Water Air Soil Pollut (2022) 233: 193
https://doi.org/10.1007/s11270-022-05667-w

# The Use of *Cupressus arizonica* as a Biomonitor of Li, Fe, and Cr Pollution in Kastamonu

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Received: 19 March 2022 / Accepted: 14 May 2022 / Published online: 23 May 2022

**Abstract** Air pollution became an important problem that severely threatens human health, especially in urban areas. Such that it was reported that 90% of global population respire polluted air and the air pollution causes the death of approx. 7 million annually. It is known that heavy metals have a significantly high share in these deaths. Heavy metals are elements, which do not easily disintegrate in nature, can accumulate in bodies of living organisms, and can be toxic, carcinogenic, and even lethal. Thus, monitoring the changes of heavy metal concentrations in the air is very important for human and environmental health. The method most preferred for determining the change of heavy metal concentration in the atmosphere is the biomonitors. The use of annual rings of trees is one of the most accurate methods because they can provide information about the long-term change of heavy metal concentrations in the air. In the present study, it was aimed to reveal the usability of annual rings of *Cupressus arizonica* as a biomonitor in determining the change of Li, Fe, and Cr concentrations. Within the scope of this study, the

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concentrations of elements in the inner bark, outer bark, and wood in the roadsides, where there is intense traffic, and opposite sides of trees were compared and the annual changes in heavy metal concentrations in annual rings were investigated. At the end of this study, it was determined that Fe and Cr concentrations in the roadside outer barks of trees were generally at very high levels, that the annual changes of elements generally followed a fluctuating course, and that annual rings of *Cupressus arizonica* are very useful for monitoring the annual change of Fe concentration in the air but not for monitoring the changes in Li and Cr concentrations.

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

## 1 Introduction

Together with the Industrial Revolution, the needs and demands of human have diversified and increased in the last century and the production aiming to meet these needs and demands resulted in the extraction of underground mineral sources and their use in industry as a raw material (Shahid et al. 2017; Koc, 2021). In this process, the concentrations of various elements, which are used as a raw material in industry, in air (Aricak et al., 2019; Sevik et al., 2019), water (Ucun Ozel et al., 2020), and soil (Bayraktar, 2021; Kaplan et al., 2021) have significantly increased. Heavy metals, which constitute a group among these materials and can be very harmful to both human and environmental health, have specific importance (Savas et al. 2020). Since heavy metals do not easily disintegrate in nature, they can accumulate in living organisms, and some of them can be toxic, carcinogenic, and even lethal even at low concentrations, and even the nutrients that are necessary for organisms have toxic effects on humans when at high concentrations; it is essential to monitor the concentrations of heavy metals (Bozdogan Sert et al., 2019; Turkyilmaz et al., 2019).

The most remarkable anthropogenic sources of heavy metals are industrial and traffic activities (Aricak et al., 2020). Hence, it is very important to monitor the heavy metal concentrations in areas having high levels of industrial and traffic activities. It is known that the traffic density is very high in urban areas hosting intense populations; and thus, it is much more important to monitor the heavy metal concentrations in these city centers. Because the heavy metal pollution in these urban areas is at a higher level and since the number of individuals living in a unit area is higher, the number of individuals influenced by this pollution is also higher (Alaquori et al. 2020). The European Environment Agency reported that the living areas of approx. 2.5 million individuals have been polluted and 14% of these areas required emergency recovery plans (Akarsu, 2019; Akarsu et al., 2019; Adiguzel et al., 2022; Bozdogan Sert et al., 2021; Varol et al., 2022; yucedag et al. 2021; Zeren cetin et al., 2020). As stated by the World Health Organization (WHO), 90% of the global population respire polluted air and air pollution is responsible for the death of approx. 7 million individuals annually (Jo et al., 2020). It is thought that heavy metals are responsible for a large portion of these deaths.

The heavy metals accumulating in the air land on the earth through precipitation, snow, or gravity and pollute the soil and water sources, and may directly harm people through respiration. In London, approx. 4000 individuals lost their lives between December 5th and 9th, 1952, due to the reasons related with air pollution and, together with the individuals losing their lives in following months, the total number of deaths reached 12,000; the investigations revealed that the lungs of victims were contaminated by very small particles incorporating heavy metals including Pb, Zn, and Fe (Shahid et al. 2017; Ateya, 2020). Thus, monitoring the heavy metal concentrations in the air is very important.

Directly monitoring the air pollution is not widely preferred since it is expensive and the direct effects on the ecosystem cannot be determined (Turkyilmaz et al., 2018). Moreover, direct measurements do not provide information about the heavy metal concentrations in previous years. Thus, the change cannot be traced and the change in pollution due to anthropogenic factors cannot be determined. Hence, biomonitors are among the methods most widely used in monitoring the heavy metal pollution (Sevik et al., 2020a).

Plants are the most widely used biomonitors since they constantly remain in a single place and can be sampled easily. However, the leaves of non-evergreen plants are not considered to be useful biomonitors because it is not known how long they have been subjected to heavy metals in the air, while the leaves of evergreen plants are also not considered useful biomonitors because they provide information only about the heavy metal concentration accumulating in a single vegetation period (Koc et al. 2021). The accumulation of heavy metals in the plants' organism is shaped by many factors and it is very difficult to interpret what the heavy metal concentrations in plants' organs do mean. Hence, the leaves of species such as pine, spruce, and fir, on which the leaves remain for a long time and age of the leaves of which can be measured, are the biomonitors that can be used in monitoring the change of heavy metal pollution (Cetin et al., 2020). However, this method can provide insight only for the change in a maximum 8 to 10 years.

One of the most useful methods to be used for monitoring the long-term changes in the heavy metal concentrations in air is the annual rings of trees. In regions where the winter season is observed, the development of trees varies between the seasons; and thus, annual rings form in the wood part. The heavy metals accumulating in the annual rings of trees in the long-term can provide important information about the change of air pollution (Zeren 2017a,b; Sevik et al., 2020a, 2020b, 2021; Cetin & Jawed, 2022; Cesur et al., 2021).

In the present study, it was aimed to determine the annual changes in concentrations of Li, Fe, and Cr in annual rings of *Cupressus arizonica* grown in the city center of Kastamonu. In this parallel, by determining the heavy metal concentrations in the inner and outer barks, the heavy metal concentrations were compared and it was examined how these changes in trees changed in different sides of trees.

## 2 Material and Method

This study was carried out on *Cupressus arizonica* grown in the city center of Kastamonu. The log cut from the trunk of *Cupressus arizonica* at a 50-cm height was taken to the laboratory by marking the roadside face. In order to make the annual rings more apparent, the log was ground in the laboratory and, by counting the number of annual rings, it was determined that the tree was 21 years old. Considering the widths of annual rings, the part covering the first 7 years was assigned as the first group and the year-based samples were taken from the next annual rings.

The samples were taken using a steel-bit drill and no instrument made of elements, which were examined in the present study, was used. In roadside and opposite side, the samples were taken from the inner bark and outer bark, and every annual ring. Then, the samples were taken to glass Petri containers and labeled. The samples were ground to sawdust, dried through room-drying for 30 days under room conditions, and then dried at 50 °C in a drying oven for a week.

The dried samples were powdered. In total, 0.5 g of dried samples was weighed and then put into tubes designed for the microwave oven. In these tubes, these samples were added with 10 ml 65% $HNO_3$ and 2 ml 30% $H_2O_2$. Security was paid significant attention during these procedures. The samples were burnt in a microwave under 280 PSI pressure and at 180 °C for 20 min. During this process, the device was programmed to increase the temperature to 200 °C in 15 min and then stay at 200 °C for 15 min. The tubes taken out of the microwave were left for cooling and then filled to 50 ml using deionized water. Then, the samples were filtered using filter paper and read using a ICP-OES (inductive coupled plasma-optic emission spectrometer) device at appropriate wavelengths. The results were multiplied by 100. The measurements were in triplicate. All the data obtained were analyzed using the SPSS package program, and variance analysis and Duncan test were applied.

**Table 1** Elements' change by organs

| Organ | Li | | | Fe | | | Cr | | |
|---|---|---|---|---|---|---|---|---|---|
| | IS | RS | M | IS | RS | M | IS | RS | M |
| OB | 848.2 a | 814.6 ab | 831.4 | 4.63 | 60.95 c | 32.79 b | 1372.2 b | 2691.3 b | 2031.7 b |
| IB | 1474.0 b | 566.7 a | 1020.4 | 8.48 | 23.56 b | 16.02 a | 1091.8 a | 829.2 a | 960.50 a |
| WO | 1291.4 b | 981.6 b | 1136.5 | 2.99 | 7.77 a | 5.46 a | 996.6 a | 1077.0 a | 1036.8 a |
| *F* | 4012 | 7269 | 3094 | 4689 | 99,819 | 27,628 | 4986 | 87,114 | 40,417 |
| Sig | 0.024 | 0.002 | 0.050 | 0.140 | 0.000 | 0.000 | 0.011 | 0.000 | 0.000 |

## 3 Results

The changes of Li, Fe, and Cr elements in different directions and by the organs were determined. The mean values, *F* value obtained from variance analysis, error rate, and groups after the Duncan test are presented in Table 1.

*IB*, inner bark; *OB*, outer bark; *WO*, wood; *IS*, inner side; *RS*, roadside; *M*, mean.

As a result of the variance analysis, it was determined that the changes of Fe values in ID direction by the organs were found to be non-significant at the confidence rate of 95%. All other changes by all directions and organs were found to be statistically at the minimum confidence level of 95%.

Given the Li values, it was determined that the highest concentrations in ID direction were found in the inner bark and the lowest value in the outer bark, whereas the highest value was found in wood and the lowest value in the inner bark in roadside direction. Considering the mean values, the highest values were found in wood. It was observed that there was no remarkable difference between inner-side and roadside directions for Li concentrations.

There were very remarkable differences between inner-side and roadside directions for Fe concentrations. Considering the mean values, Fe concentrations obtained from roadside direction were 13.1 folds of Fe concentrations obtained from the inner-side direction in the outer bark, 2.7 folds in the inner bark, and 2.6 folds in wood. Similarly, there were remarkable differences between the organs. While there was no difference between the organs in ID direction, the highest values were obtained from the outer bark in YD direction. Considering the mean values, the values obtained from wood and inner bark were in the same group, whereas the values obtained from outer bark constituted the second group.

Considering the concentrations of Cr element, it was observed that there were very remarkable differences between the values obtained from the outer bark in roadside and inner-side directions. The values obtained from the outer bark in roadside direction were approx. 2 folds of values obtained from the inner-side direction. Considering the mean values, the lowest values were obtained from the inner bark (960.5 ppb) and wood (1036.8 ppb), while the value obtained from outer bark (2031.7 ppb) was approx. 2 folds of these values.

**Table 2** Annual change of Li (ppb) element in woods

| Ages | IS | RS | M |
|---|---|---|---|
| First 7 ages | 1775.0 e | 1247.6 f | 1511.3 |
| 8. age | 959.4 ab | 1170.8 def | 1065.1 |
| 9. age | 1391.6 d | 1179.3 ef | 1285.5 |
| 10. age | 1241.2 bcd | 992.0 cdef | 1116.6 |
| 11. age | 1085.9 abcd | 1011.4 cdef | 1048.7 |
| 12. age | 1030.8 abc | 1120.0 cdefg | 1075.4 |
| 13. age | 1145.6 abcd | 963.4 bcde | 1054.5 |
| 14. age | 1357.2 cd | 866.7 abc | 1112.0 |
| 15. age | 1206.0 bcd | 709.4 ab | 957.7 |
| 16. age | 1426.4 d | 1045.1 cdef | 1235.7 |
| 17. age | 1387.5 d | 910.6 abcd | 1149.1 |
| 18. age | 1787.5 e | 677.8 a | 1232.6 |
| 19. age | 837.0 a | 881.7 abc | 859.4 |
| 20. age | 1323.6 cd | 1008.4 cdef | 1166.0 |
| 21. age | 1416.9 d | 940.8 bcde | 1178.8 |
| *F* value | 6888 | 4178 | 1748 |
| Sig | 0.000 | 0.000 | 0.063 |

The annual changes of elemental concentrations in wood were examined separately and the annual change of Li element is presented in Table 2.

Given the results of variance analysis for the change of Li element's concentrations in wood by year, it can be seen that there were statistically significant differences between years in both inner-side and outer-side directions at the confidence level of 99.9%, while there was no significant difference between years at the minimum 95% confidence level.

Given the values presented in Table 2, it can be seen that the highest Li concentration in ID direction was found in the 18th age (1787.50 ppb) and in the first 7 years of age (1775.0 ppb), whereas the lowest values were found in the 19th age (837.06 ppb) and 8th age (959.46 ppb) following those years. In the YD direction, Li concentrations followed a fluctuating course in general and ranged between 677.8 and 1247.6 ppb. The changes of Fe concentrations in woods by years are presented in Table 3.

Examining the results of variance analysis on the changes of Fe concentrations in wood by years, it was determined that there were statistically significant differences between the annual mean values in both inner-side and roadside directions (minimum confidence level of 99%).

**Table 3** Changes of Fe (ppm) concentrations in woods by years

| Ages | IS | RS | M |
|---|---|---|---|
| First 7 ages | - | 2.90 bc | 2.90 ab |
| 8. age | 10.82 h | 2.41 b | 6.62 ab |
| 9. age | 2.84 e | 2.33 b | 2.59 a |
| 10. age | 0.94 bc | 10.08 g | 5.51 ab |
| 11. age | 0.29 ab | 1.10 a | 0.70 a |
| 12. age | 0.49 ab | 3.24 c | 1.87 a |
| 13. age | 0.06 a | 1.19 a | 0.62 a |
| 14. age | 1.47 cd | 5.96 d | 3.71 ab |
| 15. age | 0.45 ab | 8.84 f | 4.65 ab |
| 16. age | 1.50 cd | 26.25 j | 13.87 c |
| 17. age | 1.98 d | 11.58 h | 6.78 ab |
| 18. age | 1.72 cd | 18.40 i | 10.06 bc |
| 19. age | 7.01 g | 6.58 d | 6.79 ab |
| 20. age | 4.94 f | 7.97 e | 6.45 ab |
| 21. age | 7.32 g | 7.69 e | 7.51 abc |
| *F* value | 161,397 | 825,394 | 2891 |
| Sig | 0.000 | 0.000 | 0.002 |

Given the values presented in Table, it can be seen that the highest value in inner-side direction was observed in the 8th age (10.828 ppm). It was also determined that the values remained at low levels until the 18th age, increased after the 19th age, and reached the level 4 folds of the value observed for the 18th age. The values remained at these levels during the next years.

In the roadside direction, the values were at approx. 1.11 ppm levels in the 11th age, increased to 26.25 ppm in the 16th age, and then decreased to the values, which were observed in the inner-side direction, between the 19th and 21st ages. The changes in Cr concentrations in woods by years are presented in Table 4.

**Table 4** Changes of Cr (ppb) concentrations in wood by year

| Ages | IS | RS | M |
|---|---|---|---|
| First 7 ages | 1029.0 d | 1036.4 cde | 1032.7 abcd |
| 8. age | 747.6 a | 991.8 bcde | 869.7 a |
| 9. age | 1258.5 e | 1119.2 e | 1188.8 de |
| 10. age | 823.0 b | 1488.0 g | 1155.5 cde |
| 11. age | 869.4 bc | 984.3 bcd | 926.8 ab |
| 12. age | 694.9 a | 1024.5 cde | 859.7 a |
| 13. age | 883.0 c | 930.1 abc | 906.5 ab |
| 14. age | 925.4 c | 940.1 abc | 932.7 abc |
| 15. age | 993.0 d | 1003.1 bcde | 998.1 abcd |
| 16. age | 1527.7 f | 1096.3 de | 1312.0 e |
| 17. age | 998.9 d | 1237.7 f | 1118.3 bcde |
| 18. age | 1033.8 d | 987.5 bcde | 1115.5 bcde |
| 19. age | 903.2 c | 820.7 a | 861.9 a |
| 20. age | 1243.5 e | 886.7 ab | 960.2 abc |
| 21. age | 1019.1 d | 1609.3 h | 1314.2 e |
| *F* value | 131,013 | 29,081 | 5027 |
| Sig | 0.000 | 0.000 | 0.000 |

Given the results of variance analysis showing the changes in Cr concentration in wood by year, it can be seen that there were significant differences between annual mean values in both inner-side and roadside directions at the confidence level of 99.9%. Examining the changes of Cr concentration in wood by years, the concentration ranged between 694.9 ppb (12th age) and 1527.7 ppb (16th age) in inner-side direction and between 820.7 ppb (19th age) and 1609.0 ppb (21st age) in roadside direction. The remarkable increase in roadside direction especially in the last 3 years draws attention.

## 4 Discussions

At the end of this study, it was determined that there were significant differences between the organs except for outward portions for Li element and inward portions for Fe element. The highest concentrations were observed in wood of Li, whereas the highest concentrations of Fe and Cr were found in the outer bark for all directions (the inward change of Fe concentrations was not statistically significant). All the values obtained for Fe and Cr in wood were in the first group in the Duncan test. Accordingly, the organ-based changes of Fe and Cr concentrations were wood < inner bark < outer bark.

Previous studies carried out on this subject reported similar results. In a study carried out on cedar, Akarsu (2019) reported that the changes in Zn, Mn, Fe, K, Ca, and B concentrations generally lined from the lowest to highest as wood < inner bark < outer bark, whereas Sevik et al., (2020a, 2020b) reported that Pb, Co, and Fe concentrations were at the highest level in the outer bark and the lowest level in wood. In different studies, the highest concentrations were detected in the outer bark (Zeren 2017a,b; Sevik et al., 2020a, 2020b, 2021; Cetin & Jawed, 2022). Savas (2021) found the lowest Ca, Cr, Al, and P concentrations in wood and the highest concentrations of most of these elements in the outer bark. Turkyilmaz et al. (2018) stated that Cr concentration found in bark was 9 folds of the concentration found in wood. However, in a study carried out on *Acer platanoides*, Zn concentration found in bark was higher than 3.5 folds of concentration found in wood. Janta et al. (2016) reported that the highest Cu, Fe, and Zn concentrations in different layers of *Cassia fistula*'s bark were found in the outer bark.

Moreover, within the scope of the present study, it was also found that the values measured in the outer bark in roadside direction were much higher than those measured in the outer bark in the opposite direction. Cr concentration was found to be 1372.2 ppb in the outer bark in inward direction and 2691.3 ppb in the outer bark in outward direction, whereas Fe concentration was found to be 4.63 ppm in the outer bark in inward direction and 60.95 ppm in the outer bark in outward direction.

Both much higher Fe and Cr concentrations in the outer bark than in the inner bark and wood and much higher concentrations found in the outward side of the outer bark in comparison to the inward side of the outer bark are thought to be largely related with particles. Previous studies revealed that heavy metals in the air adhere on and contaminate the particle materials and, as a result of these particles' adhesion to the plant organelles, the heavy metal concentrations in these organelles increase (Savas et al., 2021; Turkyilmaz et al., 2020). The rough structure of the outer bark allows the particles to easily bind.

In this study, the concentrations of Fe and Cr in the outer bark were very high, whereas the highest Li concentrations were found in the inner bark. It suggests that the Li element has no traffic-related origin; and thus, particle materials were not contaminated by Li element. Cetin et al. (2020) compared the Li concentrations in needle, bark, and branches of *Picea pungens* in different ages and the highest concentrations were found in washed needles for all the ages. The fact that the highest Li concentration was found in both needles and washed needles suggests that Li concentration is caused by traffic. However, various studies reported that Fe and Cr have traffic origins (Sevik et al., 2019; Turkyilmaz et al., 2019). Hence, very high Fe and Cr concentrations found in the outward side of the outer bark are the results of adhesion of particles, which have been contaminated by Fe and Cr on the outer bark.

As a result of this study, it was determined that the changes of concentrations by years generally followed a fluctuating course. While the change of Li concentrations by years occurred in a narrow range, Fe concentration in inward portions significantly increased in the last 3 years but the increase in outward portions started in the 13th age. For the Cr element, the highest concentration in the roadside direction was found in the last age. Akarsu (2019) reported that, although the change in concentrations of Ba, Li, Ca, Mg, Mn, and Cr in Kastamonu province generally followed a horizontal course, an increase was observed in recent years and this finding was related with the increase in population and the number of vehicles in the Kastamonu city center.

Within the scope of this study, while the highest Fe concentrations in the woods were found in the outward direction with high traffic density, the highest Cr concentrations were found sometimes inward and sometimes outward directions. Considering Li, the concentrations found in inward parts were generally higher than those found in outward parts. This finding showed that Fe concentration in annual rings varied depending on the density of traffic, whereas the accumulation of Cr and Li in woods was not affected by the traffic density.

One of the most important uncertainties regarding the use of biomonitors in monitoring the air metal concentrations in the air is the speciation and transmission of elements since the moment of entering the plant organism (Shahid et al. 2017). Since there are few studies carried out on this subject, there is limited information. Koc (2021), in a study carried out on *Cedrus atlantica*, reported that the transfer of Ni and Co was at a limited level. Zhang et al. (2019) stated that Zn and Pb concentrations migrated to a limited level in annual rings but Cu concentration did not migrate at all.

After the importance of heavy metals for human health was revealed, many studies were carried out on monitoring the change of concentration in the air. However, there are many factors influencing the inlet and accumulation of heavy metals in plant organisms. These factors include plant species, level of precipitation and humidity, plant habitus, organ structure, type of heavy metal, and interaction with plant (Aricak et al., 2020; Ozel et al., 2021; Varol et al., 2021). Hence, it is not possible to calculate what a specific heavy metal concentration detected in a plant organism means or to what extent it reflects the heavy metal level in the medium. For this reason, since the annual rings of trees allow the chance for comparison, they are considered one of the most suitable methods for monitoring the changes throughout the process. Moreover, it should be determined through the annual rings which trees can be used in monitoring the change of which heavy metal concentrations. In one of the previous studies carried out on this subject, Koc (2021) reported that the annual rings of *Cedrus atlantica* were very suitable for monitoring the change of Ni concentration but not for the change of Co concentration. Hence, the species suitable for monitoring the change of concentration should be determined separately for each type of heavy metal.

## 5 Conclusions

The results obtained here showed that the accumulations of Li, Fe, and Cr concentrations in the air in different organs of the tree trunk were at different levels and the highest Fe and Cr concentrations were found in the outer bark. However, because of its rough structure, the outer bark holds particles to a large extent and, since these particle materials are contaminated by heavy metals, it significantly changes the heavy metal concentrations detected in the bark. Furthermore, the age of the bark is related to the age of tree and the exfoliation of the bark in the course of time makes it impossible to determine which part of the bark has been subjected to air infected with heavy metals for how long. Thus, it is not recommended to use barks as a biomonitor in monitoring heavy metal pollution.

Examining the results obtained within the scope of this study, it was determined that there were very remarkable differences between Fe concentrations in the barks developed in the same year but in different directions or in the barks developing in subsequent years but in the same direction. However, the change in Li and Cr remained at a limited level. This finding suggests that the migration of Fe within the wood is very limited. But, it is very difficult to say that it applies to Li and Cr. Hence, it can be stated that the annual rings of *Cupressus arizonica* are very suitable for monitoring the change of Fe concentrations in the air but not for monitoring the changes of Li and Cr concentrations.

The method used in the present study revealed that annual rings are very useful biomonitors to monitor the change of heavy metal concentrations but not every tree can be used for every element. Hence, it should be separately determined if the annual rings of which trees are suitable for monitoring the concentrations of which heavy metals.

**Acknowledgements** This research is Alican Cesur's Master Thesis in Kastamonu University, Institute of Science, Programs of Sustainable Agriculture and Natural Plant Resources conducted one of part of this research. He sincerely thanks his Advisor's Assoc. Prof. Dr. Mehmet Cetin and Kastamonu University. In addition, we thank Bartin University collaborate with Turkish Council of Higher Education (YÖK) for 100/2000 Doctoral Scholarship. We thank Bartin University and YÖK for the 100/2000 PhD Scholarship Project and TÜBİTAK for their support.

**Author Contribution** Alican Cesur, Ilknur Zeren Cetin, Mehmet Cetin, Hakan Sevik, Halil Baris Ozel: design, resources, writing; materials, data collection and/or processing; materials, literature search, analysis and/or interpretation.

**Funding** The Scientific and Technological Research Council of Turkey (TÜBİTAK), Turkish Council of Higher Education (YOK), Bartin University: YÖK 100/2000 PhD Scholarship.

**Data Availability** The datasets used and/or analyzed during the current study are available from the corresponding author on reasonable request.

**Code Availability** Not applicable.

**Declarations**

**Ethics Approval and Consent to Participate** Not applicable.

**Consent for Publication** Not applicable.

**Conflict of Interest** The authors declare no competing interests.

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