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Long-term Monitoring of Thallium Uptake and Phytoremediation Potential of *Robinia pseudoacacia*

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ABSTRACT

This study investigates the spatial and temporal variation of thallium (Tl) accumulation in different tissues (outer bark, inner bark, and wood) of Robinia pseudoacacia growing in the city center of Düzce, Türkiye, a region known for severe air pollution. Sampling was performed by sectioning the main trunk at approximately 50 cm above ground level, and Tl concentrations were analyzed using inductively coupled plasma optical emission spectroscopy (ICP-OES). Variance analysis revealed significant differences in Tl concentrations depending on organ type and directional exposure ($p < 0.001$). The highest concentrations were generally detected in the northern orientation, with the outer bark showing particularly elevated accumulation. Temporal evaluations across twelve time periods from 1963 to 2022 indicated a decreasing trend in Tl levels after 2008, suggesting improved environmental conditions in recent years. The results highlight the ability of Robinia pseudoacacia to reflect both spatial and historical patterns of Tl contamination, supporting its use as a medium-sensitivity biomonitor. Additionally, the wood tissue showed a potential to represent long-term cumulative Tl exposure. Overall, these findings suggest that Robinia pseudoacacia could be an effective candidate for phytoremediation and biomonitoring strategies in urban and industrial areas with thallium contamination.

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Introduction

Metals are among the most important chemical compounds in the assessment of environmental pollution, and in this context, the toxic effects of heavy metals have become an increasing source of global concern (Wei et al., 2021; Shakoor et al., 2013; Zhai et al., 2018). High concentrations of heavy metals in the environment can pose serious threats to both ecosystem health and human life (Ali & Khan, 2018; Aliyev et al., 2018). Among these metals, thallium (Tl), which occurs in trace amounts in nature but is notable for its high toxicity, has drawn considerable attention (Corzo Remigio et al., 2022). The toxic effects of thallium are reported to be even stronger than those of other well-known toxic metals such as lead, mercury, and copper (Peter & Viraraghavan, 2005; Das et al., 2007; Liu et al., 2020).

The environmental behavior of thallium is mainly explained by its interference with biochemical processes, especially due to the chemical similarity of Tl(I) ions to potassium ions (Mullins & Moore, 1960; Sager, 1998). This property disrupts potassium-dependent metabolic processes, thereby enhancing its toxicity. The persistence of thallium in soil, water, and air systems poses risks both through direct exposure and through indirect effects via the food chain (Karbowska, 2016). Thallium reaching human health through contaminated water and food is considered toxic to all living organisms (Nriagu, 1998; Kazantzis, 2000).

Thallium pollution is directly associated not only with natural processes but also with anthropogenic activities such as mining, metal smelting, coal combustion, and cement production. Although its insoluble forms in soil tend to be relatively stable, dissolved thallium compounds can migrate into surface and groundwater, thereby becoming mobile (Karbowska, 2016). Furthermore, high Tl concentrations in soil can be taken up by plants, leading to accumulation in plant tissues and creating a potential biological transfer mechanism.

Therefore, it is important to develop sustainable and environmentally friendly methods to mitigate thallium pollution. In this regard, the phytoremediation approach, which relies on the capacity of certain plant species to absorb, accumulate, or detoxify environmental pollutants through biochemical pathways, has come to the forefront (Lee, 2013; Cristaldi et al., 2017; Parihar et al., 2021). Compared to traditional physical or chemical remediation

techniques, phytoremediation attracts attention due to its low cost, ecosystem-friendly nature, and lower destructive impact on land (Ali et al., 2013).

Phytoextraction, one of the most common sub-applications of phytoremediation, is based on the uptake of metals through plant roots and their accumulation in tissues (Suman et al., 2018; Antoniadis et al., 2021). For this purpose, plants with features such as high growth rates, metal tolerance, and high biomass production are preferred (Arthur et al., 2005; Shikha & Singh, 2021). In the literature, species such as Populus spp., Salix, Brassica, and certain Iberis and Biscutella species have been reported to have high thallium absorption capacity (LaCoste et al., 1999; Dmowski et al., 2002; Marmiroli et al., 2011; Fellet et al., 2012; Villette et al., 2019; Wani et al., 2020).

Determining the concentration of thallium in the environment is crucial for analyzing exposure risks and developing appropriate remediation strategies. Advanced analytical techniques such as ICP-MS, ICP-OES, GFAAS, LEAFS, and voltammetric methods are among the methods developed for this purpose (Baiocchi et al., 1994; Boughriet et al., 2007; Liu et al., 2010; Karbowska et al., 2014; Ghaderi et al., 2015; Kravkaz Kuşçu et al., 2022; Ölmez & Kuşçu, 2024; Ergül & Kuşçu, 2025a).

In this study, the variation in thallium accumulation in different tissues (outer bark, inner bark, and wood) of Robinia pseudoacacia (black locust) was determined and evaluated according to direction, organ type, and year interval. In addition, the potential suitability of this species as a bioindicator or phytoremediator in environmental remediation studies was assessed.

Materials and Methods

Study Area

The study was conducted in Düzce province (40° 56' 49" N, 31° 14' 17" E), located in the Western Black Sea Region of Türkiye. The mean annual temperature of the study area is 13.2 °C, with an average annual precipitation of 838 mm. According to the Köppen climate classification, the area is categorized as "Cfa," indicating a temperate climate with mild winters, hot summers, and precipitation throughout the year (Kottek, 2006; Bölük et al., 2023). Düzce is a region facing significant environmental pollution challenges, particularly in terms of air quality, which has reached critical levels in recent years. According to the 2021 World Air Quality Report published by Switzerland-based IQAir, Düzce was ranked among the top five most polluted cities in Europe for air quality (IQAir, 2021; Ergül & Kuşçu, 2025b). This ranking indicates that the concentrations of harmful particulate matter such as PM_2 in Düzce are far above European standards. The province's high air pollution is also strongly influenced by its geographic and meteorological characteristics. The city center is situated within a plain surrounded by mountains, where weak wind circulation prevents the dispersion of pollutants, leading to their accumulation in the atmosphere. As a result, due to both its geographical features and human activities, Düzce is considered a region highly prone to pollution. Therefore, this study was conducted in this area, considering the potential role of thallium as an invisible risk factor within Düzce's current environmental pollution profile.

Samples Collection and Analysis

This study utilized wood, inner bark, and outer bark samples of Robinia pseudoacacia growing in the city center of Düzce. Robinia pseudoacacia is a species commonly found in natural European forests and is frequently used as an ornamental plant in landscaping projects. Samples were collected toward the end of the vegetation season, in the last week of October, by identifying the north-facing side of the tree and cutting approximately 50 cm above ground level from the main trunk. In the laboratory, it was determined that the tree stem was 60 years old. Wood samples were grouped in five-year intervals, and inner and outer bark samples were also extracted using a steel drill. In similar studies, analyzing individual annual rings is generally not feasible; therefore, grouping is typically performed based on the study objectives, the total number of annual rings, and the ring widths, using groupings of 3, 5, or 10 years (Key et al., 2023; Cobanoglu et al., 2023).

The prepared samples were first pre-dried by storing them in glass Petri dishes for 15 days at room temperature, followed by drying in an oven at 45 °C for two weeks. Subsequently, 6 ml of 65% HNO_3 and 2 ml of 30% H_2O_2 were added to 0.5 g of dried sample, and pre-digestion was performed in a microwave oven. The resulting digests were transferred to volumetric flasks and diluted to 50 ml with ultrapure water. Tl (thallium) concentrations were then determined using an inductively coupled plasma optical emission spectrometer (ICP-OES) (Spectro, Kleve, Germany). This method has been widely applied in previous research in this field (Erdem et al., 2024; Ergül & Kuşçu, 2024; Isinkaralar vd., 2025a,b).

The data obtained were analyzed using the SPSS 22.0 software package, and analysis of variance (ANOVA) was applied (Baykalı & Şen, 2024). For factors showing statistically significant differences at a confidence level of at least 95% ($p < 0.05$), the Duncan test was subsequently performed.

Results

In Table 1, the spatial variation of thallium (Tl) concentrations in three different organ types (outer bark, inner bark, and wood) of Robinia pseudoacacia was evaluated using analysis of variance (ANOVA), and significant spatial differences were identified. According to the variance analysis, the organ × direction interaction showed statistically significant differences ($p < 0.001$, $F = 42.4$). The highest mean Tl concentrations were observed in the northern direction for all organ types, while the lowest means were recorded in the west. In particular, the Tl concentration in the wood tissue in the northern direction (62,975.8 ppb) was significantly higher compared to that in the west ($p < 0.001$). The distribution of organs across directions also demonstrated statistically significant differences; for instance, in the eastern direction, a significant difference at the 5% significance level was identified between the inner bark and the other organs ($F = 6.9$; $p < 0.05$). A highly significant difference was found between the north and west directions in the outer bark. Among the organs, the highest mean value was measured in the inner bark, whereas the lowest was observed in the wood tissue. Based on mean values by direction, Tl concentrations in wood tissues were ranked as west < east < south < north.

Table 1. Variation in Tl (ppb) Concentrations by Organ and Direction in Robinia pseudoacacia

	North	East	South	West	F	Mean
Outer bark	63923.7^{C}	57902.2^{aB}	59322.7^{B}	53689.4^{A}	14.3**	58709.5
Inner bark	62870.3^{D}	61379.5^{bC}	57892.6^{B}	55388.7^{A}	6.9*	59382.7
Wood	62975.8^{D}	57312.3^{aB}	58995.2^{C}	54599.2^{A}	42.4***	57312.3
F	0.0^{ns}	8.0**	0.6^{ns}	0.4^{ns}		0.1^{ns}
Mean	62870.4^{D}	56924.2^{B}	59059.8^{C}	54609.3^{A}	35.0***	

* $p < 0.05$; ** $p < 0.01$; *** $p < 0.001$; ns $p > 0.05$. Lowercase letters indicate Duncan test groups within columns, while uppercase letters indicate Duncan test groups within rows

Table 2. Variation in Tl (ppb) Concentrations by Period and Direction in Robinia pseudoacacia

Year	North	East	South	West	F	Mean
2018-2022	BDL	57946.7	59796.3	56912.3^{cde}	1.3^{ns}	58218.4
2013-2017	BDL	56537.7	58627.7	57641.7^{e}	0.7^{ns}	57602.4
2008-2012	BDL	56442.6	59201.8	55573.9^{bcde}	3.2^{ns}	57072.8
2003-2007	54036.3	57001.9	59654.8	57128.2^{de}	0.1^{ns}	56955.3
1998-2002	63687.3^{C}	55568.6^{A}	58769.5^{B}	53706.3^{abA}	21.8***	57932.9
1993-1997	67345.1^{A}	57823.5^{AB}	59618.7^{B}	54067.1^{abcdA}	22.0***	59713.6
1988-1992	64261.3^{C}	55558.0^{A}	58572.2^{B}	53915.5^{abcA}	29.3***	58076.8
1983-1987	62998.0^{C}	56647.1^{AB}	58556.7^{B}	53346.2^{abA}	13.5**	57887.0
1978-1982	63626.9^{D}	56749.5^{B}	59410.9^{C}	54553.2^{abcdeA}	46.2***	58585.1
1973-1977	63302.7^{C}	56864.6^{B}	59451.5^{B}	52950.2^{abA}	19.7***	58142.3
1968-1972	63564.4^{C}	57790.9^{B}	58048.9^{B}	52169.3^{aA}	20.5***	57893.4
1963-1967	63011.9^{C}	58159.0^{B}	59008.3^{B}	53347.4^{abA}	14.4**	58381.7
F	0.9^{ns}	0.6^{ns}	0.2^{ns}	3.4**		0.2^{ns}
Mean	62975.8^{D}	57312.3^{B}	58995.2^{C}	54599.2^{A}	42.4***	

*$p < 0.05$; ** $p < 0.01$; *** $p < 0.001$; ns $p > 0.05$; BDL: Below detection limit. Lowercase letters indicate Duncan test groups within columns, while uppercase letters indicate Duncan test groups within rows

Researchers evaluated the temporal and directional variation of thallium (Tl) concentrations in the woody tissues of Robinia pseudoacacia over twelve periods spanning 1963–2022, and the results are presented in Table 2. The findings indicate that Tl concentrations fell below the detection limits after 2008, particularly in the northern direction, suggesting a decline in Tl accumulation over time. This may point to reduced Tl exposure in the north in recent years, potentially related to changes in environmental conditions such as atmospheric emissions, soil pH, and precipitation regimes.

When comparing periods by direction, it was observed that all measurements after 2008 remained below the detection threshold in the northern direction. In earlier periods prior to 2003, however, higher Tl accumulation was recorded, with the highest value observed during 1993–1997 at 67,345.1 ppb. In the eastern direction, no statistically significant variation was detected across all periods ($F = 0.6$; $p > 0.05$), with values remaining relatively stable (ranging from a minimum of 55,558.0 to a maximum of 58,159.0 ppb). In the southern direction, Tl concentrations ranged between 58,600 and 59,600 ppb, with some temporal variation observed, although these differences were not statistically significant ($F = 0.2$, ns). Only in the western direction were statistically significant differences between periods found ($F = 3.4$; $p < 0.01$). The lowest mean value was recorded at 52,169.3 ppb (1968–1972), and the highest at 57,641.7 ppb (2013–2017), indicating the presence of temporal fluctuations in Tl accumulation in the west.

Overall, Tl concentrations were found to vary within a relatively narrow range, with the highest value measured in the north at 67,345.1 ppb and the lowest value measured in the west at 52,169.3 ppb.

Discussion

According to the findings of this study, significant differences were identified in thallium (Tl) concentrations within the outer bark, inner bark, and wood tissues of Robinia pseudoacacia, depending on directional exposure. The interaction between organ type and direction was found to be statistically significant. Based on the mean values, Tl concentrations across directions were ranked as follows: North (62,870.4 ppb)> South (59,059.8 ppb)> East (56,924.2 ppb)> West (54,609.3 ppb). This result indicates a significant spatial asymmetry in metal accumulation associated with orientation, with the northern direction demonstrating a dominant pattern of Tl accumulation ($F = 35.0$; $p < 0.001$). These findings suggest that depending on orientation, plants may exhibit differential heavy metal accumulation. Environmental factors such as sunlight exposure, prevailing wind direction, and soil moisture could influence biotic absorption processes. The distribution of Tl concentrations across plant organs according to direction was also statistically significant ($p < 0.05$).

Overall, Tl accumulation varied according to both directional and organ-related factors, with particularly high accumulation observed in the wood tissue oriented northward. This underscores the importance of considering the orientation-based selection of plant species and tissues in phytoremediation strategies and highlights the critical role of plant tissue type and spatial positioning in environmental risk assessments. The results also indicate that the wood tissue functions not merely as a transient or superficial metal sink but as a permanent and systematic reservoir. The transport of Tl through the xylem and its subsequent storage in woody structures may be interpreted

as a consequence of long-term environmental exposure. The higher accumulation observed in the northern direction may be related to microclimatic factors such as lower sunlight exposure, more extended periods of retained moisture, and local precipitation patterns. The considerable accumulation of Tl in the wood tissue suggests that this tissue could serve as a valuable component in biomonitoring studies due to its capacity to reflect cumulative pollution loads over time. Nonetheless, the lower Tl levels in the wood than in bark tissues may also indicate that the plant possesses specific mechanisms for metal filtering and retention.

When the temporal and directional variations of Tl concentrations in the wood tissue were examined, statistically significant differences were detected only in the western direction across different time periods. This finding suggests the presence of a temporal fluctuation specific to the west. On the other hand, in the northern direction, Tl accumulation in previous periods was found to be significantly higher compared to other directions and time intervals. In particular, the periods 1993–1997 and 1988–1992 emerged as groups with notably high values in the north. Based on average values, the general ranking of Tl concentrations according to direction was as follows: North (62,975.8 ppb)> South (58,995.2 ppb)> East (57,312.3 ppb)> West (54,599.2 ppb). This pattern indicates that despite temporal changes, Tl accumulation remained dominant in the north, while the west continued to exhibit the lowest concentrations ($F = 42.4$; $p < 0.001$). Another important finding is that overall Tl concentrations remained within a relatively narrow range over time. Similar observations have been reported in the literature; for example, Zhang (2019) found that Zn and Pb concentrations in the annual rings of Cedrus deodora showed some variation while Cu concentrations remained stable; Yiğit (2024) observed that Ge transfer in Pseudotsuga menziesii wood occurred within certain limits; and Kuşçu (2025) reported that Ca concentrations in wood tissues varied within a narrow range. These data suggest that Tl exhibits limited mobility within the wood tissues of Robinia pseudoacacia, indicating its potential suitability as a biomonitor species for applications requiring moderate sensitivity in Tl pollution monitoring.

The fact that the highest values were generally observed in the northern direction and in the outer bark tissue shows that this tissue is the most intense region for Tl accumulation. Since the outer bark is the first structure in direct contact with environmental exposure, it is reasonable for higher metal concentrations to be detected there (Yiğit, 2024; 2025). This supports the conclusion that the outer bark is a critical tissue for biomonitoring purposes.

In general, this study reveals that heavy metal accumulation varies not only among different species but also across different organs of the same species. The primary reason for this is that numerous factors simultaneously influence the uptake and translocation processes of heavy metals in plants. The entry of heavy metals into plant tissues is affected by parameters such as plant species, organ structure, surface area, metal–plant interactions, and meteorological conditions (Şevik et al., 2024). Heavy metals can enter plant systems through the soil via roots, from the atmosphere through leaf stomata, or directly through stem tissues (Ozturk Pulatoglu et al., 2025). Heavy metals accumulated in various plant organs are among the significant pollutants that pose risks to both human and ecosystem health, which explains the intense research interest in recent years. Moreover, Tl has been reported to show high levels of correlation with other heavy metals (Şevik et al., 2024). Regional studies have also reported elevated concentrations of Pb (Koç et al., 2025a), As (Yaşar İsmail et al., 2025), Sb (Canturk et al., 2024), Bi (Isinkaralar et al., 2024), Cr (Ozturk Pulatoglu et al., 2025), as well as Sr, Mo, and Sn (Yiğit, 2024), all of which pose threats to human health.

In conclusion, it can be inferred that the wood tissue of Robinia pseudoacacia does not directly reflect acute environmental metal exposure, but rather has the capacity to indicate long-term accumulation. The significant directional differences observed point to the influence of environmental conditions (light, moisture, wind) on metal accumulation within wood tissues. These findings highlight that wood tissue, owing to its metal storage potential, should be considered when selecting tissues for phytoremediation applications.

Conclusion

In this study, thallium (Tl) concentrations in the wood and bark tissues of Robinia pseudoacacia were evaluated. The results indicate that Robinia pseudoacacia can serve as a biomonitoring species with medium sensitivity, capable of reflecting directional and temporal patterns of environmental Tl accumulation, exhibiting organ-level variability, and benefiting from a broad distribution range. In particular, it represents a suitable candidate for tracking historical pollution levels and for conducting environmental risk assessments in urban and industrial areas. However, in cases where lower-level (micro-scale) pollution needs to be monitored or acute exposure events must be identified, it is advisable to use this species in combination with other plant species that possess high metal hyperaccumulation capacity.

Declarations

Ethical Approval Certificate
Not applicaple

Author Contribution Statement
S.G.Ş.: Data collection, investigation, methodology, formal analysis, and writing the original draft, review and editing.

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