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Temporal Variability of Trace Metal Evidence in *Cupressus arizonica*, *Platanus orientalis*, and *Robinia pseudoacacia* as Pollution-Resistant Species at an Industrial Site

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Abstract Ambient air pollution in industrial areas is one of the significant problems increasing high emissions since the 1990s. Atmospheric trace metal deposition in the industrial estate has occurred, and some have shown toxic effects on humans and non-human biota. Tree bark and annual rings have been used as convenient biomonitors due to the highly absorbing capacity of several metals for passive samplers in recent decades. It gives information about the speciation of some pollutants and their transition between organs in the trees. Systematic differences in air pollution degrees can be recorded spatial or temporal by analyzing the annual rings of the trees. In this study, *Cupressus arizonica*, *Platanus orientalis*, and *Robinia pseudoacacia* have been selected as landscape trees to identify Al, Cr, and Mn pollution during the past 30 years in the urban environments of the İzmit, Türkiye. The widespread type of landscape species was compared to perform the deposition degree of several sites. Metal concentrations have been detected in the barks and wood due to the industrialization of the territory year-to-year. The study's framework indicated that the applicability of *Robinia pseudoacacia* L. is the better choice for assessing the degree of atmospheric metal pollution, both historical and near-past, on local scales.

Keywords Air pollution · Biomonitor · Industrial zone · Trace metals · Tree rings

1 Introduction

Global growing economies that have occurred in recent years led to the spread of various pollutants, causing vital deterioration in the composition and quality of air (Liu et al., 2018; Xu et al., 2020). The developments in the industrial field explain the rapid population growth and urbanization, which is one of the biotic factors in changing the atmosphere's composition (Hu et al., 2013; Isinkaralar et al., 2017; Wu et al., 2018; Yilmaz & Isinkaralar, 2021a, 2021b). Natural resources have started to be consumed rapidly, transforming nature with an increasing population (Cohen, 2006; Nathaniel et al., 2021). Thus, atmospheric pollution is mainly caused by anthropogenic factors (Wang et al., 2021). The air pollutants have become widespread and have reached severe dimensions, causing 4.2 million premature deaths and more than 7 million deaths every year by data of the WHO (World Health Organization, 2016; World Health Organization, 2018; Johnson et al., 2021; Roberts & Wooster, 2021). The European Environment Agency stated that polluted areas cover 2.5 million regions, and 14% of these areas need urgent

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remediation planning throughout Europe (OECD I., 2016; Bose, 2010). In this context, the fight against air pollutants originating from humans has taken its place within the scope of industrial activities (Kim et al., 2017; Sun et al., 2021).

Trace metals can be dangerous in high concentrations, and the essential metal is at an average level in vital activity (Singh & Kalamdhad, 2011; Nagajyoti et al., 2010). Some of those have toxic effects according to their long half-life in nature, exposure level, and concentration levels and are included in the Priority Pollutant List (PPL) by the Agency for Toxic Substances and Disease Registry (ATSDR) (ATSDR, 2018; Derosa et al., 1993; Jiang et al., 2021). They harm humans and ecosystem's living life (Tchounwou et al., 2012). Under the local background, trace metal contamination is a vital issue that is very important for researching and monitoring resources between atmospheric and terrestrial systems (Marć et al., 2015). Regarding the research, topics have emphasized that atmospheric trace metal deposition was related to dust and anthropogenic aerosol deposition (Faggi et al., 2011; Isinkaralar, 2022a, 2022b; Nickel & Schröder, 2017). The correlation between the atmospheric deposition of mineral dust and dissolved metals' enrichment that harms several species was observed (Cui et al., 2013; De Nicola et al., 2013; Märten et al., 2015). It is known that the formation, transport, and accumulation of trace metals on living things are a trending subject in recent studies.

Technological advances in the air monitoring system have begun to cause some problems, such as the cost and continuity of multi-point measurements (Ram et al., 2015; Tan et al., 2021). Since monitoring air pollutants is not a one-time thing, continuous monitoring is essential (Kelly et al., 2012; Ravindra et al., 2021). Thus, low-cost systems and biological monitoring have come to the fore. Biomonitoring is a passive methodology that is inexpensive and easy to implement (Ghoma et al., 2022; Guéguen et al., 2012; Sánchez-Chardi, 2016; Vitali et al., 2019). On the other hand, it has started to be preferred frequently in terms of the diversity of species and many advantages (being effective, low cost, etc.). They are living organisms that accumulate environmental pollutants due to anthropogenic activities and can provide quantitative information (Baroudi et al., 2020; Harrison et al., 2021). The availability of species as biomonitors to get metals in their bodies and the fact that we can measure their levels have enabled them to become biological receptors (Eid et al., 2020; Sawidis et al., 2011). A plant must first have certain physical conditions to be used as a biomonitor. It should not be overly sensitive and should have tolerance in its structure despite heavy metal loads (Fernández-Olmoet al., 2020; Rai, 2016). It is also necessary to be easily found outside the study area, that is, in other areas (for reference purposes), and that it has enough organs for analysis (Ghosh et al., 2020; Khamesi et al., 2020). Many studies have been conducted on the usability of several species as biomonitors or bioindicators (El Hayek et al., 2015; Isinkaralar et al., 2022, 2022a; Isinkaralar et al., 2022b, 2022c, 2022d; Yap et al., 2011). Since plants are the creatures that provide these features most easily (Brázová et al., 2021; Turkyilmaz et al., 2018), some questions have emerged about using plant fragments as biomonitors (Dołęgowska et al., 2021; Farzin et al., 2017). It is unknown how long plants such as moss and lichens are exposed to heavy metal pollution in the air (Kularatne and Freitas, 2013; Van der Wat & Forbes, 2015). As the best alternative to these, annual plants that are not evergreen solved this problem. Their leaves can easily give data on heavy metal pollution during vegetation season (Bonanno & Pavone, 2015; Chellman et al., 2020; Nakazato et al., 2016). Previous research stated that the healthiest information is obtained using biomonitors by using the tissues of species such as pine, spruce, and fir, whose needles have remained on the tree for many years (Alahabadi et al., 2017; Isinkaralar, 2021; Savas et al., 2021). The maximum data of 10 years are obtained when the annual rings of the trees are used. The development of trees has been at different levels in regions where four seasons are experienced. Annual rings are formed on the bark of woody plants, which has an essential role in understanding air pollution, its contents, and the transport of pollutants.

Some studies on the usability of annual tree rings as biomonitors have gained acceleration (Alatou & Sahli, 2019; Cucu-Man & Steinnes, 2013; Isinkaralar, 2022c, 2022d; Sen et al., 2017). However, the speciation of trace metals has not been demonstrated to transport the tree between bark and wood. In the present study, tree rings as biomonitors were evaluated and a comprehensive description of Al, Cr, and Mn concentration was determined in the industrial area of

İzmit in Kocaeli city. In this context, it is aimed to compare usability biomonitors in the three resistant species as *Cupressus arizonica*, *Platanus orientalis*, and *Robinia pseudoacacia*, which have grown in the intense industrial zone. Regional and international mean values can explain the extent of the tree bark and wood accumulation. Also, it has been investigated whether to effectively biomonitor the evolution and change level of Al, Cr, and Mn concentrations in the annual rings over the years.

Table 1 Descriptive information of the species and their identification numbers

Species ID	Species	Tissue	Tissue ID
T1	*Robinia pseudoacacia* L	Wood	W1
		Inner bark	I-B1
		Outer bark	O-B1
T2	*Cupressus arizonica* G	Wood	W2
		Inner bark	I-B2
		Outer bark	O-B2
T3	*Platanus orientalis* L	Wood	W3
		Inner bark	I-B3
		Outer bark	O-B3

2 Materials and Methods

2.1 Tree Species Sampling Site

Figure 1 shows that İzmit is one of the districts in Kocaeli with intensive production activities in the Marmara region of Türkiye. The main sectors are chemicals, solvents, machines manufacturing, automobiles, wood, furniture and PVC, foundry, food, different industries, and sub-industries in the İzmit. *Cupressus arizonica*, *Platanus orientalis*, and *Robinia pseudoacacia* can be found easily in landscape afforestation. Therefore, they were used as a biological biomonitor under the cover of their annual rings in this study. All species at each sampling site were collected from a pristine landscape area near industry-centered by the end of December 2020 from 7 individual trees of about 2.5 m. The sampling points were taken from a 750-m stretch of the near sector (taking into account the distance bordering it and prevailing wind direction). Therefore, it would not harm them as a 100-cm^2-thick log sample was removed down to the vascular cambium as the northern part at the breast height (1.5 m).

The tree rings of *Cupressus arizonica*, *Platanus orientalis*, and *Robinia pseudoacacia* were appointed in different age portions as 30 years old. They have come from 1991–1993 to 2018–2020 within 3 years (considering their widths and recorded for three years since 1991). The woody tissue was divided into groups for the age ranges and systematically collected (the same age per species) over a densely industrial area in one season. Then they determined the wood and bark layer over a much extended period. Small fragments were divided into years and put polyethylene sampling tube. Then, they were transferred to the laboratory and kept at – 18 °C before analysis. Also, they were labeled for identification by assigning a number from RP1 to RP10 (1991–2020) in Table 1.

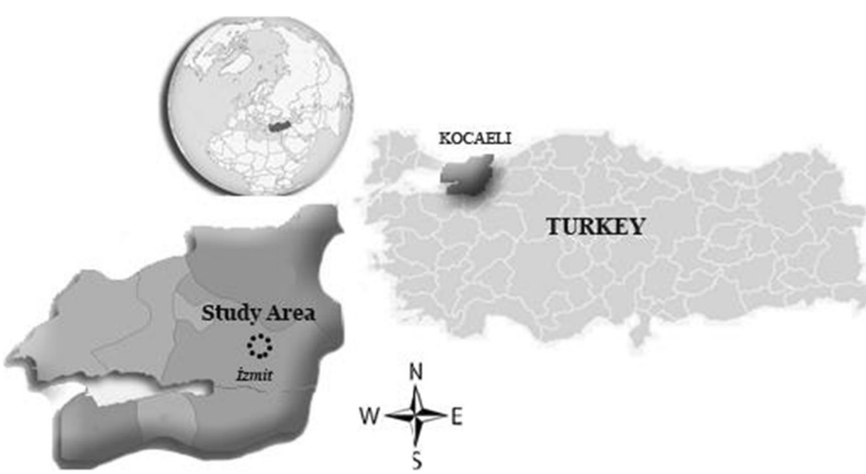

Fig. 1 Schematic diagram of the sampling locations

2.2 Analytical Methods

All samples were collected from open spaces and comprised roughly 30 subsamples of similar weight and 1–2 cm intervals with wooden tools. Once in the laboratory, log samples were separated from each other, then taken into glass containers (with deionized water before use), and kept in dry air at room temperature with their mouths open for 40 days. First, to remove the moisture content of samples, they were dried at 45 °C for 48 h. They were grinded with ball milling before analysis, ensuring no contamination. Next, roughly 0.5 g (±0.05 g) of the dry sample was weighed in the added solution tube as a digestion solution (HNO_3, HF = 3:1 v/v) and 2 mL of deionized water. Finally, the digesting was carried out in the microwave (Ethos One, Milestone GmbH, Germany) for 15 min. According to US EPA 3052 method (USEPA, 1996), the digestion procedures are as follows:

1. 1 min with the power of 1000 W to stay at 75 °C
2. 4 min with the power of 1000 W to keep at 180 °C
3. 10 min with the power of 1000 W to remain at 180 °C
4. The power of 0 W to stay at 15 min

After cooling, the resulting samples were diluted to 50 mL with ultrapure water. The concentrations of Al, Cr, and Mn elements were determined by the inductively coupled plasma mass spectrometry (ICP-MS, SpectroBlue, Spectro, Analytical Instruments GmbH, Germany) with a plasma source device (SpectroBlue, Spectro) by the EPA method 6020B (USEPA, 2013). All stock solutions were purchased from Merck (analytical grade and highest purity) and stored in a refrigerator at 4 °C. Samples from a region not close to industry or traffic emission sources were used as blank samples to determine the reference levels of the species used in the study. In addition, for analytical quality control, every five samples were checked at the same frequency to see if there was any contact with contaminants.

2.3 Statistical Analysis

The one-way analysis of variance (ANOVA) and Duncan's multiple range test were conducted on Al, Cr, and Mn concentration levels separately at the level of significance of 95% ($p < 0.05$). Whole measurements are repeated in triplicate to avoid experimental bias, and the acquisition data are analyzed using the statistical tool within SPSS version 22.0 (IBM SPSS Statistics software for Windows). The variance was demonstrated Al, Cr, and Mn concentration at a statistically significant 95% ($p < 0.05$) among samples.

2.4 Quality Assurance and Quality Control

Before starting the study, the tools for sample collection were first cleaned with acid and then rinsed with plenty of deionized water. After the pre-control procedures, the analyses were started by calibrating the devices to be measured (calibration curve > 0.999), and relative standard deviations in three replication experiments were determined as 5% for elements. Finally, measurements confirmed that the blank samples were not exposed to pollution. The detection limit of Al, Cr, and Mn was 0.12, 0.038, and 0.023 ppb, respectively.

3 Results

The ANOVA results show that the change of metals in species is statistically significant ($p < 0.001$). The Duncan test results obtained the lowest W1, W2, and W3 values, although the highest was obtained in O-B1, O-B2, and O-B3 (O-Bs). The importance of Ws and inner bark (I-Bs) was accepted as the same group in *Platanus orientalis*. However, the other species formed a separate group. The O-B values are higher than Ws and I-Bs in all the species. The variation of Al concentration based on species is statistically significant (at least $p < 0.01$) in Table 2.

Figure 2 shows that Mn concentration's accumulation levels are relatively low compared to Al and Cr deposition. The lowest Mn, Al, and Cr values of T1 in W1 are 8500, 14,100, and 15,000 ppb, respectively. On the other hand, the average concentrations of Cr, Al, and Mn of T2 in W2 are 289.9, 344.9, and 414.9 ppb, respectively. The medium values Al, Cr, and Mn of T3 in W3 are 457.5, 1424, and 1935.8 ppb, respectively. The highest values were obtained at 2,425,900 ppb in O-B1 and 93,829.7 ppb in O-B3 for Al and

Table 2 Variations in the mean value of trace metal concentration (ppb) for sampled species

Tissue	Elements			*F* value
	Al	Cr	Mn	
W1	14,100 Ba	15,000 Ba	8500 Aa	5.60**
I-B1	73,800 Cb	25,300 Bb	7200 Aa	5898.5***
O-B1	2,425,900 Cc	2,007,100 Bc	118,600 Ab	43,971***
F_1 value	55,883.6***	107,246.1***	4926.7***	
W2	344.9 Aa	289.9 Aa	414.9 Ba	6.7**
I-B2	446.9 Ba	450.4 Bb	228.4 Aa	22.6**
O-B2	10,462.3 Bb	23,022.2 Cc	1058.4 Ab	54,768***
F_2 value	18,164.2***	212,804.3***	17.6***	
W3	457.5 Aa	1424 Ba	1935.8 Ca	58.46***
I-B3	3913.8 Ab	16,976.8 Bb	47,352.4 Cc	45,285.1***
O-B3	93,829.7 Cc	88,741.0 Bc	36,755.4 Ab	9302.9***
F_3 value	18,164.2***	212,804.3***	17.6***	

Upper (horizontal) and lower case (vertical) letters indicate the directions

** significance at $p < 0.01$,*** significance at $p < 0.001$

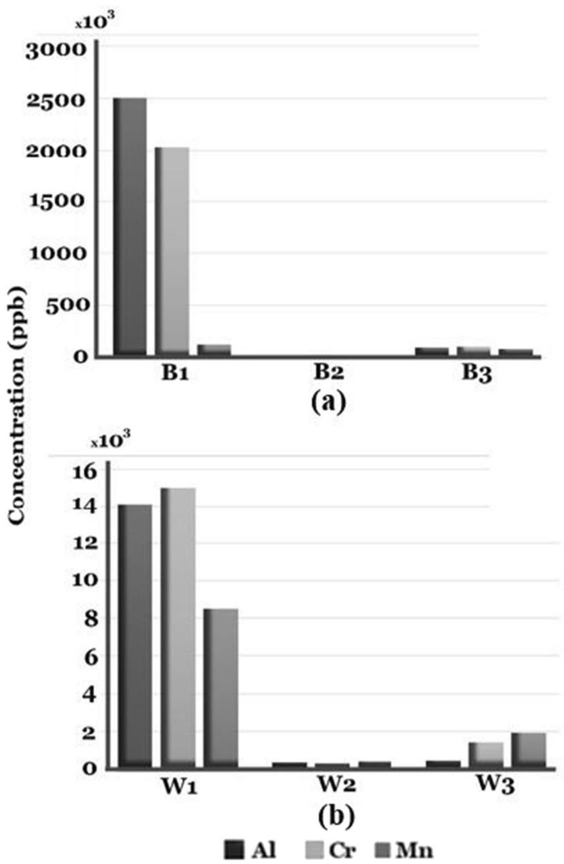

Fig. 2 The concentration of elements from species (**a** wood deposition; **b** total bark deposition)

23,022.2 ppb in O-B2 for Cr. The result also indicates that it remained consistent with the main result.

The variation of metal concentration is given according to the recorded period in tree species in Table 3. The Al concentration changes irregularly over the years. T1 obtained a maximum value of 49,400 ppb in Rp6, although the minimum value was achieved at 6600 ppb in Rp8. T2, the lowest value, is obtained with 7900 ppb between Rp7. Still, 29,700 ppb gained as the highest level in Rp5. T3 acquired the lowest value with 6300 ppb in Rp2, despite obtaining the highest value at 13,500 ppb between Rp4. In addition to industries such as mineral processing, other factors are predicted to affect its deposition on trees. The change of Cr element is examined; the highest value in T1 was 534.1 ppb in Rp3 and the lowest value with 253.8 ppb in Rp8. The lowest value in T2 was 223.6 ppb in Rp3; the highest value was 368 ppb in Rp10. Also, the highest value in T3 was 885.5 ppb in Rp6, and the lowest was obtained at 251 ppb between Rp9. The Cr concentration generally tends to increase until Rp6, but it decreases typically after these dates, and T3 reflects this change more clearly. In Mn deposition, the highest values in Rp10 with 903.4 ppb and 3462 ppb and 2482.8 ppb in Rp1 are obtained from T1, T3, and T2, respectively.

4 Discussion

The Al, Cr, and Mn values were relatively high in the samples from the industrial area. Tree bark and wood

Table 3 The mean metal concentration changes (ppb) in species for the recorded period

Recorded period	Al				Cr				Mn			
	T1	T2	T3	*F* value	T1	T2	T3	*F* value	T1	T2	T3	*F* value
Rp10	12,400 Bc	16,800 Cg	7900 Ad	28.3**	396.4 Cd	368 Be	273.2 Ab	101.5***	903.4 Ai	997.4 Bb	3462 Ch	22,133.2***
Rp9	6800 Aa	16,900 Cg	8300 Be	26,953.3***	348.9 Cc	289.2 Bc	251 Aa	103***	395.7 Ae	1127.5 Bd	2184.5 Ce	54,597***
Rp8	6600 Aa	11,400 Bd	6600 Aa	4384.2***	253.8 Aa	284.4 Bc	382.1 Cc	90.1***	327.8 Ac	1133.2 Bd	2097.2 Cd	101,970.3***
Rp7	10,800 Cb	7900 Aa	9600 Bg	849.6***	469.2 Be	284.6 Ac	671.3 Ce	728.8***	381.6 Ad	1405.1 Bf	2248 Cf	35,782.8***
Rp6	49,400 Ce	19,900 Bh	7900 Ad	547,636.6***	342.8 Bc	313.2 Ad	885.5 Cf	3725.8***	730.6 Ah	1488.8 Cg	1241.1 Ba	24,529.9***
Rp5	9500 Bb	29,700 Ci	7500 Ac	39,283.5***	284 Ab	367.8 Be	277 Ab	61.5***	562.2 Ag	1110.9 Bc	1741.5 Cc	14,461***
Rp4	10,700 Ab	11,200 Bc	13,500 Ch	4746.7***	256.8 Aa	250.4 Ab	444.3 Bd	790.1***	389.8 Ade	1223.1 Be	2576.2 Cg	78,364.3***
Rp3	20,700 Cd	12,200 Be	7300 Ab	47,311.7***	534.1 Cf	223.6 Aa	439.6 Bd	381.3***	518 Af	792.4 Ba	1326.2 Cb	11,200.5***
Rp2	7100 Ba	9100 Cb	6300 Aa	1235***	289.1 Bb	243.8 Ab	251.6 Aa	19.1**	189.6 Ab	2479.4 Ch	1242 Ba	74,632.5***
Rp1	6800 Aa	15,100 Cf	9400 Bf	61,259***	274 Ab	274.1 Ac	273.2 Ab	0.009 NS	176.4 Aa	2482.8 Ch	1239.3 Ba	86,122.4***
F value	823.3***	24,385.1***	5075.1***		293.8***	65.5***	994.6***		3522***	16,745.9***	15,112.2***	

Upper (horizontal) and lower case (vertical) letters indicate the directions

NS, there is no significant difference, ** significance at $p<0.01$, *** significance at $p<0.001$

have proven reliable tools for biomonitoring deposition of trace metals from the atmosphere. The study results showed that the data obtained were statistically significant for the organ-based change in the species ($p<0.05$). Also, it is stated that the Al, Cr, and Mn vary widely depending on the tissue changes within industrial releases. In another study, the many metals and biomonitors for underestimating the air pollution relationship emphasized the total bark of the trees was higher than in other organs (Isinkaralar & Erdem, 2021a, 2021b). Fujiwara et al. (2011) collected the tree bark of *Fraxinus pennsylvanica* to analyze heavy metal levels in Buenos Aires, Argentina. They deduced that Al and Mn levels in reference tree bark were 210,700 and 24,280 ppb, respectively. Sevik et al. (2020) conducted in parallel with this result that the accumulation of Pb, Co, and Fe elements considered heavy metals was higher in the outer bark. In some current research, it is overtly seen that there is accumulation in all tissues, but the assembly in the outer bark is higher, especially in the samples taken from industry and heavy traffic areas (Aricak et al., 2020; Isinkaralar & Erdem, 2022; Turkyilmaz et al., 2019). Koc (2021) also examined the Ni and Co changes in *Cedrus atlantica* annual rings from traffic sources. The Ni concentration was found to have significant values in wood. The familiar criteria of all these researches are that heavy metals' origins, whose absorption from the soil is low, are caused by susceptibility to air pollution (Zhang, 2019). It is derived from the use of fossil fuels and their incorporation through their release and transport.

Hubai et al. (2021) used *Plantago lanceolata* L. as biomonitors for high molecular weight PAHs and found a positive correlation. Similarly, Ortiz-Oliveros et al. (2021) showed the bioaccumulation potential of *Echeveria elegans* was very suitable for the determination of some metals (Zn, Ni, Cr, Pb, and Cs). Galal et al. (2021) selected *Ricinus communis* L. as a biomonitor to determine the presence of Cu in the leaves. Indeed, traffic and industrial facilities are the most important source of many heavy metals in plants (Al-Thani et al., 2018). The high accumulation of heavy metals has emerged primarily in the outer barks and the precipitation of particles after their transport and assembly in the atmosphere (Kumar & Khan, 2021). These studies have shown that elements in the air adhere to particulate matter and infect particulate matter with heavy metals (Cetin et al., 2020; Karacocuk et al., 2022). The rough surface structure of the outer bark facilitates the adhesion of particulate materials here and the high concentration of metals in the barks. The contamination of particulate materials is explained by heavy metals originating from traffic and industry because they are the most severe sources of toxic metals (Paraschiv & Paraschiv, 2019; Turkyilmaz et al., 2020). The relationship between particulate matter and heavy metals in plants has been demonstrated by the changes of the physiological structure of plants and the different transport and accumulation properties of each heavy metal in various studies (Sert et al., 2019; Świsłowski et al., 2020). Sevik et al. (2019) examined that the aspects of plant species in the regions with industry and vehicle density accumulate more parts and organs facing that direction. The papers show that many factors (plant habitus, organ structure, rainfall and moisture amount, etc.) can affect metal deposition (Kumar et al., 2020). While explaining the change of Al, Cr, and Mn concentrations over the years, their incorporation and accumulation can depend upon many environmental agents. The transfer of metals within the plant pathway is fast in some organs, such as the outer bark; it is slow in organs like wood.

For this reason, the fact that the trace metal in the atmosphere during the formation of woods is variable causes the accumulation in the woods formed by past pollution levels (El-Khatib et al., 2020; Locosselli et al., 2019). All studies reveal that the way of toxic metals enter cells is still a current topic of research. In summary, some findings show that there may be significant differences between organ transfers (the total bark and woods). The Al, Cr, and Mn concentrations of the varied trees hardly formed in the same year in the regions. Even though the effort of the metal transfers can be interpreted between organs as very limited, the trees can be used to estimate anthropogenic pollutants from the atmosphere or deposition on soil and the subsequent uptake by trees (Cocozza et al., 2016). Some metals move inside trees from bark to the phloem and the xylem (Martin et al., 2018). However, the information on the speciation and displacement of trace metals from their entry into the plant is quite limited.

5 Conclusion

Air quality monitoring studies in densely populated areas and cities are necessary to better understand the distribution and probable implications of pollutants. Monitoring and observation studies are expected to result from multiple points simultaneously. However, the number of sampling is kept low due to the high investment and operating costs for multi-use. The principal purpose of this paper was to examine the extent of exposure of the territory to atmospheric metal deposition of the industrial site with the use of trees as ecological indicators. It can emerge successfully in assessing air quality by multiple sample collections. This study was carried out on the three dominant species, *Cupressus arizonica* G., *Platanus orientalis* L., and *Robinia pseudoacacia* L., which are operatively utilized as landscape tree links between the industrial and the urban areas, despite the fact that the restricted number of three species measured the contamination level of metals between 1992 and 2021 in metalworking and machinery manufacturing industries. This study has pointed out that the most suitable species for Al concentration levels is *Robinia pseudoacacia*. Conversely, it has been determined that *Platanus orientalis* is the most suitable species for monitoring Cr and Mn concentrations for biomonitoring purposes due to experiencing intensely attributable to air pollution. Selected elements were shown a positive correlation with industrial emissions with all controls. This biomonitoring study helps to provide information on the increasing industrial activities in the region over the years and has endeavored for future works. This study highlights the relationship between passive monitoring methods for valorization and the exposure to air pollution over time by the long-range atmospheric transport. Further biomonitoring studies will probably lead to the impact of air pollution on trees and provide a reliable causal estimate.

Author Contribution This study is written entirely by Kaan Isinkaralar.

Data Availability The data that support the findings of this study are available from the corresponding author upon reasonable request.

Declarations

Ethics Approval Not applicable.

Conflict of Interest The author declares no competing interests.

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