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ORIGINAL ARTICLE

The large-scale period of atmospheric trace metal deposition to urban landscape trees as a biomonitor

Kaan Isinkaralar[1]

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Abstract

Traffic emissions and industrial activities cause atmospheric contamination derived from fossil fuels. The potential uptake of air pollutants affects the ecosystem and decreases air quality. Trace metal accumulation on trees shows toxicity in their cells due to root uptake, pathways of leaf deposition, and foliar absorption over the years. Trees can indeed provide data for the deposition quantity for a long time. Thus, urban landscape trees can be effectively trackable as a biomonitor to assess regional atmospheric pollution by starting a new research area. This study's aim was to perform Mn, Cr, Ni, Cu, Zn, Al, Cd, and Fe deposition to landscape trees within a region in Ankara by motor vehicles, residential areas, and small industries. *Ailanthus altissima* (Mill.) selected as native landscape tree species, an invader street tree, and overstrained species in urban areas. It was used as a biomonitor to determine the trends of trace metals and analyzed via tree rings the long-term atmospheric deposition between 1950 and 2020. The obtained results were determined as wood < inner bark < outer bark by years. The toxic metal accumulation was significantly correlated with emissions of sources as follows: Al (758.04 ppm) > Fe (275.47 ppm) > Mn (52.68 ppm) > Ni (38.09 ppm) > Cr (36.40 ppm) > Cu (32.34 ppm) > Zn (29.22 ppm) > Cd (0.50 ppm). The street tree rings of *Ailanthus altissima* (Mill.) can be used as retrospective biomonitoring for estimating metal contamination levels.

Keywords Air pollution · Biomonitor · Trace metals · Tree rings · Urban air quality

1 Introduction

Environmental health problems have shown themselves as a lifelong problem in rapidly technologically advancing, industrializing societies and motor vehicles [1, 2]. Even though ecological degradation is primarily regional, it sets the way for emerging global issues [3]. Especially air pollution is a critical trouble for the urban environment. Its monitoring and risk assessment has become unavoidable for modern civilizations and urbanization [4–7]. Although not widely used, monitoring stations and active approaches have proven costly and time-consuming in urban systems [8]. Passive sampling approaches have become more popular because authorities seek less expensive and longer term monitoring without a device [9–11]. Urban areas contain a wide variety of trace metals which constitute the significant components of air pollution [12, 13]. Manganese (Mn), chromium (Cr), copper (Cu), zinc (Zn), aluminum (Al), nickel (Ni), lead (Pb), iron (Fe), and cadmium (Cd) are trace metals with different chemical structures that can lead to harmful effects when present in high concentrations [14]. These contaminants can build up on living organisms, initially nearby, and then transfer to large distances by wet or dry deposition [15]. They can be quickly accumulated on trees and widely used in biomonitoring studies in dendrochemical methods [16–18].

Manganese (Mn) is a necessary element for humans and occurs naturally in different forms as MnO_2, Mn_3O_4, oxidized Mn^{+2}, and Mn^{+3} [19]. It is easily obtained from various sources (soil, eroded rocks, decomposed plants, dust, etc.) [20]. Chromium (Cr) is a hazardous metal that belongs to the "Group 1 carcinogen" by the International Agency for Research on Cancer [21]. It is one of the most common pollutants in the environment and is a source of concern. Although there are seven different oxidized forms, the most prevalent are Cr^{+3} (abundant in nature and plants may readily absorb) and Cr^{+4} forms. Cr^{+4} is released into the environment by anthropogenic means, particularly in the

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✉ Kaan Isinkaralar
kisinkaralar@kastamonu.edu.tr

[1] Faculty of Engineering and Architecture, Department of Environmental Engineering, Kastamonu University, 37150 Kastamonu, Türkiye

metal industry, from leather processing and dyestuff-producing facilities [22]. In acidic and alkaline environments, Cr^{+3} can become Cr^{+4} [23]. Nickel (Ni) is released into the environment through anthropogenic and natural emissions from facilities such as fossil fuel combustion, mining, waste disposal, and the cement and steel industries [24]. Copper (Cu), zinc (Zn), aluminum (Al), nickel (Ni), lead (Pb), and cadmium (Cd) are examples of metals that differ chemically because they are hazardous metals. Cu is a heavy metal in three forms (1, +2, and +3) which vary in mobility and bioavailability [25]. It is frequently employed in wire, sheet, and transfer items. Cd is a highly active hazardous metal discharged into the environment and soil due to metal mining, exhaust emissions, and other industrial processes [26, 27]. Al^{+3} is an abundant cation in the earth's crust that may bind to a wide range of macromolecules [28]. Because of its prevalence in some fertilizers, polluted soil, industrial emissions, pharmaceutical and cosmetic items, and acid rain, humans can easily absorb it through the gastrointestinal system and oral epithelium, and olfactory and skin contact [29]. Fe is a vital micronutrient for several metabolic processes, including DNA synthesis, respiration-photosynthesis, enzyme function, and biological functions [30]. The Fe has a valence of −2 to +6 depending on its oxidation state and is primarily found in nature as Fe^{+2} (ferrous) and Fe^{+3} (ferric) [31]. Fenton and Haber-Weiss reactions make free Fe ions more hazardous when they contact other radicals [32]. They are a crucial component that helps regulate metabolic activities (enzymes, oxidative stress regulation, antioxidant status, mitochondrial activity, and neurotransmitter synthesis). They are essential for plants' growth, development, and enzyme activity. In higher plants, this element is found in enzymes like glyoxalase-I and urease, which promote biotic and abiotic stress [33, 34]. When these metal levels are excessive, studies have reported they induce growth abnormalities, development deficiencies, sugar-mineral imbalance, and several other ailments in human health [35–39]. They observed that some poisonous metals adversely affect human health depending on exposure time, concentration, and pollutants [40–43]. Miscarriages, early births, developmental abnormalities, and even death have been observed in pregnant women [44, 45]. Also, they can have poisonous and toxic characteristics on DNA functions in trees at high concentrations [46, 47]. Trees absorb these trace metals, accumulating in their cells under many factors such as pH, humidity, elemental analysis, soil structure, atmospheric deposition, and acid rain [48, 49]. Therefore, it is essential to monitor the levels of pollutants and observe predominant anthropogenic sources.

Various stations have been established to monitor these actively, although they are not widely used because of the high cost of monitoring trace metals and poisonous metals [50, 51]. Most of these studies have been carried out using urban trees and their organ for biomonitoring the amounts of atmospheric metals and other air pollutants [52–54]. It is critical due to the low cost and many advantages of monitoring hazardous metals [55, 56]. As a result, passive sampling approaches have gained traction and become a hot topic [57–61]. The usefulness of living organisms can accumulate toxic metals as passive sampling because they are coming into prominence [62, 63]. The quest for animals that can live for a long time has become increasingly important so that pollution can be monitored for a long time [64–66]. Because the valuable species that may survive for a long time die for assorted reasons, the findings of the tree ring study revealed the amounts of various heavy metal contaminants throughout time [67, 68]. Current research indicates that the absorbing capacity of trees can be used as bioindicators for temporal air pollution, and they have vital roles in atmospheric pollutants' deposition [69]. The quantity of air pollutants' concentration in annual rings of species that may be utilized for biomonitoring purposes has given information about the composition and history of air pollution in areas with dense populations, traffic, and industrial emissions worldwide [70–72]. This research aims to track the changes of Mn, Cr, Ni, Cu, Zn, Al, Cd, and Fe bioaccumulated by annual rings of *Ailanthus altissima* (Mill.) species in a region with high traffic density and estimate current atmospheric levels. The purpose was to assess the trace element concentration levels of tree bark and wood, preferred primarily on roads in cities. Their suitability has been explored to gather the information to guide future generations of biomonitor studies.

2 Materials and methods

2.1 Study area and sample collection

Altindağ district, part of Ankara province, has about 400,000 people, and is the study area (Fig. 1). It is located on the rough land between Ankara, Çubuk, and Akinci plains. The district covers an area of 157.47 km^2. It is 850 m above sea level, with mountainous terrain accounting for 31% of the surface area, plain ground for 6%, and undulating land for 3%. *Ailanthus altissima* (Mill.) is a deciduous tree of the Simaroubaceae family widely utilized in many world regions. This species, which can be found in parks, gardens, urban forests, and landscape regions, has a therapeutic use for various illnesses (such as stomach diseases, colds, and bleeding). Multiple kinds of research have revealed that it contains lignans, quassinoids, carboline alkaloids, sterols, and lipids.

The neighboring trees' annual ring was collected precisely and without mixing as two to three cores per tree near the main roadside. A 12-mm-diameter increment borer was used 1.5 m above the ground at different orientations for

Fig. 1 Sampling site location

each tree. They were given numbers for labels and stored in a polyethylene vessel prior to analysis. Samples were retained when the vegetation was over in 2021 from 12 cross-dated (precision of 0.001 mm) annual ring samples from 6 trees per point as the tree's outer bark, inner bark, and wood with a length of 69 years.

2.2 Tree ring analysis

Each sample was cleaned using a soft brush with silk bristles to remove smoothy and roughness. They were kept at room temperature with their mouths open for 12 days. One last time, there is no moisture inside that they were oven-dried at 45 °C for 48 h. They were ground with ball milling before analysis without any contamination. Each sample was taken at 0.5 g and weighed 6 mL of 65% HNO_3 (Merck), 2 mL of 30% H_2O_2 (Merck), and 2 mL of deionized water. The digested process was carried out in a microwave (Ethos One, Milestone GmbH, Germany) for 15 min. It was set up as follows: (a) 1 min at 1000 W, 75 °C; (b) 4 min at 1000 W, 180 °C; (c) 10 min at 1000 W, 180 °C; (d) 0 W for 15 min according to the 3052 Method USEPA [73]. The resulting samples were made up to 50 mL with ultrapure water (for dilution), and Cu, Zn, Al, and Cd analyses were made by atomic emission spectrometry (ICP-OES from SpectroBlue, Spectro, Analytical Instruments GmbH, Germany) with a plasma source device (SpectroBlue, Spectro). To determine the reference levels of the species used in the study, samples taken from a region not close to industry or traffic emission sources were used as blank samples. In addition, for analytical quality control, every five samples were checked at the same frequency to see if there was any contact with contaminants. In addition, limits of quantification (LOQ) and limits of detection (LOD) were calculated for Mn, Cr, Ni, Cu, Zn, Al, Cd, and Fe elements.

2.3 Statistical analysis

The one-way analysis of variance (ANOVA) and Duncan test (multiple ranges) were applied to Mn, Cr, Ni, Cu, Zn, Al, Cd, and Fe concentrations. All measurements were repeated in triplicate, and the obtained data were analyzed using the SPSS v.22.0 for Windows (IBM® Armonk, NY, USA). *F* parameter is shown of all concentrations at statistical significance (p<0.001). All data were presented as mean value with three replicates.

3 Results and discussion

Analyzing tree components of *Ailanthus altissima* investigated the spatial variability of 8 trace metals' concentration. In addition, the analysis of atmospheric trace metal deposition was evaluated for their total grades within the organs (inner barks, outer barks, and woods) in Table 1.

As a result of the ANOVA, it was determined that the change of the metals' subject to the study on an organ basis was statistically significant at the 99.9% confidence level ($p<0.001$). The Duncan test is examined, and the values obtained in the outer bark for all metals in the last group are seen. Although there is no significant difference in metal concentrations in wood and inner bark, the values in the outer bark are significantly greater than those in the wood and inner bark. The LOD values are 0.007 ppm for Ni, 0.03 ppm for Fe, 0.009 ppm for Cr, and 0.013 ppm for Mn; the outer bark values are around two times higher in Cd than the inner bark values. They are more than 12 times higher in Zn, 24 times higher in Cu, and 28 times higher in Al. The LOQ values were 0.0009 ppm for Ni, 0.0019 ppm for Fe, 0.0012 ppm for Cr, and 0.0011 ppm for Mn. While the outer bark values are around six times higher in Ni than the inner bark values, they are more than 3.5 times higher in Cr, five times higher in Mn, and 22 times higher in Fe (Table 2).

At the 99.9% confidence level, it was concluded that the variations in metals were statistically significant ($p<0.001$) between 1950 and 2020. When the changes in the mean values periodically and the groupings formed due to the Duncan test are examined, the Ni concentration reached the highest level in 1973–1978. The values obtained in this period were around three times in the other periods. Nevertheless, the Ni concentration has changed in the range of 0.53–0.93 ppm at relatively limited values and has shown a falling tendency since 2000. It was determined that the Fe concentration also followed a fluctuating course, which reached its highest concentration (100.7 ppm) in 1979–1981, and changed between 17.46 and 41.75 ppm in the other periods.

Moreover, Fe concentration did not show a regular change yearly. The Cr concentration followed a fluctuating course until 1982 and entered a general upward trend after this year, reaching its highest value in 2018–2020 at 1.13 ppm. When the change in Al is analyzed periodically, it can be observed that it grew to a maximum of 12.34 ppm until 1994–1996, except for 1966–1968, and then typically followed a horizontal trajectory until 1994–1996. The Al concentrations, which only slightly decreased from 1966 to 1981, continuously increased between 1994 and 2020. In terms of Cu concentration, the maximum value was 4.03 ppm in the 1950–1952 period, while the highest value was 2.25 ppm in the 2018–2020 period. The Cu concentration is horizontal in a narrow range between 0.82 and 1.67 ppm when seen in general. While the greatest Zn concentration was 5.95 ppm in the 2018–2020 period, it was acquired that it followed a variable course in general, with the highest values (except for the last period) in 1963–1965, 1972–1974, and 1975–1977, respectively. The results indicate that the Cd concentration showed variation in the narrowest range. The lowest value was 0.11 ppm in 2018–2020, while the highest was 0.13 ppm in 1985–1987. Between the two dates, there is a 16% difference. The Mn concentration followed a fluctuating course until 1994, contrary to Cr, and the values decreased after this date. Temporal changes of all heavy metal concentrations are given in Figure 2.

The study's findings proved the effect of traffic and industry emissions on trees, and these findings are similar to previous biomonitoring studies [75, 76]. The tree rings of the *Cedrus atlantica* Manetti species were gathered from the traffic density area as north-south and west-east by Savas et al. [56]. They investigated the accumulation of Cr and Mn heavy metals in wood, inner bark, and outer bark. They discovered that the outer bark had significant Cr and Mn levels compared to other organs. Ullah and Khan [63] used ANOVA and Tukey's test to analyze the variation of Pb, Cd, Zn, and Cu heavy metals from *Xanthium strumarium* L. invasive species collected from various sites in Khyber Pakhtunkhwa, Pakistan. As a result, they collated the magnitude of the heavy metal levels as road > urban > rural sites. Trace metal deposition has commonly been controlled via biomonitors and has increased over time in several trees [77–79]. The primary feature of the trees' usability as biomonitors is investigated according to their absorption

Table 1 The variability of major and trace element concentrations (ppm) between organs

Organ	Trace metals and their records							
	Mn	Cr	Ni	Cu	Zn	Al	Cd	Fe
Outer bark	42.07 c	34.48 b	36.61 b	29.92 b	25.32 b	719.84 c	0.25 c	164.79 c
Inner bark	8.02 b	0.98 a	0.61 a	1.22 a	2.10 a	24.96 b	0.13 b	75.18 b
Wood	2.59 a	0.94 a	0.87 a	1.20 a	1.8 a	13.24 a	0.12 a	35.50 a
F value	0.89***	1.83***	0.38***	2.64***	0.67***	52.19***	0.93***	149.44***

Lowercase letters express vertical direction for each variable, and *** indicates $p<0.001$

Table 2 Investigated major and trace element concentrations (ppb) for regional pollution from 1950 to 2020

Time series	Trace metals and their rates							
	Mn	Cr	Ni	Cu	Zn	Al	Cd	Fe
2020–2018	4159.2 s	1127.9 k	611.6 b	2254.3 g	5943.9 r	23119.5 t	106 a	44450 n
2017–2015	1794.3 d	954.3 g	541.6 a	1017.7 abcd	807.2 a	17326.9 p	110.7 abcd	30780 gh
2014–2012	1719.2 c	1051.4 j	605.2 b	950.5 abc	1446.5 g	15925.4 o	116.8 cdefgh	30050 f
2011–2009	1982.5 g	1023.3 i	746.4 h	1024.8 bcd	2184.2 m	21002 s	116.4 cdefgh	45710 p
2008–2006	1947.4 f	990.3 h	650.4 cd	1185.2 de	1914.6 l	17633.5 r	113 bcde	30590 g
2005–2003	2186.4 j	1002.8 h	675.6 ef	1131.2 cde	1855.7 k	15646 n	110.2 abc	23870 c
2002–2000	1591.7 a	886.6 bcde	752.4 hi	859.4 ab	1011.6 c	15055.3 l	111.5 abcde	17460 a
1999–1997	2199.1 j	959 g	680.5 ef	980.8 abc	1746.4 i	14079.8 k	111.3 abcde	19280 b
1996–1994	2399 n	893 de	708.3 g	869.3 ab	1127 d	11768.1 i	112.6 bcde	29570 e
1993–1991	1666.8 b	894.6 e	671.6 def	961.1 abc	1875.2 k	11588 i	112.9 bcde	42330 l
1990–1988	2111.7 h	888.7 cde	657.5 cde	856.7 ab	1015.6 c	11000.8 g	116.1 cdefg	30550 g
1987–1985	1940.8 f	871.8 abc	705.4 g	861.4 ab	930.6 b	10028.7 e	122.9 h	31060 h
1984–1982	2297.4 l	862.4 a	688.2 fg	865.4 ab	939.3 b	10045.2 e	121.9 gh	32900 j
1981–1979	1834.1 e	872.7 abcd	772.3 i	838 ab	943.2 b	8406.8 a	111.4 abcde	10070 r
1977–1975	2453.4 o	896.9 e	2721.8 m	818.7 a	2539.8 n	9057.6 b	116.8 cdefgh	24040 c
1974–1972	1949.2 f	879.1 abcde	2573.1 l	868.5 ab	2564.1 o	9336.6 c	112.8 bcde	44110 m
1971–1969	2256.7 k	949.8 g	756.6 hi	1033.1 bcde	1455.7 g	9878.7 d	108.2 ab	35840 k
1968–1966	2148.3 i	950.6 g	744.4 h	1663.8 f	1406.5 f	15411.6 m	118 efgh	23980 c
1965–1963	2298.5 l	896.6 e	930 k	1111.8 cde	3906 p	10635.4 f	120 fgh	45200 o
1962–1960	2350.6 m	867.8 ab	643.3 c	1217.4 e	1420.4 f	12311.7 j	117.3 defgh	31490 i
1958–1956	3025 p	918.7 f	763.4 hi	1116.1 cde	1530.8 h	11721.5 j	114.4 bcdef	28480 d
1955–1953	3640.5 r	1060.4 j	745.4 h	989 abc	1319.3 e	11137.7 h	112.2 abcde	44190 mn
1952–1950	9698.4 t	893 de	826.2 j	4032 h	1797.2 j	12341 j	120.2 fgh	29780 ef
F value	65747.5***	132.6***	5033.8***	140.1***	20233.1***	9486.8***	4.86***	2321660 ***

Lowercase letters express vertical direction for each variable, and *** indicates $p<0.001$

capacity that some trees are able to absorb specific elements better than others [80]. Rajfur [81] researched to evaluate the possibility of using deciduous tree bark on June 2–26, 2017, in Poland. Several species have accumulated different heavy metal levels by atmospheric aerosol and were influenced by their location, such as roadside. Koc [82] used annual rings to determine the change of Ni and Co elements in *Cedrus atlantica* over time and in response to traffic circumstances. Although there was a slight change in elemental concentration in wood, the outer bark had the highest values, and the inner bark had slightly lower values. Kousehlar et al. [83] used the bark of mainly oak (*Quercus*) and maple (*Acer*) as biomonitors in southwestern Ohio. They reported that elemental concentrations of tree barks contain several accumulation levels and exhibited Cr and Os elements having a positive correlation. In their study, tree bark can be a sufficient bioindicator of Os pollution in the environment. Karacocuk et al. [84], on the other hand, compared Cr and Mn levels in the leaves and bark of *Robinia pseudoacacia*, *Platanus orientalis*, *Acer negundo*, *Ulmus minor*, and *Nerium oleander* species collected in regions with high, low, and no traffic in Samsun. They discovered that each species could hold a different amount of heavy metal. Perone et al. [85] studied the assessment of trace elements in industrial areas near the urban center in Terni, Italy. The chosen tree rings of *Quercus pubescens* as biomonitors revealed retrospective pollution between 1958 and 2009 by anthropogenic factors. Locosselli et al. [86] evaluated biomonitor feasibility for accumulating the metals in the annual ring of *Tipuana tipu* in São Paul, Brazil. They found some metal levels are high, although other elements' accumulation level was low due to uptake of ways. Jeddi et al. [87] analyzed some metals for their accumulation situation on five tree species in Gabès, Tunisia. They used the leaves and bark of the different species and calculated their metal accumulation index for each species. The levels of presence of some metals varied according to the organs. Hatami-Manesh et al. [55] (2021) used diffusion on several species to investigate the quantities of heavy metals in washed and unwashed leaves, as well as a particulate matter on leaf surfaces in Isfahan, Iran. While Cd, Cr, and Ni levels were detected in clean and unwashed *Morus nigra*, *Pterocarya fraxinifolia* had greater Zn and Cu values, while *Platanus orient*alis had higher Pb values. The amount of accumulation of heavy metals was emitted

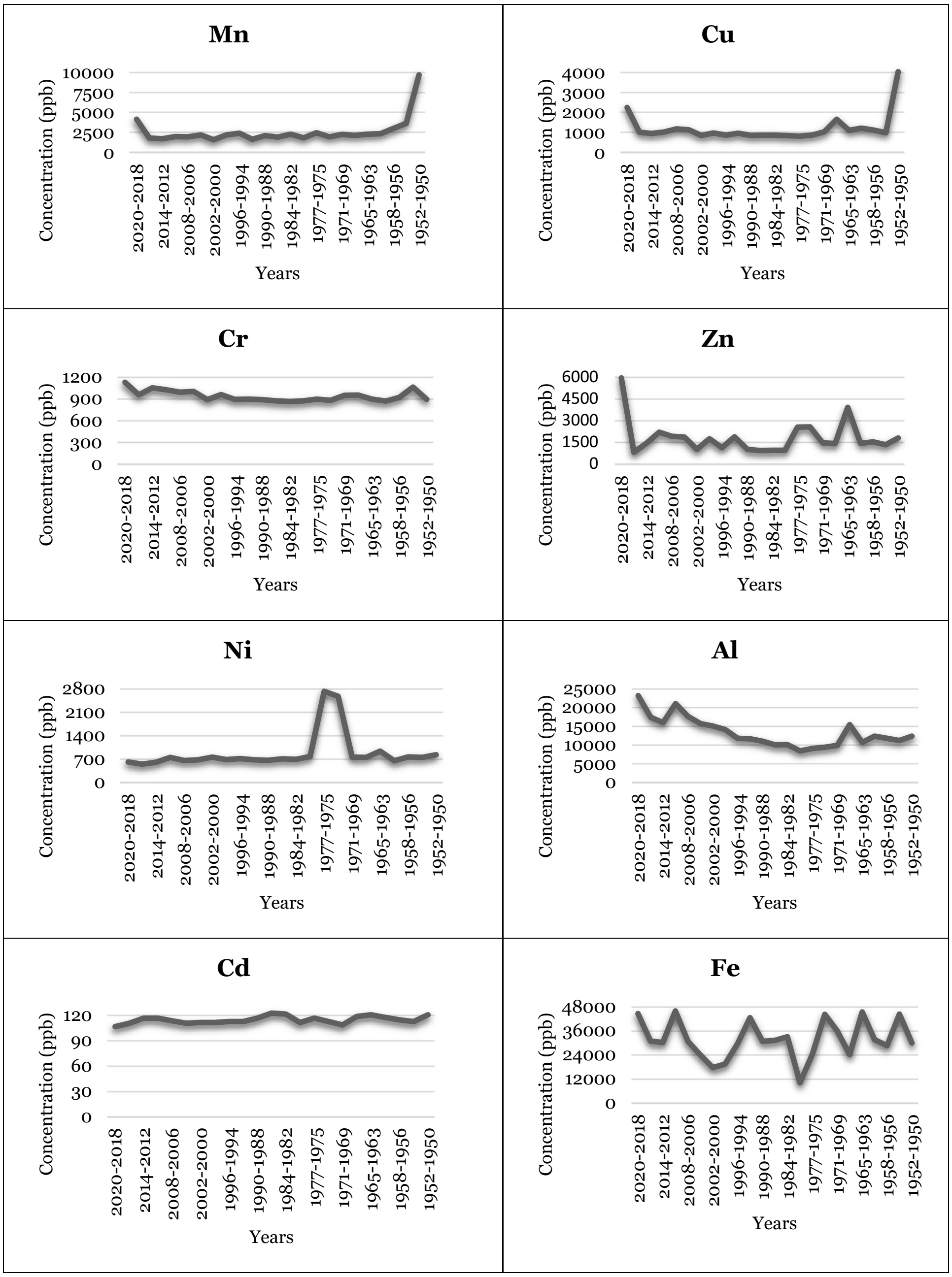

Fig. 2 Levels of trace metal deposition over the years

through transportation, industry, and other anthropogenic sources in many cities [88]. Table 3 shows the annual rings of various tree species employed in the study and their ages. The elements' accumulation was acquired in several species, corresponding with our results [91–93].

4 Conclusion

The prominence of landscape trees as bioindicators using older trees is convenient because they procure long-term tree ring records for environmental pollution. The tree ring of *Ailanthus altissima* (Mill.) as biomonitor is one of the best ways for passive sampling to evaluate trace metals' quantities and temporal records due to the capability of traversing long distances by various emissions. Accumulation in tree rings was accomplished to determine the primary sources and transport pathways of Mn, Cr, Ni, Cu, Zn, Al, Cd, and Fe level in the regional atmospheric deposition. On a local scale, they have identified that majority originate from anthropogenic activity (motor vehicles and fossil fuel combustion) and are readily transported under meteorological conditions. This paper indicates that *Ailanthus altissima* is crucially exposed and sensitive to

Table 3 Comparison of other studies about metal accumulation in tree rings

Species	Age	Elements	Range value	Study area	Ref.
Salix matsudana	~45	Magnetic particles, including heavy metals	5.97–285.71 µg/g	Xinglong, China	Zhang et al. [92]
Pinus ponderosa	350+	Heavy metals	0.002–20.51 ppm	Washington, USA	Padilla and Anderson [93]
Acer platanoides	27	Heavy metals	0.3–1500 ppm	Ankara, Türkiye	Turkyilmaz et al. [94]
Acer pseudoplatanus	111	Lead	0.45–6.64 mg/kg	Glasgow, Scotland	Patrick and Farmer [89]
Pinus massoniana	168	Heavy metals	4.402–64.938 mg/kg	Fuzhou, China	Chen et al. [91]
Quercus pubescens Willd.	26	Trace elements	-	Venafro, Italy	Cocozza et al. [95]
Cedrus sp.	39	Co, Pb, and Fe	18.6–5902.5 ppb	Kastamonu, Türkiye	Sevik et al. [90]
Ailanthus altissima (Mill.)	69	Trace elements	106–45200 ppb	Ankara, Türkiye	In this study

air pollution due to its ability to absorb trace metals in various ways. The results acquired specify the significance of a better comprehension of potential metal sources of urban air and their temporal distribution patterns in the nearby high emission region (condensed roads, lots of residential and industrial areas, etc.). The positive correlation analysis has been linked to the influence of local emissions on the organs, although relatively little is known about the dynamic relationships between trace metals and trees. The research species has a limited accumulation potential in its wood or a high amount of them due to various chemical binding forms with different strengths. The highest concentrations are associated with transportation pathways from the atmosphere to the *Ailanthus altissima* despite some elements having a natural origin, such as local pollution. Therefore, it is well known that atmospheric heavy metal deposition plays an essential role in urban air quality. Although atmospheric trace metal deposition cannot be precisely defined, biomonitoring provides an understanding of partial accumulation in the site where plants are collected. Therefore, *Ailanthus altissima* can effectively interpret local air quality in future studies.

Data availability All data generated or analyzed during this study are included in this published article. Also, the datasets are available from the corresponding author on reasonable request.

Declarations

Ethics approval Not applicable

Consent to participate Not applicable

Consent for publication Not applicable

Conflict of interest The author declares no competing interests.

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