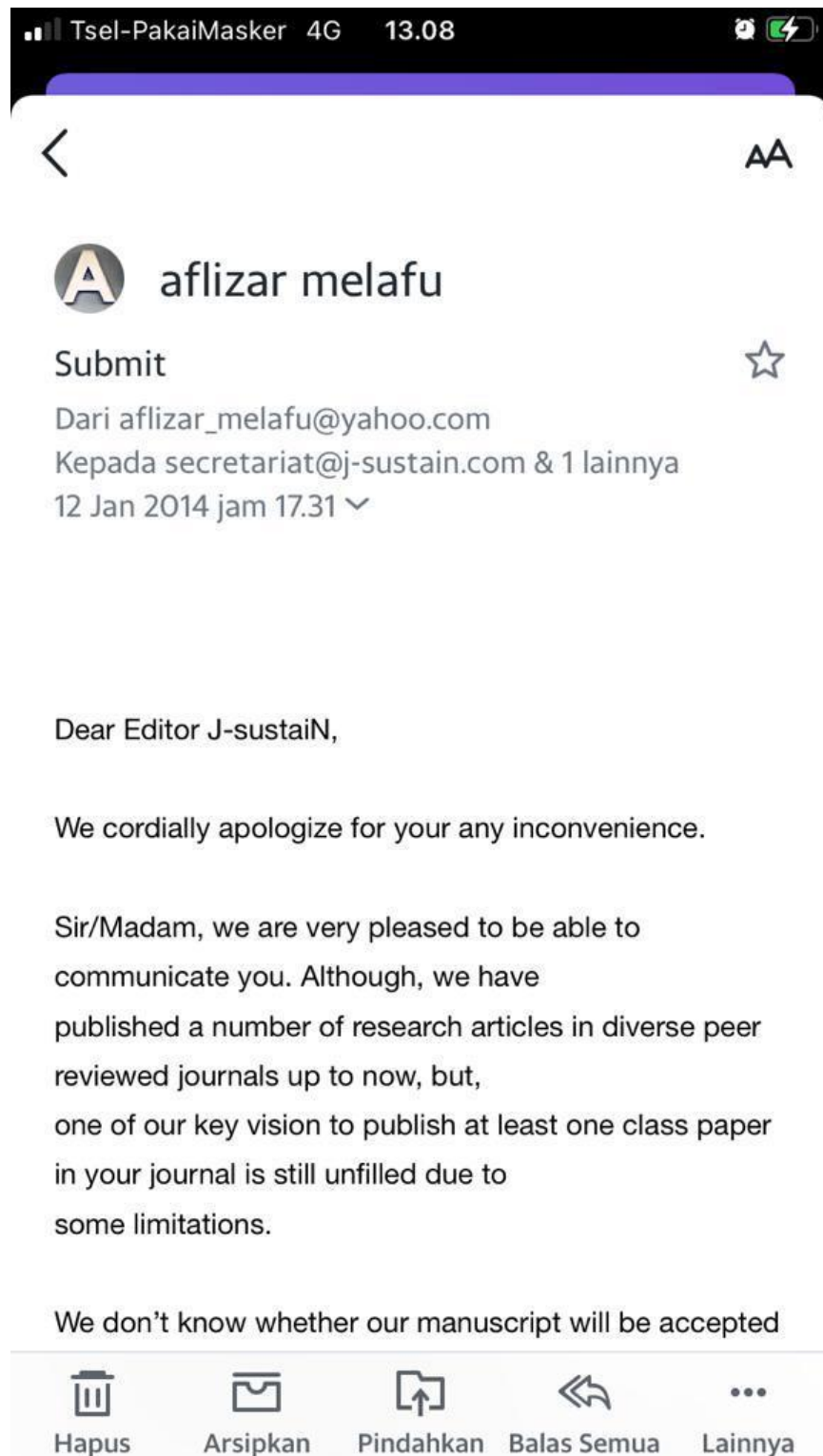


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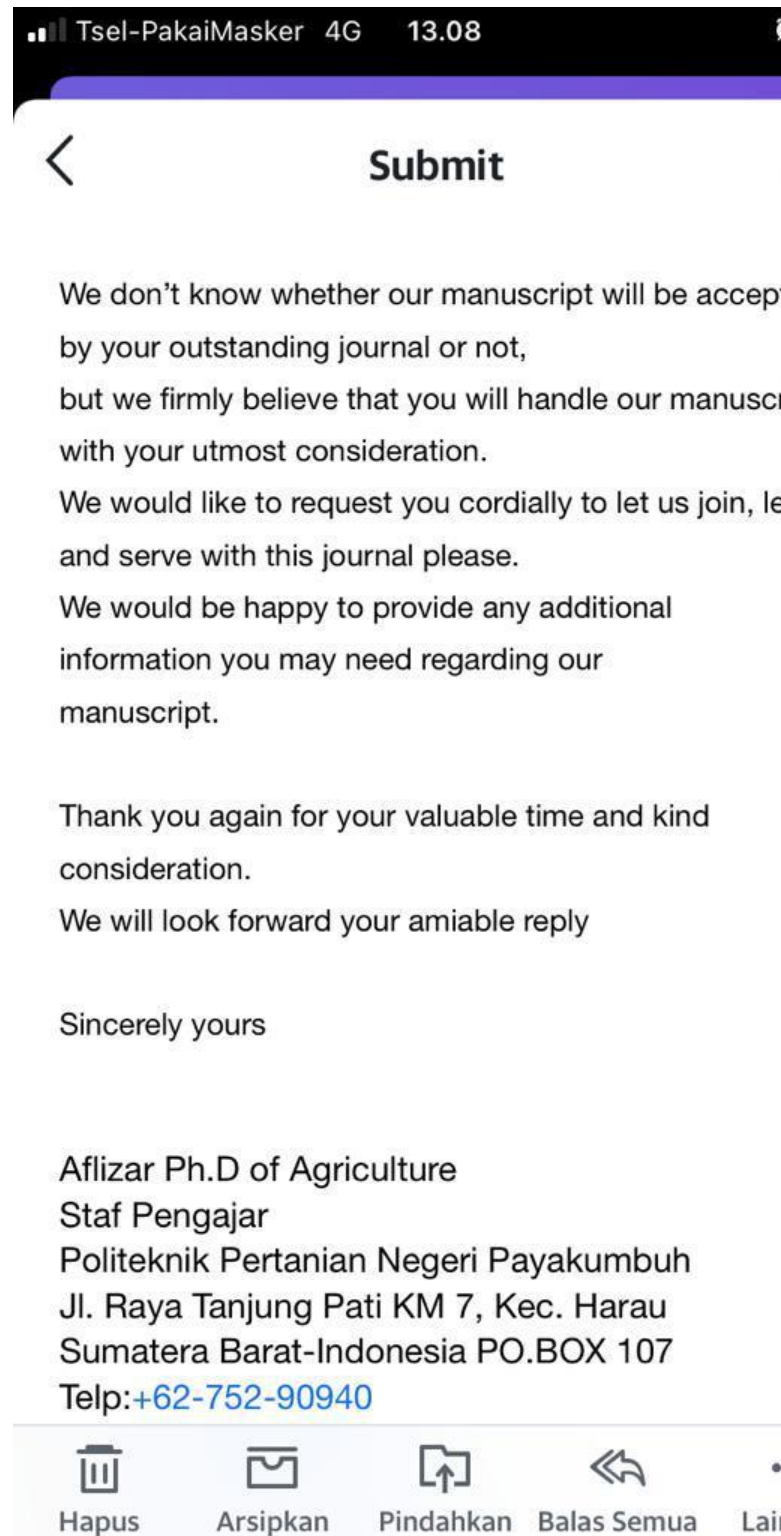


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Trace metal concentrations in An Agricultural Watershed: Case study in the Sumani Watershed, West Sumatera Indonesia

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Abstract

Selected trace metals investigation of agricultural soil at sawah, vegetable and river sediment in Sumani watershed, West Sumatra, Indonesia were conducted to determine their abundances and to assess contamination levels. pH, CEC and TC values indicates moderate to high soil fertility status. Average concentrations of Pb, Zn, Cu, TiO₂, Fe₂O₃ and P₂O₅ were greater than those of Bulk Composition Crust (BCC) and UCC in most areas. Enrichment factor values at Sawah confirms depletion to minimal enrichment with most metals, while vegetable area indicates moderate enrichment with Pb, Cu and Rb, and depletion to minimal enrichment for Zn, Ni, Cr, V and Sr. Igeo values for Pb, at Sawah were unpolluted to moderately polluted but practically unpolluted for Zn, Cu, Ni, Cr, V, while vegetable area were moderately polluted with Pb, unpolluted to moderately polluted with Cu and Rb and and practically unpolluted for Zn, Ni, Cr, V and Sr. Elevated values of Pb in vegetable area, possibly due to volcanic ash from mount Talang, excess application of fertilizers and pesticides, and high contents of Pb in gasoline, Indonesia.

Keyword: Trace elements, vegetable and sawah soil, environmental evaluation, Sumani watershed.

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Abbreviations:

BCSCST:	Bulk Composition Sediment Column Subdicting at Trenches
BCC	: Bulk Continental Crust
NCJ	: Non Contaminated Soil in Japan
CSJ	: Contaminated Soil in Japan
UCC	: Upper Continental Crust
CF	: Contamination Factor
EF	: Enrichment Factor
Igeo	: Geo Accumulation Index
PLI	: Pollution Load Index
eCEC	: Effective cation exchange capacity
SW	: Sawah
MG	: Mixed garden
Sh	: Shrub
F	: Forest
T	: Tea
St	: Settlement
Vg	: Vegetable
Rs	: River sediment
XRF	: X-ray fluorescence

Soils are complex and heterogeneous mixtures of organic and inorganic matter, as well as the differing components that determine their physical, chemical and biological properties. Soils in agricultural areas (especially gardens) may have a direct influence on public health that is unrelated to production of food, because may come into contact with humans or be transferred to them as suspended dust [1]. Contamination of agricultural soils with heavy metals through repeated use of untreated or poorly treated waste water from industrial establishments, and the application of chemical fertilizers and pesticides is one of the most severe ecological problems in Indonesia [2].

Indonesia is now at the beginning of its third Five-Year Development Plan (TFDP), commonly known as "Repelita III". While the preceding Repelita I and II plans emphasized the development of agriculture, industrialization is now assuming a more important role. Modernization of agriculture and expansion of industry could, unless handled properly, give rise to environmental pollution. Before this occurs, environmental base-line studies should be put in place [3]. Trace elements display a dualistic behavior in their interaction with the Earth's flora and fauna. Many trace elements are essential to the growth and health of both flora (e.g., Cu, Zn, Mn, Fe, Mo and B) and fauna (e.g., Se, Co – [4]), where they act as catalysts in the processes of life. However, at higher concentrations, these essential nutrients can have a

detrimental effect and can become toxic to organisms (including humans), via direct exposure, transportation and bioaccumulation in food chains [5].

Trace metals are among the most persistent environmental pollutants in water bodies and biota, but their occurrence in sediments arises from both natural and anthropogenic sources [6]. Trace metals are ubiquitous in aquatic environments [7]. Some of these metal contaminants are crucial due to their potential toxicity [8] and have potential impact on human health if they reach levels such that they constitute toxic pollutants [9]. In addition, metals including Cu, Fe, Mn, and Ni are essential micronutrients for the life processes of animals and plants [10], but imbalances of these metals may be fatal [11]. Furthermore, [12] observed that elements such as As, Pb, Zn, and Cd exhibit extreme toxicity, even at low levels. Elevated concentrations of trace metals in the environment occur as a result of anthropogenic activities such as mining, milling or smelting of ores, application of agricultural fertilizers, and combustion of fuels [13]. Elements can also be enriched by natural processes, such as redistribution and mobilization of trace elements from host rocks or regoliths by surface and subsurface waters, and by flooding events [14].

The role of trace elements in the soil system is becoming an issue of global concern, at both private and governmental levels, since soil constitutes a crucial component of rural and urban environments and constitutes a very important “ecological crossroad” in the landscape [15]. Vegetable crops cultivated in soils polluted with toxic and heavy metals may take up such metals and accumulate them in their edible and non-edible parts in quantities high enough to cause clinical problems in animal and human consumers [16]. Toxic metals are known to have serious health implications, including carcinogenesis and induced tumor promotion. Growing awareness of the health risks associated with environmental chemicals has also brought a shift in global concern towards prevention of heavy metal accumulation in soils, water and vegetables [17]. Long-term use of industrial or municipal wastewater in irrigation contributions significantly to the content of trace and heavy elements such as Cd, Cu, Zn, Cr, Ni, Pb, and Mn in surface soils [18]. Excessive accumulation of trace elements in agricultural soils through waste water irrigation may thus not only lead to soil contamination, but also affect food quality and safety [19].

The Sumani watershed in West Sumatra, Indonesia, is a prime example of a setting where the use of such water constitutes a potential threat for agricultural ecosystems and human health. Sources of heavy metals in the Sumani watershed include irrigation water, volcanic ash, and residues of fertilizers and pesticides, the use of which has increased since the “green revolution” began in 1972.

Vehicle emissions are also a potential source, as gasoline in Indonesia contains high contents of trace metals, especially Pb. Soil pH, soil physicochemical and geological characteristics could also control availability of heavy metals in agricultural soils in the Sumani watershed. Much of the agricultural area in the Sumani watershed is irrigated with river water that is partly supplied from Lake Dibawah on the west of Mount Talang (2500 m asl), which is an active volcano.

Several environmental studies have been conducted in Indonesia to determine trace metal concentrations, their rates of loading, pollution history, and the extent of contamination from trace metals. These studies have been made in different areas of Sumatra and Java, in both river and coastal sediments [20, 21, 16, 23]. However, trace metals in agricultural watersheds with varied land use have received limited attention. Consequently, our present study examines the status of trace metal pollution in the Sumani watershed, to evaluate the influence of anthropogenic activities and impacts on the agricultural soils within it. This study aimed to determine the concentrations and distribution of trace metals (Pb, Zn, Cu, Ni, Cr, V, Sr, Rb, Ce, Th and Zr) and major oxides (Fe_2O_3 , CaO, MnO, TiO_2 and P_2O_5) in representative soils, to assess the contamination level (if any) in the study area. We employed several procedures including contamination factor (CF), enrichment factor (EF), pollution load index (PLI) and geo-accumulation index (*I_{geo}*) to establish the status of selected trace metals. These data also provide the baseline information needed for developing strategies for future pollution control in an agricultural watershed.

2. Methods

2.1. Study area

The Sumani watershed is located in Solok regency (latitude $0^\circ 36'08''$ to $1^\circ 44'08''$ S, longitude $100^\circ 24'11''$ - $101^\circ 15'438''$ E), approximately 50 km east of Padang City in West Sumatra, Indonesia (Fig. 1). The watershed occupies an area of 58330 ha, and elevation ranges from 300 m to 2500 m above sea level (asl). Lake Singkarak is the outlet of the watershed. Average annual precipitation ranges from 1669 to 3230 mm [24, 25] and climate is classed as humid tropical. The Sumani watershed has a variety of land uses, including primary forest, mixed

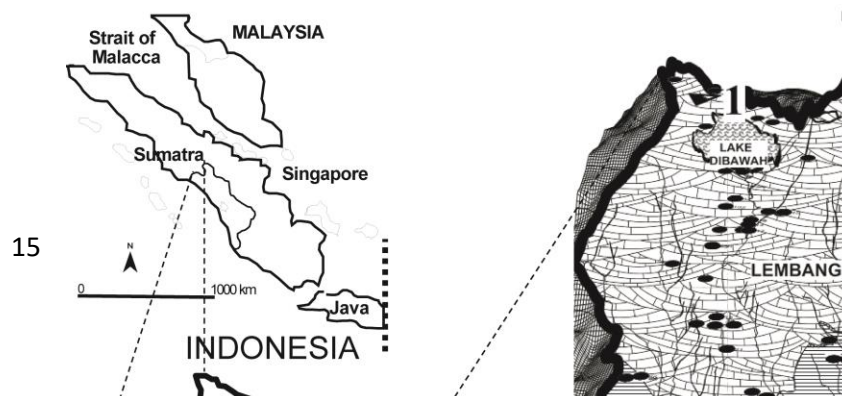


Figure 1. Study site, simplified basement geology and distribution of soil sampling points sites in Sumani watershed, West Sumatra, coordinates bases on UTM coordinate system WGS 84 Zone 47 Southern Hemisphere.

and outlet for irrigation and drainage [25]. Lowland areas of the Sumani watershed are both urban and agricultural. Water from the upland rivers, tributaries, and Lake Dibawah accumulate in Singkarak Lake, and this carry municipal wastes and chemicals from agricultural land. Flatlands in the lowlands are used commonly for sawah, and the uplands for vegetable cultivation, although in some cases sawah and vegetables are rotated to maintain soil fertility and prevent pests. Lowland areas thus receive wastewater from irrigation sourced from Lake. Dibawah in the uplands. The drainages in the lowlands are urbanized, and house small industries such as Tofu and traditional food factories, and service industries such as car and motorcycle repair workshops and gasoline stands. The Sumani watershed contains a range of land uses, including primary forests, tree crop gardens (mixed gardens, coconuts and tea gardens), vegetable gardens, sawah, bush (shrubs, grasses and alang-alang [land dominated by *Imperata cylindrica* (Poaceae)]) and settlements. Mixed garden refers to land where perennial

crops, primarily trees such as coconuts, cloves, coffee, teak, mahogany, sawo (*Achras zapota* L.), avocados, melinjo (*Gnetum gnemon*), rubber, and cinnamon are planted in a combination with annual crops [27]. Chillies (*Capsicum annum* L.), onions (*Allium cepa* L.), soybeans (*Glycine max* L.), corn (*Zea mays* L.) and sweet potatoes (*Ipomea batatas* L.) are the main crops in the vegetable gardens. The Sumani watershed contains five sub-watersheds, including Sumani, Lembang, Gawan, Aripan and Imang. Soil group distribution in the Sumani watershed consists of six groups i.e. Oxic Hapludant, Andic Humitropept, Typic Kandiodult, Aeric Tropaquept, Typic Distropept and Typic Eutropept. The geology of the Sumani watershed area consists of the andesitic volcanic and intrusive rocks that form Mount Talang, undifferentiated volcanic products (basalt to andesite), slates and shales of the Tuhur Formation, limestones (Kuantan Formation) and surficial deposits consisting of river alluvium and alluvial fans [28, 29]. A block diagram showing geology feature and sampling location in watershed was generated by kriging topographic geology map using Surfer from Golden Software [30].

2.2. Fields survey and analytical methods

Soil surveys were conducted at 149 sites occupying a variety of geomorphic positions and land use types (Fig. 1). This paper discusses representative vegetable and sawah soils, and river sediments analyzed using X-ray fluorescence (XRF). Soils were collected at depths of 0-20 cm, air dried, and sieved through a 2 mm mesh for physico-chemical analyses. Soil texture was determined by the pipette method [31]. Soil pH was measured using the glass electrode method with a soil: water ratio of 1:2.5 [32]. Exchangeable acidity was determined by first extracting with 1 mol L⁻¹ KCl and titrating with NaOH [33]. Extractable trace elements- Cd, Pb, Cu and Zn were determined by first extracting with 0.1 mol L⁻¹ HCl. Extractable trace element were determined using Inductively Coupled Plasma-Atomic Emission Spectroscopy (Shimadzu ICPS 2000, Kyoto, Japan) in the Soil and Ecological Laboratory, Shimane University. Exchangeable base cations (Ca, Mg, K and Na) were extracted using 1 mol L⁻¹ neutral ammonium acetate [34]. Exchangeable Ca and Mg were determined using Inductively Coupled Plasma-Atomic Emission Spectroscopy (Shimadzu ICPS 2000, Kyoto, Japan) and exchangeable K and Na were determined using Atomic Absorption Spectrophotometer (Shimadzu AS 680). Effective Cation Exchange Capacity (eCEC) represents the sum of the exchangeable bases and the exchangeable acidity. Four representative soil samples from upland, lowland, middle topography and river sediments were selected for XRF analysis. Dried samples were ground for 20 min in an automatic agate mortar and pestle. Selected major oxide [TiO₂, Fe₂O₃ (total iron expressed as Fe₂O₃), CaO, SiO₂ and P₂O₅] and trace element (Pb, Zn, Cu, Ni, Cr, V, Sr, and Rb) abundances were determined by XRF in the Department of Geoscience, Shimane University, using a RIX-2000 spectrometer (Rigaku Denki Co. Ltd.) equipped with Rh-anode X-ray tube. All samples were made on pressed powder disks, following [35]. Average errors for these elements are less than ±10%. Analytical results for USGS standard SCo-1 (Cody Shale) were acceptable compared with the proposed values of [36]. Contamination Factor (CF), Enrichment Factor (EF), Geo Accumulation Index (I_{geo}) [37, 38] and Pollution Load Index (PLI) [39, 40] were calculated to identify the environmental status of selected trace metals.

3. Result and Discussion

3.1. Soil characteristics

General characteristics and elemental abundances in the Sumani watershed are shown in Table 1. The Sumani watershed surface soils were mainly silty loams (55.8%)

and silty clay loams. Average pH values of the surface soils in the sawah, vegetable plots and river sediments were 4.90-5.85, 4.96 and 5.34, respectively, indicating slightly acidic soil characteristics. The CEC ranges between 9.17 to 15.32 cmol/kg in Sawah; 17.83 cmol/kg in vegetable gardens and 13.99 cmol(+)/kg in river sediment, reflecting low to very high soil fertility conditions in agricultural lands in the Sumani watershed.

The watershed was mainly dominated by Sawah, vegetable gardens, mixed gardens and forest. Drainage patterns are good indicators of land form and bedrock type, and also suggest soil characteristics of the region. The predominant drainage pattern in the watershed is dendritic, indicating the existence of rocks with uniform resistance to erosion. Dendritic pattern is the irregular branching of channels ('tree like') in many directions, and is common in massive hard rock terrains, especially in metamorphic rocks, volcanic rocks, granitoids and in flat-lying areas. This pattern develops where rocks lack marked structural control, and have uniform resistance to erosion. The drainage network, physiography, geology type, of the watershed is illustrated in Fig 1.

3.2. Major and Trace element abundance

Major and trace element analyses of the selected top soils (two sawah soils, one vegetable plot, and one stream sediment) in the Sumani watershed are given in Table 1. Because there is no standard criteria for heavy metal in agricultural soil in Indonesia, we compare the XRF data to modern sediment data from sediment columns subducting at trenches in Sumatra (BCSCST), Java BCSCST, E. Sunda BCSCST, and Japan BCSCST [49], UCC (Bulk continental crust) from [46] and BCC (Bulk continental crust) from [42]. The XRF results showed that the Sumani watershed soils average 30 mg/kg Pb, 55.75 mg/kg Zn and 23 mg/kg Cu. The Pb content of Sumani watershed soils is similar to Sumatra, Java, E. Sunda and Japan BCSCST, but higher than BCC and UCC. However, concentrations of Zn and Cu are lower than Sumatra, Java, E. Sunda and Japan BCSCST (Table 1). Nickel and Cr abundances range from 3 to 15 mg/kg and 8 to 38 mg/kg, while Sr range from 27 to 96 mg/kg, indicating depletion relative to Sumatra BCSCST. Average V and Pb concentrations in the Sumani watershed were 214.5 mg/kg and 37 mg/kg, respectively, while Zr concentration averaged 264.3 mg/kg. The concentration of Rb in the upland vegetable soil, and V

Area (code)	code/ Sample no.	Lithology			Trace element (mg/kg) mean				
		Type	pH	CEC	Pb (lead)	Zn (Zinc)	Cu (Copper)	Ni (Nickel)	Cr (Chromium)
Upland(1)	Vegetable(1)	Si	4.96	17.83	38	88.3	38.7	3	8
Middle(2)	River Sediment(2)	L	5.34	13.99	30	61.6	35.7	9	22
Side (3)	Sawah(3)	C	5.85	9.17	33	43.7	11.2	12	24
Lower(4)	Sawah(4)	L	4.90	15.32	19	34.4	6.4	15	38
Mean			5.26	14.08	30	55.75	23	9.75	23
Sumatra BCSCST					24.5	95.7	39.4	57.5	101.5
Java BCSCST					25.5	99.9	122.1	91.5	70.5
E. Sunda BCSCST					20.6	81.7	163.0	63.5	43.6
Japan BCSCST					23.68	76.8	88.5	66.6	37.3
BCC					12.6	73	24	51	119
UCC					20	71	25	20	35

BCSCST, Bulk composition sediment columns subducting at trenches [50], BCC, Bulk continental crust from [42], na not available, nd not detected, SC silty clay, FMS fine to medium sand; Si, Silty; C, Clay, L, Loam, SL, Silty loam; NCJ, non contaminated soil in Japan from [51], CSJ, contaminated soil in Japan from [51], UCC, upper continental crust measured by XRF from [46], na not available

in sawah soil were roughly double the level in Sumatra BCSCST, suggesting enrichment in the weathered parent rocks of the soils. Average SiO₂, CaO and P₂O₅ abundances in the Sumani watershed soils were lower than Sumatra BCSCST. In contrast, contents of TiO₂ and Fe₂O₃ were almost double that of Sumatra BCSCST, UCC and BCC. Average values of SiO₂, TiO₂, Fe₂O₃, CaO and P₂O₅ were 49.9, 1.0, 8.22, 0.4 and 0.09 wt%, respectively, broadly similar to Sumatra BCSCST, Java BCSCST, E. Sunda BCSCST, Japan BCSCST, UCC and BCC (Table 1).

The average concentration of Pb in all the land use types (Sawah(SW), Mixed garden(MG), Shrub(Sh), Forest(F), Tea(T), Settlement (St), Vegetable(Vg) and River sediment(Rs)) has greater values than NCSJ (Pb 2.7 mg/kg; 45.9% of total sample), but lower than CSJ (Pb 60 g/kg; 6.8% of total sample). The average concentration of Cu in all the land use types (SW, MG, Sh,F, T, St,Vg) has lower values than NCSJ (Zn 11 mg/kg; 90.5% of total sample), but Cu concentration at Rs was mostly similar to NCSJ. The average concentrations of Cu at MG, Sh,F, T, St,Vg were lower than value of Non- Contaminated soil in Japan (NCSJ, Cu 2.9 mg/kg; 67.1% of total sample) but average concentration of Cu at SW, St, Rs lower than CSJ (Cu 6.1 mg/kg).

3.3. Soil and sediment quality guidelines

The pollution status (non-polluted, moderately polluted, or heavily polluted) of the four soil samples taken at sawah and vegetable area and river sediment was evaluated based on sediment quality guidelines compared to background concentrations [41]. The XRF data show significant enrichment in Pb compared to BCC and UCC. Concentrations of Zn and Cu in the vegetable areas are significantly enriched compared to BCC and UCC (Table 1).

3.4. Assessment of metal pollution

The degree of pollution in sediments and soil can be assessed by determining the parameters CF, EF, PLI and Igeo. In the present study, these pollutant indicators were calculated based on average BCC (Bulk continental crust) from [42]. The different methods are discussed below.

3.4.1. Contamination Factor (CF)

The geochemical compositions of soils are a powerful tool for environmental diagnosis, as well as identifying contamination point sources and non-point sources and permitting monitoring. The level of contamination is

Table 2. CF and PLI value at representative land use type in Sumani

Land use/No. sample/XRF analyze	Pb	Zn	Cu	Ni	Cr	CF
Vegetable (1)	3.02	1.21	1.61	0.06	0.07	0.07
River sediment(2)	2.38	0.84	1.49	0.18	0.18	2.38
sawah(3)	2.62	0.60	0.47	0.24	0.20	2.62
Sawah(4)	1.51	0.47	0.27	0.29	0.32	1.51

or ranges, and converts the calculated numerical results into broad descriptive bands of pollution ranging from low to high intensity. The CF suggested by [44] was used to describe the concentration of given toxic substance in a basin:

$$CF = C0_1/Cn$$

where C0–1 is the mean content of the substances and Cn are the background values according to [42]. “The CF may indicate low contamination (CF<1); moderate contamination (1<CF<3); considerable contamination (3<CF<6); or very high contaminations (CF>6)” [44].

The CF values (Table 2) for Pb range from 1.51 in Sawah sample 4 to 3.02 in the vegetable soil, suggesting moderate to considerable contamination at this site. Zinc has an CF of >1 in the vegetable soil, but values of <1 in all others, whereas Cu CF is > 1 in the vegetable soil and the stream sediment, and <1 in the sawah soils. CF values for Ni and Cr are <1 in all samples, indicating nil or low contamination of these elements (Table 3). Vanadium CF is also <1 in the vegetable soil, but ranges from 1.42-2.24 in the other samples. The high CFs for Pb, Zn and Cu in the vegetable soil may be due to the influence of external discrete sources like scavenging and agricultural runoff, erosion, residue of pesticide, fertilizer and manure applied by farmers. [29] report that volcanic ash from Mount Talang contains trace elements that include Pb (16.5-27.50 mg/kg), Zn (64.88-90.60 mg/kg) and Cu (21.2-38.5 mg/kg), and this material may also constitute a source of these elements. With one exception, CF values for the non-metallic elements Sr and Rb are <1, indicating normal levels in the soils.

3.4.2. Pollution Load Index (PLI)

The pollution level of trace metals was calculated based on PLI [39] that reveal the geometric mean CF of each of its constituent samples present in the study area. The PLI is computed by the formula:

$$PLI = n(CF1 \times CF2 \times CF3 \times \dots \times CFn) ^{0.5}$$

where CF is the contamination factor and n is the number of selected metals. According to [39], PLI values of 0, 1, or >1 suggest the absence of baseline pollutants, their presence, and progressive deterioration of sediment quality, respectively. The low PLI values for all four surface samples in both sawah and vegetable plot indicate pollutants are not present (Table 2).

3.4.3. Enrichment Factors(EF)

Heavy metal enrichment factors have been proposed as indicators for diffuse soil contamination. The behavior of a given element in soil (that is, the determination of its accumulation or leaching) may be established by comparing concentrations of a trace element with a reference element [45]. In this study, the EF was also used to assess the level of contamination and possible anthropogenic impacts on the soils in the Sumani watershed. To measure the anomalous metal concentrations, TiO₂ was employed as the normalizer to calculate EF values and Igeo because it has a short residence time and low concentration in natural waters, and thus is transferred quantitatively from source to sediments and soil [46]. It is also unlikely to be contributed by anthropogenic activity. The EF is calculated as:

$$EF = (M/TiO_2 \text{ sample}) / (M/TiO_2 \text{ background});$$

where (M/TiO₂) sample is the ratio of metal and TiO₂ concentrations of the sample and (M/TiO₂) background is the ratio of metal and TiO₂ concentrations of the background. As regional geochemical background values are not available, average concentrations of Pb, Zn, Cu, Ni, Cr, V, BCC [42] were used in this study. Generally, the enrichment factor is used to assess soil contamination (enrichment) based on these criteria: EF < 2—depletion to minimal enrichment; EF 2–5—moderate enrichment; EF 5–20—significant enrichment; EF 20–40—very high enrichment; EF > 40—extremely high enrichment. EF can also be used to evaluate elemental depletions in soils. EF values of less than 1 may indicate that leaching and consumption of elements take precedence over their accumulation in soil [47].

The EF values for Zn, Cu, Ni, Cr, Sr and Rb in the sawah soils indicate depletion to minimal enrichment (EF <2), as shown in Fig. 3. This shows no significant enrichment has occurred, and may indicate that leaching and consumption over accumulation in the soils [47]. Sediment input from the uplands is the main control on the abundances of these element. EF values < 2 strongly suggest that Zn, Cu, Ni, Cr, Sr and Rb in the sawah soils originate from natural processes. However, the EF values

for Pb, V, in the vegetable soil and river sediment show moderate enrichment (EF, 2–5). For Pb this could indicate contribution from both human activities (agrochemicals and leaded petrol from vehicles) and natural sources (volcanic ash) as noted by [15, 29]. The low EF values for Zn, Cu, Ni, Cr, and Sr are classed as depletion to minimal enrichment, suggesting that no environmental problem exists.

3.4.4. Index of geoaccumulation (Igeo)

The Igeo was also calculated to determine the level of pollution, using the following equation [48]:

$$I_{geo} = \log_2 (C_n/1.5B_n),$$

where C_n is the measured concentration of the element n and B_n is the geochemical background value of element n in BCC [49]. The factor 1.5 is introduced to include possible variations in the background values due to lithogenic effects. The following classification is given for Igeo [48]: <0 =practically unpolluted, $0-1$ =unpolluted to moderately polluted, $1-2$ =moderately polluted, $2-3$ =moderately to strongly polluted, $3-4$ =strongly polluted, $4-5$ =strongly to extremely polluted, and >5 =extremely polluted.

The Igeo values of <0 for Zn, Cu, and Ni in the sawah soils and the river sediment (Figure 2) confirm that the areas are unpolluted by these metals, as shown by the indicators discussed above. Furthermore, the Igeo values for Pb all lie in the range from 0 to 1, suggesting that the soils are unpolluted to at worst only moderately polluted by this metal. The highest Igeo values for Pb, Cu, and Zn occur in the vegetable plot upland soil, suggesting that agricultural practices may be beginning to influence the soils. However, a much larger soil database and well-defined background values for the basement rocks are needed before this can be confirmed. The HFSE (high field strength elements) also show slightly elevated Igeo values. Similar results for CF and EF were obtained, but the immobile nature of these elements preclude an anthropogenic source

4. Conclusion

Soils of Sumani watershed have slight acid to acid pH, and based on their CEC, samples ranged from low to very high soil fertility conditions in agriculture land. In Sumani watershed, representative soil sample at River sediment, sawah and vegetable area analyzed by XRF showed that concentration of Pb are greater than those of BCC and UCC and concentration of Pb and Zn at Vegetable and River sediment greater than Sumatra BCSCST. The

elevated CF values are probably the result of anthropogenic activity and provide a useful means of

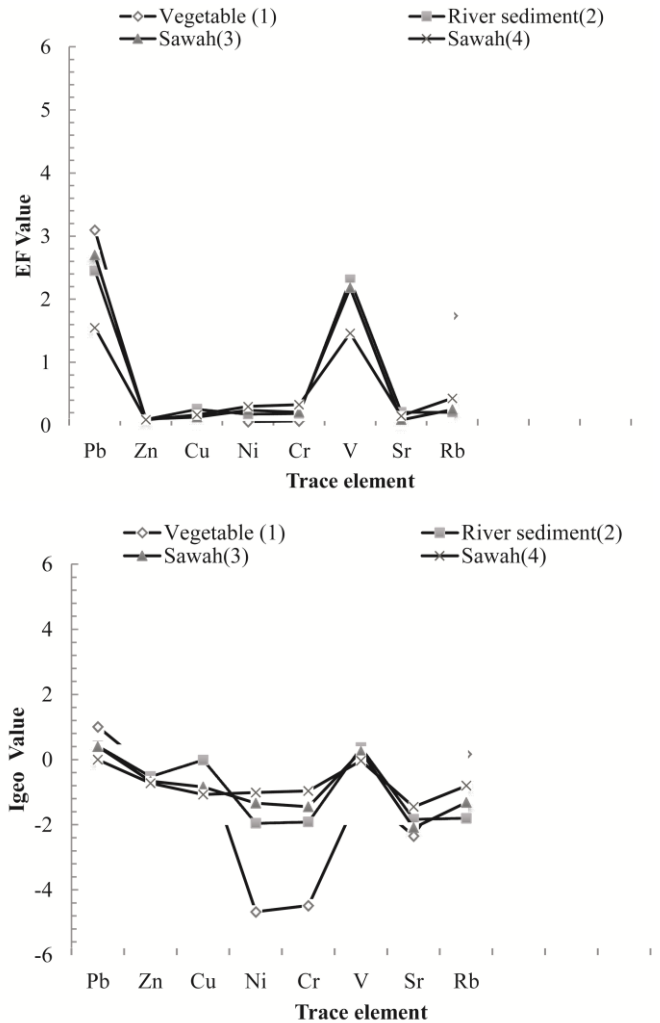


Figure 2. EF values and Igeo values in Vegetable, River sediment and sawah in Sumani watershed.

distinguishing between natural and anthropogenic metals entering the surface soil in Sumani watershed. The PLI values confirm that the quality of the soil at sawah, vegetable and river sediment area in Sumani watershed has no pollutant, and this may not impact on crop production, human life and other organism. According to the EF and the Igeo Index, the area can be considered as having considerable contaminant by Pb and moderate contaminant by Zn, Cu, Rb, at vegetables area. Sawah area can be considered as having moderate contamination by Pb, Cu and V and low contaminated or uncontaminated with respect Zn, Cu, Ni, Cr, Sr and Rb. Concentration Pb and Cu, in surface soil at vegetables and sawah area in Sumani watershed reflected the tendency of metal

contamination that seem to be related to nature additional of volcanic ash from mount Talang, organic matter-rich through agricultural practices and anthropogenic point sources such as fertilizer and pesticide residue and gas from vehicle.

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Figure 1

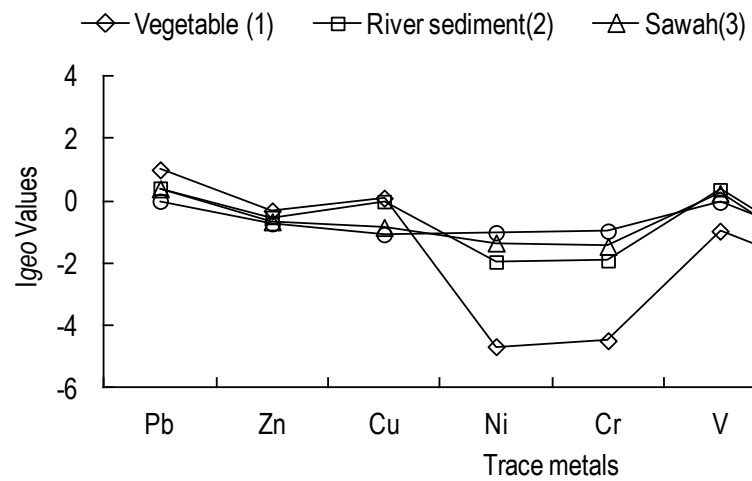
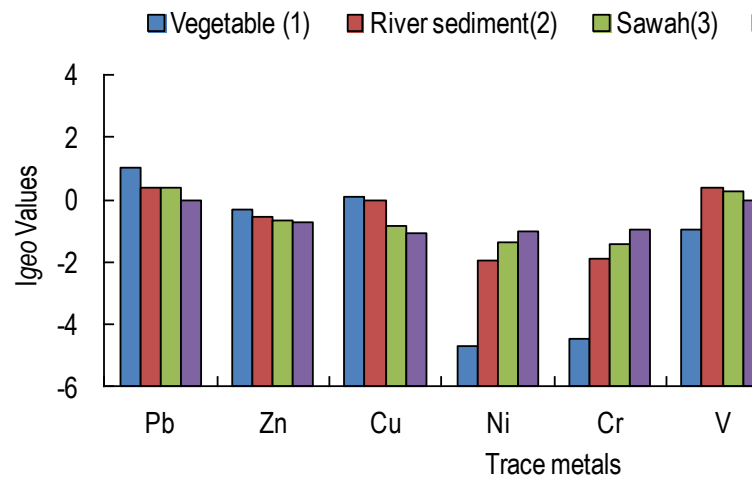
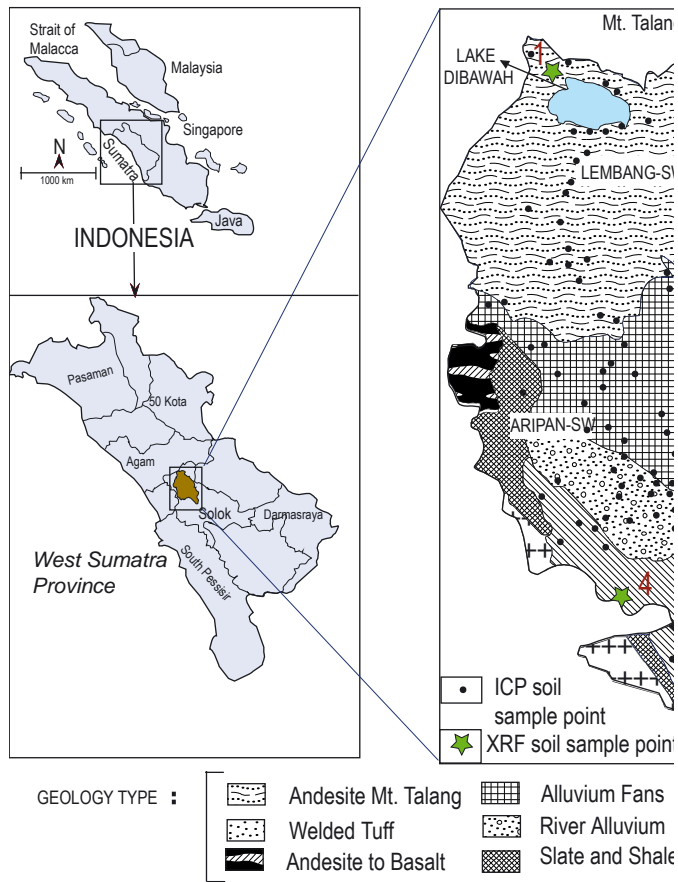
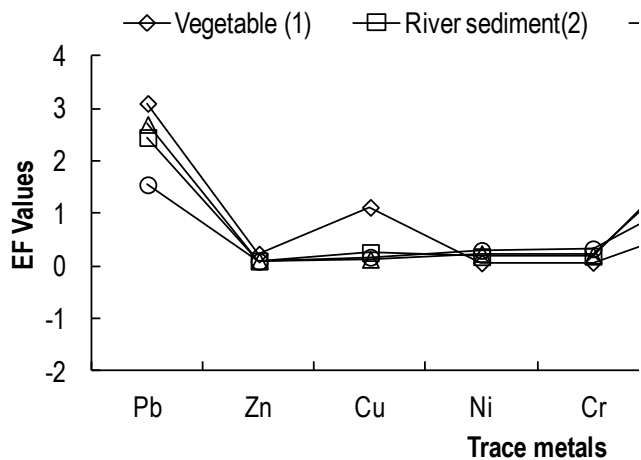
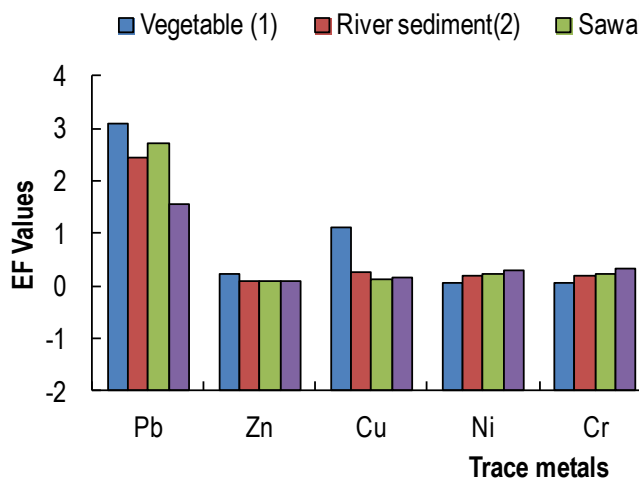


Figure 2

I have kept here both scatter and bar types graph for EF and Igeo values so that you can select the better one. However, reviewers recommended to put bar graph so I think it will be better to put bar graph instead of scatter one.

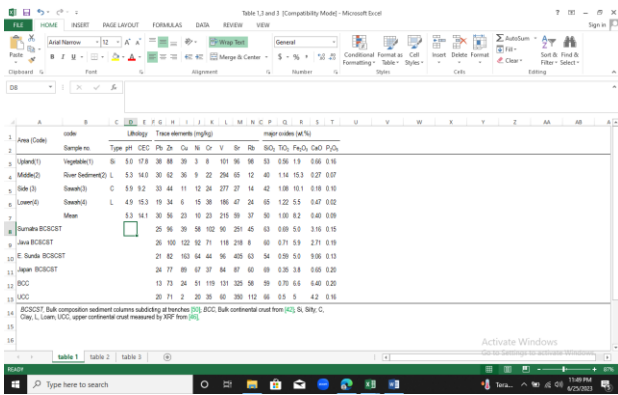
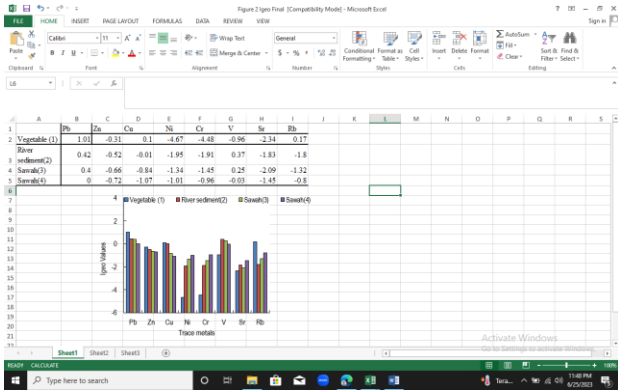


Area (Code)	code/ Sample no.	Lithology			Trace elements (mg/kg)						
		Type	pH	CEC	Pb	Zn	Cu	Ni	Cr	V	S
Upland(1)	Vegetable(1)	Si	5.0	17.8	38	88	39	3	8	101	9
Middle(2)	River Sediment(2)	L	5.3	14.0	30	62	36	9	22	294	6
Side (3)	Sawah(3)	C	5.9	9.2	33	44	11	12	24	277	2
Lower(4)	Sawah(4)	L	4.9	15.3	19	34	6	15	38	186	4
	Mean		5.3	14.1	30	56	23	10	23	215	5
Sumatra BCSCST					25	96	39	58	102	90	2
Java BCSCST					26	100	122	92	71	118	2
E. Sunda BCSCST					21	82	163	64	44	96	4
Japan BCSCST					24	77	89	67	37	84	8
BCC					13	73	24	51	119	131	3
UCC					20	71	2	20	35	60	3

BCSCST, Bulk composition sediment columns subsiding at trenches [50]; BCC, Bulk continental crust; UCC, upper continental crust measured by XRF from [46].

Table 1

ICP, CSJ, NCJ values has deleted from the table for making the table and interpretation much clear.



Thank you again for your valuable time and kind consideration.

We will look forward your amiable reply

Sincerely yours

Aflizar Ph.D of Agriculture

Staf Pengajar

Politeknik Pertanian Negeri Payakumbuh

Jl. Raya Tanjung Pati KM 7, Kec. Harau

Sumatera Barat-Indonesia PO.BOX 107

Before

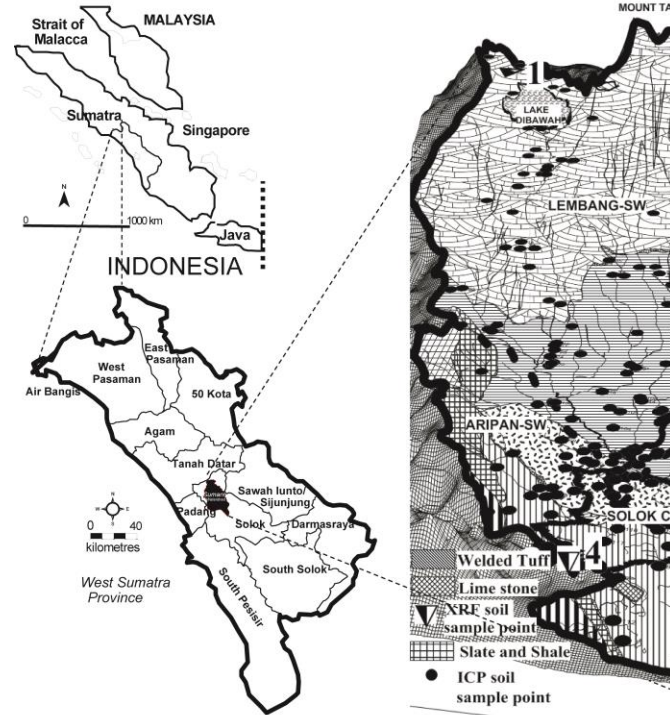


Figure 1. Study site, simplified basement geology and distribution of soil Sumatra, coordinates bases on UTM coordinate system WGS 84 Zone 47 S

Dear Editor J-sustain,

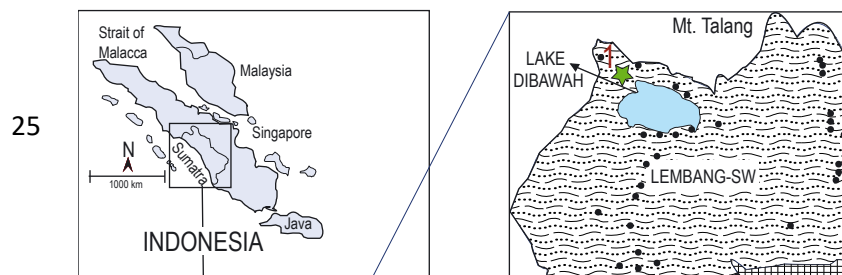
We cordially apologize for your any inconvenience.

Sir/Madam, we are very pleased to be able to communicate you.

We don't know whether our manuscript will be accepted by your outstanding journal or not, We have revised our manuscript according your suggestion.

We would be happy to provide any additional information you may need regarding our manuscript.

After



BCSCST, and Japan BCSCST [49], UCC (Bulk continental crust) from [46] and BCC (Bulk continental crust) from [42]

change to

sediment data from sediment columns subducting at trenches in Sumatra (Sumatra BCSCST), sediment columns subducting at trenches in Java (Java BCSCST), sediment columns subducting at trenches in E. Sunda (E.Sunda BCSCST), and sediment columns subducting at trenches in Japan (Japan BCSCST) [49],

The XRF results showed that the Sumani watershed soils average 30 mg/kg Pb, 55.75 mg/kg Zn and 23 mg/kg Cu.

Change to

The XRF results showed that the Sumani watershed soils average 30 mg/kg Pb (Timbal), 55.75 mg/kg Zn(Zinc) and 23 mg/kg Cu (Copper).

1 mol L⁻¹ KCl cahange to 1 mol L⁻¹ KCl

mol L⁻¹ HCl cahnge to mol L⁻¹ HCl

[TiO₂, Fe₂O₃ (total iron expressed as Fe₂O₃), CaO, SiO₂ and P₂O₅]

change to Selected major oxide [TiO₂, Fe₂O₃ (total iron expressed as Fe₂O₃), CaO, SiO₂ and P₂O₅]

sediment columns subducting at trenches in Sumatra (BCSCST), Java BCSCST, E. Sunda

Ni and Cr abundances range from 3 to 15 mg/kg and 8 to 38 mg/kg, while Sr range from 27 to 96 mg/kg, indicating depletion relative to Sumatra BCSCST.

Change to

Ni (Nickel) and Cr (Chromium) abundances range from 3 to 15 mg/kg and 8 to 38 mg/kg while Sr (Strontium) range from 27 to 96 mg/kg indicating depletion relative to Sumatra BCSCST

Before

Table 1 Lithology and elemental concentration in the surface soil sample (0-

Area (code)	code/ Sample no.	Lithology			Trace element (mg/kg) measured by XRF					
		Type	pH	CEC	Pb (lead)	Zn (Zinc)	Cu (Copper)	Ni (Nickel)	Cr (Chromium)	V (Vanadium)
Upland(1)	Vegetable(1)	Si	4.96	17.83	38	88.3	38.7	3	8	101
Middle(2)	River Sediment(2)	L	5.34	13.99	30	61.6	35.7	9	22	294
Side (3)	Sawah(3)	C	5.85	9.17	33	43.7	11.2	12	24	277
Lower(4)	Sawah(4)	L	4.90	15.32	19	34.4	6.4	15	38	186
Mean			5.26	14.08	30	55.75	23	9.75	23	214.5
Sumatra BCSCST					24.5	95.7	39.4	57.5	101.5	90
Java BCSCST					25.5	99.9	122.1	91.5	70.5	118
E. Sunda BCSCST					20.6	81.7	163.0	63.5	43.6	96
Japan BCSCST					23.68	76.8	88.5	66.6	37.3	84
BCC					12.6	73	24	51	119	131
UCC					20	71	25	20	35	60

Code/number of sample	Lithology			Trace element (mg/kg) measured by XRF					
	Type	pH	CEC	Pb (lead)	Zn (Zinc)	Cu (Copper)	Ni (Nickel)	Cr (Chromium)	V (Vanadium)
Vegetable(n=4)		4.67	17.67	13.02	7.82	1.75			
River Sediment(n=6)		5.81	29.04	11.27	11.53	3.10			
Sawah(n=79)		5.53	21.71	5.11	3.36	25.86			
Shrub(n=7)		5.56	19.32	4.83	1.74	1.07			
Forest(n=8)		5.63	20.10	5.17	3.08	1.09			
Settlement(n=2)		5.81	27.75	2.50	6.57	3.24			
Tea(n=2)		5.25	13.5	8.53	2.79	2.17			
Mixed garden(n=41)		5.54	21.10	3.44	7.48	1.85			
NCSJ	Mean	SL-L	5.6		2.7	11	2.9		
CSJ	Mean	SL-L	5.03		60	151	6.1		

Average V and Pb concentrations in the Sumani watershed were 214.5 mg/kg and 37 mg/kg, respectively, while Zr concentration averaged 264.3 mg/kg . The concentration of Rb in the upland vegetable soil, and V

Change to

Average V (Vanadium) and Pb concentrations in the Sumani watershed were 214.5 mg/kg and 37 mg/kg, respectively. The concentration of Rb (Rubidium) in the upland vegetable soil, and V

After

Table 1 Lithology and elemental concentration in the surface soil sample (0-

Area (Code)	code/ Sample no.	Lithology			Trace elements (mg/kg)					
		Type	pH	CEC	Pb	Zn	Cu	Ni	Cr	
Upland(1)	Vegetable(1)	Si	5.0	17.8	38	88	39	3	8	
Middle(2)	River Sediment(2)	L	5.3	14.0	30	62	36	9	22	
Side (3)	Sawah(3)	C	5.9	9.2	33	44	11	12	24	
Lower(4)	Sawah(4)	L	4.9	15.3	19	34	6	15	38	
Mean			5.3	14.1	30	56	23	10	23	
Sumatra BCSCST					25	96	39	58	102	
Java BCSCST					26	100	122	92	71	
E. Sunda BCSCST					21	82	163	64	44	
Japan BCSCST					24	77	89	67	37	
BCC					13	73	24	51	119	
UCC					20	71	2	20	35	

NCSJ (Pb 2.7 mg/kg; 45.9% of total sample), but lower than CSJ (Pb 60 g/kg; 6.8% of total sample).

After

non contaminated soil in Japan (NCJ)(Pb 2.7 mg/kg; 45.9% of total sample), but lower than contaminated soil in Japan (CSJ) (Pb 60 g/kg; 6.8% of total sample).

Table 2 Lithology and elemental concentration in the surface soil sample (0-20 cm) measured by ICP in Sumani watershed West Sumatra Indonesia

Area (Code)	code/			
	Sample no.	n	Type	p
Upland(1)	Vegetable(Vg)	4	Si	4
Middle(2)	River Sediment(Rs)	6	L	5
Side (3)	Sawah(Sw)	79	C	5
Lower(4)	Shrub(Sh)	7	L	5
	Forest(F)	8	SL-L	5
	Settlement(St)	2	SL-L	5
	Tea(T)	2	SL-L	5
	Mixed garden(MG)	41	SL-L	5
Sumani Watershed	Mean			5
NSJ	Mean		SL-L	5
CSJ	Mean		SL-L	5

Si, Silty; C, Clay, L, Loam, SL, Silt Loam; NCJ, non contaminated soil in Japan (CSJ) (Pb 60 g/kg; 6.8% of total sample) extracting with 0.1 mol L⁻¹ HCL and measured by ICP fr Japan from [51]

Before

The average concentration of Cu in all the land use types (SW, MG, Sh,F, T, St,Vg) has lower values than NCSJ (Zn 11 mg/kg; 90.5% of total sample), but Cu concentration at Rs was mostly similar to NCSJ. The average concentrations of Cu at MG, Sh,F, T, St,Vg were lower than value of Non- Contaminated soil in Japan (NCSJ , Cu 2.9 mg/kg; 67.1% of total sample) but average concentration of Cu at SW, St, Rs lower than CSJ (Cu 6.1 mg/kg).

Before

After

The average concentration of Cu in all the land use types (SW, MG, Sh,F, T, St,Vg) has lower values than NCJ (Zn 11 mg/kg; 90.5% of total sample), but Cu concentration at Rs was mostly similar to NCJ. The average concentrations of

Cu at MG, Sh,F, T, St,Vg were lower than value of Non- Contaminated soil in Japan (NCJ , Cu 2.9 mg/kg; 67.1% of total sample) but average concentration of Cu at SW, St, Rs lower than CSJ (Cu 6.1 mg/kg) (Table 2).

Before

Table 3. CF and PLI value at representative land use type in Sumani watershed

Land use/No. sample/XRF analyze	CF											PLI
	Pb	Zn	Cu	Ni	Cr	V	Sr	Rb	Ce	Th	Zr	
Vegetable (1)	3.02	1.21	1.61	0.06	0.07	0.77	0.30	1.69	2.07	5.54	1.77	0.004
River sediment(2)	2.38	0.84	1.49	0.18	0.18	2.24	0.20	0.21	1.86	2.50	1.89	0.000
sawah(3)	2.62	0.60	0.47	0.24	0.20	2.11	0.08	0.24	2.05	3.04	2.08	0.000
Sawah(4)	1.51	0.47	0.27	0.29	0.32	1.42	0.14	0.41	1.67	2.68	2.85	0.000

e samples indicate

Before

After

Table 3. CF and PLI value at representative land use type in Sumani watershed

Area (Code)	code/ Sample no.	Type	Contamination Factor (C)			
			Pb	Zn	Cu	Ni
Upland(1)	Vegetable(1)	Si	3.02	1.21	1.61	0.0
Middle(2)	River Sediment(2)	L	2.4	0.84	1.49	0.1
Side (3)	Sawah(3)	C	2.6	0.60	0.47	0.2
Lower(4)	Sawah(4)	L	1.5	0.47	0.27	0.2
	Mean		2.38	0.78	0.96	0.1

Before

The CF values (Table 2) for Pb range from 1.51

After

The CF values (Table 3) for Pb range from 1.51

Before

. The low PLI values for all four surface samples in both sawah and vegetable plot indicate pollutants are not present (Table 2).

The EF values for Zn, Cu, Ni, Cr, Sr and Rb in the sawah soils indicate depletion to minimal enrichment (EF <2), as shown in Fig. 3.

After

The EF values for Zn, Cu, Ni, Cr, Sr and Rb in the sawah soils indicate depletion to minimal enrichment (EF <2), as shown in Fig. 2

Before

The Igeo values of <0 for Zn, Cu, and Ni in the sawah soils and the river sediment (Figure 2)

After

The Igeo values of <0 for Zn, Cu, and Ni in the sawah soils and the river sediment (Figure 3)

Before

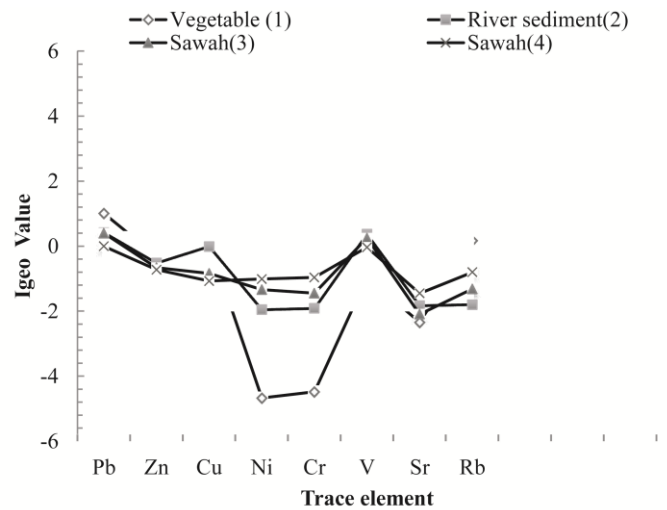
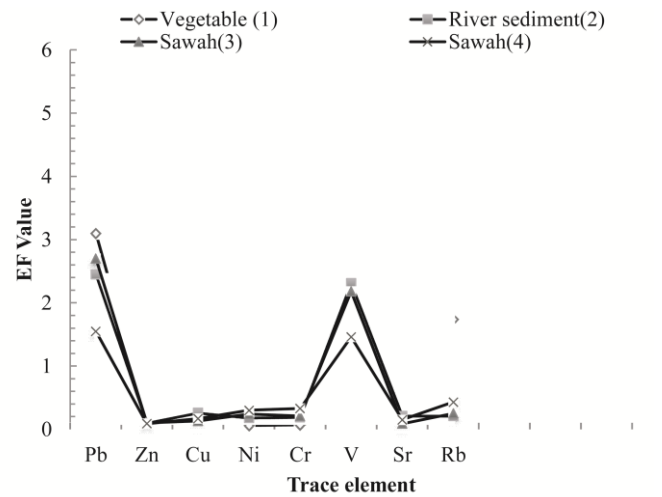


Figure 2. EF values and Igeo values in Vegetable, River sediment and sawah in Sumani watershed.

We thank Professor Tsugiyuki Masunaga of Shimane University, Japan, for their invaluable help during XRF analysis, and many helpful suggestions. We are also grateful to DIKTI-RISTEK Indonesia for their support in this research.

After

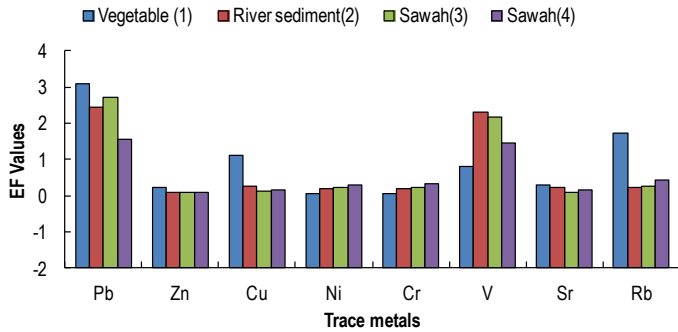


Figure 2. EF values in Vegetable, River sediment and sawah in Sumani watershed.

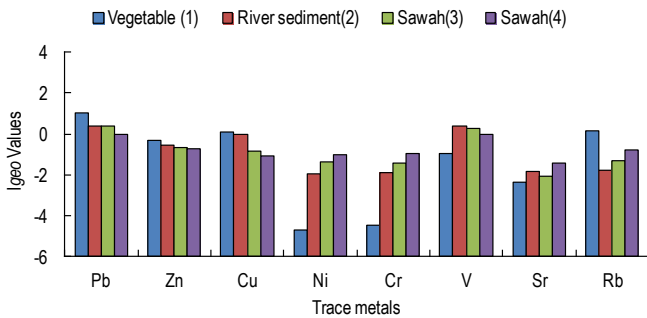


Figure 3. Igeo values in Vegetable, River sediment and sawah in Sumani watershed.

Before

Acknowledgements

We thank Professor Tsugiyuki Masunaga of Shimane University, Japan, for their invaluable help during XRF analysis, and many helpful suggestions. We are also grateful to DIKTI Indonesia for their support in this research.

After

Acknowledgements

Trace metal concentrations in An Agricultural Watershed: Case study in the Sumani Watershed, West Sumatera Indonesia

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Abstract

Selected trace metals investigation of agricultural soil at sawah, vegetable and river sediment in Sumani watershed, West Sumatra, Indonesia were conducted to determine their abundances and to assess contamination levels. pH, CEC and TC values indicates moderate to high soil fertility status. Average concentrations of Pb, Zn, Cu, TiO₂, Fe₂O₃ and P₂O₅ were greater than those of Bulk Composition Crust (BCC) and UCC in most areas. Enrichment factor values at Sawah confirms depletion to minimal enrichment with most metals, while vegetable area indicates moderate enrichment with Pb, Cu and Rb, and depletion to minimal enrichment for Zn, Ni, Cr, V and Sr. Igeo values for Pb, at Sawah were unpolluted to moderately polluted but practically unpolluted for Zn, Cu, Ni, Cr, V, while vegetable area were moderately polluted with Pb, unpolluted to moderately polluted with Cu and Rb and and practically unpolluted for Zn, Ni, Cr, V and Sr. Elevated values of Pb in vegetable area, possibly due to volcanic ash from mount Talang, excess application of fertilizers and pesticides, and high contents of Pb in gasoline, Indonesia.

Keyword: Trace elements, vegetable and sawah soil, environmental evaluation, Sumani watershed.

Abbreviations:

BCSCST:	Bulk Composition Sediment Column Subdicting at Trenches
BCC	: Bulk Continental Crust
NCJ	: Non Contaminated Soil in Japan
CSJ	: Contaminated Soil in Japan
UCC	: Upper Continental Crust
CF	: Contamination Factor
EF	: Enrichment Factor
Igeo	: Geo Accumulation Index
PLI	: Pollution Load Index
eCEC	: Effective cation exchange capacity
SW	: Sawah
MG	: Mixed garden
Sh	: Shrub
F	: Forest
T	: Tea
St	: Settlement
Vg	: Vegetable
Rs	: River sediment
XRF	: X-ray fluorescence

Soils are complex and heterogeneous mixtures of organic and inorganic matter, as well as the differing components that determine their physical, chemical and biological properties. Soils in agricultural areas (especially gardens) may have a direct influence on public health that is unrelated to production of food, because may come into contact with humans or be transferred to them as suspended dust [1]. Contamination of agricultural soils with heavy metals through repeated use of untreated or poorly treated waste water from industrial establishments, and the application of chemical fertilizers and pesticides is one of the most severe ecological problems in Indonesia [2].

Indonesia is now at the beginning of its third Five-Year Development Plan (TFDP), commonly known as "Repelita III". While the preceding Repelita I and II plans emphasized the development of agriculture, industrialization is now assuming a more important role. Modernization of agriculture and expansion of industry could, unless handled properly, give rise to environmental pollution. Before this occurs, environmental base-line studies should be put in place [3]. Trace elements display a dualistic behavior in their interaction with the Earth's flora and fauna. Many trace elements are essential to the growth and health of both flora (e.g., Cu, Zn, Mn, Fe, Mo and B) and fauna (e.g., Se, Co – [4]), where they act as catalysts in the processes of life. However, at higher concentrations, these essential nutrients can have a

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detrimental effect and can become toxic to organisms (including humans), via direct exposure, transportation and bioaccumulation in food chains [5].

Trace metals are among the most persistent environmental pollutants in water bodies and biota, but their occurrence in sediments arises from both natural and anthropogenic sources [6]. Trace metals are ubiquitous in aquatic environments [7]. Some of these metal contaminants are crucial due to their potential toxicity [8] and have potential impact on human health if they reach levels such that they constitute toxic pollutants [9]. In addition, metals including Cu, Fe, Mn, and Ni are essential micronutrients for the life processes of animals and plants [10], but imbalances of these metals may be fatal [11]. Furthermore, [12] observed that elements such as As, Pb, Zn, and Cd exhibit extreme toxicity, even at low levels. Elevated concentrations of trace metals in the environment occur as a result of anthropogenic activities such as mining, milling or smelting of ores, application of agricultural fertilizers, and combustion of fuels [13]. Elements can also be enriched by natural processes, such as redistribution and mobilization of trace elements from host rocks or regoliths by surface and subsurface waters, and by flooding events [14].

The role of trace elements in the soil system is becoming an issue of global concern, at both private and governmental levels, since soil constitutes a crucial component of rural and urban environments and constitutes a very important “ecological crossroad” in the landscape [15]. Vegetable crops cultivated in soils polluted with toxic and heavy metals may take up such metals and accumulate them in their edible and non-edible parts in quantities high enough to cause clinical problems in animal and human consumers [16]. Toxic metals are known to have serious health implications, including carcinogenesis and induced tumor promotion. Growing awareness of the health risks associated with environmental chemicals has also brought a shift in global concern towards prevention of heavy metal accumulation in soils, water and vegetables [17]. Long-term use of industrial or municipal wastewater in irrigation contributions significantly to the content of trace and heavy elements such as Cd, Cu, Zn, Cr, Ni, Pb, and Mn in surface soils [18]. Excessive accumulation of trace elements in agricultural soils through waste water irrigation may thus not only lead to soil contamination, but also affect food quality and safety [19].

The Sumani watershed in West Sumatra, Indonesia, is a prime example of a setting where the use of such water constitutes a potential threat for agricultural ecosystems and human health. Sources of heavy metals in the Sumani watershed include irrigation water, volcanic ash, and residues of fertilizers and pesticides, the use of which has increased since the “green revolution” began in 1972.

Vehicle emissions are also a potential source, as gasoline in Indonesia contains high contents of trace metals, especially Pb. Soil pH, soil physicochemical and geological characteristics could also control availability of heavy metals in agricultural soils in the Sumani watershed. Much of the agricultural area in the Sumani watershed is irrigated with river water that is partly supplied from Lake Dibawah on the west of Mount Talang (2500 m asl), which is an active volcano.

Several environmental studies have been conducted in Indonesia to determine trace metal concentrations, their rates of loading, pollution history, and the extent of contamination from trace metals. These studies have been made in different areas of Sumatra and Java, in both river and coastal sediments [20, 21, 16, 23]. However, trace metals in agricultural watersheds with varied land use have received limited attention. Consequently, our present study examines the status of trace metal pollution in the Sumani watershed, to evaluate the influence of anthropogenic activities and impacts on the agricultural soils within it. This study aimed to determine the concentrations and distribution of trace metals (Pb, Zn, Cu, Ni, Cr, V, Sr, Rb, Ce, Th and Zr) and major oxides (Fe_2O_3 , CaO, MnO, TiO_2 and P_2O_5) in representative soils, to assess the contamination level (if any) in the study area. We employed several procedures including contamination factor (CF), enrichment factor (EF), pollution load index (PLI) and geo-accumulation index (*I_{geo}*) to establish the status of selected trace metals. These data also provide the baseline information needed for developing strategies for future pollution control in an agricultural watershed.

6. Methods

6.1. Study area

The Sumani watershed is located in Solok regency (latitude $0^\circ 36'08''$ to $1^\circ 44'08''$ S, longitude $100^\circ 24'11''$ - $101^\circ 15'438''$ E), approximately 50 km east of Padang City in West Sumatra, Indonesia (Fig. 1). The watershed occupies an area of 58330 ha, and elevation ranges from 300 m to 2500 m above sea level (asl). Lake Singkarak is the outlet of the watershed. Average annual precipitation ranges from 1669 to 3230 mm [24, 25] and climate is classed as humid tropical. The Sumani watershed has a variety of land uses, including primary forest, mixed gardens, vegetable gardens, sawah, abandoned agricultural fields, and settlements. The term sawah refers to a levelled and bounded rice field with an inlet

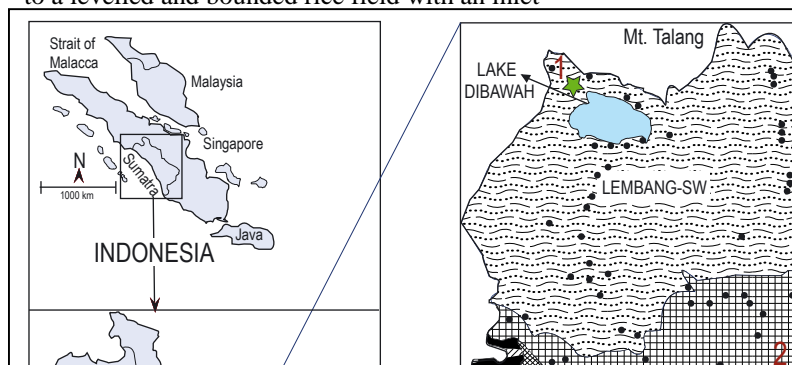


Figure 1. Study site, simplified basement geology and distribution of soil sampling points sites in Sumani watershed, West Sumatra, coordinates bases on UTM coordinate system WGS 84 Zone 47 Southern Hemisphere.

and outlet for irrigation and drainage [25]. Lowland areas of the Sumani watershed are both urban and agricultural. Water from the upland rivers, tributaries, and Lake Dibawah accumulate in Singkarak Lake, and this carry municipal wastes and chemicals from agricultural land. Flatlands in the lowlands are used commonly for sawah, and the uplands for vegetable cultivation, although in some cases sawah and vegetables are rotated to maintain soil fertility and prevent pests. Lowland areas thus receive wastewater from irrigation sourced from Lake. Dibawah in the uplands. The drainages in the lowlands are urbanized, and house small industries such as Tofu and traditional food factories, and service industries such as car and motorcycle repair workshops and gasoline stands. The Sumani watershed contains a range of land uses, including primary forests, tree crop gardens (mixed gardens, coconuts and tea gardens), vegetable gardens, sawah, bush (shrubs, grasses and alang-alang [land dominated by *Imperata cylindrica* (Poaceae)]) and settlements. Mixed garden refers to land where perennial

crops, primarily trees such as coconuts, cloves, coffee, teak, mahogany, sawo (*Achras zapota* L.), avocados, melinjo (*Gnetum gnemon*), rubber, and cinnamon are planted in a combination with annual crops [27]. Chillies (*Capsicum annum* L.), onions (*Allium cepa* L.), soybeans (*Glycine max* L.), corn (*Zea mays* L.) and sweet potatoes (*Ipomea batatas* L.) are the main crops in the vegetable gardens. The Sumani watershed contains five sub-watersheds, including Sumani, Lembang, Gawan, Aripan and Imang. Soil group distribution in the Sumani watershed consists of six groups i.e. Oxic Hapudant, Andic Humitropept, Typic Kandiudult, Aeric Tropaquept, Typic Distropept and Typic Eutropept. The geology of the Sumani watershed area consists of the andesitic volcanic and intrusive rocks that form Mount Talang, undifferentiated volcanic products (basalt to andesite), slates and shales of the Tuhur Formation, limestones (Kuantan Formation) and surficial deposits consisting of river alluvium and alluvial fans [28, 29]. A block diagram showing geology feature and sampling location in watershed was generated by kriging topographic geology map using Surfer from Golden Software [30].

6.2. Fields survey and analytical methods

Soil surveys were conducted at 149 sites occupying a variety of geomorphic positions and land use types (Fig. 1). This paper discusses representative vegetable and sawah soils, and river sediments analyzed using X-ray fluorescence (XRF). Soils were collected at depths of 0-20 cm, air dried, and sieved through a 2 mm mesh for physico-chemical analyses. Soil texture was determined by the pipette method [31]. Soil pH was measured using the glass electrode method with a soil: water ratio of 1:2.5 [32]. Exchangeable acidity was determined by first extracting with 1 mol L⁻¹ KCl and titrating with NaOH [33]. Extractable trace elements- Cd, Pb, Cu and Zn were determined by first extracting with 0.1 mol L⁻¹ HCl. Extractable trace element were determined using Inductively Coupled Plasma-Atomic Emission Spectroscopy (Shimadzu ICPS 2000, Kyoto, Japan) in the Soil and Ecological Laboratory, Shimane University. Exchangeable base cations (Ca, Mg, K and Na) were extracted using 1 mol L⁻¹ neutral ammonium acetate [34]. Exchangeable Ca and Mg were determined using Inductively Coupled Plasma-Atomic Emission Spectroscopy (Shimadzu ICPS 2000, Kyoto, Japan) and exchangeable K and Na were determined using Atomic Absorption Spectrophotometer (Shimadzu AS 680). Effective Cation Exchange Capacity (eCEC) represents the sum of the exchangeable bases and the exchangeable acidity. Four representative soil samples from upland, lowland, middle topography and river sediments were selected for XRF analysis. Dried samples were ground for 20 min in an automatic agate mortar and pestle. Selected major oxide [TiO₂, Fe₂O₃ (total iron expressed as Fe₂O₃), CaO, SiO₂ and P₂O₅] and trace element (Pb, Zn, Cu, Ni, Cr, V, Sr, and Rb) abundances were determined by XRF in the Department of Geoscience, Shimane University, using a RIX-2000 spectrometer (Rigaku Denki Co. Ltd.) equipped with Rh-anode X-ray tube. All samples were made on pressed powder disks, following [35]. Average errors for these elements are less than ±10%. Analytical results for USGS standard SCo-1 (Cody Shale) were acceptable compared with the proposed values of [36]. Contamination Factor (CF), Enrichment Factor (EF), Geo Accumulation Index (I_{geo}) [37, 38] and Pollution Load Index (PLI) [39, 40] were calculated to identify the environmental status of selected trace metals.

7. Result and Discussion

7.1. Soil characteristics

General characteristics and elemental abundances in the Sumani watershed are shown in Table 1. The Sumani watershed surface soils were mainly silty loams (55.8%),

and silty clay loams. Average pH values of the surface soils in the sawah, vegetable plots and river sediments were 4.90-5.85, 4.96 and 5.34, respectively, indicating slightly acidic soil characteristics. The eCEC ranges between 9.17 to 15.32 cmol/kg in Sawah; 17.83 cmol/kg in vegetable gardens and 13.99 cmol(+)/kg in river sediment, reflecting low to very high soil fertility conditions in agricultural lands in the Sumani watershed.

The watershed was mainly dominated by Sawah, vegetable gardens, mixed gardens and forest. Drainage patterns are good indicators of land form and bedrock type, and also suggest soil characteristics of the region. The predominant drainage pattern in the watershed is dendritic, indicating the existence of rocks with uniform resistance to erosion. Dendritic pattern is the irregular branching of channels ('tree like') in many directions, and is common in massive hard rock terrains, especially in metamorphic rocks, volcanic rocks, granitoids and in flat-lying areas. This pattern develops where rocks lack marked structural control, and have uniform resistance to erosion. The drainage network, physiography, geology type, of the watershed is illustrated in Fig 1.

7.2. Major and Trace element abundance

Major and trace element analyses of the selected top soils (two sawah soils, one vegetable plot, and one stream sediment) in the Sumani watershed are given in Table 1. Because there is no standard criteria for heavy metal in agricultural soil in Indonesia, we compare the XRF data to modern sediment data from sediment columns subducting at trenches in Sumatra (Sumatra BCSCST), sediment columns subducting at trenches in Java (Java BCSCST), sediment columns subducting at trenches in E. Sunda (E.Sunda BCSCST), and sediment columns subducting at trenches in Japan (Japan BCSCST) [49], UCC (Bulk continental crust) from [46] and BCC (Bulk continental crust) from [42]. The XRF results showed that the Sumani watershed soils average 30 mg/kg Pb (Timbal), 55.75 mg/kg Zn (Zinc) and 23 mg/kg Cu (Copper). The Pb content of Sumani watershed soils is similar to Sumatra, Java, E. Sunda and Japan BCSCST, but higher than BCC and UCC. However, concentrations of Zn and Cu are lower than Sumatra, Java, E. Sunda and Japan BCSCST (Table 1). Ni (Nickel) and Cr (Chromium) abundances range from 3 to 15 mg/kg and 8 to 38 mg/kg, while Sr (Strontium) range from 27 to 96 mg/kg, indicating depletion relative to Sumatra BCSCST. Average V (Vanadium) and Pb concentrations in the Sumani watershed were 214.5 mg/kg and 37 mg/kg, respectively. The concentration of Rb (Rubidium) in the upland vegetable soil, and V

Table 1 Lithology and elemental concentration in the surface soil sample (0-20 cm) measured by XRF in Sumani watershed West Sumatra Indonesia

Area (Code)	code/ Sample no.	Lithology			Trace elements (mg/kg)								major oxides (wt.%)				
		Type	pH	CEC	Pb	Zn	Cu	Ni	Cr	V	Sr	Rb	SiO ₂	TiO ₂	Fe ₂ O ₃	CaO	P ₂ O ₅
Upland(1)	Vegetable(1)	Si	5.0	17.8	38	88	39	3	8	101	96	98	53	0.56	1.9	0.66	0.16
Middle(2)	River Sediment(2)	L	5.3	14.0	30	62	36	9	22	294	65	12	40	1.14	15.3	0.27	0.07
Side (3)	Sawah(3)	C	5.9	9.2	33	44	11	12	24	277	27	14	42	1.08	10.1	0.18	0.10
Lower(4)	Sawah(4)	L	4.9	15.3	19	34	6	15	38	186	47	24	65	1.22	5.5	0.47	0.02
	Mean		5.3	14.1	30	56	23	10	23	215	59	37	50	1.00	8.2	0.40	0.09
Sumatra BCSCST					25	96	39	58	102	90	251	45	63	0.69	5.0	3.16	0.15
Java BCSCST					26	100	122	92	71	118	218	8	60	0.71	5.9	2.71	0.19
E. Sunda BCSCST					21	82	163	64	44	96	405	63	54	0.59	5.0	9.06	0.13
Japan BCSCST					24	77	89	67	37	84	87	60	69	0.35	3.8	0.65	0.20
BCC					13	73	24	51	119	131	325	58	59	0.70	6.6	6.40	0.20
UCC					20	71	2	20	35	60	350	112	66	0.5	5	4.2	0.16

BCSCST, Bulk composition sediment columns subsiding at trenches [50]; BCC, Bulk continental crust from [42]; Si, Silty; C, Clay, L, Loam; UCC, upper continental crust measured by XRF from [46].

Table 2 Lithology and elemental concentration in the surface soil sample (0-20 cm) measured by ICP in Sumani watershed West Sumatra Indonesia

Area (Code)	code/ Sample no.	n	Lithology			Trace elements (mg/kg) measured by ICP		
			Type	pH	CEC	Pb	Zn	Cu
Upland(1)	Vegetable(Vg)	4	Si	4.67	17.67	13.02	7.82	1.75
Middle(2)	River Sediment(Rs)	6	L	5.81	29.04	11.27	11.53	3.10
Side (3)	Sawah(Sw)	79	C	5.53	21.71	5.11	3.36	25.86
Lower(4)	Shrub(Sh)	7	L	5.56	19.32	4.83	1.74	1.07
	Forest(F)	8	SL-L	5.63	20.10	5.17	3.08	1.09
	Settlement(St)	2	SL-L	5.81	27.75	2.50	6.57	3.24
	Tea(T)	2	SL-L	5.25	13.50	8.53	2.79	2.17
	Mixed garden(MG)	41	SL-L	5.54	21.10	3.44	7.48	1.85
Sumani Watershed	Mean			5.48	21.27	6.73	5.55	5.02
NSJ	Mean		SL-L	5.60		2.70	11.00	2.90
CSJ	Mean		SL-L	5.03		60.00	151.00	6.10

Si, Silty; C, Clay, L, Loam, SL, Silt Loam; NCJ, non contaminated soil in Japan measured extracting with 0.1 mol L⁻¹ HCL and measured by ICP from [51]; CSJ, contaminated soil in Japan from [51]

abundances in the Sumani watershed soils were lower than Sumatra BCSCST.

and Fe₂O₃ were almost, UCC and BCC. Average and P₂O₅ were 49.9, 1.0, 8.22, 0.4 and 0.09 wt%, respectively, broadly similar to Sumatra BCSCST, Java BCSCST, E. Sunda BCSCST, Japan BCSCST, UCC and BCC (Table 1).

The average concentration of Pb in all the land use types (Sawah(SW), Mixed garden(MG), Shrub(Sh), Forest(F), Tea(T), Settlement (St), Vegetable(Vg) and River sediment(Rs)) has greater values than , non contaminated soil in Japan (NCJ)(Pb 2.7 mg/kg; 45.9% of total sample), but lower than contaminated soil in Japan (CSJ) (Pb 60 g/kg; 6.8% of total sample). The average concentration of Cu in all the land use types (SW, MG, Sh,F, T, St,Vg) has lower values than NCJ (Zn 11 mg/kg; 90.5% of total sample), but Cu concentration at Rs was mostly similar to NCJ. The average concentrations of Cu at MG, Sh,F, T, St,Vg were lower than value of Non-Contaminated soil in Japan (NCJ , Cu 2.9 mg/kg; 67.1% of total sample) but average concentration of Cu at SW, St, Rs lower than CSJ (Cu 6.1 mg/kg) (Table 2).

7.4.1. Contamination Factor (CF)

The geochemical compositions of soils are a powerful tool for environmental diagnosis, as well as identifying contamination point sources and non-point sources and permitting monitoring. The level of contamination is expressed by a CF [43] that estimates the impact scales or ranges, and converts the calculated numerical results into broad descriptive bands of pollution ranging from low to high intensity. The CF suggested by [44] was used to describe the concentration of given toxic substance in a basin:

Table 3. CF and PLI value at representative land use type in Sumani watershed

Area (Code)	code/ Sample no.	Type	Contamination Factor (CF)									PLI
			Pb	Zn	Cu	Ni	Cr	V	Sr	Rb		
Upland(1)	Vegetable(1)	Si	3.02	1.21	1.61	0.06	0.07	0.77	0.30	1.69	0.004	
Middle(2)	River Sediment(2)	L	2.4	0.84	1.49	0.18	0.18	2.24	0.20	0.21	0.000	
Side (3)	Sawah(3)	C	2.6	0.60	0.47	0.24	0.20	2.11	0.08	0.24	0.000	
Lower(4)	Sawah(4)	L	1.5	0.47	0.27	0.29	0.32	1.42	0.14	0.41	0.000	
	Mean		2.38	0.78	0.96	0.19	0.19	1.64	0.18	0.64	0.001	

7.3. Soil and sediment quality guidelines

The pollution status (non-polluted, moderately polluted, or heavily polluted) of the four soil samples taken at sawah and vegetable area and river sediment was evaluated based on sediment quality guidelines compared to background concentrations [41]. The XRF data show significant enrichment in Pb compared to BCC and UCC. Concentrations of Zn and Cu in the vegetable areas are significantly enriched compared to BCC and UCC (Table 1).

7.4. Assessment of metal pollution

The degree of pollution in sediments and soil can be assessed by determining the parameters CF, EF, PLI and Igeo. In the present study, these pollutant indicators were calculated based on average BCC (Bulk continental crust) from [42]. The different methods are discussed below.

$$CF = C0_1/Cn$$

where C0–1 is the mean content of the substances and Cn are the background values according to [42]. “The CF may indicate low contamination (CF<1); moderate contamination (1<CF<3); considerable contamination (3<CF<6); or very high contaminations (CF>6)” [44].

The CF values (Table 3) for Pb range from 1.51 in Sawah sample 4 to 3.02 in the vegetable soil, suggesting moderate to considerable contamination at this site. Zinc has an CF of >1 in the vegetable soil, but values of <1 in all others, whereas Cu CF is > 1 in the vegetable soil and the stream sediment, and <1 in the sawah soils. CF values for Ni and Cr are <1 in all samples, indicating nil or low contamination of these elements (Table 3). Vanadium CF is also <1 in the vegetable soil, but ranges from 1.42-2.24 in the other samples. The high CFs for Pb, Zn and Cu in the vegetable soil may be due to the influence of external discrete sources like scavenging and agricultural runoff, erosion, residue of pesticide, fertilizer and manure applied by farmers. [29] report that volcanic ash from Mount

Talang contains trace elements that include Pb (16.5-27.50 mg/kg), Zn (64.88-90.60 mg/kg) and Cu (21.2-38.5 mg/kg), and this material may also constitute a source of these elements. With one exception, CF values for the non-metallic elements Sr and Rb are <1, indicating normal levels in the soils.

7.4.2. Pollution Load Index (PLI)

The pollution level of trace metals was calculated based on PLI [39] that reveal the geometric mean CF of each of its constituent samples present in the study area. The PLI is computed by the formula:

$$PLI = n(CF1 \times CF2 \times CF3 \times \dots \times CFn)^{0.5}$$

where CF is the contamination factor and n is the number of selected metals. According to [39], PLI values of 0, 1, or >1 suggest the absence of baseline pollutants, their presence, and progressive deterioration of sediment quality, respectively. The low PLI values for all four surface samples in both sawah and vegetable plot indicate pollutants are not present (Table 3).

7.4.3. Enrichment Factors(EF)

Heavy metal enrichment factors have been proposed as indicators for diffuse soil contamination. The behavior of a given element in soil (that is, the determination of its accumulation or leaching) may be established by comparing concentrations of a trace element with a reference element [45]. In this study, the EF was also used to assess the level of contamination and possible anthropogenic impacts on the soils in the Sumani watershed. To measure the anomalous metal concentrations, TiO₂ was employed as the normalizer to calculate EF values and Igeo because it has a short residence time and low concentration in natural waters, and thus is transferred quantitatively from source to sediments and soil [46]. It is also unlikely to be contributed by anthropogenic activity. The EF is calculated as:

$$EF = (M/TiO_2 \text{ sample}) / (M/TiO_2 \text{ background});$$

where (M/TiO₂) sample is the ratio of metal and TiO₂ concentrations of the sample and (M/TiO₂) background is the ratio of metal and TiO₂ concentrations of the background. As regional geochemical background values are not available, average concentrations of Pb, Zn, Cu, Ni, Cr, V, BCC [42] were used in this study. Generally, the enrichment factor is used to assess soil contamination (enrichment) based on these criteria: EF < 2—depletion to minimal enrichment; EF 2–5—moderate enrichment; EF

5–20—significant enrichment; EF 20–40—very high enrichment; EF > 40—extremely high enrichment. EF can also be used to evaluate elemental depletions in soils. EF values of less than 1 may indicate that leaching and consumption of elements take precedence over their accumulation in soil [47].

The EF values for Zn, Cu, Ni, Cr, Sr and Rb in the sawah soils indicate depletion to minimal enrichment (EF <2), as shown in Fig. 2. This shows no significant enrichment has occurred, and may indicate that leaching and consumption over accumulation in the soils [47]. Sediment input from the uplands is the main control on the abundances of these element. EF values < 2 strongly suggest that Zn, Cu, Ni, Cr, Sr and Rb in the sawah soils originate from natural processes. However, the EF values for Pb, V, in the vegetable soil and river sediment show moderate enrichment (EF, 2–5). For Pb this could indicate contribution from both human activities (agrochemicals and leaded petrol from vehicles) and natural sources (volcanic ash) as noted by [15, 29]. The low EF values for Zn, Cu, Ni, Cr, and Sr are classed as depletion to minimal enrichment, suggesting that no environmental problem exists.

7.4.4. Index of geoaccumulation (Igeo)

The Igeo was also calculated to determine the level of pollution, using the following equation [48]:

$$Igeo = \log_2 (Cn/1.5Bn),$$

where Cn is the measured concentration of the element n and Bn is the geochemical background value of element n in BCC [49]. The factor 1.5 is introduced to include possible variations in the background values due to lithogenic effects. The following classification is given for Igeo [48]: <0=practically unpolluted, 0–1=unpolluted to moderately polluted, 1–2=moderately polluted, 2–3=moderately to strongly polluted, 3–4= strongly polluted, 4–5=strongly to extremely polluted, and >5=extremely polluted.

The Igeo values of <0 for Zn, Cu, and Ni in the sawah soils and the river sediment (Figure 3) confirm that the areas are unpolluted by these metals, as shown by the indicators discussed above. Furthermore, the Igeo values for Pb all lie in the range from 0 to 1, suggesting that the soils are unpolluted to at worst only moderately polluted by this metal. The highest Igeo values for Pb, Cu, and Zn occur in the vegetable plot upland soil, suggesting that agricultural practices may be beginning to influence the soils. However, a much larger soil database and well-defined background values for the basement rocks are needed before this can be confirmed. The HFSE(high field strength elements) also show slightly elevated Igeo values.

Similar results for CF and EF were obtained, but the immobile nature of these elements preclude an anthropogenic source

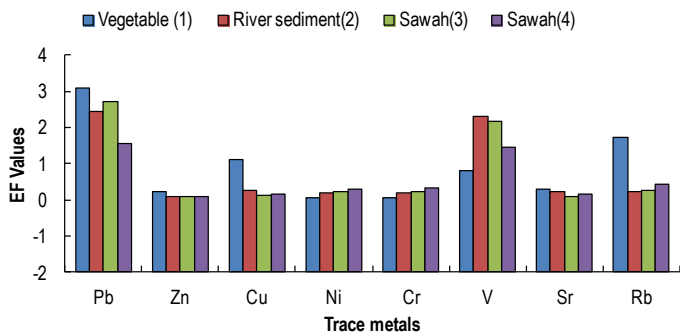


Figure 2. EF values in Vegetable, River sediment and sawah in Sumani watershed.

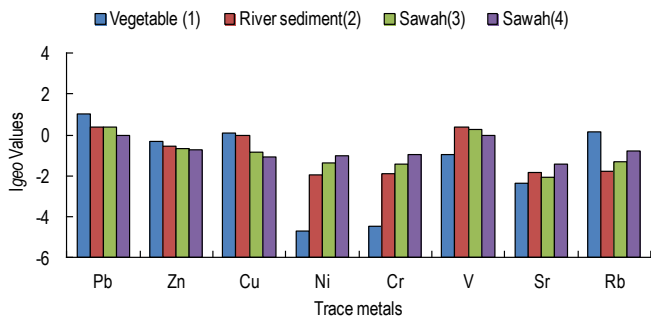


Figure 3. Igeo values in Vegetable, River sediment and sawah in Sumani watershed.

8. Conclusion

Soils of Sumani watershed have slight acid to acid pH, and based on their CEC, samples ranged from low to very high soil fertility conditions in agriculture land. In Sumani watershed, representative soil sample at River sediment, sawah and vegetable area analyzed by XRF showed that concentration of Pb are greater than those of BCC and UCC and concentration of Pb and Zn at Vegetable and River sediment greater than Sumatra BCSCST. The elevated CF values are probably the result of anthropogenic activity and provide a useful means of distinguishing between natural and anthropogenic metals entering the surface soil in Sumani watershed. The PLI values confirm that the quality of the soil at sawah, vegetable and river sediment area in Sumani watershed has no pollutant, and this may not impact on crop production, human life and other organism. According to

the EF and the Igeo Index, the area can be considered as having considerable contaminant by Pb and moderate contaminant by Zn, Cu, Rb, at vegetables area. Sawah area can be considered as having moderate contamination by Pb, Cu and V and low contaminated or uncontaminated with respect Zn, Cu, Ni, Cr, Sr and Rb. Concentration Pb and Cu, in surface soil at vegetables and sawah area in Sumani watershed reflected the tendency of metal contamination that seem to be related to nature additional of volcanic ash from mount Talang, organic matter-rich through agricultural practices and anthropogenic point sources such as fertilizer and pesticide residue and gas from vehicle.

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