JOURNAL OF AGRICULTURE AND APPLIED BIOLOGY
2024, Vol. 5, No. 2, 164 - 177
http://dx.doi.org/10.11594/jaab.05.02.03
E-ISSN: 2723-5106

Research Article

Impact of agricultural inputs on the abundance of heavy metals (Cu, Zn) in soil, water, and plants in the south of Algeria

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Article history:

Submitted 28 May 2024
Accepted 19 June 2024
Revised 21 June 2024
Published 09 August 2024

Keywords:

Contamination
Environment
Fertilizer
Pesticide
Pollution

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Abstract

The aim of our study is to assess the impact of agricultural inputs on the levels of heavy metals, specifically copper (Cu) and zinc (Zn), in soil, water, and plants in the oasis of El Ghrous, Branis, and Sidi Okba in Algeria. The existence of perilous substances in the surroundings is a significant ecological issue, endangering both human well-being and agricultural output. The overuse of fertilizers and pesticides may be causing heavy metals build-up. The aim will be to highlight a better understanding of the concentrations of copper (Cu) and zinc (Zn), considering as detrimental contaminants, after extensive utilization of agricultural inputs. In order to examine the levels of trace metals (TMs), we performed additional tests by comparing field samples using metal analysis techniques that involved ammonium acetate and EDTA, and detection by atomic absorption spectrophotometry. Our findings suggest that in soils affected by fertilizers and phytosanitary agents, the concentrations of Cu^{++} and Zn^{++} are below levels that would cause toxicity. Indeed, the levels of Cu^{++} are measured at 6.14 µg/g and the levels of Zn^{++} are measured at 4.97 µg/g. Nevertheless, the copper contents in irrigation waters from sites 01 and 02 beyond the levels that are considered dangerous. Similarly, copper levels in plants are categorized as hazardous. The findings emphasize the significance of implementing integrated pest management strategies in order to reduce the negative consequences of excessive use of agricultural inputs. Additionally, our study imposes the need to closely monitoring agricultural practices in order to mitigate environmental and public health hazards. Implementing sustainable farming practices and employing effective resource management strategies is crucial for safeguarding agricultural ecosystems and guaranteeing sustainable food production in the long run. Adopting an all-encompassing strategy is crucial for preserving the well-being of both the ecosystem and the populace.

How to cite:
Guesmia, H., Tarai, N., Ouamane, A. T., Guimeur, K., Masmoudi, A., & Djellouli, A. (2024). Impact of agricultural inputs on the abundance of heavy metals (Cu, Zn) in soil, water, and plants in the south of Algeria. *Journal of Agriculture and Applied Biology, 5*(2): 164 - 177. doi: 10.11594/jaab.05.02.03

1. Introduction

The environment is getting increasingly polluted as a direct result of the great economic expansion and quick growth that has occurred in many different fields, such as agriculture and industry (H. Ali et al., 2019; Naidu et al., 2021; Sharma et al., 2023). Heavy metals and pesticides are two of the most frequent types of pollutants, both of which pose a threat to the entire ecosystem by disrupting its function and destroying its structure (Donald et al., 2022; Timothy & Tagui Williams, 2019). Agroecosystems are commonly exposed to a diverse array of pollutants, including agricultural pollutants that arise from farming practices. As a consequence, the presence of these pollutants results in contamination and deterioration of the surrounding agroecosystem. Fertilizers, pesticides, and sewage sludge are among the most prevalent agricultural sources of heavy metals. The toxic heavy metals exhibit varying characteristics and modes of accumulation, whether in the soil or plants (Alengebawy et al., 2021; Srivastava et al., 2017).

Soil plays a crucial role in agricultural production by not only providing support for plants but also serving as a reservoir for vital nutrients (Hou et al., 2020). However, due to the intensification of agriculture, the soil is threatened by erosion, pollution, and declining fertility, as well as poor and inappropriate agricultural practices (Duarte et al., 2017; Miseckaite, 2022; Upadhyay et al., 2020). The widespread use of chemical products in agriculture, particularly mineral fertilizers and plant protection products, has made possible the intensification of agriculture with a spectacular increase in crop yields, including plasticulture, which is one of the most intensive agricultural practices (Bibi & Ilyas, 2020; Upadhyay et al., 2020).

The use of pesticides and mineral fertilizers is a fact that can affect the quality of soils through their physicochemical modification and contamination (de Jesus et al., 2023; Schaeffer & Wijntjes, 2022; Streletskii et al., 2022). In addition, these chemical products can lead to the pollution of surface water as well as groundwater (Javaid et al., 2023; Rasool & Thakur, 2023; Yadav et al., 2022). These chemicals contain relatively high levels of trace metals (TMEs), often with high levels of copper and zinc. Repeated applications to the same plot can potentially alter the loadings and distributions of these metals in the different soil fractions, and subsequently disrupt soil-solution equilibria (Cagnarini et al., 2021; Halli et al., 2021; Szostek et al., 2023).

In Algeria, the cultivation of vegetable crops in greenhouses has experienced rapid growth. Currently, the Biskra region is a major producer of early vegetables with high yields (Aidat et al., 2023; Belhadi et al., 2016). This significant production is attributed to the extensive area dedicated to greenhouse farming, comprising approximately 49% of the total national greenhouse cultivation area. Furthermore, the production accounts for around 60% of the overall greenhouse production in Algeria, including common crops such as tomatoes, peppers, eggplant, melon, courgettes and cucumbers (Aidat et al., 2023; Bakroune et al., 2023). Unfortunately, the rise in crop yields has led to increased use of plant protection agents and fertilizers, especially in regions like M'Ziraa and El Ghrous, as well as Sidi Okba and Branis. These regions are known for their abundant vegetable production and have numerous points of sale for plant protection products (Bettiche et al., 2019; Ramdani et al., 2009; Salah Bouchemal, 2021).

Today, soil contamination due to agricultural activity is a serious problem. It is imperative to understand the abundance of heavy metals in soils and water after long-term fertilizer and pesticide use, as well as their effects on the environment. In this context, our work aims to study the toxic effects of phytosanitary products and fertilizers on the abundance of heavy metals, specifically copper and zinc, in soil, water, and plants. For this purpose, we selected three study sites in the Biskra region: Branis, El Ghrous, and Sidi Okba.

2. Materials and methods

2.1 Presentation of the study area

The Wilaya of Biskra is located in the southeast of Algeria, in the southern foothills of the Saharan Atlas. It covers an area of 21,671 km^2 and is situated at an altitude of 87 meters. Its latitude is 34° 48' and its longitude is 5° 44' (Figure 1). The climate in this region is semi-arid to arid, with an average annual rainfall of 150 mm. The soils in the study sites are predominantly of alluvial origin and are characterized by clay-loam textures in Branis and Sidi Okba, and loamy-sand textures in El Ghrous. These soils are frequently saline and contain significant amounts of gypsum and limestone. Additionally, they are generally poor in organic matter (Khadraoui, 2010).

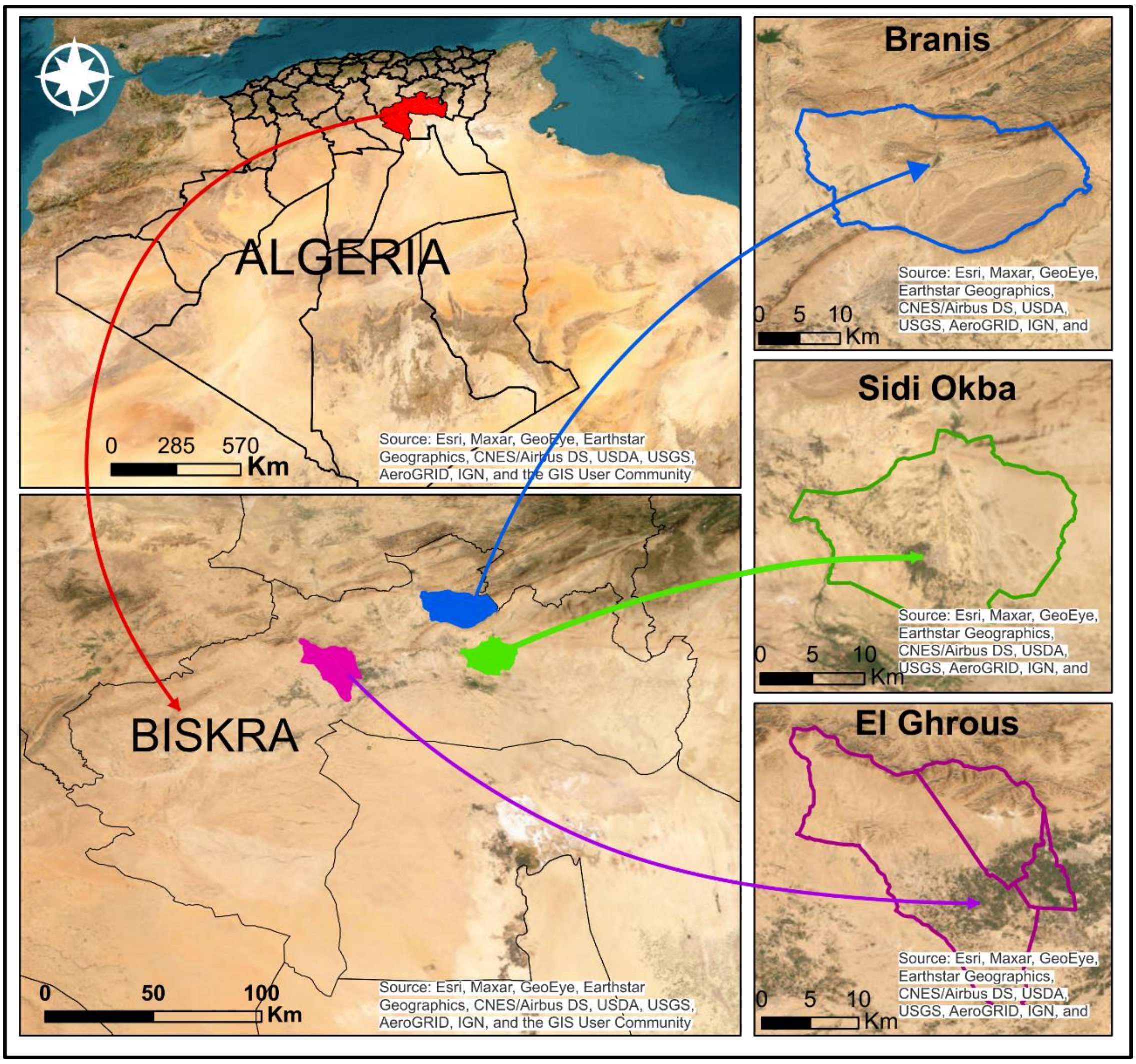

Figure 1. Map of the geographical location of the study area

2.2 Location of the experiment

After prospecting the study area, three locations with distinct water and soil characteristics representing significant agricultural potential were selected (Figure 1). The first site is in Branis (Dar Aarous), 20 kilometers north of Biskra; the second site is in El-Ghrous, 47 kilometers west of

Biskra; and the third site is in Sidi Okba (Aalb Charmat), 25 kilometers east of Biskra. The sites are homogeneous and have been used for vegetable production for over a decade, resulting in the intensive use of pesticides and fertilizers.

2.3 Sampling

Under greenhouses, soil samples were collected at two depths, 0-30 cm and 30-60 cm, with six samples/site for the soil affected by pesticides and fertilizers and two samples/site for the control. There were 24 soil samples, 3 irrigation water samples, and 12 physiologically mature plant samples from four organs, including leaves, roots, stems, and fruits. The soil samples were collected using an Edelman auger and preserved in plastic bags with the date and site number clearly labeled. The three water samples were transferred to plastic receptacles bearing the date and site number. Vegetable cultivars (courgette for sites 01 and 02, and tomato for sites 03) were used for the plant study. The four plant elements (Root (R), Stem (S), Leaves (L), and Fruit (Fr)) were analyzed.

Water		
Site 01	**Site 02**	**Site 03**
B_W	L_W	S_W

Soil								
Site 01			**Site 02**			**Site 03**		
B_1 0-30	B_2 0-30	B_3 0-30	L_1 0-30	L_2 0-30	L_3 0-30	S_1 0-30	S_2 0-30	S_3 0-30
Bt_1 30-60	Bt_2 30-60	Bt_3 30-60	Lt_1 30-60	Lt_2 30-60	Lt_3 30-60	St_1 30-60	St_2 30-60	St_3 30-60

Plant											
Site 01				**Site 02**				**Site 03**			
B_R	B_S	B_L	B_F	L_R	L_S	L_L	L_F	S_R	S_S	S_L	S_F

Figure 2. Diagram of the experimental set-up (factorial design), Site 01: Branis site (B), Site 02: Leghrous site (L), Site 03: Sidi Okba site (S), W: water sample, t: soil control, $_R$: Root, $_S$: Stem, $_L$: Leaves, $_F$: Fruit

2.4 Soil sample preparation

Following the drying process, the samples underwent grinding in a mortar and subsequent sifting through a 2mm sieve to ensure their representativeness for the analysis.

2.5 Analysis of heavy metals (Zn, Cu)

The method utilized for the analyses was as described in reference (Mathieu & Pieltain, 2003), wherein the specimens were dissolved using a combination of ammonium acetate and EDTA.

2.6 Statistical analysis

A multi-factor analysis of variance (ANOVA) was employed in our study to investigate the factors that contribute to the variability in Cu and Zn levels. Figure 2 was used to illustrate these causes. The Newman-Keuls test was used to determine if there were any significant differences between the group means based on a factorial design. The Pearson correlation method was employed to evaluate the magnitude and orientation of the correlation between Cu and Zn, and pH and EC. The generation of graphs and computations is facilitated through the utilization of R. Studio and IBM SPSS Statistics 25.

3. Results and discussions

The ANOVA results indicate significant differences in Cu and Zn concentrations among the sites, depths, and plant parts, with Pr > F values confirming the reliability of these differences. Post hoc SNK tests further identify homogeneous groups, as indicated by the letters on the bars in Figures 3, 4, 5, and Table 1. The results shown in Figure 3 demonstrate that for Site 1, there is always a significant increase in the copper content of the treated soils compared to the control soils; this increase can range from 5.74 µg/g for the depth 0-30 cm to 6.15 µg/g for the depth 30-60 cm. However, for Sites 02 and 03, the concentration of copper in the soils is slightly lower compared to the control soils, with values ranging from 4,99 to 5,69 µg/g for the depth 0-30 cm and from 4.67 µg/g to 5,77 µg/g for the depth 30-60 cm. On the other hand, when comparing the means of all samples from the same site, as illustrated in Table 1, it is observed that Sidi Okba has the highest mean of copper concentration, followed by Branis, and finally Elghrous, with respective values of 5.418, 5.178, and 4.615 µg/g.

Copper may be depleted from treated soils due to organic matter and irrigation techniques (Shaheen et al., 2015; Suszek Gonçalves et al., 2020), because Copper is a highly soluble mineral, and it can easily leach from the soil when excessive water is applied or when the soil has inadequate drainage (Nicol et al., 2022b). Copper, however, can be adsorbed by clay minerals and organic matter in the soil, rendering it unavailable for plant absorption (Palanisamy et al., 2022). Over time, the quantity of copper adsorbed can increase, resulting in a decline in the soil's available copper (Filipović et al. 2023; Nicol, Welham, and Senanayake 2022a).

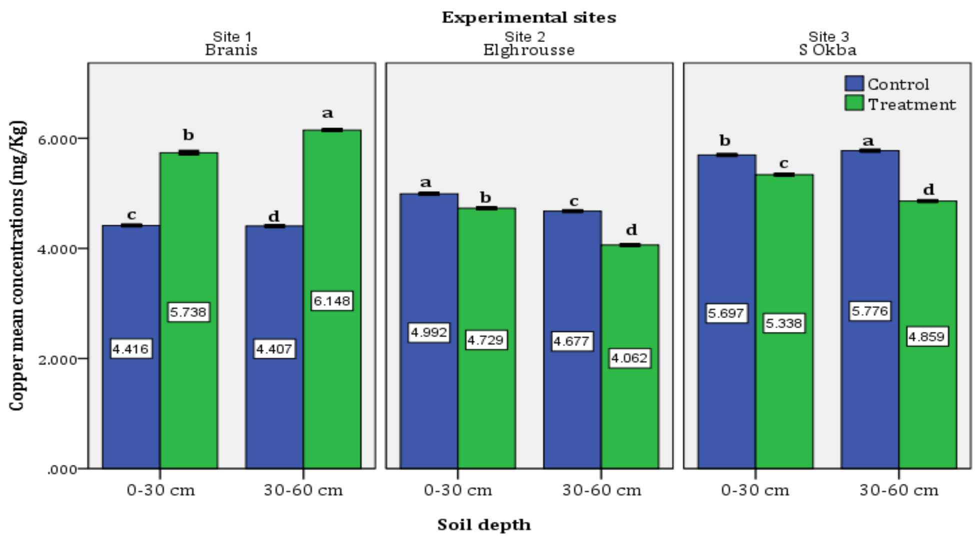

Figure 3. Variation of copper in soil (CI: 95% confidence interval)

The concentration of copper is comparatively higher in the top soil layer of 0-30 cm as opposed to the subsoil layer of 30-60 cm. The average of the whole set of samples collected from the topsoil was 5.15 µg/g, whereas the average of the samples obtained from the subsoil was 4.99 µg/g (Table 1). The phenomenon under consideration can be elucidated by the limited mobility of heavy metals, as documented by Coppenet (1981), Baize (2018) and Salam (2021).

Copper concentrations may be higher in soil strata containing more organic matter. Because organic matter can bind to copper and prevent it from leaching into deeper soil strata, it can prevent copper from leaching (Lekfeldt et al., 2017; Oorts, 2013). In contrast, soil texture can influence the transportation of water and nutrients through the soil profile. Copper may be more likely to accumulate in finer-textured soils, such as clay, as these soils have a greater capacity to retain nutrients (Dutta & Lodhari, 2018). Based on the copper toxicity threshold of 70 mg/kg proposed by Coïc (1989), Coppenet (1981), and Baize (2018), it can be concluded that the three study sites are significantly distant from the threshold.

Table 1. Cu and Zn Concentrations in Water, Soil, and Plants with ANOVA Results

Type	Factors	Terms and conditions	$\overline{[Cu]}$ +/- DS (ppm)	$\overline{[Zn]}$ +/- DS (ppm)
Soil	Experimental site	S Okba	5,418 +/- 0,372	3,288 +/- 1,073
		Branis	5,178 +/- 0,8	2,107 +/- 0,719
		Elghrousse	4,615 +/- 0,35	2,957 +/- 0,944
		Pr > F	< 0,0001	< 0,0001
		Significant	Yes	Yes
	Phytosanitary treatment	Treated	5,146 +/- 0,698	3,13 +/- 1,232
		Untreated	4,994 +/- 0,57	2,438 +/- 0,651
		Pr > F	< 0,0001	< 0,0001
		Significant	Yes	Yes
	Profiler	0-30 cm	5,152 +/- 0,495	3,265 +/- 1,24
		30-60 cm	4,988 +/- 0,751	2,303 +/- 0,415
		Pr > F	< 0,0001	< 0,0001
		Significant	Yes	Yes
Water	Experimental site	Branis	0,218 +/- 0,002	0,375 +/- 0,001
		Elghrous	0,248 +/- 0,003	0,267 +/- 0,002
		S. Okba	0,165 +/- 0,002	0,267 +/- 0,002
		Pr > F	< 0,0001	< 0,0001
		Significant	Yes	Yes
Plant	Experimental site	Branis	41,03 +/- 2,778	44,116 +/- 16,046
		Elghrous	36,528 +/- 3,988	46,329 +/- 17,4
		S. Okba	45,96 +/- 5,776	28,972 +/- 10,295
		Pr > F	< 0,0001	< 0,0001
		Significant	Yes	Yes
	Organ	Leaves	44,687 +/- 1,242	53,135 +/- 7,564
		Roots	44,125 +/- 8,29	49,828 +/- 12,39
		Stems	38,555 +/- 4,347	37,645 +/- 12,799
		Fruits	37,323 +/- 2,442	18,613 +/- 0,652
		Pr > F	< 0,0001	< 0,0001
		Significant	Yes	Yes

$\overline{[X]}$ +/− *SD: mean concentration +/- standard deviation. The illustrated means for the soil and plants indicate the overall averages of each factor individually, while considering the treatments of the other factors as repetitions.*

It can be observed from Figure 4 that there is always an increase in zinc levels for site 03, compared to the soils at sites 01 and 02, and this increase can range up to 4.97 µg/g for the treated soils and 3.28 µg/g for the control soils. This can be confirmed by the fact that the average of zinc concentration in the entire set of samples collected from the site 3 (around 3.29 µg/g) is significantly higher than those at the other two sites (Table 1).

Furthermore, at all three sites, zinc was found to be higher at depths of 0-30 cm than at depths of 30-60 cm, except for the control soil at site 01, where the opposite was found. There was a greater concentration of zinc at the depths of 30-60 cm (2.82 µg/g) compared to 0-30 cm (1.26 µg/g).

A maximum value of 4.97 µg/g of zinc was recorded for the treated soil at site 03 at a depth of 0-30 cm, whereas a minimum value of 1.265 µg/g was recorded for the control soil at site 01.

Several factors can influence the concentration of zinc in soil, including the abundance of organic matter. The presence of organic matter can enhance the availability of zinc in soil by binding to the nutrient and gradually releasing it over an extended period (Fan et al., 2016; Opfergelt et al., 2017). Soil texture and soil moisture are two important soil factors that affect the availability of zinc to plants (Natasha et al., 2022; Noulas et al., 2018). Specifically, Zinc has a greater tendency to bond with clay particles in soils with certain textures, thereby reducing its availability (Mossa et al., 2021; Recena et al., 2021; Thingujam et al., 2019). Conversely, Zinc is more readily accessible in soils that are well drained and have sufficient moisture content (Lv et al. 2022 ; Wen et al. 2019).

The increase in zinc concentrations in the 0-30 cm depths compared to the 30-60 cm depths is due to the low mobility of heavy metals in soil, which is mainly due to their tendency to bind to soil particles, especially those with high surface area and negatively charged surfaces, such as clay and organic matter. This adsorption reduces their mobility and availability to plants (Nieder et al., 2018).

The use of pesticides and fertilizers can have both positive and negative impacts on the concentration of zinc in the soil. Fertilizers containing zinc can increase the concentration of zinc, while pesticides can also affect the concentration of zinc (Alengebawy et al., 2021; Hassan et al., 2022). Soil pH can also affect the availability of zinc to plants, while leaching can lead to the loss of zinc and other nutrients from the soil (Adamczyk-Szabela & Wolf, 2022; Amrani et al., 1997; Neina, 2019). In accordance with the soil thresholds suggested by Coïc and Coppenet (1989), Coppenet (1993), and Baize (2018), it can be inferred that the samples analyzed in this study exhibit a significant distance from the zinc toxicity levels of 100 µg/g or 50 µg/g.

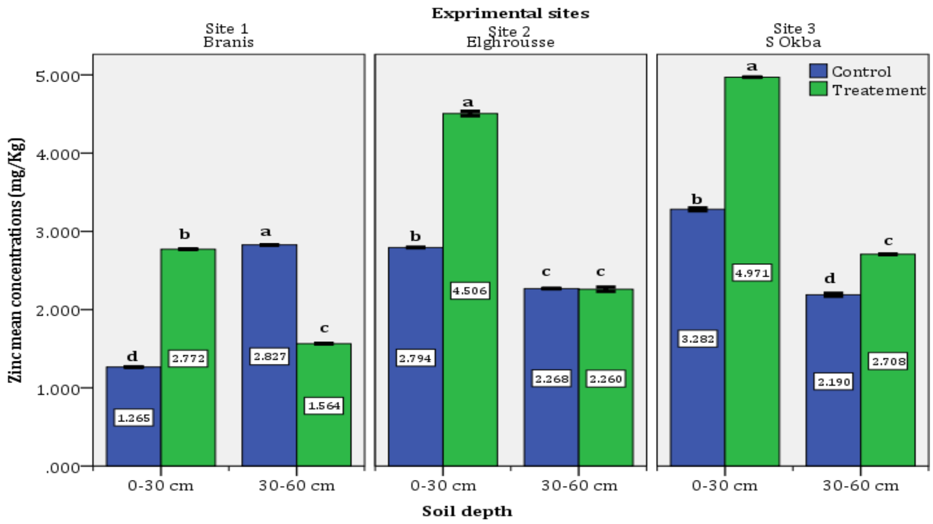

Figure 4. Variations in soil Zinc at the three sites (95% confidence interval).

The findings depicted in Figure 5 indicate that the levels of copper present in the irrigation water at sites 01 and 02 exceed the concentration observed at site 03, which was estimated to be 0.17 µg /ml. The copper concentration at site 03 is far below the water toxicity thresholds, whereas sites 01 and 02 are very near to Peterson's (Couture, 2004) proposed toxicity standard (0.2 mg/l). In contrast, based on the State Secretariat for Energy, Water, and Environment's proposed standards (2 mg/l) (Secretary of State for the Ministry of Energy and Environment, 2002), the irrigation water of the three sites is well below the toxicity thresholds. For zinc, Figure 5 and Table 1 demonstrate that the values for sites 02 and 03 are identical, with a concentration of 0.267 g/ml. In contrast, site 01 has the greatest zinc concentration (0.3752 g/ml). The results from the three sites are well below the 2 ppm toxicity thresholds cited by the Secretary of State for the Ministry of Energy and Environment (2002) and Couture (2004).

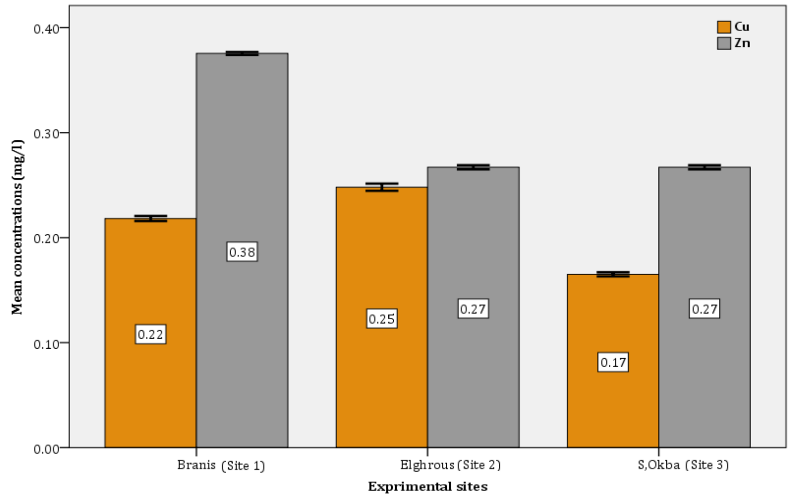

Figure 5. Copper and zinc variations in irrigation water for the three sites (CI: 95% confidence interval)

The results indicate (Figure 6 and Table 1) that the concentrations of copper are greatest in the leaves, with values ranging from 43.03 µg/g to 45.85 µg /g, and in the roots, with values ranging from 35.88 µg/g to 54.97g/g. On the other hand, the stems and fruits contain significantly less copper, with concentrations ranging from 33.06 µg/g to 43.25 µg/g for stems and from 34.14 µg/g to 39.78 µg/g for fruits. It is observed that the highest copper content is found in the tomato roots of site 03, with a concentration of 54.97 µg/g, and it is additionally noted that the copper concentration is always higher in tomato crops than in zucchini crops.

According to the standards proposed by Loué (1993) and Tremel-Schaub and Feix (2005), the results obtained for copper levels in roots, stems, leaves, and fruits are toxic.

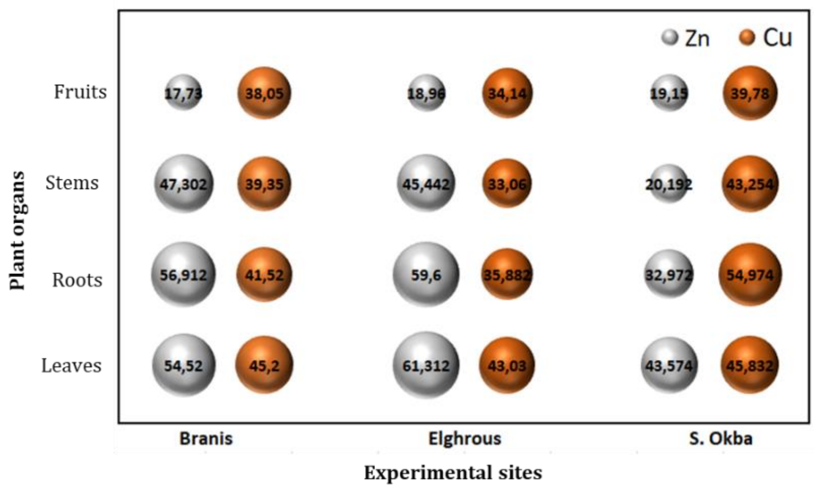

Figure 6. Cu and Zn variations in zucchini and tomato crops at the three sites

Zinc concentrations are greater in the leaves, with values ranging from 43.57 µg/g to 61.3 g/g, as well as in the roots, where values range from 32.97 µg/g to 59.6 µg/g (Figure 6 and Table 1). In contrast, the Zinc concentrations in the stems and fruits are practical, ranging from 20.19µg/g to 47.3µg/g for the stems and from 17.73 µg/g to 19.14 µg/g for the fruits. The highest zinc concentration was found in the leaves of site 2 with a value of 61.3 g/g, whereas the lowest zinc concentration was found in the fruits of site 1 with a value of 17.73 g/g. It is also noted that, contrary to copper, the concentration of zinc is always lower in tomato crops than in zucchini crops. Upon classification of the results in accordance with Loué (1993), it was determined that all samples exhibited typical Zn concentrations. However, based on the findings of Tremel-Schaub and Feix (2005), the majority of the results surpassed the toxicity threshold, with the exception of four outcomes. These four results, which were 17.73 µg/g, 18.96µg/g, 19.14 µg/g, and 20.19 µg/g, were obtained from the fruits of sites 1, 2, 3, and the stems of site 3, respectively.

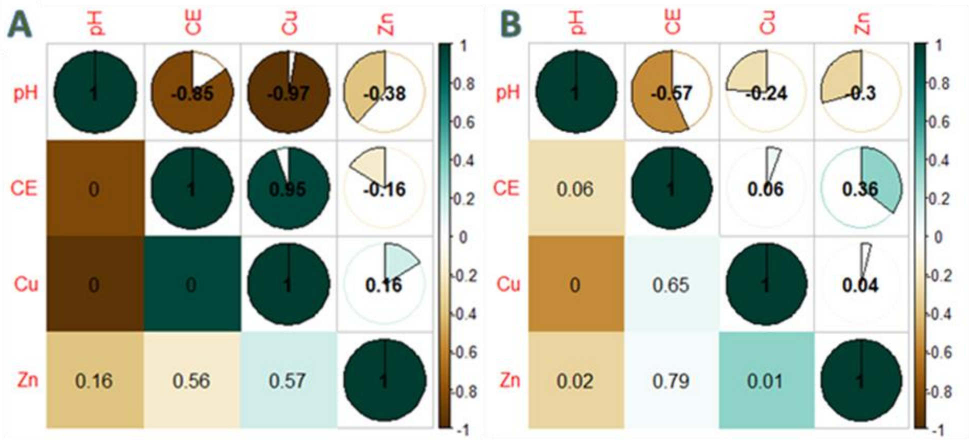

Figure 7. Pearson correlation matrix of Cu, Zn, pH, and EC, in irrigation water (A), and soil (B), the correlation coefficients (r) appear on the top triangles with a pie chart display of the proportions of these values, while the P-Value values appear on the bottom triangles

According to the correlation matrix of Cu, Zn, pH, and EC in soil and irrigation water (Figure 7), the amount of zinc in either the soil or irrigation water has no effect on copper concentration. However, it appears that the level of acidity (pH) in irrigation water can significantly (R=-0.97/p0.001) explain a large portion of the variation in copper content, whereas in the soil, pH is significantly more related to zinc, but with a lower level of significance, estimated at R=-0.30/p0.05. These findings concur with those of several other works (Alengebawy et al., 2021; A. Ali et al., 2019; Alloway, 2013).

4. Conclusion

This study evaluates the potential harmfulness of plant protection chemicals and fertilizers by analyzing their effects on the concentrations of heavy metals (Copper and Zinc) in soil, water, and plants. The results of our study show that there are higher concentrations of copper and zinc in soils that have been treated, particularly at a depth of 0-30 cm. This is mainly because heavy metals have limited ability to move within the soil, and also because fertilizers and phytosanitary products have been applied.

The highest concentrations of copper and zinc were found in the leaves and roots of plants, respectively, while stems and fruits contained lower levels. Specifically, tomato roots showed the highest copper concentration, and zucchini leaves exhibited the highest zinc levels. These variations can be attributed to factors such as the chemical composition of agrochemicals, the physiological and nutritional needs of the plants, and the properties of the metals themselves.

Levels of copper in roots, stems, leaves, and fruits are considered toxic, suggesting a substantial risk of phytotoxicity in all study areas. On the other hand, zinc levels in plant samples typically fall within normal ranges according to some standards, although they may exceed toxicity thresholds according to others, except for fruits and tomato stems at certain sites.

Our study emphasizes that the occurrence of soil toxicity from Cu and Zn is not universally observed. Nevertheless, the presence of phytotoxicity can be observed as a result of the extensive utilization of fertilizers and plant protection agents, which have a tendency to accumulate in the environment and potentially pose risks to both human health and the ecosystem. To address toxicity concerns, it is advisable to follow recommended pesticide application levels, adopt integrated pest management techniques, establish structured treatment regimens, and refrain from using highly hazardous products.

Author's contributions

TN (Professor of Agronomy) supervised the research work. GH (Post Graduate Student) carried out laboratory work, analysed data, and writing original draft. OAT (Senior Researcher) application of Software, Formal analysis, and Data curation. GH & MA (Professors of Agronomy) advised about the laboratory technique and conducted manuscript proofreading before submission. DA (Post Graduate Student) carried out laboratory work. All authors read and approved the final version of the manuscript.

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