



Proof of concept to characterize historical heavy-metal concentrations in atmosphere in North Turkey: determining the variations of Ni, Co, and Mn concentrations in 180-year-old *Corylus colurna* L. (Turkish hazelnut) annual rings

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Abstract

Heavy metals (HMs) are among the pollutants posing a significant threat to human, animal, and environmental health. Therefore, monitoring HM concentration changes, especially in the air, is crucial. This study used *Corylus colurna* L. (Turkish hazelnut) annual tree rings cut in 2020, and it was intended to define the 180-year variations in concentrations of Ni (nickel), Co (cobalt), and Mn (manganese) that are among the most harmful HMs. This study analyzed HMs concentrations in the wood, outer and inner bark in the north, south, east, and west directions and the seasonal variations in HM concentrations in tree rings. It was determined that, for all the three elements, the wood and barks significantly differed ($P < 0.001$) only in the north side, and the concentrations in wood were much lower than in the bark. The maximum concentrations were usually observed in annual rings in the west and south directions. The changes by both organ and direction can be related to the iron and steel factory and the highway, which are defined as HM sources. The results also revealed that the relocation of Co, Ni, and Mn in the wood of *C. colurna* tree remained at a limited level. The results suggest that *C. colurna* annual tree rings are very useful in tracking the variation of Ni, Co, and Mn concentrations.

Keywords Annual ring · Biomonitors · Heavy metal · Turkish hazelnut

Introduction

As a result of the increased population worldwide, the industrial revolution in the last century, advancements in technology, and natural sources were severely consumed (Shahid et al. 2017). During this duration, excessive use of all-natural resources and the release of minerals, which are used in the industry as raw material, into nature caused severe contamination in the soil (Liu et al. 2022; Chen et al. 2023), water (Ucun Ozel et al. 2020; Wang et al. 2022), and atmosphere (Cetin et al. 2019; Elsunousi et al. 2021; Koç 2021a; Isinkaralar et al. 2022a; Key et al. 2022). Among these types of pollution, especially air pollution, is of significant effect on human health and, as reported by World Health Organization, approx. 90% of people inhale the contaminated air. Air pollution has been reported to cause mass mortality, such as cardiac diseases, stroke, and lung cancer, in several regions in the past (Jo et al. 2020; Ozel et al. 2021). Moreover, due to the changes in the atmosphere's composition by air pollution, it directly or indirectly has adverse impacts

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on the world ecosystem (Canturk and Kulaç, 2021; Varol et al. 2021).

Among heavy metals (HMs), cobalt (Co) is a ductile and hard metal element naturally found in the earth's crust, water, and soil and is widely distributed in the environment. The chemical properties are very similar to nickel (Ni) and iron (Fe) (Barceloux and Barceloux 1999). Manganese (Mn) is a necessary nutrient and the 12th most abundant element (Agency for Toxic Substances and Disease Registry (ATSDR), 2012). Although Mn can naturally exist in the earth's crust, water, and soil in both organic and inorganic form, it can cause toxicity in humankind and animals under high exposure (Agency for Toxic Substances and Disease Registry (ATSDR), 2012). Nickel is used as a catalyzer in the chemistry and food industry and various metallurgic processes (production of nickel–cadmium batteries and alloy, electro-deposition). Due to some features, such as chemical properties, brightness, and affordability, Ni compounds are used in several commercial and industrial sectors (Genchi et al. 2020).

The entrance and increment of HMs into the body of plants is a complex mechanism formed by the mutual interaction of numerous factors. In addition, the mechanisms by which these elements play a role in the plant may be of great importance in plant development. For example, the elements (Ni, Co, and Mn) that are the research subject are classified as HM and micronutrients necessary for plant growth (Wani et al. 2018). In addition, these elements have been listed in the major contaminant category by the ATSDR (Agency for Toxic Substances and Disease Registry) because they are poisonous to living organisms at elevated amounts (Badea et al. 2018). The toxic impacts of HMs on tree species are commonly manifested as growth inhibition, low biomass production, altered water, and mineral nutrient balance, chlorosis, and, eventually, plant senescence (Wani et al. 2018).

HMs are the most poisonous group regarding human and environmental health among air pollutants. These three HMs, which accumulate within living organisms, can be harmful and lethal even at low amounts, and many are carcinogenic (Turkyilmaz et al. 2020). For instance, although these metals are used in several industrial productions (production of grinding, paint, and hard metals) (Barceloux and Barceloux 1999; Agency for Toxic Substances and Disease Registry (ATSDR), 2012; Genchi et al. 2020), they are considered to be one of the most dangerous HMs (carcinogenic for people) causing various respiratory and hematological health problems, such as cardiovascular diseases, asthma, contact dermatitis, respiratory tract cancer, and lung fibrosis (Barceloux and Barceloux 1999; ATSR 2012; O'Neal and Zheng 2015; Genchi et al. 2020). Furthermore, even though some elements (Zn, Cu, Fe, Mn, Ni, and Cr) are necessary as micronutrient elements for living things, they can be toxic at

low concentrations (Sevik et al. 2020). The very long half-life of HMs causes them not to be degraded easily in nature and to remain non-degraded for a prolonged period (Cesur et al. 2021).

The HM pollution in the air is considered more important than in any other medium from many aspects. HMs, which coalesce into the air after leaving their sources, can be transferred long distances and pollute other media, such as water and soil (Rajendran et al. 2022; Yang et al. 2022). Moreover, HMs inhaled into the liver pose a more critical hazard to human health (Ghoma et al. 2022). Hence, it is critical to observe the HM concentration in the air.

Besides monitoring HM concentration changes in the environment, reducing them is one of the priority research topics. Studies show that HMs cause stresses that result in reduced plant growth and even lead to death (Shahid et al. 2017; Wani et al. 2018). However, some plants can tolerate high HM concentrations and accumulate heavy metals in various organs while continuing their lives (Kowalik et al. 2021; Yaashikaa et al. 2022). These plants have a high potential for reducing heavy-metal pollution in the atmosphere, soil, and water (Hejna et al. 2021; Parihar et al. 2021). Monitoring and reducing HM pollution is highly critical for human and environmental health. For this purpose, identifying and effectively using plants that can accumulate pollution factors in their bodies but do not lose their lives due to pollution factors is one of the priority research subjects.

However, contrary to the water and soil environments, it is challenging to define the HM concentration in the atmosphere because the amount of HMs in the air is not stable. It is very costly and difficult to make a direct measurement and multiple measurements which take a long time but are necessary to gain information about the changes during a process. Thus, biomonitors are used in observing HM concentration variation in the air (Karacocuk et al. 2022; Cui et al. 2022; Isinkaralar et al. 2022b).

However, a method that can be used effectively in observing air pollution using biomonitors has yet to be developed. By making use of bryophytes as biomonitors, it cannot be determined how long the non-evergreen plant species have been subjected to HM contamination in the air. Non-evergreen plants reflect the 1-year amount of pollution, and since it is impossible to compare it with the available data, it cannot be easily interpreted (Cesur et al. 2021). The needles of evergreen plants such as *Picea* or *Abies* allow such a comparison and provide proof of the change of HM contamination in the air in the near past. However, using this method, one can achieve data about approx. 8–10 years of the period at the longest (Cetin et al. 2020).

It was reported that the most efficient method to be used in tracking HM pollution in the troposphere is the annual tree rings (Isinkaralar 2022; Key et al. 2022). Previous studies approved the usage of these annual tree rings in tracking,

especially the chemical contamination, and reported a connection between the element concentrations and environmental pollution in tree rings (Lee et al. 2015; Key and Kulaç 2022). Hence, on the condition of determining the accurate and suitable sampling strategy and selecting the suitable tree species, the use of annual rings is an efficient instrument for monitoring air pollution (Liu et al. 2018). The current study was designed to determine the variations in the concentrations of Ni, Co, and Mn, among the most dangerous and critical HMs for environmental health and human, in Kastamonu, using 180-year-old *Corylus colurna* L. annual tree rings.

C. colurna L. has been chosen because it can live for many years, has a robust root system, and does not rot for many years; therefore, its annual tree rings are very proper for the study. In some previous studies, the use of this

species as a biomonitor was investigated in monitoring Cd, Fe, Al, Pb, Cr, and Zn (Key and Kulaç 2022; Key et al. 2022) concentrations. Not much work has been done on the species that has been effective in choosing the species as the study material.

Materials and methods

In this study, a *C. colurna* L. tree (Turkish hazelnut) grown within the borders of Müsellimler Village of Ağlı province (coordinate: 41°38'13.14" N, 33°30'13.60" E) of Kastamonu city was used as material (Fig. 1). In the 2020 growing season, the northern direction was marked on the main trunk, and a tree with 69.7 cm diameter at breast height and 19.3 m height was cut down as it damaged the power line

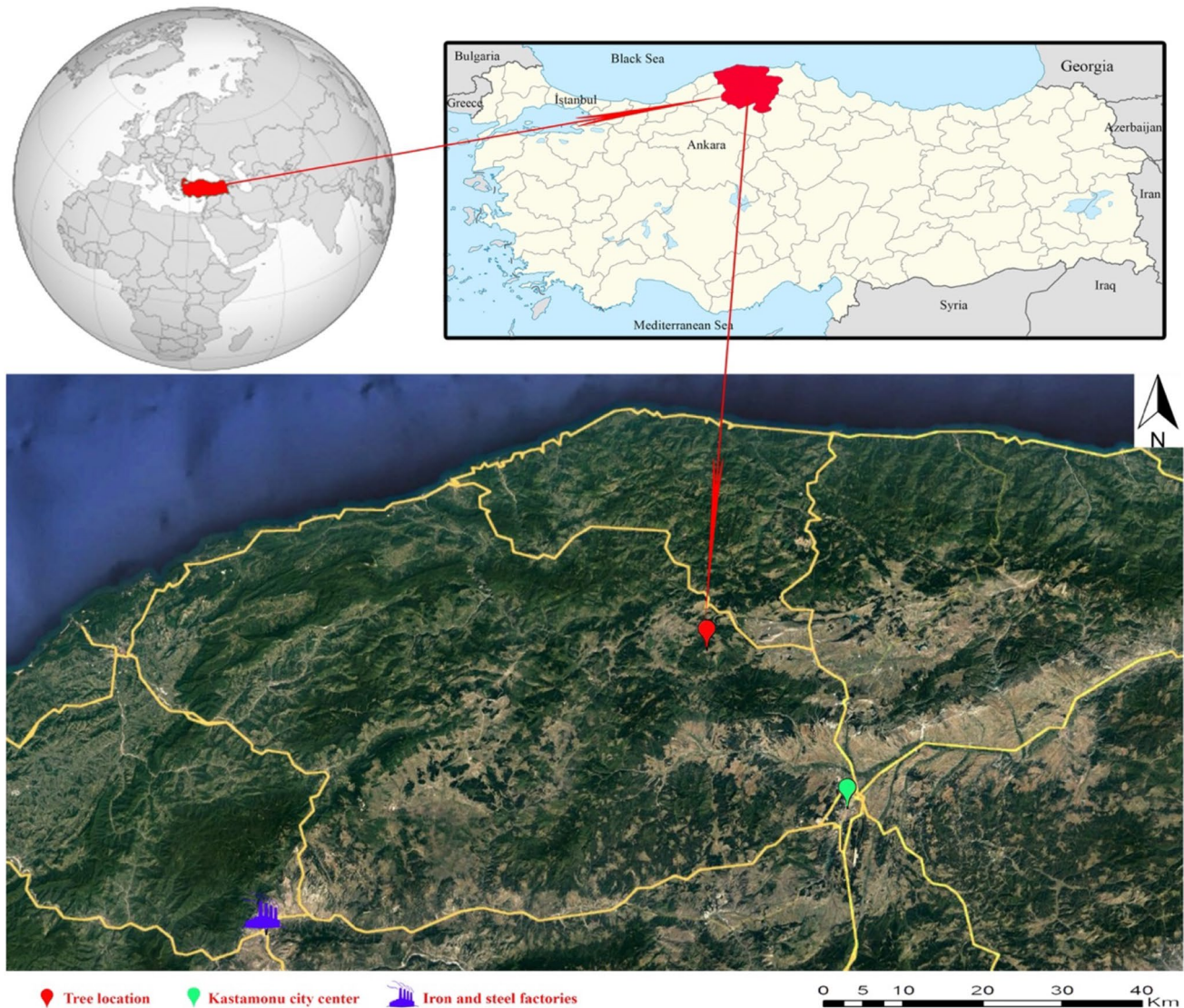


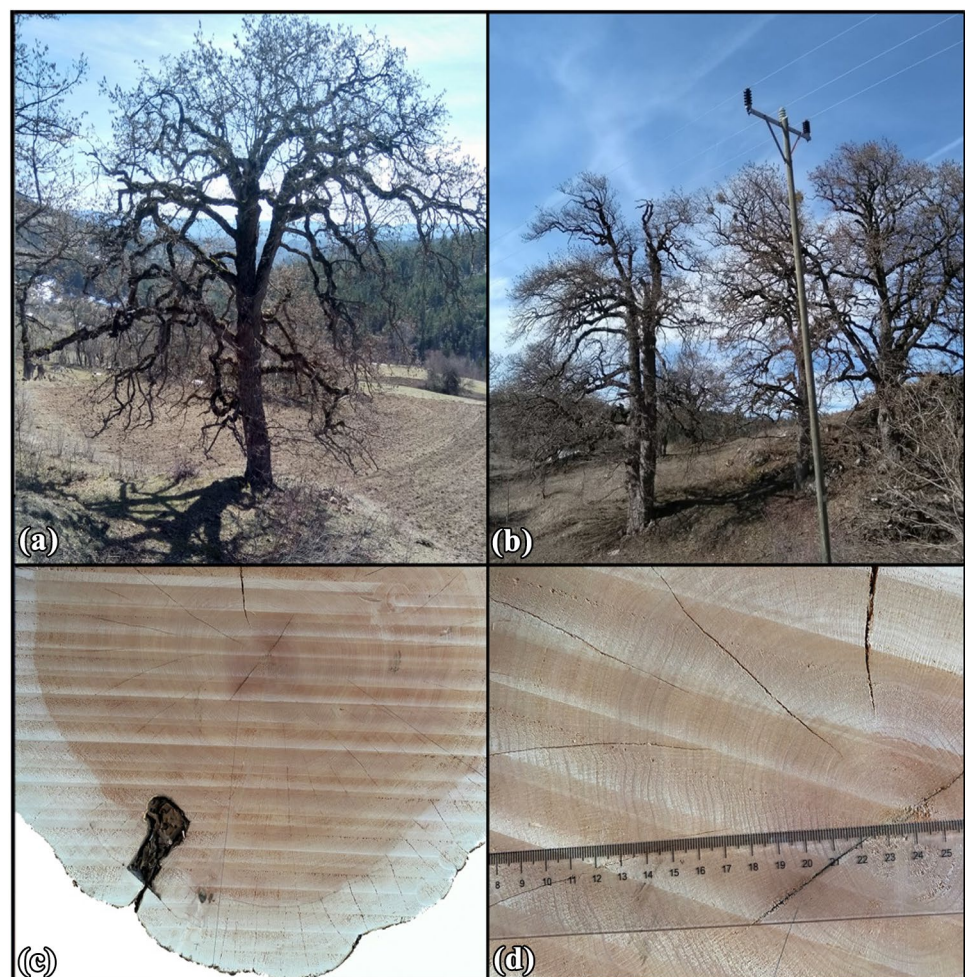
Fig. 1 Location of iron & steel plants and *Corylus colurna* tree (Key et al. 2022)

(Fig. 2a, b). Then, a 10-cm-thick log sample was obtained from 50 cm distance from the ground. The radius of the log sample measured 38.2, 40.9, 36.9, 38.7 cm from the north, south, west, and east, respectively (Fig. 2c, d).

The log sample was brought to the laboratory, each surface of the log was sanded off, and annual rings were made clearly visible. After the counting and analysis, it was approved that it was 180-years old. Regarding the ring widths, the annual tree rings were grouped as 10-year-old clusters, and outer bark, inner bark, and wood samples were taken from each age group by using a steel drill. The sampling was repeated three times in each organ and age group. Though it can be specified in which year the tree rings were constructed, specimens of wood chips cannot be obtained from the ring-shaped yearly because of the width of the annual ring. Hence, grouping tree rings on a log sample by their width and the tree's age were an easy solution used in the prior studies. In previous studies, 180, 55, 39, 30, 20, and 20-year-old tree species were separated as 10 (Key and Kulaç 2022; Key et al. 2022), 5 (Yigit 2019), 3 (Cobanoğlu et al. 2023), 3 (Koç 2021a; Savas et al. 2021), and 2 years (Turkyilmaz et al. 2019), respectively.

The samples, which were taken in the form of sawdust, were placed into glass petri containers and keeping the lids of containers open, the samples were air-dried for 15 days. Then, the samples of sawdust were dried for a week at 45 °C in a drying oven. Taking 0.5 g of dried sawdust samples, 6 ml 65% HNO₃ and 2 ml 30% H₂O₂ were added, and they were placed into the microwave designed for these analyses. Then, the samples, which were prepared as a solution, were taken into balloon flasks and filled to 50 ml using ultrapure water. The sample preparations were analyzed using the ICP-OES instrument (GBC Integra XL -SDS-270) (GBC Scientific Equipment Pty Ltd., Melbourne, Australia). Prior to the analysis of samples, the plasma of the ICP-OES instrument was burned, and ultrapure water was run through the device for 15 min to sterilize and equilibrate the system. After the calibration graph was built utilizing the standard solutions of the elements to be analyzed, the specimens were put into the device, and the reading procedure was implemented. Individual calibration graphs were generated at ppb or ppm levels, and re-reading was acted for test results that were not included in the calibration graph.

Fig. 2 a *Corylus colurna* tree, b its ecosystem, c wood specimen, d cross section of wood (Key et al. 2022)



The dilution factor was taken into account to recalculate the element concentrations. This method has been frequently used recently (Karacocuk et al. 2022; Key et al. 2022; Cobanoglu et al. 2023).

Statistical analysis

The obtained records were evaluated using SPSS 21.0 package software and conducting analysis of variance (ANOVA), and the Duncan test was performed for the factors having meaningful differences at the minimum confidence interval level of 95% ($P < 0.05$). The results were simplified and presented in graphics and tables.

Results

The ANOVA and Duncan test results on the variations of Ni concentration by plant part and direction are illustrated in Table 1. As a result of ANOVA, the variation in Ni amount by direction was significant ($P < 0.001$) for all plant parts. In all plant parts, the concentrations obtained from the east were clustered in the lowest groups as an outcome of the Duncan test, whereas the maximum Ni concentrations were seen in the north direction for inner bark and the west direction for outer bark. The wood results were clustered in two groups; the values received from the north and east directions were clustered in one group, and those obtained from the south and west were clustered in the other group.

It was found that the difference in Ni concentration by plant part was noteworthy only in the west and north sides ($P < 0.05$). In the Duncan test, the concentrations obtained from inner bark and wood were clustered in a group, and those taken from outer bark were clustered in the second group for the west direction. However, for the north direction, the values obtained from different organs constituted separate groups; the minimum values were seen in wood, and the maximum values were seen in the inner bark. The variations in Ni concentration in woods by season and facing side are illustrated in Table 2.

The results obtained from the ANOVA showed that there was a statistically meaningful ($P < 0.001$) change in Ni concentrations by directions in each period and by periods in each direction. The highest Ni concentrations were acquired mostly in the south and west directions. It was found that there was a significant decrease in the east direction after 1921–1930, while there was a remarkable increase in the north direction after 1971–1980. Moreover, there were notable variances among the directions in the same age category. For instance, it was determined that, in the period between 1841 and 1959, Ni concentration was found to be 1799.0 ppb in the west and 189.4 ppb in the north. Similarly, in the period between 2001 and 2010, Ni concentration was found to be 2243.6 ppb and 331.9 ppb in the south and east, respectively. The diagram illustrating the variation of Ni amount by directions and periods is illustrated in Fig. 3.

The results of the ANOVA and Duncan test on the changes of Co concentration by plant part and facing side are illustrated in Table 3.

Examining the change of Co amount by directions and tree part, there were statistically noteworthy ($P < 0.001$) variations by directions in all the tree parts. Duncan's test showed that, in all the organs, the values obtained from the east were clustered in the first group and those obtained from the west in the last group. Examining the changes by organs, the differences between organs were found statistically notable ($P < 0.01$) only in the north. In the north direction, the concentrations acquired from wood parts were clustered in the first group, and those obtained from barks were in the second group. The variations of Co amount in the wood part by facing side and periods are illustrated in Table 4.

Examining the variation of Co concentration by period and directions, the changes by directions and by period were significant ($P < 0.001$) in all periods and directions, respectively. Given the mean concentrations, it can be concluded that the Co amount coursed almost horizontally in the south and west directions. While it coursed almost horizontally in the north until the period 1951–1960, it started increasing since then. In the east route, the maximum Co concentrations were seen until the period 1911–1920, and remarkable

Table 1 The amount of Ni concentration (ppb) by part and direction

Organ	South	West	North	East	F value
Outer bark	1122.4 B	1938.5 Db	1466.8 Cb	643.7 A	652.485***
Inner bark	1043.8 B	1413.8 Ca	1989.5 Dc	440.2 A	7967.368***
Wood	1454.0 B	1307.8 Ba	707.3 Aa	630.3 A	54.874***
F value	1.324 ns	3.900*	29.737***	0.506 ns	

Means with a line and column for a direction and organs followed by the same letter are not different. Vertical and horizontal directions were expressed in lowercase and capital letters, respectively

*** and * indicate that means within a selection are significantly different at $P \leq 0.001$ and $P \leq 0.05$, respectively, while ns refers to no significant difference at $P > 0.05$

Table 2 Variation of Ni (ppb) amount by season and direction

Periods	South	West	North	East	F value
1841–1850	1565.5 Cgh	1799.0 Dn	189.4 Aa	671.4 Bh	5855.6***
1851–1860	1555.9 Cgh	1078.4 Bef	514.2 Acd	503.7 Ag	5137.3***
1861–1870	913.1 Ba	1183.6 Dh	538.3 Ade	1020.9 Cij	1538.1***
1871–1880	1131.0 Dc	1082.6 Cf	577.5 Aef	1043.2 Bj	650.8***
1881–1890	3315.2 Dj	933.3 Bb	616.5 Af	1041.2 Cj	9475.6***
1891–1900	1497.9 Cf	993.6 Bc	674.8 Ag	1012.4 Bi	1099.0***
1901–1910	1119.2 Cc	1047.2 Bd	540.0 Ade	1113.8 Ck	1739.1***
1911–1920	1101.1 Bc	1217.2 Ci	529.4 Ad	1236.2 Cl	2077.5***
1921–1930	1540.6 Dg	1319.6 Cj	528.5 Bd	476.3 Af	1747.3***
1931–1940	1575.5 Ch	1669.2 Dm	519.5 Bcd	368.3 Acd	5947.4***
1941–1950	1037.0 Cb	1770.4 Dn	547.7 Bde	383.8 Ad	3918.6***
1951–1960	1222.1 Dd	981.2 Cc	462.9 Bb	348.8 Abc	3553.3***
1961–1970	1400.9 De	1362.8 Ck	481.8 Bbc	335.2 Ab	12191.5***
1971–1980	1244.4 Cd	1116.0 Bg	1103.0 Bh	340.4 Ab	4233.2***
1981–1990	1215.0 Bd	1623.5 Dl	1292.5 Cj	419.6 Ae	1593.1***
1991–2000	928.7 Ba	2434.4 Do	1390.8 Ck	393.0 Dd	2507.3***
2001–2010	2243.6 Ci	1051.4 Bde	1079.1 Bh	331.9 Ab	6244.7***
2011–2020	1564.8 Dgh	876.6 Ba	1146.7 Ci	306.2 Aa	498.8***
F value	2753.179***	1574.634***	537.931***	1652.1***	

Means with a line and column for a direction and organs followed by the same letter are not different. Vertical and horizontal directions were expressed in lowercase and capital letters, respectively

***Indicates that means within a selection are significantly differ at $P \leq 0.001$

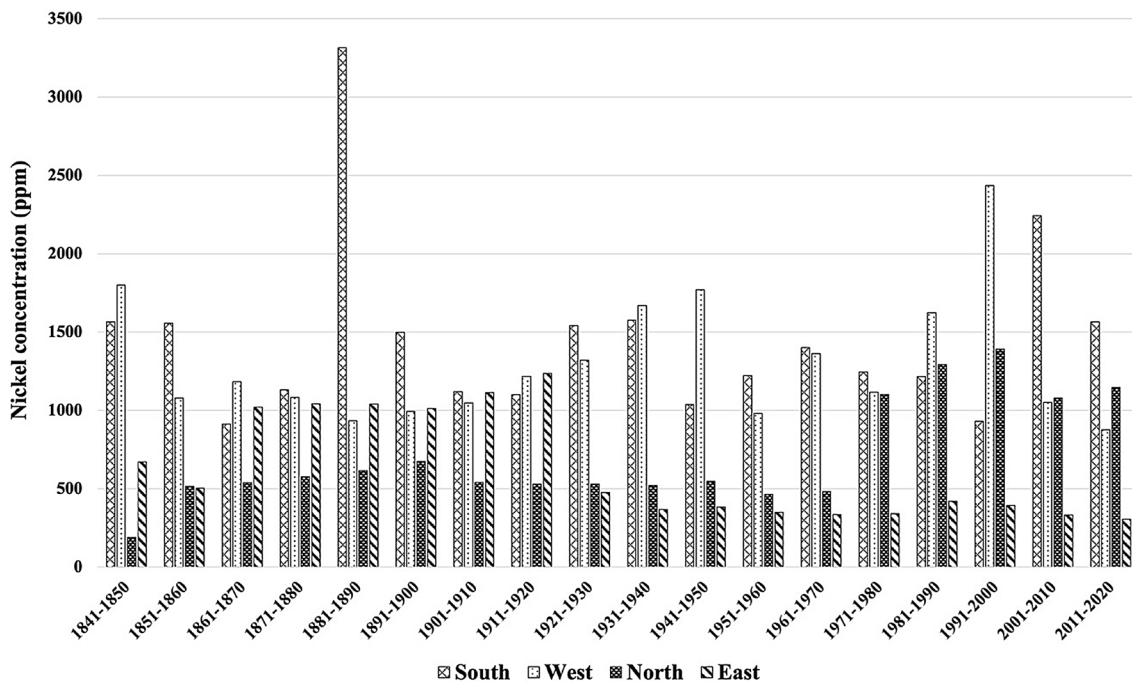


Fig. 3 Ni concentration (ppm) variation in wood by directions and periods

Table 3 The amount of Co concentration (ppb) by direction and plant part

Organ	South	West	North	East	F value
Outer bark	367.5 B	565.6 C	352.2 Bab	274.5 A	414.368***
Inner bark	372.3 B	491.8 C	381.9 Bb	179.7 A	651.153***
Wood	373.4 B	406.0 B	247.1 Aa	262.2 A	32.117***
F value	0.077 ns	2.051 ns	6.400**	0.941 ns	

Means with a line and column for a direction and organs followed by the same letter are not different. Vertical and horizontal directions were expressed in lowercase and capital letters, respectively

*** and ** indicate that means within a selection are significantly differ at $P \leq 0.001$ and $P \leq 0.01$, respectively, while ns refers to no significant difference at $P > 0.05$

decreases were observed since that period. The variation of Co amount by directions and age category is shown in Fig. 4.

The ANOVA and Duncan test results on the variation of Mn concentrations by tree part and facing side are illustrated in Table 5.

Examining the alteration of Mn concentrations by organ and direction, as in Ni and Co, the variations by directions were found statistically meaningful ($P < 0.001$) in the whole tree parts. The lowest concentrations were taken from the east side, and the maximum values from the north and west in barks. In woods, the values were gathered in two groups; those obtained from the north and east were clustered in

the first group, and those obtained from the south and west constituted the second group. The ANOVA results by organ revealed that the change of Mn concentration was meaningful ($P < 0.001$) in all directions. The lowest Mn concentrations were acquired from woods, and the maximum values from outer bark in all directions. It is attention-grabbing that there were tenfold differences between the values obtained from wood in the north and west directions. The variation of Mn amounts in the wood section by facing side and period is illustrated in Table 6.

The results obtained from ANOVA showed that, as in Ni and Co concentrations, Ni concentrations were statistically meaningful ($P < 0.001$) and varied by direction in all periods and by the period in all directions. Evaluating the changes by period, it is clear that there was a fluctuating and almost-horizontal course in the east but an upwards course in other directions in general. This increase is at a higher level in the south and west directions. The variation of Mn amount by directions and age category is illustrated in Fig. 5.

Discussion

In the current study, the variation of Ni, Mn, and Co element amount in outer bark, inner bark, and 180-year-old annual tree rings of *C. colurna* L. by direction was determined. It was found that Ni (west and north), Co (north), and Mn

Table 4 Co concentration variations (ppm) in wood by age and direction

Periods	South	West	North	East	F value
1841–1850	420.1 Cfg	506.8 Df	182.5 Aabc	226.1 Bd	337.1***
1851–1860	387.6 Cde	533.8 Dg	169.6 Aa	229.0 Bd	636.2***
1861–1870	393.6 Bde	415.2 Ce	169.9 Aa	383.4 Be	725.0***
1871–1880	388.2 Cde	360.7 Bcd	174.3 Aab	413.5 Dg	508.0***
1881–1890	359.2 Babc	372.7 Bd	248.1 Ae	410.9 Cg	280.0***
1891–1900	365.1 Cbc	348.4 Babc	195.6 Acd	397.2 Df	328.7***
1901–1910	352.3 Cab	322.8 Ba	207.8 Ad	401.8 Dfg	409.3***
1911–1920	341.1 Ba	337.7 Babc	189.9 Abc	447.4 Ch	258.6***
1921–1930	347.6 Cab	358.0 Dbed	188.8 Bbc	155.6 Aa	1322.1***
1931–1940	362.0 Dabc	343.5 Cabc	210.3 Bd	181.8 Ac	323.5***
1941–1950	349.0 Cab	341.0 Cabc	238.0 Be	163.7 Aa	655.6***
1951–1960	378.4 Ccd	342.8 Babc	181.9 Aabc	190.7 Ac	277.9***
1961–1970	361.0 Cabc	352.7 Cabcd	245.6 Be	186.4 Ac	318.6***
1971–1980	365.0 Bbc	353.2 Babcd	354.2 Bf	187.2 Ac	222.8***
1981–1990	366.2 Bbc	348.9 Babc	369.9 Bg	166.1 Aab	164.2***
1991–2000	356.0 Babc	986.0 Ch	356.9 Bfg	221.3 Ad	1872.4***
2001–2010	401.5 Cef	339.0 Babc	345.2 Bf	177.6 Abc	85.5***
2011–2020	426.9 Cg	334.2 Bab	420.1 Ch	181.1 Ac	530.6***
F value	12.485***	487.916***	255.114***	630.031***	

Means with a line and column for a direction and organs followed by the same letter are not different. Vertical and horizontal directions were expressed in lowercase and capital letters, respectively

***Indicates that means within a selection are significantly differ at $P \leq 0.001$

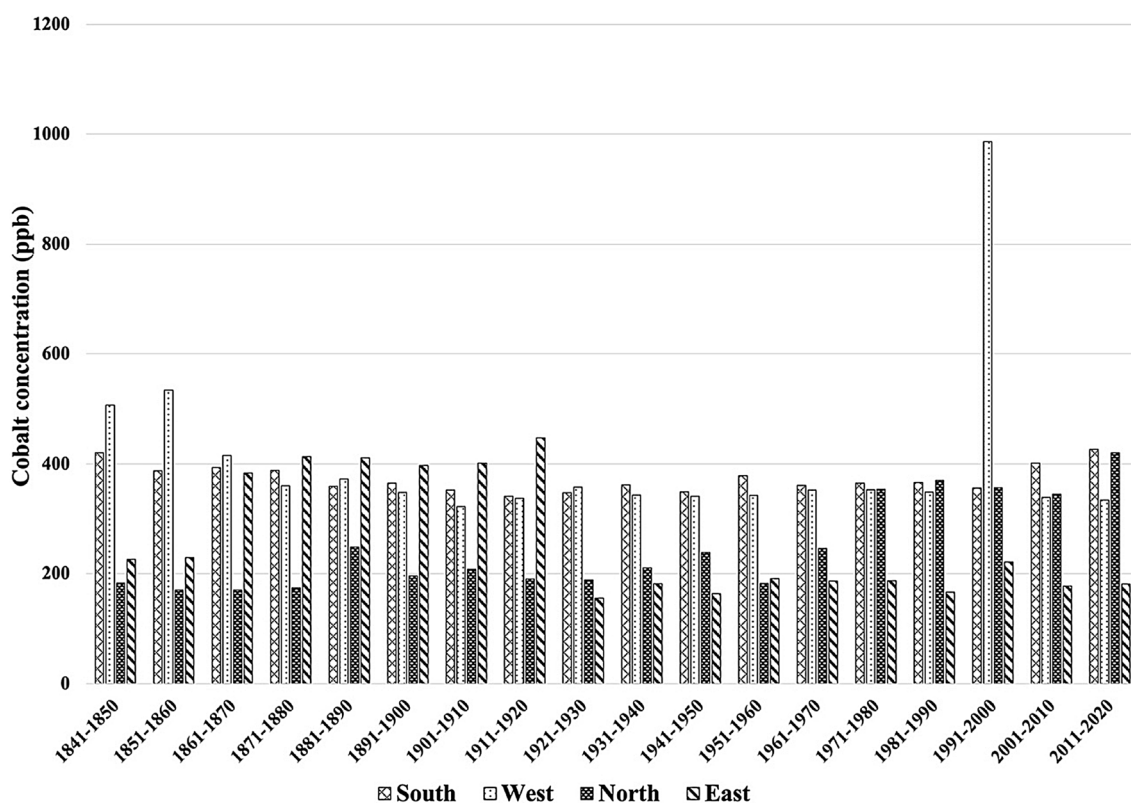


Fig. 4 Co amount (ppb) in wood part by direction and periods

Table 5 The amount of Mn (ppb) concentrations by tree part and facing direction

Organ	South	West	North	East	F value
Outer bark	10647.9 Bc	18062.8 Dc	14400.2 Cc	9689.6 Ac	3361.077***
Inner bark	8137.2 Bb	8787.6 Cb	9749.9 Db	4816.3 Ab	17690.716***
Wood	1786.8 Ba	1722.3 Ba	1434.8 Aa	1507.3 Aa	7.444***
F value	570.780***	1253.890***	3177.678***	2854.106***	

Means with a line and column for a direction and organs followed by the same letter are not different. Vertical and horizontal directions were expressed in lowercase and capital letters, respectively

***Indicates that means within a selection are significantly differ at $P \leq 0.001$

(all directions) concentrations obtained in barks were higher than in wood. Especially the outer bark element concentrations were much greater than in wood for many elements (Cesur et al. 2021). This is mainly because of the particles in the air (Hozhabralsadat et al. 2022). The HMs investigated in the present study are those determined to generally increase with the traffic (Jin et al. 2022), and there is a very busy highway north of the location where the samples were collected. After leaving the source, HMs can be spread out over long distances by adhering to the particles, and the heavy-metal-contaminated particles adhering onto the rough outer bark can cause high levels of HM concentrations (Turkylmaz et al. 2018). Hence, previous studies on this subject determined that Ni and Co concentrations in outer

bark were very high at high-traffic locations (Sevik et al. 2020; Koç 2021a).

The current study pointed out that Ni, Co, and Mn elements' concentrations in the wood remained higher than the measurable limits in all periods and all years. It suggests that *C. colurna* has a high potential to accumulate Ni, Co, and Mn elements in wood. Tracking the air metal contamination in the atmosphere is very difficult and pricy. Thus, generally, biomonitors are used to observe the HM pollution airborne. Nevertheless, it was reported that, among the biomonitors currently used, the most appropriate one is the annual rings of trees because their use can deliver substantial proof of the variation of HM over time (Savas et al. 2021; Mulenga et al. 2022). Besides, since the research is performed on the same

Table 6 The amount of Mn concentrations (ppb) in wood part by facing side and period

Periods	South	West	North	East	F value
1841–1850	1260.5 Ba	1635.3 Dg	955.0 Aa	1323.3 Cc	1103.1***
1851–1860	1237.9 Ca	1289.2 Dc	1047.6 Ab	1181.0 Ba	389.5***
1861–1870	1287.2 Cb	1259.5 Bb	1161.0 Ad	1325.0 Dc	195.3***
1871–1880	1327.5 Bc	1414.6 Ce	1195.4 Ae	1598.4 Di	4051.7***
1881–1890	1514.0 Ce	1373.4 Bd	1358.0 Ag	1687.4 Dk	2224.0***
1891–1900	1433.6 Cd	1174.4 Aa	1192.1 Be	1474.7 De	1473.1***
1901–1910	1585.0 Cg	1247.0 Bb	1087.1 Ac	1723.4 Dl	9766.8***
1911–1920	1827.4 Bk	1257.3 Ab	1220.5 Af	2012.0 Cm	582.9***
1921–1930	3343.3 Do	1480.1 Af	1388.7 Bhi	1633.8 Cj	153921.3***
1931–1940	1686.0 Di	1437.3 Be	1374.6 Ah	1543.0 Cg	1964.2***
1941–1950	1545.1 Af	1969.8 Dj	1610.4 Cj	1577.4 Bh	2540.1***
1951–1960	1717.0 Cj	1656.7 Bg	1395.9 Ai	1402.3 Ad	3469.6***
1961–1970	2301.4 Dm	1995.7 Cj	1382.2 Bhi	1284.6 Ab	21777.0***
1971–1980	1904.1 Cl	1881.3 Ci	1752.3 Bl	1643.1 Aj	42.4***
1981–1990	1718.2 Cj	2305.6 Dl	1701.0 Bk	1513.0 Af	4980.6***
1991–2000	1658.6 Bh	3833.6 Dm	1991.0 Cm	1598.7 Ai	31964.6***
2001–2010	1922.1 Bl	2045.9 Dk	1980.8 Cm	1281.5 Ab	6135.9***
2011–2020	2894.2 Dn	1745.2 Bh	2033.8 Cn	1329.6 Ac	17525.7***
F Value	4670.323***	4406.031***	4116.517***	1523.561***	

Means with a line and column for a direction and organs followed by the same letter are not different. Vertical and horizontal directions were expressed in lowercase and capital letters, respectively

*** indicates that means within a selection are significantly differ at $P \leq 0.001$

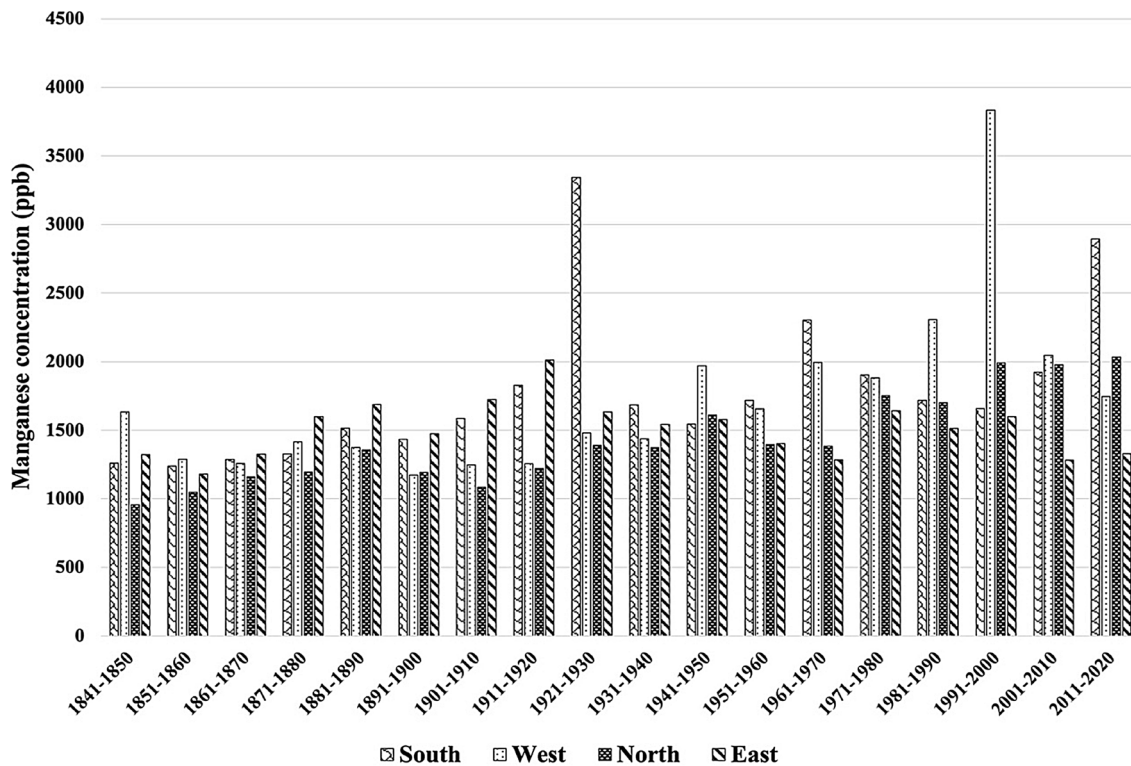


Fig. 5 Mn concentration (ppb) variation in wood part by period and direction

plant species, it avoids deviations from the genetic makeup. Plant expansion is formed by the interplay between environmental elements and genetic structure (Koç et al. 2021). Therefore, plant species with different genetic makeup may have different phenotypic and physiological traits even under the same environmental conditions (Koç 2021b; Koç and Nzokou 2023). Genetic makeup can also influence the HM accretion potential of the plant species (Karacocuk et al. 2022).

Many HMs, including Ni, Co, and Mn, used in this research are necessary micro or macronutrients for plant expansion (Riaz et al. 2021). Nevertheless, high element concentrations, particularly in the soil, cause plant stress and reduce plant expansion (Wani et al. 2018). HMs can store in the tree parts by being captivated by belowground organs, through the atmosphere via the tree leaves, and straight penetrating the tree trunk (Chen et al. 2021). HM ions join the plant body via the symplastic or apoplastic pathway depending on the absorbed ions classification. The plant species can inhibit HM accretion by precipitating metal elements or making complex metals in the rhizospheric area (Wani et al. 2018).

The studies carried out on the same tree eliminate the differences arising from the genetic makeup. However, it suggests that the plant species have dissimilar HM accretion potentials (Cesur et al. 2021). Therefore, it is necessary to determine which annual tree rings of individual plants can be used as biomonitors for which metals. In some of the experiments carried out to date, in determining the change of concentrations over time, *Populus* spp. was used for Pb, Hg, and Cd (Bardule et al. 2020), *Pinus halepensis* for Hg, Cd, and Pb (Martin et al. 2018), *Tilia platyphyllos* and *Populus nigra* for Zn and Pb (Kiss et al. 2019), *Pinus montezumae* for Zn and Pb (Baieta et al. 2021), *Populus bonatii* and *Ailanthus altissima* for Zn and Cd (Liu et al. 2018), *Cedrus atlantica* for Mn and Cr (Savas et al. 2021), *Larix decidua* for Hg (Nováková et al. 2022), *Swietenia mahagoni* for Fe, Zn, Pb, and Cu (Edusei et al. 2021a, b), *Picea abies* and *Pseudotsuga menziesii* for Pb, Sr, Co, and Ni (Milosevic et al. 2022), *Quercus robur* for Mn, Cu, Cd, Pb, Zn, and Ni (Nechita et al. 2021), *Quercus pubescens* for Cu, Pb, Cr, Co, Ni, Tl, Hg, U, Mo, W, V, and Zn (Perone et al. 2018), *Pinus massoniana* for Zn, Mn, Cd, Fe, Cu, Pb, Cr, Sr, Ni, and Co (Chen et al. 2021).

Previous research claimed that the most important lack of knowledge is about transferring of HMs within the plant (Shahid et al. 2017). Examining the data obtained in the present study, it can be stated that the transfer of elements, which were investigated in this study, within the wood is very limited. For instance, it was reported that Ni concentration was 1799.0 ppb in the west and 189.4 ppb in the period 1841–1950 and, similarly, it was 2243.6 ppb and 331.9 ppb in the south and east in the period of 2001–2010. Similar

results were also achieved for Co and Mn. It suggests that the transfer of elements, which are examined here, within the wood is limited. There is limited information since only a few studies are focused on this subject. Koç (2021a) pointed out that the movement of Ni and Co elements in the wood of *C. atlantica* was restricted. Zhang (2019) determined that Pb and Zn concentrations shifted slightly between the annual tree rings, but the concentration of Cu element did not shift at all. Cesur et al. (2021) claimed that the transport of Ni and Cd element concentrations in the wood of *Cupressus arizonica* was restricted, but the transfer of Bi concentration was at a higher level.

The element movement in the plant wood part is primarily associated with the cell wall and structure. The CWPM (cell wall–plasma membrane) interface defines an apoplastic mechanical obstacle, and an adjustable structure takes part in stress signaling, sensing, and perceiving the metalloids and metal stress. The CWPMs, involved in answering several abiotic strains, have been widely identified and indicated among diverse agricultural plants. Under various unfavorable environmental situations, the main (CWPM) contains transcription factors, phospholipases, dehydration-sensitive element-binding proteins, C-repeat binding factor, SOS (salt excessively sensitive kinases), mitogen-activated protein kinases and phosphatases, C-repeat binding factor, and abscisic acid-responsive-binding elements. The CWPM edge is considered the possible spot of HM tolerance as it collects big HMs particles (Wani et al. 2018).

The highest amounts of Co, Ni, and Mn in wood were obtained in the south and west directions in the current study. Although the sampling was performed at a location that is moderately far from the HM resources, such as mining plants (Alaquori et al. 2020), traffic (Aricak et al. 2020; An et al. 2022), and urban areas (Huang and Gergel 2022), the largest steel and iron plant in Turkey is located at approx. 88 km southwest of the sampling area. After leaving their sources, previous studies revealed that HMs could be conveyed to far distances via wind (Key and Kulaç 2022; Key et al. 2022). Moreover, it was also noted that iron and steel factories are among the primary sources of HMs examined here (Dai et al. 2015). Hence, high HM concentrations obtained in wood in the west and south routes can be correlated to these factories.

Conclusions and suggestions

This research focused on specifying the usage of the annual tree rings of *C. colurna* in monitoring the 180-year alteration of Ni, Co, and Mn element concentrations. The results achieved here suggest that the annual tree rings of *C. colurna* are proper in tracking the alteration of Ni, Co, and Mn concentrations. To individually determine the most suitable

tree species to be used in observing which HM pollution, it is necessary to carry out further studies on the tree species widely grown in city centers, high-traffic areas, mining areas, and urban areas, where the HM pollution is at a high level.

In the method used in this study, the annual rings must be obtained. The most efficient way for this purpose is to cut the tree. However, the samples can be taken without cutting the trees but by using an increment borer. Hence, examinations can be made on different trees, and more detailed evidence about the alteration of HM concentrations in the region can be achieved. Moreover, substantial evidence about the variation of HM concentration in the recent period can be obtained by taking samples from side branches instead of the main trunk.

It was concluded that *C. colurna* annual tree rings are appropriate for observing the variation in the amounts of Ni, Co, and Mn elements in the atmosphere. In addition, some elements are very detrimental to individual and environmental health, so observing the alteration in their concentrations in the atmosphere is crucial. It should be investigated whether the annual tree rings of *C. colurna* are proper for examining the alteration of these elements. In studies to be carried out on this issue, it is recommended to give priority to some elements (Hg, Ag, Pb, Pd, As, Zn, Al, Ba, Sb, Be, Cd, Sr, Cr, U, Pu, Se, Cu, Tl, Th, and V), which are on the priority contaminant checklist by ATSDR, since they are poisonous to living organisms at elevated concentrations.

The results show that there has been a substantial accumulation in Co and Ni concentrations since 1971, especially in the north direction. It is estimated that this rise is most likely related to the highway in this direction. In this case, it can be seen that the Ni and Co concentration in the air rises significantly depending on the vehicle traffic. Co and Ni are highly harmful elements to individual and environmental health, and high concentrations of these elements are hazardous, especially in areas with heavy traffic. It has been determined that the species subject to the study can store these hazardous elements at a high rate in the wood part. Therefore, this species can be actively used in monitoring and reducing Ni and Co pollution. Moreover, these results indicate that Mn is not directly related to the traffic.

Author contribution statement KK: design, raw material collection, performed the analysis, writing. ŞK: design, raw material collection, performed the analysis, and reviewing. İK: data analysis writing, reviewing, and editing. HS: contributed to the study designed, data analysis, and reviewing and editing. All authors read and approved the final manuscript.

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