



The use of perennial needles as biomonitors for recently accumulated heavy metals

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Abstract

The measurement of heavy metal concentrations in plants is important both for determining their ability to remove these pollutants from the air to increase its quality, and for the monitoring of air quality. Previous studies have mostly focused on the use of annual leaves of broadleaved species as biomonitors. In this study, the aim was to determine the heavy metal accumulation of the perennial needles of some conifers. In almost all the species studied, concentrations of heavy metals were found to increase with the age of the tree, but this increase was not linear. There were significant differences between the species in terms of heavy metal accumulation. The highest concentrations were for Fe in *Pinus nigra*, Zn in *Picea pungens*, Pb in *Pinus sylvestris*, and for all the other heavy metals in *Abies bornmülleriana*. This study indicates that *A. bornmülleriana* is particularly useful as a biomonitor for many heavy metals, and also has the potential to remove heavy metals from the air.

Keywords Conifer · Air quality · Pollutant · Fossil fuel · Emissions

Introduction

With the rapid growth of the world population, industrial activities are increasing. Increasing air pollution, especially due to the use of fossil fuels, has reached dangerous levels for the environment (Sevik et al. 2015; Cetin 2016a, b, c; Cetin and Sevik 2016a, b; Cetin et al. 2017a, b), and plants that are most exposed to air pollution are the best indicators of this. They show the changes in concentrations of heavy metals emitted by burning fossil fuels, especially in areas where traffic is intense, with their accumulation of these pollutants in trunks and leaves (broadleaves and needles).

Heavy metal accumulation in plants is mainly seen in the roots, but some plants accumulate heavy metals in their leaves. Trees are not necessarily better indicators of heavy metals than mushrooms, algae and lichen, but as they are planted in cities and may live longer than other bioindicators,

they can give more information on changes in environmental pollution (Sawidis et al. 2011).

Heavy metals are particularly important components of air pollution as they do not decompose or disappear, and also tend to bioaccumulate. Therefore, the monitoring of heavy metal concentrations is of great importance for the identification of areas and levels of risk of air pollution. There are many methods for the determination of air pollutants. Direct measurement requires expensive measuring devices and carries more risk of contamination than the use of bioindicators. Bioindicators are both cheap and allow for the easy collection of samples, and their heavy metal concentrations are higher than those in air and rainwater (El-Hasan et al. 2002).

Plants are frequently used for the biomonitoring of heavy metal concentrations. However, for biomonitoring, heavy metals are measured which accumulate in the leaves during the vegetative period of the year in which the specimens are collected. However, heavy metal concentrations accumulated on a yearly basis are also extremely important. Few recent studies have determined concentrations of heavy metals accumulated in tree trunks, and most of these took samples from the annual rings, which usually involves cutting trees (Cocozza et al. 2016; Panyushkina et al. 2016), thus this is not a suitable method for sustainable monitoring.

Studies on the use of plants for the biomonitoring of heavy metal concentrations have mostly focused on

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broadleaved, evergreen ones. However, in many species with needle leaves, the needles remain on the trees for several years. One of the advantages of using species with needles is their ability to accumulate traffic-induced heavy metals that cause air pollution throughout the year, and thus are good indicators for year-round pollution. They also have significant potential for the determination of recent heavy metal accumulation.

In this study, we aimed to determine the concentration of heavy metals in needles of different ages in species for which needles remain on the tree for 3 or more years, to examine the potential of using these species for the determination of the recent accumulation of heavy metals. Our study was carried out in Kastamonu, the city with the largest forested area in Turkey and lowest industrial activity in the region. Thus the vast majority of data we obtained here reflect only heavy metal pollution from traffic.

Materials and methods

Plant materials were collected from the city centre of Kastamonu. The samples were taken by cutting side branches of *Pinus nigra* (Pn), *Pinus sylvestris* (Ps), *Abies bornmülleriana* (Ab) and *Picea pungens* (Pp)—species frequently used in landscape studies—and brought to the laboratory. The branches were cut and classified in the laboratory on the basis of age, and the needles were collected from the different age groups.

Needle samples were air-dried at room temperature, then dried at 50 °C in a drying oven for 1 week. Two grams of

dried sample was weighed and stored in 10 ml of concentrated HNO₃ at room temperature for 1 day, then boiled at 180 °C for 1 h. Then 20 ml of distilled water was added to the solution, and the solution filtered through 45-µm filter paper. The Fe, Co, Ni, Zn, Cd, Hg and Pb analyses were performed for the filtrates using a GBC Integra XL-SDS-270 inductively coupled plasma atomic emission spectroscopy device. The results are reported as means.

Results

Heavy metal levels (parts per billion; p.p.b.) depended on the ages of the needles (Table 1; Fig. 1).

The concentrations of the heavy metals generally increased with age, but this increase was not linear (Table 1). For example, Co was 13887 p.p.b. in 1-year-old Ps needles, 4222 p.p.b. in 2-year-old Ps needles, and 4741 p.p.b. in 3-year-old Ps needles; almost all of the data showed the same pattern. Moreover, the concentrations of some of the heavy metals in different aged needles were very similar. For example, Zn, which was 1368 p.p.b. in 1-year-old aged needles, increased to 1403 p.p.b. in 2 and 3-year-old aged needles. However, the increase in some metals in some of the species was almost linear. For example, the concentration of Zn in Ab was 3735 p.p.b. in 1-year-old needles, 4205 p.p.b. in 2-year-old needles and 7220 p.p.b. in 3 year-old needles. Similarly, the concentration of Zn in Pp was 1191 p.p.b. in 1-year-old needles, 4430 p.p.b. in 2-year-old needles, and 6257 p.p.b. in 3-year-old needles. In order to better interpret the different concentrations of the heavy metals between

Table 1 Change in the amount of heavy metals depending on tree species and age (p.p.b.)

Species ^a	Fe	Co	Ni	Cu	Zn	Cd	Hg	Pb
1 Ps	4114	3887	179.5	345.3	838.2	2580	39.51	67.29
2 Ps	5388	4222	231.5	379.8	1176	2858	46.83	271.3
3 Ps	5343	4741	265	522.9	1706	3820	56.59	205.3
Ps average	4948.3	4283.3	225.3	416	1240.1	3086	47.6	181.3
1 Pn	4361	25.35	262.3	465.1	922.4	2912	42.44	57.47
2 Pn	7264	26.34	297.9	559.2	1002	3125	59.51	73.37
3 Pn	8009	28.94	377	587.9	1244	3243	121	84.67
Pn average	6544.7	26.9	312.4	537.4	1056.1	3093.3	74.3	71.8
1 Ab	4178	4528	459.6	491.4	1368	3735	28.29	62.51
2 Ab	5766	4708	1097	515	1403	4205	49.76	72.49
3 Ab	7144	6641	1801	2462	1402	7220	164.7	99.85
Ab average	5696	5292.3	1119.2	1156.1	1391	5053.3	80.9	78.3
1 Pp	2547	2351	430.7	541	3412	1191	12.2	59.83
2 Pp	3300	2720	619.2	533.2	3674	4430	18.05	83.46
3 Pp	3831	2739	648.1	619.7	4322	6257	22.44	94.52
Pp average	3226	2603.3	566	564.6	3802.7	3959.3	17.6	79.3

Ps *Pinus sylvestris*, Pn *Pinus nigra*, Ab *Abies bornmülleriana*, Pp *Picea pungens*

^a Numbers indicate needle age (years)

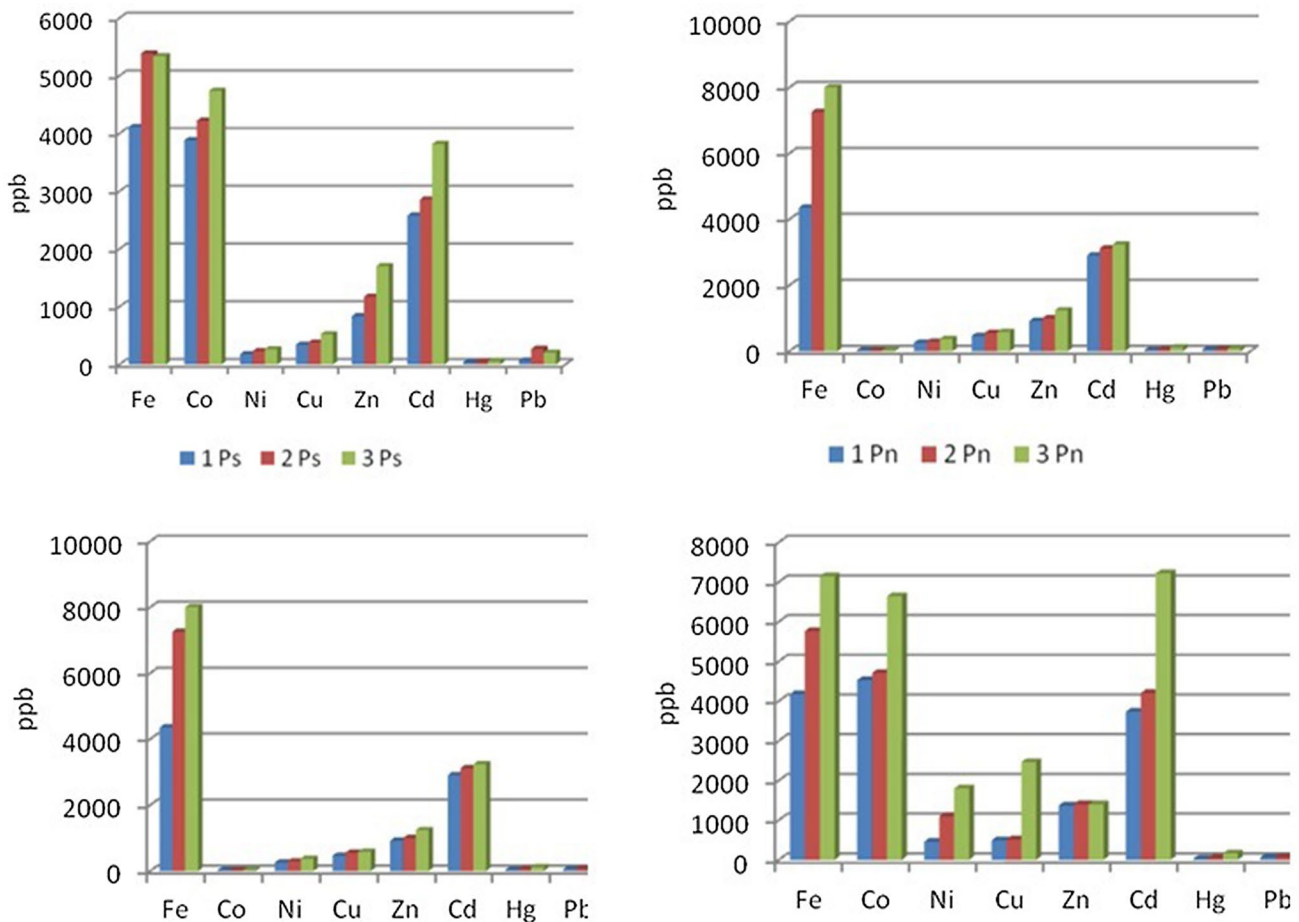


Fig. 1 Change in the amount (parts per billion; p.p.b.) of heavy metals with tree species and age. *Ps* *Pinus sylvestris*, *Pn* *Pinus nigra*, *Ab* *Abies bornmülleriana*, *Pp* *Picea pungens*

species, the variations in these, calculated on a species basis, are given in Table 2 and Fig. 2.

The heavy metal concentrations differed significantly from species to species (Table 2; Fig. 2). Ni was 225.3 p.p.b. in Ps and 119.2 p.p.b. in Ab; Zn was 1240.1 p.p.b. in Ps and 3802.7 p.p.b. in Pp; Co, Ni, Cu, Cd and Hg were remarkably high in Ab. The highest concentration for Fe was in Pn, that for Zn in Pp, and that for Pb in Ps.

Discussion

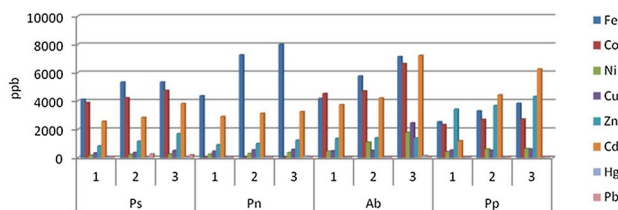
Although the main source of air pollution in cities is industrial activity, the increasing number of vehicles in cities also contributes significantly to air pollution. There is little industrial activity in Kastamonu city, where we conducted our study. However, the largest iron and steel factory in Turkey is also located in Karabük province, 100 km from Kastamonu, and pollutants such as Fe, Co, Ni, raw materials of steel, are emitted there and airborne to Kastamonu with the wind.

These emissions would have increased the amounts of these metals in our samples. The increase in the amount of Cu is due to pollution from the Küre Copper mine, 30 km from Kastamonu. Other pollutants in our samples, such as Pb, Cd and Hg, resulted from exhaust emissions and are related to the increase in traffic density in Kastamonu. According to the Turkish Statistical Institute (2017), the number of vehicles in Kastamonu was 82,334 in 2009 and reached 116,55 in 2015, i.e. a 41 % increase in 2015 compared to 2009. Thus, the emission of exhaust gases increased at the same rate as the increase in heavy metal accumulation in the needles of trees where the traffic was intense.

Accordingly, our results indicate that, in almost all species, the amounts of heavy metals increase with increasing of age of needles, but this increase is not linear. For example, in Ps Co was 3887 p.p.b. in 1-year-old needles, 4222 p.p.b. in 2-year-old needles, and 4741 p.p.b. in 3-year-old needles. Similar results were found for other heavy metals in studies on different species. Beramendi-Orosco et al. (2013) found in the annual rings of *Prosopis juliflora* that Cu was

Table 2 Variation in the concentrations (p.p.b.) of heavy metals depending on age of needles and different tree species

Needle age	Species	Fe	Co	Ni	Cu	Zn	Cd	Hg	Pb
1	Ps	4114	3887	179.5	345.3	838.2	2580	39.51	67.29
	Pn	4361	25.35	262.3	465.1	922.4	2912	42.44	57.47
	Ab	4178	4528	459.6	491.4	1368	3735	28.29	62.51
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	Pn	7264	26.34	297.9	559.2	1002	3125	59.51	73.37
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3	Ps	5343	4741	265	522.9	1706	3820	56.59	205.3
	Pn	8009	28.94	377	587.9	1244	3243	121	84.67
	Ab	7144	6641	1801	2462	1402	7220	164.7	99.85
	Pp	3831	2739	648.1	619.7	4322	6257	22.44	94.52
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	Pn	6544.7	26.9	312.4	537.4	1056.1	3093.3	74.3	71.8
	Ab	5696	5292.3	1119.2	1156.1	1391	5053.3	80.9	78.3
	Pp	3226	2603.3	566	564.6	3802.7	3959.3	17.6	79.3

**Fig. 2** The heavy metal concentrations differ significantly from species to species

1.09 p.p.m. in 1988–1992, but 1.27 p.p.m. in 2003–2007; Pb was 0.35 p.p.m. in 1993–1997, and 0.46 p.p.m. in 1998–2002.

In other studies, similar results were obtained for monthly variations. Norouzi and Khademi (2015) reported heavy metal accumulation in *Platanus orientalis* on a monthly basis and found 15.1 mg Cu kg⁻¹ in May, 15.7 mg Cu kg⁻¹ in July and 16.6 mg Cu kg⁻¹ in November; similar results were obtained for Fe, Mn, Ni, Zn and Pb. Gao et al. (2015) reported similar results when they investigated the variation of Pb, Cd and Cr in the annual rings of *Platyclusus orientalis*, *Populus tomentosa* and *Sophora japonica* over many years.

It the present study, heavy metal accumulation varied considerably between species. Similar results have been shown in a number of previous studies, e.g. in *Aesculus hippocastanum* (for Pb and Cu) (Tomasevic and Anicic 2010; Anicic et al. 2011); *Betula pendula* (Cd, Cr and Zn) (Petrova et al. 2014); *Elaeagnus angustifolia* (for Pb, Cd and Zn) (Aksoy and Sahin 1999); *Fraxinus excelsior* (Pb, Cu, Ni, Zn, Cr) (Aksoy and Demirezen 2006); *Pinus pinea* (Cr, Cu and Pb) (Oliva and Mingorance 2006); *Robinia*

pseudoacacia (heavy metals) (Celik et al. 2005); *Tilia* sp. (Pb, Cu) (Tomasevic and Anicic 2010), (Cr, Ni, Pb) (Piczak et al. 2003); *Platanus orientalis* (Pb, Zn, Ni, Cu, Cr, Cd) (Ozel et al. 2015); landscaping plants (Pb, Zn, Ni, Cu, Cd) (Ozturk and Bozdogan 2014, 2015); and *Quercus ilex* (Zn) (Gratani et al. 2008).

Various parts of plants can be used as biomonitors, e.g. the leaves of highly structured plants (Monaci et al. 2000; Gratani et al. 2008; Anicic et al. 2011), their bark (Fujiwara et al. 2011; Sawidis et al. 2011), and wood (Gao et al. 2015); in addition, lichens (Conti and Cecchetti 2001) and algae (Ceburnis and Steinnes 2000) can be used as biomonitors (Ugolini et al. 2013).

Conclusion

We determined the variation in the concentrations of eight heavy metals accumulated in the last 3 years in species in which needles stay on the trees for many years. The heavy metal levels increased with the age of the tree in almost all species. This type of information can be used to determine the level of influence of factors that cause a sharp change in recent heavy metal accumulation. For example, similar techniques can be used to determine how levels of airborne heavy metals change with the establishment of a factory with high emissions.

This study shows that there are significant differences between species in terms of heavy metal accumulation. Species with higher concentrations of heavy metals can be used more effectively for monitoring. For example, the most suitable biomonitors indicated in this study were Pn for Fe, Pp for Zn, Ps for Pb, and Ab for the other heavy metals measured.

One of the most important results is the highest concentration of each of these heavy metals, with the exception of Zn, Pb and Fe, in Ab. Although Ab is endemic to Turkey, it is grown as a Christmas tree, as well as for landscaping, in many parts of the world (Cetin 2016d; Sevik et al. 2016a, b; Cetin 2017; Sevik et al. 2017; Cetin et al. 2017b). In addition to being used as a biomonitor for many heavy metals, this species has also been found to have an important potential for the removal of heavy metals from the air. Therefore, it is advisable to plant it in urban environments, and especially along avenues.

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