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Determination of the Changes in Airborne Ba and Mn Concentrations in Düzce City Center in the Last 40 Years with the Help of *Cedrus atlantica* Annual Rings

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Abstract: Heavy metals are pollutants that pose a great threat to humans, other living things, and the ecosystem. All barium (Ba) compounds, one of the most dangerous and harmful heavy metals, are poisonous. Manganese (Mn) is a harmful heavy metal that can cause hallucinations, fatigue, insomnia, weakness, forgetfulness, nerve damage, Parkinson's, and lung embolism. Therefore, observing the changes in Ba and Mn concentrations in the air is essential. This study aimed to determine the change of Ba and Mn heavy metals in the Cedrus atlantica tree based on direction, period, and organ. As a result, the annual rings of *Cedrus atlantica* show that the transfer of both heavy metals into the wood of *Cedrus atlantica* was limited. Therefore, the species is a suitable biomonitor that can be used to monitor the changes in Ba and Mn concentrations in the air. In addition, the study results revealed that the concentrations of both elements in the outer barks are quite high, and Ba pollution is primarily caused by traffic.

Keywords: Atlas cedar, Annual ring, Biomonitor, Barium, Manganese

Öz: Ağır metaller insanlar yanında hem diğer canlılar hem de ekosistem için büyük tehdit oluşturan kirleticilerdir. Ağır metaller içerisinde en tehlikeli ve zararlılarından olan Ba'un bütün bileşikleri zehirlidir. Mn ise halusinasyonlar, bitkinlik, uykusuzluk, güçsüzlük, unutkanlık, sinir hasarları, parkinson, akciğer ambolisi gibi etkilere sebep olabilen zararlı ağır metallerden birisidir. Bundan dolayı havadaki Ba ve Mn konsantrasyonunun değişiminin izlenmesi büyük önem taşımaktadır. Bu çalışmada Ba ve Mn ağır metallerinin *Cedrus atlantica* ağacında yön, dönem ve organ bazında değişiminin belirlenmesi amaçlanmıştır. Çalışma sonucunda *Cedrus atlantica* yıllık halkalarının her iki ağır metalin, *Cedrus atlantica*'da odun içerisindeki transferinin sınırlı düzeyde olduğu, dolayısıyla türün havadaki Ba ve Mn konsantrasyonlarının değişiminin izlenmesi amacıyla kullanılabilecek uygun bir biyomonitor olduğunu göstermektedir. Ayrıca çalışma sonuçları her iki elementin dış kabuklardaki konsantrasyonlarının oldukça yüksek olduğunu ve Ba kirliliğinin büyük oranda trafik kaynaklı olduğunu ortaya koymaktadır.

Anahtar Kelimeler: Atlas sediri, Yıllık halka, Biyomonitor, Baryum, Mangan

1. Introduction

While the proportion of the population living in urban areas worldwide was 9% in the 1900s, it is around 50% today and is estimated to reach 90% in 2030 [1, 2]. Urban areas have many problems, and the most critical problems in these areas are environmental pollution, especially air pollution [3-6], which causes more than 7 million people to die worldwide yearly [7].

Among the air pollution components, heavy metal pollution is the most harmful and dangerous to human health [8, 9]. Due to factors such as the high number of people living per unit area in urban areas, activities carried out to meet the demands and needs of these people and traffic density, some heavy metal concentrations in the air increase significantly, and this increase threatens human health [10].

While some of the heavy metals, many of which are the primary raw materials of industrial and agricultural activities, can be toxic, carcinogenic, and fatal to living creatures even at low concentrations [11], it is stated that even some of them, which are the basic building blocks of living organisms, are harmful at high concentrations [12, 13]. Heavy metals, which do not easily deteriorate through natural processes after their release into nature, pose a great threat to living organisms and the ecosystem [14, 15].

Barium (Ba) and manganese (Mn) are among the most dangerous and harmful heavy metals for living organisms and the ecosystem, especially humans [16]. Ba, which plays a crucial role in the production of many products in the industry, produces many products such as rubber, brake pads, ink, paint, radio vacuum tubes, medicine, machine oil, photo paper, optical glass, plastic and textile products, detergents, batteries, and petroleum industry. Although it is used in production, it is one of the most dangerous heavy metals, and all Ba compounds are poisonous [17]. Mn, one of the heavy metals that is dangerous and harmful to human health, affects mainly the respiratory system and brain when it reaches humans through the food chain. Mn may cause effects such as hallucinations, fatigue, insomnia, weakness, nerve damage, bronchitis, forgetfulness, lung embolism, Parkinson's, and impotence in men [18, 19]. Due to these dangers, it is of great importance to observe the change in heavy metal pollution in urban centers where the population is dense [20].

Monitoring heavy metal pollution in the atmosphere can be done directly or indirectly. However, direct determination of pollution is not preferred because it is pricy, and the direct effect of atmospheric contamination on the ecosystem cannot be determined [21, 22]. The most effectively used method for monitoring the indirect effect of heavy metal contamination is biomonitors. Plants, especially those used as biomonitors, accumulate heavy metals in different plant parts, and determining the metal concentration in these organs provides essential evidence about heavy metals concentration in the airborne [23]. In regions where trees enter dormancy, the development of plants varies depending on the season, and thus, annual tree rings are made in the wood. By comparing the amounts of heavy metals accumulated in the annual rings of trees over a long period, unequal environmental factors and genetic structure can be equalized, and the variation in the long term can be evaluated. Though studies on the usability of annual rings of trees as biomonitors have been carried out for a while, the amount of evidence on the speciation of heavy metals within the plant species and their alteration between organs after their entry into the plant body is relatively restricted [24].

By evaluating the studies on using plants as biomonitors, deficiencies in the literature were identified, and this study was planned. This study determined changes in Ba and Mn concentrations, two of the most hazardous heavy metals for human and environmental health, in the annual rings of a *Cedrus atlantica* tree growing in Düzce, based on organ, period, and direction.

2. Material and Method

The study was carried out on a *Cedrus atlantica* tree growing in Düzce. The log samples used in the study were obtained by marking the north direction on the log and taking its coordinates during the cutting in a park in the center of Düzce province in 2022. The log sample was examined and determined to be 40 years old. The upper surface of the log was smoothed in the laboratory to make the annual rings more clearly visible. Considering the annual ring widths, they are grouped between 1 and 8 age clusters, from outside to inside, for 5 years. After the wood surface was divided into groups and age ranges were determined, samples of wood in each age range were taken with the help of a steel-tipped drill and placed in glass petri dishes. The samples taken into glass containers were kept open in laboratory conditions until they became room dry, and the samples that became room dry were taken to the oven and dried at 45 °C for two weeks. 0.5 g of the dried samples was taken, 6 ml of 65% HNO3 and 2 mL of 30% H2O2 were added and placed in the microwave oven. The program of the microwave device was set to rise to 200 °C for 15 minutes and remain at 200 °C for 15 minutes. After the samples were burned in the microwave oven, the samples in solution were taken into scaled balloon test tubes diluted to 50 mL with ultrapure water. Ba and Mn heavy metal analyses were performed with the ICP-OES device, and the results were calculated by multiplying by the dilution factor. The raw data were evaluated with the help of variance analysis (ANOVA) and the Duncan test using the SPSS 22.0 package program. This method has been frequently used in studies on heavy metal analysis in recent years and is used in both soil [25, 26] and plant parts [27, 28] is frequently used for analysis.

3. Result

The change of Ba concentration based on organ and direction is given in Table 1.

Table 1. Change of Ba concentration based on organ and direction

Organ	North	East	South	West	Average
Outer bark	14232.4^{b}	273.3	5472.9^{b}	23015.0^{c}	10748.4^{b}
Inner bark	553.8^{a}	50.9	78.2^{a}	115.0^{a}	199.4^{a}
Wood	643.0^{a}	181.6	956.8^{a}	1509.4^{b}	822.7^{a}
F value	2104.4***	0.8 ns	65.4***	976.0***	66.1***
Average	1993.0^{AB}	177.7^{A}	1320.5^{A}	3520.5^{B}	3.6*

According to the outcomes of Duncan's test, concentrations followed by the different letters (a, b, and A, B) refer to significant differences among organs within each direction. ns = not significant; * = p<0.05; *** = p<0.001. Capital letters indicate a horizontal direction, while lowercase letters refer to the vertical direction.

According to the results of ANOVA, the change in Ba concentration was statistically noteworthy on an organ basis in all directions except the East. The highest Ba values were obtained in the outer bark. Considering the average values, the highest value was obtained in the West direction, while the lowest was in the East and South direction. The change of Ba concentration based on period and direction is given in Table 2.

Table 2. Change of Ba concentration based on period and direction

Age cluster	North	East	South	West	Average
2018-2022	589.4^{c}	70.5^{a}	2354.4^{g}	1785.5^{e}	1199.9
2013-2017	772.4^{e}	58.3^{a}	1635.6^{f}	1506.8^{d}	993.2
2008-2012	560.6^{c}	45.9^{a}	788.6^{cd}	2125.7^{f}	880.2
2003-2007	659.5^{d}	52.8^{a}	918.4^{d}	1064.1^{c}	673.7
1998-2002	543.8^{c}	648.0^{c}	17.6^{a}	2679.7^{g}	972.3
1993-1997	1142.9^{f}	41.7^{a}	727.3^{c}	1621.8^{d}	883.4
1988-1992	396.4^{a}	33.9^{a}	1062.1^{e}	549.0^{a}	510.3
1983-1987	479.4^{b}	502.0^{b}	150.4^{b}	742.5^{b}	468.5
F value	165.8***	293.5***	306.3***	210.6***	1.5 ns

According to the outcomes of Duncan's test, concentrations followed by the different letters (a and b) refer to significant differences among periods within each direction. ns = not significant; *** = p<0.001.

Ba concentration change was statistically significant in all directions on a period basis. There was no statistically significant difference when evaluating the average Ba values according to the periods. The change of Mn concentration based on organ and direction is given in Table 3.

Table 3. Change of Mn concentration (ppb) based on organ and direction

Organ	North	East	South	West	Average
Outer bark	46820.5^{b}	25772.6^{b}	32826.0^{c}	51295.9^{c}	39178.7^{b}
Inner bark	45325.2^{b}	61515.3^{c}	23162.9^{b}	39837.4^{b}	42460.2^{b}
Wood	8637.0^{a}	7158.0^{a}	7150.9^{a}	8493.9^{a}	7860.0^{a}
F value	161.8***	248.1***	47.0***	107.2***	201.2***
Average	16124.2	14455.2	11319.6	15908.4	0.6 ns

According to the outcomes of Duncan's test, concentrations followed by the different letters (a and b) refer to significant differences among organs within each direction. ns = not significant; *** = p<0.001.

When the Mn concentrations were examined, Mn concentration change was statistically significant in all directions on an organ basis. When evaluating the average values, the highest values were observed in wood, while the lowest values were obtained in barks. There was no statistically noteworthy alteration between directions when the directions were examined according to the average values. The change of Mn concentration based on period and direction is given in Table 4.

Table 4. Change of Mn concentration (ppb) based on period and direction

Age cluster	North	East	South	West	Average
2018-2022	5708.9^{c}	4103.7^{b}	12529.4^{c}	6801.7^{b}	7285.9^{bc}
2013-2017	16951.5^{g}	8213.8^{c}	16401.9^{d}	6657.4^{b}	12056.1^{ef}
2008-2012	13098.1^{f}	14426.4^{f}	11483.4^{c}	16451.7^{d}	13864.9^{f}
2003-2007	5474.4^{c}	12387.0^{e}	4939.7^{b}	18283.3^{e}	10271.1^{de}
1998-2002	9429.3^{d}	9064.5^{d}	3752.6^{a}	11268.9^{c}	8378.8^{cd}
1993-1997	11484.5^{e}	2812.5^{a}	2853.1^{a}	2566.7^{a}	4929.2^{ab}
1988-1992	4017.2^{b}	2593.7^{a}	2594.7^{a}	2765.1^{a}	2992.7^{a}
1983-1987	2932.4^{a}	3662.7^{b}	2652.4^{a}	3156.3^{a}	3100.9^{a}
F value	304.1***	370.1***	226.0***	555.0***	16.2***

According to the outcomes of Duncan's test, concentrations followed by the different letters (a and b) refer to significant differences among periods within each direction. *** = p<0.001.

Considering the results, it was found that the Mn concentration change was statistically significant in all directions on a period basis. While the highest value in the North and South was observed in 2013-2017, the highest value in the East and West was obtained in 2003-2007. According to the average values, it was seen that the Mn concentration is inversely proportional to age, and the concentrations are higher in wood formed in recent years.

4. Discussion

This study examined the usability of *Cedrus atlantica* annual rings in observing the alterations in Ba and Mn concentrations in the air. Since the effects of heavy metals on human and environmental health are known, many studies have been conducted on the usability of tree annual rings in monitoring the change in heavy metal concentrations in the air over time [23, 28, 29]. These studies mainly examined elements such as Pb, Cr, Ni, Co, and Cd [30, 31]. However, elements such as Sr, Pd, As, Be, Pu, Sb, Sr, Th, Tl, and V, which are not subject to much of these studies, are also included in the list of ATSDR's priority pollutants due to their dangers to human and environmental health [15]. Mn and Ba are among the priority pollutants in this list regarding human and environmental health, but the number of studies on these elements is relatively low.

As a result of the study, the highest values were generally obtained in the barks, especially the outer bark. Studies have determined that the maximum concentrations of many heavy metals are obtained in the outer barks. This situation is generally due to the structure of the outer bark and the adhesion of particulate matter contaminated with heavy metals to the outer bark. Particulate matter in the air is contaminated with heavy metals, and since the outer shell surface is rough and cracked, these particles can adhere to the shell surface [11, 29]. As a result of the study, it was determined that Mn concentrations in the inner bark were relatively high. This situation is related to the entry of heavy metals into the plant body. Heavy metals enter the plant body mainly through roots, leaves, and stem parts [32]. It can be said that Mn in the inner bark enters through the trunk parts, and therefore, the concentrations in the inner bark, which is not in contact with air, are higher than in the wood.

The element concentrations subject to the study were quite variable, and there were significant alterations between their concentrations in different directions in the same period or different periods in the same direction in this study. For example, while the concentration of Mn in the western direction in 1993-1997 was 2566.7 ppb, the concentration obtained in 1998-2002 was 11268.9 ppb. Similarly, the concentration obtained in the south direction in the same period was 3752.6 ppb. Similar results were obtained for Ba. These results show that the transfer of Ba and Mn into wood is limited. This result is significant because it emphasizes that the essential need for more information about the usability of biomonitors in monitoring heavy metal pollution is about the transfer of elements in wood. As a matter of fact, studies have shown that the displacement potential of different elements in the wood of different species is at different levels [33-35].

The elements transport within the plants wood is mainly related to the cell structure, especially the cell wall (apoplastic pathway) and stress factors [36]. Plants frequently face abiotic stress factors throughout their life cycle. Among the stress factors that plants encounter most frequently are those related to climatic parameters such as drought [37] and frost [38]. Because plant development depends on the interaction of genetic structure [39] and environmental conditions [40]. Rigid changes in the climate constitute the most important source of stress factors in plant development, and this effect may even result in individual, species, and population losses in large areas [41]. Factors that cause significant and permanent changes in climatic parameters, such as global climate change [42], trigger the stress mechanisms of plants [63]. In addition, increasing UV-B stress due to climate change [43], anthropogenic radiation [27], and heavy metal pollution [18] are important sources of stress for plants. These stress sources inevitably affect plant metabolism and, therefore, plants' heavy metal accumulation potential [36].

The current study obtained the highest Ba concentrations in the western direction. There is a highway located on the west side where the study was conducted. It is stated that Ba is spread into the atmosphere mainly due to anthropogenic sources [69]. This situation is actual for many heavy metals. Studies show that the primary sources of heavy metals are human activities such as traffic, urbanization, industry, and mining activities [44, 45].

5. Conclusion and Suggestions

The study examined the usability of Cedrus atlantica annual tree rings in observing the alterations in Ba and Mn concentrations in the air. The study revealed that Cedrus atlantica is a suitable biomonitor that can monitor the change of both elements. However, reducing heavy metal pollution in the air is as important as monitoring it, and the most suitable instruments that can be used for this purpose are plants. In order to determine the plants that can be used to reduce Ba and Mn concentrations in the air, studies comparing many plants should be carried out. Heavy metals are one of the most critical environmental problems regarding human and environmental health. However, studies have been conducted to monitor and reduce heavy metal pollution, focusing on specific elements. It is recommended that elements such as Be, Sr, Th, Tl, and V should be included in studies on the subject, especially in addition to Ba.

Conflict of Interest

The authors have no conflicts of interest to declare.

Ethics Committee Approval

Not applicable

Author Contribution

Conceptization: SU, NY; methodology and laboratory analyzes: SU, NY; writing draft: SU, NY; proof reading and editing: SU, NY. Other: All authors have read and agreed to the published version of manuscript.

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