



Research article

Stability and heavy metals accumulation of soil aggregates under different land uses in the southwest coastal Bangladesh

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ABSTRACT

Agricultural soil contamination is increasing day-by-day and becoming a major problem over the globe. Trace elements accumulation in the bulk soil is frequently documented, however, there is no precise mechanism of their distribution in the different soil aggregates level. We collected twelve composite soil samples from banana fields, fallow land, rice cultivated with pond water (rice field-I), and rice cultivated with rain water (rice field-II). We separated soil samples into four different size of aggregates (4-2, 2-0.25, 0.25-0.053, <0.053-mm) and then, aggregate stability (MWD), soil organic carbon (SOC), and heavy metals content (Pb, Cd, Cr, As, Fe, Mn, Zn, Ni, Co, Cu) in the soil samples were measured with different techniques. Results showed that MWD was higher in the rice-based land use, which was significantly contributed by SOC ($p < 0.001$). The concentration of Pb, As, Cd, Fe, and Mn were increased, while Cu and Zn concentration were reduced with increasing aggregate size ($p < 0.05$). In contrast, aggregate size did not influence on Ni and Co accumulation ($p > 0.05$). Moreover, macroaggregate acted as an accumulator for Fe, Mn, and As, while all the aggregate fractions acted as accumulators for Cu and Zn. Our study indicated that MWD, SOC, aggregate size and composition, and metal species were the controlling factors of trace elements accumulation and distribution in the various sizes of soil aggregates.

1. Introduction

Soil aggregates are the building block of soil structure, which plays a vital role in different ecological processes, such as plant growth, yield, root function, and microbial activity [1,2]. Soil aggregates are classified into four fractions for study: large macroaggregates (4-2 mm), small macroaggregate (2-0.25 mm), micro-aggregate (0.25- 0.053 mm), and silt plus clay fractions (<0.053 mm) [3,4]. Soil organic matter (SOM) is a potential binding agent for soil aggregate formation [5]. Gluing soil particles together by different transient and persistent binding agents forms larger aggregates and increase the soil aggregate stability [5].

Soil aggregate stability is expressed by the mean weight diameter (MWD) of aggregates, which is an indicator of soil structure formation [6-8]. Many natural and anthropogenic factors, such as tillage, land use, cropping systems, organic (manure, crop residue, compost from waste) and inorganic fertilization, impact the soil physical, chemical, and biological processes, which can regulate MWD

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and the composition of soil aggregates [9]. It has been reported that land uses have a great influence on soil structure and quality [10, 11]. For instance, it has been reported that land use change (conversion of natural forests to cultivated lands) resulted in significant decreases in MWD, soil pore space, and clay content [12]. In recent decades, the land use in the soils of southwestern Bangladesh has been changed tremendously due to population expansion, industrialization, new construction of roads and highways, intensive agricultural practices, and deforestation [13]. These land uses around these areas are promoting numerous ecological problems such as soil erosion, soil quality degradation, and soil carbon mineralization [14]. Moreover, industrialization, vehicle emission, and use of inorganic fertilizer in intensive agricultural practices are leading to accumulation of different toxic metals in the nearby agricultural soils [15]. Once these metals enter ecosystems, they cannot be decomposed by soil microbes, resulting easily migrate and enrich in

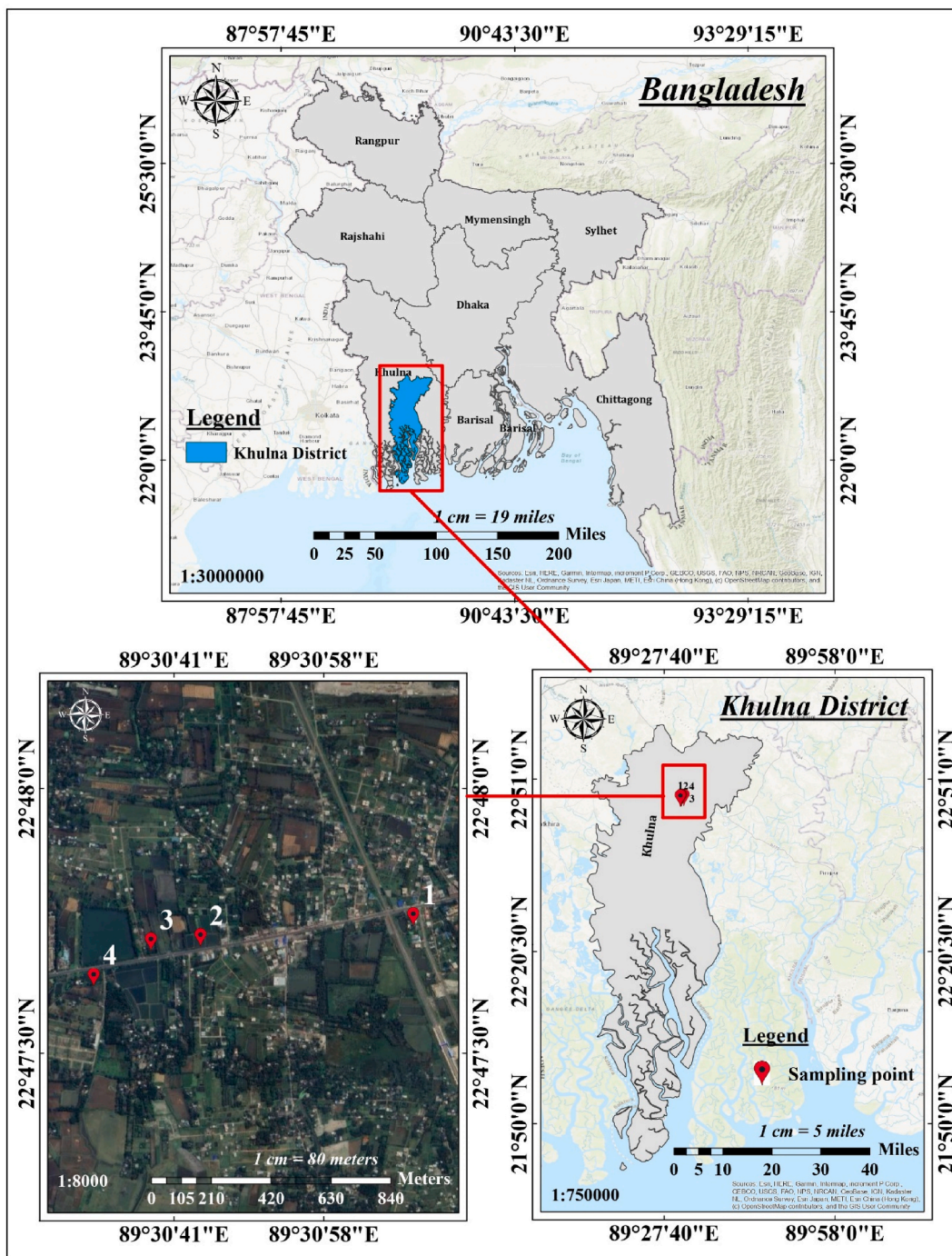


Fig. 1. The study map under the current investigation.

different components of ecosystem with different forms [16]. These metals also can migrate from soil to surface and sub-surface water bodies during recharging with rainwater [17]. Consequently, these toxic metals are entering biological food chain leading to different diseases in human health [15,16,18].

The trace metals accumulation also can deteriorate the soil inherent quality for crop production [19]. Different agricultural approach is applied to improve the sustainable soil quality [19–21]. The application of organic amendments (manure, crop residue) to enhance the soil physical properties like soil aggregation is frequently used soil conservation option [8,22,23]. A lot of research findings suggested that incorporation of inorganic fertilizer coupling with organic amendments is the promising and sustainable practice to enhance the soil organic carbon and MWD [19,20,22,24]. These inorganic fertilizers contain substantial amounts of trace metals, which can contaminate agricultural soils [25]. Human activities such as pesticides application in agriculture, industrial processing, mining, and fossil fuel combustion also can release substantial amounts of heavy metals into the surface soils [26]. A holistic understanding of the environmental behavior of metal elements should not be limited to bulk soil but must also focus on the aggregate level [27]. Previous studies have indicated that heavy metal concentrations in soils are strongly influenced by soil aggregate-size distribution thus the stability of soil aggregates [28–31]. The ability of various sizes of aggregates to adsorb metal elements is different due to their differences in physicochemical characteristics [32,33]. In most cases, fine aggregates have a greater ability to bind heavy metals in the soil than macroaggregates [34,35]. Fine particles in surface soils accumulate a higher content of heavy metals due to their higher reactive surface area, and presence of Fe/Mn oxide phases [31]. In contrast, some researchers found that coarse sand-sized soil aggregates had the highest content of heavy metals due to their greater content of SOM [33,36]. SOM has different charge carbon functional groups, which have the affinity to absorbed heavy metals [37,38]. In addition, SOM acts as a chelating agent, which form chelation with metals and reduce their bioavailability resulting enhance their concentration in the macroaggregates [39]. Moreover, following aggregate hierarchy theory, smaller charged clay particles bind with trace metals and form microaggregates to macroaggregates and thus leading to greater content of heavy metals in macroaggregates [5,37,39]. However, these contrasting findings call for a detailed investigation of how the heavy metals are distributed along the different sizes of soil aggregates.

In our current investigation, we hypothesized that SOM in the aggregates controls the heavy metals accumulation and distribution in the soil aggregates. The specific objectives of the current study were: (i) to quantify the distribution of aggregates in the different land uses, (iii) to explore the levels of heavy metals contamination in the soils of different land uses, and (ii) to measure the heavy metals content in the different sizes of soil aggregates.

2. Materials and methods

2.1. Sampling site

Soil samples were collected from the vicinity of the Khulna-Satkhira highway road under the Khulna district of Bangladesh with different land uses (Fig. 1). The collected soil samples were classified as Cambisols [40]. Samples were collected from 4 different sampling sites belonging to 4 different land uses (Table 1). The four different land use fields were a banana field, a roadside bare field, and two rice fields (rice fields– I, and II). In banana fields, bananas are cultivated over the year. In roadside bare fields, the field is kept bare over the year. In rice fields, rice is cultivated from March to April every year. Irrigation water sources in rice field-I and rice field-II are groundwater and rainwater, respectively. Mostly inorganic fertilizers are applied (TSP, Urea, DAP, Potash), and no organic amendments are applied.

2.2. Sample collection and processing

Soil samples were collected from topsoil (0–15 cm) of the above-mentioned land-use fields. At first, the upper grass layer was cleaned using a spade and then samples were taken and stored in polyethylene bags. For each land use (for example banana field land use), three same land use (three Banana field) were selected. Then, a composite soil sample from each field (each of three banana field) of each land use were collected. Thus, three composites soil samples from three field of each land use were collected. Total 12 composites soil samples from four land uses were collected. Then, unwanted materials were separated from the soil samples and

Table 1
Sampling locations, bulk soil pH, conductivity (EC) and soil organic carbon (SOC), and clay percentage of the studied land use.

Land use	Coordinate	pH	EC (dS m ⁻¹)	SOC (g kg ⁻¹)	Clay (%)
Banana field	22° 47' 45.474" N 89° 31' 8.1624" E	6.93 ± 0.2b	0.51 ± 0.1b	19.9 ± 2.8b	21.33 ± 4.18b
Fallow land	22° 47' 40.4952" N 89° 30' 36.0792" E	7.33 ± 0.2a	0.32 ± 0.14b	10.9 ± 2.2c	33.02 ± 7.42a
Rice field I	22° 47' 41.2512" N 89° 30' 40.0896" E	7.27 ± 0.1a	2.56 ± 0.64a	28.1 ± 3.8 ab	38.61 ± 3.23a
Rice Field II	22° 47' 39.5232" N 89° 30' 30.4164" E	7.23 ± 0.1 ab	0.73 ± 0.11b	30.2 ± 8a	35.33 ± 3.85a

The means ± SD (standard deviation) (n = 3) with different lowercase letters within the same column indicate significant differences among land use (P < 0.05).

broken down into smaller chunks by using a hammer and kept for air drying. Then large clods were broken down and passed through 4 and 2-mm sieves for wet sieving and soil physicochemical analysis, respectively.

2.3. Soil analysis

2.3.1. Soil physicochemical properties

The soil pH and electrical conductivity (EC) were measured with the soil: water ratio of 1:2.5 and 1:5 [21]. Soil organic carbon (SOC) was estimated through the chromic acid wet oxidation method proposed by Halder et al. [21]. Briefly, 2 g of air-dried soil sample was oxidized by 1 N $K_2Cr_2O_7$ solution. The reaction was assisted by the heat generated when H_2SO_4 was mixed with $K_2Cr_2O_7$ solution in a 2:1 ratio (v/v). The remaining $Cr_2O_7^{2-}$ was titrated with an aqueous solution of ferrous sulfate. The titer was inversely related to the amount of organic carbon present in the soil sample [21]. The clay content was determined by following the modified hydrometer method proposed by Beretta et al. [41]. Shortly, 50 g of air-dried soil sample was taken to remove organic matter by using hydrogen peroxide (H_2O_2). After plunging, the hydrometer reading was taken at 40 s and 2 h of agitation, and clay content was calculated [41].

2.3.2. Soil aggregate stability (MWD)

Soil samples were separated into four different aggregate fractions following the wet-sieving method of Liu et al. [7]. The four following water-stable aggregate fractions were separated: (i) 2–4 mm (large macroaggregates), (ii) 0.25–2 mm (small macroaggregate), (iii) 0.053–0.25 mm (micro-aggregate), (iv) < 0.053 mm fractions. Briefly, soil samples were immersed in pure water for 5 min before moving the sieve up and down to a depth of about 3 cm. Then, the sieves containing soil samples were moved up and down to a depth of about 3 cm 50 times in 2 min. The fractions remaining on each sieve were collected and oven-dried for 24 h at 45 °C. The aggregate stability indicated by mean weight diameter (MWD) was calculated as:

$$MWD = \sum_{i=1}^n X_i W_i \quad (1)$$

Where X_i is the mean diameter of each aggregate fraction, W_i is the mass proportion of the aggregate fraction remaining on each sieve, and n is the number of fractions.

2.3.3. Total heavy metals analysis

For the determination of total content of globally concerned 10 heavy metals such as Pb, Cd, Cr, Cu, Co, Ni, As, Zn, Fe, and Mn, the bulk soil and the soils of different aggregate fractions were digested in glass beakers using a mixture of concentrated nitric acid (HNO_3) and perchloric acid ($HClO_4$) in a 2:1 ratio [42]. About 5 g of air-dried soil sample was mixed with the acid mixture and heated on an electric hot plate (at ~100 °C) until the mixture was almost dry. The process was repeated until getting a transparent solution. The sample solution was then diluted to the desired volume (50 mL) in a calibrated volumetric flask by filtering and rinsing the sample container with deionized water (electrical conductivity <0.5 $\mu S cm^{-1}$) and finally, stored at 4 °C transferring it into non-transparent plastic (polyethylene) bottle. A sample blank was also prepared following the same scheme (without sample) for quality control.

From the digested sample, heavy metal contents were measured by Atomic Absorption Spectrophotometers (AAS, Models: a. AA240FS and b. SpectrAA220, Varian, Australia). Except for As, which was determined using hydride vapor generation technique with electrothermal temperature controller, all metals were measured in air-acetylene flames following our earlier work [13]. Traceable certified reference materials (CRM) were used for the preparation of calibration standards to construct the calibration curves (linearity >0.998) for the heavy metal determinations. While preparing calibration standards, 2 % aqueous HNO_3 was used for the dilution of 1000 mg/L of stock CRM solution obtained from Fluka Analytical, Sigma-Aldrich, Germany. The standard quality control samples (CRM) and method bank samples were measured after five and ten samples, respectively. The sample blanks and spike samples were also measured sequentially for analytical quality check. The percentage of spike recovery of the metals was within 93–107 %. The quantification limit of the metals Pb, Cd, Cr, Cu, Co, Ni, As, Zn, Fe, and Mn were 0.1, 0.02, 0.1, 0.1, 0.1, 0.1, 0.005, 0.05, 0.2, and 0.05 mg/L, respectively which were sufficient enough for the accurate and precise detection of the metal concentrations present in the digested soil samples by the employed AAS. Each sample was measured in trice and the mean concentration with relative standard deviation less than 5 % was considered to report.

2.3.4. Contamination factor (CF)

Soil pollution due to anthropogenic activities is indicated by contamination factor (CF) [43]. CF is defined as the metal content in soil to background metal concentration [13]. This contamination index is extensively applied to evaluate the level of soil contamination [13].

$$CF = \frac{C_m}{B_m} \quad (2)$$

where, C_m and B_m refer to the metal content in the samples and the background metal content, respectively. The value of CF is considered as follows: (I) if $CF = 0$, no pollution, (II) $CF = 1$, none to moderate pollution, (III) $CF = 2$, moderate pollution, (IV) $CF = 3$, moderate to strong pollution, (V) $CF = 4$, strong pollution, (VI) $CF = 5$, strong to very strong pollution, (VII) $CF = 6$, very strong pollution [44].

2.3.5. Pollution load index (PLI)

PLI is an integrated approach to assessing soil contamination in respective to trace metal contamination [45]. In our current investigation for the calculation of PLI, we followed the following equation as follows [10]:

$$PLI = (CF_1 \times CF_2 \times CF_3 \times \dots \times CF_n)^{1/n} \tag{3}$$

Here, $PLI > 1$ indicates pollution exists while $PLI < 1$ indicates no pollution load [46].

2.3.6. Accumulation factor (AF)

The accumulation factors (AF) of metals in different aggregates with respect to the bulk soil were calculated by following the formula.

$$AF = \frac{HM_{Aggregate}}{HM_{Bulk}} \tag{4}$$

Where $HM_{Aggregate}$ is the particular metal content in a given size of aggregate ($mg\ kg^{-1}$) and HM_{Bulk} is the particular metal content in bulk soil ($mg\ kg^{-1}$). $AF > 1.0$ indicates the accumulation of metals in certain sizes of aggregates (aggregates act as an accumulator).

2.3.7. Statistical analysis

All the statistical analysis was performed using SPSS 16.0. Shapiro-Wilk test was performed to test the data normality. Analysis of variance (ANOVA) was used to explore the impacts of different land uses on pH, EC, aggregate size fractions, MWD, SOC, and heavy metal content in the bulk soil and aggregate fractions. The least significant difference (LSD at $p < 0.05$) test was applied to assess significant differences among the means of the three replicates ($n = 3$). Pearson’s correlation was performed to explore the relationship among the investigated parameters. Principal component analysis (PCA) and cluster or dendrogram analysis were performed to explore the potential source analysis of trace metals.

3. Results

3.1. Soil physicochemical properties

The soil pH, EC, and clay content in the studied soil of different land uses are presented in Table 1. The pH values represented the neutral soil condition across the investigated land use. The fallow land showed the highest pH followed by rice field I and the lowest in the banana field (Table 1). On the other hand, the EC value in rice-I was found significantly higher than other land uses ($p < 0.05$, Table 1). Clay content in the soil samples followed in the following order of magnitude as rice field-I > rice field-II > fallow land soil > banana field (Table 1). The proportions of 2–4 mm and 0.25–2 mm fractions were significantly higher in the rice-based land uses compared to Banana and fallow land (Fig. 2; $p < 0.05$). A greater proportion of macroaggregates were found in the rice-based land use followed by Banana fields and the lowest in the fallow land soil (Fig. 2). On the other hand, the micro-aggregate content followed the following order of magnitude as fallow land > banana field > rice field-I > rice field-II. Thus, the MWD was 200 % greater in the rice-

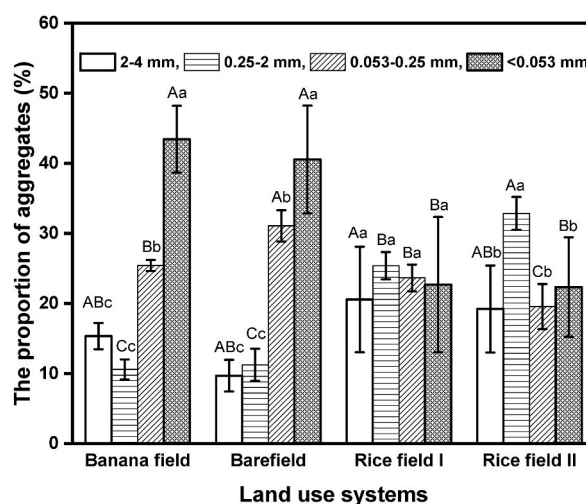


Fig. 2. Aggregate size distribution in the soil samples of different land uses. The vertical bar represents the mean ± SD (standard deviation) of three replicates ($n = 3$). Different lowercase letters denote significant difference among the proportion of different size of aggregates under the same land uses ($p < 0.05$). Different capital letters denote significant differences among the proportion of same size of aggregates under different land uses ($p < 0.05$).

based land uses compared to other land uses (Fig. 3; $p < 0.001$).

3.2. Soil organic carbon (SOC)

The SOC content in the bulk soil samples across the different land uses is presented in Table 1. The SOC content was significantly higher in the rice-based land uses followed by banana fields and the lowest SOC was found in the fallow land soil ($p < 0.05$; Table 1). The SOC in the rice-based land use was 3 and 1.5-fold higher in comparison to fallow land and banana fields, respectively ($p < 0.05$). The SOC content in different aggregate size fractions was reduced by reducing the size of the aggregates (Fig. 4; $p < 0.05$). The highest SOC in macroaggregate was found in the rice field-I, while the lowest SOC was observed in the bare field soil. In addition, in microaggregates, SOC peaked in the rice-based land use fields in comparison to other land use fields. The SOC content was reduced in the microaggregates in the following order of magnitude as rice field-II > rice field-I > banana field > fallow land soil (Fig. 4).

3.3. Heavy metals content in soil

The total Cd content in the bulk soil was lower than $<0.01 \text{ mg kg}^{-1}$ across all land use (Table 2). The Pb content was significantly higher in the banana field in comparison to other land use ($p < 0.05$; Table 2). Pd in banana fields were 5, 5, and 3-fold greater than fallow land, rice field I, and rice field II land use, respectively. The Fe, Zn, and As content ranged from 572 to 617, 6 to 11, and 206 to 312 mg kg^{-1} , respectively across the studied land use. There was no significant difference in As, Zn, and Fe content in the bulk soil samples among the land use fields of fallow land, rice field-I, and rice field-II ($p > 0.05$). The highest Cr content was found in rice field II followed by fallow land and the lowest Cr content was found in the banana field (Table 2). The Cr concentration in rice field II was 2.8, 2.1, and 2 times higher than banana, fallow land, and rice field I land use, respectively. Moreover, Ni, Cu, Co, and Mn content were also found significantly higher in the rice-based land use in comparison to banana and fallow land fields. Table 3 explore the comparison between our study (Table 2) and different studies in Bangladesh with respect to heavy metals in the agricultural field and their guidelines values. The CF of Pd, Cr, Cu, and As were greater than 1 while the CF of Co, Ni, Fe, Mn, and Zn were less than 1 (Table 4). The CF of Pb was 5 times higher in the banana field in comparison to other land uses. The CF of As was found to be highest in the rice-based land use followed by fallow land and the lowest in the banana field. Moreover, the PLI followed in the order of rice II > banana field > rice I > fallow land (Table 4).

3.4. Heavy metals in different soil aggregates

Total heavy metal content in the different aggregate fractions across different land use fields is displayed in Table 5. All the measured heavy metal content was reduced with reducing aggregate size except Cu and Zn ($p < 0.05$), while there was no impact of aggregate size on Co accumulation in the investigated land use ($p > 0.05$). Large macro and small macro aggregates showed higher content of trace metals followed by silt and clay fractions and the lowest content was found in the microaggregates in the investigated land use (Table 5). Pb and As content was reduced significantly with reducing aggregate size in all land use except bare field ($p < 0.05$). Pb and As content was reduced by 2.5, 1.1, 1.30, 1.5, and 8.21, 1.11, 1.94, and 1.55 times lower in the banana, bare, rice I, rice II, respectively due to the reduction of aggregates size from large macroaggregates to silt plus clay size aggregates (Table 5). Fe and Mn content were increased by 18, 20 in the banana, bare field, and 45, 27 % in the rice I and rice II, respectively as a result of increasing aggregate size from <0.053 to $>4 \text{ mm}$ aggregates (Table 5). In contrast, Cu and Zn content was increased by 1.13, 1.25, 1.53, and 1.60

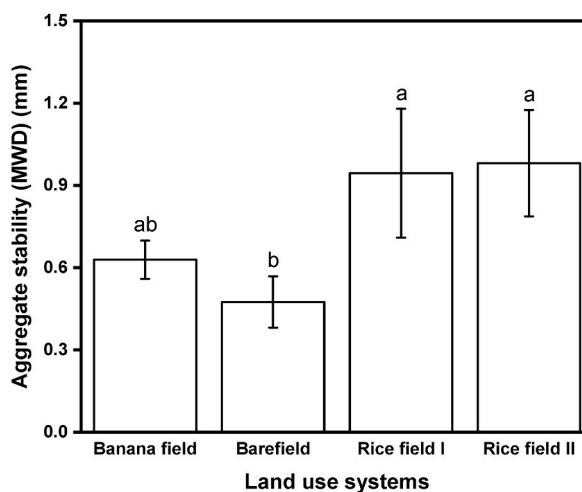


Fig. 3. Soil aggregate stability (MWD) under different land uses. The vertical bar represents mean \pm SD (standard deviation) of the three replicates ($n = 3$). With different lowercase letters above the bar denote significant differences among the MWD of different land uses at $p < 0.05$.

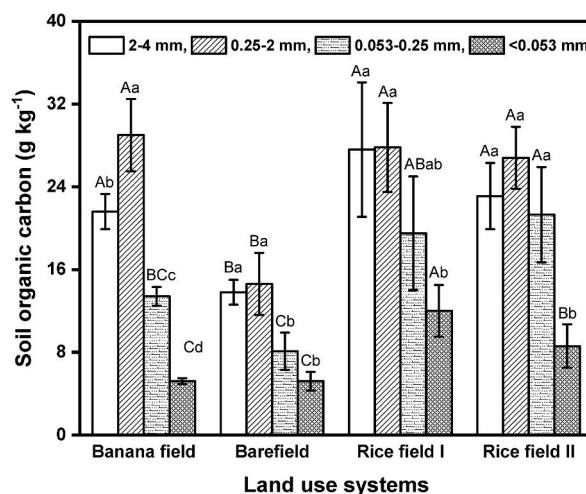


Fig. 4. The distribution of SOC in the soil aggregate size fraction in soil samples of different land uses. The vertical bar represents the mean \pm SD (standard deviation) of three replicates ($n = 3$). Different lowercase letters denote significant difference among the SOC of different size of aggregates under the same land uses ($p < 0.05$). Different capital letters denote significant differences among the SOC of same size of aggregates under different land uses ($p < 0.05$).

Table 2

Total heavy metals content across the bulk soil of the studied land use.

Heavy metals (mg kg^{-1})	Banana field	Fallow land	Rice field I	Rice field II
Pb	$1179 \pm 185a$	$242 \pm 55b$	$238 \pm 52b$	$356 \pm 113b$
Cd	<0.01	<0.01	<0.01	<0.01
Cr	$93 \pm 15b$	$131 \pm 51 ab$	$121 \pm 21b$	$261 \pm 143a$
Cu	$45 \pm 7b$	$38 \pm 12b$	$57 \pm 13 ab$	$74 \pm 1 a$
Co	$7 \pm 1b$	$8 \pm 1 ab$	$8 \pm 1 ab$	$9 \pm 1a$
Ni	$21 \pm 4b$	$25 \pm 5b$	$30 \pm 5a$	$29 \pm 1a$
As	$206 \pm 53a$	$240 \pm 34a$	$266 \pm 57a$	$312 \pm 82a$
Zn	$6 \pm 1a$	$6 \pm 3a$	$6 \pm 2a$	$11 \pm 6a$
Fe	$572 \pm 85a$	$617 \pm 46a$	$653 \pm 5a$	$640 \pm 16a$
Mn	$17 \pm 2b$	$23 \pm 2 ab$	$28 \pm 6a$	$28 \pm 3a$

The means \pm SD (standard deviation) ($n = 3$) with different lowercase letters within the same row indicate significant differences among the different land use ($P < 0.05$).

times greater with reducing aggregate size from large macroaggregates to silt plus clay fractions. Moreover, Cr and Cd concentrations were also reduced with reducing aggregate size but were not significant except for rice II ($p > 0.05$). On the other hand, aggregate size did not influence the Co and Ni accumulation in all investigated land use ($p > 0.05$; Table 5). Pb and Cd content were found to be higher in the aggregate fractions of banana fields while Cr content was dominated in the aggregates of rice-based land use. There was no significant impact of land use on Co, Ni, Zn, Fe, and Mn accumulation in the different aggregates ($p > 0.05$).

3.5. Heavy metals in aggregates in relation to bulk soil

Fig. 5 shows the accumulation factors (AF) of Pb, Cd, Cr, Cu, Co, Ni, As, Zn, Fe, and Mn in soil aggregates across different land uses. The results revealed that even for the same elements, AF values from different soil aggregates (>2.00 mm, 0.25 – 2.00 mm, 0.053 – 0.25 mm, <0.053 mm) varied across all land use (banana field, fallow land, rice field-I, and rice field-II). The AF of Pb and Cr in all aggregate fractions across the studied land was very lower (<1) (Fig. 5). AF of Cu and Zn were higher than 1 in all sizes of aggregates over all the land use (Fig. 5). Within soil aggregates, greater AF values (>1) of Cu and Zn were found in <0.053 mm aggregates in comparison to 0.25 – 4 mm aggregates in all land use. In contrast, the AF values of As, Fe, and Mn were observed >1 in the macroaggregates of all land use except rice II. In the case of Fe and Mn, the AF was >1 in all land use except rice II. For, Co and Ni the AF values were >1 only for banana and bare fields, while rice-based cropping fields showed lower AF (<1). The AF of Co and Ni were found >1 in the <0.053 mm size aggregates.

3.6. Multivariate analysis among the trace metals

Multivariate analysis (Pearson's correlation (CM), principal component analysis (PCA), and cluster analysis (CA)) have been

Table 3Literature values of trace elements concentration (mg kg^{-1}) in agricultural soils in the different parts of Bangladesh along with their guideline values.

Study sites	Pb	Cd	Cr	Cu	Co	Ni	As	Zn	Fe	Mn	References
Agricultural field (Potuakhali)	24	3.1	19	34	N/A	27	9.3	N/A	N/A	N/A	Islam et al., 2017
Vegetable cultivated soil (Dhaka)	41.5	0.87	50.1	58.7	N/A	16.1		286	N/A	N/A	Mizan et al., 2023
Farmland (Dhaka) dry season	28	0.007	50	N/A	N/A	N/A	4073	N/A	N/A	N/A	Rahman et al., 2012
Farmland (Dhaka) wet season	10	1.0	34	N/A	N/A	N/A	2326	N/A	N/A	N/A	Rahman et al., 2012
Dhaka (Hazariabagh)	50	0.45	976	N/A	N/A	N/A	2.0	N/A	N/A	N/A	Mottalib et al., 2016
Agricultural land (Dhaka)	66	14	1457	57	50	47	20	54	12232	455	Bhuiyan et al., 2021
Agricultural land (Jamalpur)	17	0.32	63	32	N/A	21	N/A	79	31882	472	Bushra et al., 2022
Guidelines values											
Dutch soil quality standard (Target value) (VROM, 2000)	85	1.0	100	36	N/A	35	29	N/A	N/A	N/A	
Dutch soil quality standard (Intervention value) (VROM, 2000)	530	12	380	190	N/A	210	55	N/A	N/A	N/A	
WHO/FAO (2001)	100	3.0	100	100	N/A	50	N/A	300	N/A	N/A	
WHO (1989)	10	0.3	N/A	N/A	N/A	N/A	14	50	N/A	200	
Ministry of Environmental Protection of the People's Republic of China (2018)	120	0.6	200	100	N/A	100	N/A	250	N/A	N/A	

Table 4

Contamination factor (CF) and pollution load index of investigated heavy metals across the bulk soil of the studied land use.

Heavy metals (mg kg^{-1})	Banana field	Fallow land	Rice field I	Rice field II
Pb	25.63	5.26	5.17	7.74
Cd	–	–	–	–
Cr	1.03	1.46	1.34	2.90
Cu	1.00	0.84	1.27	1.64
Co	0.37	0.42	0.42	0.47
Ni	0.31	0.37	0.44	0.43
As	61.49	71.64	79.40	93.13
Zn	0.40	0.40	0.40	0.73
Fe	0.03	0.03	0.03	0.03
Mn	0.45	0.61	0.74	0.74
PLI	90.71	81.02	89.22	107.82

performed among the metal content of the study land uses. The results of the correlation matrix are displayed in Fig. 6. SOC showed a positive correlation with MWD, Cu, and As, while MWD showed a positive relationship with As, Mn, Cu, and Cr. Among metals, As showed a strong and significant correlation with Co, Ni, and Mn, whereas Mn showed a significant relationship with Pb, Fe, Co, and Ni. Cr showed a significant positive correlation with Cu and As ($p < 0.01$). Pb was negatively related to Mn, Ni, and Co, while Zn did not show any correlation with any metals or soil properties. The results of the PCA for trace element concentrations in soils of different land uses are shown in Table 6 and Fig. 7. The PCA of elements in the soils of different land uses explained the cumulative variance of 83.86%. Three principal factors were extracted from the analytical data of trace elements. The soils of different land use, the Co, Ni, Mn, and As were positive, Pb was negatively loaded in PC1 and explained 43.41% of the variance, while the PC2 which loaded positively with Zn explained 20.70% of the total variance (Table 6). The PC3 loaded positively with Cr and Cu and explained 19.77% of the variance. The results of CA of the measured trace elements for soils of different land use demonstrated a similar trend of grouping and depicted clearly with a dendrogram (Fig. 8) in which three main clusters (with the Phenon line set to a rescaled distance of about 10) can be visualized. The elements such as Co, Ni, Mn, and As, constitute the same cluster (Cluster-1) whereas the elements viz., Cr, Cu, Fe, and Zn were grouped into another cluster (Cluster-2). Finally, Pb alone showed a separate cluster (Cluster-3). These elemental grouping similarities were strongly supported by the results of the PCA. In-depth observation from CA was that Co and Ni were closely related with each other rather than Mn and As (Fig. 8). Cu was closely associated with Cr, while Fe was more closely related with Zn (Fig. 8).

Table 5
Total heavy metals content in different aggregate fractions across the studied land use systems.

Heavy metals	Aggregate Size (mm)	Banana field (mg kg ⁻¹)	Barefield (mg kg ⁻¹)	Rice field-I (mg kg ⁻¹)	Rice field-II (mg kg ⁻¹)	Level of sig. (p < 0.05)
Pb	>2.00	330 ± 47Aa	37 ± 17Ba	49.56 ± 19Ba	54 ± 12Ba	*
	0.25–2.00	209 ± 31Ab	49 ± 26Ba	26.89 ± 4Bab	22 ± 3Bb	*
	0.053–0.25	125 ± 22Ab	28 ± 19Ba	13.61 ± 11Bb	24 ± 6Bab	*
	<0.053	130 ± 30Ab	29 ± 7Ba	44.69 ± 15Bab	36 ± 20Bab	*
Level of sig. (p < 0.05)	*	ns	*	*		
Cd	>2.00	0.28	0.05	0.03	<0.01	ns
	0.25–2.00	1.25	<0.01	<0.01	<0.01	ns
	0.053–0.25	0.03	<0.01	<0.01	<0.01	ns
	<0.053	0.02	<0.01	<0.01	<0.01	ns
Level of sig. (p < 0.05)	ns	ns	ns	ns		
Cr	>2.00	5 ± 1Aa	4 ± 2Aa	10 ± 5Aa	11 ± 9Aab	ns
	0.25–2.00	5 ± 2Ba	6 ± 4Ba	10 ± 3Ba	23 ± 8Aa	*
	0.053–0.25	3 ± 0.2Aa	4 ± 2Aa	8 ± 3Aa	10 ± 6Aab	ns
	<0.053	3 ± 0.7Ba	5 ± 0.6ABa	5 ± 0.9Aba	5 ± 1Ab	*
Level of sig. (p < 0.05)	ns	ns	ns	*		
Cu	>2.00	161 ± 26Ab	100 ± 14Aa	111 ± 64Aab	94 ± 30Aa	ns
	0.25–2.00	461 ± 72Aa	111 ± 81Ba	71 ± 8Bb	92 ± 32Ba	*
	0.053–0.25	64 ± 10Bc	100 ± 26ABa	89 ± 8ABb	113 ± 38Aa	*
	<0.053	182 ± 53Ab	125 ± 9Aa	170 ± 2Aa	149 ± 56Aa	ns
Level of sig. (p < 0.05)	*	ns	*	ns		
Co	>2.00	8 ± 1Aa	6 ± 3Aa	8 ± 1Aa	6 ± 1Aa	ns
	0.25–2.00	7 ± 1Aa	8 ± 1Aa	8 ± 0.3Aa	7 ± 1Aa	ns
	0.053–0.25	6 ± 1Aa	7 ± 2Aa	8 ± 1Aa	7 ± 1Aa	ns
	<0.053	7 ± 1Ba	9 ± 0.3Aa	8 ± 0.2ABa	7 ± 1Ba	*
Level of sig. (p < 0.05)	ns	ns	ns	ns		
Ni	>2.00	19 ± 5Aa	17 ± 7Aa	25 ± 5Aa	23 ± 3Aa	ns
	0.25–2.00	22 ± 4Aa	27 ± 6Aa	29 ± 1Aa	26 ± 3Aa	ns
	0.053–0.25	17 ± 10Aa	23 ± 8Aa	25 ± 2Aa	26 ± 4Aa	ns
	<0.053	28 ± 8Aa	29 ± 1Aa	27 ± 1Aa	25 ± 4Aa	ns
Level of sig. (p < 0.05)	ns	ns	ns	ns		
As	>2.00	230 ± 31Aab	210 ± 105Aa	339 ± 114Aa	231 ± 62Aa	ns
	0.25–2.00	282 ± 30ABa	326 ± 54Aa	227 ± 33Bab	224 ± 9Ba	*
	0.053–0.25	209 ± 26Ab	224 ± 49Aa	178 ± 20Ab	181 ± 16Aab	ns
	<0.053	203 ± 47ABb	297 ± 93Aa	175 ± 12Bb	149 ± 22Bb	*
Level of sig. (p < 0.05)	*	ns	*	*		
Zn	>2.00	17 ± 4Abc	15 ± 1Aa	14 ± 11Aa	12 ± 5Aa	ns
	0.25–2.00	26 ± 6Ab	31 ± 20Aa	10 ± 3Aa	10 ± 7Aa	ns
	0.053–0.25	13 ± 3Ac	16 ± 8Aa	8 ± 1Aa	13 ± 6Aa	ns
	<0.053	37 ± 6Aa	17 ± 2Ba	13 ± 3Ba	16 ± 10Ba	*
Level of sig. (p < 0.05)	*	ns	ns	ns		
Fe	>2.00	727 ± 105Aab	783 ± 250Aab	721 ± 180Aa	652 ± 69Aa	ns
	0.25–2.00	957 ± 351Aa	944 ± 178Aa	613 ± 11Aa	565 ± 22Aa	ns
	0.053–0.25	423 ± 83Bb	553 ± 81Ab	578 ± 15Aa	643 ± 66Aa	*
	<0.053	599 ± 135Aab	629 ± 7Ab	646 ± 79Aa	607 ± 112Aa	ns
Level of sig. (p < 0.05)	*	*	ns	ns		
Mn	>2.00	18 ± 2Ba	22 ± 2Ba	34 ± 8Aa	26 ± 3ABa	*
	0.25–2.00	20 ± 4Ba	28 ± 6Aa	28 ± 3Aab	25 ± 1ABa	*
	0.053–0.25	16 ± 3Aa	21 ± 6Aa	21 ± 2Abc	23 ± 4Aab	ns
	<0.053	21 ± 3ABa	24 ± 1Aa	19 ± 2Bc	19 ± 1Bb	*
Level of sig. (p < 0.05)	ns	ns	*	*		

The means ± SD (standard deviation) (n = 3) with different lowercase letters within the same column indicate significant differences among the different size of aggregate fractions under the same land use system (P < 0.05). Different capital letters within the same row indicate significant differences among land use (P < 0.05).

4. Discussions

4.1. Impacts of land use on soil aggregate stability

Soil organic carbon (SOC) and aggregate stability (MWD) were found to be higher in rice-based land use in comparison to other investigated land use. No organic matter was added to the bare soil and banana field soil, while rice roots and stable were kept in the rice-based land use. As a result, SOC in the rice-based land use was higher than that of other investigated land use. Consequently, MWD and SOC were significantly correlated with each other (P < 0.001; r = 0.88). This result was consistent with the findings reported by Halder et al. [21] and Rahman et al. [6]. Halder et al. [21] reported that MWD was increased by 2 times in the straw treatments in comparison to the control leading to a positive correlation between SOC and MWD in a field study under clay soil of Bangladesh. Rahman et al. [6] also reported that MWD increased resulting significant positive relationship between SOC and MWD under 28-day

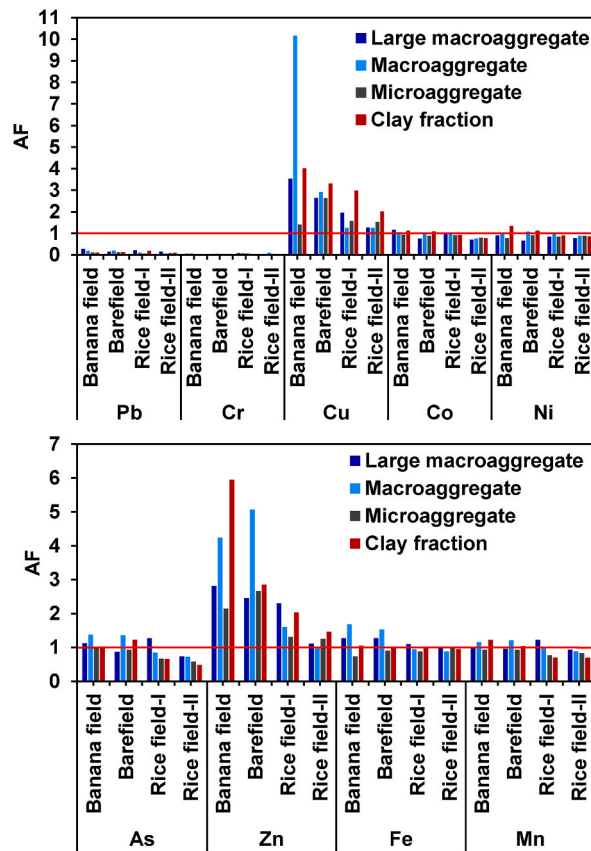


Fig. 5. Accumulation factor (AF) of heavy metals in different aggregate fractions across the studied land uses. Every bar represents mean value (n = 3). AF>1.0 indicates the accumulation of metals in certain sizes of aggregates.

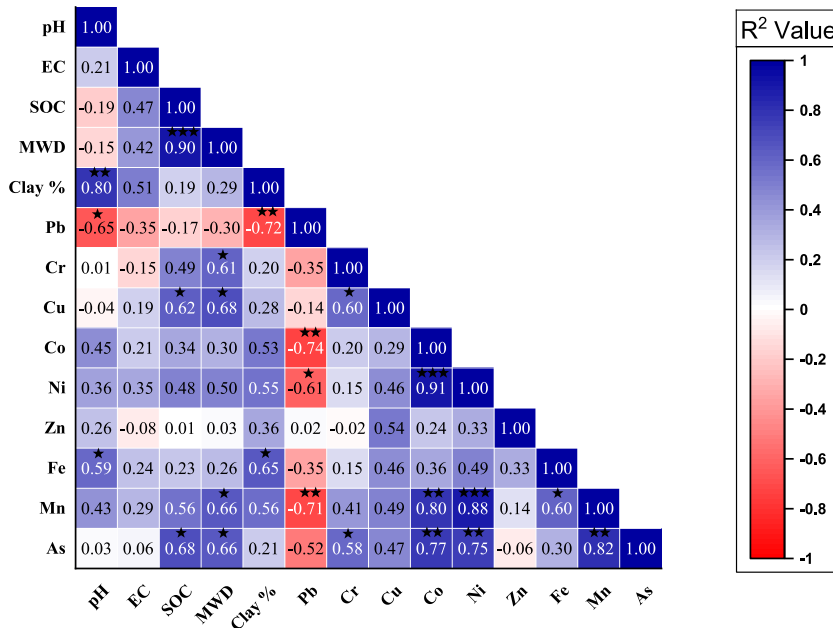
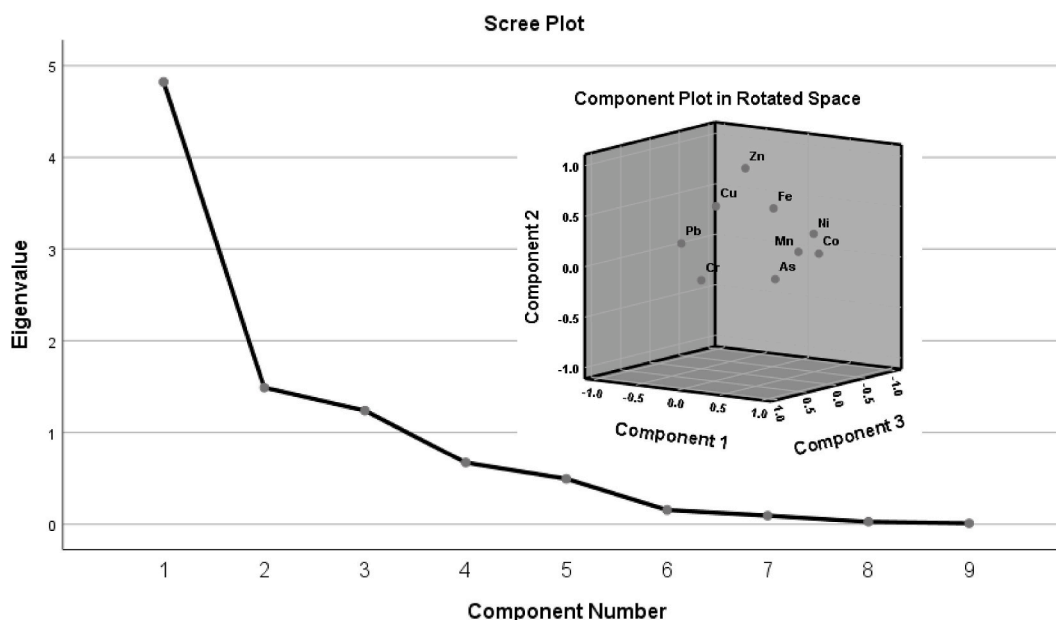


Fig. 6. Heat map of Pearson correlation matrix among soil properties and heavy metal contents across the studied land uses. The blue, red and white color correspond positive, negative and neutral correlation, respectively. *, **, and *** represent significance levels at $p < 0.05$, $p < 0.01$, and $p < 0.001$, respectively. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

Table 6

Varimax rotated factor loadings and communalities of trace elements in soils (strong loadings are in bold face).

Parameters	PC1	PC2	PC3	Communalities
Pb	-0.826	0.080	-0.127	0.704
Cr	0.166	-0.029	0.950	0.932
Cu	0.151	0.664	0.679	0.925
Co	0.932	0.180	0.039	0.902
Ni	0.883	0.376	0.059	0.925
Fe	0.427	0.586	0.090	0.533
Zn	-0.009	0.919	-0.052	0.847
Mn	0.876	0.232	0.309	0.917
As	0.759	-0.018	0.536	0.863
Eigenvalue	4.823	1.489	1.237	
% of total variance	43.408	20.701	19.766	
Cumulative % of variance	43.408	64.109	83.875	

**Fig. 7.** Principal component analysis and scree plot of measured trace metals in rotated space.

incubation study using the red clay soil of China. Rice root and stable residue, source of soil organic matter, and their incorporation increase the soil microbial function and their decomposition [6,23]. Microbial degradation of added organic matter (rice roots and stables) produces extracellular polymeric substances (EPS) [47]. These ESP compounds are glue-like polysaccharides, which encrust the smaller soil particles and thereby leading to formation of microaggregates [47]. The microaggregates binding together form macroaggregates resulting enhance the MWD [5]. Macroaggregates are formed enmeshing the microaggregates by plant roots and fungal hypha [48]. Among the microbial diversity, the bacterial community enhances the aggregate stability by gluing the clay and silt-size soil particles while fungi enhance the MWD by enmeshing the larger particles [47,48]. Particularly, mycorrhiza fungal hypha enmeshes and entangle the soil particle for soil aggregation [49]. After turnover of mycorrhiza fungal hyphae synthesizes glomalin protein, which enhances the MWD through gluing the soil particle [48]. Moreover, rice residue profound with cellulose, O-alkyl and di-O-alkyl carbon, which enmeshes the soil particle and increases the MWD [23,50]. In addition, the hydrophobic nature of organic matter enhances the stability of soil aggregates by reducing the wettability of aggregates [51]. We found that the higher content of organic carbon in the larger soil aggregates (large and small macroaggregates). This result was corroborated by the findings of Rahman et al. [6], Halder et al. [21], and Bravo-Garza et al. [52]. Rahman et al. [6] found that the macro-aggregates retained a greater amount of added carbon in comparison to < 0.25 mm aggregates using ^{13}C isotopic tracer technique. Bravo-Garza et al. [52] concluded that this carbon is stored as particulate organic matter ($\approx 50\%$ of the total soil carbon) in the macro-aggregates. This particulate organic matter is an easily decomposed substance leading to a faster turnover rate of macroaggregates [5,53]. Finally, soil aggregation can be enhanced with the addition of organic amendments (rice root and stables), which can enhance the SOC [20]. Islam et al. [22] found that application of balanced inorganic fertilizer with organic amendments has the potentiality to enhance SOC significantly boosting microbial activity than soul crop residue addition. This inorganic fertilization is the potential source of heavy metals contamination in agricultural soil, which is leading to ecological and human health risk [25].

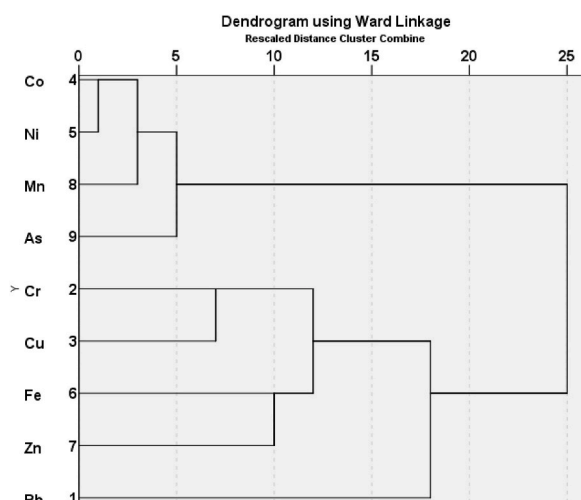


Fig. 8. Dendrogram obtained by hierarchical clustering analysis of analyzed trace elements from different land use (Note: Cd content was excluded during analysis due to very lower concentration).

4.2. Heavy metal accumulation and its ecological risks

The evaluated heavy metals contents were found to be higher in the rice-based land use except Pb in comparison to other land use (Table 2). Higher content of Pb in fallow land and banana field (very close to highway than rice field) than rice field soil has resulted from vehicle emission and Pb deposition [54]. The concentration of Cu, Co, Ni, Cr, and Mn contents were significantly greater in the rice based agricultural field compared to banana and fallow land. Iig et al. [55] found that these differences as a result of biogeochemical cycling and anthropogenic influence. Another possible reason is higher content of soil organic matter in the rice-based land use than banana and fallow land, which can form chelation with metals [56]. On the other hand, Ogunwole and Ogunleye [25] suggested that the higher content of Cu, Ni, Mn, and Cr in the agricultural rice field in comparison to fallow and banana field soil as a result of anthropogenic application of chemical fertilizer. In the current study, As and Pb content were higher than that of a study conducted in the agricultural field of different parts of Bangladesh (Table 4). The trace metal contents were within the recommended values of different organizations except As and Pb (Table 4). The CF values of Pb and As were >1 in all land use, which indicates very strong pollution of Pb and As by anthropogenic-induced metal deposition [44]. Wang et al. [26] found that a CF value greater than 2 was an indication of anthropogenic deposition of heavy metals. All the investigated land use was located on the side of the Khulna-Satkhira highway thereby leading to higher CF of Pb in all land use [13]. Pb is emitted from the exhausts and dust of private cars, long-distance transport, trucks, and other vehicles that cause potential Pb emission and contamination in the nearby agricultural field [57–59]. Another possible reason for Pb contamination in investigated agricultural rice and banana cultivated field was indiscriminate use of inorganic fertilizer to enhance crop productivity [25,58,60]. Moreover, we found CF of As is also >1 in all investigated land use (Table 4). Southwest Bangladesh is very near to the sea, which causes potential limitation of fresh water for rice cultivation [61]. The farmers use As-contaminated groundwater for irrigation purposes in this area resulting enhances the As content in the agricultural field [61]. Another possible reason for arsenic contamination is the application of As-contaminated fertilizer and pesticides during cultivation in the field [62]. This higher values of As and Pb can lead to phytotoxicity, ecological risk and human health risk [63–68]. The surface and subsurface water bodies also can be contaminated due to runoff and percolation during rainfall [69–71]. Subsequently, these metals migrate to the food chain and accumulate in human body thus leading to different diseases such as abdominal pain, asthma, cancer, respiratory, kidney and cardiovascular disorder, arsenicosis, etc. [67,72,73].

Moreover, it has been found that the soil aggregate stability was positively correlated with only As, Mn, Cu, and Cr accumulation, which indicates that all the metals accumulation in soil do not depend on the soil aggregate stability [28,74]. This result was consistent with Cui et al. [28], who found that trace metals accumulation does not depend on MWD in the four-year field experiment under different organic and inorganic amendment applications. Aggregate stabilization depends on different biotic (e.g., organic matter; microbial diversity), abiotic factors (e. g., Fe/Al oxides; clay mineralogy, wetting drying cycles, etc.), management practices in different soil conditions thereby leading to the different magnitude of MWD and metal content in the different land use [28,47].

4.3. Distribution of heavy metals in different soil aggregates

MWD of soil aggregates is the most widely used indicator to explore the water erosion resistance of soil [75]. The soil of southwestern is very prone to water erosion due to lower SOC and MWD [14]. Wet sieving methods of soil aggerate separation is a useful procedure not only to quantify the SOC and microbial function but also the heavy metals distribution in the soil [76]. Heavy metals accumulation in the soil aggregates varied based on specific metals [74]. Fe, Mn, and As tended to accumulate in larger aggregates (AF is >1), which acted as an accumulator for these respective metals [74]. Results showed that the Pb, Cd, Cr, Ni, As, Fe, and Mn content

were found to be higher content in the >0.25 aggregates followed by <0.053 mm aggregates, and the lowest content was found in the $0.25\text{--}0.053$ mm aggregates. Our study findings were consistent with previous studies findings [60,77]. The >2 and $0.25\text{--}2$ -mm fractions contained a higher amount of soil organic matter (SOM). Newly added heavy metals in soil are initially absorbed by micro-aggregates coupling with finest particles surface [77]. Based on the aggregate's hierarchy model, the micro-aggregates and clay plus silt attached together by SOM form macroaggregates thereby leading to higher content of heavy metals in macroaggregates [5, 28]. Moreover, SOM contributes a lot of non-specific and specific binding sites for heavy metals and then forms stable SOM-metals complex resulting increase in the heavy metals in macroaggregates [38,39]. Shirvani et al. [39] suggested that humic and fulvic acids play a crucial role in the accumulation of heavy metals in soil. Humic acid competes with clay minerals for metal sorption resulting in the release of different metals from clay particles by humic acids in turn attenuate the metal content in the finest particles [78].

Moreover, an AF value greater than 1 expressed that the soil aggregate was an accumulator, while if <1 , the aggregate was an excluder [74]. For Cu and Zn, soil smaller aggregates acted as a Cu accumulator (Fig. 4). Cu and Zn accumulated in all soil aggregates an average of 3 times higher than bulk soil. On the other hand, we found that Cu and Zn were accumulated in the finest particles (Table 5). These findings were consistent with the results of previously published reports [28,59,79]. Cui et al. [28] found that Cu was significantly accumulated in the <0.053 mm aggregates fractions in comparison to larger soil aggregates. The finest particles have a greater specific surface area, which enhances the proportion of reactive substrates [79]. The aggregates size <0.053 -mm fundamentally comprised of silt plus clay fractions, and metals may act as the binding agents for clay-polyvalent metal-organic matter complexes [75]. Yu et al. [80] found that the trace metals tended to accumulate in the finest particles due to their greater reactive surface coupling with surface different functional groups. Thus, heavy metals can be easily accumulated on their large surfaces by adsorption, forming the chelating complexes with the organic-mineral colloidal particles in the finest fractions [77]. Shen et al. [81] suggested that smaller soil fractions have greater affinities for the sorption of trace elements due to the presence of Fe/Mn oxides in higher proportions. The dominant clay minerals in his areas are 2:1 expanding type (smectite) [82]. The negative charge of this mineral has the potential for the retention of heavy metals [59].

A lot of studies showed that SOM influences speciation, movement, and availability of metals [39,74]. However, our study findings indicated that greater content of SOM in soil aggregates is not the only controlling factor of heavy metal accumulation and distribution in the soil aggregates [74]. Metals accumulation also depends on the metal species and size of the particles [28,74]. The soil properties, texture especially clay content, and composition of SOM, Fe/Mn/Al oxide, influence metal accumulation in clay soil, while SOM is considered as the main factor for heavy metals distribution in soil aggregates in the sandy soil [83].

4.4. Potential sources of heavy metals

Multivariate statistical tools (PCA, CA, CM) were used to identify the possible sources of the heavy metal contamination in the studied different land uses following our previous work [13]. The loading in the principal components is assigned as weak, moderate, and strong for the factor scores of $0.30\text{--}0.50$, $0.50\text{--}0.75$, and >0.75 , respectively [84]. The significant loadings of the trace elements in the same principal component indicate their common source of origin in the studied soil samples under different land uses [42,85]. The strong positive loading in PC1 of Co, Mn, Ni, and As metals implies their common sources as a results of anthropogenic activities [62, 86]. The soil of southwest Bangladesh has been suffering from P deficiency for decades [87]. The indiscriminate use of P fertilizer to enhance rice yield in the soil of these land uses may contribute to these metal contamination [62]. In addition, another agricultural input in this land use field is pesticide which has recently been applied in large quantities to reduce pest attacks in rice fields [88]. These pesticides might be another possible source of these metals in the studied land use [88,89]. On the other hand, Cr, Cu, Fe, and Zn content showed cluster-2 implying their identical sources. The sources of these elements are either natural or anthropogenic [90–94]. The soil sampling areas under different land uses were free from industrial activities leading to reduced metal contamination from anthropogenic activities [13]. These metals (Cr, Cu, Fe, and Zn) content in the study areas might be geologic in origin [90,91]. On the contrary, Pb showed an entirely identical source to the other two groups of identified trace elements. A highway (Khulna Satkhira highway road) was very near the sampling sites [13]. Different types of vehicles like buses, trucks, private cars, taxis, and three-wheeler lead-acid batteries containing bikes are traveled very frequently on the highway [13]. These vehicles emit a large concentration of Pb from their fumes and waste [95,96]. The Pb release from these vehicles might be the potential source of Pb in the studied land use [54].

5. Conclusions

Aggregate stability was enhanced in the rice-based land use, which was mainly contributed by soil organic carbon. All the measured heavy metals in the investigated land uses were found to be greater in the rice-based land use except Pb. Moreover, Pb and As contamination in the studied land uses were found which can contaminate soil, water and plant. Subsequently, their migration through food chain can promote different human health risks such as abdominal pain, cancer, asthma, kidney and cardiovascular disorder. To reduce ecological and health risk, it is essential to reduce the bioavailability. Local farmers can apply in-situ stabilization and fixation techniques to attenuate the heavy metals bioavailability using various chelating and stabilizing agents, which can reduce their mobility and convert them toxic to non-toxic form. The Pd, As, Cd, Fe, and Mn were increased with increasing SOC in aggregates coupling with the size of aggregates. On the other hand, the contents of Cu and Zn were increased with reducing aggregate size, while Ni and Co accumulation had no relation with aggregate size. Furthermore, macroaggregate acted as an accumulator for the Fe, Mn, and As, while all the measured aggregates acted as an accumulator for Cu and Zn. The study findings demonstrate that MWD and SOC

content were not only controlling factors of trace metals accumulation in the soil aggregates but also aggregates size and individual metal species were also important factors for heavy metals distribution and accumulation in the soil aggregates.

Data availability

Data will be available upon reasonable request from the corresponding authors.

CRediT authorship contribution statement

Md. Piash Sarder: Writing – original draft, Formal analysis, Data curation. **Md. Kamruzzaman:** Software, Investigation, Formal analysis, Data curation. **Md. Abu Bakar Siddique:** Methodology, Formal analysis, Data curation, Visualization, Writing – review & editing. **Milton Halder:** Writing – review & editing, Writing – original draft, Visualization, Supervision, Methodology, Investigation, Funding acquisition, Conceptualization.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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