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RESEARCH ARTICLE

Identification of proper species that can be used to monitor and decrease airborne Sb pollution

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Abstract
Heavy metal concentrations, which have increased continuously in the environment this century due to anthropogenic factors, severely threaten human and environmental health. Antimony (Sb) is one of the most toxic and harmful heavy metals in terms of human and environmental health. Therefore, the priority research subjects are monitoring the alteration of Sb pollution in the airborne and reducing pollution. This study was conducted to conclude the proper species to monitor and drop airborne Sb contamination on trees grown in Düzce, the 5th most polluted European city. This study examined samples taken from *Pseudotsuga menziesii*, *Cupressus arizonica*, *Pinus pinaster*, *Picea orientalis*, and *Cedrus atlantica*, and the Sb concentration changes based on tree species, route, tissue, and age range in the last 40 years were evaluated. The study hypothesizes that Sb concentration varies depending on (1) tree species, (2) direction, (3) plant tissue, and (4) age range, all confirmed in this study. In conclusion, the maximum concentrations were achieved in the outer bark and east (5.45 μg g^{-1}) and north directions (6.72 μg g^{-1}), with high traffic density. In addition, the mining and industrial places (sources of metal pollution) are not close to the study area. Therefore, it was concluded that traffic pollution was the primary source of Sb pollution in the study area. The study revealed that *C. arizonica* is the most suitable species for monitoring and reducing the change in Sb pollution because the highest Sb concentration (4.47 μg g^{-1}) in wood (the largest organ) was obtained in *C. arizonica*.

Keywords Antimony · Air pollution · Biomonitor · Düzce · Heavy metal

Introduction

Today, heavy metal (HM) concentrations in soil, water, and air are gradually increasing due to anthropogenic factors such as vehicles, solid fuel burning, mining, and industrial activities, especially in urban regions (Ucun Ozel et al. 2020; Cetin et al. 2022). HMs are among the most dangerous environmental pollutants that threaten ecosystems and the health of other living things, particularly humans (Isinkaralar et al. 2022a; Sulhan et al. 2023). Especially when HMs in the air are taken into the human body by respiration, they become much more harmful. HMs are the major perilous components of air contamination, which cause the death of around 7 million people yearly (Ghoma et al. 2022).

Sb, one of the critical and common HMs, is a metalloid and relates to group 15 in the periodic table. It is widely used in materials, drugs, and flame retardants (Lai et al. 2022). Fossil fuel combustion is the major source of anthropogenic Sb atmospheric emissions (Vallero 2014; Dousova et al. 2020; Nishad and Bhaskarapillai 2021).

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It is known that there are high levels of Sb concentration in urban areas. A study determined that the Sb concentration obtained in urban roadside soil was approximately 3.2 times that obtained in forest topsoil (Thestorf and Makki 2021). People in these dense areas are exposed to Sb through the air, air dust, water, and food sources (Voutsa et al. 2002; Belzile et al. 2011). It is thought that exposure to Sb changes the hormone levels in the human body and may increase the risk of cancer (Lai et al. 2022). Chronic exposure to airborne Sb is thought to aggravate irritation of the skin, eyes, and lungs, and prolonged inhalation may aggravate stomach vomiting, diarrhea, stomach ulcers, and pain (Cooper and Harrison 2009). It is stated that Sb causes symptoms such as conjunctivitis, upper respiratory tract chronic bronchitis, inflammation, pleural adhesions, and chronic emphysema (Gad 2014). Due to its effects on human health, Sb is on the ATSDR's (Agency for Toxicity Substance and Disease Registry) priority pollutants list (Savas et al. 2021). It is also among the 13 priority pollutants that the EPA (United States Environmental Protection Category) considers the most dangerous (Hsieh et al. 2004). Therefore, monitoring and reducing the change in the concentration of Sb in the environment, especially in the air, is a priority research topic (Parviainen et al. 2020; Ishimaru et al. 2021; Morton-Bermea et al. 2021; Pleijel et al. 2023).

In this study, the determination of the variation in the Sb concentration in the air in the last 40 years in Düzce, which is the 5th most air-polluted European city, the explanation of the factors that may be effective in this change, and the possibilities of reducing the Sb pollution in the air were investigated. Contrary to receptive environments such as soil and water, it is quite challenging to detect the variation in HM concentrations in the air (Kuzmina et al. 2023; Erdem et al. 2023). Because direct measurement of HM pollution in the air is costly and quite complex, it cannot give an idea about the shift of HM levels in the process and their impacts on the ecosystem (Cesur et al. 2021; Isinkaralar et al. 2022b). Therefore, biomonitors are frequently used to screen the variation in HM levels in the airborne (Key et al. 2022). Annual rings of trees are defined as the most suitable biomonitors to be used for this purpose (Yayla et al. 2022; Cobanoglu et al. 2023). However, the potential to accumulate different HMs in different organs of the species is at different levels (Karacocuk et al. 2022). Therefore, it is essential to define the species of biomonitor that can be used to monitor the change of each HM separately.

Based on the World Air Pollution Report 2021, Düzce city, located in the northwest of Türkiye, between İstanbul and Ankara, is among Europe's five highest air-polluted towns (average PM2.5 was 44.4 μg/m^3) (IQAir 2021). The highest PM2.5 and PM10.0 were measured at 244.83 and 386.31 μg/m^3 in Düzce air (Çevre, Şehircilik ve İklim Değişikliği Bakanlığı 2023). D-100 highway passes through the city, and the TEM highway passes around the city of Düzce. Therefore, this study was carried out to monitor and reduce the level of Sb pollution in the air and to define the suitable plant species that can be used for phytoremediation.

The hypotheses in the study are as follows: (1) Sb concentration varies depending on tree species. (2) Sb concentration changes in different directions of the trees (transfer of Sb in the wood is limited). (3) Sb concentration varies on a plant tissue basis. (4) Sb concentration changes depending on age range (Sb concentration in the air in the study area has changed in the last 40 years).

Material and method

Study area

The current research was conducted in a park in Düzce (Fig. 1), in Türkiye's western Black Sea region (140 m a.s.l.), whose industry is developing day-to-day. The city is shaped like a bowl and surrounded by high mountain ranges on both sides, which reduced the wind speeds (Özdede and Ak 2022) that caused contamination to settle over the urban site and not scatter during the winter. Air pollution is increasing gradually and has recently been a crucial environmental problem for Düzce. Studies indicate that Düzce's air pollution level has increased significantly in recent years. In a study conducted in 2018, although Düzce was the 6th most polluted city in Türkiye, it was determined that it was one of the cities whose air pollution decreased in the 2016–2019 period (Aykaç and Yasin 2021). However, studies conducted in recent years have stated that Düzce is the 2nd most polluted city in Türkiye (5th in European town) and that air pollution continues to increase (Canturk 2023; Isinkaralar et al. 2023; Koc et al. 2024).

Sample collection

In the study, five coniferous species *Cupressus arizonica* Greene (Cpa) [*Cedrus atlantica* (Endl.) Maanetti ex Carriere (Cda), *Pinus pinaster* Aiton. (Pp), *Pseudotsuga menziesii* (Mirb.) Franco (Pm)] and *Picea orientalis* L. (Po) which show good growth and are adapted to the Düzce climate were used. These species have high aesthetic values due to durability, low maintenance cost, and various benefits for landscaping in urban sites. In selecting these trees, it was considered that they were healthy, were about 5 m from each other, were similar in age and height, and their crowns did not touch each other.

After marking the direction on the trunk, cuttings (one single tree from each species) were carried out in the late autumn of 2022. Five thick discs (10 cm) from each tree species were taken from around a half meter above the

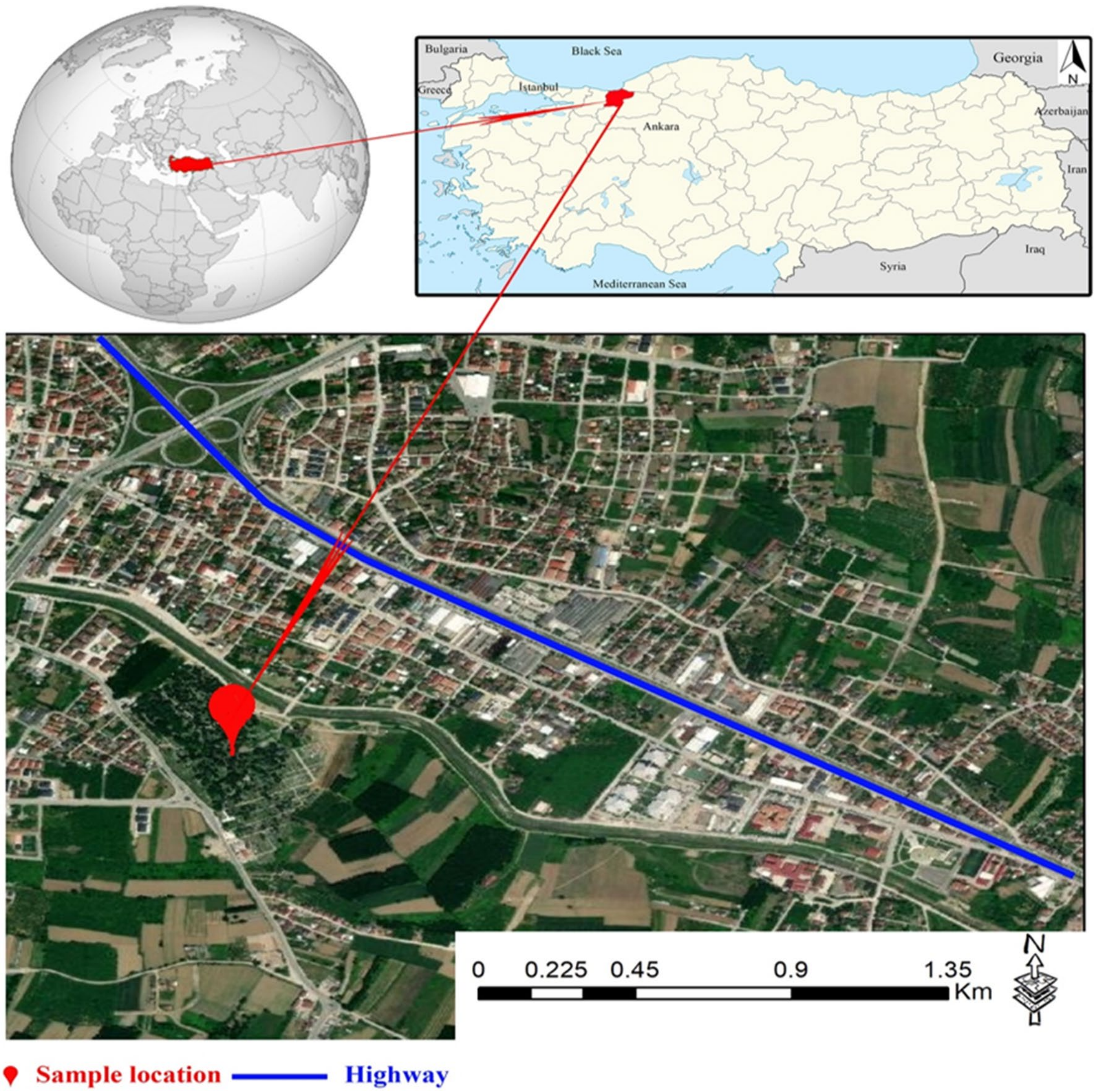

Fig. 1 Location of study site

ground level, and discs were taken to the laboratory for preliminary work. The thickness of wood, outer bark, and inner bark were measured, and the proportion of tissues is provided in Table 1. In order to see the annual rings on the disc samples, the discs were smoothed with the help of a planer. At the end of the check, it was concluded that the species was 40 years old. Due to the narrow annual ring widths, the wood samples were divided into age groups of 5 years.

Performing Sb element analysis

The samples [5 species × 4 route × 5 disc sample × 10 (8 age category + outer bark + inner bark) × 3 repetitions = a total of

Table 1 The ratios of plant parts (%) by plant species

Tissues	*Cedrus atlantica*	*Cupressus arizonica*	*Pinus pinaster*	*Pseudotsuga menziesii*	*Picea orientalis*
Wood	93.1	97.6	91.3	89.3	96.5
Inner bark	3.3	1.1	2.2	4.8	1.2
Outer bark	3.6	1.3	6.5	5.9	2.3

3000 samples] were taken with the help of a steel drill. The samples were kept in the laboratory environment (20–25 °C) for 15 days in glass petri dishes to turn into air-dry. Afterward, the samples were ground first, oven-dried at 45 °C for a week, and prepared for analysis. Six milliliters of nitric acid (65%) and 2 ml of hydrogen peroxide (30%) were added to the samples and allowed to react in the microwave oven (Ethos brand). The microwave device's program was set to rise to 200 °C for 15 min and then remain at 200 °C for 15 min. The prepared samples were burned in a microwave at 280 PSI pressure and 180 °C for 20 min. The samples that became solutions were moved to the flasks and filled with ultrapure H_2O to 50 ml. The prepared solutions were analyzed for Sb metal with an ICP-OES machine (Inductively Coupled Plasma-Optic Emission Spectrometer, GBC Scientific Equipment Pty Ltd., Melbourne, Australia), and the concentration values were obtained. Sb concentration values were calculated by multiplying the obtained values with the dilution factor. The method used is commonly preferred in current reports (Turkyilmaz et al. 2020; Karacocuk et al. 2022; Canturk 2023).

Statistical analysis

The obtained values from 3000 samples were evaluated with the help of the SPSS 22.0 statistical program. Analysis of variance (ANOVA) was primarily applied to the data, and the Duncan test was performed for the factors with statistically significant differences when the alpha was 0.05. Thus, the difference in Sb concentration in the airborne in Düzce province based on tree species, route, tissue, and year group was defined and evaluated independently.

Results

Variation of Sb concentration based on a species

Sb element concentration in annual tree rings of five trees by period and species are presented in Fig. 2. Sb concentration in wood tissues decreased in Cda throughout the experiment period. Sb concentration in Cpa decreased until 2000 and then had an increasing trend, while Sb concentration in the rest of the species increased until 2005 and decreased afterward.

Variation of Sb concentration based on direction

Sb concentration based on species and direction is presented in Fig. 3. While the uppermost concentration was found in Cpa (5.95 μg g^{-1}) and Cda (6.01 μg g^{-1}) in the north route, the maximum value was seen in Pp (6.23 μg g^{-1}) and Cpa (6.15 μg g^{-1}) in the east side. The smallest concentration in the south route was found in Pp, while in the west, it was obtained in Pp, Po, and Cpa. Sb concentration in Cpa, Po, and Pp are ordered as west < south < north < east. According to the averages, the minimum values were determined in Po and Pp species.

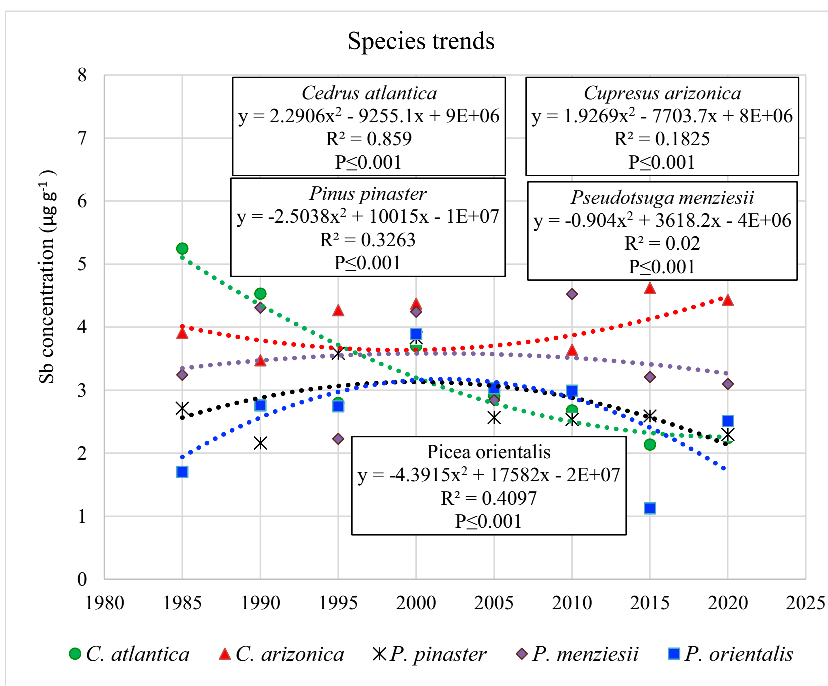

Fig. 2 Sb concentration rate of annual tree rings of five tree species grown in Düzce, Türkiye, throughout 1985–2020 (5-year interval)

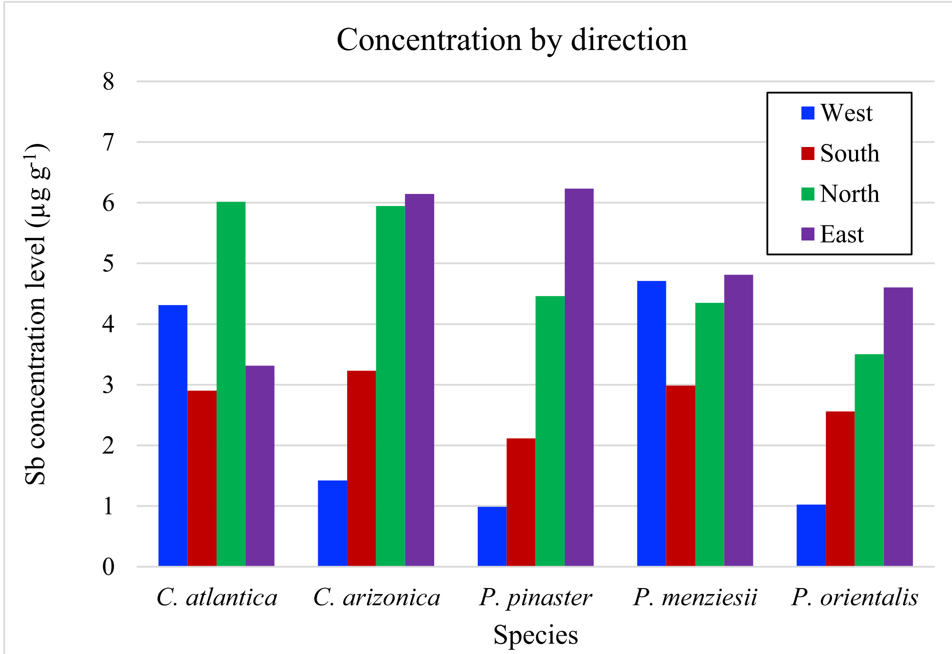

Fig. 3 Sb element concentration in wood tissues of five trees by direction

Variation of Sb concentration based on plant tissue

The alteration of Sb amount by plant species and tissues is shown in Fig. 4. While the maximum Sb concentrations in the inner bark were observed in Po and Pp species, the maximum in wood was in Cpa, Cda, and Pm. According to the averages in organs, the lowest concentrations were mostly seen in Po species.

Variation of Sb concentration based on age period

The trend of Sb element concentration in annual rings by period and route is shown in Fig. 5. The uppermost Sb concentration is mainly seen in the east route, while the smallest values are seen in the south from 1985 to 2005 and then in the west afterward. The Sb concentration decreased in the west direction from 1985 to 2020. The Sb concentration

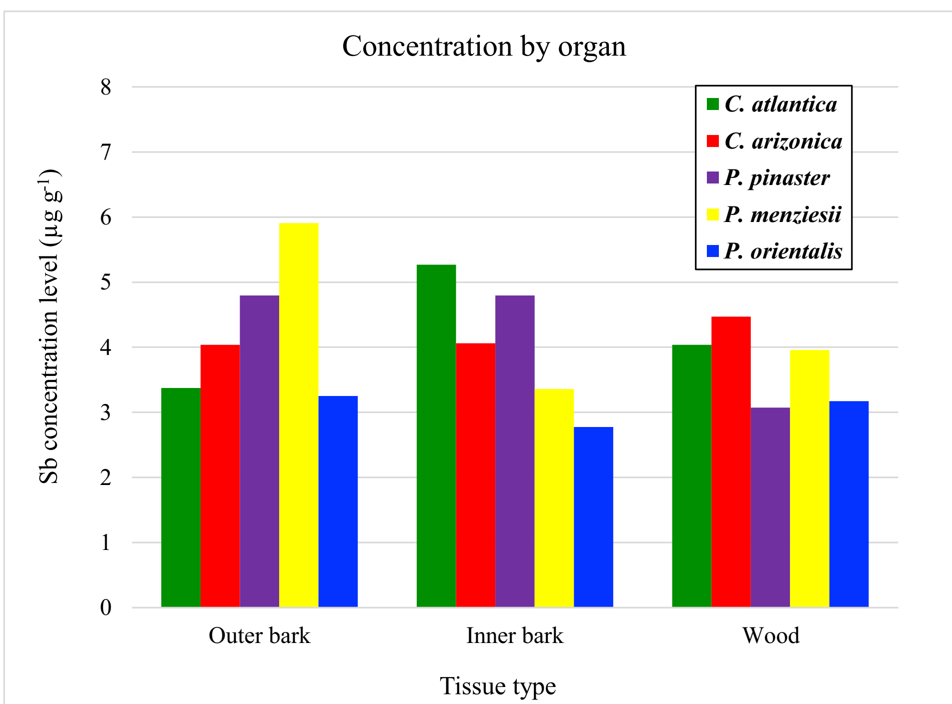

Fig. 4 Alteration of Sb element amount by organs and species, developed in Düzce, Türkiye

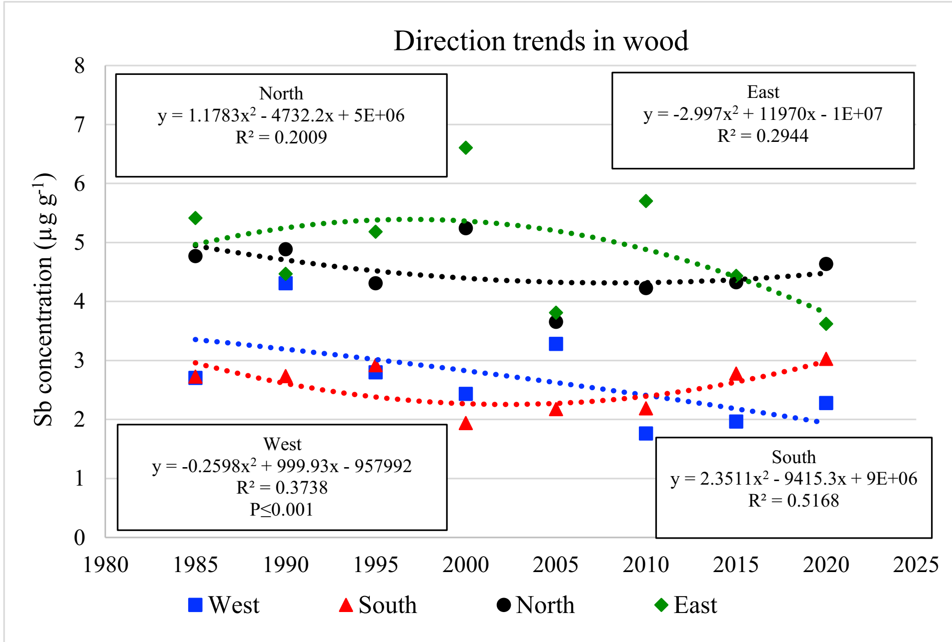

Fig. 5 Sb element concentration of five conifer species wood (tree rings) by direction throughout 1985–2020 (5-year intervals)

decreased from 1985 (2.85 µg g^{-1}) to 2000 (1.94 µg g^{-1}) and then increased until 2020 (3.03 µg g^{-1}) in the south.

Variation of Sb concentration based on species, plant tissue, and direction

The variation of Sb element concentration by organ and direction is presented in Fig. 6. The highest Sb values in organs were observed in the north and east, while the lowest values were obtained in the west and south. The maximum Sb amount in the north direction was observed in the outer and inner bark, while the lowermost values were achieved in the wood. While the lowest Sb amount in the west was found in the outer bark (1.48 µg g^{-1}), the maximum values in the west direction were observed in the inner bark (3.37 µg g^{-1}) and wood (2.61 µg g^{-1}).

Sb concentration was affected by plant species, direction, organ, and their interactions (Table 2). The alteration of Sb concentration in five plant species by direction and organ is shown in Table 3. The Sb concentration change in

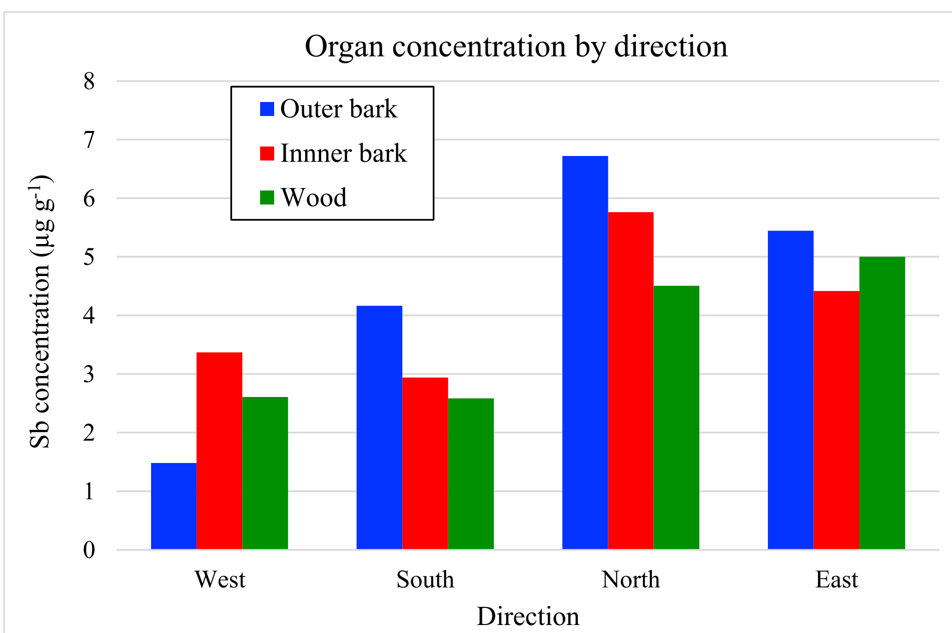

Fig. 6 Sb element concentration in organs of 5 species by direction

Table 2 Summary of ANOVA for Sb concentration for organs of five conifer species under four direction

Source of variation	df	*p*-value
Species (S)	4	0.0001
Organ (O)	2	0.0001
Direction (D)	3	0.0001
S x O	8	0.0001
S x D	12	0.0001
O x D	6	0.0001
S x O x D	20	0.0001

Table 3 Sb element concentration (μg g^{-1}) in organs of 5 plant species grown in Düzce, Türkiye, sampled by direction

Cedrus atlantica				
Plant parts	West	South	North	East
Outer bark	1.26 Aa	2.98 B	6.34 Cab	2.92 B
Inner bark	6.72 Cb	3.54 B	7.53 Db	3.28 A
Wood	4.63 Bb	2.81 A	5.79 Ba	3.37 A
Cupressus arizonica				
Outer bark	2.74 Ac	4.22 B	5.15 C	UL
Inner bark	2.44 Ab	3.03 B	6.72 C	UL
Wood	0.84 Aa	3.13 B	5.95 C	6.15 C
Pinus pinaster				
Outer bark	1.29 Ab	2.74 Bb	6.84 Cb	8.31 Db
Inner bark	1.23 Ab	2.83 Bb	7.43 Cb	7.70 Dab
Wood	0.92 Aa	1.89 Ba	3.79 Ca	5.73 Da
Pseudotsuga menziesii				
Outer bark	1.17 Aa	7.60 Cb	9.74 Db	5.11 B
Inner bark	5.55 b	2.31 a	2.22 a	UL
Wood	5.05 Cb	2.49 Aa	3.94 Ba	4.74 BC
Picea orientalis				
Outer bark	0.96 A	3.27 B	5.53 Cb	UL
Inner bark	0.91 A	3.00 C	4.91 Db	2.28 Ba
Wood	1.12 A	2.42 B	3.07 Ba	4.99 Cb

According to Duncan's test outcomes, numeric values followed by the different letters (A and B or a and b) specify significant differences among organs within each direction. Lowercase letters demonstrate vertical ways, whereas capital letters specify horizontal routes. *UL* under detection limit

Cda was significant ($p < 0.05$) in all organs based on route. Sb concentration change was not significant ($p > 0.05$) in routes other than west and north. The maximum Sb amount in Cda in the north was observed in the bark, whereas the uppermost Sb in the west was achieved in the inner bark (6.72 μg g^{-1}) and wood (4.63 μg g^{-1}). The lowermost Sb values were detected in the south (2.81 μg g^{-1}) and east directions (2.92 μg g^{-1}), while the uppermost ones were achieved in the north (7.53 μg g^{-1}).

The alteration in Sb amount in Cpa was significant ($p < 0.05$) only in the west route. The change in Sb concentration based on the route was substantial ($p < 0.05$) in all organs. The smallest Sb amount in Cpa in the west was seen in the wood (0.84 μg g^{-1}), while the uppermost is in the outer bark (2.74 μg g^{-1}). The smallest values in Cpa among the directions were in the west, while the highest was obtained in the north and east. Sb concentration variation in Cpa in the outer and inner bark in the east route remained below the detectable limits.

The variation in Sb concentration on organ basis in Pp was significant ($p < 0.05$) in all directions, and the change in directional basis was significant ($p < 0.05$) in all organs. The average lowest Sb value was detected in the west route (1.15 μg g^{-1}), while the highest was in the east (7.25 μg g^{-1}). The change of Sb concentration in Pp is west < south < north < east. Notably, the minimum amount in all directions was obtained in wood.

The variation in Sb concentration in Pm is significant ($p < 0.05$) in all tissues based on route and tissues in all routes except the east route. The smallest Sb value in Pm in the south and north directions was detected in the inner bark (2.31 and 2.22 μg g^{-1}) and wood (2.49 and 3.94 μg g^{-1}), while the uppermost was found in the outer bark (7.60 and 9.74 μg g^{-1}), respectively. In the west direction, the lowest Sb amount was detected in the outer bark (1.17 μg g^{-1}), while the highest was observed in the inner bark (5.55 μg g^{-1}) and wood (5.05 μg g^{-1}). Sb amount variation in the inner bark in the east route remained below the detectable limits. In the eastern route, the alteration in Sb amount in the inner bark was at undetectable limits.

The change in Sb amount in Po is substantial ($p < 0.05$) in all organs based on route. In routes other than the east and north, the change of Sb amount based on organs was not significant ($p > 0.05$). The smallest Sb amount in Po in the north direction was observed in the wood (3.07 μg g^{-1}), while the lowermost amount in the east side was detected in the inner bark (4.91 μg g^{-1}). Sb amount variation in the eastern outer bark remained below the detectable ranges. The change of Sb concentration in Po was seen in west < south < north < east.

Discussion

The source of Sb pollution

This research investigated the variation in Sb concentrations across different plant types, directions, organs, and wood in select trees in the downtown zone of Düzce. This study found the maximum Sb element concentrations on the east and north sides. The highway, located northeast of the research site, is thought to have caused this situation because it is stated that traffic is one of the primary HM sources

(Sevik et al. 2019; Cetin et al. 2020). Studies show that the primary sources of heavy metals are anthropogenic sources such as mining activities, industrial areas, urban areas, and traffic (Istanbullu et al. 2023; Sevik et al. 2024). However, since there is no mining area or industrial establishment in the area close to where this study was conducted, traffic comes to the fore as a source of pollution. On the other hand, in a study, although Sb concentrations in *Cupressus macrocarpa* annual rings increased in the polluted area in central Chile, they did not differ statistically significantly compared to the control area (Muñoz et al. 2019). Reports show that anthropogenic sources such as the wear of motor vehicle surfaces or brake pads are a source of Sb (Amereih et al. 2005). In addition, by examining the grass and soil samples, it was determined that Sb accumulation is intense in places close to traffic (Dietl et al. 1997), and a significant part of the Sb in the soil is caused by vehicle emissions (Amereih et al. 2005; Dousova et al. 2020).

Primary sources of Sb in high-traffic regions come mainly from wear of tires (Adachi annd Tainosho 2004), brake pads (Torre et al. 2001; Amereih et al. 2005), tailpipe emissions (Lough et al. 2005), and engine bearings (Belzile et al. 2011). The uppermost concentrations of Sb were achieved in areas where the traffic is mostly passenger cars and on streets that show a "stop-and-go" pattern during rush hour (Gómez et al. 2005). For instance, a research study investigated heavily congested intersections in three distinct districts of the Czech Republic (Central Europe), monitoring the presence of Sb in topsoils, road dust, and reference soils over 2 years (Dousova et al. 2020). The results uncovered a primary source of Sb pollution linked with braking activities in high-traffic zones, where the resulting abrasion dust exhibited an average chemical composition of up to 5%. Sb concentrations varied from 5 to 70 μg g^{-1} in topsoils and from 20 to 350 μg g^{-1} in road dusts. The average Sb concentration was up to 60 folds the background value in that study. High volume and heavy truck fractions in traffic also cause an increase in airborne emissions (Lough et al. 2005). It is stated that 70% of the most critically identified sources in urban areas are diesel exhaust (Iijima et al. 2008). It is stated that vehicle-derived metals (Pb, Cu, Ba, Cd, Sb, and Zn) are primarily caused by abrasion rather than combustion; Sb may also be caused by brake wear (Sternbeck et al. 2002), and Sb is found in particulate matter in brake dust (Adachi and Tainosho 2004).

In this study, the change in Sb concentration on an organ basis was also noteworthy. The highest average Sb amounts were found in the outer bark, especially in the north and east routes, where traffic is heavy. The concentration of Sb in tissues that accumulated within determinable limits varied between 0.96 and 9.74 μg g^{-1} in the outer bark and between 0.91 and 7.53 μg g^{-1} in the inner bark in different species and directions in the current study (Table 3). These results are generally higher than the results obtained in similar studies. In a study conducted in China, it was determined that the Sb concentration in the outer barks of trees varied between 0.07 and 1.49 μg g^{-1}, and the concentrations in the inner barks varied between 0.01 and 0.07 μg g^{-1} (Liu et al. 2023). In a study conducted on eight dominant species in mangrove forests in India, the highest Sb concentration in wood was obtained in *Avicennia officinalis* wood with 12.8 μg g^{-1} (Mandal et al. 2021). A study on waste wood in Finland determined that the Sb concentration varied between 1.20 and 15.0 μg g^{-1}, with an average of 6.46 μg g^{-1} (Bajamundi et al. 2015). A study on *Pinus nigra* wood determined that the Sb concentration varied between 6.33 and 10.00 μg g^{-1} (Sevik et al. 2024).

Elevated Sb concentrations in the outer bark are correlated to particulate matter polluted with Sb due to the bark structure and traffic density. Reports have stated that HMs in the atmosphere adhere to particulate matter and contaminate particulate matter with HMs. HM concentrations in plant organs boost as these particulate substances stick to plant tissues (Turkyilmaz et al. 2019; Dousova et al. 2020). The rough outer bark surface structure facilitates the adhesion of particulate substance here, and the elevated HM concentrations in the bark, especially in the route of traffic jam, are clarified by the contagion of particulate matter in this path with HMs initiated by vehicle (Koç 2021; Cesur et al. 2022; Canturk 2023).

Suitable species for monitoring and mitigating Sb pollution

This study obtained the highest Sb concentrations in Cpa, Cda, and Pm according to the mean values. Generally, there is a significant difference between the Sb concentrations determined in the woods that occurred in different routes in the same period or successive periods in the same direction. However, the species where this difference is most apparent is Cpa. This species also obtains the highest average Sb concentrations in the wood part and all organs. Therefore, Cpa is the proper tree species to monitor the change of Sb concentration in the airborne and reduce Sb pollution.

Report on the speciation of HMs in the tree and their movement between tree parts, starting from entering the plant body, is limited (Savas et al. 2021). It has been stated that the allocation of some elements in wood may alter from species to species. For example, in *Cedrus deodara* woods, Zn and Pb are replaced, but the Cu element remains constant (Zhang 2019). In *Corylus colurna* woods, the displacement of Ni, Pb, Zn, and Cr elements is limited, but Cd can be replaced (Key and Kulaç 2022; Key et al. 2022). In *Cedrus atlantica* woods, the transfer of Ni element is quite limited, but this is not valid for Co (Koç 2021). Therefore, defining the appropriate species separately for each HM pollution monitoring is essential.

Conclusion

Sb contamination in the airborne is high in the study site. It was determined how the Sb concentrations changed based on direction, and generally, the uppermost concentrations were found in the east and north routes. The north and east sides are the direction with a main highway with high traffic density, and therefore, the accumulation of Sb in the trees is caused by traffic.

It was also revealed that the Sb concentrations differed considerably in diverse routes in the consecutive year intervals and same year range in all trees subject to the research. However, the species in which this difference is most apparent is Cpa. This species also obtains the highest average Sb concentrations in the wood part and all organs. HMs taken into the wood are trapped here and removed from the ecosystem. Therefore, plants with high HM accumulation potential in the wood portion can effectively decrease HM contamination in the air. Therefore, Cpa is a suitable tree to use to monitor the alteration of Sb concentration in the airborne and reduce Sb pollution. Using this species in afforestation and landscaping practices in urban zones will reduce Sb concentration and thus prevent Sb-related diseases and disorders in humans.

Author contribution All authors contributed to the study conception and design. Material preparation, data collection, and analysis were performed by U.C. and İ.K. The first draft of the manuscript was written by U.C. and İ.K., and all authors commented on previous versions of the manuscript. The final draft of the manuscript was edited by İ.K., H.B.O., and H.S. All authors read and approved the final manuscript.

Data Availability The data that support the findings of this study are available from the corresponding author, upon reasonable request.

Declarations

Ethical approval Not applicable.

Consent to participate Not applicable.

Consent for publication Not applicable.

Competing interests The authors declare no competing interests.

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