Phytoremediation and Long-term Metal Uptake Monitoring of Silver, Selenium, Antimony, and Thallium by Black Pine (*Pinus nigra* Arnold)

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The usability of black pine (*Pinus nigra* Arnold) in both monitoring the changes in the concentrations of silver (Ag), selenium (Se), antimony (Sb), and thallium (Tl), and in reducing soil or air pollution was investigated. In the study, annual rings of a *Pinus nigra* tree, cut in 2023 and identified as 356 years old, were grouped into 10-year intervals, and then the changes in these heavy metals throughout the process were determined by analysing the concentrations of these elements. Additionally, the relationship of these elements with other elements was also detected in the scope of the study. The study results suggested that *Pinus nigra* was not a suitable bio-monitor for monitoring the changes in Ag, Se, Sb, and Tl concentrations in the soil or air, but it was a highly suitable species for phytoremediation studies aimed at reducing the pollution of these elements. Moreover, it was determined that the relationships of the studied elements with essential nutrients, such as Mg, Ca, P, K, Al, Zn, Ni, and Fe, were not statistically significant and were very weak; whereas they exhibited positive and very strong relationships with elements known to be highly harmful for health such as V, Pb, and As.

DOI: 10.15376/biores.19.3.4824-4837

Keywords: Heavy metal; Bio-monitors; Phytoremediation; Pinus nigra Arnold

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INTRODUCTION

The industrial revolution and technological advancements occurring around the world in the past century have had a profound impact, causing radical changes in almost every domain of study. While the industrial revolution enabled rapid increases in production, it also escalated the demand for raw materials, energy, and labour. To meet the demand for labour, rural-to-urban migration has occurred, leading to the concentration of populations in specific areas and raising the issue of urbanization (Dogan *et al*. 2022; Key *et al*. 2023). Further, the use of fossil fuels to meet the required energy needs has significantly increased the atmospheric CO_2 levels and become a major contributor to global climate change (Sevik *et al*. 2015; Tekin *et al*. 2022). The consequences of this process, which are urbanization and global climate change, have been defined as irreversible problems worldwide (Varol *et al*. 2022; Zeren Cetin *et al.* 2023)

One of the most essential impacts of this process is environmental pollution, particularly air pollution. The extraction and utilization of elements from underground reserves as raw materials for industrial production have led to the release of many harmful elements into the environment. Most of these elements are heavy metals that are likely to pose serious threats to human and environmental health even at low concentrations (Erdem *et al.* 2023; Sulhan *et al.* 2023). The issue of heavy metal-induced air pollution has reached such serious levels that it is reported that 90% of the world's population now breathes polluted air. In addition, air pollution causes almost 6 million premature births, almost 3 million underweight babies and around 7 million premature deaths worldwide each year (Isinkaralar *et al.* 2022; MedicalNewsToday 2024; WHO 2024).

Most heavy metals are highly hazardous to human and environmental health even at low concentrations. However, among heavy metals, some are significantly more hazardous and harmful than others, causing them to be included in priority pollutant lists by international organizations such as Agency for Toxic Substances and Disease Registry (ATSDR) and Environmental Protection Agency (EPA). The elements subjected to this study, namely silver (Ag), selenium (Se), antimony (Sb), and thallium (Tl), are included in the priority pollutant lists of both ATSDR and EPA due to their potential hazards (Savas *et al.* 2021).

Among these elements, silver (Ag) is highly toxic to mammalian cells. It may cause damage to brain cells, liver cells, and stem cells as well as skin diseases such as argyria or argyrosis. Silver is also extremely toxic to fish, algae, certain plants, fungi, crustaceans, and bacteria (Panyala *et al*. 2008). Thallium (Tl) is an element that is not essential for humans, and the human body does not have a biological use for it. It is considered one of the most poisonous metals, posing a significant threat to human and environmental health and being more toxic to humans than mercury, cadmium, lead, copper, or zinc (Blain 2022). Negative impacts of Tl pollution emerge as symptoms including exhaustion, muscle and joint pains, vision impairment, hair loss, and chronic Tl poisoning (Xiao *et al*. 2012). Thallium poisoning can lead to gastrointestinal dysfunction, ascending paralysis, and mental disorders, while chronic poisoning may result in polyneuritis (Duri *et al.* 2020). Selenium (Se) poisoning can show acute symptoms, such as vomiting, pain, nausea, and abdominal symptoms, as well as garlicy breath and heart symptoms. In cases of severe toxicity, cardiac and pulmonary symptoms may occur, which then potentially results in death (Hadrup and Ravn-Haren 2020).

It is reported that exposure to antimony (Sb), an important and widespread heavy metal, may change the hormone levels in the human body, and may increase the risk of cancer (Lai *et al.* 2022). Further, Sb is said to cause symptoms such as conjunctivitis, upper respiratory tract inflammation, chronic bronchitis, chronic emphysema, and pleural adhesions (Gad 2014). Due to these effects, Ag, Se, Sb, and Tl are included in the hazardous heavy metals list alongside other heavy metals, like As, Cr, Cd, and Hg, by ATSDR and EPA.

It is also noted that the inhalation of heavy metals into the body through the respiratory system can lead to much more serious health problems (Ghoma *et al.* 2023). Thus, monitoring the changes in heavy metal concentrations in the air and reducing pollution are of great importance in terms of human health. Direct determination of heavy metal pollution in the atmosphere is not widely preferred due to its costliness and the inability to directly assess the impact of atmospheric pollution on ecosystems, and instead biomonitoring is used for this purpose, in general (Karacocuk *et al.* 2022).

Some of the heavy metals in the air enter the plant body through the leaves during respiration. Some of them enter directly into the plant by adhering to the plant organs with the help of particulate matter. Some of them mix into the soil and water with the effect of rain and gravity. This portion can enter the plant body from water and soil through the roots. Thus, heavy metal pollution in air, water, and soil is reduced (Shahid *et al.* 2017;

Hlihor *et al.* 2022). Heavy metals entering the plant body may change form, form compounds with other elements, or degrade in the process. Thus, heavy metal pollution is reduced in a long process. These studies are called phytoremediation studies (Tangahu *et al.* 2011; Asati *et al.* 2016). Thus, phytoremediation, the study of how plants may reduce heavy metal pollution in the air, have been carried out for a long time. However, mosses, lichens, and herbaceous plants are generally used in these studies (Duan *et al.* 2023). The reason for this is that these plants can accumulate a lot of heavy metals in their bodies (Sevik *et al.* 2019). However, herbaceous plants are tiny in mass compared to trees. The mass of an adult tree is much greater than the combined mass of tens of thousands of herbaceous plants. In addition, herbaceous plants die quickly, and the heavy metals they contain are mixed with the soil. However, heavy metals accumulated in the wood of trees can remain away from nature for hundreds of years. Therefore, trees can be used more effectively in phytoremediation studies (Wei *et al.* 2021; Duan *et al.* 2021; Koç *et al.* 2024).

In this study, it was aimed to determine the usability of *Pinus nigra* in monitoring and reducing the concentrations of the extremely harmful heavy metals, such as Ag, Se, Sb, and Tl, in the air, by taking their impact on human and environmental health into consideration. Additionally, an attempt was made to determine the relationship of these elements with other elements.

EXPERIMENTAL

The study was conducted on black pine (*Pinus nigra* Arnold) trees growing in Kastamonu province. Black pine is an important forest tree for Türkiye and the world. However, it is also used extensively in landscaping studies due to its features, such as being able to grow in dry and cold areas, being durable, being evergreen, requiring little water and maintenance, and having aesthetic value due to its monopodial appearance (Topacoglu *et al.* 2016; Akkemik 2018; Yigit *et al.* 2023). For this reason, black pine is also heavily exposed to pollution factors. Urban areas only account for 4% of the global land area but provide space for 90% of land human activities (Duan *et al.* 2021).

The area where the studied tree grew is in the İhsangazi district of Kastamonu. No heavy traffic or urban pollution were present in the area where the tree grew. However, a large iron and steel factory is approximately 75 km away. The factory in question started operating in 1939 (Yurtoglu 2017). Studies have shown that air pollution has increased rapidly in the last century in parallel with the developments in industry and technology (Istanbullu *et al.* 2022; Özel *et al.* 2024). The study determined the changes in heavy metal concentrations in the air since 1678. The fact that the tree is so old is important because it reveals the connection between the change in heavy metal pollution in the air and the industrial revolution.

At the end of the 2023 vegetation season, a sample log was obtained by cutting approximately 50 cm above the ground. To clearly observe the annual rings in the log sample, it was wiped clean, and the annual rings were counted. The tree was identified as being 356 years old, and considering the widths of the annual rings, it was grouped into ten-year age ranges from outer to inner (from recent to old). Using a steel drill bit for each age range, samples were taken from the wood in three directions. After the collected samples were placed in glass petri dishes and air-dried at room temperature for 15 days, they were dried in an oven at 45 °C for an additional week (Key *et al.* 2023; Erdem *et al.* 2024; Koç *et al.* 2024).

The dried samples, each weighing 0.5 g, were taken, and 6 mL of 65% HNO_3 and 2 mL of 30% H_2O_2 were added. Each specimen was then placed in a microwave oven designed for the specified analyses. Once the samples turned into a solution, they were transferred to balloon flasks and filled up to 50 mL with ultra-pure water. The prepared samples were analysed using an ICP-OES device, and the obtained values were multiplied by the dilution factor to calculate the concentrations of Ag, Se, Sb, and Tl. Furthermore, other element analyses were performed within the study to determine the relationship levels of these elements with other elements. The method used in the study is a commonly employed technique in recent studies on this subject (Isinkaralar *et al*. 2022; Key *et al*. 2022). Variance analysis was applied to the data using the SPSS package program, and for factors with statistically significant differences at a minimum of 95% confidence level ($p < 0.05$), the Duncan test was applied. Moreover, correlation analysis was conducted using the SPSS package program to identify the relationship levels between the elements and other elements.

RESULTS AND DISCUSSION

The average values and statistical analysis results regarding the periodic changes in Ag, Se, Sb, and Tl concentrations in *Pinus nigra* wood are given in Table 1. The concentration of Ag changed between 3626 ppb and 5649 ppb. The highest Ag concentration was approximately 1.56 times the lowest value. The lowest Ag concentrations were obtained between the years 1734 and 1763, while the highest Ag concentration was obtained in the immediately following period of 1774 and 1783. Similar changes are observed for other elements as well. According to the study results, it was found that the concentration of Se varied between 2719 ppb and 4615 ppb, Tl concentration between 2014 ppb and 3179 ppb, and Sb concentration between 6332 ppb and 9999 ppb. The calculated differences between the lowest and highest concentrations were approximately 1.7 times for Se and 1.58 times for both Sb and Tl. When examining the changes in elemental concentrations from the past to the present, a similar trend is observed for all element concentrations. There is a slight decrease in concentrations until the period of 1764 and 1773, followed by a relatively stable period, whereas a significant increase is noted during the period of 1774 and 1793. The most striking result obtained within the study was the accumulation of Se, Ag, Tl, and Sb concentrations from the black pine wood within detectable limits. Because the elements under study are considered extremely harmful to human and environmental health, they are included in the priority pollutant lists of both ATSDR and EPA. In addition, these elements are naturally found in very low amounts, and their concentrations in nature increase due to anthropogenic effects, like in other heavy metals. Further, studies have shown that the most significant sources of heavy metals are anthropogenic sources (Ucun Ozel *et al.* 2019) including industrial activities (Istanbullu *et al.* 2023), mining activities (Kuzmina *et al*. 2023), and traffic (Aricak *et al.* 2019). Plants, especially trees with high biomass, are considered among the most effective instruments in reducing heavy metal pollution in air, soil, and water (Koc *et al.* 2023). However, studies conducted indicate that the concentrations of various heavy metals, similar to the elements within the scope of the study, remain below detectable limits in bio-indicator plants (Key *et al*. 2022).

Table 1. Periodic Changes in Ag, Se, Sb, and Tl Concentrations

Date Range	Ag (ppb)	Se (ppb)	Sb (ppb)	Tl (ppb)
2014-2023	4419.3cd	2801.2ab	6631.7b	2226.3bc
2004-2013	4564.7de	3038.3cd	7372.1d	2221.8bc
1994-2003	4707.1efg	3117.3de	7376.6d	2267.5bc
1984-1993	5244.4ijk	3399.8f	7989.7e	2430.2def
1974-1983	5098.7hi	3390.3f	8023.6e	2447.5ef
1964-1973	5137.8hij	3462.2fg	8260.1efg	2489.2ef
1954-1963	5300.9jklm	3566.2g	8467gh	2516.8ef
1944-1953	5051.5h	3806.4h	9460.8lmno	2722.5g
1934-1943	5262.3ijkl	3892.4h	9221.3kl	2710.6g
1924-1933	5439.9lmnop	4360.9lmn	9531.1mno	3070.5jklmn
1914-1923	5526.1opq	4548.2op	9500.2lmno	3125.8klmn
1904-1913	5622.6pq	4236.4jkl	9299.1klm	3016.4hijkl
1894-1903	5558.9pq	4384.9mn	9297.7klm	3046.5ijklm
1884-1893	5460.9mnopq	4199.1ijk	9282.4klm	3003.2hijk
1874-1883	5560.2pq	4198.9ijk	9547.6mnop	2982.8hij
1864-1873	5572.1pq	4480.9nop	9592.1nop	3178.7n
1854-1863	5534.6opq	4614.6p	9372.4klmn	3174.8n
1844-1853	5338.8klmn	4482nop	9105.7jk	3129.1lmn
1834-1843	5306.6jklm	4229.9jkl	9322.6klmn	3038.1hijklm
1824-1833	5482.2mnopq	4070.5i	9375.5klmn	2947hi
1814-1823	5596.7pq	4106.2ij	9494.4lmno	2924.4h
1804-1813	5360.4klmno	4386mn	9703op	3064.2ijklmn
1794-1803	5505.6nopq	4425.2mno	9806.3pq	3057.7ijklmn
1784-1793	5339.3klmn	4417.9mno	9999.4q	3153mn
1774-1783	5649.1q	4282.7klm	9999.4q	3120.1klmn
1764-1773	3625.8a	2718.9a	6332.3a	2024.3a
1754-1763	3883.9b	3055.6d	6932.2c	2049.9a
1744-1753	4039.2b	2905.2bc	6945.2c	2014.3a
1734-1743	4304.8c	3217.8e	7489.4d	2199.5b
1724-1733	4595.7ef	3416.9f	8084.6ef	2323.2cd
1714-1723	4733.5efg	3517.1fg	8298.2fg	2510ef
1704-1713	4795.5g	3463.9fg	8208.3efg	2397.2de
1694-1703	4761.8fg	3602.2g	8731.1hi	2530.5f
1684-1693	5007h	3610.9g	8730.1hi	2505.7ef
1678-1683	5032.1h	3858.1h	8938.5ij	2652.3g
F Value	88,317	141,748	131,304	106,281
Sig.	0,000	0,000	0,000	0,000

According to Duncan's test results, numbers followed by the same letters are not statistically different at P > 0.05.

Especially, the wood part of plants is usually the part where heavy metals accumulate the least (Karacocuk *et al.* 2022). In fact, plants that can accumulate heavy metals in the wood part are crucial in phytoremediation studies, because the wood part is usually the largest organ of the plant in terms of mass (Koc *et al.* 2023; Key *et al.* 2023). The study results show that Se, Ag, Tl, and Sb concentrations in black pine wood were quite high, and thus suggesting that this species is suitable for phytoremediation studies to reduce the pollution of these elements.

Later, another relatively stable but slightly decreasing trend was observed. This result indicates a potential correlation among these elements. For this reason, the correlation between the elements was determined, and the correlation analysis results are provided in Table 2.

Table 2. Results of Correlation Analysis among Heavy Metals

	Se	Ag	Tl	Sb
Se	1	0.864**	0.970**	0.944**
Ag	0.864**	1	0.882**	0.887**
Tl	0.970**	0.882**	1	0.934**
Sb	0.944**	0.887**	0.934**	1
Mg	0.104	0.077	0.077	0.227*
Ca	0.054	0.209*	0.033	0.285**
K	0.058	0.109	0.073	0.144
S	0.297**	0.566**	0.358**	0.403**
Al	-0.033	0.013	-0.026	0.032
As	0.966**	0.779**	0.948**	0.885**
Ba	-0.326**	-0.287**	-0.320**	-0.204*
Bi	0.778**	0.904**	0.846**	0.789**
B	-0.877*	-0.844*	0.145	-0.963**
Cd	0.510**	0.658**	0.580**	0.435**
Cr	0.034	-0.073	0.057	0.084
Co	0.226*	0.330**	0.276**	0.345**
Cu	-0.449**	-0.203*	-0.350**	-0.467**
Ga	0.841**	0.984**	0.849**	0.879**
In	0.909**	0.943**	0.909**	0.970**
Fe	-0.177	-0.121	-0.172	-0.221*
Pb	0.907**	0.948**	0.929**	0.924**
Li	0.618**	.0873**	0.667**	0.640**
Mn	-0.259**	-0.217*	-0.247*	-0.044
Ni	-0.026	-0.074	-0.011	0.114
P	-0.083	0.212*	-0.033	-0.041
V	0.818**	.941**	0.836**	0.872**
Zn	0.070	0.164	0.113	0.073

*Significant at 0.05 level; **significant at 0.01 level

When the levels of relationships among the elements were examined, it was observed that the elements, within the scope of the study, had statistically significant and very strong relationships with each other. While the weakest relationship was observed between Se and Ag (0.864), the strongest relationship was observed between Se and Tl (0.970). It is noteworthy that even the weakest relationships were profoundly strong. Additionally, all relationships were seen to be positive. When the relationships of the elements under study with other elements were examined, it was observed that they do not show statistically significant relationships ($p > 0.05$) and had very weak correlations with essential nutrient elements, such as Mg, Ca, P, K, Al, Zn, Ni, and Fe, whereas there were positive and very profound relationships with elements known to be highly harmful to health, such as V, Pb, As, and In. In this context an important result obtained in this study is that the concentrations of all the elements, under study, in adjacent wood samples were at very close levels. For a species to be utilized as a bio-monitor in monitoring the changes in heavy metal pollution, the transfer of elements between tissues in the wood parts should be limited. The displacements of elements within the wood change depending on the relationship between the element and the structure of the wood, in other words the tree species. In some studies conducted previously, different species were suitable bio-monitor plants for different heavy metals. For instance, *Cedrus deodora* was found to be a suitable bio-monitor for Cu (Zhang 2019), *Picea orientalis* for Tl (Canturk 2023), *Cedrus atlantica* for Ni, Cr, and Mn (Koc 2021; Savas *et al.* 2021), *Cupressus arizonica* for Cd, Ni, Cr, Tl, Fe, and Zn (Cesur *et al.* 2021, 2022; Canturk 2023; Cobanoglu *et al.* 2023; Koc *et al.* 2023), *Corylus colurna* for Cd, Ni, Zn, Co, Pb, Cr, Mn, and Zn (Key *et al.* 2022; Key and Kulac 2022; Key *et al.* 2023), and *Pseudotsuga menziesii* for Cr (Koc *et al.* 2023), and indicated that the displacement of these elements in the wood of these species were limited. However, in some studies, it is mentioned that Pb and Zn can be displaced within *Cedrus deodora* (Zhang 2019), Co within *Cedrus atlantica* (Koc 2021), and Bi, Li, and Cr within *Cupressus arizonica* (Cesur *et al.* 2021, 2022; Cobanoglu *et al.* 2023), indicating that these species are not suitable for being utilized as bio-monitor plants.

The entry of heavy metals into plant tissues may occur through the roots from the soil, through the leaves from the air, and directly through adsorption from stem parts (Key *et al.* 2022). In contrast, the transfer of elements within the wood is mainly related to the structure of cell, especially the cell wall (apoplastic pathway). The apoplast, which is present between the cell wall and the plasma membrane (CWPM) in plants, not only acts as an apoplastic membrane barrier but also serves as a flexible structure that perceives and produces signals during metal/metalloid stress. Cell wall proteins (CWP) activate under various abiotic stresses (Wani *et al.* 2018). Plants often face stress factors throughout their lives, especially abiotic stress factors. The most common stress factors that plants face are climatic stress factors such as drought (Gur *et al.* 2024; Erturk *et al.* 2024), frost (Sevik and Karaca 2016; Yigit *et al.* 2021), UV-B (Ozel *et al.* 2021a), radiation (Ozel *et al.* 2021b), heavy metal (Isinkaralar *et al.* 2023), salinity (Nazari *et al.* 2023), toxins (Gull *et al.* 2019), nanoparticles (Özel *et al.* 2024). This is because plant development depends on the interaction between genetic structure (Hrivnak *et al.* 2023; Kurz *et al.* 2023) and environmental conditions (Sevik *et al.* 2021; Tandogan *et al.* 2023; Cetin *et al.* 2023a). For this reason, factors that cause significant and permanent changes in climatic parameters, such as global climate change, trigger stress mechanisms in plants (Varol *et al.* 2022; Tekin *et al.* 2022). In addition, factors, such as increased UV-B stress due to climate change (Ozel *et al.* 2021a), radiation generated by anthropogenic sources (Ozel *et al.* 2021b), and heavy metal pollution (Isinkaralar *et al.* 2023) are significant stress sources for plants. Cell wall proteins (CWP) activated under various abiotic stresses have been comprehensively identified and characterized among different plant species. In CWPM, main phospholipases, salt overly sensitive kinases (SOS), transcription factors, C-repeat binding factors, dehydration-sensitive element binding proteins, mitogen-activated protein kinases and phosphatases, and binding factors responding to abscisic acid are found. As CWPM interface accumulates a large portion of heavy metals, it is believed to be a potential part responsible for heavy metal tolerance (Wani *et al.* 2018).

The study results indicate that black pine trees are not suitable bio-monitors for monitoring the changes in the concentrations of the studied elements in the air. As a result of the study, the differences, not exceeding twice the amount, were found between the lowest and highest concentrations in the wood. However, studies conducted on this topic have shown significantly higher differences in tissues adjacent to each other. For instance, Canturk (2023) reported that in *Cedrus atlantica*, the concentration of Tl in the wood formed on the northern side was 9780 ppb during the same period, while it was 1483 ppb in the wood formed on the western side. Similarly, Cetin *et al*. (2023b) stated that in *Pinus pinaster*, the concentration of Sn in the wood formed on the eastern side was 3216 ppb during the same period, while it was 404 ppb on the southern side. In similar studies, the significant differences in element concentrations obtained in adjacent wood tissues have been interpreted as limited transfer of elements within the wood (Yayla *et al.* 2022). However, the results obtained in this study suggest that elements can indeed transfer within the wood.

The study results revealed that there was a profoundly strong and positive relationship between Se, Ag, Tl, and Sb elements. Additionally, it was also revealed that these elements had a positive and very strong relationship with other elements, such as V, Pb, As, and In, which are highly dangerous and harmful to human and environmental health. This finding particularly highlights that industrial and traffic-related sources may serve as simultaneous contributors to numerous harmful elements. In fact, previous studies have consistently identified industrial activities and traffic as major sources of heavy metal pollution (Savas *et al.* 2021). The material used in the study is far from roads and urban areas with heavy vehicle traffic, which shows that the main source of pollution may be the iron and steel factory, which is approximately 75 km away from the region. In fact, a study conducted on a tree 88 km away from this factory stated that heavy metal pollution originated from the factory in question (Key and Kulaç 2022).

CONCLUSIONS

1. Studies show that each heavy metal accumulates in different concentrations in different plants. Many plants are not able to accumulate various heavy metals, especially in the wood, and the concentrations of heavy metals in the wood of these plants remain below the detectable limits. Plants that cannot accumulate heavy metals in the wood at a certain level cannot be used for phytoremediation or as biomonitors. In this study, black pine trees (*Pinus nigra*) accumulated Se, Ag, Tl, and Sb concentrations within detectable limits, reaching significantly high levels. This finding suggests that black pine trees are suitable for phytoremediation studies aimed at reducing concentrations of these elements in the environment, including the air, soil, and water.

2. The elements under study may pose a serious threat to human and environmental health even at low concentrations, and thus reducing the concentrations of these elements in the environment is crucial. The use of black pine trees in landscaping projects in areas with high pollution levels could contribute greatly to reducing pollution.
3. The concentrations of the studied elements in the wood of black pine trees were quite similar in adjacent tissues, with differences between the lowest and highest values not exceeding twice the amount. This result indicates that the studied elements can transfer within the wood part of black pine tree. Therefore, the species studied may not be a suitable bio-monitor for identifying the changes occurring in the concentrations of these elements in the air over time.
4. The results also indicate a positive and profoundly strong relationship between the studied elements and elements such as V, Pb, As, and In. This result suggests that in areas, where the pollution levels of these elements are high, other harmful elements are also present at significantly high concentrations and thus radiating from similar sources. However, the number of studies conducted on this topic so far is quite limited. Further comprehensive studies are recommended to be conducted on this topic.

DATA AVAILABILITY

The datasets used and/or analyzed during the current study are available from the corresponding author upon reasonable request.

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Article submitted: February 11, 2024; Peer review completed: April 27, 2024; Revised version received: April 29, 2024; Accepted: May 2, 2024; Published: May 31, 2024.
DOI: 10.15376/biores.19.3.4824-4837