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RESEARCH ARTICLE

The usability of *Cupressus arizonica* annual rings in monitoring the changes in heavy metal concentration in air

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Abstract

Air pollution, which has been increasing in recent years, has reached significant dimensions and has become one of the most important agenda topics of present day. Among air pollution components, heavy metals are of particular importance, since they are not easily decomposed, they tend to bioaccumulate, and some of them have toxic or carcinogenic effects even at low concentrations. Therefore, it is an extremely important subject to monitor the changes in heavy metal concentrations found in air. The most preferred method in determining the changes in heavy metal concentrations in the atmosphere is the use of biomonitors. From past to present, trees have been good biomonitors in determining the increase in heavy metal concentrations in the atmosphere. Particularly, with the help of the annual growth rings of trees, vital information can be obtained on the changes in heavy metal concentrations in air. In this study, after the annual rings were determined on the log taken from the main body of the cypress (*Cupressus arizonica*) tree cut from the Kisla park located in Kastamonu province, the concentrations of Bi, Cd, and Ni in the outer bark, inner bark, and wood were compared in the inward-facing and road-facing parts of these sections. Also, the changes in heavy metal concentrations in the annual rings were evaluated on a yearly basis. As a result of the study, it was found that the element concentrations in the outer bark of the road-facing part were generally at a higher level, and that the changes in the elements on a yearly basis generally followed a fluctuating course, but there had been a general increase in the Cd and Ni concentrations in recent years.

Keywords Annual ring · Biomonitor · *Cupressus arizonica* · Heavy metal

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Introduction

In order to survive, people's needs such as life, health, food, and shelter are increasing at the same rate as the rapid increase in the world population. Therefore, the environment is getting polluted at the same rate as the increasing needs. Environmental pollution takes place in two ways. One of them is the natural pollution, that is, the waste of all creatures other than humans, and the direct waste of humans. In natural pollution, nature can clean itself in a short time through recycling mechanism (Turkyilmaz et al. 2018a; Kilicoglu et al. 2021). However, human-caused pollution, especially the pollutants caused by industrial activities or vehicle exhaust fumes, remain in nature for a long time and have adverse effects on human health. Earth's atmosphere is mainly composed of oxygen (O_2), nitrogen (N_2), and carbon dioxide (CO_2). However, the rapid economic development, urbanization, and industrialization processes, which took place within the last 30 to 40 years, caused the diffusion of various pollutants into the atmosphere, and substantially impaired the quality of the atmosphere, that is to say, the air we breathed (Shahid et al. 2017; Sevik et al. 2019a; Cetin et al. 2019; Kilicoglu et al. 2020).

The atmospheric pollution has reached such a serious level that more than 6.5 million people are reported to die due to air pollution each year (Sevik et al. 2019b). The European Environment Agency states that there are polluted areas covering 2.5 million areas across Europe, and that 14% of these areas need urgent improvement planning (Akarsu 2019).

Heavy metals are of particular importance among air pollution components, since they are not easily decomposed, they tend to bioaccumulate, and some of them have toxic or carcinogenic effects even at low concentrations (Turkyilmaz et al. 2018b). Heavy metals such as As, Ni, Zn, Cr, Pb, Cd, and V are mostly of industrial origin and are carcinogenic. In terms of their potential toxicity and effects on living organisms in particular, Pb, Cr, As, Cd, and Hg are among the most toxic heavy metals (Ucun Ozel et al. 2019; Cetin et al. 2020).

Studies conducted show that almost all metals have toxic effects when taken above a certain amount. Even though micronutrients such as Mn, Zn, Cr, Cu, Fe, and Ni are necessary for living organisms, including plants, they may also have harmful effects at high concentrations, whereas heavy metals such as Hg, Cd, As, and Pb can cause serious toxicity to living organisms even at low concentrations (Sevik et al. 2020a).

Especially when heavy metals, which accumulate in the air, come down to the earth through rain water, they cause many diseases in people by being inhaled through respiration. Heavy metals can remain in nature for a long time without decomposition, and their concentration in the environment is ever increasing. For this reason, determining the heavy metal concentration is of great importance in identification of the risk zones as well as the risk levels (Turkyilmaz et al. 2020; Ucun Ozel et al. 2020).

However, there are two important issues in determining atmospheric pollution directly. The first one is the high cost, and the other is the inability to determine the direct effect of atmospheric pollution on the ecosystem (Sevik et al. 2020b). Moreover, with direct measurements, it is not possible to obtain information on the level of heavy metal concentration in the atmosphere in the past years. For this reason, biomonitors are frequently used in monitoring heavy metal pollution (Turkyilmaz et al. 2019).

Plants provide important information on the concentration of heavy metals in air through accumulation of such metals in various organelles. For this reason, especially the leaves of plants, which are not evergreen, are extensively used in order to compare regions with different characteristics (for instance, regions with different traffic density), since it is known how long the plants are exposed to heavy metals in air (Aricak et al. 2020). However, since this method cannot give an idea on the changes in heavy metal concentration, it is not considered as an effective method in monitoring heavy metal pollution (Sevik et al. 2020c).

Monitoring the changes in heavy metal concentration in a region is possible primarily by analyzing the changes in the process in a comparable way. In this study, it was aimed to determine the changes in some heavy metal concentrations found in the annual rings of a 21-year-old *Cupressus arizonica* tree growing in the city center of Kastamonu province on a yearly basis. Within the scope of the study, the heavy metal concentrations found in the bark and inner bark were also compared with the heavy metal concentrations in the wood, and the changes in heavy metal concentration in the region were also tried to be determined on the basis of years and organelles.

Materials and methods

This study was carried out on the samples of *Cupressus arizonica* (Blue Cypress) annual rings taken from the Kisla Park located in Kastamonu province in with the coordinate of 41° 23′ 20.2488″ north latitude and 33° 46′ 45.9912″ east longitude (height above sea level: 774 m). The sample was taken from the main body of *Cupressus arizonica* tree, which grew in the Kisla Park, about 50 cm above the ground. Before the sample was taken, its main road-facing side was marked on the log, and the tree stump of approximately 20-cm thickness was brought to the laboratory. In the lab, the top surface of the tree stump was sanded in order to make the annual rings more visible, and thus, the annual rings were smoothed.

The tree was found to be 21 year-old as a result of the census made. With the help of a steel-tipped drill, samples were taken from the outer bark, the inner bark, and the wood of all ages, and were placed in glass petri dishes. Since the annual rings were large enough, the wood samples were taken from annual rings of the last 14 years, separately for each year. However, the tree grew very slowly in the first years, and thus, the annual rings formed in the first years were quite narrow. For this reason, the annual rings of the first 7 years in the center of the tree were taken as a single sample. The samples were taken separately from the road-facing and inward-facing directions. In total, 17 samples were taken from each direction, including 15 wood samples from both directions, and one sample from the inner bark and outer bark; all in all 34 samples were evaluated within the scope of the study.

The wood samples taken were cut into wood chips. During these processes, care was taken to avoid using tools made of metals subjected to the study. The samples were kept for 30 days until they became air-dried, and afterwards they were dried in the drying oven at 50 °C for a week.

Afterwards, the samples were powdered, and 0.5 g of dry sample was weighed and put into tubes designed for microwave. In the fume cupboard, 10 ml of 65% HNO_3 and 2 ml of 30% H_2O_2 were added to the samples placed in the tubes. The prepared samples were then burned in a microwave at 280 PSI pressure and 180 °C for 20 min. The program of the microwave device was set in a way to rise to 200 °C in 15 min and to remain at 200 °C for 15 min. After the tubes taken out of the microwave were cooled, deionized water was added until it became 50 ml. The samples were then filtered through filter paper and read at appropriate wavelengths on the ICP-OES (inductively coupled plasma–optical emission spectrometer) device. In the study, all measurements were made in triplicate.

The data obtained were evaluated with the help of SPSS package program and was subjected to variance analysis. Homogeneous groups were obtained by applying Duncan test to the data in which statistically significant differences at minimum of 95% confidence level were found as a result of the variance analysis. The data obtained were simplified and interpreted after being tabulated.

Results

The organelle-based changes in Bi element, one of the studied elements, depending on the direction were determined, and the *F* value, obtained as a result of the variance analysis and the mean values based on organelles, error rate, and the groupings formed as a result Duncan test are given in Table 1.

IB inner bark, *OB* outer bark, *W* wood, *IF* inward facing, *RF* road facing, *MEAN* mean value

When the organelle-based changes of the elements are examined, it is seen that according to the results of the variance analysis, there are significant differences at 95% confidence level between the IF parts of the organelles of the Bi and Ni elements, and at 99.9% in the RF and MEAN parts of the Ni element. While the highest Bi concentration in the IF part is obtained in IB with 5721.8 ppb, Bi concentration is seen to be 5426.4 ppb in the outer bark and 3802.0 ppb in the wood. As a result of Duncan test, while outer and inner bark values are found to be in the same homogeneous group, wood is found to be in the other homogeneous group.

When the changes in Cd concentration are examined, it is seen that there are no statistically significant differences between organelles at a minimum of 95% confidence level. In Ni concentrations, it is seen that the lowest value is obtained in wood, while the highest values in both IF and RF parts are obtained in OB. It is especially noteworthy that the value obtained in the RF part in DC is two times more than the value obtained in the IF part.

The changes in the Bi element are determined on a yearly basis, and the mean values on a yearly basis and the *F* value obtained as a result of the variance analysis, as well as the error rate and the groupings formed as a result of Duncan test are given in Table 2.

When the values of Table 2 are examined, it is seen that the lowest value of the Bi concentration in the IF part is reached in the 12th year (1186.53 ppb), while the highest value is reached in the first 7 years (5243.46 ppb), whereas the highest value in the RF part is reached in the 10th year (5534.73 ppb), and the lowest value in the 15th year (1512 ppb). When the values are examined, it is seen that the changes in the Bi element on a yearly basis generally follow a fluctuating course, that it begins to decrease starting from the 10th year in the samples taken from the RF part, decreases to the lowest level in the 15th year, then increases until the 17th year, and then starts to decrease again and continues to decrease until the 19th year. Later on, it seems to increase again. When assessed in general, it can be said that the values in the IF part are higher than those in the RF part. The changes in the Cd element are given in Table 3.

When the table values are examined, it is seen that there are significant differences at 99.9% confidence level between the years in terms of IF, RF, and MEAN. When the mean values are analyzed, it is seen that the Cd concentration peaked in the 10th year in the RF part, apart from that it generally increases in the 16th year in the IF part and in the 18th year in the RF part, but it tends to decrease after these years. In the IF part, while the lowest value is obtained in the 10th year (52.53 ppb) and the highest value in the 16th year (304.6 ppb), the lowest value in the RF part is obtained in the 13th year (81.93 ppb) and the highest values in the 10th (409.53 ppb) and 18th (352.0 ppb) year. The changes observed in the element are given in Table 4.

When the changes in the Ni concentration are examined on a yearly basis, it is observed that the values in the IF part vary between 460.0 ppb (12th year) and 1434.8 ppb (20th year), while they vary between 595.07 ppb (15th year) and 1492.2 ppb (17th year) in the RF part. While the Ni concentration in the IF part generally reaches a plateau, it is seen to reach the highest value in the 20th year and decreases in the

Table 1 Organelle-based changes of the elements

	Bi			Cd			Ni		
Organelles	IF	RF	MEAN	IF	RF	MEAN	IF	RF	MEAN
OB	5426.4 b	2492.4	3959.4	140.40	131.33	135.86	1076.06 b	2184.20 b	1630.13 b
IB	5721.8 b	1552.2	3637.0	80.53	131.00	105.76	1039.73 b	704.20 a	871.97 a
W	3802.0 a	3394.51	3598.2	127.65	163.32	145.49	791.85 a	881.93 a	836.89 a
F	6.889	2.991	0.190	0.757	0.348	0.720	3.727	42.83	23.69
Error	0.002	0.060	0.827	0.475	0.708	0.489	0.031	0.000	0.000

21st year. Whereas in the RF part, it is seen that the values start to decrease after having the highest values in the first years, that it reaches the lowest value in the 15th year, and then starts to rise again and reaches the highest value in the 17th year. Afterwards it starts to decrease again.

Discussions

As a result of the study, it was found that the organelle-based changes in the elements subjected to the study were not statistically significant at a minimum of 95% confidence level in both directions in the Cd element and in the road-facing part in the Bi element. When the organelle-based changes in the values were examined, the highest values of Bi and Ni concentrations were seen to be obtained in the outer bark according to the mean values.

In studies conducted on the organelle-based changes of heavy metals, it is reported that the accumulation of different elements in different organelles differ from each other. As a result of the study conducted by Akarsu (2019) on the *Cedrus* tree, it was found that the highest concentrations of Zn, Mn, Fe, K, Ca, and B elements were in the outer bark, which was then followed by the inner bark. It was even stated that there were great differences between these values, for example, that the Mn concentration in the inner bark was 10 times more than the values in the wood part, and the values in the outer bark was 50 times more than the values in the wood part (Akarsu 2019).

Many studies suggest that heavy metal concentrations, especially obtained in the bark, are at higher levels than of those in the other organelles. Turkyilmaz et al. (2019) state that there is almost a 9 times difference between wood and bark in terms of Cr concentration. In another study, it is reported that while

Table 2 Changes in the Bi (ppb) element on a yearly basis

Age	Inward facing	Road facing	Mean
First 7 years	5243.46 i	5288.80 gh	5266.13 d
8th year	3260.60 cd	4990.533 g	4125.56 bcd
9th year	3655.13 de	5454.86 h	4555.00 cd
10th year	2742.80 bc	5534.73 h	4138.767 bcd
11th year	2269.33 b	4228.60 f	3248.96 abc
12th year	1186.53 a	3964.60 f	2575.56 ab
13th year	4061.06 ef	3395.80 e	3728.43 abc
14th year	4265.33 fg	1667.66 a	2966.50 ab
15th year	4379.06 fgh	1512.00 a	2945.53 ab
16th year	4677.86 ghi	2079.46 b	3378.66 abc
17th year	4189.46 efg	2922.80 d	3556.13 abc
18th year	4956.75 hi	2291.66 bc	3624.2 abc
19th year	2884.06 c	1614.93 a	2249.5 a
20th year	4546.06 fgh	2556.53 cd	3551.30 abc
21st year	4712.40ghi	3414.66e	4063.53 bcd
F value	36.775	105.221	2.676
Error	0.000	0	0.003

Table 3 Changes in the Cd (ppb) element on a yearly basis

Age	Inward facing	Road facing	Mean
First 7 years	134.33 de	104.07 bc	119.20 ab
8th year	128.80 de	111.67 cd	120.23 ab
9th year	73.33 ab	97.80 b	85.57 a
10th year	52.53 a	409.53 i	231.03 cd
11th year	58.33 a	109.67 bcd	84.00 a
12th year	73.40 ab	157.87 f	115.63 ab
13th year	75.66 ab	81.93 a	78.80 a
14th year	113.66 cd	142.07 e	127.87 ab
15th year	165.00 f	113.13 cd	139.07 abc
16th year	304.60 h	110.93 cd	207.77 bcd
17th year	99.86 bc	193.33 g	146.60 abc
18th year	144.33 ef	352.00 h	248.17 d
19th year	144.46 ef	121.00 d	132.73 ab
20th year	255.86 g	157.27 f	206.57 bcd
21st year	90.66 bc	187.67 g	139.17 abc
F value	71.074	510.972	3.341
Error	0	0	0

Table 4 Changes in the Ni (ppb) element on a yearly basis

Age	Inward facing	Road facing	Mean
First 7 years	854.06 fg	1375.00 g	1114.53cd
8th year	549.40 ab	889.20 e	719.30a
9th year	658.40 bcde	823.47 d	740.93a
10th year	612.20 bc	1065.20 f	838.70ab
11th year	617.40 bcd	902.07 e	759.73a
12th year	460.00 a	906.40 e	683.20a
13th year	742.06 cdef	754.80 c	748.43a
14th year	745.06 cdef	613.80 a	679.43a
15th year	755.73 def	595.07 a	675.40a
16th year	862.73 fg	847.13 d	854.93abc
17th year	789.73 efg	1492.20 h	1140.97d
18th year	1020.08 h	842.87 d	931.48abcd
19th year	912.00 gh	738.67 c	825.33ab
20th year	1434.80 i	651.53 b	1043.17bcd
21st year	864.06 fg	659.29 b	797.83ab
F value	27.711	35.487	3.183
Error	0	0	0.001

the Zn concentration measured in wood of *Acer platanoides* is 3.59 ppb on average, this figure increases to 14.79 ppb in bark samples (Turkyilmaz et al. 2018a). Janta et al. (2016) found in their study conducted on *Cassia fistula* that the highest values of Cu, Fe, and Zn concentrations in different layers of the bark were obtained in the outer bark. Ugulu et al. (2016) reported that the Ni concentration in the bark of *Ficus carica* was 5.5 times higher than the concentration in the leaf, whereas the Cr concentration was more than 5.1 times.

As a result of the study, the Ni concentrations in the road-facing part were found to be higher than the concentrations in the inward-facing part. In a study carried out by Akarsu (2019), it is revealed that the element concentrations in the road-facing parts are higher than the element concentrations in the inward-facing part in terms of Ni, Li, Cu, Cr, Co, Cd, and Ba elements, and even that the difference is very high for some other elements. For instance, it is stated that while the Cd concentration in the inward-facing outer bark is 116.6 ppb, this value increases to 2601.2 ppb in the road-facing outer bark; similarly, there is a 17-times difference between the values of the road-facing outer bark and the values of the inward-facing outer bark.

The fact that the metal concentrations in both the outer bark and the road-facing part are higher than the other parts is associated with the particulate matter concentration in the air (Sevik et al. 2020a, 2020c). In various studies, it is reported that heavy metal concentrations in the outer bark are directly related to environmental conditions and caused by atmospheric pollution (Janta et al. 2016; Akarsu 2019). As a matter of fact, in studies conducted, it is found that the heavy metals in the air adhere on particulate matter, the particulate matter gets contaminated with heavy metals, and with the adhesion of this particulate matter on plant organs, the heavy metal concentrations in these organs also increase (Shahid et al. 2017; Sevik et al. 2020c).

In studies conducted, it is also reported that heavy metals adhere to various organelles of the plant by adhering to particulate matter in the air, they enter inside the plant in various ways, and that the potential of plants to capture and intake particulate matter, and thus heavy metals is closely associated with the physical and physiological characteristics of species and their organelles. Features such as roughness, villus, area, and the structure of the plant organelles affect the accumulation of heavy metals on organelles, especially on leaf surfaces (da Cunha and do Nascimento 2009; Ataabadi et al. 2011; Shahid et al. 2017).

From this point forth, the settling of particulate matter contaminated with heavy metals on the outer bark, which has a rough structure, may cause high levels of heavy metal concentrations in these organelles. Furthermore, it is possible that the adhesion of particles contaminated with traffic-induced heavy metals to both road-facing bark and the leaves of the tree subject to the study cause these substances to enter into the tree by means photosynthesis, osmosis, etc., and thus increase the heavy metal concentrations in the road-facing part.

The heavy metal exposure of the air and thus the particulate matter in the air as well as the plants is also related to the traffic density. Studies show that traffic is one of the most important causes of heavy metal pollution (Turkyilmaz et al. 2018c). In many studies conducted to date, the heavy metal concentrations in plants grown in areas with heavy traffic are found to be at higher levels than those grown in areas with no or less traffic (Alaqouri et al. 2020a, 2020b).

As a result of the study, it was found that the changes in the elements subjected to the study on a yearly basis generally followed a fluctuating course, and that the concentrations of Cd and Ni elements had generally been increasing in recent years. Similar results were obtained in another study conducted in the same region. Akarsu (2019) states that while there is generally a horizontal change in Ba, Li, Ca, Mg, Mn, and Cr elements in Kastamonu province, an increase has been observed in recent years. Akarsu (2019) indicates that this situation is related to the increase in the population and the number of vehicles in Kastamonu city center in recent years, and that the population of Kastamonu city center increased from 115,332 people in 2007 to 123,972 people in 2010 and to 146,103 in 2016 (Akarsu 2019).

There are many studies in which Cd and Ni elements, of which concentrations have been increasing in recent years, are associated with traffic density. In their study, El-Hasan et al. (2002) report that the Cd concentration is higher in areas with heavy traffic. Cd is generally released by vehicles, and they are more concentrated in particulate matter found in areas with heavy traffic (Jaradat et al. 1999). In studies conducted, it is stated that heavy metal concentrations in soil and plant samples in the regions alongside the road are mostly caused by traffic density (Aksoy and Demirezen 2006). In many studies, it is reported that heavy metal concentrations in various organelles of plants used as biomonitors increase with traffic density (Aricak et al. 2019; Sevik et al. 2019c).

As a result of various studies conducted on the subject, it is revealed that the concentrations of heavy metals that adversely affect human health increase with age (Yigit 2019). However, the changes in heavy metal concentrations on a yearly basis are at different levels. It is also stated that the fact that the increase in the amount of heavy metals in the tree annual rings is high in some years and low in others is because of the morphological structure of the tree (Yigit 2019).

In conclusion, it is known how dangerous heavy metals can be to human health. Nevertheless, it is a fact that heavy metal pollution in the air is increasing day by day (Turkyilmaz et al. 2018c; Shahid et al. 2017). For this reason, it is of utmost importance to follow the changes in heavy metal concentrations in the air. Tree annual rings act as a good biomonitor in monitoring this change, and especially in monitoring the change from past to present.

Conclusions

In this study, the usability of annual rings of perennial trees was tried to be determined in observing the changes in heavy metal concentration. For this purpose, heavy metal concentrations in annual rings were determined retrospectively. The study results show that the annual rings of trees are suitable for monitoring heavy metal concentration. In various studies conducted on this subject, this situation has been reported. The annual rings of trees can provide important information on the changes over the years in not only heavy metal concentrations but also the nutrients required by the plant.

In order to use annual rings in determining heavy metal concentrations, these annual rings must somehow be extracted from the tree. It is clear that the most effective method for this is cutting the tree. However, this process should be carried out without causing the death of the trees. In different studies carried out with this purpose, it is possible to obtain more detailed information on the changes in heavy metal concentrations in the region using annual rings of tree obtained by means of increment borer without damaging the tree or causing the death of the tree. In addition to this, it is also considered possible to obtain important information on the changes in heavy metal concentrations in the region at least 20 to 30 years ago by using the materials obtained as a result of the pruning performed on the side branches rather than the main trunk of the trees.

As a result of this study, it was presented that by using the data obtained and the method used in this study, it was possible to monitor the heavy metal concentrations in different regions easily. By using this method, retrospective changes in heavy metal concentrations can be monitored, especially in industrial areas or regions where traffic pollution is ever increasing.

With the method used in this study, it is possible to obtain hundreds of years of historical data about a region. However, the number of studies conducted on which species are more suitable for this purpose is not sufficient. For this reason, it is recommended to diversify and increase similar studies on different regions and different species.

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