields are referred as 'anthropogenically transformed'. Samples from uncontaminated sites (control site) were collected from Hooghly (22°54′24.14′′ N, 88°21′52.12′′ E, about 45 km away from ECW area) where either rain water or ground water is used to raise the agricultural produces.

Sample collection

Surface (0-10 cm) soil samples were collected from eight cultivated lands of ECW and Hooghly area by a hand-held core sampler (having a diameter of 5 cm). Surface soils were collected as it generally associated with root rhizosphere and were influenced

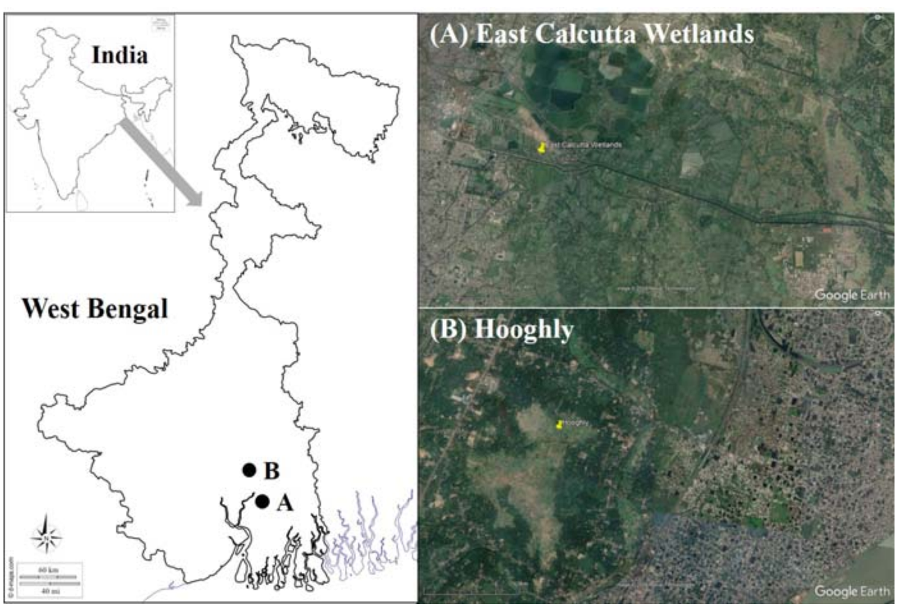

Fig. 1. Map of the study sites: East Calcutta Wetlands (A) and control site, Hooghly (B)

by the root activity (Gregory 2006). Three soil samples were collected from both control and experimental sites. Soils of each of the agricultural fields of eight different vegetables *viz.*, brinjal, cauliflower, cucurbit, jute, maize, mustard, red amaranth and spinach were collected on vernal months *i.e.* February and March 2018 between 08:00 AM to 10:00 AM. After sample collection they were brought in laboratory for further analyses.

Sample analysis

Physical properties of soil

For the estimation of bulk density (BD), moisture and porosity samples were dried in hot air oven at 60 °C until constant sample weight was achieved. Both wet and dry weights were considered for necessary calculations. Sediment texture analysis was carried out following Wentworth (1922) and as per their set standard soil samples were fractioned into sand (>63 μm) and silt-clay (<63 μm).

Chemical properties of soil

Soil pH and electrical conductivity (EC) were estimated following soil: solution ratio of 1:2 and measured using pH-Conductivity meter (WTW Multi 3420). Spectrophotometric estimation of the organic carbon in soil was carried out using a UV-VIS spectrophotometer (Lamda-25, Perkin-Elmer, USA) following Walkley and Black (1934) method. Obtained values were multiplied with 1.724 to get the percentage values of SOM. Elemental analyses for six prevalent metals in the concerned areas *viz.*, Cu, Fe, Mn, Zn, Cr and Pb (Chattopadhyay *et al.* 2002), were done by atomic absorption spectrophotometer (Perkin-Elmer AAnalyst-100) using element specific hollow cathode lamps in default condition. Standard protocol recommended by Perkin-Elmer was followed for sample preparation *i.e.*, elemental extraction and elemental detection (Welz and Sperling 1999).

Statistical analyses

Post-Hoc measures (Tukey's HSD) were performed by using STATISTICA 8.0 software to highlight significant differences between the soil properties of ECW agricultural sites and control sites. Canonical Correspondence Analysis (CCA) was carried out by CANOCO (version 4.57) software and represented as bi-plot to evaluate the SOM-metal association. For graphical representations, Origin 6.1 software was used.

Results and Discussion

Physical properties of soil varied between different agricultural fields of ECW and control study sites (Table 1). Bulk density of soil samples was marginally higher in control sites (0.91±0.13 Mg m^{-3}) than the samples of ECW (0.79±0.06 Mg m^{-3}). Both moisture and porosity values were significantly higher in agricultural soils of ECW. Average proportion of

Table 1. Soil physical properties of different agricultural farming at contaminated ECW and control sites

Agricultural farming	Bulk density (Mg m^{-3})	Moisture (%)	Porosity (%)	Sand (%)	Silt and clay (%)	pH	EC (dS m^{-1})
ECW							
Brinjal	0.80±0.06	13.35±7.80	10.41±5.49	54.67±3.37	45.33±3.37	6.44±0.02	0.08±0.01
Cauliflower	0.70±0.08	28.51±3.36	20.12±4.71	73.41±1.59	26.59±1.59	6.52±0.05	0.19±0.03
Cucurbit	0.75±0.07	29.62±5.81	21.96±2.56	75.63±1.69	24.37±1.69	6.77±0.04	0.09±0.03
Jute	0.83±0.08	21.32±2.69	17.55±0.44	71.26±1.74	28.74±1.74	6.75±0.06	0.10±0.03
Maize	0.80±0.04	20.18±1.41	16.19±1.98	66.15±3.21	33.85±3.21	6.57±0.04	0.08±0.02
Mustard	0.81±0.04	22.79±4.32	18.36±2.73	73.78±3.50	26.22±3.50	6.74±0.03	0.10±0.05
Red Amaranth	0.75±0.06	21.63±2.55	16.12±0.72	73.96±2.75	26.04±2.75	6.70±0.04	0.12±0.02
Spinach	0.84±0.03	18.75±4.90	15.68±3.76	77.67±2.63	22.33±2.63	6.56±0.03	0.23±0.04
Control							
Brinjal	0.89±0.12	14.21±5.44	13.09±6.69	74.73±4.50	25.27±4.50	6.98±0.14	0.27±0.03
Cauliflower	1.06±0.27	9.84±4.27	11.00±7.32	61.23±2.79	38.77±2.79	6.83±0.15	0.26±0.04
Cucurbit	0.90±0.04	16.82±6.26	15.13±5.55	78.77±3.08	21.23±3.08	6.83±0.17	0.13±0.02
Jute	0.79±0.02	11.31±6.42	8.91±4.96	63.22±2.56	36.78±2.56	6.99±0.12	0.12±0.02
Maize	0.87±0.05	8.66±1.94	7.52±1.67	43.57±7.53	56.43±7.53	7.10±0.07	0.06±0.02
Mustard	0.86±0.06	18.33±3.07	15.81±3.25	61.65±5.65	38.35±5.65	7.00±0.06	0.08±0.01
Red Amaranth	0.85±0.06	19.40±6.38	16.70±6.61	67.47±7.58	32.53±7.58	6.75±0.06	0.16±0.02
Spinach	1.07±0.10	9.00±3.47	9.85±4.37	70.54±2.18	29.46±2.18	7.16±0.05	0.10±0.03

sand was comparatively higher in ECW than silt-clay. Soils of the agricultural fields of ECW area were comparatively slightly acidic (pH 6.62±0.12) than control sites (pH 6.95±0.17). However, EC values were considerably higher in control sites (0.16±0.09 dS m^{-1}) than ECW sites (0.12±0.06 dS m^{-1}). Bulk density of soil is an important soil characteristic that is influenced by several other factors like amount of solid phase in soil (proportion of sand or silt-clay, organic matter) and the pore space or available water content (Chaudhari *et al.* 2013). In present study BD of soil samples from ECW were lower in comparison to control sites due to the higher moisture content and porosity. Tanveera *et al.* (2016) reported that sand particles showed positive correlation with BD whereas clay, porosity and total organic matter were negatively correlated. However, in present study higher percentage of sand particles were recorded from ECW area where BD values were comparatively low. Zhao *et al*. (2012) reported that BD of the compost decreased as particle size increased. However, with the increase of particle size there is a trend to increase some soil parameters such as pH, porosity, saturated water holding capacity, OC content, and Ca, Fe, Mg, and Mn contents.

The SOM values ranged from 73.5 g kg^{-1} to 154.0 g kg^{-1} in ECW with an average of 108.9±18.1 g kg^{-1}. Average SOM value was much lower in control sites (83.1±17.5 g kg^{-1}) and ranged from 34.6 g kg^{-1} to 106.0 g kg^{-1} (Fig. 2). Results obtained from Post-Hoc analysis revealed that except two agricultural fields (that cultivated brinjal and maize) all the values of SOM were significantly different between contaminated ECW and control sites (Fig. 2). The SOM values were significantly higher in most agricultural fields at ECW areas. Minimum tillage and retention of crop residues played an important role in carbon storage in the agricultural fields of ECW ecosystem. Chemical fertilizers were not commonly used in ECW areas as the soil was loaded with municipal solid wastes contain inorganic matter and various quanta of elements including essential elements for plant growth. However, in control sites of Hooghly intense agricultural activity were practiced including tillage and use of chemical fertilizers *etc*. Such differences in the mode of agricultural practices were important factor to bring about spatial differences in the SOM. Besides, wastewater containing higher concentration of organic matters, used for irrigation, also increased the organic load in soils of agricultural fields of ECW compared to the control fields.

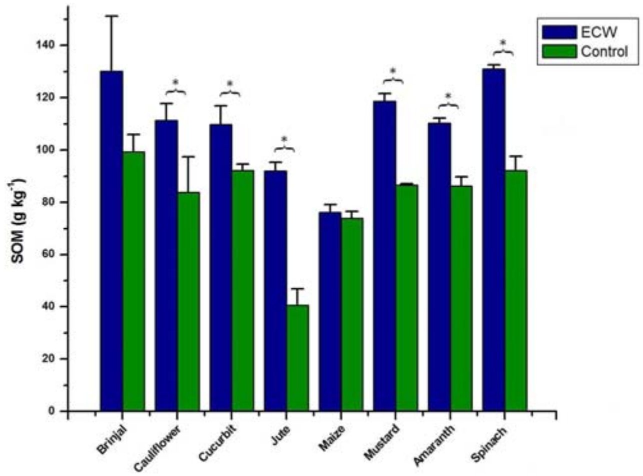

Fig. 2. Soil organic matter present in different agricultural farming sites at contaminated ECW and uncontaminated control areas (* marked fields showed significant differences at *p<0.05* in Post-Hoc analyses)

Metal concentrations were comparatively higher in ECW samples except the values of Fe in five agricultural fields (Fig. 3). Metal accumulation in soils of ECW showed the trend Fe>Zn>Mn>Cr>Pb>Cu. Similar trend was also recorded from control sites like Fe>(Zn=Mn)>Cr>(Pb=Cu). Post-Hoc analyses (with Tukey HSD) clearly pointed out field wise significant differences (at $p<0.05$) for Mn, Cu, Zn and Pb between ECW and control sites. Soil concentrations for Cr were also significantly higher (except cauliflower and cucurbit fields) in ECW soils than control soils. Contrary to this, Fe values were significantly different between contaminated and control study sites only in a single case, *i.e.*, the brinjal field (Fig. 3). Result obtained from CCA biplot clearly pointed out the close association of selected metals (Cu, Mn, Pb and Zn) with SOM (Fig. 4). However, in control site SOM was associated with BD and sand particles. Metals like Mn and Zn were associated with pH in the control sites. Position of Fe was near the interception of two axes in the CCA plot in both ECW and control sites. Physical properties of soil like moisture and porosity were coupled in both cases and soil and silt-clay contents were placed in opposite direction in the biplot. Bradl (2004) reported that SOM formed complexes with metals by ion-exchange, surface adsorption, chelation, and complex coagulation and peptization reactions. However, little was known concerning the nature of the ligands in polymeric components of SOM which could chelate metals, but carboxyl, hydroxy, and amide groups were probably involved. In the present study, high metal

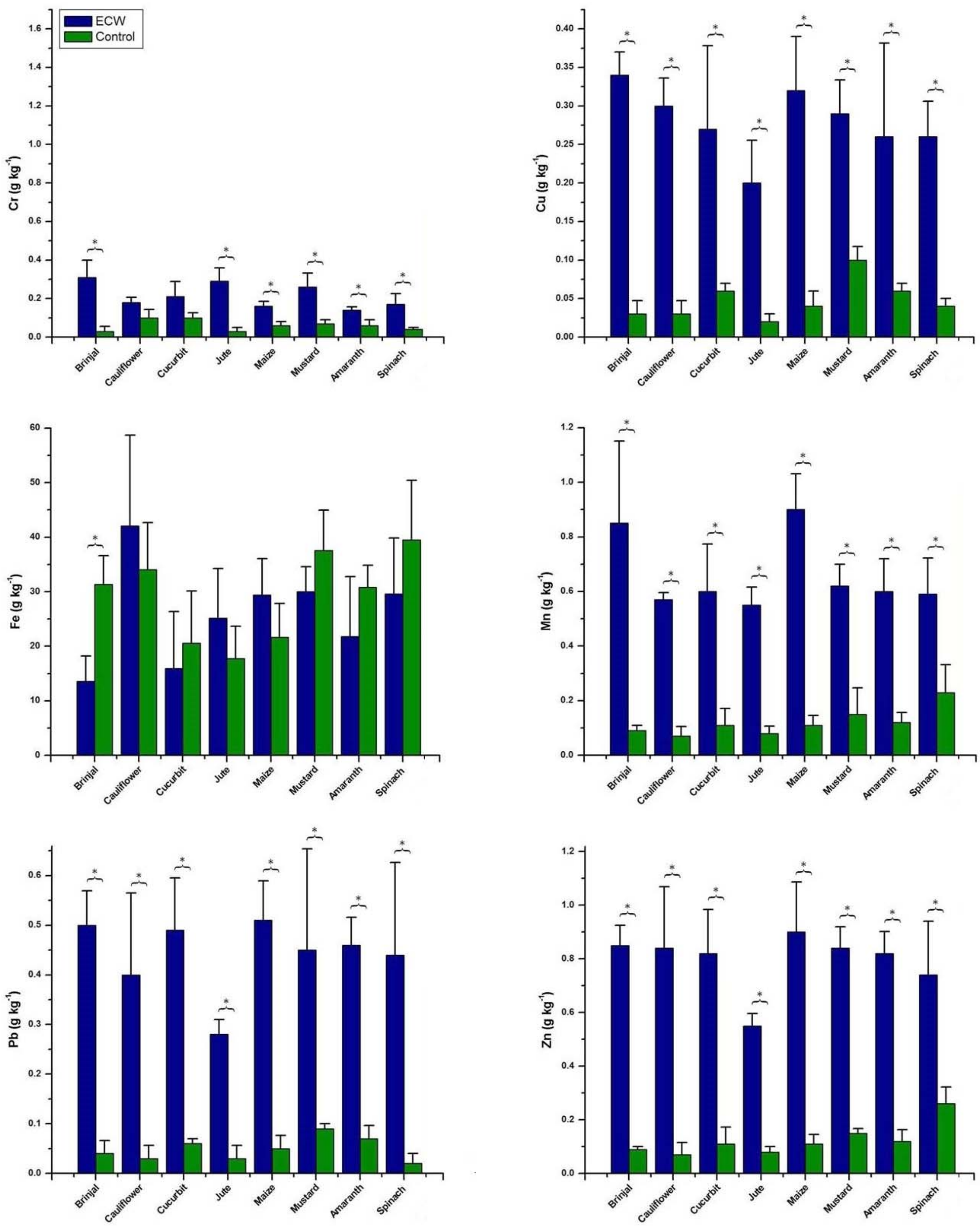

Fig. 3. Concentration of different metals present in different agricultural farming sites at contaminated ECW and uncontaminated control areas (* marked fields showed significant differences at *p<0.05* in Post-Hoc analyses)

contents in the ECW soils showed a definite affinity to SOM as depicted in CCA (Fig. 3). Possibly due to negligible metal contents of soils at the control sites, especially Cr, Mn, Cu, Zn and Pb, no such relationship was apparent in CCA biplot. Quenea *et al.* (2009) investigated the interactions of four metals (Pb, Zn, Cu and Cd) with the SOM associated to five different size fractions (between 2000 µm and <2 µm) of a sandy top soil contaminated by waste water. Higher affinity of metals to bind with SOM at the ECW soils where particles were predominantly sandy over silt-clay corroborated the findings of Quenea *et al.* (2009).

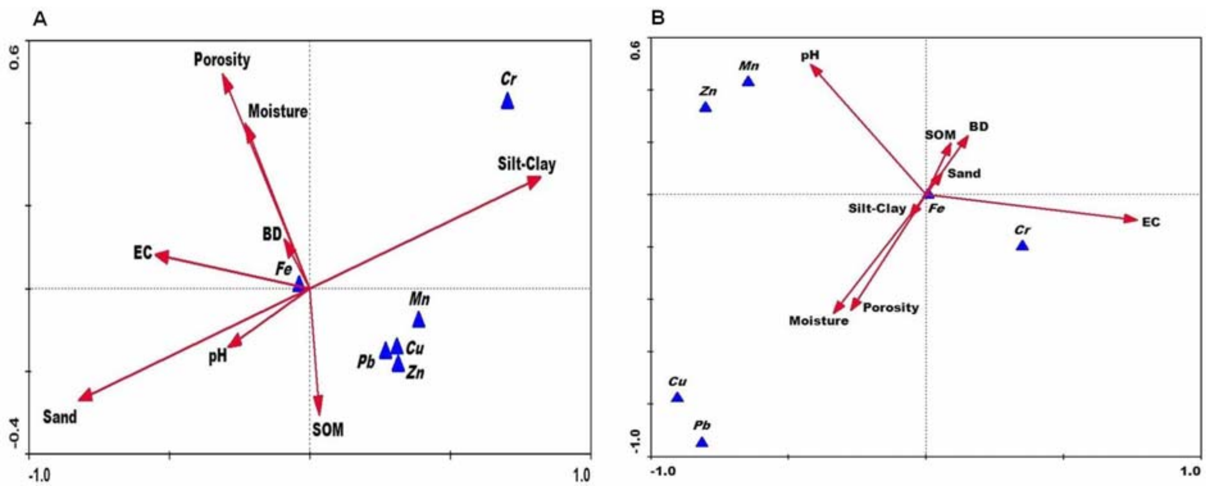

Fig. 4. Canonical correspondence analysis (CCA) bi-plot for ECW (A) and control (B) sites

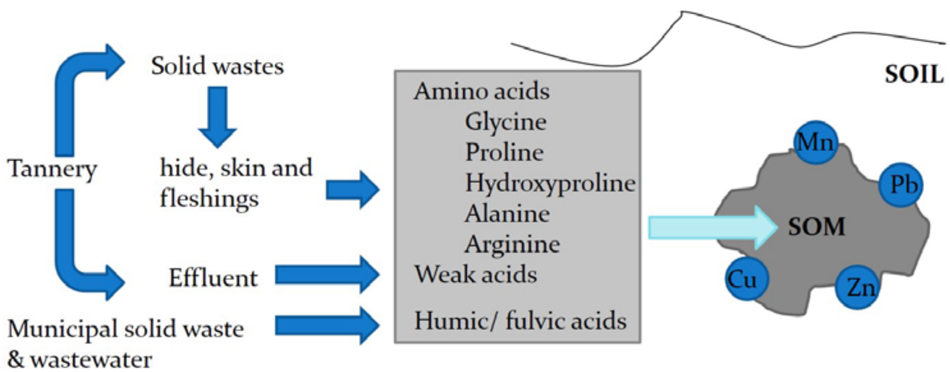

Fig. 5. Probable strategy of SOM-metal association in anthropogenically transformed contaminated agricultural field soils of ECW ecosystem

Hugar and Soraganvi (2014) recorded that soil OC helped to aggregate fine particles leading to the increased soil porosity. The ECW agricultural fields were prepared by the sludge and dredged sediment of wastewater carrying canals along with solid wastes that provided the characteristics of municipal solid waste compost. Thereby, ECW agricultural soil exhibited higher porosity, moisture content, organic matter and metal contents than the control soil.

The ECW received huge amount of solid waste and composite wastewater from several large- and small-scale industries especially from leather industry. Organic load in the form of hide, skin and fleshings contains collagen which was rich in amino acids like glycine, proline, and hydroxyproline (Shoulders and Raines 2009). Apart from different enzyme residues used in leather processing two other amino acids like alanine and arginine were also added to the soil with tannery solid wastes and wastewater (Paul *et al.* 2013). Untreated municipal wastewater contained bio-degradable matters that produced humic/fulvic acids resulting acidic pH of soil (Krishna *et al.* 2016). Soluble humic/fulvic acids and amino acids played a key role in metal chelation in soil. do Nascimento *et al.* (2006) reported the performance of low molecular weight organic acids as naturally occurring chelates in enhancing extraction of metals. Waste metal contamination of soil was one of the most important environmental problems throughout the ECW areas (Chattopadhyay *et al.* 2002). Heavy metals were non-biodegradable and could remain almost indefinitely in the soil environment. However, their availability to biota could change considerably depending on their chemical speciation in the soil and soil characteristics such as organic matter and organic acid contents (Wuana *et al.* 2010). High SOM and considerable high contents of low molecular weight organic acids in the organic wastes filled agricultural soils of ECW significantly arrested the waste metals reducing the phytoavailability (Fig. 5). Thereby, the agricultural practices in the contaminated soils of ECW thrived sustainably for nearly a century.

Conclusions

From present study it was revealed that anthropogenically transformed soils of ECW was highly contaminated by waste metals than the control sites. Besides higher concentration of waste elements SOM content was also high in the agricultural fields of ECW ecosystem. By employing multivariant constrained ordination statistical analysis like CCA it was shown that selected metals had affinity with SOM in ECW soils. According to the result it may be concluded that SOM-metal association in turn reduce phytoavailability of unwanted loads of elements. In the highly contaminated anthropogenically transformed soils of ECW sustainably yield agricultural crops and vegetables for the last century for human consumption in and around Kolkata in cheap costs, due to reduction of cost of manure, irrigation *etc*., recognizing Kolkata as 'ecologically subsidized city'. However, there was no such recorded evidence of health-related hazards by consuming such agricultural produces. Present study will serve as a basis for future studies on soil-metal-plant association in ECW ecosystems.

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