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BIOACCUMULATION OF HEAVY METALS IN GARLIC BULBS (*ALLIUM SATIVUM* L.) IN CORRELATION WITH SOIL FROM PRIVATE GARDENS IN THE COPȘA MICĂ AREA, ROMANIA

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ABSTRACT. Heavy metal contamination of soil and plants is a worldwide concern. Copșa Mică in Romania is one of the areas in the country known to have a high level of pollution. Even if metallurgical activities have been reduced or stopped the soil is still polluted. This study aimed to estimate the bioaccumulation of heavy metals (Cd, Pb, Zn and Cu) in soil from garlic bulbs (*Allium sativum* L.) from individual gardens in the Copșa Mică polluted area, Romania. The bioaccumulation of heavy metals in garlic bulbs was estimated based on a data set collected from 44 individual gardens. The value of the linear correlation coefficient between the total cadmium content in the soil and that in the plant was significantly different from zero, indicating a close correlation between the two variables ($r = 0.775^{***}$). In addition, for zinc ($r = 0.649^{***}$) and lead ($r = 0.423^{**}$), simple power-type regressions were found to be best for estimating the bioaccumulation of these elements in garlic bulbs. Only for copper, the value of the linear correlation coefficient was not significantly different from zero ($r = 0.274^{ns}$), indicating that the estimation of copper accumulation in garlic cannot be described by simple power-type regressions. The results of this study are important for estimating the accumulation of heavy metals in garlic bulbs (head), which are often consumed by the population.

Keywords: heavy metals; soil; garlic; pollution; bioaccumulation.

INTRODUCTION

Numerous studies have shown that the accumulation of heavy metals in various vegetables constitutes a serious

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danger to human health and is associated with great concern (Ali *et al.*, 2021; Gebeyehu and Bayissa, 2020; Gupta *et al.*, 2021; Khan *et al.*, 2015; Nimyel and Chundusu, 2021). Heavy metal toxicity is a serious danger because it can lead to cardiovascular system disorders, kidney stones and damage to neurons in the brain (Bathla and Jain, 2016). Food safety and security issues are seen by many countries as key factors for sustainable development. According to Rai *et al.* (2019), “Conventional anthropogenic contaminants pose major risks to human health through dietary consumption of contaminated food crops through root transfer from soil to plant tissues”. A high concentration of heavy metals strongly affects the soil, resulting in a high level of heavy metal toxicity (Gupta *et al.*, 2021).

Copșa Mică in Sibiu County is one of the known areas in Romania with a high level of pollution. The main sources of pollution in the Copșa Mică area are due to the industrial activities of two economic agents: SC SOMETRA SA, with a profile of non-ferrous (zinc, lead, cadmium) and ferrous metals, which, before 1990, was considered the largest unit profile from the country, and SC CARBOSIN SA with a chemical profile (Vrînceanu, 2009). Even though metallurgical activities have been reduced or stopped, the soil remains polluted, and the local communities continue to carry out their agricultural activities, which can harm the health of the local people. Heavy metal soil pollution affected about 8400 ha of a total studied area of 8500 ha in Copșa Mică. In the area, the highly polluted surfaces amount to around 2257 ha, of which 1500 ha are polluted with Cd, 422 ha with Zn and 335 ha with Pb. Another 33 ha are highly polluted with Cu (Gamenț *et al.*, 2010).

Garlic (*Allium sativum* L.) is one of the most widely used plants for both culinary and medicinal purposes. It is the most used bulb crop, after onions. It is widely used as a spice in many foods (Tegegne and Mengiste, 2016a, b; Addis and Abebaw, 2017; Ata *et al.*, 2013; Phuengphai *et al.*, 2022).

Some studies have shown that intercropping garlic with other plants (*P. vittata, Conyza canadensis* and *Lolium perenne*) alters heavy metal uptake and bacterial diversity in neighbouring plants. This leads to a higher accumulation in roots and lower in shoots of Pb and Cd, which indicates that interplanting could be a good phytoremediation that can achieve a higher metal extraction rate in plants compared to cultivation a single plant (Hussain *et al.*, 2021).

Due to its greater mobility compared to other heavy metals, cadmium is easily transmitted through soil and plants (Mawari *et al.*, 2022).

The paper presents a study carried out during 2021–2022 regarding the bioaccumulation of heavy metals in garlic bulbs (head) in correlation with the polluted soil in the Copșa Mică area located in central Romania. The area is recognized as having a high degree of historical pollution due to the Copșa Mică industrial platform, namely, the SC SOMETRA SA and CARBOSIM SA factories, which stopped their activity, but the source of pollution remains. The consumption of vegetables, especially garlic, poses a risk to human health because it is not subject to control according to EU directives.

MATERIALS AND METHODS

The bioaccumulation of heavy metals in garlic bulbs (*Allium sativum* L.) was estimated from soil and plant samples collected from 44 individual households, located in the seven localities marked in *Figure 1*. The samples were analysed with instruments and methods reported previously (Vrînceanu *et al*., 2022). The statistical analyses were conducted in Excel 2002.

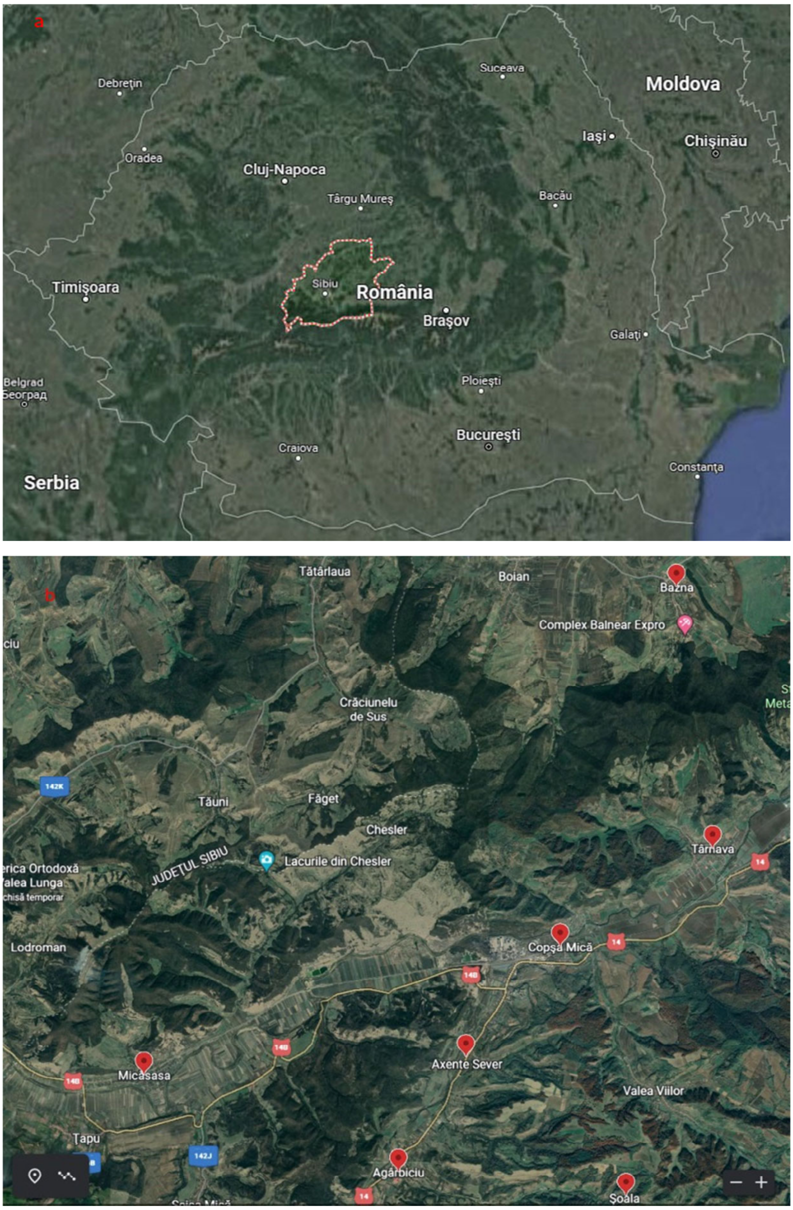


Figure 1 - The location of Sibiu County within Romania (a) and the localities studied in the Copsa Mica area (b)
(Source: Google maps).

RESULTS

The results of the study carried out in the Copșa Mică area regarding the accumulation of heavy metals in the soil are presented in *Table 1*.

The content of cadmium in the soil at depths of 0–20 cm varies between 0.10 mg·kg^{-1} (minimum value) and 27.11 mg·kg^{-1} (maximum value), with a coefficient of variation of 99.8%. Lead values ranged between 17 mg·kg^{-1} and 770 mg·kg^{-1}, with a median of 130 mg·kg^{-1} and a coefficient of variation of 89.0%, and zinc values were between 71 mg·kg^{-1} (minimum value) and 1472 mg·kg^{-1} (maximum value), with a coefficient of variation of 72.0%. The values of the average content of Cd, Pb and Zn exceeded the alert threshold for sensitive land use according to Order 756/1997. The content of Cu in the soil varied between 25 mg·kg^{-1} and 163 mg·kg^{-1}, with an arithmetic mean of 65.5 mg·kg^{-1} and median of 61.5 mg·kg^{-1}, which were below the alert threshold.

Garlic was one of the most common vegetables in the individual households included in the study.

The cadmium content of garlic bulb samples varied from 0.005 mg·kg^{-1} (minimum value) to 0.671 mg·kg^{-1} (maximum value), with a median of 0.092 mg·kg^{-1} and with a coefficient of variation of 104.8%. Lead content ranged from 0.014 to 0.230 mg·kg^{-1}, median (0.081 mg·kg^{-1}), with a coefficient of variation of 57.0%. The zinc values in garlic bulbs were between 3.1 mg·kg^{-1} (minimum value) and 18.4 mg·kg^{-1} (maximum value), with a coefficient of variation of 42.4%. Copper ranges from 0.45 to 2.09 mg·kg^{-1}with a coefficient of variation of 34.2 % (*Table 2*).

Table 1 - Values of statistical parameters that characterize the central tendency and the variability of the total cadmium, lead, zinc and copper contents in soil (layer 0–20 cm)

Variable	Minimum	Maximum	Median	Geometric mean	Arithmetic mean	Standard deviation	Coeficient of variation
Cd_{soil}	0.10	27.11	4.90	2.83	5.94	5.93	99.8%
Pb_{soil}	17	770	130.5	119.3	168.9	150.4	89.0%
Zn_{soil}	71	1472	391.5	361.2	457.1	329.3	72.0%
Cu_{soil}	25	163	61.5	60.4	65.5	27.2	41.5%

Table 2 - Values of statistical parameters that characterize the central tendency and the variability of the total cadmium, lead, zinc and copper contents in the garlic bulbs

Variable	Minimum	Maximum	Median	Geometric mean	Arithmetic mean	Standard deviation	Coeficient of variation
Cd_{garlic}	0.005	0.671	0.092	0.073	0.125	0.131	104.8%
Pb_{garlic}	0.014	0.230	0.081	0.083	0.100	0.057	57.0%
Zn_{garlic}	3.1	18.4	8.0	7.5	8.2	3.48	42.4%
Cu_{garlic}	0.45	2.09	1.14	1.10	1.17	0.40	34.2%

According to EU Regulation 2021/1323, the maximum allowed level of cadmium in garlic bulbs is 0.05 $mg \cdot kg^{-1}$. In the studied area, the cadmium content in garlic bulbs varied between 0.005 $mg \cdot kg^{-1}$ (minimum value) in Bazna 9 and 671 $mg \cdot kg^{-1}$ (maximum value) in Copșa Mică 2, which is near the pollution source and presents a level that is far above the maximum allowed (*Figure 2a*). In 15 individual households out of the 44 studied the cadmium content values were below the maximum concentration, varying between 0.005 $mg \cdot kg^{-1}$ and 0.045 $mg \cdot kg^{-1}$. Cd varied between 0.058 $mg \cdot kg^{-1}$ and 0.093 $mg \cdot kg^{-1}$, then increased from 0.107 $mg \cdot kg^{-1}$ in Axente Sever15 to 0.145 $mg \cdot kg^{-1}$ in Micăsasa 1, which was above the maximum allowed concentration. Cd content increased to two times above the maximum allowed concentration, from 0.154 $mg \cdot kg^{-1}$ in Axente Sever 6 to 0.172 $mg \cdot kg^{-1}$ in Copșa Mică 6. The closer we sampled to the source of pollution, the Cd content increased from 0.209 $mg \cdot kg^{-1}$ (minimum value) in Copșa Mică 12 to 0.971 $mg \cdot kg^{-1}$ (maximum value) in Copșa Mică 2.

According to EU Regulation 2021/1317, the maximum permissible limit of lead content in root and bulb vegetables is 0.10 $mg \cdot kg^{-1}$. Based on *Figure 2b*, the minimum value was 0.014 $mg \cdot kg^{-1}$ in Bazna 9 and the maximum value was 0.230 $mg \cdot kg^{-1}$ in Axente Sever 6, which shows that in more than half of the collection areas the lead content was below the maximum limit. Thus, in the other localities the values were between 0.107 $mg \cdot kg^{-1}$ (minimum value) in Târnava 6 and 0.230 $mg \cdot kg^{-1}$ (maximum value) in Axente Sever 6.

Logarithmic plots for power-type regression curves estimating the stochastic dependence between total soil Cd, Pb, Zn and Cu contents in soil and garlic are shown in *Figure 3*.

The linear correlation coefficient ($r = 0.775^{***}$) indicates a significant relationship between soil and plant cadmium content. In addition, for lead with a linear correlation coefficient $r = 0.423^{**}$ and for zinc $r = 0.649^{***}$, simple power-type regressions were found to be the best for estimating the bioaccumulation of these elements in garlic heads. Regarding the accumulation of copper in the studied plant, the linear correlation coefficient was not significantly different from zero ($r = 0.274^{ns}$), showing that this process cannot be described by power type regressions.

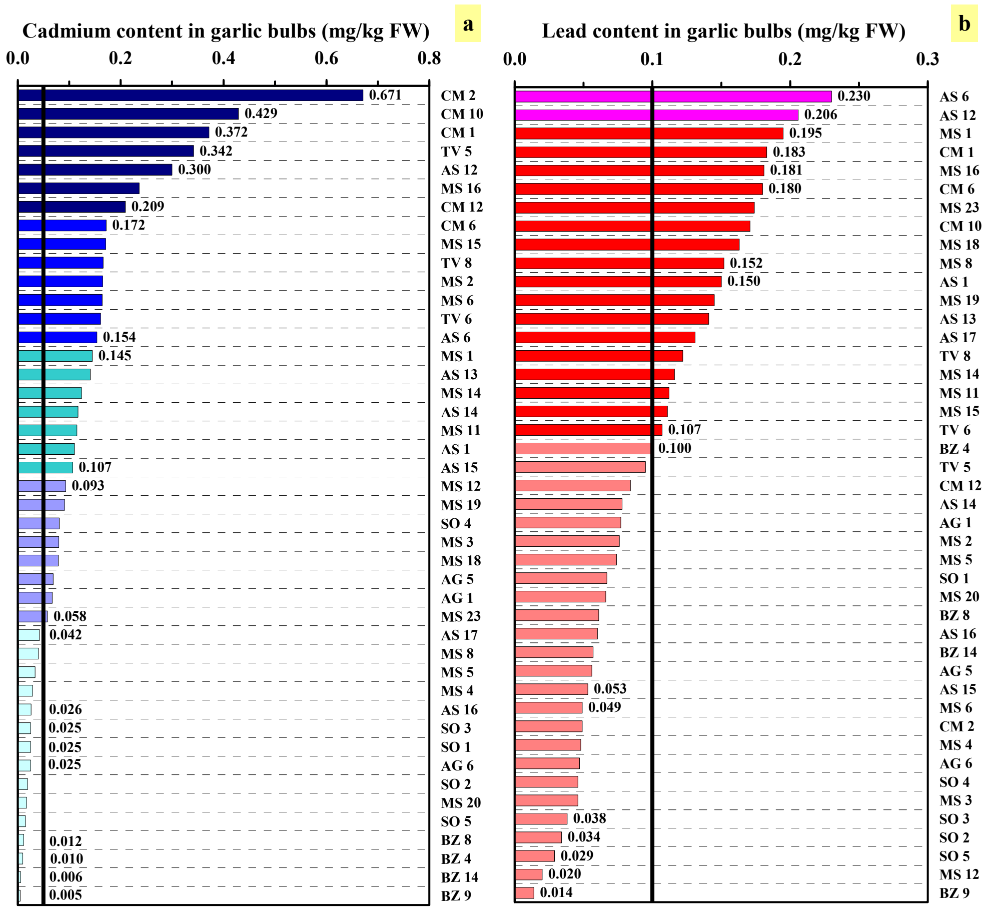
DISCUSSION

According to Commission Regulation (EU) 2021/1323, Law 228/13 and Law 286/1, Cd content in garlic bulbs in more than half of the samples exceeded the maximum permissible limit (0.05 $mg \cdot kg^{-1}$).

As can be seen from *Figure 1b*, in half of the analysed samples, the lead content was below the maximum limit allowed according to Regulation (EU) 2021/1317 (0.10 $mg \cdot kg^{-1}$).

The results carried of Tegegne and Mengiste (2016b) showed that the concentration of copper ions in garlic bulbs varied between 4.21 and 7.16 mg/kg, with a significant difference between the means, and from 5.41 to 8.44 in leaves, and the zinc level was in the range of 31.17–35.39 $mg \cdot kg^{-1}$ in bulbs and 49.10–71.39 mg·kg^{-1}in garlic leaves. Mean lead concentrations ranged from 2.02 to 1.07 mg·kg^{-1} in bulbs and 1.87 to 2.84 mg·kg^{-1} in leaves. Cadmium was between 0.16 mg·kg^{-1} (maximum value) and 0.10 mg·kg^{-1} (minimum value) in garlic bulbs and 0.17 and 0.12 in garlic leaves.

Thus, the authors found that there were higher concentrations of heavy metals accumulated in the leaves than in the bulbs of garlic. Also, other authors reported that a higher concentration had accumulated in the leaves and stem compared to the roots. According to several authors, the translocation of heavy metals from the soil to the plant is controlled by soil and plant pH (Atsarou and Economou, 2012; Shibdawa *et al.*, 2019; Street, 2010).



——— Maximum level of cadmium*/lead in garlic bulbs** **FW** - **F**resh **W**eight

* **Commission Regulation (EU) 2021/1323 of 10 August 2021**

** **Commission Regulation (EU) 2021/1317 of 9 August 2021**

Notes: **AS** - Axente Sever, **AG** - Agârbiciu, **SO** - Şoala (villages of Axente Sever commune)
BZ - Bazna, **MS** - Micăsasa, **TV** - Târnava (communes in Sibiu County)
CM - Copşa Mică (town in Sibiu County)

Figure 2 - Cadmium and lead contents in garlic bulbs harvested from the Copşa Mică area

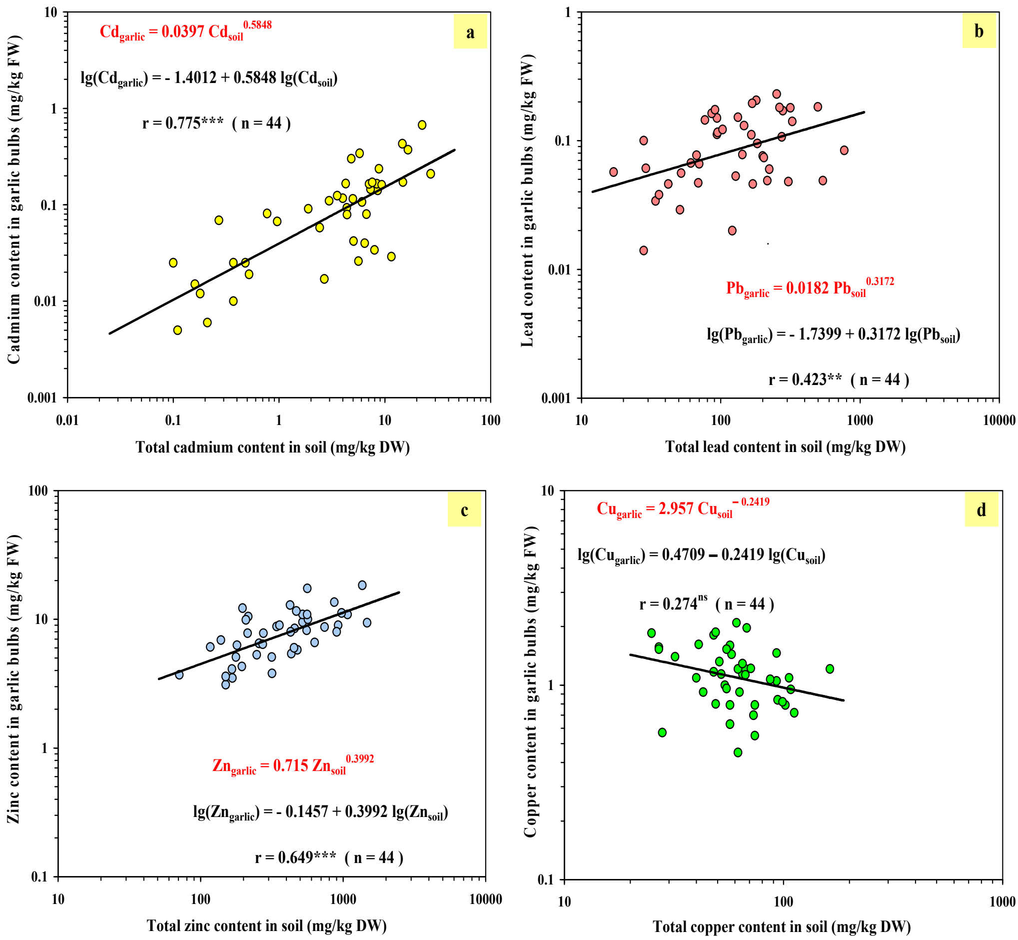


Figure 3 – Stochastic dependency between total heavy metal content in soil and garlic bulbs

Another study showed that in garlic (*A. sativum*) grown hydroponically with cadmium chloride and sodium hydrosulfide, Cd caused growth inhibition and reduced biomass with high accumulation in roots and shoots (Gharehbaghli and Sepehri, 2022). Absorption and accumulation of Cd in roots, bulbs and shoots of garlic varied with Cd concentration and increased with increasing the concentration of cadmium (Jiang *et al.*, 2001).

The results presented by Bakht (2016) showed that different vegetables (including garlic) have heavy metal contents above the maximum allowed limits, which can affect human health.

Consistent with the results published by Liu *et al.* (2009), the influence of lead concentrations on the growth and development of *A. sativum* L. plants, as well as its accumulation mainly in the roots and less in the bulbs and shoots, were noticed.

Copper ion concentrations ranged from 0.45 to 2.09 mg·kg^{-1}. According to Akhter *et al.* (2022), the range of the health risk indexes of *A. sativum* in differently treated pots was between 0.0006 and 2.89. Health risk index values of the treatments were in the following order: Cr < Mn < Fe < Co < Cu < Zn < Pb < Cd.

Studies conducted by Kamyabi *et al.* (2021) showed that garlic waste (garlic peel and stem) was used as a bio-adsorbent for the biosorption of Hg(II) ions. As the dose of garlic was raised, the effectiveness of Hg(II) absorption improved. Garlic garbage may work well as a bio-adsorbent to remove mercury from contaminated waters. Shaikhiev (2022) presented the use of ground garlic peels, leaves and roots as sorption materials to remove various metal ions such as As^{3+}, Cd^{2+}, Cr(VI), Cu^{2+}, Fe^{2+}, Hg^{2+}, Mn(VII), Ni^{2+}, Pb^{2+}, Sn^{2+} and W(VI) and dyes and antibiotics from aqueous media. Liu et al. (2014) suggested a comparison study utilizing native and mercerized garlic peel as adsorbents for Pb^{2+} removal and discovered that mercerized garlic peel can be a more effective adsorbent because of its quicker absorption capability.

CONCLUSIONS

Garlic is one of the most common and consumed vegetables in individual households in the study area.

A statistically significant difference from zero of the bioaccumulation of total cadmium concentration from soil to plant indicates a strong link between the two variables. Simple power regressions worked best for calculating the bioaccumulation of lead and zinc in lightbulbs (garlic head). The value of the linear correlation coefficient was only statistically different from zero for copper, showing that basic power regressions cannot adequately represent the estimation of copper accumulation in garlic.

Author Contributions: Conceptualization VC, BSO; methodology VC, DMM, GP, MC; analysis DMM, NOV, GP, MC, BSO, VT; writing VC, BSO, VT, review BSO, VT; supervision VC, VT. All authors declare that they have read and approved the publication of the manuscript in this present form.

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Conflicts of Interest: There are no conflicts of interest associated with this study or work.

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