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RESEARCH ARTICLE

Use of tree rings as a bioindicator to observe atmospheric heavy metal deposition

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Abstract
Trees can be used as good indicators to evaluate the increase in atmospheric heavy metal concentrations. In the last two decades, air pollution in the city of Ankara has rapidly increased with the ever-increasing traffic density. In the present study, the depositions of aluminum (Al), zinc (Zn), copper (Cu), cobalt (Co), iron (Fe), manganese (Mn), chrome (Cr), cadmium (Cd), sodium (Na), calcium (Ca), barium (Ba), phosphor (P), magnesium (Mg), arsenic (As), and boron (B) in the rings of oak trees were analyzed using a GBC Integra XL–SDS-270 ICP-OES device. The study found that heavy metal concentrations in tree rings varied over the past 20 years; furthermore, there was a significant relationship between the heavy metal concentrations in tree rings and the atmospheric heavy metal concentrations. There was an increase in the concentrations of nutritional elements (Na, P, and Mg) in 2010 when there was excessive precipitation. As a result, the concentrations of all elements in the woods of different ages were significantly different at a confidence interval of 95% for As, 99% for Cd, and 99.9% for other elements.

Keywords Atmospheric pollution · Annual ring · Heavy metal · Bioindicator · Oak

Introduction

The life, health, food, and shelter needs of people have been rapidly increasing with the increase in the world's population, which has caused considerable environmental pollution. Environmental pollution occurs in two forms. Natural pollution is caused by the wastes of all living things except humans and direct wastes of humans. Natural pollutants are eliminated by natural recycling mechanisms in a short period of time. However, anthropogenic pollutants, particularly those arising from industrial activities and exhaust smoke of vehicles, are retained in the environment for long periods and have unfavorable effects on human health. Atmospheric heavy metals are transported into the soil by precipitation, and air-borne pollutants when inhaled cause various diseases in humans. The concentrations of heavy metals in the environmental sources are constantly increasing. These heavy metals also tend to show bioaccumulation. It is therefore important to determine heavy metal concentrations and identify the regions at risk and determine their risk level. However, two problems arise while directly evaluating atmospheric pollution. First, it is expensive and second, it is not possible to determine direct effects of atmospheric pollution on the ecosystem (Kaya 2009; Kaya et al. 2009; Cetin et al. 2010; Cetin 2016; Cetin 2017; Alahabadi et al. 2017; Turkyilmaz et al. 2018a; Cetin et al. 2017; Sevik and Cetin 2015; Cetin et al. 2018; Kaya et al. 2018).

Plants accumulate heavy metals within various organelles and thus provide important data about concentrations of heavy metals in the air. Particularly in areas with high traffic intensity, trees accumulate heavy metals arising from fossil fuels in their trunks, roots, fruits, barks, and leaves and thus indicate the increase in heavy metal concentrations in the air in the

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course of time. Plants are the best bioindicators of heavy metal pollution in the air (Shahid et al. 2017; Janta and Chantara 2017).

Heavy metal deposition essentially occurs in the soil. However, the uptake of heavy metals from the soil into the roots and the deposition of heavy metals in the above-surface parts render plants a good bioindicator. Various organelles of the trees have been used for a long time to determine heavy metal concentrations (Turkyilmaz et al. 2018a; Shahid et al. 2017; Sawidis et al. 2011; Turkyilmaz et al. 2018b).

Plant foliage is the most commonly used organelles to determine heavy metal concentrations. The plant leaves uptake heavy metals during photosynthesis through their stomas; removal of the leaves does not cause permanent damage to the tree, and the period during which a particular heavy metal was deposited can be estimated as we know the age of the tree. In fact, trees are not as good indicators as fungi, algae, and mosses. However, considering the fact that trees are found in every part of a city and they survive longer than other indicators, trees provide more information about the increase in atmospheric heavy metal pollution from past to present (Turkyilmaz et al. 2018a; Shahid et al. 2017; Sawidis et al. 2011).

Trees living for a long time particularly in areas with high traffic intensity provide important information about heavy metal deposition in that particular area. Coniferous trees such as pine tree, spruce tree, and fir that retain their needles for a long period of time, the age of which can be approximated accurately, are suitable to determine heavy metal deposition in the recent past (Turkyilmaz et al. 2018a; Sevik et al. 2017a). However, this would provide information for the past 8–10 years; other organelles of the trees can be used to obtain information for older periods of time.

Heavy metals deposited in the tree rings through long ages provide important information about the history of air pollution. Annual rings of the trees are in fact related to the tree's age and there are trees that can survive for thousands of years. Tree rings can be used as an indicator of pollution, and they provide important information about the distribution of elements and chronologic order of pollution in that particular area wherein the tree grows (Beramendi-Orosco et al. 2013). Many studies have shown that trees are capable of accumulating the pollutants in their annual growth rings (Perone et al. 2018). Dendrochemistry is an area of research that focuses on the chemical composition of tree rings and the transport and history of chemical elements (Liu et al. 2018).

The aim of the present study is to determine heavy metal concentrations in annual growth rings of oak tree reflecting the 20-year ring history in samples obtained from the Keçiören district of the province of Ankara, the capital city and one of the largest cities in Turkey, which is a highly populated area and has the highest traffic intensity. We therefore aimed at determining the changes in heavy metal concentrations in the air derived from the traffic that cannot be directly measured. In the scope of the study, we attempted to determine the utility of this plant type in evaluating heavy metal concentration in the recent past.

Material and method

The study was conducted on samples collected from the Keçiören district of the province of Ankara, which is one of the districts with the highest traffic intensity. The samples were cut in the month of December following the completion of vegetation at the end of 2016 and were brought to the laboratory. Disks (1 cm thick) were cut from the samples at the laboratory, and the structure of tree rings was rendered visible by rubbing the disk surface. The samples were then divided into 2-year sections. The samples obtained from two opposing sides of the tree were classified and this yielded 11 samples, including ten 2-year rings and bark.

Wood samples were processed into woodchip. A particular attention was paid to avoid the use of instruments made from the elements analyzed in the study. The samples were allowed to sit for 30 days for room drying and then dried in an oven at 50 °C for one week. Six milliliters of 65% HNO_3 and 2 mL of 30% H_2O_2 were added on to 0.5 g of dried samples and then placed in a microwave oven. The microwave device was heated to 200 °C for 15 min and the temperature was maintained for 15 min. The samples were incinerated in a Milestone brand Ethos one model microwave oven, and the dispersed samples

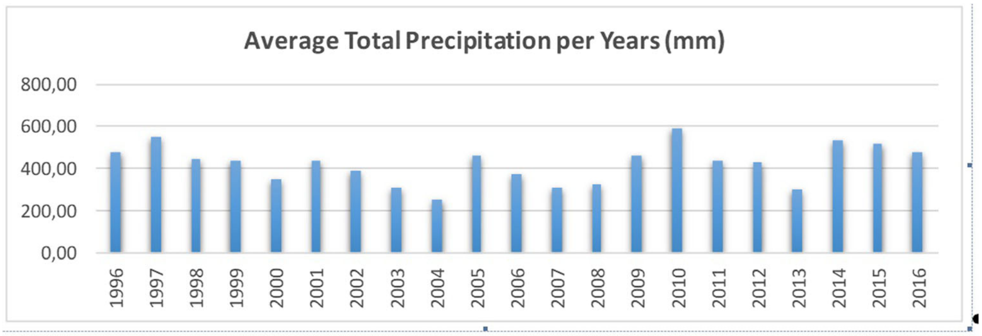


Fig. 1 Precipitation graph for the period from 1996 to 2016 in the study area

within the solution were transferred to flasks and diluted in a total volume of 50 mL with ultrapure water, which was used for the analysis of Fe, Co, Ni, Zn, Cd, Hg, and Pb concentrations using a GBC Integra XL–SDS-270 ICP-OES device. Then, plasma of the ICP device was incinerated for the analysis of the samples, and the system was rinsed with ultrapure water for 15 min to maintain the balance. Standard solutions were prepared for the elements to be analyzed and a calibration chart was created. The samples were fed into the system and read after creating the calibration chart. For the analysis results that did not fall into the calibration chart, the samples were re-read by creating different calibration charts at ppm and ppb levels. Detection limits for GBC Integra XL–SDS-270 ICP-OES device are as follows: Pb, 0.377 ppb; Cu, 0.639 ppb; Ca, 0.00208 ppm; Mg, 0.00758 ppm; Cd, 0.063 ppb; Cr, 0.311 ppb; Ni, 0.171 ppb; Fe, 0.00068 ppm; Mn, 0.00015 ppm; and Zn, 0.00634 ppm. All measurements in the study were repeated three times. The data were then processed using the SPSS software package.

The studies to date have established that heavy metal deposition in the plants is related to precipitation. Thus, the data obtained from the study were interpreted in relation to the amount of rainfall. Precipitation graph for the period from 1996 to 2016 in the study area is presented in Fig. 1.

Results

A total of 17 elements were studied; among these elements, the results for Ni could not be evaluated as the values were below the detection limits. The data obtained for other elements were analyzed using analysis of variance and Duncan test; mean values obtained for wood and bark and *F* values calculated by the analysis of variance are presented in Table 1.

When the results in Table 1 were evaluated, there was no significant difference in the concentration of As among other elements between wood and bark at a confidence level of 95%; Pb concentrations in the wood were below the detection limits, and for all other elements, there were statistically significant differences between wood and bark. According to the *F* values, concentrations of all elements other than As and Pb are significantly different between wood and bark at a confidence interval of 99.9%.

The data show that concentrations of all elements measured in the bark are higher than those measured in the wood. On average, values measured in the bark are 6-fold higher than those measured in the wood. Among elements that showed statistical significance, the difference was the lowest for (2.35-fold) Al (2.72-fold) and Zn (3.13-fold), and the highest for Fe (16.36-fold), Cr (10.38-fold), and P (9.79-fold).

The tree examined in the study was a 20-year-old tree and annual rings were grouped in pairs; the samples were obtained

Table 1 Mean values for wood and bark and results of the analysis of variance>

	Al ppb	Zn ppb	Cu ppb	Co ppb	Fe ppb	Mn ppb	Cr ppb	Cd ppb	Pb ppb	Na ppb
bark	25,567	13,224	5823	418,6	186,299	19,667	1297,43	111,66	1914	227,833
wood	9402	4229	1839	116,4	11,389	2519	125,03	16,37	–	68,396
F	974,293***	59,271***	180,748***	539,140***	2699,901***	140,007***	4162,748***	1300,755***		438,801***

	Ca ppb	Ba ppb	P ppb	Mg ppb	As ppb	B ppb
bark	3205,20	21,266	669,233	1166,60	1300	25,566
wood	1364,33	3603	68,326	142,56	1273	6186
F	224,851***	1728,921***	1066,477***	1957,689***	0,303 ns	1287,681***

***significant at 0.001 level

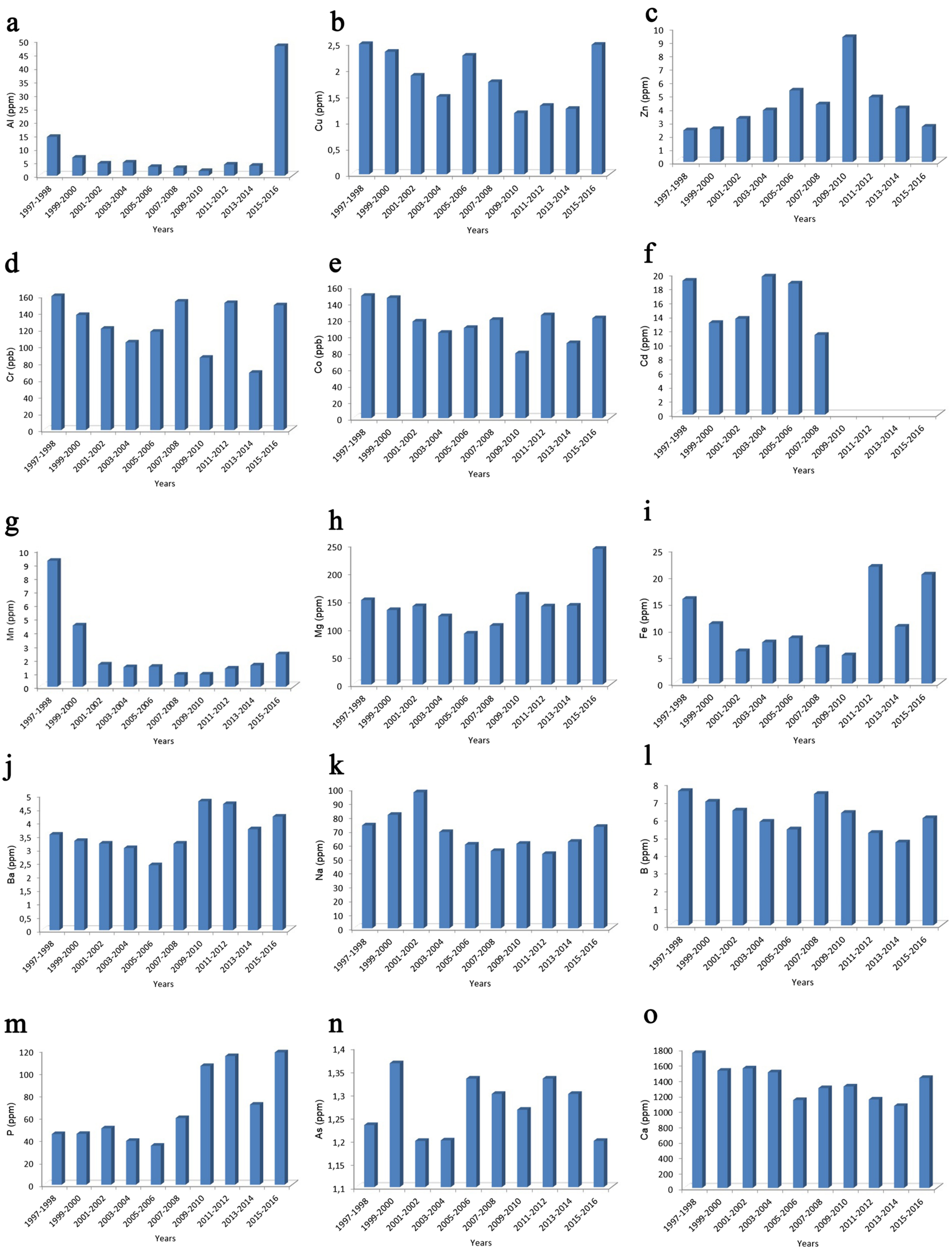
Table 2 Mean values in the wood samples according to age and the results of the analysis of variance

Age	Al ppb	Zn ppb	Cu ppb	Co ppb	Fe ppb	Mn ppb	Cr ppb	Cd ppb	Na ppb	Ca ppb
1997–1998	14,364i	2358a	2483g	148,6f	15,788h	9195g	159,6f	19,0b	73,600f	1742,766g
1999–2000	6689h	2449a	2334f	146,0f	11,109g	4462f	137,0e	13,0a	81,200g	1513,766e
2001–2002	4523f	3230b	1879e	117,3cde	6030b	1623d	120,6d	13,6a	97,300h	1543,466f
2003–2004	4883g	3857c	1481c	103,6bc	7708d	1429c	104,3c	19,6b	68,833e	1,493,033e
2005–2006	3258c	5336e	2261f	109,6cd	8482e	1456c	117,0d	18,6b	59,833c	1133,566b
2007–2008	2864b	4295cd	1761d	119,3de	6748c	0,879a	153,0f	11,3a	55,233b	1288,466c
2009–2010	1757a	9306f	1168a	78,6a	5268a	0,891a	86,3b	–	60,433c	1309,633c
2011–2012	4136e	4827de	1308b	125,0e	21,806j	1328b	151,3f	–	53,133a	1,141,233b
2013–2014	3691d	4002c	1249ab	91,0ab	10,593f	1556d	68,3a	–	61,866d	1057,466a
2015–2016	47,849j	2628a	2468g	121,3de	20,354i	2373e	148,6ef	–	72,533f	1419,933d
F	30,029,071***	122,411***	227,691***	22,634***	8037,494***	10,330,412***	52,173***	8348**	1227,833***	623,412***

Age	Ba ppb	P ppb	Mg ppb	As ppb	B ppb
1997–1998	3533e	45,066c	150,83f	1233a	7566h
1999–2000	3300d	45,233c	133,00d	1366b	6966g
2001–2002	3200c	50,166d	139,90e	1199a	6466f
2003–2004	3033b	38,966b	121,86c	1200a	5832d
2005–2006	2400a	34,666a	91,30a	1333ab	5400c
2007–2008	3200c	59,333e	104,80b	1300ab	7399h
2009–2010	4766i	105,833g	160,83g	1266ab	6333f
2011–2012	4666h	114,666h	139,56e	1333ab	5200b
2013–2014	3732f	71,333f	140,86e	1300ab	4666a
2015–2016	4200g	118,000i	242,66h	1199a	6033e
F	631,482***	4724,603***	2288,618***	2504*	262,778***

The letters a, b, c, etc. means according to Duncan test results; show that the group is located. It is statistically different from the values contained in different groups, starting with the letter a numerical value grows

*significant at 0.05 level; **significant at 0.01 level; ***significant at 0.001 level


a
Al (ppm)
0 5 10 15 20 25 30 35 40 45 50
b
Cu (ppm)
0 0,5 1 1,5 2 2,5
c
Zn (ppm)
0 1 2 3 4 5 6 7 8 9 10
d
Cr (ppb)
0 20 40 60 80 100 120 140 160
e
Co (ppb)
0 20 40 60 80 100 120 140 160
f
Cd (ppm)
0 2 4 6 8 10 12 14 16 18 20
g
Mn (ppm)
0 1 2 3 4 5 6 7 8 9 10
h
Mg (ppm)
0 50 100 150 200 250
i
Fe (ppm)
0 5 10 15 20 25
j
Ba (ppm)
0 0,5 1 1,5 2 2,5 3 3,5 4 4,5 5
k
Na (ppm)
0 10 20 30 40 50 60 70 80 90 100
l
B (ppm)
0 1 2 3 4 5 6 7 8
m
P (ppm)
0 20 40 60 80 100 120
n
As (ppm)
1,1 1,15 1,2 1,25 1,3 1,35 1,4
o
Ca (ppm)
0 200 400 600 800 1000 1200 1400 1600 1800
1997-1998 1999-2000 2001-2002 2003-2004 2005-2006 2007-2008 2009-2010 2011-2012 2013-2014 2015-2016
Years

◀ **Fig. 2** **a** Showing of changes in Al concentrations over years. **b** Changes in Cu concentrations over years. **c** Changes in Zn concentrations over years. **d** Changes in Cr concentrations over years. **e** Changes in Co concentrations over years. **f** Changes in Cd concentrations over years. **g** Changes in Mn concentrations over years. **h** Changes in Mg concentrations over years. **i** Changes in Fe concentrations over years. **j** Changes in Ba concentrations over years. **k** Changes in Na concentrations over years. **l** Changes in B concentrations over years. **m** Changes in P concentrations over years. **n** Changes in As concentrations over years. **o** Changes in Ca concentrations over years

from the grouped growth rings. The mean values obtained from the evaluations in the wood part of the tree according to age, *F* values calculated by the analysis of variance, and homogeneous groups created with Duncan's test are presented in Table 2.

The results in Table 2 suggest that the amounts of each element in wood samples of different ages are statistically different; the difference was significant at a confidence level of 95% for As, 99% for Cd, and 99.9% for other elements. Cd was not included in the analysis as Cd concentrations in wood samples of 1–4 years of age were below the detection limits. According to the results presented in Table 2, grouping was more prominent for Al, Mn, and Fe, and particularly for Al; each age group formed a separate homogeneous group. The graphs created to facilitate understanding as to how concentrations of heavy metals change by age are shown in Fig. 1.

Discussion

Root uptake is the most efficient way of heavy metal uptake from soil into trees. Acid accumulation has the potential to influence atmospheric heavy metal deposition but it also significantly contributes to heavy metal deposition in tree rings and passive absorptions of active Al^{+3} by the roots (Cui et al. 2013). The tables show that higher heavy metal concentrations between 1997 and 1998 when compared to the other years suggest that these metals were transported to the plants by root uptake from the soil rather than foliage uptake from the atmosphere. Some elements (CI, K, Mg, P, and S) can be transported actively during the metabolic processes of the plant, whereas transport of some elements (B, Ba, Ca, Cu, Fe, Li, Mn, Mo, Zn) in the phloem is limited (Perone et al. 2018). In the last century, quality and intensity of heavy metal concentration in the environment have increased due to human-derived activities such as melting, artificial fertilization, and traffic. The number of data suggests that climate change is not only affected by pollution but is also affected by plant metabolism and distribution of elements in the plants. Thus, transport of the element within the organelles of the plant also changes (Cui et al. 2013).

Figure 2o, m, h, and c shows that the concentration of nutritional metals (i.e., Ca, P, Mg, and Zn) is considerably higher. Concentrations of heavy metals such as Al, Zn, Cu, Co, Fe, Cr, and Cd in the annual growth rings of the trees have increased As from 1999 (Table 2). Increased concentrations of nutritional elements (Ca, P, Mg, and Zn) between 2009 and 2010 during which precipitation was the highest suggest that solubility of these elements in the soil has increased with excessive precipitation and these elements were transported to the growth rings by root uptake. Decreased concentrations of other heavy metals (Cu, Co, Fe, Mn, Cd, and Al) during the same period reflect that atmospheric concentrations of these elements have significantly decreased with excessive precipitation and these elements were not accumulated in the roots or growth rings due to dilution in the soil. Heavy metals are transported from the atmosphere to the tree structure via two main routes. The first route involves absorption of the atmospheric heavy metals by the foliage, and the second route involves stomatal penetration of the metal (Shahid et al. 2017). This supports the results of the current study.

Other studies have reported similar results. In a similar study, Penninckx et al. (1999) reported a negative correlation between metal concentrations in the growth rings and the amount of precipitation and suggested that soil is diluted in the summer season when there is excessive precipitation. In opposition to nutritional elements, concentrations of heavy metals such as Co, Cr, Cd, Ba, Al, and Pb emitted from human-derived sources increase with the growth of the tree (Beramendi-Oroscoet al. 2013). Beramendi-Oroscoet al. (2013) suggested that increased Pb concentrations in the growth rings from past to present are caused by urbanization. As shown in Table 1, there was a significant difference in the heavy metal deposition in the bark and growth rings. This can be explained by the porous structure of the bark that retains the pollutants from the environment. As a result, deposition of heavy metals in the growth rings and barks throughout the plant growth provides retrospective information about atmospheric heavy metal concentrations. Thus, trees used for landscaping in areas with high traffic intensity and industrial activities are good bioindicators. Similar results have been reached and similar suggestions have been made in the studies conducted on this subject. As a result of their study on Acer platanoides, the annual rings indicate a good biomonitor to monitor the change in heavy metal concentration (Turkyilmaz et al. 2018a, b). Similar results were obtained by different researchers (Beramendi-Orosco et al. 2013; Norouzi et al. 2015)

Environmental pollution is one of the most important problems of our time, and this issue has been the subject of many studies in many aspects such as environmental impact, monitoring, and economic dimension. The recent studies represent that information about international trade of environmental implications make the bonding of three effects. One of these may lead to a non-hostile change in the output composition resulting from a combination of trade, industry, and composition. The other ones, the technical effect shows that the change in the emission reduction criteria in the economy is determined by an increase in trade direction. Another one is that it can adversely affect the environment with the increasing scale of the environmental pollutant sector (Mutlu 2016; Latif et al. 2018; Mutlu et al. 2016; Baloch et al. 2018; Danish and Wang 2018; Khan et al. 2018).

As indicated in many studies conducted to date, heavy metal pollution has numerous health hazards for humans (Cetin and Sevik 2016a). Bioindicators are the most important indicators reflecting the changes in atmospheric heavy metal deposition from past to present. Particularly in the past years, trees have been the focus as a bioindicator due to the lack of technological resources that could determine heavy metal concentrations in the atmosphere. Various plant parts can be used as a biomonitor such as the foliage of higher plants (Stolpe et al. 2017; Ozel et al. 2015; Ozturk and Bozdogan. 2015), trunk barks (Sawidis et al. 2011; Ugulu et al. 2016), and woods (Gao et al. 2015) as well as lichens (Conti and Cecchetti 2001) and mosses (Ceburnis and Steinnes 2000).

Many studies conducted to date have investigated the interaction between the plant and air pollution (Sevik et al. 2017b; Cetin and Sevik. 2016b). Studies evaluating *Robinia pseudoacacia* (Serbula et al. 2012; Celik et al. 2005), *Aesculus hippocastanum* (Tomasevic and Anicic 2010; Anicic et al. 2011), Sophora japonica (Li et al. 2007), *Betula pendula* (Petrova et al. 2014), *Elaeagnus angustifolia* (Aksoy and Sahin 1999), *Fraxinus excelsior* (Aksoy and Demirezen 2005), Pinus nigra (Turkyilmaz et al. 2018a), *Pinus pinea* (Aksoy and Demirezen. 2005), and *Quercus ilex* (Gratani et al. 2008) as biomonitors of traffic-derived air pollution have investigated the opportunities to utilize these plants as a biomonitor.

Conclusions

The results of this study suggest that concentrations of all heavy metals increase in relation to age, and heavy metal accumulation in the wood rings has increased due to increased traffic intensity between 1997 and 2016 in Ankara. However, this increase was not linear. Variable concentrations of heavy metals in the growth rings (higher in some years, lower in some years) are caused by morphological features of the tree. Studies on different species have also yielded similar results. In a study conducted by Beramendi-Orosco et al. (2013) on *Prosopis julifora*, Cu concentrations in annual growth rings were found to be 1.09 ppm between 1988 and 1992 and 1.27 ppm between 2003 and 2007, and Pb concentrations were found to be 0.35 ppm between 1993 and 1997 and 0.46 ppm between 1998 and 2002. Studies evaluating monthly changes have also reported similar results (Norouzi et al. 2015). Norouzi et al. (2015) Evaluated monthly changes in heavy metal concentrations in *Platanus orientalis* and reported that Cu concentration was 15.1 $mg.kg^{-1}$ in May, 15.7 $mg.kg^{-1}$ in July, and 16.6 $mg.kg^{-1}$ in November. Similar results have been reported for Fe, Mn, Ni, Zn, and Pb (Gao et al. 2015). Gao et al. (2015) investigated the changes in Pb, Cd, and Cr concentrations in annual growth rings of *Platycladus orientalis*, *Populus tomentosa*, and *Sophora japonica* in Jinan region across years and they reported similar results.

Compliance with ethical standards

Conflict of Interest The authors declare that they have no conflict of interest.

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