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RESEARCH ARTICLE

Fertilizer-driven heavy metal contamination in tomato soils: Insights from multivariate source apportionment

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Abstract

Excessive use of chemical fertilisers has increased the accumulation of heavy metals in agricultural soils, posing serious threats to soil health and food security. However, multi-index ecological risk assessments and depth-wise contamination profiles of soils under intensively fertilised tomato systems in India remain poorly characterized. This study quantified the concentrations of Fe, Mn, Zn, Cu, Cd, Cr, Pb and As, along with associated physicochemical and biological properties, in one hundred soil samples collected from intensively cultivated tomato fields in Chintamani, Karnataka, at both surface (0-20 cm) and subsurface (20-40 cm) depths. Pollution levels were assessed using the geo-accumulation index (I_{geo}), enrichment factor (EF), pollution index (PI) and ecological risk index (E_i and RI), owing to their ability to account for geogenic variability, anthropogenic inputs and ecological hazards. Potential sources of metals were identified using multivariate statistical methods. Most soil attributes and metal concentrations showed highly significant depth-wise differences ($p < 0.001$), indicating pronounced anthropogenic enrichment in surface soils. Surface soils exhibited elevated concentrations of Cd (0.66-5.40 mg kg^{-1}), As (9.44-30.80 mg kg^{-1}), Pb (18-39.40 mg kg^{-1}), Mn (525-1870 mg kg^{-1}) and Cu (38.41-83.29 mg kg^{-1}), while Fe, Zn and Cr were below levels of concern. EF and RI identified Cd, As and Pb as priority pollutants, with Cd accounting for more than 60 % of the total ecological risk, despite classification of most soils as "uncontaminated" by $I_{geo.}$ Subsurface soils had reduced anthropogenic influence, with Cd, Pb, As and Mn remaining the concern. Principal component and cluster analysis distinguished anthropogenic (Cd, As, Pb, Mn and Cu) from geogenic (Fe, Al, Zn and Cr) sources. Depth-specific management, region-specific background values and future work on metal speciation, bioavailability testing and isotopic source tracing are critical for improving ecological risk assessments and safeguarding soil sustainability and food safety in the study area.

Keywords: agglomerative cluster analysis; background concentration; ecological risk; heavy metal pollution; intensive cultivation; principal component analysis

Introduction

The ecological health and sustainability of ecosystems are closely associated with soil quality and functionality, particularly in a country like India, where 55 % of the population depends on agriculture (1, 2). Indian soils are under increasing pressure due to intensive farming and frequent application of agrochemicals. According to national evaluations, over 52 % of Indian soils have some degree of degradation and nutrient imbalances affecting about 37 % of cultivated soils, largely because of the overuse of nitrogen, phosphorus, potassium (NPK) fertilisers (3, 4). Unlike the atmosphere and aquatic systems, elements in the soil tend to be more stable, unless the thresholds are crossed, leading to long-term or even irreversible pollution (5). These elements are distributed among different fractions in heterogeneous soil systems, including water-soluble, exchangeable, carbonate-bound, oxide and hydroxide-associated (Fe and Mn bound), organic matter-bound and residual mineral-bound forms, with their potential mobility governed by numerous chemical and biochemical processes as well as the physicochemical properties of both the soil and the elements themselves (5, 6). Over time, their content gradually decreases due to processes such as leaching, plant uptake, erosion and evaporation (5-7). In acidic soils, Cd and Pb exhibit mobility levels that are approximately 2-5 times higher than those in neutral soils (8). Long-term fertiliser trials in India have demonstrated that consistent phosphate fertiliser application can increase plant available Cd by 18-32 % over 10-15 years, especially in vegetable production belts (8, 9). Unlike macronutrients, anthropogenically introduced metals persist in soils for decades, as only 5-15 % enter mobile fractions annually, while the remaining becomes associated with residual minerals, organic matter or Fe/Mn oxides (5, 6).

Phosphate fertilisers are the primary source of Cd because most sedimentary phosphate rocks (PRs), which make up 85-90 % of the world's PR supply, contain 20-200 mg of Cd per kg of P_2O_5, whereas igneous PRs typically contain less than 1 mg of Cd per kg of P_2O_5 (10). The finished Phosphorous fertilisers retain trace elements such as Pb, As and other trace elements along with Cd during processing of PR (4, 10). India depends on approximately 95 % imported mineral P with a P-use efficiency of about 32 %. This makes the fertiliser context even more pertinent (4).

Commercial NPK (and compound) fertilisers may contain trace metals (Cd, Pb, Zn, Cr and Ni), with concentrations varying widely depending on fertiliser type, origin and manufacturing processes (11, 12). The highest burdens are typically found in NPK (7-20-30) formulations and commercial N and NPK products carry multiple metals, such as (2.0-8.0 mg kg^{-1}), Cd (11.5-31.3 mg kg^{-1}), Cr (29.8-118.5 mg kg^{-1}), Cu (7.8-26.3 mg kg^{-1}), Fe (16.5-2209 mg kg^{-1}), Mn (20.3-5290 mg kg^{-1}), Ni (6.2-27.8 mg kg^{-1}), U (2.0-82.8 mg kg^{-1}) and Zn (1.4-166 mg kg^{-1}) (11-13).

Heavy metals enter soils through different pathways such as contaminated fertilisers, herbicides, irrigation water, atmospheric deposition and organic amendments in cultivation systems. For instance, depending on the rock source, Indian phosphate fertilisers contain 2.3-78.9 mg kg^{-1} of Cd and frequent application results in gradual metal build-up in soil. According to monitoring programs conducted across India, Cd was found in 28 %, Pb in 35 % and As in 22 % of intensively cultivated vegetable soils above baseline values (4). These findings indicate an increase in the deposition of Cd, Pb and As in high-input agricultural regions (3). Hence, in peri-urban horticulture systems with intensive fertiliser application, heavy metals can bioaccumulate in root zones and provide long-term phytotoxic, ecological and human health hazards (8, 9). Although both fertilisers and plant protection chemicals are the primary sources of heavy metals in agricultural soils, many pesticide formulations break down or dissipate in the environment rapidly. Hence, the impact from pesticides is frequently temporary, whereas the contribution from fertilisers is more enduring but frequently disregarded (9, 10, 14).

The dominant parent rocks in our study area-granite and gneiss contain significantly lower Zn, Cu, Mn and Fe than basalt (15–17). Although elemental composition is largely determined by parent materials, anthropogenic activities associated with rapid urbanization, intensive cultivation, such as application of inorganic fertilizers, pesticides and vehicular emissions, have a greater influence, on agricultural and vegetable soils of Chintamani since there is no presence of major industries to contribute heavy metal addition in the study area (8, 9, 11, 16). Consequently, the bioaccumulation of such metals can lead to increased and varied health risks among different population categories of the region (18).

Chintamani, in the Chikkaballapur district of Karnataka, represents a peri-urban agricultural hub with a strong horticultural economy, where tomato cultivation is especially intensive. Chintamani tomato market is the largest in Karnataka, supporting mostly small and marginal farmers. Cultivation peaks in summer (May-August), facilitated by favourable agro-ecological conditions for intensive tomato cultivation coupled with the over-dependency on chemical fertilizers and hybrid cultivars, which demand higher fertilizers but lesser pesticide application than high-yielding tomato varieties. The widespread preference for chemical fertilizers in our research area has been supported by the findings of previous studies, which revealed that only 20 % of farmers prefer organic fertilizer application to their fields, where 43.75 % of farmers prefer compost and 25 % of farmers prefer manure (19). This trend is further amplified by additional policy factors. For example, the NBS (nutrient-based subsidy scheme) increased fertiliser subsidies by 42 %, from ₹30.8 per kg to ₹43.6 per kg for phosphorus for the *kharif*, 2025, while maintaining the subsidy rates for potassium (K) and nitrogen (N) at the same level (20).

The peri-urban horticulture systems in India, where fertiliser intensity, farmer practices and market forces interact, have received little attention even though fertiliser contamination of soils has been researched worldwide. Furthermore, source apportionment and depth-specific contamination patterns are still poorly understood. In this context, the present study focuses on the multivariate assessment of heavy metal contamination in soils subjected to excessive fertilizer application in tomato-growing areas of Chintamani, Karnataka, at surface and subsurface levels. The overall pollution status was quantified using the various pollution indices of heavy metals, taking the background concentration of the concerned metals in the earth's crust and both Indian soils and world soils. A multivariate statistical approach was adopted to assist the interpretation of geochemical data using PCA and CA. This study connects the statistical results to real-world agro-geochemical conditions while recognizing the combined effects of both natural processes and human activities.

Materials and Methods

Description of the study area

Chintamani lies between 13° 6' to 13° 42' N latitude and 77° 51' to 78° 12' E longitude, encompassing a geographical area of 867 km^2. The study area is classified as the eastern dry zone of Karnataka (zone 5) and receives an annual precipitation of 400-750 mm and experiences the lowest temperature of 15 °C in December and the maximum temperature of 35 °C in April. The soils are sandy loam in texture, moderate in moisture holding capacity and allow deep furrowing. The predominant red soil overlying the granite and gneiss from which it is derived is shallow, well-drained to excessively drained, reddish-brown to yellowish-brown, gravely sandy loam to sandy clay loam and moderately to severely eroded. A major part of the studied area (87.10 %) is covered by alfisols and 12.6 % by inceptisols (21).

Soil sampling and analytical procedures

Before laboratory analysis, a preliminary survey was conducted to identify farmers who produce tomatoes intensively and specific data was gathered about how often they grow tomatoes each year and how much fertilizer and insecticide they use. In 2023, fifty soil samples, each at subsurface (20-40 cm) and surface (0-20 cm), hence a total of 100 samples, were collected using random sampling across tomato fields with Global Positioning System marking. Each sample was a composite of five sub-samples. The location details of the soil samples collected are shown on the map (Fig. 1).

Collected soil samples were air-dried, ground using a wooden pestle and mortar to avoid metal contamination and sieved using a 2 mm sieve. Processed samples were stored in clean, labelled polyethene bags. Further samples were analysed using acid-washed containers and contact with metallic tools was avoided. Physicochemical properties such as soil particle size, bulk density, particle density, porosity, pH, electrical conductivity, soil organic carbon, soil available nitrogen, phosphorus, potassium, sulphur, exchangeable cations, exchangeable acidity and soil biological properties like microbial biomass carbon, microbial biomass nitrogen, enzyme activities (dehydrogenase, phosphatase and urease) were assessed using standard analytical procedures (Supplementary Table S1), while their role in influencing heavy

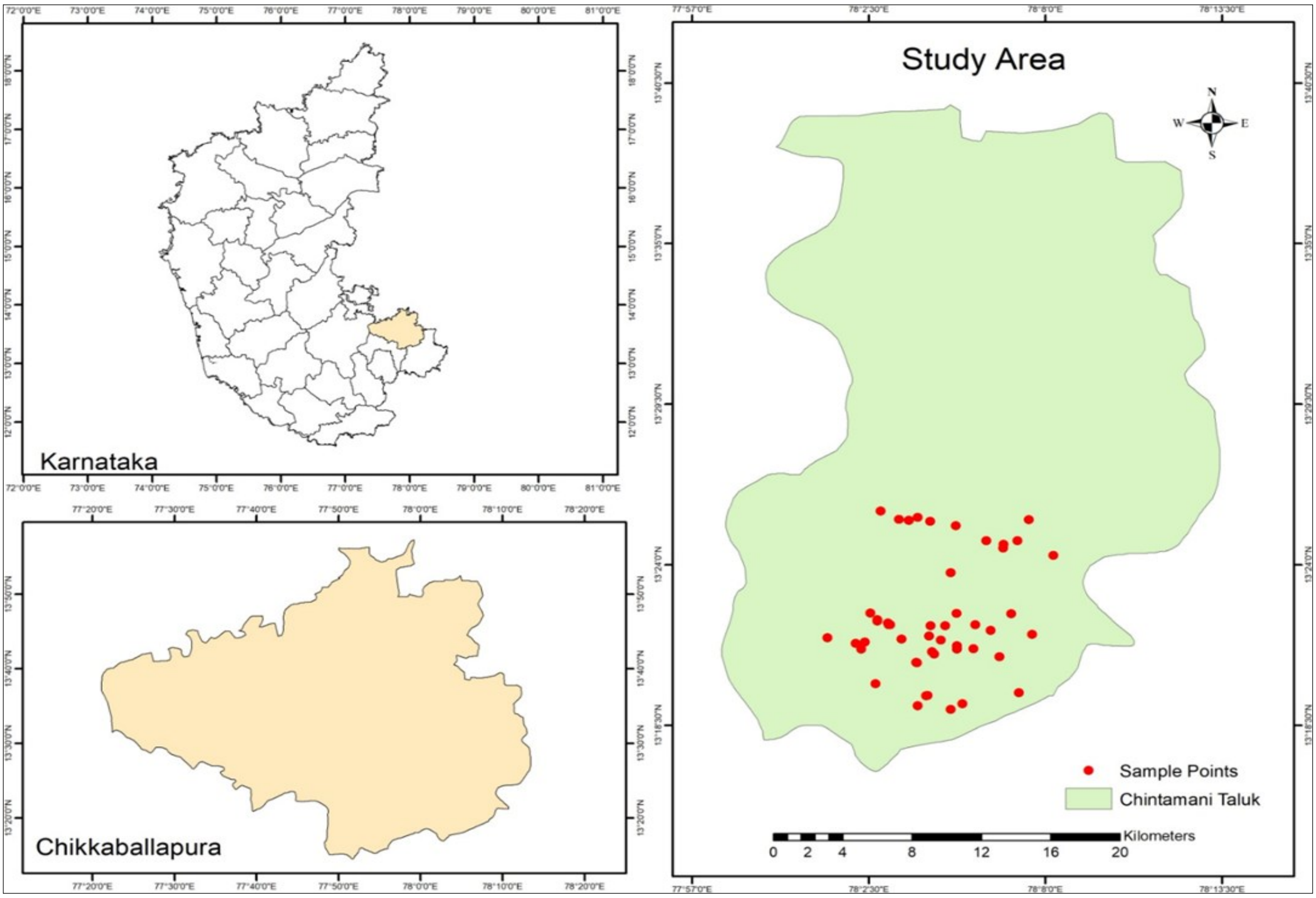

Fig. 1. Location of the study area and geo-referenced soil sampling points (red dots) in tomato-growing soils of Chintamani, Karnataka, India.

metal distribution is explained in the discussion section. The analytical results are presented in Supplementary Table S2.

Total heavy metal analysis

United States Environmental Protection Agency (EPA) method 3050B was used to prepare the soil samples to determine the concentrations of Al, Fe, Mn, Zn, Cu, Cd, Cr, Pb and As (22). After weighing 1 g of dry soil, 10 mL of nitric acid was applied. The samples were heated for 10 to 15 min at 95 °C. To reduce effervescence, 5 mL of hydrochloric acid and 10 mL of hydrogen peroxide were added gradually after cooling. Particulates were eliminated by centrifugation (2000-3000 rpm for 10 min), followed by filtration using 0.25 µm filter paper. The filtrates were collected in 100 mL volumetric flasks and volumes were adjusted.

ICP-OES (inductively coupled plasma–optical emission spectroscopy; Thermo Scientific iCAP 7000 Series) was used to determine the elemental concentration under the following operating conditions: RF power at 1250 KV, a stability time of 20 sec, a flush pump rate of 35 rpm, an analysis pump rate of 20 rpm, an auxiliary gas flow of 1.0 L min^{-1}, a nebulizer flow of 0.65 L min^{-1}, a cooling gas flow of 15 L min^{-1} and a radial view height of 8.0 mm. We used the standard multi-element solution (1000 ppm) provided by Bengaluru-based Sigma-Aldrich-Merck.

The below detection limit (BDL) is defined as three times the standard deviation of blank measurements and it varied from 0.0016 µg g^{-1} (Mn) to 0.042 µg g^{-1} (Pb) for particular elements. The limits of quantitation (LOQ) were ten times the standard deviation. Calibration curves of all the elements displayed outstanding linearity ($r \geq 0.9988$). Precision, which represents high analytical repeatability and is expressed as relative standard deviation (RSD) of triplicate results, was < 2.5 % for all analytes (Supplementary Table S1). The spike recovery was computed by adding known quantities of each analyte to previously examined soil samples and re-digesting them under the same circumstances. The accuracy of the process was confirmed by the fact that all recoveries were within the USEPA acceptability limits (75-125 %).

Fig. 2 was generated using R Studio version 2025.05.1 and it displays the distribution of As, Cd, Cr, Cu, Fe, Mn, Pb and Zn in Fig.2 (a) surface and Fig.2 (b) subsurface soils while highlighting the top five hotspots for each element across the study area.

Contamination assessment methods

Geo accumulation index

The I_{geo} value was calculated to assess soil pollution levels by comparing current soil metal concentrations with national background values (Table 1). I_{geo} was calculated as follows (23, 24):

$$I_{geo} = \log 2\frac{Cx}{1.5\times cb} \qquad \ldots (1)$$

Where C_x is the concentration of individual heavy metal in sample x and C_b is the concentration of heavy metal in reference soil (national background value), since background metal concentration of the specific study area was not available. Factor 1.5 was introduced in equation 1 to minimize the effect of possible lithogenic variations in the background metal concentration (24).

Enrichment factor

To assess the extent of metal contamination, background concentrations in the Earth's crust were used as a benchmark, with Al serving as the reference element under the assumption that its crustal content remains unaffected by human activities (Table 1)

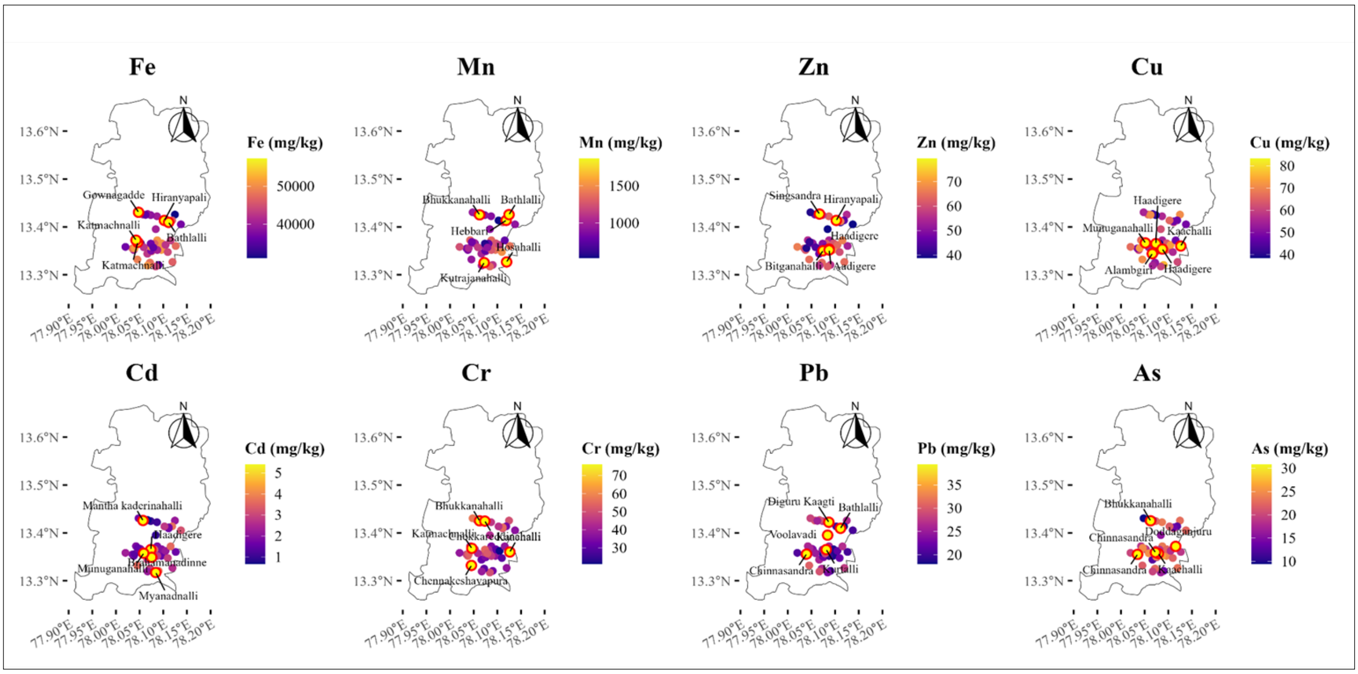

Fig. 2(a). Spatial distribution of heavy metals (Fe, Mn, Zn, Cu, Cd, Cr, Pb and As) in surface soil samples from tomato-growing soils of Chintamani, Karnataka, India. Warm colours indicate higher concentrations and the five highest sampling points per metal are highlighted.

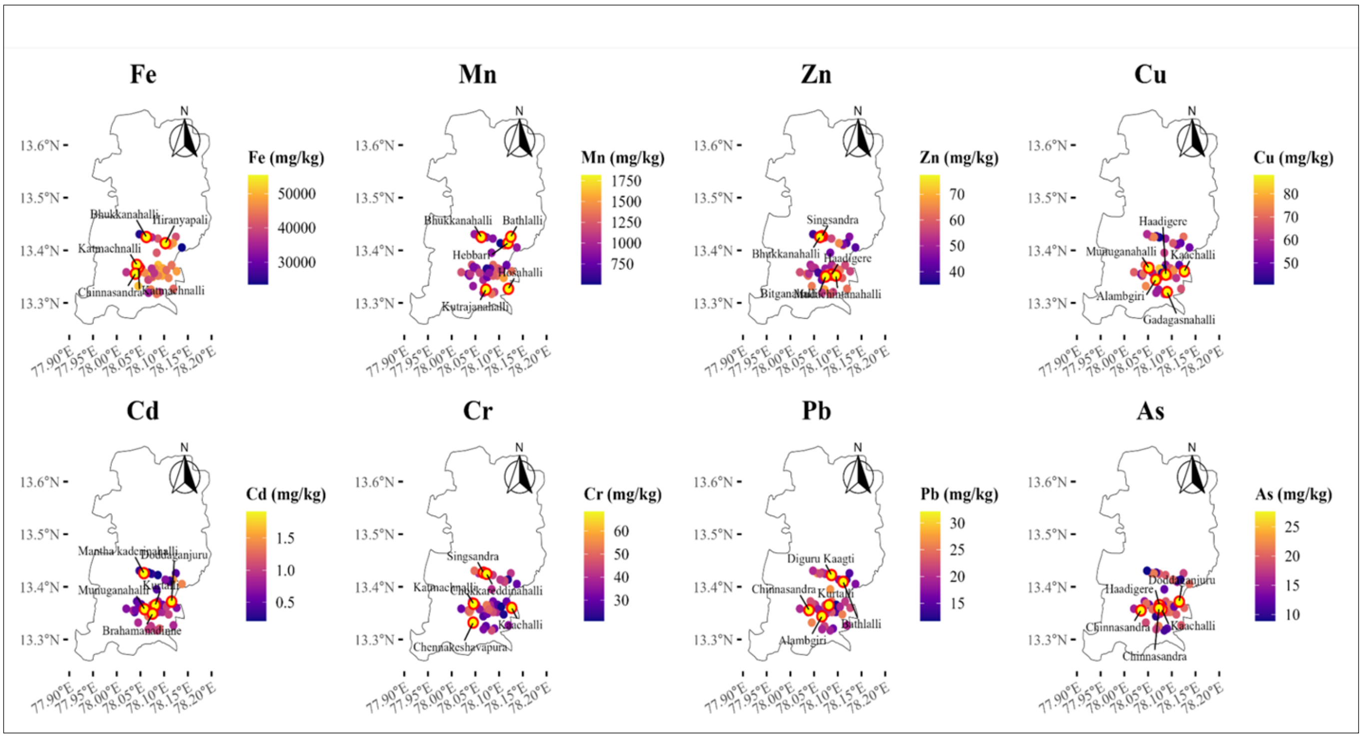

Fig. 2(b). Spatial distribution of heavy metals (Fe, Mn, Zn, Cu, Cd, Cr, Pb and As) in sub-surface soil samples from tomato-growing soils of Chintamani, Karnataka, India. Warm colours indicate higher concentrations and the five highest sampling points per metal are highlighted.

(25,26). EF were computed using the equation adapted from Reimann and de Caritat (25).

Where M (sample) = concentration of the examined heavy metal in the sample, M (crust) = concentration of the examined heavy metal in the crust, Al (sample) = concentration of aluminium in the sample and Al (crust) = concentration of aluminium in the crust.

$$\mathrm{EF} = \left[\frac{\mathrm{M(sample)}}{\mathrm{Al\ (sample)}}\right] / \left[\frac{\mathrm{M(crust)}}{\mathrm{Al\ (crust)}}\right] \quad \ldots\ldots\ldots (2)$$

Pollution index

The pollution level of a specific heavy metal (M) was assessed using the single PI, defined as the ratio of the metal concentration (M) in a soil sample to its corresponding reference value, i.e., (M) background (27).

Where PI = single PI, M (sample) = concentration of the examined heavy metal in the sample, M (background) = concentration of the examined heavy metal in the background (Indian national background and world soil background, since the background metal concentration of the specific study area was not available (Table 1).

$$\mathrm{PI} = \frac{\mathrm{M(sample)}}{\mathrm{M(background)}} \quad \ldots\ldots\ldots (3)$$

Table 1. Average natural concentrations of heavy metals in continental crust, Indian soils, worldwide soil and crust (mg kg^{-1}) (5, 66-70)

Elements	Continental crust		Indian soil	Worldwide soil
		Clemente et al. (2003)		
Fe	43200	-	32015	-
Mn	716	900	209	488
Zn	65	70	22.1	70
Cu	25	55	56.5	38.9
Cd	0.102	0.1	15.2	0.41
Cr	126	100	114	59.5
Pb	14.8	15	13.1	27
As	2	1.8	13.8	6.83

Ecological Risk Index (RI)

The potential ecological risk posed by heavy metal pollutants in the soil samples was assessed using the RI (28). The RI was determined as the sum of the individual risk factors for the heavy metals.

$$E_i = T_i f_i = T_i \frac{C_i}{B_i} \quad .. (5)$$

where Ei is the single risk factor for heavy metal i and is defined as:

$$\mathrm{RI} = \sum \mathrm{Ei} \quad \ldots (4)$$

Here, T_i represents the toxic-response factor for heavy metal i. The T_i values for Hg, Cd, As, Ni, Cu, Pb, Cr and Zn are 40, 30, 10, 5, 5, 5, 2 and 1, respectively (28). The ratio f_i denotes the metal pollution factor, which is calculated as the ratio of the measured concentration (C_i) to the background concentration (B_i) using Indian national background values and world soil background values (Table 1).

The various pollution indices used to assess heavy metal contamination in soils provide an understanding of the degree of pollution and associated risks (29). The assessed PI are represented graphically by box plots and violin charts generated using Python 3.13.

Statistical analysis

Multivariate statistical analysis was performed using XLSTAT-Student 2024.4.2.1426 software, where specific techniques such as Pearson correlation, followed by PCA and agglomerative hierarchical cluster analysis (CA) based on Ward's minimum-variance method, were performed. Results were represented in biplots, profile plots and dendrograms.

Results and Discussion

Concentration of total content of heavy metals in tomato-growing soils of Chintamani

Even though soil testing is crucial for managing nutrients, just 11.8 % of surveyed farmers tested their soils every year, 22.4 % only once every three years and 65.9 % never tested. More than 50 % of the surveyed farmers revealed relying on chemical fertilisers and plant protection chemicals more than the recommended package of practices. Only 22 out of the 50 farmers applied farmyard manure, though not at the recommended rates. Furthermore, 36 % did not use micronutrient fertilisers and 58 % did not use biofertilizers at all. The use of marigolds as a trap crop in tomato cultivation was notably unknown to 45 % of farmers.

Descriptive statistics (mean, standard deviation, skewness and kurtosis) were used to examine the levels of heavy metals in

Table 2: Descriptive statistics of total concentration of studied heavy metals and non-parametric tests (sign test and Wilcoxon signed-rank test) for depth-wise comparison of heavy metals in the surface and subsurface soil samples in study area of Chintamani, Karnataka, India

Total metal (mg kg^{-1} soil)	Surface soil (0-20 cm)						Sub-surface soil (20-40 cm)						Sign test (p-value)	Wilcoxn signed rank test (p-value)	Significance level
	Minimum-maximum	Mean	Median	SD	Skewness	Kurtosis	Minimum-maximum	Mean	Median	SD	Skewness	Kurtosis			
Al	35,745-63,019	51858.12	51682.50	5503.84	-0.35	1.19	38226-85194	56057.14	56111.50	6666.24	1.07	7.70	<0.0001	<0.0001	***
Fe	31127-57238	44205.28	44116.00	6204.44	-0.05	-0.81	23442-55396	44298.55	45336.50	7033.19	-0.92	1.10	0.001	0.071	ns
Mn	525-1870	1104.52	1044.00	312.96	0.48	-0.04	502-1816	1079.62	1044.50	310.20	0.48	-0.07	<0.0001	<0.0001	***
Zn	38.80-79.40	56.21	56.20	10.96	0.07	-1.02	35-77.40	55.23	55.55	10.24	0.17	-0.49	0.044	0.066	ns
Cu	38.41-83.29	61.34	61.62	12.08	-0.10	-0.65	40.68-88.11	64.25	65.70	12.47	-0.16	-0.62	<0.0001	<0.0001	***
Cd	0.66-5.40	2.53	2.30	1.14	0.56	-0.24	0.21-1.96	0.90	0.87	0.48	0.65	-0.29	<0.0001	<0.0001	***
Cr	21.10-76.10	45.80	45.19	11.56	0.43	0.04	20.89-68.40	41.04	41.48	10.70	0.20	-0.27	<0.0001	<0.0001	***
Pb	18-39.40	25.17	24.70	4.97	0.69	0.22	11.70-32.10	20.96	19.80	4.26	0.48	0.35	<0.0001	<0.0001	***
As	9.44-30.80	**21.26**	22.25	4.92	-0.41	-0.39	8.89-27.60	17.81	19.40	4.81	-0.17	-0.70	<0.0001	<0.0001	***

Note: ns = not significant ($p > 0.05$), * = significant at $p < 0.05$, ** = highly significant at $p < 0.01$, *** = very highly significant at $p < 0.001$.

both surface and subsurface soils (Table 2). Heavy metal pollution was significantly heterogeneous, according to high standard deviations with Mn and Fe exhibiting the highest variability. Metals with skewness values between -1 and 1 had normal distributions, whereas those with slightly positive skewness had anomalous distributions (30). The spatial distribution of As, Cd, Cr, Cu, Fe, Mn, Pb and Zn in surface and subsurface soils (Fig. 2).

For each element, the top five sampling sites with the highest concentrations above the respective background concentration in crust and Indian soils are highlighted. Strong vertical stratification was shown by the non-parametric tests (sign test and Wilcoxon signed-rank test), which showed substantial depth-related anomalies for Al, Mn, Cu, Cd, Cr, Pb and As (p < 0.0001). Conversely, there were no appreciable variations in Fe and Zn between surface and sub-surface soils (p > 0.05). Significant regional variations in elemental concentrations were caused by soil heterogeneity, which was influenced by local geomorphology, pedology and lithology of the respective area (16, 17, 30).

The slight variation in Mn and Zn with depth may be due to the compensation of leaching losses through external inputs, the differences in metal concentrations (Cd, Pb, As, Cr, Mn, Zn, Al and Fe) between surface and subsurface soils may be attributed to anthropogenic activities, such as agricultural inputs and atmospheric deposition, which were less pronounced in subsurface soils (11, 23, 31). The modest increase in Cu with depth in our study is consistent with previous findings that Cu remains in soils for years, with long-term trials demonstrating elevated levels even 21 years after amendment (32, 33).

In our study, As contamination may have originated from the use of As-containing pesticides and animal manures (9, 34). Another contributing factor was the naturally occurring elevated background concentration of As, which is around double the concentration in world soils and 6.9-7.66 times higher than that of the continental crust. In these soils, As binds to iron (hydroxides) as inner-sphere complexes as arsenate and arsenite (9, 34, 35). Under anoxic conditions, microbial reduction of As-rich Fe (III) (hydroxides) can dissolve As-bearing ferrihydrite, mobilizing geogenic As, though secondary Fe minerals may sequester some As through direct microbial Fe (III) reduction (9, 35).

The total concentration of Pb in surface and subsurface soils may be attributed to its association with clay minerals, Mn oxides, Fe and Al hydroxides and organic matter (11, 27). Since tomato crops need Ca, application of Ca sources may be the reason for retention of Pb in calcium carbonate along with phosphate particles (36). The heavy metal profile of tomato soils at Ajiwa Fadama farms in Nigeria showed a similar pattern, with Pb predominated and was above FAO/WHO allowed limits, while other metals remained within safe ranges (37).

In India, 47 % of soils are deficient in Zn and Zn contamination is found mostly in industrial soils (8, 38). In our study area, surface soils, having more negative charges than subsurface soils, showed greater Zn adsorption across all pH levels (31).

The total Cr content in both surface and subsurface soils was lower than the background levels reported for the Earth's crust, Indian soils and global soils, which indicated minimal pollution concern (Table 3). This aligns with the lack of significant industrial discharges, such as leather or tannery effluents, which are prevalent sources of Cr (39). In soils, Cr typically exists as the stable trivalent form [Cr (III)], characterised by low mobility and restricted bioavailability owing to its association with Fe/Mn oxides and organic matter (35, 39, 40). However, the elevated Mn concentration in our soils may facilitate the oxidation of Cr (III) to the more toxic and mobile [Cr (VI)] in aerobic environments, depending upon Mn reactivity and [Cr (III)] availability (39, 40). On the other hand, bacterial reductase enzymes and plant root activity in the rhizosphere can convert [Cr (VI)] back to [Cr (III)]. This process is particularly active in our study location, where intensive tomato cultivation takes place (35, 40).

Furthermore, putting our findings in a larger agro-environmental framework gives them more depth. For instance, heavy metals- As, Pb, Cr, Cu, Hg and Cd-in highland rice-producing soils in northern Thailand under rotational shifting cultivation were found to have soil concentrations of Cu, Cr, As, Pb and Hg that stayed below reference limits, but Cd was not found (41). In contrast, agricultural soils of Southeast Romania growing tomato had observed considerable Cd and Mn pollution, whereas Pb and Cr concentrations were comparatively low (42).

Al and Fe concentrations in soils increased with depth, suggesting that they were primarily pedogenic or geogenic in origin with weathering as a contributing process (5, 15). This aligns with the observation that the soils of the Deccan Peninsula in South India are rich in iron oxides and originated from laterite (43). Moreover, the difference in mean clay concentrations (Supplementary Table S2) between surface and subsurface soils in our study suggests that the process of clay illuviation in alfisols may have contributed to the accumulation of Al and Fe at greater depths (15, 31).

The type of soil also has a great influence on the total content of heavy metals in soils such as alfisols in the study area, which have a higher pH-dependent charge for metal retention (5, 6, 10). In comparison to more affected areas, the soil metal levels often indicated a comparatively low pollution in temperate regions, indicating a comparatively limited risk of heavy metal buildup (44).

Extent of the heavy metal contamination in the surface and subsurface soil samples

The I_{geo}, EF and PI results of heavy metals in tomato-growing soils (surface and subsurface) are represented in Fig. 3-5. The samples were classified into pollution categories using the soil contamination assessment methodology based on the heavy-metal indices (29) (Table 3).

Surface soils

The geo-accumulation index (I_{geo})

The I_{geo} values of Fe ranged from -0.63 to 0.25, with 74 % of the samples categorized as uncontaminated and the remaining 13 samples were categorized as uncontaminated to moderately contaminated. In contrast, I_{geo} values of Mn ranged from 0.74 to 2.58, with 64 % moderately contaminated samples, 32 % moderately to strongly contaminated samples and 4 % samples were uncontaminated to moderately contaminated with Mn. Zn showed I_{geo} values ranging from 0.23 to 1.26, with 84 % of the samples being uncontaminated to moderately contaminated and the remaining 16 % moderately contaminated. All the surface soil samples were uncontaminated with Cu, Cd, Cr and As since their I_{geo} values were ≤ 0. However, Pb exhibited significant surface soil pollution

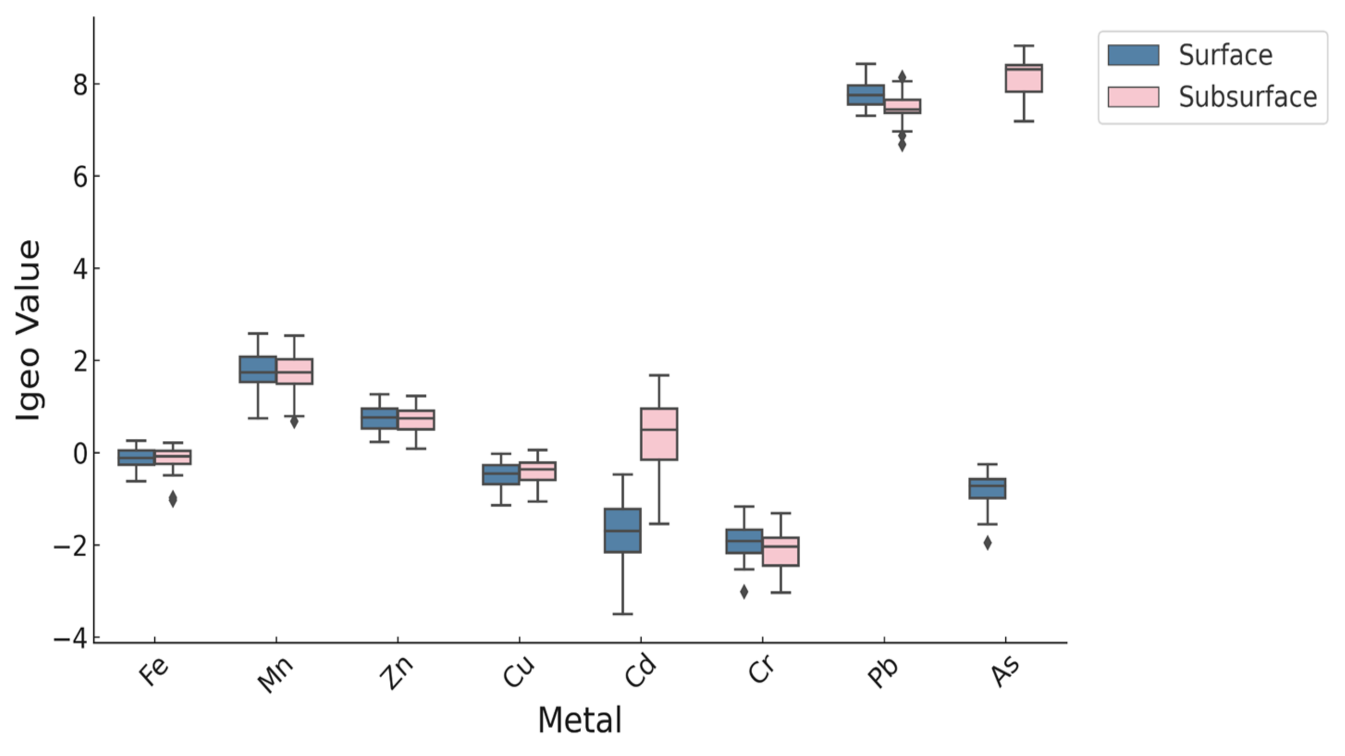

Fig. 3. Boxplots of geo-accumulation index (I_{geo}) values for heavy metals in surface and subsurface soil samples from tomato-growing soils of Chintamani, Karnataka, India, illustrating differences in contamination levels between depths.

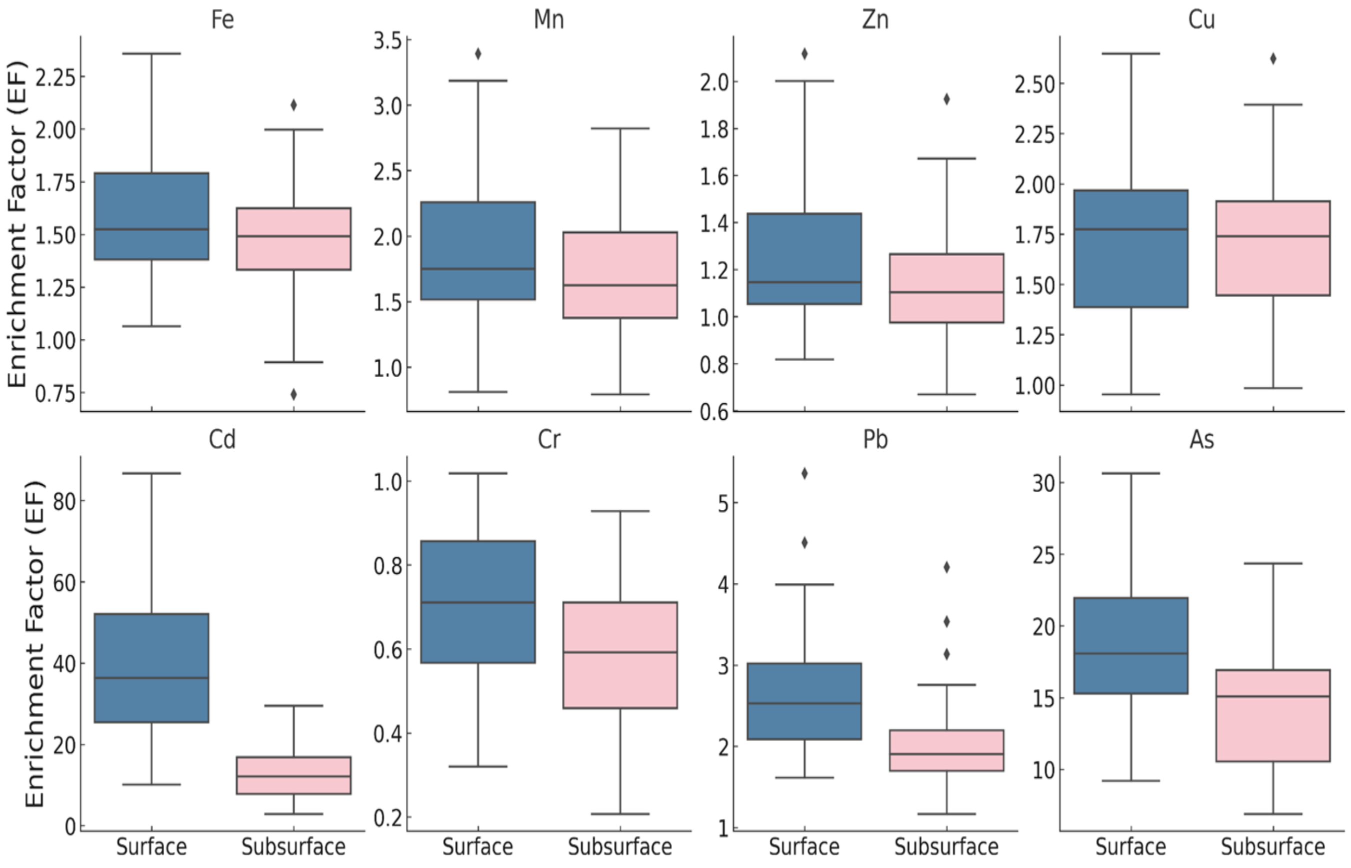

Fig. 4. Box-plot showing EF of heavy metals (log scale) in surface and subsurface soil samples from tomato-growing soils of Chintamani, Karnataka, India.

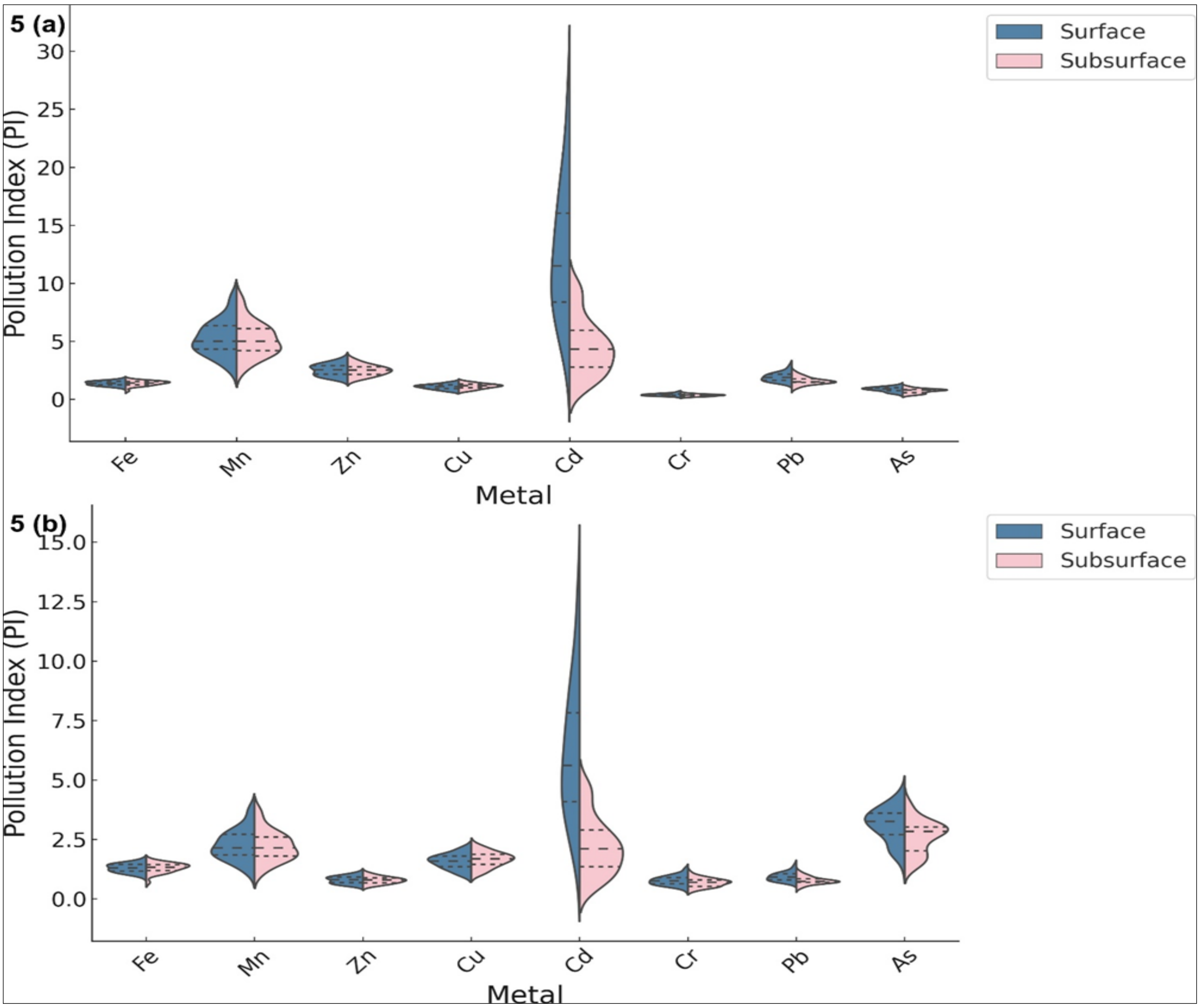

Fig. 5. Violin plots showing pollution indices of heavy metals in surface and subsurface soil samples from tomato-growing soils of Chintamani, Karnataka, India: (a) Pollution Index based on Indian soil reference values (PI′ = Ms / Mis), (b) Pollution Index based on world soil reference, values (PI″ = Ms / Mws), where, Ms = concentration of the metal in the sample; Mis = metal concentration in Indian soils; Mws = metal concentration in world soils.

Table 3. Classes of Heavy metal pollution indices: Igeo, EF, PI and RI (29)

Pollution indices	Value	Soil quality
I_{geo}	$I_{geo} \leq 0$	Uncontaminated
	$0 \leq I_{geo} < 1$	Uncontaminated to moderately contaminated
	$1 \leq I_{geo} < 2$	Moderately contaminated
	$2 \leq I_{geo} < 3$	Moderately to strongly contaminated
	$3 \leq I_{geo} < 4$	Strongly contaminated
	$4 \leq I_{geo} < 5$	Strongly to extremely contaminated
	$I_{geo} > 5$	Extremely highly contaminated
EF	EF < 2	Deficiency of minimal mineral enrichment
	EF = 2–5	Moderate enrichment
	EF = 5–20	Significant enrichment
	EF = 20–40	Very high enrichment
	EF > 40	Extremely high enrichment
PI	PI < 1	Unpolluted, low level of pollution
	1 ≤ PI ≤ 3	Moderate polluted
	3>PI	Strong polluted
E_i	$E_i < 40$	Low ecological risk
	$40 < E_i \leq 80$	Moderate ecological risk
	$80 < E_i \leq 160$	Considerable ecological risk
	$160 < E_i \leq 320$	High ecological risk
	$E_i > 320$	Serious ecological risk

I_{geo} values exceeding 5 in all the soil samples. The order of contamination was determined to be as follows: Pb > Mn > Zn > Fe.

Enrichment factors (EF)

The EF of Fe, Mn, Zn, Cu and Cr were in the range of 1.06–2.36, 0.81-3.39, 0.82-2.12, 0.95-2.65 and 0.32-1.02, with an average of 1.59, 1.9, 1.25, 1.73 and 0.71 respectively, exhibiting minimal metal enrichment. Pb exhibited moderate enrichment in the surface soils, with an average EF value of 2.63. The EF results indicated that As had significant contamination levels with a mean EF value of 18.36. The maximum contamination was observed for Cd, which showed EF values in the range of 10.04 - 86.64, with a mean of 38.95.

Pollution index (PI)

PI assessment of metals, compared with their Indian national background concentration, indicated widespread moderate pollution with Fe, Zn and Pb in majority of the soil samples (98 %) and 72 % of soil samples showed moderate pollution with Cu and 28 % were unpolluted with Cu. All the soil samples showed PI > 3 for Mn and Cd, indicating strong pollution of Mn and Cd, whereas all the samples and more than half the soil samples showed PI < 1 for Cr and As respectively, indicating minimal pollution with Cr and As. None of the samples were categorised as unpolluted for any of the metals, indicating a concerning level of anthropogenic impact. Finally, the overall order of pollution based on PI in the soil was Mn = Cd > Fe = Zn = Pb > Cu > As > Cr.

In contrast, when PI values were calculated taking the natural background concentration of each metal in world soil, most soil samples (98 %) showed moderate pollution with Fe and Cu, whereas the majority of soil samples showed low pollution due to Zn (88 %), Pb (74 %) and Cr (78 %). In contrast, soil samples showed severe pollution, with Cd and As having 70 % and 64 % of soil samples, respectively, exhibiting PI values > 3. The mean PI values for Fe, Mn, Zn, Cu, Cd, Cr, Pb and As were 1.3, 2.26, 0.80, 1.58, 6.16, 0.77, 0.93 and 3.11 respectively, giving a PI order of Cd > As > Mn > Cu > Fe > Pb > Zn > Cr.

Potential ecological risk index

The potential ecological risk indices (Ei' and Ei'') for individual heavy metals and multi-metals in the surface and subsurface soils in the study area are represented in Table 4. The mean E_i' values ranged from 2.54 for Zn to 378.96 for Cd, indicating a variation in potential ecological risk. With a maximum Ei' of 810.00, Cd showed the highest risk, suggesting a significant ecological risk. Similarly, the ecological risk (E_i'') of Cd, considering global soil background levels of metals, had the greatest mean (184.86), followed by As (31.13) and Pb (4.66). These findings indicated that Cd posed the greatest environmental hazard due to its high concentrations and steady contribution to the overall ecological risk.

The cumulative risk indices (RI' and RI'') showed corresponding average values of 406.02 and 230.78 respectively. Based on Indian soil background concentrations, RI' indicated a very high ecological risk, while the global soil background-based RI'' indicated a moderate ecological risk. Further, a maximum RI' of 849.91 emphasized the risk associated with Cd. Overall, the data distribution was stable among samples, as evidenced by the tight alignment of median and mean values. All metals showed a modest potential ecological risk, except for Cd.

Subsurface soil

Geo accumulation index (I_{geo})

All the subsurface soil samples were 100 % extremely highly contaminated with Pb and As, with average I_{geo} values of 7.49 and 8.13, respectively. In contrast, all the samples exhibited an I_{geo} value < 1 for Cr, implying they were 100 % uncontaminated with Cr. Mn showed

Table 4: Potential Ecological Risk Index of individual heavy metals and multi-metals in the surface and sub-surface soils in the study area of Chintamani, Karnataka, India

Surface soils	Individual metal												Multi-metal	
	Ei'						Ei''						RI'	RI''
	Zn	Cr	Pb	As	Cu	Cd	Zn	Cr	Pb	As	Cu	Cd		
Mean	2.54	0.80	9.61	8.68	5.43	378.96	0.80	1.54	4.66	31.13	7.88	184.86	406.02	230.87
Maximum	3.59	1.34	15.04	12.57	7.37	810.00	1.13	2.56	7.30	45.10	10.71	395.12	849.91	461.91
Minimum	1.76	0.37	6.87	3.85	3.40	99.00	0.55	0.71	3.33	13.82	4.94	48.29	115.25	71.65
Median	2.54	0.79	9.43	9.08	5.45	345.00	0.80	1.52	4.57	32.58	7.92	168.29	372.30	215.69
Subsurface soils	**Individual metal**												**Multi-metal**	
	Ei'						**Ei''**						**RI'**	**RI''**
	Zn	**Cr**	**Pb**	**As**	**Cu**	**Cd**	**Zn**	**Cr**	**Pb**	**As**	**Cu**	**Cd**		
Mean	2.50	0.72	8.00	7.27	5.69	135.60	0.79	1.38	3.88	26.07	8.26	66.15	159.77	106.52
Maximum	3.50	1.20	12.25	11.27	7.80	294.00	1.11	2.30	5.94	40.41	11.33	143.41	330.02	204.50
Minimum	1.58	0.37	4.47	3.63	3.60	31.50	0.50	0.70	2.17	13.02	5.23	15.37	45.14	36.98
Median	2.51	0.73	7.56	7.92	5.81	129.75	0.79	1.39	3.67	28.40	8.44	63.29	154.28	106.00

$Ei' = T_i f_i = T_i \frac{Ci}{Bi}$ where B_i = Indian national background values of metals & $RI' = \sum Ei'$

$Ei'' = T_i f_i = T_i \frac{Ci}{Bi}$ where B_i = global soil background values of metals & $RI'' = \sum Ei''$

[a] $E_i < 40$ indicates a low potential ecological risk; $40 < E_i < 80$ is a moderate ecological risk; $80 < E_i < 160$ is a considerable ecological risk; $160 < E_i < 320$ is a high ecological risk and $E_i > 320$ is a very high ecological risk.

[b] RI< 95 indicates a low potential ecological risk; 95 < RI< 190 is a moderate ecological risk; 190 < RI< 380 is a considerable ecological risk and RI > 380 is a very high ecological risk

moderate contamination in 66 % of samples and moderate to strong contamination in 28 % of subsoil samples, having a mean I_{geo} of 2.29. Zn showed no contamination to moderate contamination in 86 % of samples, exhibiting a mean I_{geo} of 0.71. Cd in subsurface soils was distributed across moderate contamination (22 %), uncontaminated (34 %) and uncontaminated to moderately contaminated (44 %) categories, with maximum, minimum and mean I_{geo} values of 1.67, -1.55 and 0.33 respectively. Cu and Fe posed no contamination concern since most samples showed I_{geo} values in the range of $0 \leq I_{geo} < 1$. Overall, the order of metal contamination in subsurface soils based on I_{geo} was As > Pb > Mn > Zn > Cd > Fe > Cu > Cr.

Enrichment factors (EF)

The EF of subsurface soils revealed minimal enrichment for Cr, Fe and Zn with mean EF values of 0.59, 1.47 and 1.13 r espectively. With EF values ranging from 2.82 to 29.52 (mean 12.89), Cd showed the largest anthropogenic influence, placing most samples in the significant to very high enrichment category. Additionally, As showed significant to very high enrichment in all samples with EF ranged from 6.88 to 24.35 (mean 14.30). Pb showed specifically moderate enrichment (EF: 1.16-4.21) while Mn (EF 1.72) and Cu (EF 1.68) showed minor to moderate enrichment. The order of enrichment in subsurface soils based on mean EF values was As > Cd > Pb > Mn > Cu > Fe > Zn > Cr.

Pollution index (PI)

The PI based on Indian background values demonstrated that subsurface soils were primarily contaminated by Mn and Cd, with mean PI values of 5.17 and 2.20, respectively, suggesting moderate to strong pollution. Fe exhibited low to moderate contamination (mean 1.30), whereas Cu showed moderate pollution in 64 % of samples (mean 1.58). On the other hand, most samples were unpolluted (PI < 1) with respect to Zn, Pb, As and Cr levels. Based on mean PI values, the contamination order in subsurface soils was Mn > Cd > Cu > Fe > Pb > As > Zn > Cr.

A contrasting pattern became apparent when global background values (PI) were used for evaluation. While Fe and Mn frequently exhibited moderate contamination, the majority of subsurface samples were unpolluted with Zn and Pb (PI < 1). Cd and As showed higher levels of pollution levels with 22 % and 32 % of samples respectively, falling into the strongly polluted category (PI > 3). The mean PI values followed the order, As > Mn > Cd > Cu > Fe > Zn > Pb > Cr. Overall, Mn and Cd were the dominant pollutants under the Indian background PI. As, Mn and Cd were the dominant ones under the global background PI.

Potential ecological risk index

Subsurface soil samples showed high ecological risk due to Cd, similar to surface samples as per Ei′ (135.60), but low potential ecological risk as per the Ei″ value of Cd (Table 4). Similar to surface soil samples, subsoil samples also exhibited low potential ecological risk of Zn, Cr, Pb, As and Cu as per the Ei′ and Ei″ values of respective metals. The cumulative risk indices, RI' and RI'', were averaged at 159.77 and 106.52, respectively, categorizing the subsoil samples under very high ecological risk according to both Indian soil background-based RI' and world soil background-based RI''.

Comparison of different heavy metal pollution indices in surface and subsurface soils

The mineralogical composition of atmospheric dust has a significant impact on EF (25). The differentiation between biogenic dust- derived from plant material or organic horizons and is enriched in Pb, As, Bi, Cd, Sb and Zn- and minerogenic dust, originating from soil horizons or rock weathering and is enriched in crustal elements such as Al, Fe, Li, Sc, Ti and Zr (25). Hence, dust composition alone can provide high EF values even in pristine surroundings.

The comparison of different heavy metal pollution indices in surface soil samples clearly stated that Cd posed significant level of contamination and potential ecological risk which exhibited extreme enrichment (10.04 - 86.64, mean: 38.95), severe pollution (highest among all metals) from PI assessment and very high ecological risk (Ei′ and Ei″) contributing the most to cumulative risk indices (RI′ and RI″). However, the geo accumulation index (I_{geo}) values of Cd in almost all the surface and sub-surface samples were ≤ 0. Instead, Pb was found to be concerning according to I_{geo}. Cd pollution in the studied area originated likely from anthropogenic sources since the Cd in soil is typically introduced artificially through deposition rather than Cd derived from geogenic materials found in the remaining insoluble fraction (9, 10, 45).

Following Cd, Mn and As showed moderate to severe pollution levels according to PI, I_{geo} values. As posed the second-highest ecological risk (Ei′ and Ei″) after Cd. The natural background concentration of As in Indian soils is 50.61 % higher than in world soils, coupled with long-term P application in the study area may have contributed to As accumulation in soils, despite reports of relatively low concentrations of As in P fertilizers (Table 3) (9, 34, 46). The frequent application of agrochemicals used to protect tomato crops, such as mancozeb (contains Mn and Zn), maneb, metiram, zineb (contains Zn and Mn) and various herbicides may have increased the Mn content in our soils (47, 48). However, the contribution of irrigation water to the observed rise of Mn and As in our soil samples was found to be minimal.

Even though Pb was found to be a major pollutant, its ecological risk was less than Cd and As. However, the I_{geo} values indicated severe Pb pollution in all samples, which may be due to the peculiar nature of Pb being considerably varying with soil type (5, 31). Fe and Cu showed moderate concerns because of their PI and E_i values, but minor contamination based on their I_{geo} and EF values. Zn and Cr were the least concerning heavy metals in our soils. In contrast to our findings, high chromium levels (average of 2652 mg kg^{-1}) resulted in a mean I_{geo} of 3.14 and EF ranging from 4.27 to 222.73 in the Ganga plain, where intense agricultural activity takes place (38).

The final pollution ranking in surface soils, from most to least concerning, was Cd > Mn> As > Pb > Cu > Fe >Zn > Cr. Similarly, in subsurface soils, the most concerning heavy metals were Cd, Pb, As and Mn whereas Cr, Cu, Zn and Fe exhibited low contamination and minimal ecological risk. The results corroborate that the components found in surface soil can frequently be linked to those found in subsurface soil (26). Natural processes such as leaching, weathering and capillary action, as well as water movement, biological activity or even human activity like farming or construction, may have caused elements from deeper layers to migrate upward or downward.

Identification of the sources of heavy metals by multivariate analysis

Correlation matrix of heavy metals in surface and subsurface soils

Al and heavy metals in surface and subsurface soils had varying associations, according to the Pearson correlation matrix (Table 5).

Table 5. Pearson correlation coefficient matrix for Al and heavy metals in the surface and subsurface soil samples in study area of Chintamani, Karnataka, India

Surfaces soils									
Variables	**Al**	**Fe**	**Mn**	**Zn**	**Cu**	**Cd**	**Cr**	**Pb**	**As**
Al	1	0.052	0.111	0.006	-0.03	0.119	0.057	-0.371	-0.001
Fe	0.052	1	0.051	-0.023	0.013	-0.115	-0.001	0.026	-0.067
Mn	0.111	0.051	1	0.007	0.102	-0.107	-0.093	-0.128	-0.083
Zn	0.006	-0.023	0.007	1	-0.174	-0.116	-0.023	-0.275	-0.031
Cu	-0.03	0.013	0.102	-0.174	1	0.042	0.078	-0.094	-0.068
Cd	0.119	-0.115	-0.107	-0.116	0.042	1	-0.314	-0.178	0.096
Cr	0.057	-0.001	-0.093	-0.023	0.078	-0.314	1	0.167	0.039
Pb	-0.371	0.026	-0.128	-0.275	-0.094	-0.178	0.167	1	0.011
As	-0.001	-0.067	-0.083	-0.031	-0.068	0.096	0.039	0.011	1

Subsurface soils									
Variables	**Al**	**Fe**	**Mn**	**Zn**	**Cu**	**Cd**	**Cr**	**Pb**	**As**
Al	1	0.195	0.244	0.113	-0.048	0.119	-0.173	-0.169	-0.111
Fe	0.195	1	0.154	0.197	0.159	-0.056	0.146	0.063	0.077
Mn	0.244	0.154	1	0.012	0.099	-0.058	-0.251	-0.121	-0.147
Zn	0.113	0.197	0.012	1	-0.044	-0.179	0.084	-0.041	0.038
Cu	-0.048	0.159	0.099	-0.044	1	-0.083	0.093	0.046	0.034
Cd	0.119	-0.056	-0.058	-0.179	-0.083	1	-0.404	-0.022	0.205
Cr	-0.173	0.146	-0.251	0.084	0.093	-0.404	1	0.164	0.18
Pb	-0.169	0.063	-0.121	-0.041	0.046	-0.022	0.164	1	0.014
As	-0.111	0.077	-0.147	0.038	0.034	0.205	0.18	0.014	1

Values in bold are different from 0 with a significance level alpha = 0.05.

In surface soils, a weak negative correlation between Al and Pb (r = -0.371) indicated their distinct origins. While Pb most likely may have originated from anthropogenic sources like long-term agri-input uses or atmospheric deposition, Al was likely the result of lithogenic inputs from alfisols derived from granite and gneiss (5, 16, 17). The positive correlations of Al with Fe and Mn in subsurface soils highlighted the dominance of natural mineral matrices in deeper horizons, which are less impacted by surface disturbances and are common geogenic origins (15-17).

Zn showed a negative correlation with Pb in surface soils and with Cd in subsurface soils, implying distinct sources. While Zn may have originated from both natural weathering and agricultural amendments such as phosphate fertilizers and agrochemicals (33, 37). The presence of Pb may have stemmed from both fertilizers and non-fertilizer sources, such as vehicular emissions, which reduces its spatial overlap with Zn-rich zones and Cd from fertilizer inputs (33, 36, 45). In contrast to surface soils, the stronger negative correlation between Cd and Cr in subsurface soils (-0.404 vs. -0.314 in surface soils) indicated enhanced antagonism, possibly due to differential leaching or immobilization processes (31, 35).

The subsurface Cd-As association, which indicates possible downward leaching, raises concerns regarding groundwater contamination (48, 49). The lack of strong Cu correlations (e.g., Cu-Fe: r = 0.159) suggested limited fertilizer-derived Cu inputs consistent with reduced usage of Cu-based agrochemicals in the study area (32, 33).

The majority of metals lacked strong positive correlations, suggesting that diverse sources contributed, with some elements possibly having overlapping origins (18, 23). The Pearson correlation matrices in our study showed relationships between variables, but these relationships may not be actual cause-and-effect mechanisms but rather indirect connections, common environmental processes or confounding factors (23, 27). Therefore, in order to separate overlapping sources and differentiate fertilizer-derived contributions from other anthropogenic activities, multivariate techniques like PCA and CA were crucial.

Principal component analysis (PCA)

Factor loadings showed the strength of association between each metal and the principal components (F1 to F5 = PC1 to PC5), with loadings above ± 0.5 typically considered significant. The extraction of factors was performed using PCA, followed by Varimax rotation with Kaiser normalization (eigenvalue > 1), which retained five principal components. The results of PCA revealed that these five components explained 69.67 % and 72.35 % of the cumulative variance in surface and subsurface soils, respectively. Among these components, PC1 and PC2 showed the highest percentage of variance in both soil layers (Fig. 6, 7). The rotated component matrices of heavy metals in the surface and subsurface soils are presented in Table 6.

Surface soils

In surface soils, PC1 was dominated by Pb, indicating a possible anthropogenic source, such as vehicle emissions and agricultural amendments, along with natural lithogenic components (Al, Cr) in surface soils (50). The dominance of Pb in our soils was also confirmed by its high I_{geo} values, which may be attributed to its strong association with clay minerals, Mn oxides, Fe and Al hydroxide and organic matter (31). Additionally, the use of Ca sources in growing tomatoes can increase Pb accumulation through co-precipitation with Ca-carbonates and phosphate particles (36).

PC2 loaded with Cd and Mn may be linked to agricultural inputs. PC3, explaining 13.689 % variance and influenced by Cu and Zn, may indicate differing sources or mobility or availability of these metals (49). PC4 loaded with Cr, As and Al reflected geogenic sources or residual soil content derived from parent materials (3, 5). PC5 showed strong loadings from Fe (0.780) and may be governed by a distinct process, possibly redox dynamics or parent material weathering (35).

Subsurface soils

In subsurface soils, PC1 (Cr, Pb, Mn, Al and Cd) and PC3 (Cd, Fe and As) reflected a mixed anthropogenic-metal signature with background geogenic variation. PC2 (Fe, Mn and Zn) may be linked to geogenic origin, related to parent material weathering or iron-manganese nodules in the subsurface (49, 51). PC3 (Cd, Fe and As) and PC4 (Cu and Zn) indicated differential mobility or adsorption patterns under subsurface redox conditions (35). PC5, having isolated loading on Pb, may have arisen, possibly from contamination persisting in deeper layers and was attributed to vehicular emissions and fossil fuel burnings (10, 11, 26).

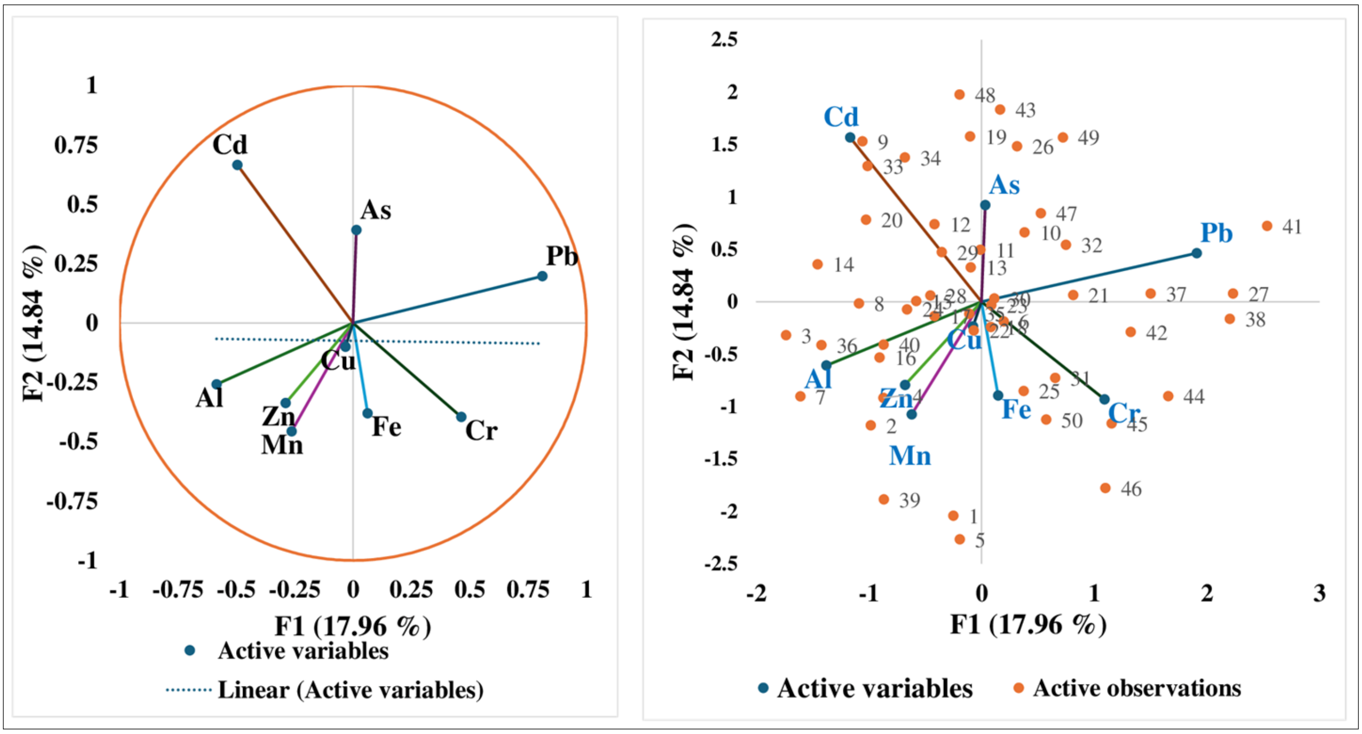

Fig. 6. PCA biplots (axes F1 and F2: 32.80 % of the total variance) illustrating relationships among heavy metals and the spatial distribution of soil samples in surface soil samples from tomato-growing soils of Chintamani, Karnataka, India.

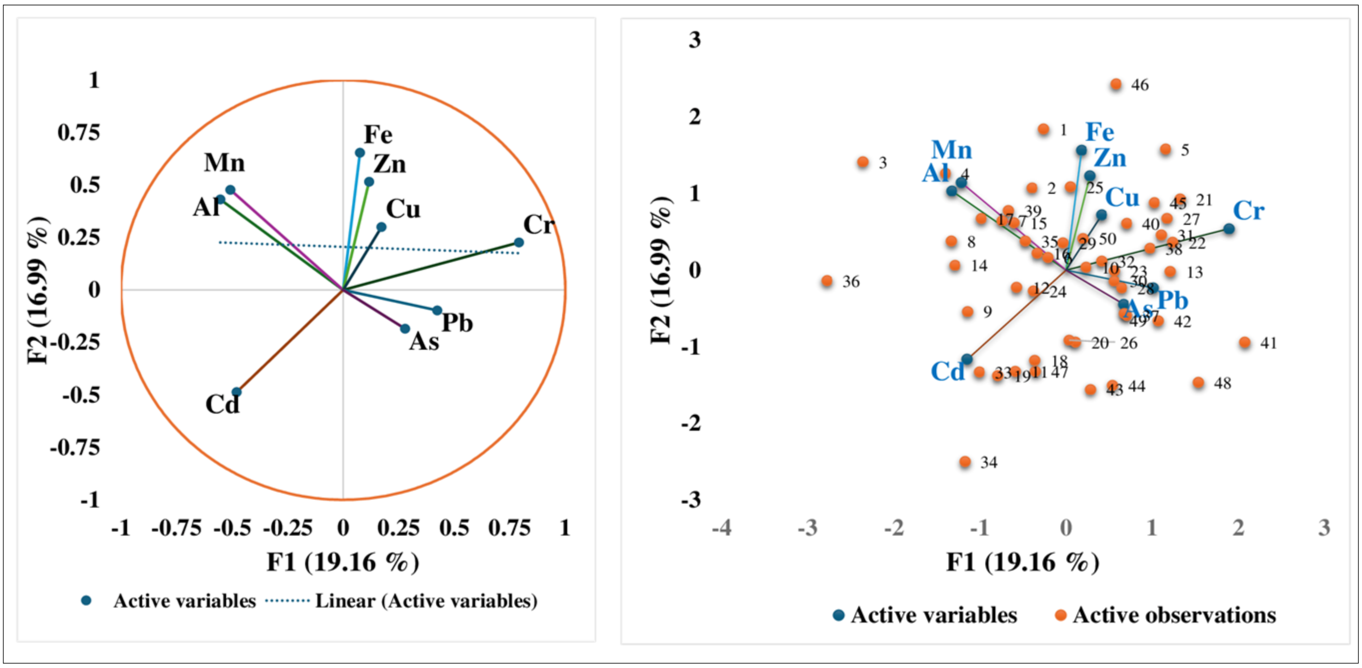

Fig. 7. PCA biplots (axes F1 and F2: 36.15 % of the total variance) illustrating relationships among heavy metals and the spatial distribution of soil samples in sub-surface soil samples from tomato-growing soils of Chintamani, Karnataka, India.

Table 6. The rotated component matrix of heavy metals in the surface and subsurface soils of tomato growing soils of Chintamani, Karnataka, India

	Surface soils (0-20 cm)					Subsurface soils (20-40 cm)				
Elements	**PC1**	**PC2**	**PC3**	**PC4**	**PC5**	**PC1**	**PC2**	**PC3**	**PC4**	**PC5**
Al	-0.584	-0.258	0.063	0.493	0.295	-0.553	0.431	0.188	-0.165	0.157
Fe	0.063	-0.380	0.175	-0.190	0.780	0.076	0.654	0.453	0.121	0.212
Mn	-0.263	-0.456	0.336	-0.237	-0.119	-0.507	0.477	-0.123	0.311	-0.048
Zn	-0.288	-0.337	-0.691	-0.161	-0.248	0.117	0.515	0.133	-0.570	0.072
Cu	-0.032	-0.100	0.720	0.170	-0.389	0.173	0.300	0.125	0.719	-0.346
Cd	-0.495	0.666	0.189	-0.001	0.091	-0.479	-0.486	0.576	0.086	0.146
Cr	0.464	-0.396	-0.049	0.657	-0.107	0.791	0.226	0.010	-0.092	-0.105
Pb	0.811	0.197	0.071	-0.142	0.123	0.424	-0.097	0.057	0.326	0.787
As	0.014	0.392	-0.215	0.506	0.175	0.279	-0.185	0.766	-0.088	-0.295
Eigenvalues	1.617	1.336	1.232	1.097	0.989	1.725	1.529	1.212	1.110	0.936
% of variance	17.962	14.841	13.689	12.192	10.986	19.165	16.987	13.467	12.333	10.395
Cumulative %	17.962	32.804	46.493	58.685	69.671	19.165	36.152	49.619	61.952	72.347

Overall, the PCA results showed that anthropogenic inputs (Pb, Cd, Cu, Mn and As) exerted a stronger effect on surface soils, while geogenic sources (Fe, Mn, Al, Zn and Cr) and residual anthropogenic contamination (Pb, Cd, As and Cu) had a larger impact on subsurface soils. This pattern indicated that subsoils represented both the lithogenic background and the downward movement or persistence of some metals, including Pb, Cd and As, whereas surface pollution was primarily caused by contemporary agricultural practices and atmospheric deposition (23, 37, 42).

Agglomerative cluster analysis (CA)

Hierarchical clustering of the sampling points

Based on information analysed from PCA, agglomerative hierarchical cluster analysis was performed by determining the Euclidean distance and agglomeration was conducted through Ward's method on surface and subsurface samples and organized in the dendrogram and profile plots to identify the identical geochemical groups and thereby help detect specific locations of contamination (Fig. 8, 9) (52).

Surface soils

Fig. 8(a, b) illustrates the dendrogram and the profile plot of cluster analysis for surface soil samples in the study area. Four clusters were found to be optimal (Silhouette index = 0.174, Calinski-Harabasz index = 9.854).

Cluster 1 (C1) included sample numbers 5, 8, 13, 14, 15, 24, 30, 35, 46 and 50 and showed low Pb and Cd pollution, low Fe levels, but significant levels of Cu in soils. Cluster 1 was possibly less affected by anthropogenic inputs or redox-poor environments (33, 53–55) and was likely a "low-pollution" group.

Cluster 2 (C2) comprised sample numbers 2, 3, 4, 6, 7, 11, 16, 17, 18, 23, 25, 36, 39 and 40, which recorded moderate pollution and showed enrichment with Fe. Cluster 2, associated with Pb, Cd and Cu and rich in Fe, exhibited possibly more geogenic influence and well-drained soil conditions (54, 55).

Cluster 3 (C3) contained the highest number of samples, which were sample numbers 9, 10, 21, 26, 27, 28, 29, 31, 32, 33, 34, 38, 43, 44 and 48 and represented the most contaminated or impacted group-high Cu, Pb suggesting multiple anthropogenic sources (30, 33).

Cluster 4 (C4) was the lowest polluted group and contained the lowest number of samples, including 12, 19, 20, 22, 41, 42, 45, 47 and 49, which were possibly depleted in Cu and Zn and could reflect less disturbed soils or specific pedogenic factors (e.g., leaching) (5, 33)

Subsurface soils

The dendrogram and cluster profile plot distinguished subsurface soils into two clusters based on PCA scores (Fig. 9a, b). Cluster 1, high in F1 (PC1), F3 (PC3), F4 (PC4) and F5 (PC5) and low in F2 (PC2) represented metal-enriched subsurface soils, particularly in Cr, Cd, As, Cu and Pb, implying anthropogenic impact with complex interactions (31, 50).

Cluster 2, low in F1, F3, F4 and F5 and positive in F2, reflected more geo-genic influence and was dominated by natural Fe-Mn-Zn concentrations (15, 21). This cluster represented less contaminated subsurface profiles predominantly shaped by weathering and minimal anthropogenic disturbances (53, 55).

Hierarchical clustering of the metals

Agglomerative hierarchical clustering (CA) was performed on the heavy metals along with Al in the soil samples, where proximity was determined by measuring the squared Euclidean distance and agglomeration by Ward's method. This ensured robust cluster formation by prioritizing intra-cluster homogeneity and standardization (centring and reduction) and it mitigated biases from scale differences between metals (27, 30). These clustering levels indicated the relative similarity of spatial patterns for each metal across the fifty surface and subsurface soil samples. Cluster analysis conducted on the metals revealed that the elements that were grouped in one branch can have similar geochemical behaviour (11, 30).

Surface soils

The dendrogram in surface soil samples clustered Al and eight heavy metals (Cd, As, Zn, Cu, Mn, Fe, Pb and Cr) into two major branches, based on their similarity in spatial distribution across surface soil samples (Fig. 10).

The initial clusters were formed when Cd merged with Al and As joined shortly after at a lower dissimilarity (≈ 80–90). Group 1, containing Cd and As, is indicative of input from phosphate fertilizers, especially those derived from sedimentary PR, which carry trace

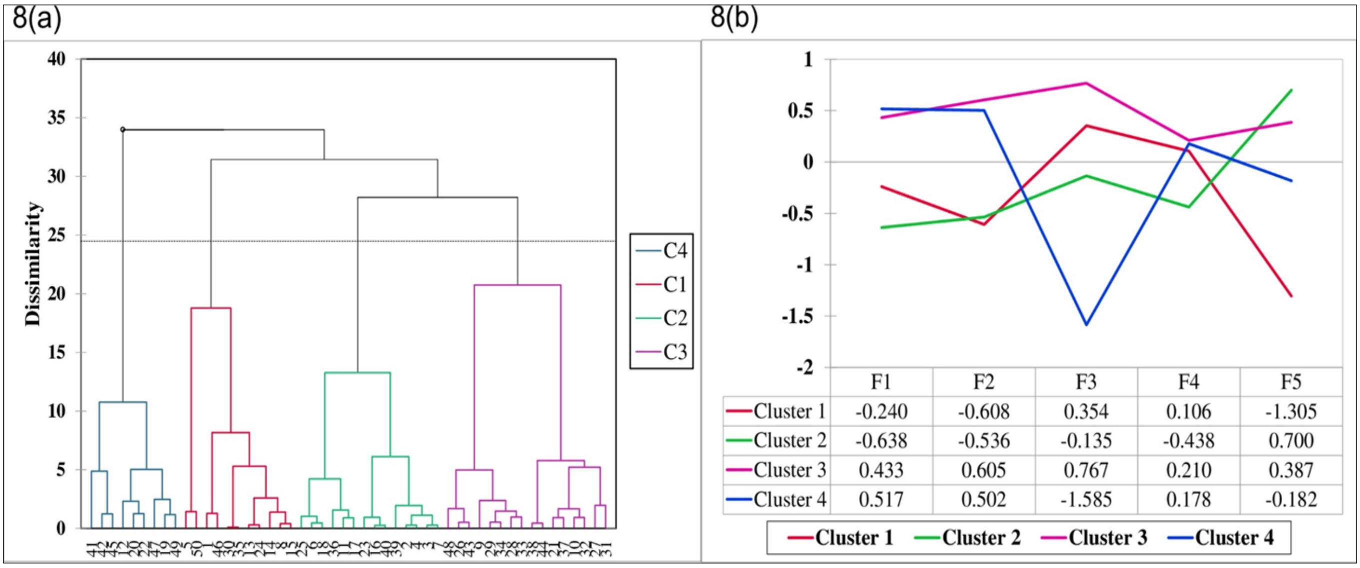

Fig. 8. Agglomerative hierarchical cluster analysis of Surface soil samples from tomato-growing soils of Chintamani, Karnataka, India: (a) the dendrogram identifying four clusters and (b) the corresponding profile plot derived from factor scores.

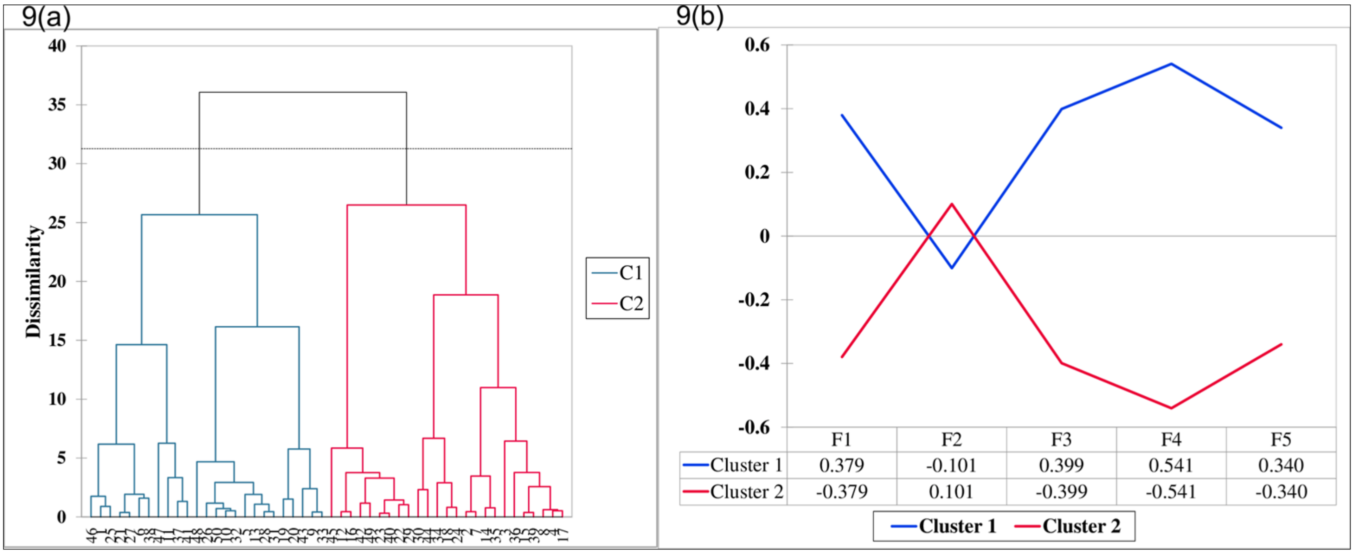

Fig. 9. Agglomerative hierarchical cluster analysis of sub-surface soil samples from tomato-growing soils of Chintamani, Karnataka, India: (a) the dendrogram identifying two clusters and (b) the corresponding factor-score profile plot.

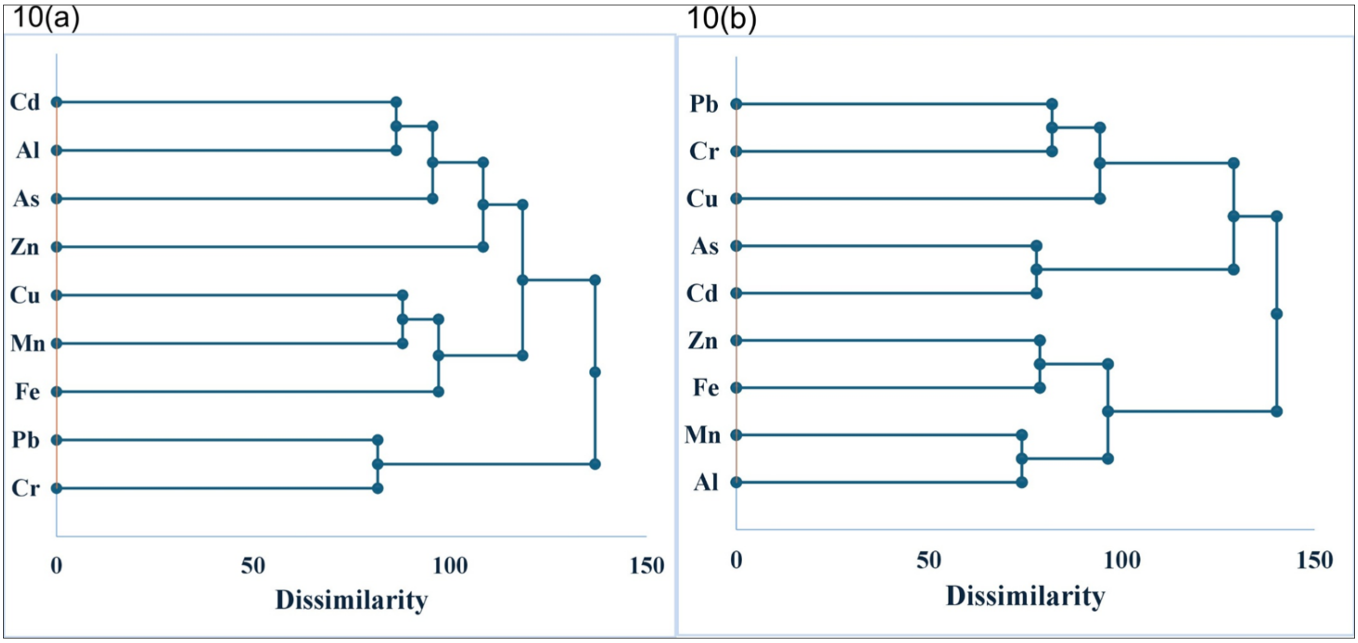

Fig. 10. Dendrograms of agglomerative hierarchical cluster analysis (Ward's method) for heavy metals and Al in tomato-growing soils of Chintamani, Karnataka, India. (a) Surface soils (0-20 cm) and (b) Subsurface soils (20-40 cm).

impurities (45, 46). Although Al present in this group is primarily geogenic, its association here may have resulted from soil acidity (common in tropical alfisols), which mobilized Al and facilitated its interaction with Cd and As (5, 43).

In group 2, Mn and Cu formed a pair, with Zn joining them at a slightly higher dissimilarity (~100). The co-existence of Zn, Mn and Cu in this group reflects their role as essential micronutrients and may have been added through commercial formulations (8, 11). Cu-based fungicides (e.g., copper oxychloride), Zn-enriched NPKs and Mn $(SO_4)_2$ were standard inputs in tomato cultivation (37). Their co-clustering suggests shared application practices across the fields.

Group 3 represented mixed lithogenic and anthropogenic sources having Fe intrinsic to the parent material of Chintamani soils (15, 21) and Pb, in contrast, may have entered via lead arsenate pesticides, atmospheric deposition (e.g., traffic emissions from nearby roads), contaminated organic manures, or municipal compost (36, 44, 50). Cr merged with the rest only at the final step (node 17, dissimilarity ≈137), confirming its most distinct spatial distribution. Consequently, the late clustering of Cr implies its distinct geochemical behaviour and less frequent external input (39, 40).

Subsurface soils

Similar to the surface dendrogram, the dendrogram of subsurface soils splits the nine metals into two main branches (Fig. 10b). At the lowest dissimilarities, Al and Mn formed the first tight pair (merged at dissimilarity ≈74), while Fe and Zn formed another pair (merged at ≈78). The first cluster (Al, Mn, Fe and Zn) likely represented natural soil background and nutrient inputs (30, 37). Al and Fe were primarily lithogenic, while Mn and Zn, being essential micronutrients, were naturally present but often supplemented through fertilizers (15, 51). Long-term use of phosphate and micronutrient fertilizers may have elevated Mn, Zn and Cu levels (42, 46).

Separately at a similar level (~77-79), Cd clustered with As. This produced three small clusters at low dissimilarity (Al, Mn), (Fe, Zn) and (Cd, As). In the mid-range (dissimilarity ~82), Cr joined Pb (forming a Cr-Pb pair). By dissimilarity ≈94, Cu merged the Cr-Pb pair, yielding a Cu-Cr-Pb cluster. The second cluster (Cd, As, Cu, Cr and Pb) implied anthropogenic contamination. Cu-based amendments and herbicides may have contributed to Pb and other metals with Mn, Zn and Pb being major agrochemical inputs (56). In our findings, Cu grouped with Cr and Pb, indicating potential co-occurrence from both anthropogenic and lithogenic sources.

Finally, at high dissimilarity (≈129–140), the two groups merged the (Al, Mn, Fe and Zn) cluster merged with the (Cd, As, Cu, Cr and Pb) cluster to form the full set. Al and Mn were most similar (lowest dissimilarity), implying they co-vary strongly, followed by Fe-Zn and Cd-As, but these three pairs remained distinct from each other at lower dissimilarity. In contrast, Cr and Pb (with Cu) clustered only at higher dissimilarity, indicating a different distribution pattern than the Al-Mn-Fe-Zn group. Overall, clustering revealed two broad groups: a geochemically dominated group (Al, Mn, Fe and Zn) and an anthropogenically influenced group (Cd, As, Cu, Cr and Pb) (30, 52).

Largely, PCA and CA validated and enhanced the interpretations from the pollution indices. Both the methods clearly distinguished between metals with geochemical behaviour (Al, Fe, Mn and Zn) and those predominantly influenced by anthropogenic inputs (Cd, As, Pb, Cu and Cr), except for Mn, which showed significant concern in our soils according to EF and PI values. Long-term agrochemical application and movement through the soil profile created complex interactions, as evidenced by the spatial distribution and covariation of these metals, particularly the strong connections of Cd-As and Cr-Pb-Cu in subsurface soils. The accuracy of the source attribution was confirmed by strong correlations between pollution indices and multivariate approaches.

Conclusion

The study provided a comprehensive assessment of heavy metal contamination in tomato-cultivated soils of Chintamani, Karnataka. The variations observed in EF, I_{geo}, PI, E_i and RI values across metals and soil depths indicated heterogeneous metal distribution with considerable spatial and depth-wise variability, strongly influenced by anthropogenic inputs. The results of I_{geo} were quite different from the results of other pollution indices. Likewise, the PI assessment taking Indian national background concentration metals, especially for As, gave varied results from taking global soil concentration of metals, emphasising the importance of selecting appropriate reference values. Overall, As, Pb and Cd were identified as priority pollutants in our soils, whereas Zn and Cr were the least concerning heavy metals. The adopted multivariate analyses (PCA and CA) effectively delineated sources: geogenic (Al, Fe, Mn, Zn and Cr) and anthropogenic (Cd, As, Pb and Cu). The risk of downward migration and long-term persistence was highlighted by elevated levels of Cd, As, Pb and Cu in subsurface clusters and emphasize the need for depth-specific mitigation, establishment of regional background values and future research on metal speciation, bioavailability and isotopic tracking to improve ecological risk assessment and sustainable soil management. The identification of key pollutants in the study area underscores the need for safer fertiliser use, routine soil testing and better nutrient management, all of which are directly relevant to the farming community of Chintamani. The findings also provide evidence-based recommendations for local authorities to improve soil health monitoring and fertiliser quality regulation.

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Authors' contributions

MR Conceptualized and carried out the investigation, formal analysis, prepared all figures and tables and wrote the original manuscript, reviewed and edited. KMR, DVN, VP and PK guided in method and data curation, validation, writing, reviewing and editing. All authors read and approved the final manuscript.

Compliance with ethical standards

Conflict of interest: The authors declare that the research was conducted in the absence of any potential conflict of interest.

Ethical issues: None

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