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RESEARCH ARTICLE

The possibility of using Scots pine needles as biomonitor in determination of heavy metal accumulation

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Abstract
One of the biggest problems of modern world is the air pollution which causes the death of millions of people every year. The heavy metals included in the component of air pollution occupy an important position in human health since they could remain intact in nature for a long time and build bioaccumulation, and also some of them are toxic or carcinogenic even at some low concentrations. Even the heavy metals functioning as micronutrient element could create toxic effect for human beings at the high concentrations. For this reason, the determination of the heavy metal concentration has an important role in terms of the detection of risky regions and risky level. One of the primary sources of heavy metals is industrial plants where the heavy metal ores are processed. Those regions might have risks at high level in terms of particular heavy metals. Consequently, it is significant to find out in which level they influence the area around these plants and to what extent the heavy metal pollution is effective. In this study, Ba, Zn, Cd, K, and Na concentrations are determined by analyzing the samples which are taken from 1- and 2-year-old needles of Scots pines (*Pinus sylvestris* L.) growing in 1 km, 3 km, 10 km and 25 km distances around a processing and mining of magnesite ore in Russia. In the end of the study, it is concluded that generally, the concentrations of heavy metals subject to the study are increasing depending upon the distance, and this increase is quite apparent in some elements, and finally in many points, the concentrations determined in 2-year-old needles have higher levels than 1-year-old needles.

Keywords Biomonitor · Heavy metal · *Pinus sylvestris* · Scots pine

Introduction

Together with the increase in the world population, industrial activities have increase in the same level, and air pollution increased in parallel with these activities has reached to dangerous extent (Adiguzel et al. 2020; Cetin 2019; Turkyilmaz et al., 2018a, b). The increased air pollution causes the destruction of the nature; the pollution in the air, water, and soil; and degradation of ecological balance (Mutlu et al. 2016; Ozel et al. 2019; Bayraktar 2019; Mutlu and Kurnaz 2017; Bayraktar et al. 2019; Bozdogan Sert et al. 2019; Adiguzel et al. 2020; Cetin 2019; Yucedag and Kaya 2017). Air pollution affects the human health significantly, such that air pollution today has become a global problem causing the death of millions of people every year (Cetin et al. 2019a; Sevik et al. 2019a, b; Aricak et al. 2019; Kaya 2009; Yucedag and Kaya 2016; Kaya et al. 2019).

Even though air pollution has many components, the heavy metals as one of those components have a particular importance because they tend to be bioaccumulation, and they are toxic for human health even in the low concentrations (Aricak et al. 2020; Sevik et al. 2019c, d). In addition to them, heavy metals can remain intact and not decompose in nature for a long time. The metals like Hg, Cd, As, and Pb have critical toxic effects in organism even at the low levels (Shahid et al. 2017; Turkyilmaz et al., 2018a, b). Although micronutrients such as Mn, Zn, Cr, Cu, Fe, and Ni are necessary for the living organisms including the plants, those also could create

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detrimental effects at high concentrations. The conducted studies show that when almost the whole metals are taken above the specific amount, they can create toxic effect (Niazi et al. 2011; Harguinteguy et al. 2016).

The industrial activities are shown as the most important responsible for air pollution (Shahid et al., 2017). Despite the fact that mineral resources are highly important for socioeconomic development, the mineral extraction and its usage in different industrial treatments play a leading role in the increasing of environmental pollution and especially air pollution (Niazi and Burton 2016; Turkyilmaz et al. 2018c,d). Therefore, the determination of the environmental damage caused by industrial facilities shown as the most important source for heavy metal pollution is of capital importance for all living organism, especially humans and ecosystem.

The change of heavy metal pollution can be determined by direct or indirect methods. The usage of bioindicators within these methods is the most preferred and leading method because of the fact that it is cheap and simple, and also it provides more reliable data about periodical change of heavy metal concentration and gives an idea about the effect of heavy metal pollution on ecosystem (Sevik et al. 2020a,b,c; Aricak et al. 2020). The plants growing where the pollution is intense indicate the progress of increase in heavy metal concentration in the air in the course of time by accumulating the heavy metals in their stem, boughs, and needles (Turkyilmaz et al. 2018a). Accordingly, it is much more common method to use bioindicator as the pollution indicator instead of determination of directly heavy metal pollution transiently (Cetin et al. 2019b; Turkyilmaz et al. 2018e).

In this study, it is aimed to determine the distance and age-related variations of some heavy metal concentrations in 1- and 2-year-old needles of Scots pines growing in 1 km, 3 km, 10 km, and 25 km distances around a processing and mining of magnesite ore in Russia.

Materials and methods

In the conditions of modern cities, aerotechnogenic pollution is a permanent environmental factor that has a negative impact on the environment and human health. One of the objects of large foci of destruction of forest vegetation is located in the Southern Ural, Chelyabinsk City, Satka, Magnesite Combine (Fig. 1a, b). Studies were conducted in the area of influence of the plant in the experimental plots (ES) established in 1980–1988 by the Ural Experimental Station.

In the pollution gradient to the northeast, in the direction of the prevailing winds from the emission source, the following areas are located: ES-2 at 1 km from the plant in the zone of strong influence, ES-5 at 3 km - zone of average influence and ES-4 at 10 km - zone of weak influence at the Magnesit and near the village of Sibirka is located Control at 25 km from the emission source, background conditions (Fig. 2).

The raw material for the production of sintered periclase powders is raw magnesite. High-carbonate feedstock is burned in such a way that raw magnesite of Satkinsky groups of deposits (fractions 0–40 mm) is loaded into a rotary kiln. After burning natural magnesite, a large amount of caustic dust is formed which undergoes repeated burning. When burning dust, a large amount of flue gases, carbon monoxide, alkali, fluorine and sulfuric anhydride, nitrogen oxides are released into the atmosphere. Near the plant and within a radius of 1.5–2 km, the forest completely died (Fig. 1b). The maximum volumes of magnesite dust into the atmosphere in 1963 reached 182.5–328.5 thousand tons per day. In 1978, new electrostatic precipitators were installed at the plant, and dust emissions dropped to 70–90 tons per day.

The relevance of biochemical research is currently associated with the need to understand the ecological situation in general and is an integral part of the study of natural and man-made modified terrestrial ecosystems. To assess the impact of the source of pollution, plant organisms, including woody ones, are often used. Scotch pine (*Pinus sylvéstris* L.) has proved itself as a promising species indicator of aerotechnogenic pollution (Mokhnachev et al. 2015; Kazantsev 2005).

The samples which were brought to the laboratory after being collected and labeled were subjected to the separation process by laying out on carton. The needles from different distances and ages were placed in glass petri dishes after being separated and labeled. The samples prepared in this way were kept for 15 days to become room dry, and within this period, the laboratory was ventilated on a daily basis. The air-dried samples were dried in the oven at 45 °C for 1 week to dry completely. In the next phase, the hand samples were crushed into powder by grinding and 0.5 grams were put into the tubes designed for microwaves by weighed. After adding 10 mL 65% HNO_3 over the samples, it was burned in the microwave at the pressure of 280 PSI and at the 180 °C degree for 20 min. By adding deionized water over the samples, 50 ml was completed, which were taken from microwave and cooled. The prepared samples were read at proper wavelength in the ICP-OES device after being filtered from filter paper.

The obtained data was assessed by means of SPSS package program; the variance analysis was applied to the data, and homogeneous groups were gained by applying Duncan test to the values with the statistical differences of at least 95% confidence level. The obtained data were interpreted by simplifying and tabulating.

Results

The change of Ba element was determined depending upon the age of needles and distance; and the groupings formed as a

Fig. 1 The location for Satkinsky, Magnesite Combine (**a**) and the surrounding forests (**b**)

result of Duncan test, F value obtained in consequence of variance analysis, and the error rate were given in Table 1.

When examining the table, it is seen that the changes in the concentration of Ba element based on the distance are statistically significant ($p < 0.01$ in the 1-year-old needles and $p < 0.001$ in the 2-year-old needles). When examining the average values and the results of Duncan test, it could be seen that the concentration of Ba increases prominently in 1-year-old hand and decreases generally in 2-year-old needles together with the distance. Moreover, it can be seen that in terms of the age of needles, the changes in the distances of 1 km, 3 km, and 10 km are statistically significant (at least $p < 0.01$), and also the values found in 2-year-old needles in the close range (1 km and 3 km) are much higher than the values specified in 1-year-old needles. The difference between 1- and 2-year-old needles at the distance of 1 km is higher than 10 times.

The change of Zn element was determined depending upon the age of pointer and distance; and the groupings formed as a result of Duncan test, F value obtained in consequence of variance analysis, and the error rate were given in Table 2.

It is also seen that the changes in the concentration of Zn element based on the distance at both 1- and 2-year-old needles are statistically significant ($p < 0.001$). Furthermore, when examining the groupings formed by Duncan test and the average values, it might be said that there is a linear relationship between the distance and Zn concentration and also Zn

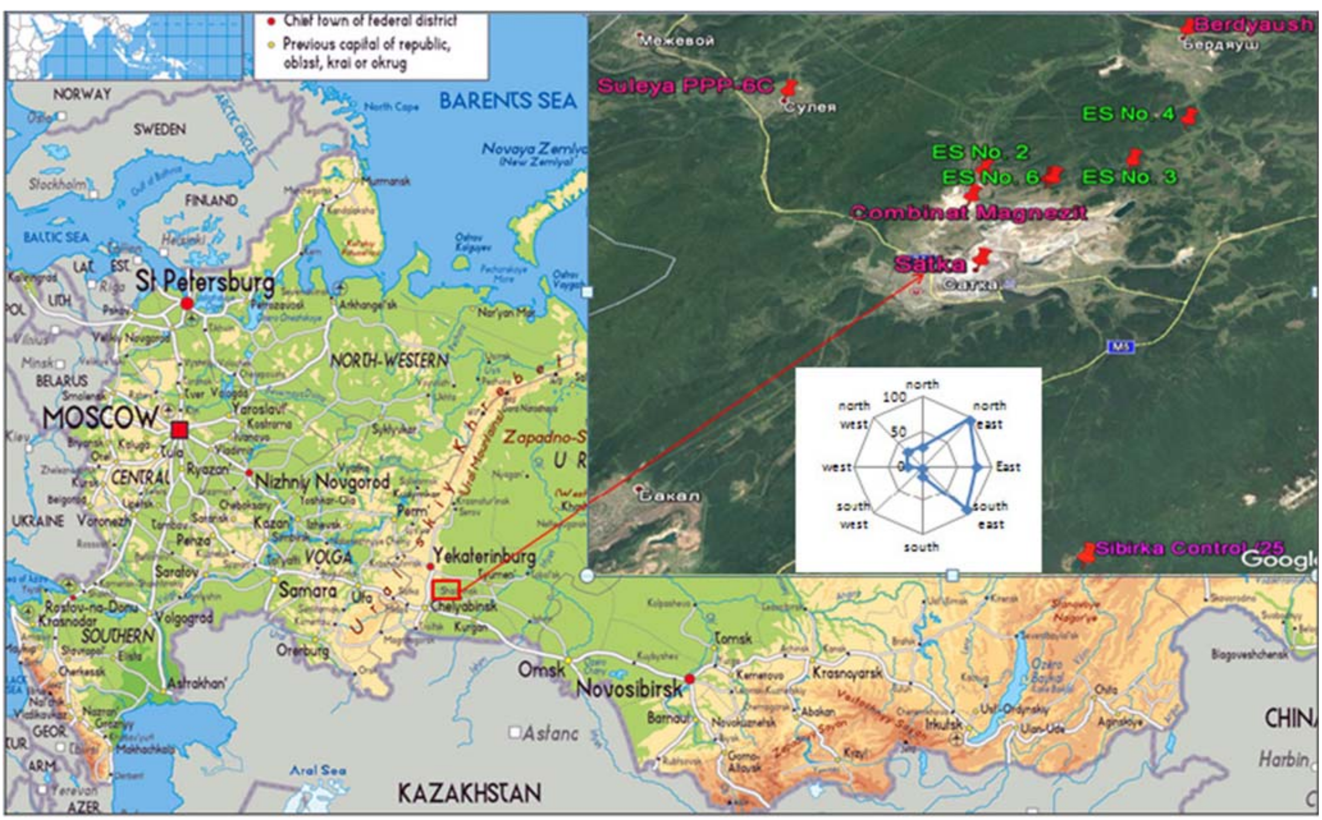

Fig. 2 Location of experimental sites depending on the Magnesite Combine

Table 1 The change of Ba (ppb) element based on the distance and age of needles

Distance	1-year needles	2-year needles	F value	Sig.
1 km	190.40 Aa	2184.13 Bc	64.989	0.000
3 km	524.11 Aab	3125.33 Bd	42.496	0.000
10 km	847.48 Bb	203.51 Aa	17.929	0.001
25 km	864.48 b	1112.0 b	1033	0.324
F value	6195	27.233		
Sig.	0.002	0.000		

Table 3 The change of Se (ppb) element based on the distance and the age of needles

Distance	1-year needles	2-year needles	F value	Sig.
1 km	155.11 a	241.51 a	2766	0.116
3 km	640.31 Ab	1490.26 Bb	9839	0.006
10 km	993.91 Ab	1360.26 Bb	7188	0.016
25 km	1537.73 c	1490.53 b	0.090	0.768
F value	17.274	35.916		
Sig.	0.000	0.000		

concentration increases with increasing distance. As a result of the variance analysis, it is determined that there is statistically significant difference ($p > 0.05$) only between the concentration of Zn in the needles from different ages at the close distances (1 km and 3 km). At these distances, the concentrations determined in the 2-year-old needles are higher than the concentrations detected in 1-year-old needles.

The change of Se element was determined depending upon the age of hand and distance; and also, the groupings formed by Duncan test, F value obtained in consequence of variance analysis, and the error rate were given in Table 3.

According to the results of the variance analysis, it could be seen that the change in the concentration of Se element based on the distance is statistically meaningful ($p < 0.001$). According to the average values and the results of Duncan test, it is observed that the concentration of Se increased together with the distance generally, and this increase is much more clear in 1-year-old needles. As a result of variance analysis, it is found that only the changes between the distances of 3 km and 10 km are statistically significant in regard to the age of hand, and at these distances, the values determined in 2-year-old needles are much higher than the values in 1-year-old needles.

The change of Cd element was determined depending upon the age of hand and distance; and also, the groupings formed by Duncan test, F value obtained as a result of variance analysis, and the error rate were given in Table 4.

It is stated that the changes in the concentration of Cd element depending on the distance are also statistically in significant level ($p < 0.001$) at the 1- and 2-year-old needles. When the average values and the results of Duncan test were evaluated, it can be seen that there is a clear-cut increase together with the distance in general. In consequence of the variance analysis, it is found that the changes at all distances except 10 km are statistically in meaningful level (at least $p < 0.05$) in terms of the age of hand; and also, at all distances, the values determined in 2-year-old needles are higher than the values in 1-year-old needles.

The change in K element was determined based on the age of hand and the distance; and also, the groupings formed by Duncan test, F value obtained as a result of variance analysis, and the error rate were given in Table 5.

Based on the results of the variance analysis, the changes of K element depending on the distance are also statistically in significant level ($p < 0,001$) at both 1- and 2-year-old needles. Moreover, when the average values and Duncan test results are examined, it is found that the concentration of K generally increases with the distance except 3 km. According to the results of the variance analysis, it is determined that the changes at all distances in terms of the age of hand are statistically significant, and at the all distances, the values found in 1-year-old needles are higher than the values in 2-year-old needles.

The change of Na element was determined based on the age of hand and the distance; and also, the groupings formed by Duncan test, F value obtained as a result of variance analysis, and the error rate were given in Table 6.

According to the results of the variance analysis, it is stated that the changes of Na element based on the distance is

Table 2 The change of Zn (ppm) element based on the distance and age of needles

Distance	1-year needles	2-year needles	F value	Sig.
1 km	22.57 Aa	25.83 Ba	5001	0.040
3 km	26.97 Aa	36.82 Bb	6301	0.023
10 km	39.82 Ab	59.82 Bc	130.044	0.000
25 km	53.91 c	57.16 c	0.497	0.491
F value	34.367	57.305		
Sig.	0.000	0.000		

Table 4 The change of Cd (ppb) element based on the distance and the age of needles

Distance	1-year needles	2-year needles	F value	Sig.
1 km	149.06 Aa	168.37 Ba	7056	0.017
3 km	195.11 Ab	318.75 Bc	19.906	0.000
10 km	282.48 c	295.73 b	3040	0.100
25 km	328.26 Ad	354.80 Bd	11.682	0.004
F value	35.090	143.189		
Sig.	0.000	0.000		

Table 5 The change of K (ppm) element based on the distance and the age of needles

Distance	1-year needles	2-year needles	F value	Sig.
1 km	4592.00 Ba	2770.31 Aa	112.673	0.000
3 km	5723.40 Bc	4194.26 Ac	53.089	0.000
10 km	4844.88 Ba	3527.20 Ab	56.318	0.000
25 km	5177.33 Bb	3606.17 Ab	54.229	0.000
F value	20.768	13.151		
Sig.	0.000	0.000		

statistically in significant level ($p < 0.001$) only at 2-year-old needles. Further, when examining the average values and Duncan test results, it is seen that the concentration of Na increases together with the distance in general. In the results of the variance analysis, it is determined that only the changes at 10-km distance in terms of the age of hand is statistically significant, and also the values found in 2-year-old needles are higher than the values in 1-year-old needles.

Discussion

Within the scope of the study, the concentrations of Ba, Zn, Se, Cd, K, and Na elements in the 1- and 2-year-old needles of the Scots grown around 1 km, 3 km, 10 km, and 25 km distances around the "process and mining of magnesite ore" were determined. Even though these elements have been the subject of many studies about the heavy metals until today, it is seen that the elements of Zn and Cd have become generally the more common subject of the studies because of their potential hazards (Turkyilmaz et al. 2018e). Along with these elements, the elements which are examined more commonly in the studies about the heavy metals are the elements like Ni, Pb, Cd, and Co (Sevik et al. 2019e). The main reason of why those elements come into prominence in the studies is because they can cause the toxic effect even at the low concentrations (Aricak et al. 2020).

On the other hand, the conducted studies show that even the heavy metals, which are the nutrient elements, can be quite dangerous for human health at the high concentrations (Shahid et al. 2017; Sevik et al. 2020b). For instance, Fe and Al, which are nutrient elements, are defined as carcinogenic in human beings (Göney 2018). Additionally, it is stated that the elements necessary as nutrient element can cause serious health hazards in case taken orally or by inhalation (Batır 2019). For this reason, monitoring the concentration of those elements in the air is extremely important.

Table 6 The change of Na (ppm) element based on the distance and the age of needles

Distance	1-year needles	2-year needles	F value	Sig.
1 km	138.28 a	135.20 a	0.112	0.743
3 km	154.48 ab	194.17 b	1799	0.199
10 km	179.20 Ab	205.64 Bb	5045	0.039
25 km	183.06 b	195.60 b	1552	0.231
F value	2579	8129		
Sig.	0.071	0.000		

The results of the study show that the concentrations of the elements increase generally depending upon the distance. However, a great number of studies conducted up to today indicate that heavy metal concentrations decrease with becoming distant from their source. Besides, there are also various related studies showing no clear relations between heavy metal concentration and the distance to pollution source (Mossi 2018; Saleh 2018).

It is thought that there are a few explanations for this situation. The first of them is that the pollution source subject to the study is not the pollutions' source in terms of the elements in question. The second possibility is that since the heavy metals subject to the study are also the nutrient elements, the reason behind why the concentration is low in the individuals who are close to the source of heavy metal pollution is not scarcity of the heavy metal concentration but the deficit of nutrient element. In short, the study results can be interpreted as the lack of nutrient in the individuals who are close to the pollution source. For example, the highest Na concentration is determined as 205.64 ppm in the study. In the similar study, while the highest value of K concentration is determined as 5177 ppm, Cetin et al. (2019a) state that K concentration varies between 5566 and 15.303 ppm in the leaf of *Acer platanoides*. In conclusion, the study results might be interpreted as the decrease in the concentrations of some nutrients near the pollution source in plants subject to the study.

Further, it is also known that the changes of heavy metal concentrations in the plants are a complex and not yet fully solved mechanism shaped under the influence of many factors (Sevik et al. 2020a,b,c; Turkyilmaz et al., 2018e). On the other hand, the conducted studies indicate that the accumulation of heavy metal concentration in the plants shapes under the influence of many factors and mutual interaction of those factors (Mossi 2018; Turkyilmaz et al. 2018b,c). The potential for heavy metal accumulation of the plants grown in the same environment changes depending upon the factors like the structure of the organelle, physicochemical properties of the metals, morphology and surface areas of the organelle, surface texture and size of the organelle, habitus of the plant, exposure time to the heavy metal, and the amount of particulate matter along with type and organ of the plant (Xu et al. 2016; Shahid et al. 2017). In addition to them, environmental conditions, especially air humidity and raining, also affect the heavy metal entry into the plant significantly (Shahid et al. 2017; Özel 2019).

In the end of the study, it is concluded that the concentrations of the elements subject to the study in the 2-year-old needles are higher than the concentration in the 1-year-old needles to a large extent. This situation can be explained by the longer exposure of the 2-year-old needles to the same environmental conditions and so to heavy metals (Shahid et al. 2017). In fact, the similar results were obtained in the studies about the needles from different ages on the same plant (Turkyilmaz et al. 2018c; Cobanoğlu 2019; Keçeci 2019).

Although so many studies were carried out about the usage of plants as biomonitor to observe heavy metal pollution in the air, it is still undetermined which plants are more proper for monitoring the concentration of heavy metals. In the conducted studies, 8–10 plants at most could be contrasted, and therefore, only the plants subject to the study can be compared with each other. On the other hand, the conducted studies assert that there is a difference in high level among the types grown in the same environment (Mossi 2018; Sevik et al. 2019b; Saleh 2018).

Heavy metals can enter into the body of the plant through roof or leaf; however, it is difficult to distinguish whether heavy metals in the plant's internal tissues are taken from the soil or atmosphere since the two intake ways could work simultaneously (Pourrut et al. 2013; Shahid et al. 2017). Thus, it is thought that the most appropriate way to monitor heavy metal pollution in the air is through the leaves. The reasons of that are because the leaves are exposed to heavy metal pollution in the air the most and also they are the most affected organs from heavy metal pollution in the air because of the air intake through their stomas during photosynthesis (Cetin et al. 2019b; Shahid et al. 2017; Saleh 2018).

On the other hand, there are so many factors which affect the intake and accumulation of heavy metals into the plant. Some of them are plant type, the amount of raining and humidity, habitus of the plant, structure of the organelle, the type of heavy metal, and its interaction with the plant (Sevik et al. 2019a; Aricak et al. 2020). In addition to those factors, there are also many other factors which might influence heavy metal concentration either directly or indirectly. To illustrate, the change of heavy metal concentration based on the plant type was shown in many studies (Saleh 2018). However, it might be expected that heavy metal concentrations should be in different levels in subtype of the plant, its form, variety, and origins, also. Yet, the conducted studies present that phenological, morphological, and anatomic structures change depending on those features (Ozkazanc et al. 2019; Cetin et al. 2018a, b; Ertugrul et al. 2019). In this case, change in the plant metabolites is also likely possible, and this situation might affect heavy metal absorption (Shahid et al. 2017; Sevik et al. 2020b).

Heavy metal absorption in the plants is also closely related with the plant metabolites (Shahid et al. 2017). Therefore, it is greatly possible that many factors like stress level of the plant which affects the plant metabolites dramatically (Sevik and Cetin 2015; Yigit et al. 2019), origin of the plant (Sevik and Topacoglu 2015), the subspecies (Yucedag et al. 2019), genetic structure of the plant (Hrivnák et al., 2017; Yigit et al. 2016a, b), and hormone applications might affect heavy metal absorption in the plant and so heavy metal concentration. On the other hand, the factors influencing the accumulation of heavy metal concentration in the plants and their impact level have not been determined completely until today. Consequently, studies on this issue should be continued by diversifying and increasing.

Conclusions

In this study, it was tried to determine the change of some heavy metal concentrations in the Scotch pine (*Pinus sylvestris*) whose needles could remain on the tree for a few years, depending upon age of the needles and its distance to the point considered as the pollution source. The study results show in general that many elements subject to the study increase based on the distance significantly. The probable reasons of this conclusion were explained in the study; however, it is suggested that detail researches should be conducted in this topic.

One of the most important results of the study is that the concentrations obtained from 2-year-old needles are quite higher in most of the elements subjected to the study than the concentrations in 1-year-old needles. This situation could be explained as the old needles are exposed to the heavy metals in the air longer than the young needles. According to this result, the needles of the trees like *Abies* and *Picea* whose the age of needles could be calculated exactly and whose needles could remain on the tree up to 8–10 years might be a good biomonitors to monitor the recent heavy metal pollution.

The change in heavy metal concentration is a vital subject especially for human health and biomonitors can be used very effectively to monitor this change in this subject. There is no sufficient information about the mechanism which is effective in the intake of heavy metals to the plants and the factors which are influential in this process. Therefore, it is suggested that studies on this issue should be continued by diversifying and increasing.

The heavy metals subject to this study are generally defined as elements which are the building blocks for plants and animals. However, the conducted studies indicate that the concentrations of those elements in the air are increasing. On the other hand, besides their concentrations in the air, there are no sufficient studies which are about the probable harms of inhalation exposure to these elements for human being and other livings. Studies on this issue should be continued by diversifying and increasing.

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Compliance with ethical standards

Conflict of interest The authors declare that they have no conflict of interest. None of the authors have any competing interests in the manuscript.

Human and animal rights and informed consent This article does not contain any studies with human participants or animals performed by any of the authors. Informed consent was obtained from all individual participants in the study.

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