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Potential of Organic Amendments for Heavy Metal Contamination in Soil–Coriander System: Environmental Fate and Associated Ecological Risk

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Abstract: Pollution by organic wastes and manures is an important problem in tropical and subtropical countries and novel solutions for their proper management and valorization are needed. Waste-derived organic manures may increase metal load in the soil–plant ecosystem and food chain, with potential risks to public health. The aim of this work was to evaluate the impact of three manures (poultry waste (PW), press mud (PM), and farmyard manure (FYM)) on heavy metals (HMs) (Cd, Co, Cr, Cu, Pb, Zn, Fe, Mn) toxicity in a soil and coriander (*Coriandrum sativum* L.) system and their environmental impact (bioaccumulation, pollution load) and the consequent risk to human health via consumption. Results demonstrated that HMs in coriander fluctuated from 0.40 to 0.43 for Cd, 1.84 to 3.52 for Co, 0.15 to 0.16 for Cr, 1.32 to 1.40 for Cu, 0.05 to 0.09 for Pb, 1.32 to 2.51 for Fe, 0.10 to 0.32 for Mn, and 2.01 to 8.70 mg/kg for Zn, respectively. Highest pollution load index value was 2.89 for Cd and Mn showed the lowest (0.005). Daily intake of metal was noticed to be higher for Zn (0.049 mg/kg/day) for PW and lower for Mn (0.0005) at FYM treatment. The health risk index value was <1 and in the range of 2.30–2.50 for Cd showing potential carcinogenicity. It was concluded that as the organic amendments have the widest application in vegetables, it should be prudent to avoid their contamination and mobilization in plant–soil ecosystems to protect public health perspectives.

Keywords: bioaccumulation; *C. sativum*; vegetable; organic manures; risk assessment; public health

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1. Introduction

Growing economic and demographic pressure in coming years needs to enhance crop yield. However, the predominant challenges of 21st century are global scarcity of water and food resources, alarming environmental pollution, land degradation due to increasing soil and water salinity, reduction in cultivated area, and drastic increase in human population [1,2]. The presence of heavy metals (HMs) in soil may pose major issues because of their absorption, uptake, and accumulation in vegetables and food crops in the different tissues such as leaves, stem, roots, and fruits [3]. In some cases, the level of toxic HMs surpasses permissible limits. Constant utilization of household and industrial wastewater for irrigation of vegetables can pose serious risk to the entire food chain and it is a real pathway through which HMs enter into the terrestrial ecosystem [4]. Heavy metals and metalloids are harmful substances that impact the nutrition quality and yield of food crops, cereals, and vegetables. Accumulation of HMs may induce a harmful effect on plant metabolic pathways, for examples, chlorophyll pigments, N metabolism, fruit yield, and quality [5]. Metal uptake by vegetables is influenced by soil physiochemical properties, for examples, pH, temperature, Ec, and nutrient availability, and exhibits dangers to ecological and human health [5–7].

Recently, most vegetable growers have begun to use a variety of non-traditional natural organic manures to increase crop profitability and to meet the shortfall in the world's need for food sustenance. Amendments of organic manure reduce the mobility and accumulation of traces metals in plants tissues [8,9]. The animal's compost contains basic supplements for vegetables nutrition. The utilization of organic manures including animals compost as a biofertilizer is most popular among farmers to use in organic vegetable production. Press mud is a waste material discharge after filtration of sugar stick juice at handling of sugar clear squeeze, and ascends to the best and is used for assembling of sugar. A strong waste material remains at the base known as press mud or channel cake [10]. In Pakistan, the production of press mud was around 1.28 million tons in 2012 [11]. Press mud and natural fertilizer can reduce the exchange of poisonous metals into nourishment crops [12]. Toward this path, the principal motivation behind this examination is to determine the heavy metal gathering in soil and vegetables and furthermore decide the impact of non-conventional biofertilizers, for example, PW, PM, and FYM on vegetables.

A significant amount of micro and macronutrients are present in natural manure. It is an imperative worry to keep up and increase the agribusiness profitability and satisfy the interest of nourishment in the not-so-distant eventual fate of expanding populace [13]. Overabundant utilization of synthetic chemical fertilizer increases soil contamination, which eventually goes into water and disrupts the natural pecking order, causing health-related issues [14]. The addition of organic amendments is a common practice to reduce HMs accumulation in food, vegetables and cereal crops and to enhance the soil fertility and crop productivity [15]. These organic residues act as soil conditioner and affect the chemical and physical properties of soil positively, which is useful for immobilizing HMs in soil [15,16]. They help in the mitigation of environmental pollution by reducing their mobility through increased sorption. Supplementation of soil with organic amendments such as poultry waste can decrease the HMs uptake by plants [17,18].

There are several organic wastes products and biological nutrient fertilizers that can assist soil sustainability and plant growth. The PW, PM, and FYM are good examples of organic fertilizers. Domestic sewage waste and solid sludge contain vast amounts of organic and inorganic supplements for plants development [19]. Accessibility of sewage sludge and industrial waste material is available near big cities areas and industries without any legal compulsion in Pakistan. Consequently, this organic and inorganic matter has often high pH, basic solvent salts, high oxygen demand, and all out dissolvable salts, for example, nitrites, nitrates, and cation on Na, K, Mg, and Ca reported by [20]. Pakistan is an extremely populated and horticultural country [21,22]. Lands of Pakistan are generally utilized for cultivating to meet the interest of nourishment for future difficulties. Good yield varieties of vegetables and chemical fertilizers are insufficient. For the reinforcing of agribusiness, it is imperative to find viable and ecologically friendly ways to deal with using of poultry waste rather than chemical fertilizer. The poultry sector has an incredible impact to address poverty, lack of healthy sustenance, and joblessness-related issues. In another way, poultry ranches generate feces, feathers, eggs, and undigested material during meat cleaning. Sometimes, this compost is used as a biofertilizer in growing vegetables [23].

In another way, this waste contains vital measures of harmful metals. Higher presence of harmful metals in the soil moves into vegetables [24]. Cd, Pb, and Ni are remarkably poisonous metals among all other elements for people and animals [25]. Cd enters into the human body through nourishment or water and remains in body for long time potentially causing kidney diseases, vomiting, stomach disorders, and anemia and other blood disorders. High contents of Pb in sustenance and water can cause sickliness and other blood issues in people [26]. Permissible limits for Pb are very low; even a small quantity of Pb has clear toxic effects as compared to other HMs [27]. High concentrations of Ni deliver lethal indications including lung and blood malignancy, while Cr actuates mucodermal ulceration, diseases of the respiratory tract, and hypersensitivity such as in the skin [28].

Some other elements, for example, Fe, Cu, Co, and Zn are essential for humans within a tolerable range; if their concentration increases from the prescribed dose, it may cause metabolic disorder. The vegetables, which are mostly consumed in Pakistan, were used to compare with possible limits, set up by different organizations.

The annual herb coriander (*C. sativum* L.) is most typically used for seasoning. Its plant seeds, leaves, and roots are edible; however, their flavors and applications are quite different. It is a flavoring ingredient in confections, beverages, and baked goods. Linalool, borneol, citronellol, camphor, geraniol, coriandrin, dihydrocoriandrin, coriandrin A-E, coumarins, phthalides, flavonoids, and other phenolic acids and terpinene are all major components of coriander's volatile oil [28,29]. Coriander can be used as a whole plant or processed to increase the palatability of ripe fruits (seeds). Coriander is used in cooking as a whole plant, primarily fresh leaves, and ripe fruits. The coriander plant is a rich source of micronutrients, linoleic acid, α-tocopherol, and vitamin K. The plant's leaves are high in vitamins, while the seeds are high in polyphenols and essential oils. Coriander's flavor comes from its essential oil, which contains a lot of linoleic acid and furanocoumarins (coriandrine and dihydrocoriandrine). Coriander is also used in meals because of its antioxidant, anti-diabetic, anti-mutagenic, anti-anxiety, and antibacterial properties, as well as its analgesic and hormone-balancing effects [28,29].

Coriander has attractive attributes acquiring a higher growth rate and large biomass when grown at soils that received different organic fertilizers. However, site-specific detailed information is not available on the use of *C. sativum* plant in metal phytoremediation from the polluted sites. Thus, the present study aimed to evaluate the uptake of HMs (Cd, Co, Cr, Cu, Pb, Fe, Mn) by amending PW, PM, and FYM as natural mobilized organic materials and their impact on health via food consumption by humans. The *C. sativum* was selected for biomonitoring of contaminated sites with HMs through organic amendments and screening the pollution load characteristics in this promising species with a wide range of geographical distribution. Subsequently, these plants could be used in a wide range of pollution monitoring networks. The present research work was planned to determine the effect of PW, PM, and FYM on the metal accumulation in *C. sativum*, bioconcentration of metals, pollution status of soil, and health risk due to consumption of this contaminated *C. sativum.*

2. Material and Methods

2.1. Experimental Area

The place selected for experiments was the Botany Department of the University of Sargodha during winter season of 2016 to spring season of 2017. Sargodha is a major city in Pakistan's Punjab province, in the northeast (32°8′0″ N, 73°7′0″ E) (Figure 1). Sargodha, having a total area of 5854 square kilometers, is an agriculturally focused metropolis. The multipurpose pot experiment was conducted to study the agronomical growth performance and toxic trace metals bioaccumulation and pollution load in coriander. The coriander was cultivated in soil and different waste-based organic fertilizers such as PW, PM, and FYM were applied in the experiment pots and mixed with clay soil before seed sowing.

2.2. Pot Experiment, Sowing, and Treatment Application

Clay pots were selected for the current experiment. Each pot containing clay and loamy soil was amended with 50% press mud, poultry waste, and farmyard manure. The material was blended or mixed prior to use to ensure uniformity. The PW, PM, and FYM were collected from sub urban area of Sargodha, Pakistan. Surface-sterilized (sodium hypochlorite at 10% concentration) seeds of coriander were sown manually in individual pots (10 seeds/pot), containing clay loam soil mixed with corresponding organic amendment and kept in a glasshouse maintaining 25/20 °C day and night temperature, respectively, with a 55–60% relative humidity. The germination (emergence of radicle and plumule), was checked at the end of 10 days. Organic manures (PW, FYM, and PM) were added to each pot in three replicates and total 12 pots were designated for each treatment by following completely randomized design (CRD). All the organic manures were applied to each pot as 1:1 (50% organic manure + 50% loamy clay garden soil). Each pot was watered with tap water until the first leaves developed. The plants were then watered with groundwater to keep the soil moist. After 90 days after sowing, the coriander was harvested.

Figure 1. The map of study area.

2.3. Sample Collection and Preparation

The soil samples (12) were collected from control and treatment pots. Twelve vegetable samples were randomly collected from pots and cleaned with distilled water and diluted HCl to remove dust particles and impurities. These samples were first air dried before being placed in labeled polyethylene bags and stored in a 70 °C oven for three days. The incubated soil and vegetable samples were digested using the wet digestion method. First, 1 g of soil and plant samples were digested at 80 °C with a 5:1:1 mixture of HNO_3, H_2SO_4, and $HClO_4$ until a clear solution was obtained. After cooling, filter the solution through Whatman filter paper #42 to make the final volume 50 mL. Following sample digestion, the HMs levels in the vegetation samples and in the treatments were determined via atomic absorption spectrophotometry [29]. The soil physio-chemical properties and heavy metal contents (pre- and post-treatment) are reported in Supplementary Table S1.

2.4. Spectroscopic Analysis

The heavy metal content of soil and *C. sativum* samples was determined using an atomic absorption spectrophotometer (AA-6300 Shimadzu Japan). The metals investigated were Pb, Cu, Co, Mn, Cd, Cr, Zn, and Fe. The AAS experimental procedures and analytical methods were carried out in accordance with the manufacturer's instrumentation and applications guide, as well as the European Commission [30] guidelines, El-Ansary and El-Leboudy [31] and Liu et al. [32].

Limit of detection (LOD) values were calculated using standard literature methods [33]. LOD was defined as the value at which the blank solution's standard deviation (SD) and signal/noise ratio were both 10. The detection limits are 0.001 (Cd), 0.007 (Co), 0.012 (Cr), 0.01 (Cu), 0.04 (Pb), 0.036 (Fe), 0.012 (Mn) and 0.052 (Zn) (mg/kg) as reported in supplementary Table S2.

2.5. Quality Assurance

Diagnostic mark standardization values obtained from Merck were used for apparatus calibration (Germany). Deionized water was used throughout the work, and crystal pupillages were meticulously cleaned. Value declaration was completed by assessments of consistent Specialized Position Quantifiable (SRM-2711 for soil and SRM NIST 1577b for vegetable) and approximations of copies for individual groups of samples to certify the consistency of results. The mean SRM recoveries for soil were 104 percent, 96 percent, 93 percent, 92 percent, 95 percent, 98 percent, and 90 percent for Pb, Cu, Co, Mn, Cd, Cr, Zn, and Fe, respectively, and the mean SRM recoveries for vegetable were 95 percent, 91 percent, 94 percent, 109 percent, 92 percent, 97 percent, and 88 percent.

2.6. Soil-to-Plant Transfer Factors

2.6.1. Bioconcentration Factor (BCF)

The BCF was calculated by Equation (1) [8].

$$\mathrm{BCF} = \frac{\mathrm{M_c\ (Crop)}}{\mathrm{M_s\ (Soil)}} \tag{1}$$

where M_C indicates the metal concentration (mg/kg) present in the crop and M_S represent concentration of metal in agricultural soil where crops were grown. Toxic metals were detected in high levels in crops if the BCF value was more than one. Non accumulator, moderate accumulators, and hyperaccumulator plants had BCF values of 0.01, 0.1–1, and 1–10, respectively.

2.6.2. Daily Intake of Metals (DIM)

The daily intake of HMs was being determined by using the method described previously [28,34–36] through the following equation;

$$\mathrm{DIM} = \frac{\mathrm{C\ (Metal)} \times \mathrm{D\ (Food\ intake)}}{\mathrm{B\ (average\ weight)}} \tag{2}$$

where D (food intake) is the daily intake of a crop 0.345 (kg/person), C (metal) is the heavy metal concentration in the crop (mg/kg), and B is the average body weight (65 kg for this study).

2.7. Environmental Pollution Indices

2.7.1. Pollution Load Index (PLI)

The pollution load index was calculated as reported previously [35].

$$\mathrm{PLI} = \frac{(\mathrm{M})^{\mathrm{IS}}}{(\mathrm{M})^{\mathrm{RS}}} \tag{3}$$

where M is the metal content (mg/kg), IS is the metal content (mg/kg) in the examined soil where crops were grown, RS is the reference value of the metal in the soil. The background sample values were Cd (1.49 mg kg^{-1}), Cr (9.07 mg kg^{-1}), Cu (8.39 mg kg^{-1}), Fe (56.90 mg kg^{-1}), Ni (9.06 mg kg^{-1}), Mn (46.75 mg kg^{-1}), and Zn (44.19 mg kg^{-1}).

2.7.2. Health Risk Index (HRI)

The health risk index (HRI) was calculated by dividing DIM by the reference oral dose, as shown in Equation (4).

$$HRI = \frac{DIM}{RfD} \quad (4)$$

where DIM is the daily intake metal and RfD is the oral reference dose for metal that was 0.003 for chromium (USEPA 2000). The HRI is less than 1 if safe for health, if its value is greater than one, the consumer will be at risk [36,37].

2.8. Statistical Data Analysis

One-way ANOVA was used in SPSS 23 to estimate the significant difference between actions, and the numerical implication was verified at the 0.05, 0.01 and 0.001 levels [34]. Hierarchical cluster analysis was also performed using IBM SPSS 23 software. The clustering analysis organizes the similarity/dissimilarity relationships between the samples. Principal component analysis (PCA) was performed on different parameters means following exposure to organic manures PW, PM, and FYM application and compared to un-amended control using JMP 12.0 software.

3. Results and Discussion

3.1. Element Concentrations in Soil

Vegetables are vital constituents of public food and are daily consumed by the human beings to meet the body requirements of nutrients, minerals, and vitamins. However, bioaccumulation of HMs in higher quantities will result in several sickness and clinical problems to public via food consumption [5,6,38]. Different metals such as cadmium (Cd), chromium (Cr), and arsenic (As) can cause cancer in human beings. The plants grown on polluted soils can results in bioaccumulation of toxic metals in different plant tissues. The excess intake of the HMs can damage the skeletal, cardiovascular, endocrine, nervous, enzymatic, and resistant system in human body [22].

In soil samples, HMs (Cd, Co, Cr, Cu, Pb, Fe, Mn, and Zn) ranged from 0.332 to 0.457, 0.488 to 0.562, 0.135 to 0.779, 1.222 to 1.310, 0.108 to 0.179, 0.719 to 5.68, 0.023 to 0.537, and 4.53 to 17.63 mg/kg, respectively (Table 1). The order of metal contamination in soil was Pb < Cr < Mn < Cd < Co < Cu < Fe < Zn. As a result of three organic fertilizer amendments (T1, T2, and T3), Zn, Fe, Cu, and Co showed higher concentrations while Pb, Cd, Mn, and Cr showed the lower concentrations (Table 1). Various organic amendments when applied to the soil showed different behavior regarding the concentration of HMs in the soil. Some organic fertilizers increase the levels of some HMs and decrease the others and vice versa. As expected, organic amendments increased Zn level while Pb was lowest in the soil profile. However, it was lower than the European Union standard (Table 1). Application of PW in the soil causes accumulation of Zn and Fe, PM cause significant increase in soil Zn and Fe while FYM elevated the Zn and Cu concentrations in soil profile. ANOVA results were non-significant for Cd, Co, Cr, Pb, and Zn while being significant for Cu levels in the soil samples.

The organic amendments application might influence soil pH, Ec, and available heavy metal concentrations. After harvesting, an increase in soil Ec (1.438 s/m after amendment with PW, 2.35 s/m after amendment with PM and 1.89 s/m after amendment with FYM). The pH was observed from for PW (8.1), PM (8.18), and FYM (7.6). The pH plays an important role in the availability of different nutrients to plants. Absorption of nutrient by plants increases at pH (6.0–9.4) while below 6.0 and above 8.8 significantly reduces the availability of nutrients to plants [42]. In the present study, pH was in the range of 8.58–8.80 following treatment with PW, PM, and FYM and cause facilitation of diverse nutrients availability to *C. sativum*. Several authors reported bioaccumulation and absorption of nutrients and HMs following an increment in the pH [8,35]. Other researchers also reported positive effects on the soil pH and Ec [43]. Contrary to the present study, Zn and Pb were significantly higher in the soil samples that exhibited organic amendments as reported by

Amadi et al. [44]. Various factors (pH of the soil, organic matter, redox possible, Ec, and bioaccumulation) might favor the concentration of HMs in the soil [7]. It was observed that compost application caused a significant reduction in the soil bulk density, enhanced water holding capacity, and improved gas diffusion and air permeability by increasing organic carbon sequestration [45].

Table 1. Heavy metal concentrations (mg/kg ± S.E.) in soil *and C. sativum*.

Metals in Soil (mg/kg)	Treatments				Maximum Permissible Limits
	Control	Poultry Letter	Sugarcane Press Mud	Farmyard Manure	
Cd	0.448 ± 0.003	0.332 ± 0.068	0.457 ± 0.004	0.394 ± 0.010	3
Co	0.537 ± 0.026	0.455 ± 0.093	0.562 ± 0.005	0.488 ± 0.005	50
Cr	0.135 ± 0.014	0.146 ± 0.004	0.779 ± 0.006	0.1896 ± 0.004	50
Cu	1.222 ± 0.006	1.310 ± 0.041	1.251 ± 0.006	1.254 ± 0.005	100
Pb	0.1791 ± 0.015	0.1266 ± 0.020	0.1083 ± 0.010	0.1416 ± 0.01	100
Fe	1.868 ± 0.007	5.682 ± 1.500	5.682 ± 0.100	0.7196 ± 0.01	21,000
Mn	0.537 ± 0.006	0.023 ± 0.004	0.0829 ± 0.030	0.0529 ± 0.020	2000
Zn	4.535 ± 0.057	17.633 ± 1.120	12.692 ± 0.020	10.713 ± 2.700	300
Metals in *C. sativum*					
Cd	0.422 ± 0.004	0.435 ± 0.003	0.430 ± 0.008	0.401 ± 0.001	0.1–0.2
Co	1.849 ± 0.075	2.405 ± 0.200	1.941 ± 0.100	3.523 ± 0.595	0.01
Cr	0.160 ± 0.021	0.1662 ± 0.021	0.153 ± 0.001	0.169 ± 0.001	2.3
Cu	1.328 ± 0.022	1.335 ± 0.010	1.326 ± 0.005	1.405 ± 0.001	20
Pb	0.0512 ± 0.003	0.0637 ± 0.010	0.0954 ± 0.002	0.092 ± 0.001	0.3
Fe	1.481 ± 0.030	2.2913 ± 0.040	1.3257 ± 0.080	2.515 ± 0.002	425.5
Mn	0.3237 ± 0.010	0.1829 ± 0.004	0.2808 ± 0.001	0.1020 ± 0.006	500
Zn	2.0166 ± 0.010	8.603 ± 0.130	3.6750 ± 0.712	8.7045 ± 0.645	100

Sources: [30,39–41]; ±S.E.: Standard Error.

The presence of HMs in soil profile and subsequent absorption may increase or decrease following incorporation of organic amendments [46]. For the solid waste containing organic matter, composting was a very inexpensive and dependable method [28]. The compost was harmful to the environment if it contained contaminants such as HMs. If the concentration of HMs in the compost was high, then they were poisonous to plants, soil, human health, and marine life. By affecting key microbial processes, HMs display toxic effects towards soil biota and HMs also reduce the activity and microorganism numbers present in soil. The functional metabolism of a plant might constrain even low concentration of HMs. A potential hazard to human health was accumulation of HMs by plants resulting in gathering along the food chain. If excess amount was ingested through food, the HMs were capable of producing human health complications. The HMs are non-decomposable and insistent, have a long organic half-lives, and may be bio-accumulated through biological chains [47,48].

BCF estimated for Co demonstrated the highest at T3. It was lowest for Cd and range of metals was 0.1 to 8.48. DIM value was observed higher for Zn as 0.049 (mg/kg/day) at T1 treatment and lower for Mn as 0.0005 at T3 treatment. HRI estimations of HMs in *C. sativum L.* was found the higher for Cd at T1 treatment and the least for Cr at T2 treatment. PLI for Cd demonstrated the most astounding worth (2.89) and Mn demonstrated the lowest esteem (0.005). The order of PLI estimations of metals was Mn < Pb < Co < Cu < Zn < Fe < Cu < Cd.

3.2. Heavy Metal Concentrations in Coriander

In the present study, the HMs concentrations (measured in the whole plant of coriander) ranged from 0.401 to 0.435, 1.849 to 3.52, 0.153 to 0.169, 1.326 to 1.405, 0.0512 to 0.0954, 1.355 to 2.515, 0.102 to 0.3237, and 2.016 to 8.704 mg/kg for Cd, Co, Cr, Cu, Pb, Fe, Mn, and Zn, respectively (Tables 1 and 2). Among various metals, Zn concentration was found highest at FYM treatment and Pb concentration was found lowest at poultry waste.

The values of Pb metal varied from 0.0637 to 8.7045. Ismail et al. [49] reported that high concentrations for Cu, Zn, Fe, and Pb in vegetables were grown in soil contaminated with biosolids. The orders of different metals at T1, T2, and T3 treatments were Pb < Mn < Cr < Fe < Cd < Cu < Co < Zn, Pb < Cr < Mn < Fe < Cd < Cu < Co < Zn, and Pb < Mn < Cr< Cd < Fe < Cu < Co < Zn (Table 1).

Table 2. Analysis of variance of metals in soil and *C. sativum*.

Metal	Soil	Plant
Cd	0.442 ns	0.001 **
Co	0.448 ns	3.543 ***
Cr	0.432 ns	0.002 *
Cu	1.542 **	0.009 ***
Pb	23.49 ns	0.003 **
Fe	0.519 ns	0.075 *
Mn	0.324 *	0.059 *
Zn	1.118 ns	48.635 ***

*, **, ***: Significant at 0.05, 0.01, and 0.001 levels; ns: non-significant.

According to reports of Alam et al. [43], HMs in chili pepper and coriander roots were in the range of 754–808 and 188–196, but it significantly differed in shoots of the target vegetables. FAO and WHO recommend that the maximum permissible limits of Fe in the vegetables grown in any agricultural field is 425 (mg kg^{-1}). It was shown that the vegetable was contaminated with heavy metals pollutants and humans would have to avoid consumption of such vegetables to reduce the health risk impact. Adequate supply of Fe to human body helps to prevent anemia and maintains a healthy nervous system [48]. However, higher doses of Fe dust might lead to the development of serious sickness and respiratory problems. In the present study, among various metals, concentration of Zn was found to be highest at the T3 site as compared to other treatment and concentration of Pb was found to be lowest at the T1 site. The concentration of heavy metals in various vegetables varied significantly following sub-surface irrigation with treated municipal wastewater [38]. Srinivas et al. [50] observed the seedlings of three plant species, Coccinia (*Coccinia indica*—Crcubitaceae), menth (*Menth viridis*—Lamiaceae), and Trigonella (*Trigonella foenum*—Graecum—Fabaceae) showed substantial reduction in growth when exposed to 500 and 300 mgl^{-1} of Ni and Pb, respectively, and plant growth decreased progressively with increasing concentration of Ni and Pb. Bao et al. [51] found that the long-term sewage irradiations increase the soil organic matter content. The zone that was irrigated with it for 40 years exhibited the greatest accumulation of Hg, Pb, and Cu in the top soils (0 to 30 cm). Ghoneim et al. [52] studied that discharged water contains important levels of contaminants consider hazardous to the ecosystem.

3.3. Soil-to-Plant Transfer Factors and Environmental Pollution Indices

The BCF shows the bioavailability of HMs at particular location for different varieties of vegetable. In the present study, BCF of Zn, Co, and Fe showed higher values and BCF of Cd, Mn, and Cu showed lower values in *C. sativum*. The bioconcentration factor (BCF) is an important way to know availability of HMs transferred from soil to the plants and vegetables [3,34,38,53]. BCF has its own importance that showed the HMs transfer from soil to the plant [54]. However, in the present research, Zn was significantly higher in *C. sativum* (Table 3). Several reports indicate that BCF is a vital indicator to inform about the transfer level of a particular HMs in soil to vegetable [34]. BCF values of various HMs such as Co, Cu, and Mn were >1 following all treatments. Other researchers also reported higher BCF values > 1 that document greater ability of metal transfer from soil to vegetal [8]. Heavy metal toxicity was a great problem from ecological, evolutionary, and environmental aspects [55]. Singh et al. [28] gave the view that the soils of Pakistan in wheat production were not exempt from the deficiency of zinc reported worldwide. About 50% of soils of the world were zinc deficient if used for cereal production. The percentage was even higher in areas of calcareous soils. If pH increased to more than 5.5, the Zn could be desorbed back into solution which decreases not only the nourishing quality of grains but also grain yield. Consequently, almost two billion people suffer from Zn deficiency in all over the world.

Table 3. Bioconcentration factor (BCF) for vegetables/soil system.

Metals	Treatments			
	Control	Poultry Letter	Sugarcane Press Mud	Farmyard Manure
Cd	0.0985	0.101	0.100	0.173
Co	2.2756	5.901	3.143	8.489
Cr	0.4740	0.507	0.467	0.489
Cu	1.6708	1.345	1.566	1.489
Pb	0.1563	0.392	0.208	0.136
Fe	0.3785	0.171	0.290	0.705
Mn	1.1458	0.746	1.191	0.292
Zn	0.1241	0.639	0.207	0.752

Pollution load index (PLI) is determined to evaluate the concentration of HMs in soil [56]. In the present study, Cd showed higher concentration while Mn showed lower value in all treatments for PLI (Table 4). High value for HMs increases the PLI in soil. Hooda et al. [57] announced higher PLI esteems for traces metals in Nigeria when contrasted with the present examination. Consumption of vegetables growing in contaminated soil may increase the risk for public health. The PLI values were highest in poultry waste treatment for Cd (2.89) and lowest (0.0028) for Pb in poultry waste treatment (Table 4). Pollution load index demonstrates the presence of ruining the soil profile [35,58]. If the value of PLI is >1, then the vegetable is contaminated with HMs [6,38]. In the present study, the levels of Zn, Fe, Cd, Co, and Mn were lower than the values reported by Ahmed et al. [59] for contaminated vegetables from wastewater irrigated in Bangladesh. Ogoko et al. [60] reported comparable values of PLI that ranged from 1.15 to 2.19, indicating high pollution status of the sampling stations. In another study, the PLI value of arsenic was higher in the contaminated soil and vegetables [61]. Other studies also reported PLI values in different crops and vegetables [3,42]. The HMs not only contaminate the food crops, water, and air, but are also hazardous to human and animal health [62]. For example, the submission of wastewater that is salty/sodic results in decreased yield of crop and weakening of soil corporal/biological possessions [28]. By the exogenous increase in lead level, the accumulation of lead in plants also increases. Lead can cause a comprehensive variety of biochemical and physiological dysfunctions on seed sprouting, standing of water, plant growth, and nitrate accommodation [63,64].

DIM and health risk assessment showed the entry of HMs into food chain. There are many sources responsible for contaminated soil and underground water. Heavy metals are not easily degradable so they become a permanent part of ecosystem and are continuously taken up by vegetables and enter into human body via food chain. Oral references dose (RfD) is the value about the toxic pollutants that an individual consumes daily which cannot pose considerable dangerous effect for their lifetime

The HRI was higher in case of Cd in the T1 treatment and minimum HRI was obtained for Cr in T2 (Table 5). Other researchers [7,8,15,32,34] also reported higher values of HRI for Pb and Cd in different food crops, cereals, and vegetables. Cd and Pb are non-essential metals and are highly toxic even at lower levels. Cd has shown chronic effects including lung cancer, respiratory diseases, hair, and bone problems [8,34].

Table 4. Pollution load index (PLI) of metals for cultivated soil.

Metals	Treatments			
	Control	Poultry Letter	Sugarcane Press Mud	Farmyard Manure
Cd	2.8724	2.89	2.8768	1.552
Co	0.0892	0.0447	0.0678	0.045
Cr	0.225	0.2183	0.2183	0.23
Cu	0.1194	0.1182	0.2530	0.425
Pb	0.0057	0.0028	0.0080	0.011
Fe	0.4800	0.6432	0.5604	0.437
Mn	0.0060	0.0052	0.0050	0.007
Zn	0.3675	0.3043	0.4000	0.261

Table 5. DIM and HRI for human of heavy metals by consumption of vegetables samples collected from experiment site.

Metals	Treatments	Control	Poultry Letter	Sugarcane Press Mud	Farmyard Manure
Cd	DIM	0.0024	0.0025	0.0024	0.0023
	HRI	2.4265	2.5012	2.4725	2.3057
Co	DIM	0.0106	0.0138	0.0111	0.0202
	HRI	0.2472	0.3215	0.2595	0.4710
Cr	DIM	0.0009	0.0009	0.00088	0.0138
	HRI	0.0006	0.0006	0.0005	0.0092
Cu	DIM	0.0096	0.0076	0.0191	0.0138
	HRI	0.2407	0.1919	0.4781	0.3457
Pb	DIM	0.0002	0.0003	0.0005	0.0005
	HRI	0.0841	0.1046	0.1567	0.1511
Fe	DIM	0.0085	0.0131	0.0076	0.0144
	HRI	0.0121	0.0188	0.0108	0.0206
Mn	DIM	0.0018	0.0010	0.0016	0.0005
	HRI	0.0453	0.0256	0.0393	0.0143
Zn	DIM	0.0115	0.0494	0.0211	0.0500
	HRI	0.0313	0.1336	0.0571	0.1352

3.4. Cluster Analysis

Based on the cluster method (between group linkage), a cluster analysis was performed to determine the similarities between the variables and to show the dendrogram of contaminated soil and coriander from all the amendments (T0: control, T1: poultry waste (PW), T2: press mud (PM), and T3: farmyard manure (FYM) (Figure 2). As a measure of similarity, this method employs linear correlation coefficients. The variables with the highest similarities are cluster/linked first, and variables are only connected if they are highly correlated. The correlations between two variables are averaged after they have been clustered.

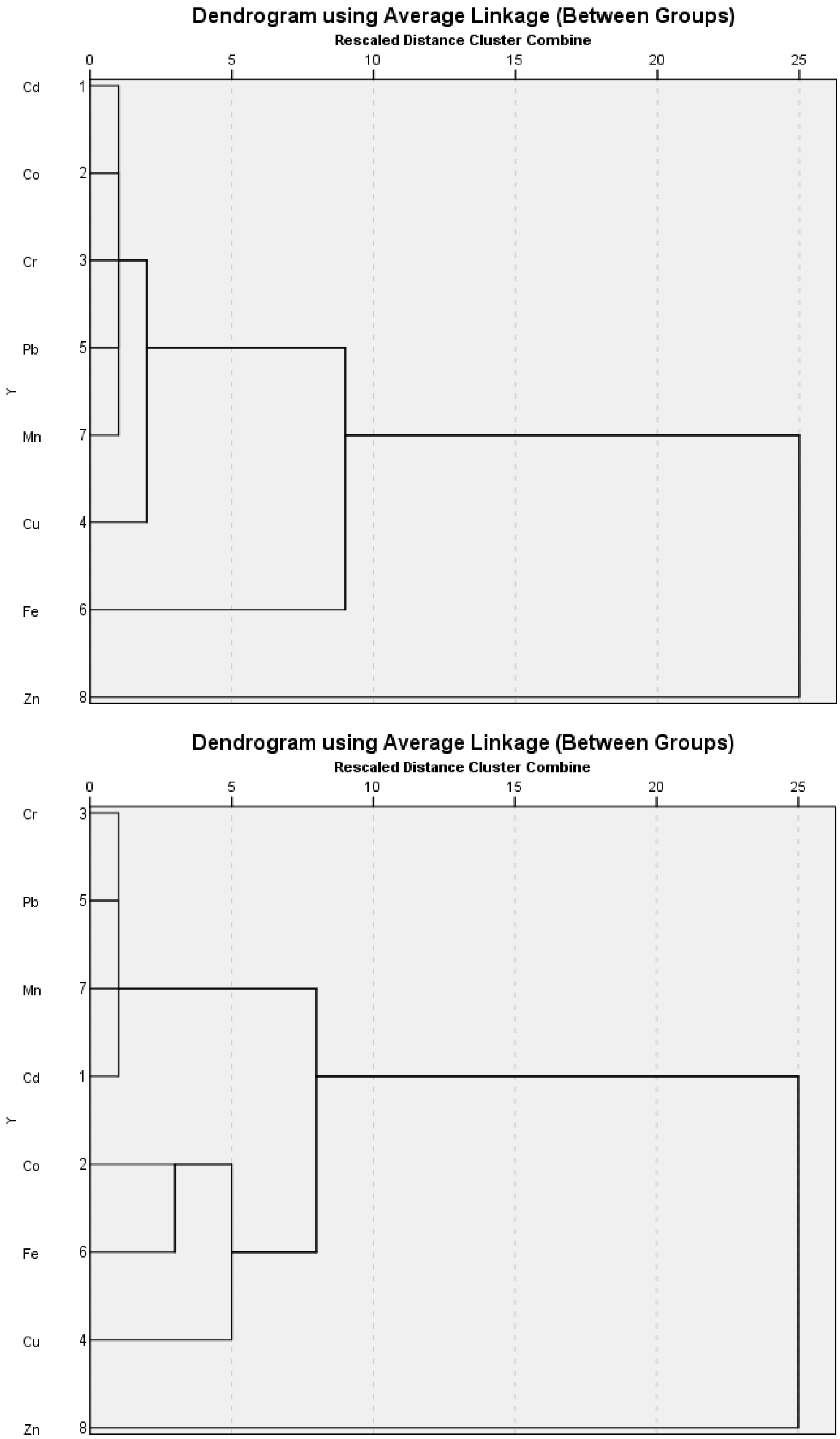

Figure 2. Dendrogram constructed from totally 8 metals based on the four treatments. It is generated with a hierarchical cluster analysis with Average Linkage (Between Groups).

In terms of similarities, the current cluster analysis results show that the contaminants studied fall into four groups: (I) Zn; (II) Fe; (III) Cu; and (IV) Cd-Co-Cr-Pb-Mn. Cluster 1 (Zn) is, however, moderately associated with Clusters II (Fe) (Figure 2). This suggests that the pollutants in Cluster I (Zn) may have some anthropogenic sources in common.

However, cluster II (Fe) is partially associated with cluster III (Cu), indicating that the hazard metals in cluster II and III may also have some anthropogenic sources in common. Furthermore, other contaminants such as Cd, Co, Cr, Pb, and Mn appear to be derived in part from sources other than Zn, Fe, and Cu, and to have originated primarily from other organic fertilizer sources.

In the case of plants, CA results show that pollutants are divided into four clusters based on similarities: (I) Zn; (II) Cu; (III) Fe-Co; (IV) Mn; and (IV) Cr-Pb-Cd. Cluster 1 is occupied by Zn, and it is associated with clusters II (Cu) and III (Figure 2). This revealed a common source of pollution for all of the HMs. Cluster III (Fe-Co) is partially associated with Cluster II (Cu) and demonstrates that hazard metals have a common anthropogenic source. Meanwhile, Cd, Cr, and Pb appear to derive from sources other than Zn, Fe, and Cu, and exhibit other organic fertilizer sources.

3.5. Principal Component Analysis (PCA)

The soil and plant samples were subjected to PCA analysis to determine the HMs content. Principal factors were chosen from variables with eigenvalues greater than one. Figure 3 depicts the rotated component matrix. Multivariate analysis (i.e., principal component analysis (PCA)) has been shown to be a useful tool for identifying the source of HMs [65]. The first two principal components, PC1 and PC2, explained the majority of the data variance (73 percent of total variance, eigenvalue 1) and are important in explaining HMs contamination in soil (Figure 3).

PC1 explained 97.4% of the total variance and was significantly positively loaded with Zn and Fe (in the first rotated matrix); Cu in the second rotated matrix and correlated with T1: poultry waste (PW), T2: press mud (PM). PC2 explained 2.11 percent of the total variance and was found to be significantly correlated with Cd, Co, Cr, Pb, and Mn. PC1 explained 93.3% of the total variance in *C. sativum* and was significantly positively loaded with Zn (in first rotated matrix) and correlated with T1 and T3. Fe, Co, and Cu are in the second rotated matrix and are correlated with T3. The PC2 explained 6.4 percent of the total variance, was significantly correlated with Cd, Cr, Pb, and Mn, and had a negative correlation with pollutant source (Figure 3).

It is also worth noting that HMs from the same source are always clustered together with high loadings [66]. Other authors [67,68] have suggested that the presence of Cr and Ni in soil is most likely due to natural enrichment by weathering processes. According to Facchinelli et al. [69], Ni and Cr inputs in agricultural soils are as low as the concentrations already present in the soil. A similar mechanism of origin could be proposed for Zn, Cu, and Fe. PC2 could be interpreted as anthropogenic sources, implying that Cu and Fe were derived from organic amendments.

The higher level of Zn, Fe, and Cu in the analyzed soils could be attributed to the overuse of organic fertilizers such as PW, PM, and FYM. Tian et al. [70] discovered that Cd is most likely a result of large amounts of manure applied by individual households engaged in intensive agriculture. Furthermore, Cu in the soil may be derived from manure utilization, which is known to increase Cu quantity in the treated soil as it is derived from feed additives used in livestock diets [71].

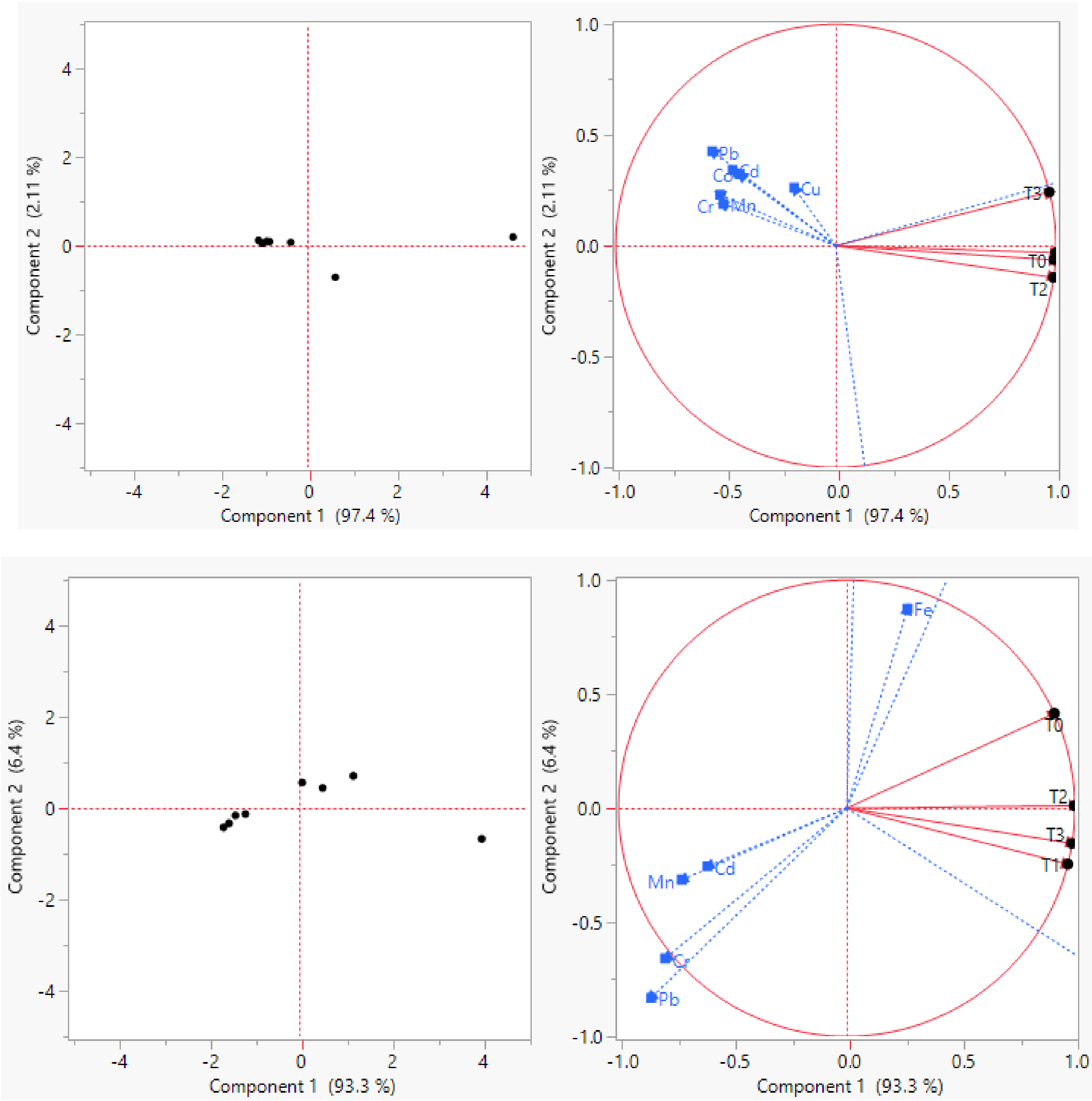

Figure 3. Principal component analysis (PCA) shows the effect of four organic fertilizers (T0, T1, T2, T3) amendments on correlation among different heavy metals in coriander via PCA (PC 1 and PC 2). Treatments (T0, T1, T2, T3) represent, control, poultry letter, sugarcane press mud, and farmyard manure.

4. Conclusions

The research aimed to evaluate the PW, PM, and FYM HM bioaccumulation in coriander (*C. sativum* L.) to evaluate the pollution severity of soil and to examine the health risk due to the consumption of coriander in Sargodha, Pakistan. Co, Cu, Fe, and Zn metal concentrations in *C. sativum* were higher than for other major pollutants. Ecological risk was found to be minimal due to lower concentration of HMs in soil profile than the declared USEPA permissible limits. Health risks to humans were also very low because *C. annuum* exhibits low levels of bioaccumulation of HMs (except Cd) that were lower than World Health Organization standard limits. The farmers should try to avoid irrigation of their vegetables with untreated wastewater and minimize the use of unauthenticated or untreated organic manures. Different management practices and microbial-based heavy metal remediation solutions should be included in the farming system to minimize the HMs toxicity, bioaccumulation, and to ensure food safety for consumers. Multivariate analysis suggests that HMs such as Zn and Fe metals and to some extent Co and Cu have some common anthropogenic origins. Excess application of PW, PM, and FYM as organic fertilizer in the soil should be avoided to minimize the ecological risks. Knowledge of the various types of pollutants, metal contaminants, metalloids, and critical plant stages at which HMs can enter in ecosystem and in the food chain and the sickness resulting from them are vital in order to protect public health.

Supplementary Materials: The following supporting information can be downloaded at: https://www.mdpi.com/article/10.3390/su141811374/s1, Table S1: Soil physio-chemical properties and heavy metal contents; Table S2: Operating conditions for the analysis of metals using flame atomic absorption spectrometry.

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