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A meta-analysis and experimental survey of heavy metals pollution in agricultural soils

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ABSTRACT

Heavy metal (HM) pollution in agricultural soils represents a hidden danger to food security worldwide. In this paper, the spatio-temporal trends of heavy metals from eight countries and 50 soil samples from agricultural farmland were evaluated through a combination of field surveys and meta-analysis to provide a comprehensive report on heavy metal pollution. The soil samples were analysed using inductively coupled plasma–mass spectrometry (ICP-MS) (Perkin Elmer Nixon 300Q). The contamination factor (CF) and pollution load index (PLI), and diagnostic tests on the extracted data were calculated. The results of the CF in the soils indicate extreme contamination for Cr, suggesting ecotoxicological effects, while the PLI values range from baseline to moderate pollution for Cd, Hg, Cu, Zn, and Ni, except for Cr, which shows very high pollution, suggesting that the soils have undergone some form of deterioration. The meta-analysis results of the 50 reviewed articles published between 2010 and 2023 showed increasing trends for all the HMs. The weighted mean values of Cd, Cr, Hg, Cu, Zn, As, and Ni were in the range of 0.0-222.7, 0.08-289.2, 0.03-193, 2.94-198.1, 0.0-771.1, 0.0-231, and 1.71-99.75.6 mg/kg, respectively. The mean values of Cd, Hg, Zn and As exceeded two to three times the values of China National Environmental Monitoring Centre (CNEMC) European Union's most cited guideline (MEF), and the rock crust guideline. The results of the correlation matrix heatmap revealed a highly positive correlation between Cr and Ni ($R^2 = 0.66$), suggesting that these elements have the same source and are likely prevalent in agricultural soils. The spatial origin of the publications reveals that 82% of the studies were from China followed by South Africa and Italy accounting for 4% respectively while Nigeria, Egypt, Morocco, Iran, and Turkey account for 2% each. The findings of this study have important implications for environmental regulation on agricultural food protection from heavy metal pollution. Unlike previous meta-analysis studies which often adopt a "silos" method, this study highlights a nexus approach that integrates both meta-analysis and experimental studies which could establish a more comprehensive understanding of heavy metal pollution in agricultural soils.

1. Introduction

Agricultural soils play a vital role in supporting human society through food production and ecosystem functioning. However, this essential natural resource is rapidly being degraded globally at an unprecedented rate due to heavy metal contamination [1,2]. The United Nations-Sustainable Development Goals (UN-SDGs) mandate poverty elimination and improved standards of living, which can be achieved through agricultural intensification and expansion [3,4]. It is estimated that approximately 20 million hectares of land are contaminated annually [2], while current projections suggest an increase of 70 to 110% for all related agricultural products and consumption, which inevitably increases the rate of crop contamination [5–7].

The core role of agricultural soils is to produce crops and safeguard the quality of cultivated crops to acceptable standards while minimising soil pollution. However, reports from most scientific scholars attest that heavy metal contamination in agricultural farmland poses a significant threat to global food security due to its irreversibility in the environment, bioaccumulation in nature, and inhibitory effects on multiple physiological processes in plants [2,8,9]. Consequently, efforts aimed at

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expanding agricultural food production through land conversion will subsequently be thwarted by elevated heavy metal concentrations in the soils, which are also transferred into the crops through the food chain or the food web, with deleterious consequences on human health [7,10,11]. Therefore, heavy metals pollution in agricultural soils should be prioritised as a primary concern, as their toxicity poses significant nutritional, ecological and environmental risks [8,12]. Due to their toxicity, bioaccumulation, and low degradability, the United States Environmental Protection Agency (USEPA) considers certain metals such as Cd, Cr, As, Hg, Pb, Cu, Zn, and Ni, among others, as high priority contaminants [13].

This is also true, as polluted agricultural soils directly and indirectly inhibit eight of the seventeen goals set by the UN-SDGs to be reached by 2030. Fig. 1 illustrates the impact of heavy metal pollution in agricultural soils on sustainable development goals. The continuous accumulation of heavy metals in agricultural soils creates an inverse relationship between agricultural expansion and the quality of available cultivated crops, thus raising concerns regarding the United Nations Sustainable Development Goals (UN-SDGs) aimed at reducing hunger, which is inevitably linked to poverty (UN-SDG #1). Contaminated surface and groundwater with heavy metals, resulting from the use of chemical fertilisers and pesticides for crop irrigation, negatively affects clean water and sanitation (UN-SDG #6). The long-term accumulation of heavy metals in cultivated soils will disrupt the soil-carbon cycle and have repercussions on climate action (UN-SDG #13), leading to nutrient loss, erratic rainfall, and deforestation [14]. Additionally, elevated concentrations of heavy metals will result in derelict urban agricultural land and inequitable exposure to heavy metals, hindering the ability of cities and communities (UN-SDG #11) to achieve environmentally friendly sustainable development and livelihoods. Furthermore, heavy metal pollution reduces the ability of crops to grow properly, leading to crop shortages and contributing negatively to hunger reduction, affecting the zero-hunger campaign (UN-SDG #2) and increasing the likelihood of poverty among individual countries (UN-SDG 1). This will inevitably affect people's good health and well-being (UN-SDG 3) through exposure to heavy metals via dermal contact, inhalation, and ingestion, causing various health-related illnesses such as impaired cognitive development, lung cancer, and kidney failure, among others [15,16]. Consequently, life on land (UN-SDG 15) will also be affected, leading to ecotoxicological effects on marine and vegetal organisms and preventing progress toward ensuring sustainable production and consumption patterns (UN-SDG 12). Soil pollution negatively impacts sustainability, specifically hindering progress on several of the Sustainable Development Goals (SDGs) set out by the United Nations.

The number of scientific reports published worldwide documenting heavy metal concentrations in various agricultural soils also highlights the need for more in-depth analysis [17,18]. Different advanced heavy metal soil analysis techniques such as flame atomic absorption spectroscopy (FAAS) and picosecond laser-induced breakdown spectroscopy (Ps-LIBS) [19,20], atomic absorption spectrometry (AAS), inductively

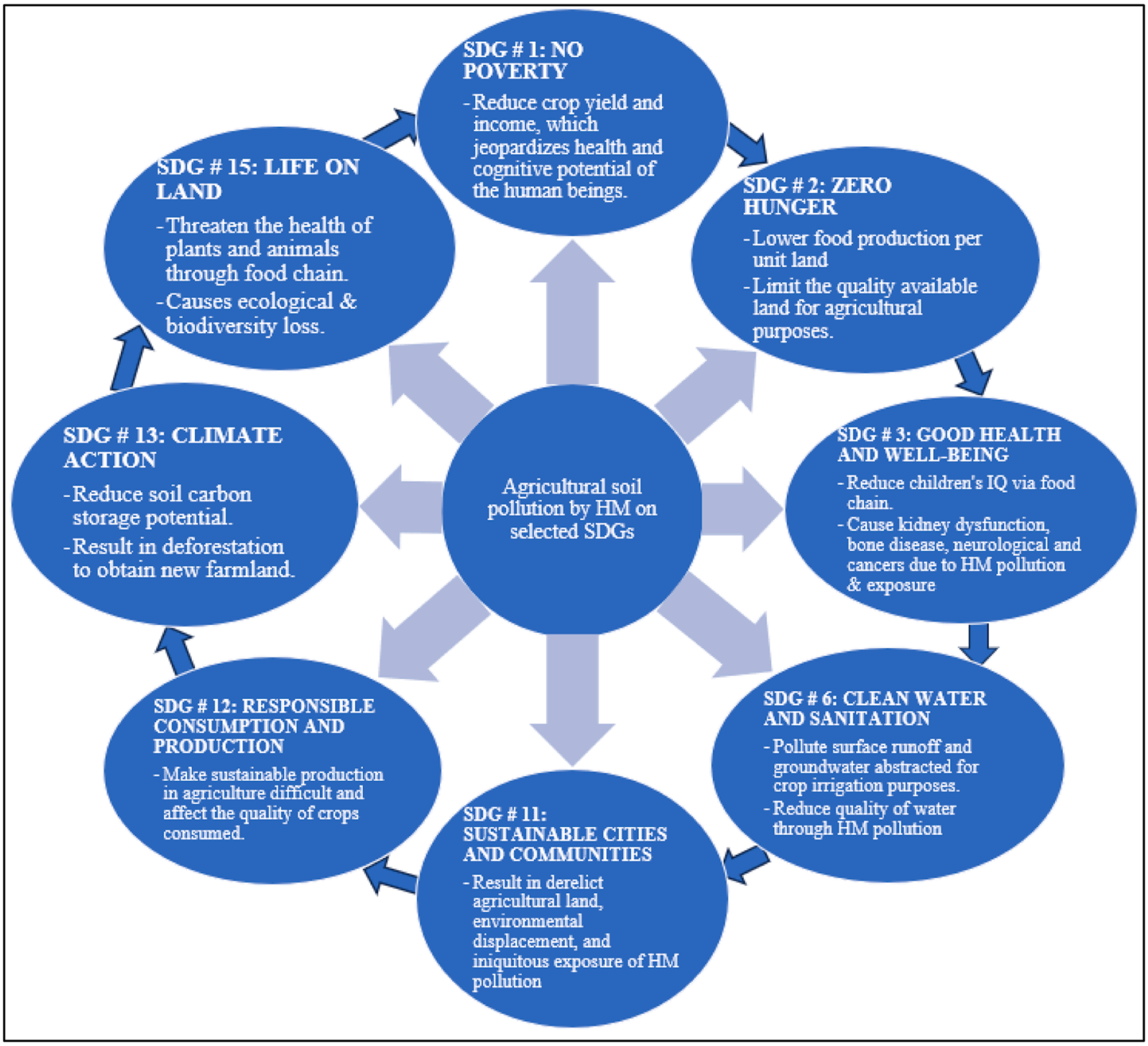

Fig. 1. Impact of agricultural soil pollution by heavy metals on SDGs. Source: Author, 2024.

coupled plasma optical emission spectrometry (ICP-OES) [21,22], and laser ablation inductively coupled plasma mass spectrometry (LA-ICP-MS) [23] have been widely used as measuring instruments due to their accuracy. Similarly, models such as geostatistical model, carcinogenic dynamic risks model, high-precision X-ray fluorescence (HDXRF), isotopic ratio analysis, positive matrix factorisation (PMF), advanced multicriterion decision analysis and the analytical hierarchy process (MCDA-AHP), multivariate statistical models incorporating principal component analysis (PCA), Monte Carlo Simulation (MCS) and potential toxic risk area identification method based on sediment source aggregation class (SLISA-SCA) have extensively been used for heavy metals analytical studies [10,24–28]. Recently, a new wave of research based on existing literature has been proposed as an approved scientific method to comprehensively understand heavy metal pollution in agricultural soils [29–31]. This method is based on meta-analysis techniques, which synthesise information extracted from primary studies for increased inference power [32]. The extracted data from the original research are synthesised to obtain an overall trend for quantitative and comprehensive conclusions [33].

While methods like meta-analysis might be good in providing a quantitative assessment of heavy metals, Huang, et al. [32] suggest that the results of such analyses can sometimes be misleading due to different reporting data sources and limited spatial scope (state or country) [34]. Consequently, these studies often provide insights from a “silos” perspective, failing to fully illuminate the situation and characteristics of heavy metal pollution. This limitation arises because heavy metal contamination in agricultural soils varies dynamically across locations, influenced by various factors such as source types, environmental conditions, and reporting units [35,36]. Therefore, bridging this gap requires adopting a nexus approach that integrates both meta-analysis and experimental studies, exploring the relationships in between to establish a more comprehensive understanding of heavy metal pollution in agricultural soils. This approach would provide a reliable database to mitigate heavy metals contamination, thereby justifying the importance of this study. The objectives of this study are: (1) to evaluate the level of heavy metal pollution in agricultural soils using various pollution indices, (2) to analyse the spatio-temporal trend of heavy metals pollution in agricultural soils from 2010 to 2023, and (3) to identify the reasons for the variation through a combination of experimental analysis and meta-analysis techniques.

2. Materials and methods

2.1. Study area, sampling, and analysis of primary investigation

The sampling area for the experimental survey is located within the Sub-Saharan Africa region, particularly in the extreme north region of Cameroon (Fig. 2A & B). The area spans approximately 4 hectares. The choice of the study site was motivated by two main factors: Firstly, there has not been any reported study on the state of heavy metal pollution in agricultural soil from Cameroon, making this study the first of its kind. Secondly, if food safety measures must be preserved, there is need to accurately monitor the state of agricultural soils to determine if they are safe for crop cultivation. The climatic condition of the area is threatened by extreme surface water shortages due to extended dry spell period, and cultivation rely on boreholes for irrigation purposes. The length of the wet season in most cases lasts for three months (May-July) characterised by heavy thunderstorms at the peak of the raining period while the remaining months are dry [36]. The average temperature of the area ranges between 18 to 45°C and the land use in the area is highly intensive agriculture [36].

One of the basic requirements for the layout of the sampling points is that the sampling site should be representative and homogeneous to reflect the pollution status of heavy metals in the area [1]. The following procedure proposed by Mao, et al. [1], Nde, et al. [10], Chen, et al. [37], was modified and adopted for the sampling points. The soil samples

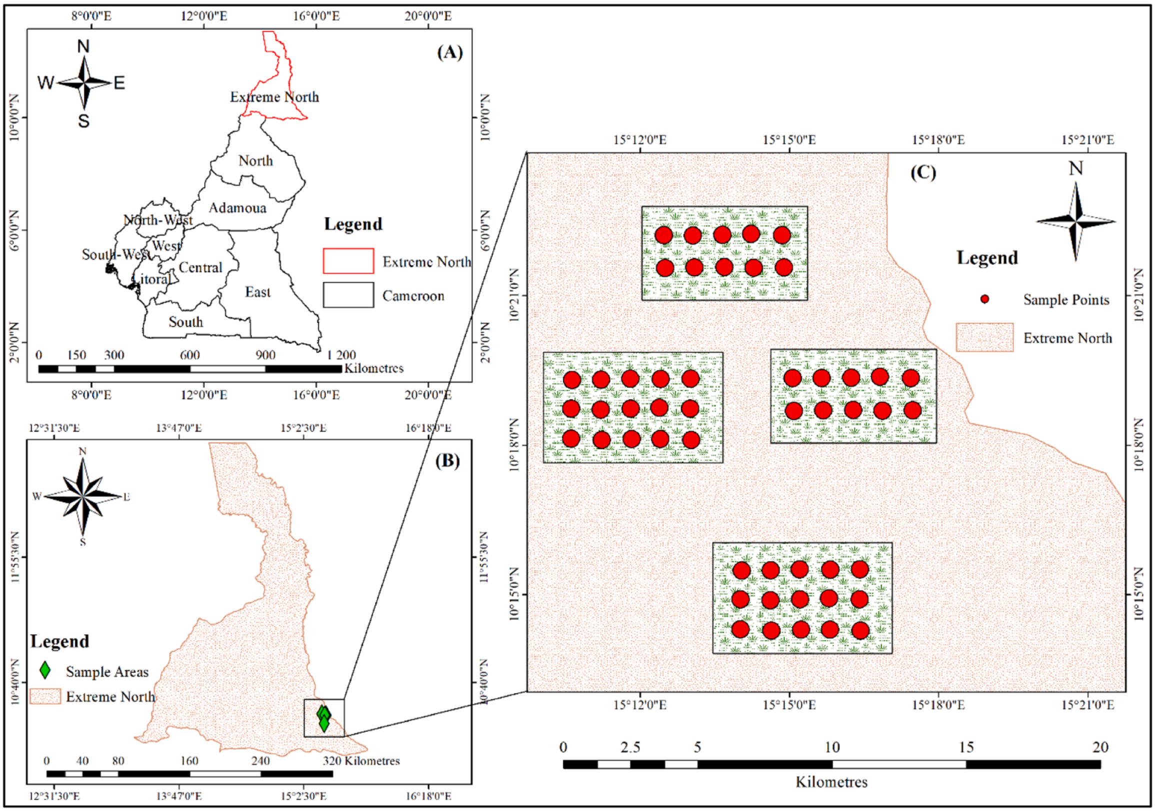

Fig. 2. Map of the study area (A) shows the map of Cameroon, (B) shows the Extreme North Province and (C) shows the sampling areas and sampling points from the agricultural fields.

were collected from different areas according to a grid layout method. The paddy fields in each sampling area were divided into 25 m × 25 m unit grids to represent the soils of the entire study area and their spatial distribution. A systematic sampling pattern with regular spacing intervals (5 m) between each sampling point was then used to cover a wide range within each sampling area (Fig. 2C). A total of 50 surface soil samples from a depth of 0-20 cm, approximately 500g in weight, were collected. The collected samples were pooled together to form composite samples for homogeneity.

In the laboratory, the soil samples were air-dried in an oven at a temperature $< 60^\circ$C, gently disaggregated using ceramic mortar and pestle, and passed through a 2 mm sieve as prescribed by [38]. Subsequently, the soil samples were analysed using Inductively Coupled Plasma-Mass Spectrometry (ICP-MS) (Perkin Elmer Nixon 300Q). The samples were digested using a Microwave Multiwave 3000, Anton Paar, Aqua-regia digestion on a 1 g aliquot of the sample using an acid mixture of 70% hypochlorite acid, 69% nitric acid, and 30% hydrogen peroxide as prescribed by Nde, et al. [10]. The instrument was calibrated by using a calibration solution as the Atomic Spectrometric standard, whose specifications are that: In the Total Quantitative method, the mass calibration stability is measured using a 10 mg/L multi-element standards solution Al, Ba, Ce, Co, Cu, In, Li, Mg, Mn, Ni, Pb, Tb, U, and Zn. For every measurement, the instrument is set to run a blank and a standard check at every ten samples for quality control (Nde et al., 2021). The detection limit (mg/l (ppb)) of the selected metals; Ni ($<$ 0.5), Fe ($<$ 1.5), Cu (0.5) and As ($<$ 0.25) and were then converted to mg/kg.

2.2. Contamination indices

In order to assess the magnitude of pollution levels in the cultivated soils, different pollution indices such as contamination factor (CF) pollution load index (PLI), which are widely used [39,40]. The standard background [41] was used as the reference materials to calculate the CF as described in other studies [40,42] (Eq. (1)).

$$CF = \frac{C_s}{C_{ref}} \tag{1}$$

where, C_s and C_{ref} is the total metal content in the soil (mg/kg) and the latter is the reference content in the pristine soil. The interpreted classes used were; $CF < 1$, indicate pristine environment, $1 \leq CF < 3$, indicate moderate contamination, $3 \leq CF < 6$, indicate high contamination and $CF \geq 6$ denote very high contamination [43].

Also, the PLI (unitless) was computed according to Eq. (2) to assess the overall pollution and based on the following classes ($PLI > 2$ denotes significant deterioration, $0 < PLI < 1$ denotes baseline level pollution), where CF_1, CF_2, CF_3, are CF of elements 1,2,3 …., n; [44].

$$PLI = \sqrt[n]{CF_1 \times CF_2 \times CF_3 \ldots. \times CF_n} \tag{2}$$

2.3. Quality control and quality assurance of the experiment procedure

This study has established a sound quality control/quality assurance over a similar study and is referenced therein [10].

2.4. Auxiliary variables for the meta-analysis

2.4.1. Literature selection and data extraction

A comprehensive literature approach analysis was carried out on primary research papers (only in English) published between January 2010 and December 2023, reporting the content of heavy metals in agricultural soils. The use of previously reported studies allowed researchers the opportunity to gather past results to ascertain the current status quo of heavy metal concentrations in agricultural soils, their distribution, and to inform their point of view [45]. The bibliography search was conducted according to the procedures prescribed by Huang, et al. [31], exploring the database of "Scopus" using the keywords "heavy metal" and "agricultural soil" or "farmland" or "cultivated land". The following inclusion criteria were adopted for each assessed research publication: (i) it must be a field study carried out on an agricultural farmland or soil, (ii) the farming operation must state one of the different cropping activities being carried out (vegetables, grains, etc.), (iii) the number of soil samples collected should be greater than or equal to twenty (sample size $\geq$ 20), (iv) the laboratory procedures of the sample preparation and analytical tools used should be reported and be within the acceptable recommended standards, (v) the mean values/range of the elements contents/standard deviation/variance/coefficient of variation should be reported, (vi) the study area or its geographical coordinates should be stated [32]. Of the 289 reviewed literature, only 50 literatures met all the inclusion criteria, and the extracted data are presented in supplementary material.

2.4.2. Diagnostics tests and calculation of the weighted value

Before calculating the weighted mean, a standard diagnostic test was used to address issues that might arise due to publication bias and sensitivity analysis, as well as to account for the existence of extreme values reported in other studies that might affect the overall results to some extent due to heterogeneity in distribution. These extreme values could be outliers or higher values, which may distort the weighted mean values. Similar concerns have been recognised and addressed in similar meta-analysis studies [32,33]. In view of this, the weight (w_i, mg/kg) was calculated according to Equation 3.

$$w_i = A_i \times \frac{N_i}{SD_i} \tag{3}$$

where, A_i is the size of the study area, $N_i$is the number of soil samples, and SD_i is the calculated standard deviation of the heavy metals reported in each study. Therefore, the weighted mean ($\mathbb{C}$, mg/kg) was calculated as given in Equation 4.

$$\mathbb{C} = \mathbb{C}_i \times w_i \Big/ \sum_{i=1}^{n} w_i \tag{4}$$

where, c_i is the calculated mean contents of the heavy metals reported in each study.

During the pre-processing phase, a standard diagnostic test was also carried out to address the concerns stated above. Logarithmic conversion was used to address the problem of skewness that might arise, which could lead to overreliance on several studies with extreme higher values, and to bring the weights closer to a normal distribution as prescribed by Huang, et al. [18]. Thus, the logarithmic transformation weights (w_i^*) was solved by using Equation 5 and the final weighted mean content $\mathbb{C}$ was recalculated as given in Equation 6 [18].

$$w_i = \mathrm{I}_g\left(A_i \times \frac{N_i}{SD_i}\right) \tag{5}$$

$$\mathbb{C} = \mathbb{C}_i \times w_i^* \Big/ \sum_{i=1}^{n} w_i^* \tag{6}$$

where, A_i, N_i, SD_i and $\mathbb{C}_i$have the same meaning as formulas (4) and (5).

2.5. Data analysis

To obtain a more vivid information of the general distribution of the heavy metals from the reported studies, we adopted the procedure similar to Yang, et al. [34]. Descriptive statistics were computed in Microsoft Excel (version 2010) and related figures were drawn. The distribution of the heavy metal content and the correlation matrix heatmap (CMHM) were performed in a manner similar to Yang, et al.

[34], Han, et al. [46], Dutta, et al. [47]. Additionally, a hierarchical clustering heatmap (HCHM) was developed to offer further insights beyond those provided by the correlation heatmap. The data distribution histograms, CMHM, and HCHM were generated using the Pandas, Seaborn, and the pyplot modules of the Matplotlib libraries of Python programming. The HCHM employed a hierarchical clustering algorithm to perform agglomerative clustering on the heavy metal's dataset, displaying a dendrogram on either side of the CHM. The dendrogram depicts the hierarchical relationships between the heavy metals based on their similarities. The length of the branches in the dendrogram indicates the degree of dissimilarity between clusters of heavy metals, with shorter lengths indicating higher similarity. These clusters represent groups of heavy metals that exhibit similar occurrence patterns across the various soils examined in the studies. While the numbers and colour intensity in the CHM represent the degree of correlation or similarity between pairs of heavy metals, the dendrograms in the HCHM reveal clusters of heavy metals that occur in similar manners.

3. Results and discussion

3.1. Concentrations of heavy metals in agricultural soils

The metal contents (mg/kg) of the measured heavy metals in the agricultural soils are presented in Table 1A. The mean contents of the metals were in order of Cr > Zn > Ni > Cu > As > Cd > Hg. Comparison with previously published studies in agricultural soils reveals that Cd, Cr, Cu, Zn, and As in this study were found to be lower than those reported in other studies (Table 1A). For instance, the metal contents level of Cr was (262.45±1.94 mg/kg), (100.6±23.97 mg/kg) and 102.30± 50.87 mg/kg for South Africa, India and Burkina Faso respectively, were four to eight-fold higher than the current study. Similarly, the same was observed for Cu and As with concentrations exceedingly higher than those in this study. However, the values of Cd (0.23 ± 0.0 mg/kg), Cr (5.08 ± 9.39 mg/kg), Hg (0.26 ± 1.00 mg/kg), Zn (34.71 ± 30.98 mg/kg) and As (4.57 ± 1.64 mg/kg) from Kenya were slightly above the current study (Table 1B).

Although the results of this study suggest that all the heavy metals were generally low compared to other reported studies, an in-depth analysis of the contamination indices proved the contrary (Table 1B). The results of the contamination factor (CF) reveal that Cd, Hg, Cu, Zn, Ni, and As contamination factors were < 1, signifying a pristine environment. However, the value of Cr was > 6, indicating very high contamination. The results of the pollution load index (PLI) indicate baseline to moderate pollution for Cd, Cu, Zn, Ni, and As, except for Cr, with very high pollution suggesting ecotoxicological effects on vegetal organisms [10] (Table 1B). Similar to these findings, Mamat, et al. [48] reported moderate to high pollution levels in farmland soils, with maximum values emanating from regions with rapidly developing economies containing industries and areas with busy traffic, from a study of 146 cities in China. The implication of the baseline to moderate pollution is that it might progressively cause high pollution in the future [10]. Additionally, these findings underscore the significance of anthropogenic activities which might have contaminated the soils through the use of fertilizers, pesticides, and other forms of agrochemicals [49].

3.2. Publication bias and sensitivity analysis of the meta-analysis

As stated previously, there was publication bias in this study that could arise due to unusual studies reporting extreme values that might affect the results to some extent. The results shown in Fig. 4 indicate that the heavy metal content reported indicates extreme values. Similar observations have been reported by Yuan, et al. [33], and the existence of such extreme values in the published literature might be from results obtained from "hot spots" or contaminated areas. The standard delate residual to outlier was performed to exclude the outliers before further analysis, as shown in Fig. 3.

3.3. The spatio-temporal variation of heavy metals pollution agriculture soils

The results of the spatio-temporal variation of heavy metals pollution from the published literature spanning from 2010 - 2023 are presented in Fig. 4. Individual metal pollution for Cd from the published survey (> 44) showed mean contamination of approximately less than 5 mg/kg, while few studies (≈ < 3) reported mean concentrations > 250 mg/kg. It was observed that most of the published surveys for Hg reported mean concentrations of ≈ < 1 mg/kg, whereas very few studies (< 2) reported mean values of ≈ 170 mg/kg. Cr and Zn results show similar patterns, as a greater number of studies' mean concentrations were ≈ > 250 mg/kg, and very few studies (≈ < 2) reported concentrations ranging from 270 mg/kg for Cr and 740-780 mg/kg for Zn.

Similarly, for As and Cu, the concentrations from the analysis reveal that a greater number of studies reported mean concentrations of approximately 60 mg/kg for As and 50 mg/kg for Cu, while very few studies (≈ < 2) reported concentrations of ≈ > 200 mg/kg. Most of the published literature reported mean concentrations of approximately 50 mg/kg of Ni, while few publications (≈ >2) reported concentrations ranging from 80 to 100 mg/kg. These findings are consistent with Meng, et al. [52], who found that the temporal variations of heavy metal pollution in cultivated soils in China could be due to atmospheric distribution by climatic variables triggered by the emergence of China's industrial activities and accelerated economic growth [53]. According to Yuan, et al. [33], Cd is the most heavily polluted heavy metal on the soil of China, with 33.54% and 44.65% of the pollution on farmland. This statement also corroborates the increasing trend of Cd concentration as shown in Fig. 4. If stricter regulation of Cd pollution is not implemented in the near future, the food safety of agricultural soils would be threatened.

Figures 5 and 6 provide the correlation analysis of the heavy metals

Table 1

(A) Concentration of heavy metals in agricultural soils and in other reported studies.

Country		Average heavy metals content in soils (mg/kg)						Sampling size	References
	Cd	Cr	Hg	Cu	Zn	Ni	As		
Cameroon	0.02±00	25.46±2.49	001±00	7.52±0.19	22.35±0.97	14.52±14.42	0.80±0.01	50	This study
South Africa	0.05±0.0	262.45±1.94	VNR	20.75±0.11	22.24±0.01	83.26±0.60	1.01 ± 0.05	64	Nde, et al. [10]
India	VNR	100.6±23.97	VNR	31.2±16.23	92.4±25.05	13.7±9.87	3.5±0.66	15	Adimalla, et al. [39]
Burkina Faso	VNR	102.30±50.87	3.83±1.45	57.13±30.43	58.51±19.15	38.87±12.02	49.67±62.88	12	Sawadogo, et al. [50]
Kenya	0.23±0.09	5.08 ±9.39	0.26±1.00	5.05 ± 13.42	34.71± 0.98	2.35±3.32	4.57 ± 1.64	72	Mungai, et al. [51]

(B)

Contamination indices							
CF	0.18	**8.49**	0.00	0.53	0.43	0.40	0.78
PLI	1.12	7.71	0.04	1.92	1.74	1.67	2.34

*VNR = value not reported.

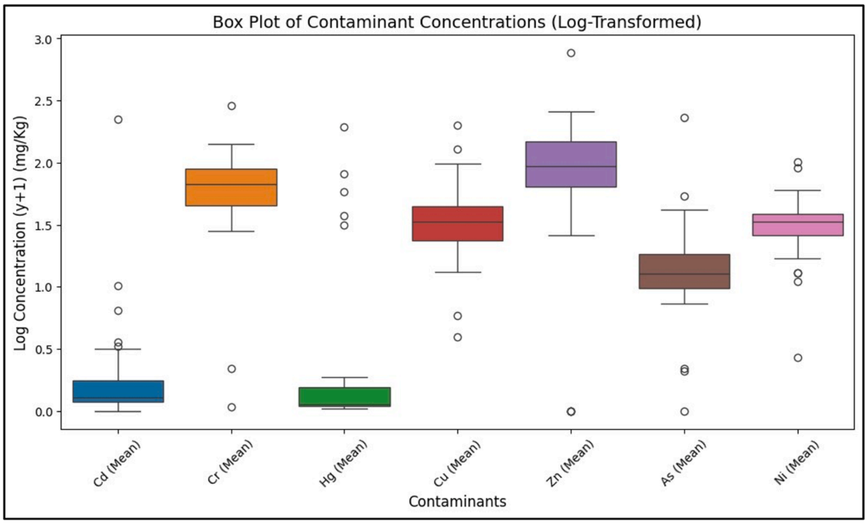

Fig. 3. Boxplot of heavy metals concentrations with extreme values.

from the different studies in pairs and as a cluster (more than 2). The results reveal a highly positive correlation between Cr and Ni (R^2 = 0.66) (Fig. 5), suggesting that these elements have the same source and are likely prevalent in agricultural soils. Shao, et al. [54] found a similar correlation between Cu, Zn, and Ni in agricultural soils in China due to the intense application of chemical additives in farming operations against the antimicrobial effect. On the other hand, the highest negative correlation was observed with Hg and Cr, suggesting that these two contaminants are very unlikely to exist together in the same soil or their anthropogenic sources could be different [55]. These results are further confirmed by the length of the dendrograms on these metals in the HCHM (Fig. 6). Additionally, this study observed that Cu, Zn, and As have the most similar occurrence (shorter cluster lengths) compared to other metal clusters, while clusters of Cd and Hg clusters are highly dissimilar to the rest of the heavy metals. The highest pair of contaminants are Cd and Hg with 0.94 followed by Ni and Cr (0.66) while the lowest pair of correlating pair is Hg and Cr (-0.40). Cd and Hg are highly dissimilar from the occurrence of the other heavy metals in the study, while Cu, Zn, and As are the closest and similarly occurring groups.

The spatial origin of the publications reveals that 82% of the studies were from China followed by South Africa and Italy, accounting for 4% respectively, while Nigeria, Egypt, Morocco, Iran and Turkey account for 2 % each (Fig. 7). The high percentage of published reports from China is mainly attributed to its economic expansion in the past 10 decades, due to intense mining activities and the high application of agrochemical pesticides [33,34]. Anthropogenic sources of heavy metal contamination in agricultural soils remain the most widely cited, such as coal mine deposition near cultivated sites [56,57], mining and industrial deposition, including smelting plants [58], roadside deposition emanating from vehicle exhaust in urban and semi-urban areas in close proximity to agricultural sites [18,59,60], chemical sprays such as insecticides, pesticides, and untreated irrigation water [61,62]. For example, in China, atmospheric deposition from coal mining is responsible for 43-85% of heavy metal inputs into agricultural soils.

3.4. The weighted mean contents of heavy metals in agricultural soils

The results of the weighted mean values of the different studies are presented in Table 2. The results are presented in order of Zn < Cu < Ni < As < Cu < Hg < Cr < Cd. Additionally, the weighted mean values of Cd, Cr, Hg, Cu, Zn, As, and Ni were in the range of 0.0-222.7, 0.08-289.2, 0.03-193, 2.94-198.1, 0.0-771.1, 0.0-231, and 1.71-99.75.6 mg/kg, respectively (Table 2).

Compared to different backgrounds, the results appear to exceed all CNEMC (GB 15618–2018) standards indicating increased levels of heavy metal content as reported in similar studies [33,63,64]. Similarly, the weighted mean metal content of Cd doubled the standard of the MEF (European Union's most cited guideline) and the crust rock guideline, while the metal content of Hg and As were two to three times higher than the same guidelines. A plausible cause of the increase in Cd concentration in agricultural soils is the use of phosphate fertilisers, as most studies have documented an increase in Cd accumulation in agricultural soils under the intensive application of P fertilisers. Yuan, et al. [33] found increased concentrations of Cd, Zn, Hg, Cr, Ni, and As in agricultural soils in China over the past 20 years, thus corroboration of these findings. Therefore, it implies that Cd, Zn, and As should be prioritised as critical toxic metals in agricultural soils [17]. Similarly to these findings, Tang, et al. [65] reported an increasing concentration of Cd in agricultural farmlands in China, while Wen, et al. [63] attest that Cd and Hg were the main pollutants in Chinese farmlands [66]. Similarly, the Ministry of Environmental Protection (MEP) and the Ministry of Land and Resources (MLR) of China by reports show that 16.1% of all sampling sites were above the recommended threshold for agricultural soils, of which 82.4% of all the polluted soils were loaded with heavy metals [32]. However, the metal content of Cr, Cu and Ni was within the MEF standards; nevertheless, it is important to recognise that even at lower concentrations, the US-EPA considers Cr, Ni, As, and Cd as the most toxic metals in the environment [67].

Table 2 further shows the status of heavy metal levels in this study compared to related meta-analysis studies. Interestingly, the results of the meta-analysis of other published studies, compared to the current meta-analysis study, showed a slight disparity in the reported metal content for Cd, Zn, and As (Table 2). This highlights the severity of metal pollution in agricultural soils, particularly in China, as it is the region with the most reported cases of metal pollution (Fig. 7 and Figures 9A).

4. Prospects and implications of heavy metal pollution in agricultural soils

The results of this study have shown that agricultural soils are heavily polluted with heavy metals. The increasing number of scientific reports on heavy metal pollution also attest to these findings. For example, the search term in Scopus 'heavy metals' and "agricultural soils" and 'contamination' and "pollution" of published articles for the period 2010-2023 shows a general upward trend, with an annual average of 633 articles indicating that most agricultural soils have become 'hotspot' for heavy metal contamination (Fig. 8).

In terms of scholarly published literature by countries, China

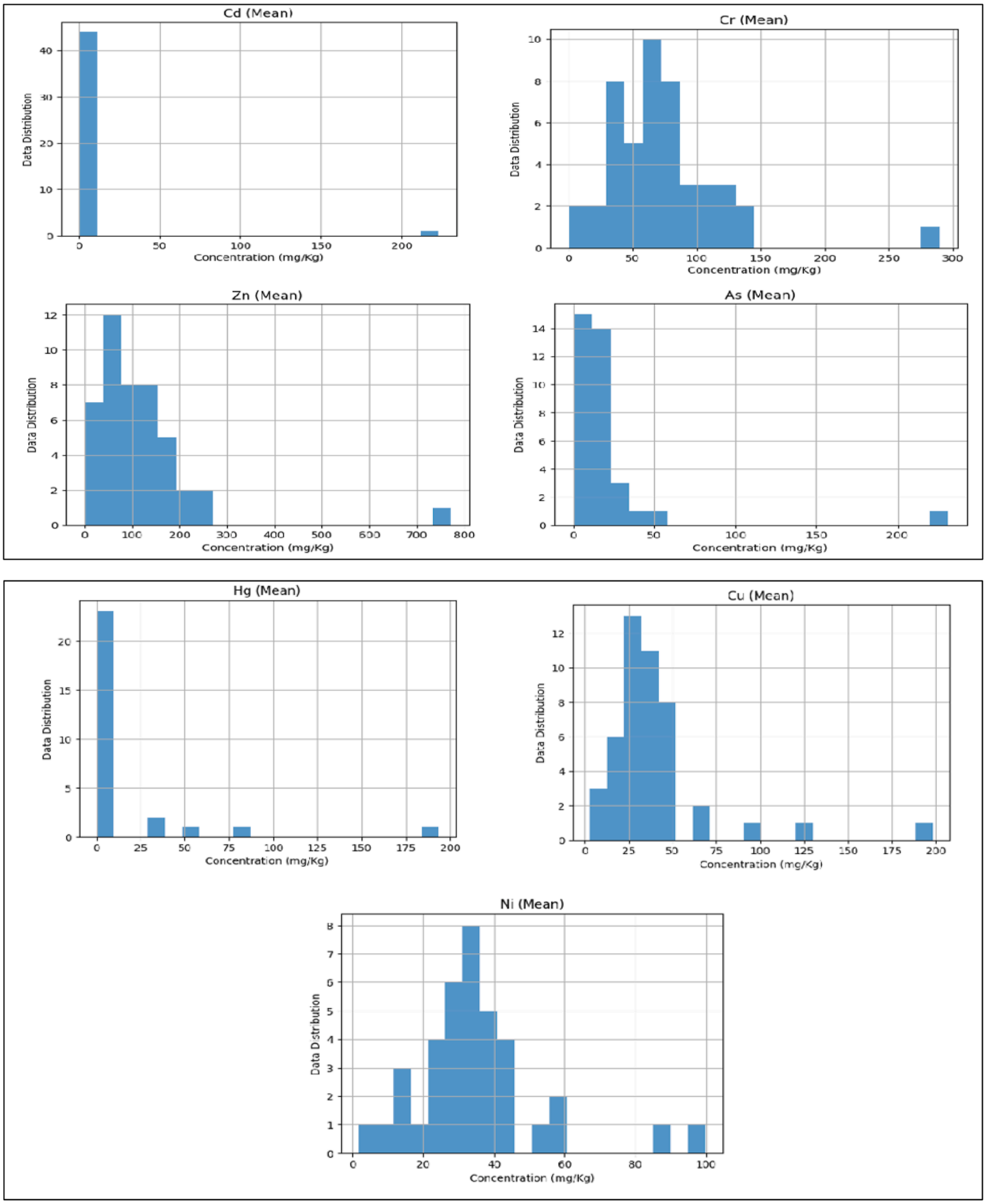

Fig. 4. Temporal variation of heavy metals concentrations in agricultural soils (2010 - 2023).

dominates exponentially published reports on heavy metal contamination, followed by India and the United States of America (Fig. 9A), while on the African continent, Egypt, Nigeria and Tunisia had the highest number of publications closely followed by Morocco and South Africa (Fig. 9B). This observation, in other words, further confirms the initial assertion that heavy metal contamination of agricultural soils represents a hidden danger to agricultural productivity, food safety, and ecosystem health worldwide [2,70].

The increased number of scientific reports on heavy metal pollution highlights two principal scenarios: first, there is a persistent increase in toxic heavy metal contamination in agricultural soil; and second, if not controlled, this pollution will eventually lead to global environmental collapse in nutrition, food chains, and ecological well-being. Studies have shown that infants, and adults are susceptible to carcinogenic risks of metals exposure through pathways such as ingestion, dermal contact and inhalation [67]. According to US Environmental Protection (US-EPA) agency, soils ingestion rate of 100 mg/day for children and 50 mg/day for adults in a population are susceptible to high health risks [11]. However, it is worth noting that these reports pose a serious concern about the credibility of the nature and types of reported studies, which might skew scientific narratives to believe that Chinese agricultural soils are highly polluted with heavy metals. For instance, Huang, et al. [32] is of the view that extensive attention and reports on the rate of heavy metal pollution in China might lead to an overexaggeration of

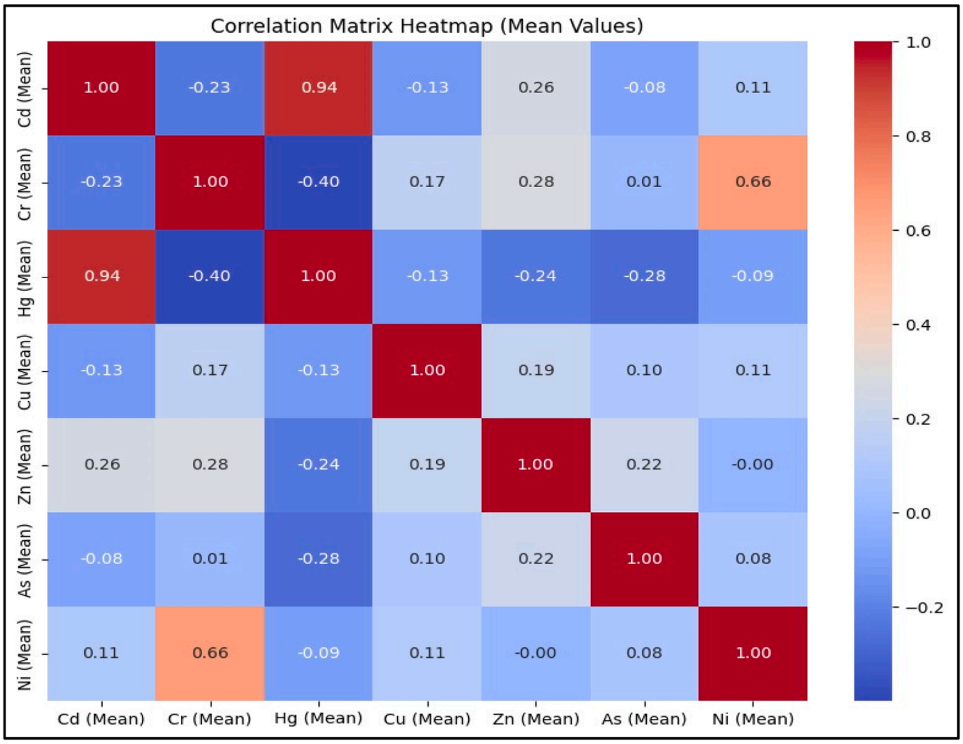

Fig. 5. The heat map of the heavy metals, $P < 0.05$ significance level.

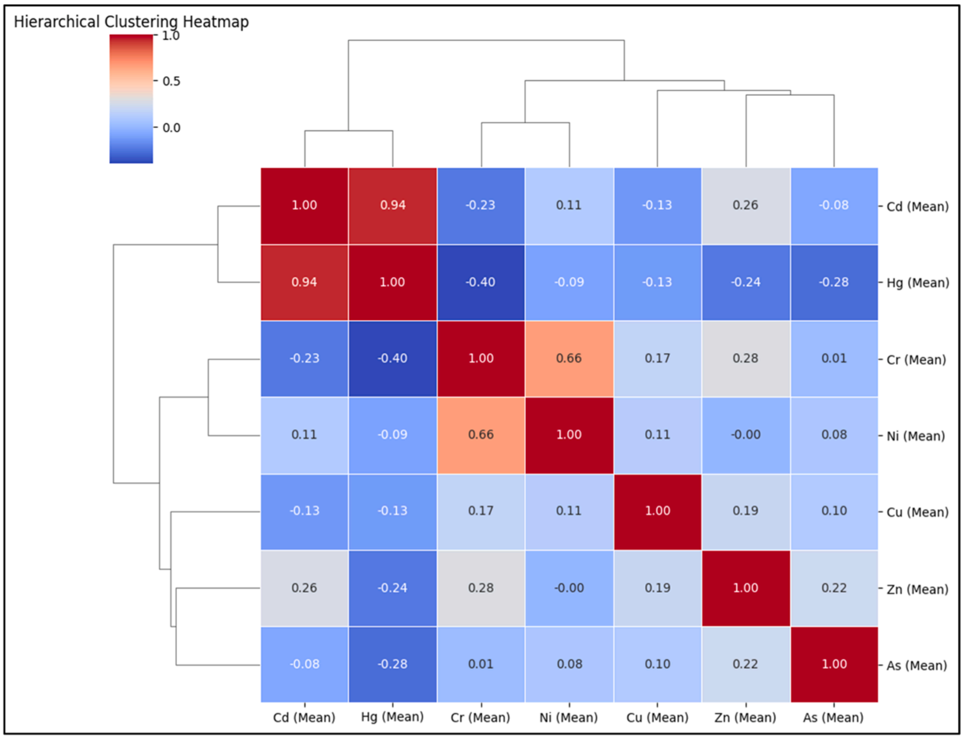

Fig. 6. Hierarchical cluster heat map of the heavy metals.

the real results. However, it is important to reckon that areas close to mining activities or adjacent to agricultural land might have a significant impact on heavy metals content in reported agricultural soils. Other authors have reported increased concentrations of heavy metals in nearby agricultural soils emanating from mining sites [56,57]. Consistent with these findings, it is evident that individual studies might report elevated concentrations that are two or three-fold higher than the contents in normal agricultural soils. However, such an assertion might not be entirely true because studies of heavy metal pollution are not based on subjective norms but rather on standardised experimental results that follow strict scientific protocols. However, the concentration of heavy metals in agricultural soils exceeded all standards as earlier mentioned.

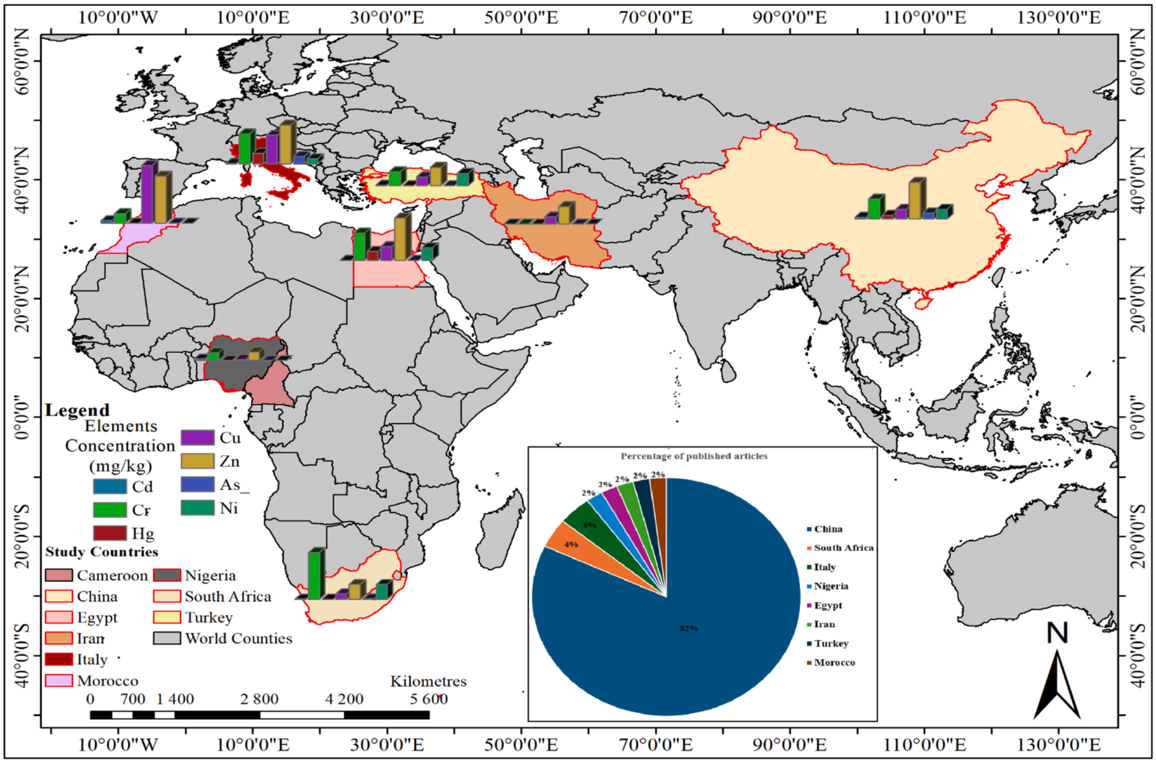

Fig. 7. Spatial distribution of heavy metals from the study countries.

Table 2

Literature analysis of heavy metal content (mg / kg) in agricultural soils (n = 50).

Countries/ regions	Average heavy metal contents in soils (mg/kg)							Reference articles	Study period	Sample size
	Cd	Cr	Hg	Cu	Zn	Ni	As			
8 Countries	2.17 ±0.19	5.32 ±1.10	5.89 ±0.38	37.38 ±0.66	111.91 ±2.19	14.74 ±0.32	34.19 ±0.61	This study	2010-2023	50
Minimum (mg/kg)	0.00	0.08	0.03	2.94	0.00	1.71	0.20	/	/	/
Maximum (mg/kg)	222.70	289.20	193.40	198.10	771.11	99.75	26.30	/	/	/
Number of missing values	6	3	22	4	8	12	16	/	/	/
Number of outliers	4	2	4	2	1	6	3	/	/	/
China	0.18	66.81	0.07	25.73	83.87	27.67	8.45	Yuan, et al. [33]	2000-2019	410
China	0.24	62.40	0.13	28.40	83.40	28.40	10.80	Huang, et al. [32]	2005-2017	336
34 Countries	0.23	*VNR	*VNR	25.83	88.38	29.21	*VNR	Shao, et al. [54]	2000-2014	68
China	0.275	66.33	0.143	29.94	80.10	*VNR	11.83	Ren, et al. [68]	1998-2019	112
Background values in China [a]	0.10	61.00	0.10	22.60	74.20	26.90	11.20	/	/	/
MEF threshold [b]	1	100	0.5	100	50	200	5	/	/	/
Crust rock [c]	0.2	59.5	0.07	38.9	70	29.0	6.83	/	/	/

[*] Values not reported.

[a] Standard value adopted the secondary standard of heavy metal content (GB 15618–2018). The background values were selected from the CNEMC, 1990 (China National Environmental Monitoring Centre).

[b] The threshold values and guidelines of the Ministry of Environment of Finland for metals in soils (extract; MEF, 2007).

[c] Crust rocks: The contents (mg/kg dry weight) of the elements in global averages for crustal rocks by Kabata-Pendias [69].

For example, a comprehensive study in the European Union (EU) estimates that 137,000 km^2 of agricultural land are polluted, and 6.24% of the 280 topsoil samples have varying types of heavy metal concentrations above the guidelines set for agricultural land [71]. Zabel, et al. [5] observed that the suitable area for agriculture farming is decreasing in terms of quality, and further pollution could exacerbate the current rate of contamination of existing cultivated land due to urbanisation.

The statistical summary of the results of the experimental study in Cameroon, as shown in Table 1, indicates low concentrations compared to other reported studies and the MEF and crust rock standards. Similarly, observations from the statistical summary in Table 2 show that the weighted mean values from this study were slightly different compared to those of Yuan, et al. [33], Huang, et al. [32], Shao, et al. [54] and Ren, et al. [68], which demonstrates the reasonableness of using the weighted mean values to calculate heavy metals. Thus, one could, at face value, conclude that heavy metal pollution is of major international concern. However, relying solely on meta-analysis to draw a generic conclusion might sometimes be misleading. This is because areas of 'hotspot' pollution are not inherent to anthropogenic activities or geogenic processes but straddle across a set of interacting phenomena such as meteorological activities, land use types, particle size, absorption rate, soil redox, etc. For example, in a study carried out by Shao, et al. [54] in

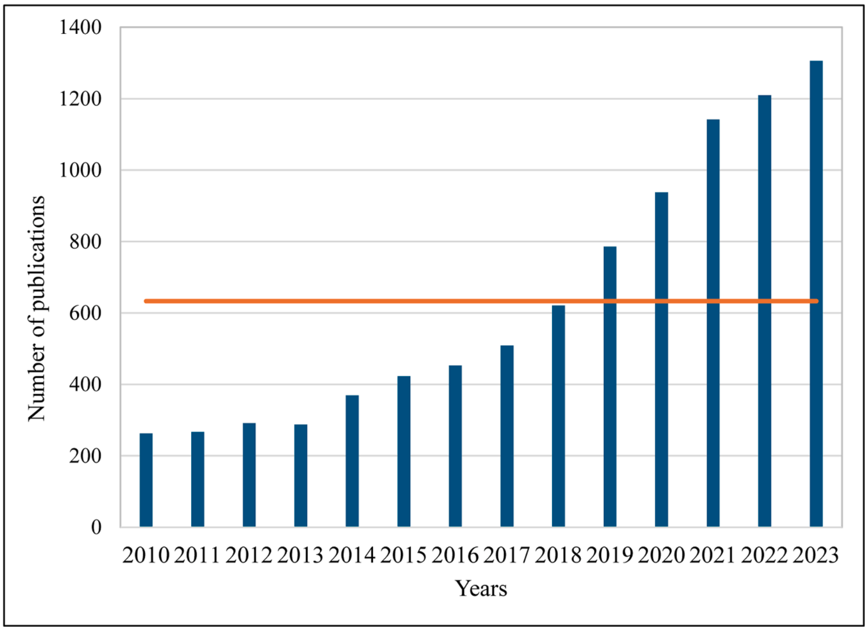

Fig. 8. Publications reporting heavy metal contamination of agricultural soils from 2010-2023.
Source: Author 2024.

Fuyang County, the results showed extremely lower Cd concentrations of Cd (0.24 mg/kg) than those of previous studies (1.12 mg/kg), which was attributed to sampling differences. Thus, any scientific argument should not be about whether reported studies are exaggerating their reports because even in small concentrations, heavy metals are still dangerous. This view has been unanimously accepted and is well established in contemporary literature [10,13].

Although this paper has collected a large amount of empirical literature in the field of heavy metals, the conclusions obtained may be biased due to the small amount of valid data in some regions. For example, the evaluation results of this article show that there is an increase in the number of cases of heavy metals contamination in agricultural soils. Hou, et al. [14] argue that soils, whether for agricultural purposes or not, contain an estimated 4.1 trillion tonnes, nearly five times the estimated mass of atmospheric carbon, which, when contaminated, might not be able to fulfil its role in carbon recycling, thus aggravating climate change, which inevitably has a spiral effect on food security, human health and ecological well-being. Since some of the sampling locations in the statistical literature are special (82% from China alone), with South Africa and Italy each accounting for 4% each, and Nigeria, Egypt, Iran, Turkey, and Morocco accounting for 2%, respectively, it can objectively cause the global pollution level to be overestimated. Therefore, it is necessary to pay attention to developing mitigation measures for heavy metal pollution control and remediation to ensure that the future situation does not become worse. Therefore, it is necessary that the contamination status of heavy metal pollution contamination status in any given country is based not only on experimental studies or meta-analysis, but also on synergy through a nexus approach to reduce sectoral trade-offs in data generation, processing, analysis, and interpretation. In this light, comprehensive pollution studies of the major pollution sources for each region can be identified for effective pollution management.

5. Conclusion

In this study, the status of heavy metals (Cd, Cr, Hg, Cu, Zn, Ni, and As) in agricultural soils was analysed using a combination of meta-analysis and an experimental case study. Spatiotemporal analysis of heavy metals from 2010 - 2023 reveals a significant increase in metal concentrations of Cd, Hg, Zn and As above the recommended guidelines for MEF and crust rock. The spatial perspectives shows that a higher number of studies were conducted in China (82%), with Italy and South Africa accounting for 4% each and Iran, Nigeria, Egypt, Morocco, and Turkey with 2%, respectively. It was observed that the close relationship between a large number of publications from China could be attributed to industrial expansion over the past few decades. Similarly, a positive correlation between Cr and Ni ($R^2 = 0.66$) suggests that these elements have the same source and are likely prevalent in agricultural soils. The results of the contamination indices reveal that agricultural soils are highly contaminated with Cr, supported by the PLI ranging from baseline to moderate pollution for Cd, Cu, Zn, Ni, and As, except for Cr with very high pollution. The implications of these findings would be of great significance for the development of heavy metal pollution control strategies to reduce environmental contamination.

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Ethical statement

In line with research ethical commissioning of the NWU, this research adheres to the research ethos and does not involve any human or animal subject. Thus, the research was considered of no risk.

CRediT authorship contribution statement

Samuel Che Nde: Writing – review & editing, Writing – original draft, Validation, Conceptualization. **Obono Mba Felicite:** Writing – review & editing, Validation, Formal analysis. **Gabriel Sanjo Aruwajoye:** Writing – review & editing, Validation, Methodology, Data curation. **Lobina Gertrude Palamuleni:** Writing – review & editing, Writing – original draft, Supervision, Conceptualization.

Declaration of competing interest

The authors declare that they have no known competing financial

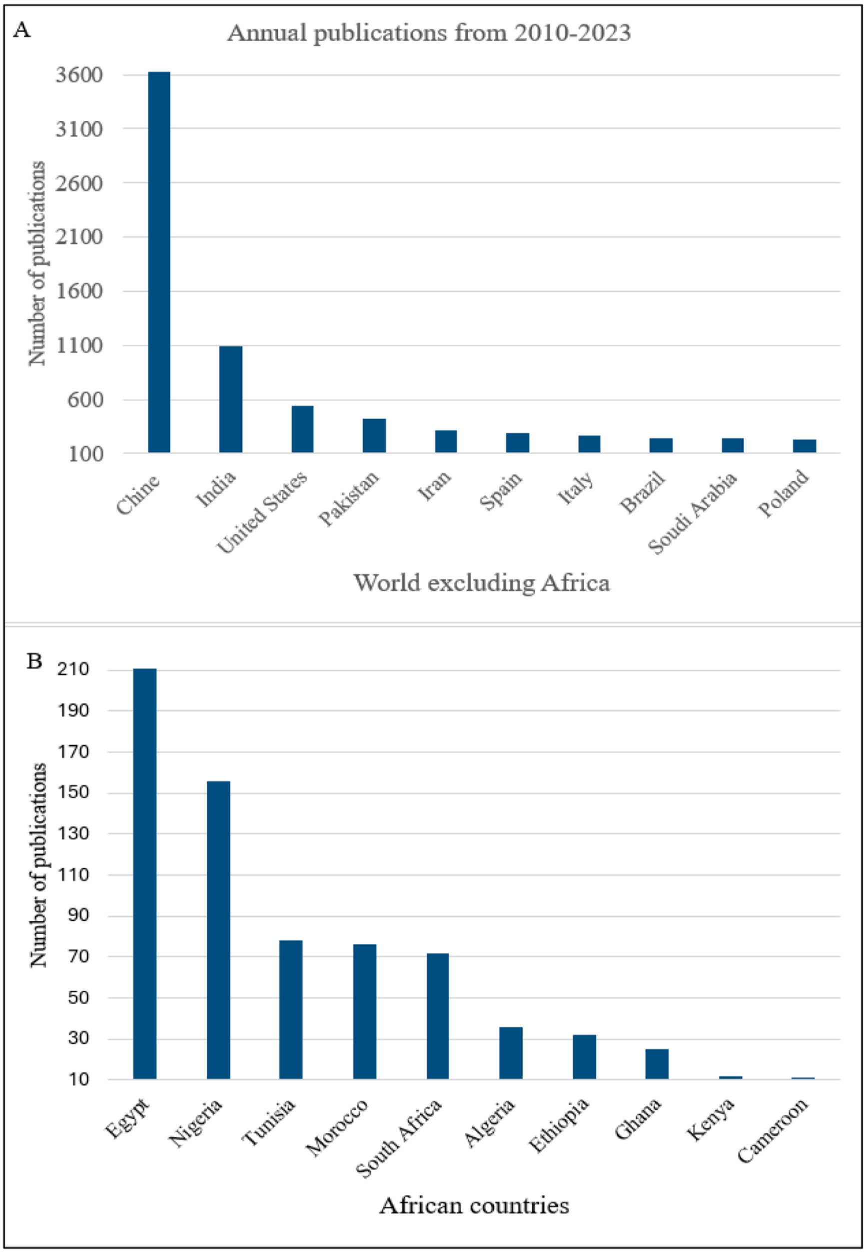

Fig. 9. Publications reporting heavy metal pollution from 2010-2023.
NB: A, represent selected countries in the world, and B, represent selected African countries. Source Author: 2023.

interests or personal relationships that could have appeared to influence the work reported in this paper.

Supplementary materials

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.jtemin.2024.100180.

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