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Analyzing of usability of tree-rings as biomonitors for monitoring heavy metal accumulation in the atmosphere in urban area: a case study of cedar tree (*Cedrus* sp.)

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Abstract It is important to monitor the heavy metal pollution in order to identify risk zones and to determine the change in the heavy metal concentration of the atmosphere within the process. For this, it is necessary to carry out measurements for many years; however, this is not possible. Especially from past to present, one of the most effective methods to determine the changes of heavy metal concentrations in the atmosphere is to use the annual tree rings as biomonitors. Perennial plants growing in our country create annual rings, and it is possible to gain information regarding the changes of heavy metal concentrations in that region by determining the heavy metal concentrations in these rings. In this study, it was aimed to determine the annual changes of Pb, Co, and Fe elements' concentrations in these sections by determining the annual rings on the logs taken from the main stem of the cedar tree (*Cedrus* sp.), which was cut by the end of 2016, in December, 2016, in Kastamonu province. Within the scope of the study, the element concentrations were also determined in the inner and outer bark. As a result of the study, it was found that the heavy metal values in the organelles taken from the road-facing part, especially the heavy metal concentrations in the outer bark were higher than the metal concentrations in the inward-facing part, and that the concentrations changed significantly on organelle and year basis.

Keywords Annual ring · Cedar tree · *Cedrus* · Heavy metal

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Introduction

Recent global developments have brought along many environmental problems with them. Environmental pollution is one of the problems caused by this process. Air pollution and particularly the heavy metal pollution are of great importance among environmental pollution issues. Although heavy metals are naturally present on earth, the amount of heavy metals in the air increases under the influence of anthropogenic sources, and this increase causes the pollution. The heavy metals accumulated on earth contain some significant biological toxic metals (Cetin 2015a, b; Cetin 2017; Ahmed 2017; Mossi 2018; Erdem 2018; Cetin 2019; Cetin et al. 2019).

These toxic heavy metals are already naturally found in the environment, but their amount is rapidly increasing as a result of human activities, and they pose a serious threat to human health. The main sources of this pollution are coal, natural gas, paper, textiles, cosmetics, food packaging, electroplating and metal refining industries, mining and waste incineration plants, etc. Heavy metals are accumulated throughout the food chain because of their long biological half-lives and the fact that they are not biodegradable in the environment, and they may cause danger for humans (Cetin 2016a, b, c; Ozaktas 2015; Cetin 2017;).

Particularly, the heavy metals accumulated in the air cause many diseases for humans by being inhaled through respiration. Heavy metals can remain in nature for a long time without degradation, and their concentration in the ambient environment is ever-increasing. Also, they tend to bioaccumulate. For this reason, the determination of heavy metal concentrations is of great importance in terms of identifying the risk zones and the risk levels. However, there are two important problems in determining atmospheric pollution directly. The first one is that it is expensive, and the other one is the inability to determine the direct effect of atmospheric pollution on the ecosystem (Cetin 2016a; Cetin 2017; Cetin et al. 2017; Saleh 2018; Turkyilmaz et al. 2018a; Cetin et al. 2018).

Plants provide important information about the concentration of heavy metals in the air by accumulating such metals in various organelles. Trees show the increase of heavy metal concentration in the air over the years by accumulating heavy metals caused by fossil fuels, especially in areas with dense traffic, in their bodies, roots, fruits, barks, and leaves (Shahid et al. 2017; Erdem 2018; Bozdogan Sert et al. 2019; Sevik et al. 2019a, b). For this reason, plants have been used as biomonitors for many years. Especially, the trees, which live in highly polluted places for many years, can provide important information about the heavy metal accumulation in this area. The leaves of trees, which are not evergreen, used in the studies carried out on this subject are collected at the end of the vegetation season, and important information can be obtained about the heavy metal pollution occurring within the vegetation season. In addition, many studies have been carried out on this subject (Mossi 2018; Saleh 2018; Ozel et al. 2015; Bozdogan Sert et al. 2019; Sevik et al. 2019a, b).

The needles of trees such as pine, spruce, and fir, which are the species whose needles remain on the tree for many years and whose needle ages can be determined with certainty, are suitable for determining the recent heavy metal accumulation. However, only information from the past 8 to 10 years can be obtained in this way (Turkyilmaz et al. 2018b).

Other organelles of trees are used in order to obtain information from a long time ago. In regions like our country, where the winter season is observed, the development of trees is at different levels depending on the season, and thus annual rings are formed in the wood parts of such trees. In the long run, the heavy metals accumulated in the annual rings of trees can provide us with important information on the history of air pollution. For as much, the annual rings in the trees are related to tree age, and there are trees that can live for thousands of years. The annual rings of trees can be used as indicators of pollution and also can provide important information on the distribution and chronology of the elements causing pollution at the growing place of the tree (Beramendi-Orosco et al. 2013; Turkyilmaz et al. 2018c; Turkyilmaz et al. 2019; Akarsu 2019).

In this study, it was tried to determine the annual change of some heavy metal concentrations in the annual rings of a cedar tree grown in Kastamonu province. Within the scope of the study, the heavy metal concentrations in the bark and inner bark were compared with the heavy metal concentrations in the log, as well as the road-facing and inward-facing parts of the trees.

Material and method

This study was carried out on the cedar tree (*Cedrus* sp.) samples taken from Kisla Park located in Kastamonu province (the coordinates of 41° 23′ 20″ N and 33° 46′ 45″ E). The study was conducted on the log taken from the main stem of the tree, which was dried by the end of 2016 within the scope of the study, during December, 2016.

Kisla Park, where cedar trees, from which the study samples are taken, are grown, is one of the parks most preferred by people for recreational purposes in Kastamonu province. Kisla Park, which also contains a children's playground, is located next to the busiest streets that enable access to the center in terms of traffic density. Figure 1 shows the locations of Kastamonu province and Kisla Park.

The samples were taken from the stem of a cedar tree which was about 80 cm above the ground level and 45.7 cm in diameter. The upper surface of the log taken was smoothed by sanding in the lab with the purpose of making the annual rings more visible. The width of the road-facing part of the 39 year-old cedar tree was measured to be 27.4 cm, while it was measured to be 18.3 cm for the inward-facing part. The annual rings of the tree were grouped ranging between 1 and 13 in a way to be from stem to pith in three-year periods, taking their width into account. After the wood surface was divided into groups, samples were taken from the outer and inner bark as well as the wood at each age range by means of a steel drill and placed inside glass petri dishes.

The wood samples taken were shaved into wood dust. During these processes, care was taken not to use tools made of metals subjected to the study. Samples placed into the glass dishes without lids were kept in the laboratory for 15 days to become air-dried. Samples, which became air-dried at the end of the 15th day, were taken into the drying oven and were dried at 45 °C for a week. A total of 0.5 g of dried samples was mixed with 6 ml of 65% HNO_3 and 2 ml of 30% H_2O_2, and then was placed into the microwave oven. The microwave program was set to heat up to 200 °C in 15 min and then to remain at 200 °C for 15 min. After the samples were burned in the Milostone Ethos One model microwave oven, the solution sample was taken into balloons and was prepared for heavy metal analyses with the help of GBC Integra XL–SDS-270 ICP-OES device by adding ultrapure water until reaching 50 ml. In order to analyze the samples, the plasma of the ICP device was then burned and ultrapure water was passed through the system for 15 min for equilibration. Standard solutions were prepared according to the elements to be analyzed, and a calibration graph was created. After the creation of calibration graph, the samples were added to the system and the reading was carried out. As 0.5 g of sample was taken and filled with water to reach 50 g, the analysis results were multiplied by 100. Different calibration graphs were created at parts per million (ppm) or parts per billion (ppb) levels according to the analysis results that did not fall into the calibration graph, and they were re-read. All measurements were performed in triplicates. Afterwards, the data were evaluated with the help of SPSS package program.

Findings

Changes of elements on the basis of organelles

The *F* value, obtained as a result of variance analysis, error rate, mean values, and Duncan test results regarding the changes of elements on the basis of organelles are given in Table 1.

According to the results of variance analysis, it is seen that the change of the elements subjected to the study on the basis of organelles is statistically significant at 99.9% confidence level in all organelles. It is also seen that three homogeneous groups are formed in the road-facing parts of all the elements, and that the wood forms the first homogeneous group, the second homogenous group of the inner bark, and the last homogenous group of the outer bark. In the inward-facing parts, it is seen that the concentration of the Co element is below the determinable limits and the concentration in the inner bark is higher than the outer bark, whereas the concentration of the Pb element in the outer bark is below the determinable limits and the concentration in the inner bark is higher than the wood. As for the Fe element, it is seen that the lowest concentrations in the road-facing part are in wood, whereas in the inward-facing part, the highest values are in outer bark; however, the values obtained in the inner bark and outer bark are included in the same homogeneous group.

When the table values are examined, it is seen that the Co concentration in the outer bark sample in the road-facing part is approximately 10.00 times higher than the outer bark concentration in the inward-facing part, and that the Pb concentration in the inward-facing bark remains below the determinable limits, while the highest Pb concentration is obtained in the outer bark in the road-facing part. As for Fe concentration, it is seen that the values obtained in the wood and inner bark in the road-facing part are lower than the values obtained in the wood and inner bark in the inward-facing part. It is also seen that the concentration obtained in the outer bark in the road-facing part is almost twice the

Fig. 1 The locations of Kastamonu province and Kisla Park

values obtained in the outer bark in the inward-facing part.

Changes of heavy metals on a yearly basis

The changes of concentrations of the elements subjected to the study in the wood were determined on a yearly basis, and the mean values, *F* value obtained as a result of variance analysis, error rate, and the groupings formed as a result of the Duncan test, are given in Table 2.

According to the values given in Table 2, it is seen that the change of the element concentrations on a yearly basis is statistically significant at 99.9% confidence level according to the variance analysis results. When changes of Co concentration by years is examined, it is seen that the Co concentration, which is below 50 ppb between 1978 and 1986, increases to 200.00 ppb between 1987 and 1989, and the Co concentration, which decreases to 100.00 ppb between 1990 and 1992, increases to 150.00 ppb again between 2014 and 2016.

Table 1 Changes of elements on the basis of organelles

Elements	Organelles	Outer bark	Inner bark	Wood	*F* value	Error
Co	Inward-facing	67.8[a]	340.6[b]	–	1315.378	0.000
	Road-facing	682.0[c]	194.7[b]	102.5[a]	167.407	0.000
	Mean	374.9[b]	267.7[b]	102.5[a]	15.861	0.000
Pb	Inward-facing	–	3341.6[b]	980.1[a]	31.781	0.000
	Road-facing	5902.5[c]	3775.8[b]	2160.9[a]	43.198	0.000
	Mean	5902.5[c]	3558.7[b]	1619.7[a]	43.417	0.000
Fe	Inward-facing	80.1[b]	62.8[b]	21.7[a]	30.369	0.000
	Road-facing	154.8[c]	28.5[b]	18.6[a]	536.095	0.000
	Mean	117.5[c]	45.6[b]	20.2[a]	110.324	0.000

The letters a, b, c, etc. means according to Duncan's 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.

Pb concentration remains below the determinable limits after 2010 in the inward-facing part. While Pb value is 2584.0 ppb between 1978 and 1980 in the inward-facing part, it is 1743.8 ppb in the road-facing part. However, in the following years between 1981 and 1983, the Pb concentration in the inward-facing part decreases to 954.2 ppb, while the value in the road-facing part increases to 2896.0 ppb. Between 1987 and 1989, the Pb concentration decreases to 1700.0 ppb in the road-facing part, and increases to 1200.0 ppb in the inward-facing part. It reaches its highest value of 3620.7 ppb in the road-facing part between 1987 and 1989, and continues until the years between 2011 and 2013 with the value of 1500.0–2000.0 ppbs. In the inward-facing part, it increases again to 1823.1 ppb and continues to decrease linearly until 2008–2010. Between 2005 and 2007, it decreases to its lowest value of 70.2 ppb in the inward-facing part. Pb concentration, which decreases between 2011 and 2013, increases to 3164.4 ppb between 2014 and 2016 in the road-facing part.

Table 2 Changes of heavy metals on a yearly basis

	Co	Pb			Fe		
Age	Road-facing	Inward-facing	Road-facing	Mean	Inward-facing	Road-facing	Mean
1978–1980	10.0[a]	2584.0[g]	1743.8[de]	2163.9[bcd]	38.0[h]	7.9[a]	23.0[bc]
1981–1983	37.5[b]	954.2[d]	2896.0[h]	1925.1[bc]	16.4[de]	26.2[i]	21.3[bc]
1984–1986	16.3[a]	1261.0[e]	1870.4[ef]	1565.7[ab]	3.2[a]	10.9[c]	7.0[a]
1987–1989	203.3[g]	1823.1f	3620.7[j]	2721.9[cd]	66.8[i]	12.8[d]	39.8[d]
1990–1992	104.4[cd]	1292.8[e]	1963.7[f]	1628.3[ab]	14.4[c]	17.0[e]	15.7[abc]
1993–1995	103.1[cd]	635.2[c]	2338.1[g]	1486.6[ab]	18.7[f]	26.4[i]	22.5b[c]
1996–1998	97.0[c]	902.4[d]	1547.1[bc]	1224.7[ab]	26.0[g]	18.1[f]	22.0[bc]
1999–2001	110.2[cd]	445.0[bc]	1661.3[cd]	1053.2[bc]	14.5[c]	8.8[b]	11.6[ab]
2002–2004	121.0[d]	404.2[b]	1668.0[cd]	1036.1[bc]	15.4[cd]	12.9[d]	14.2[abc]
2005–2007	122.2[d]	70.2[a]	1493.3[b]	781.7[a]	17.8[ef]	20.9.[g]	19.3a[bc]
2008–2010	167.2[f]	409.2[b]	2990.9[h]	1700.0[abc]	16.3[cde]	28.7[k]	22.5[bc]
2011–2013	91.9[c]	–	1134.7[a]	1134.7[ab]	9.5[b]	24.8[h]	17.1[abc]
2014–2016	149.0[e]	–	3164.4[i]	3164.4[c]	25.6[g]	26.9[j]	26.2[c]
F value	84.188	110.512	209.6	3.661	647.077	2186.284	3.595
Error	0.000	0.000	0.000	0.000	0.000	0.000	0.000

The letters a, b, c, etc. means according to Duncan's 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.

In the inward-facing part, Fe concentration increases to 38.0 ppm between 1978 and 1980, and to 66.8 ppm between 1987 and 1980, and then it decreases to 3.2 ppm (its lowest value) between 1984 and 1986. In the following years, it does not exceed 27.0 ppm. While the road-facing Fe values increase until the years between 1981 and 1983, they decrease to 10.9 ppm between 1984 and 1986. They then increase again until the years between 1993 and 1995 but decrease to 10.0 ppm again between 1999 and 2001. They continue to increase linearly below 30.0 ppm until the years between 2014 and 2016. The changes in the concentrations of the elements subjected to the study in the wood on a yearly basis are given in the following graphs (Graphs 1, 2, and 3).

Results and discussion

As a result of the study, the concentration of Co element in the inward-facing wood and the concentration of Pb element in the inward-facing outer bark remained below the determinable limits. When the changes of the elements in the inward-facing organelles were examined, it was seen that the lowest value of Co element (below the determinable limits) was found in the wood and the highest value in the inner bark, whereas the lowest value of Pb element (below the determinable limits) was found in the outer bark and the highest value in the inner bark, and the lowest value of Fe element was found in the wood and the highest value in the outer bark. It was also found that the inner bark and outer bark values were included in the same group according to the Duncan test.

When the changes of the elements in the outer bark were examined, it was found that the lowest concentrations of all elements were found in the wood and the highest concentrations in the outer bark. According to these results, while the concentrations of the elements in the inward-facing outer bark were not significant, the highest concentrations were obtained in the road-facing outer bark. In fact, the values obtained are much higher than the concentrations in the inward-facing part and the concentrations in the organelles of the outward-facing part. This situation is mostly associated with the particulate matter in the environment.

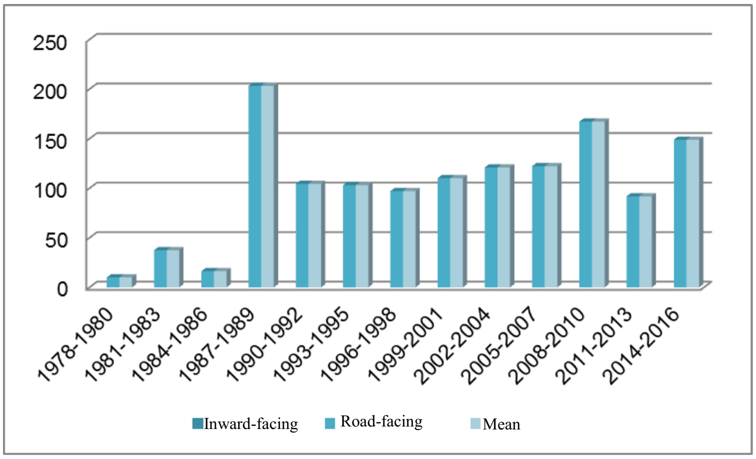
In two separate studies, Turkyilmaz et al. (2018a, d) report that some heavy metal concentrations in the outer bark are much higher than in other organelles. Janta et al. (2016) state that heavy metal concentrations in the outer bark are directly related to environmental conditions and are caused by atmospheric pollution. In fact, the studies conducted show that the heavy metals in the air cling to particulate matter, that the particulate matter is contaminated with heavy metals, and that the heavy metal concentrations increase in plant organelles after this contaminated particulate matter settles on such organelles (Shahid et al. 2017; Saleh 2018; Mossi 2018).

A study carried out on *Cupressus sempervirens* showed that the concentrations of Pb, Zn, Mn, Cr, Ni, Cd, and Cu elements in the bark were different in various regions, and that the main source of heavy metal pollution was traffic emissions. In addition, it was reported that Pb concentration was highest in areas with dense traffic (El-Hasan et al. 2002). The results obtained in this study also show that some of the heavy metal concentrations in the bark may be related to the particulate matter.

In the studies conducted on this subject, in order to determine how much of the heavy metal concentrations in organelles originate from particulate matter, the samples are washed, and the washed and unwashed samples are compared. In many studies conducted this way, the heavy metal concentrations in the washed samples were found to be much lower than those in the unwashed samples, and it was also found that the difference of Pb concentration between the washed plant and unwashed plant samples could even be 13 times (Aksoy and Öztürk 1997; Abechi et al. 2010; Buachoon 2014; Mossi 2018).

When the changes of the elements in the outer bark were examined, it was found that the lowest concentrations of all the elements were in the wood and the highest concentrations were in the outer bark. However, it was determined that the concentrations among the organelles in the inward-facing part were at different levels and that the Pb concentration in the outer bark and the Co concentration in the wood remained below the determinable limits. This situation is considered to be related to the entry mechanism of heavy metals into the plant body.

While there are a number of studies regarding the heavy metal uptake to the plant body from the root, the number of studies regarding the airborne uptake of heavy metals to the plant body is quite

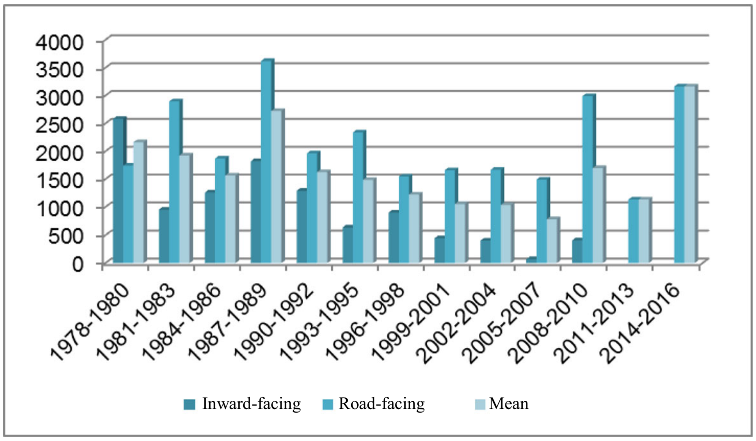


Graph 1 Changes of the Co element on a yearly basis

inadequate. The studies conducted show that heavy metals cling to various organelles of the plant by clinging to the particulate matter in the air, and then enter into the plant body in various ways (Shahid et al. 2017). The plants' potential to retain and take particulate matter, and thus heavy metals, is closely related to the physical and physiological characteristics of species. Characteristics such as roughness on the plant organelles, trichomes, organelle surfaces, and structure affect the settling of heavy metals on the organelles, especially on leaf surfaces (Schreiber and Schoenherr 1992; Cunha and do Nascimento 2009; Ataabadi et al. 2011).

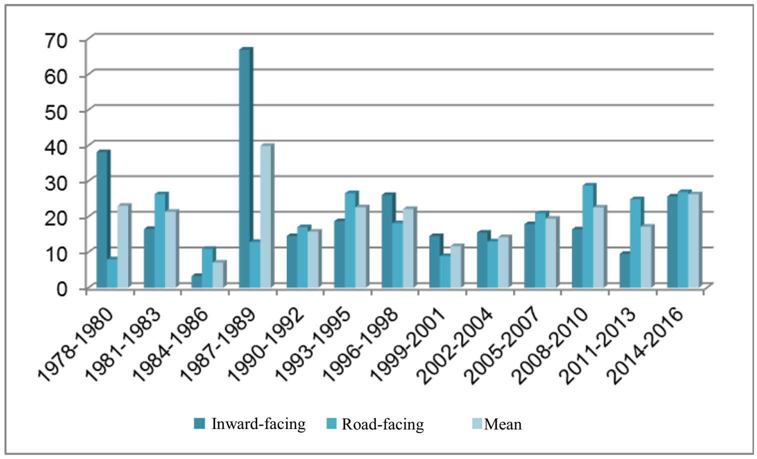
Therefore, it is possible that particulate matter contaminated with traffic-induced heavy metals cling to both the bark and the leaves of the road-facing part of the tree subjected to the study and that these substances enter into the tree by means of photosynthesis, osmosis, etc., and thus increase the concentrations of heavy metals in the road-facing part. Likewise, in many studies conducted to date, the heavy metal concentrations in plants grown in areas with dense traffic are found to be higher than the plants grown in areas with little or no traffic (Buachoon 2014; Huber et al. 2016; Yang et al. 2017; Sevik et al. 2019a, b, c; Kaya et al. 2019; Bozdogan Sert et al. 2019; Yucedag et al. 2019). Moreover, in numerous studies, the elements subjected to the study are found to be increasing interrelatedly with traffic density (Jaradat et al. 1999; Sawidis et al. 2011; El-Hasan et al. 2002; Aricak et al. 2019a, b).

As a result of the study, it was found that the change of all elements on a yearly basis was statistically significant at 99.9% confidence level. Although there was a horizontal change in element concentrations, an increase was observed in recent years. This situation is considered to be related to



Graph 2 Changes of the Pb element on a yearly basis

Graph 3 Changes of the Fe element on a yearly basis



the recent increase in population and the number of vehicles in Kastamonu city center. While the population of Kastamonu city center was 115,332 people in 2007, it went up to 123,972 in 2010 and to 146,103 in 2016. It is obvious that this increase will lead to an increase in both traffic density and air pollution (Akarsu 2019).

The study results also show that Co, Pb, and Fe elements reached high concentrations between the years of 1987 and 1989. These elements are generally known as industrial heavy metals which spread through traffic (Mossi 2018; Shahid et al. 2017; Turkyilmaz et al. 2018c, 2019). In the years in question, a development out of industry- or traffic-related routine is likely to be in place. In fact, it is reported that the population of Kastamonu city center went from being 35,465 people in 1980 to 51,560 people in 1990 (Ibret and Aydinozu 2009). The increase in some heavy metals may be related to the increase in population in the mentioned years, and consequently the increase in traffic and industry.

Recommendations

The study results show that the annual rings of the trees are suitable for monitoring the heavy metal concentration. As a result of the study, especially in certain periods (between 1987 and 1989), the concentrations of Co, Pb, and Fe in the elements, which are very harmful for human health, are found to be reaching high levels. In the literature study conducted, it was determined that there was a significant increase in the population of the city center during the relevant years, and that coal was burned intensively. As a matter of fact, also the locals state that there was an abnormal increase in air pollution in the mentioned years. Thus, it is possible to say that annual rings are very useful in the heavy metal concentration changes in annual rings.

Through dendrochronological or dendrochemical studies carried out on annual rings, various information can be obtained regarding the data on the climate of that region or the factors that stress the plant. For this purpose, annual rings of trees should be examined in more detail by experts. In order for the annual rings to be used in the determination of heavy metal concentrations, these annual rings must be taken from the tree. It is clear that the most effective method for this is cutting the tree. However, this process can be carried out without ending the lives of trees. In different studies to be carried out with this purpose, the annual rings of the tree can be taken with the help of an increment borer without damaging the tree or ending its life.

In this way, more detailed information on the changes in heavy metal concentrations of the region can be obtained by conducting studies on different trees. In addition to this, it is thought that important information can be obtained on the changes in heavy metal concentrations of the region from at least 20 to 30 years before by using the materials obtained as a result of the pruning works, not from the main stem of the trees but from the side branches. By using this method, the recent changes in heavy metal concentrations can be determined in industrial areas, around factories or in areas with dense traffic.

As a result of the study, it was found that although there was a general horizontal change in the concentrations of the elements subjected to the study, there was an increase in the recent years. This situation is considered to be related to the increase in population in recent years, and consequently the increase in human-induced pollutants. Therefore, the pollution levels should be controlled in areas with ever-growing population size and necessary precautions should be taken in this regard.

As a result of the study, the heavy metal concentrations, which are of great importance especially for human health, were found to be higher in the road-facing bark than in the inward-facing bark. Considering that the bark is not a living organelle, this situation can be said to be related to particulate matter, which is contaminated with heavy metals especially in the road-facing part. The importance of particulate matter for human health has been the subject of many studies. Especially in areas with dense traffic or industry, particulate matter contaminated with heavy metals can cause serious health problems. For this reason, changes in heavy metal concentrations and particulate matter concentrations should be evaluated together in determining risk zones, and studies should be carried out on this subject to determine both the risk zones and the necessary precautions.

Author contributions Hakan and Hatice conceived and designed the experiments. Halil, Ilknur, Hatice, and Hakan performed the experiments. Hatice and Hakan analyzed the data. Hakan, Ilknur, Hatice, Mehmet, and Halil contributed reagents/materials/analysis tools. Halil, Hakan, and Mehmet wrote the paper.

Compliance with ethical standards

Ethical approval This article does not contain any studies with human participants or animals performed by any of the authors.

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

Informed consent Informed consent was obtained from all individual participants in the study.

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