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RESEARCH ARTICLE

Availability of Annual Tree Rings for Determining Cr Concentration in Air

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ABSTRACT

According to the "IARC (International Agency for Research on Cancer)," naturally occurring chromium (Cr) in nature has been classified as a carcinogenic element. Cr pollution poses a significant threat to our environment and natural resources, particularly in terms of water and soil contamination. In recent years, this pollution has been increasing due to the influence of anthropogenic sources. In this study, we aimed to define the alterations in Cr over the years using a log sample obtained from a 39-year-old cedar tree in the center of Kastamonu province. Additionally, we aimed to assess the difference in Cr concentration between the parts of the tree facing the road and the parts not facing the road, as well as the distribution of Cr concentration in the plant's wood, bark, and inner bark. The study revealed that the Cr level in the bark on the roadside was 17 times higher than in the non-roadside area. Furthermore, when evaluated on a yearly basis, it was observed that Cr exhibited a certain increase during the years 1987-1992 and 2008-2010 compared to other years. This increase was found to be proportional to the human population growth during those periods. Therefore, the annual tree rings are considered to help determine heavy metal concentrations.

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1. Introduction

Chromium (Cr) was first identified and isolated as a metal by the French chemist Vauquelin in 1798, working with a rare mineral called Siberian red lead (crocoite, $PbCrO^4$) (IARC, 1990). Although Cr is found in several minerals, the sole commercial source of Cr is chromite (Stern, 1982). Chromium, which is one of the most familiar elements in in seawater and Earth's crust, exists in different oxidation states in our environment, primarily as metallic (Cr0), trivalent (Cr^{+3}), and hexavalent (Cr^{+6}) chromium compounds (Cefalu & Hu, 2004). Hexavalent Cr, especially at sufficiently high concentrations, is carcinogenic and toxic (Achmad & Auerkari, 2017). Prolonged exposure to hexavalent Cr is known to pose a cancer risk in the lungs (Keegan et al., 2008; Alvarez et al., 2021). According to the "IARC (International Agency for Research on Cancer)," Cr is categorized as a carcinogenic element (Sharma et al., 2020). The acute and chronic toxicity of Cr is primarily attributed to hexavalent compounds. The most significant toxic effects after exposure to hexavalent Cr compounds through ingestion, inhalation, or contact are known to include gastroenteritis, bronchial carcinomas, dermatitis, hepatocellular insufficiency, nasal septum perforation, eczematous and allergic skin

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reactions, renal oligo-anuric failure, allergic asthma reactions, and mucous and skin membrane ulcerations (Baruthio, 1992).

Anthropogenic sources, particularly emissions from coal combustion and other combustion processes, play a significant role in causing this type of pollution. Approximately 20% of these sources of nickel, arsenic, and selenium, which participate in the global cycle, can constitute approximately 10% of metals such as Cr, cobalt, and cadmium. During ore and metal processing in the industry (as well as other metals), approximately 20% of inputs of arsenic, cadmium, and selenium are disproportionately released into the environment. Significant amounts of Cr and nickel are also released into waters during disposal and incineration processes (Merian, 1984). In this way, the presence of Cr in the atmosphere poses serious threats to human health. Instant measurements can determine the accumulated heavy metal concentrations in the atmosphere over a specific period. However, it is not possible to obtain impressions about the past years' heavy metal concentrations and their processes by looking at the accumulated heavy metal concentrations in the current time frame.

Recent studies shed some light on this issue. Various studies have been conducted to determine past heavy metal concentrations using plants as biomonitors (Sevik et al., 2020; Alttaher Ateya et al., 2023). In some studies, the determination of sulfur changes in the air was achieved using annual rings of *Cedrus atlantica* (Sevik et al., 2023), and the concentrations of Al, Fe, and Cd elements in the atmosphere were determined using the tree rings of Turkish hazelnut (Key et al., 2022). Furthermore, annual rings have been utilized in studies examining not only metal concentration levels but also the relationship of the plant with nutrient elements (Çobanoğlu et al., 2022; Koç et al., 2022; Saleh & Cobanoğlu, 2022). Previous studies have used annual rings of cedar to determine Cd, Ni, and Zn concentrations in other locations (Cobanoglu et al., 2023), as well as Ni and Co concentrations in *Cedrus atlantica* (Koç, 2021), and Cr and Mn concentrations (Savas et al., 2021). Studies show that various plant species can be used as biomonitors in determining nutrient and heavy metal concentrations (Turkyilmaz et al., 2019).

Therefore, this study is aimed to reveal the Cr concentration variation in the Kastamonu center in 39 years by using the annual rings of the cedar tree.

2. Materials and Methods

Our study was conducted using trunk samples from a 39-year-old cedar tree that was cut down due to landscape maintenance in the city center of Kastamonu. Samples were taken from the trunk at breast height, with a diameter of 45.7 cm. The side facing the road and the side facing away from the road were marked on the log. In order to facilitate more efficient sampling of the log, one side was sanded to create a smooth surface. The age of the tree was determined by counting the annual rings on the smooth surface, which revealed a total of 39 rings. In order to prevent the mixing of the samples, the annual rings were grouped from outer to inner in three-year intervals (1-13 age groups).

After the grouping process, samples were taken from the outer bark, inner bark, and each age group's wood section using a steel drill to avoid affecting the concentration variation. The collected samples were then converted into sawdust and left to air dry in glass petri dishes in the laboratory for 15 days. The average ambient temperature ranged from 25 ± 5 °C. Once the samples were air-dried, they were kept in an oven at 45 °C for 7 days. After 7 days, the dried samples were taken, and 0.5 g of each sample was mixed with 2 ml of H_2O_2 (30%) and 6 ml of HNO_3 (65%). The samples with added solutions were burned in a microwave oven. Once the samples were thoroughly dissolved, they were transferred to volumetric flasks and diluted to 50 ml with ultra-pure water. Since the weight of the samples taken from the sawdust was 0.5 g and diluted to 50 ml with water, the analysis results were multiplied by 100 (dilution factor). All samples were analyzed with three replications.

The study data was evaluated using the SPSS software package based on the results of ANOVA and Duncan tests. The study data was compared and interpreted by comparing the inward-facing (non-road-facing) and road-facing sections.

3. Results

The average values and Duncan test results for the directional change of Cr element at the organelle level, evaluated using the SPSS software package, are provided in Table 1.

Table 1. Change of Cr element concentration (ppb) levels on the basis of organelles.

Organelle	Side facing the road direction (inward)	Side facing opposite the road direction (outward)	Average
Outer bark	458.400 a	7838.800 c	4148.600 c
Inner bark	2092.800 b	2281.133 b	2186.967 b
Wood	652.333 a	935.584 a	793.959 a
F value	130.209	978.957	34.999
p value	0.000	0.000	0.000

According to the Duncan test results, a,b, and c show that they are in the different group, and same letter means there is no difference between them.

Upon examining the values in Table 1, statistically significant differences in the analysis of variance results were determined for the concentration of the Cr element in average values and inward and outward directions ($p<0.001$). According to the Duncan test results, there was no significant difference in the Cr concentrations between the outer bark and wood samples on the side facing the road direction. The highest concentration of Cr element was found in the outer bark sample in the outward section, while in the inward section, it was detected in the inner bark sample. Based on the table values, the Cr concentration level in the outer bark in the outward section was determined to be 17 times more elevated than that in the outer bark in the side facing the road direction. The higher concentration of Cr in the outer bark in the outward section is thought to be due to direct exposure to particulate matter.

The average values and Duncan test results for the yearly variation of Cr element, among the elements studied, at the organelle level are provided in Table 2.

Table 2. Change of Cr element concentration (ppb) by year.

Age periods	Inward	Outward	Average
1978-1980	822.600 g	752.000 abc	787.300 ab
1981-1983	692.733 f	802.000 bc	747.367 ab
1984-1986	694.000 f	733.200 ab	713.600 ab
1987-1989	1020.400 h	1318.133 d	1169.267 c
1990-1992	635.600 de	1337.733 d	986.667 bc
1993-1995	688.133 f	758.867 abc	723.500 ab
1996-1998	695.000 f	727.400 a	711.200 ab
1999-2001	662.067 ef	770.667 abc	716.367 ab
2002-2004	627.333 de	715.533 a	671.433 ab
2005-2007	606.867 d	743.600 abc	675.233 ab
2008-2010	528.600 c	1309.667 d	919.133 abc
2011-2013	456.400 b	815.133 c	635.767 a
2014-2016	350.600 a	1378.667 d	864.633 ab
F value	152.304	154.996	2.445
p value	0.000	0.000	0.011

According to the Duncan test results, a,b, and c show that they are in the different group, and same letter means there is no difference between them.

When the Cr concentration on a yearly basis was evaluated in compliance with the concentration levels in Table 2, statistically significant differences were determined in both inward and outward directions based on the variance analysis results ($p<0.001$). Significant differentiations were also observed in the average values ($p<0.05$). According to Table 2, as shown in Figure 1, the concentration variation of Cr element on a yearly basis generally ranged between 600.00-700.00 ppb over the past 39 years. However, it was observed that in the outward direction, the Cr concentration reached values as high as 1300.00 ppb during the years 1987-1992, 2008-2010, and 2014-2016. In contrast, in the inward direction, a decrease in Cr concentration was observed during the years 1990-1992 (635.600 ppb), 2008-2010 (528.600 ppb), and 2014-2016 (350.600 ppb). It is even seen that it decreases to the lowest ppb values. The Cr concentration level was usually elevated in the outward direction throughout all years. Therefore, vehicles in traffic are believed to be a source of heavy metal accumulation in the atmosphere.

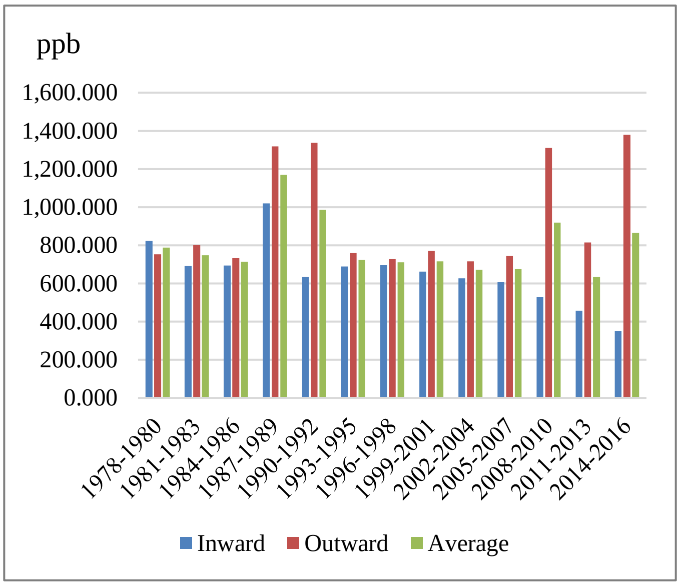

Figure1. Change in concentration (ppb) of Cr element by year.

4. Discussion and Conclusion

Global climate change (Sargıncı & Beyazyüz, 2022), general and local temperature increases (Koç, 2022a,b; Cetin et al., 2023), drought (Koç, 2022c; Koç & Nzokou, 2022; Yılmaz et al., 2022; Koç & Nzokou, 2023), extreme temperatures, population growth, and pollution (Frondel et al., 2018) are the world's leading environmental problems. Due to increasing population, industrialization, and environmental degradation, the air in the atmosphere is becoming increasingly polluted every day. This pollution, which is not limited to a decrease in air quality, reaches even more harmful and potentially lethal levels by introducing heavy metals into the atmosphere through various pathways. In our study, it has been revealed how the Cr concentration has changed significantly over the years. Although all the factors contributing to Cr concentration in the atmosphere are unknown, traffic is believed to have a significant impact. The study shows that, particularly during certain periods (1987-1989), the Cr concentration reached highly hazardous levels for human health.

Within the scope of the study, when evaluating the side facing the roadside and opposite direction sections separately, it was determined that the Cr concentration in the inward section was higher than in the outward section. Other studies have also shown that heavy metal concentrations can vary at the organelle level (Erdem, 2018; Sevik et al., 2018; Mossi, 2018). Similar studies indicate that metal concentration levels accumulated in the bark are generally much elevated than those in the wood (Mossi, 2018). In fact, there can be differences of up to 9 times in Cr concentration between the wood and the bark (Turkyilmaz et al., 2018). This study determined that the Cr concentration level in the bark sample facing the road was 17 times higher than in the outward section. This is believed to be due to the greater exposure of the roadside-facing bark to particulate matter from the road. It is thought that particulate matter suspended in the atmosphere can significantly affect the concentration of heavy metals.

Additionally, the potential of trees to absorb and retain particulate matter, including heavy metals, is strictly related to the physiological and physical traits of the plants. Factors such as surface area, roughness, trichomes, and plant organelle structures also affect the deposition of metals, especially on leaf surfaces and other organelles (Schreiber & Schönherr, 1992; Cunha & do Nascimento, 2009; Ataabadi et al., 2011). It is known that plant tissues have the potential to transform potentially toxic Cr (VI) species into non-toxic Cr (III) species (Wang et al., 2011).

Therefore, it is possible that the contamination of the bark and leaves of the tree under study with traffic-related heavy metal-contaminated particles, their penetration into the tree through processes such as photosynthesis and osmosis, and thus the elevation of heavy metal concentrations in the roadside-facing section. Numerous studies have shown that metal concentration levels in trees grown in regions with dense traffic are higher compared to trees grown in regions with no or less traffic (Çelik et al., 2005; Buachoon, 2014; Huber et al., 2016; Yang et al., 2017; Erdem, 2018; Sevik et al., 2018; Isinkaralar et al., 2022a). Moreover, it is known that Mn, Zn, Ni, and Co elements have higher values in industrial areas, while Pb, Cd, Fe, and Cr concentrations are higher in areas with heavy traffic. In a study, they stated that while the Cr concentration was at the level of 16,595 ppm in areas with no traffic, it increased to 23,716 ppm in areas with heavy traffic (El-Hasan et al., 2002).

The study concludes that while there is a general horizontal variation in Cr elements, there has been an escalation in recent periods. This increase is believed to be associated with the increased human population and the quantity of automobiles in the Kastamonu recently. The human population in the center of Kastamonu was reported to be 115,332 people in 2007, 123,972 people in 2010, and 146,103 people in 2016 (Anonymous, 2023). This status is likely to be attributed to the current population growth and the consequent increase in anthropogenic contaminants. Thus, contamination levels should be maintained in regions where population density is rapidly increasing, and necessary precautions should be taken.

It is known that Cr is typically associated with particulate matter in ambient air at concentrations ranging from 0.001 to 0.1 $\mu g/m^3$ (Fishbein, 1976). Average chromium levels in the range of 0.01 to 0.04 $\mu g/m^3$ (maximum value of 0.54 $\mu g/m^3$) were reported at 15 stations during the period of May 1972 to April 1975. These values were considered to reflect background pollution and represent levels in the air inhaled by most of the population (Kretzschmar et al., 1977). Coal, soil, and rocks can contain Cr up to 54 ppm (mg/kg), similar to many other sources. The burning of coal can contribute to Cr levels in the air, especially in urban areas (Fishbein, 1976; Merian, 1984). Particles emitted from coal-fired power plants can contain Cr in the range of 2.3 to 31 ppm (mg/kg), depending on the type of boiler combustion, while the emitted gases can contain 0.22 to 2.2 mg/m3 of chromium (Fishbein, 1976). According to these studies, it can be inferred that Cr concentration may increase due to population growth, and coal burning can contribute to the increase in Cr concentration, supporting the idea that Cr concentration can be released into the atmosphere through various human activities, including traffic. The literature review conducted in this study revealed a considerable boost in the human population of the city center zone throughout the mentioned periods, along with intense coal burning (İbret & Aydınözü, 2013; Isinkaralar et al., 2022b). Indeed, it is reported by the residents that there was a significant escalation in air contamination during those periods. Therefore, it can be stated that tree rings are convenient in assessing the variation in heavy metal concentrations over the years.

5. Suggestions

Dendrochemical and dendrochronological investigations conducted on annual tree rings can provide valuable information about the environment of a province or the characteristics that stress the trees. In order to obtain data on heavy metal concentration levels within the annual rings, experts must examine the tree's annual rings more thoroughly. In order to determine heavy metal concentrations within the annual rings, it is necessary to obtain these rings from inside the tree in some way. The most practical approach for this is clearly through tree cutting. Nevertheless, this procedure can be done without preventing the trees from continuing their lives. In various examinations carried out for this goal, tree rings can be taken from the tree using an increment auger without harming the plants or ending the tree's life by causing minimal damage.

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Conflict of Interest

The authors declare that they have no conflict of interest.

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