HEAVY METAL LEVELS IN PINE *(Pinus eldarica Medw.)* TREE BARKS AS INDICATORS OF ATMOSPHERIC POLLUTION

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Bio-monitoring of air quality in TehranCity was investigated by analyzing 36 pine tree (*Pinus eldarica* Medw.) barks. The samples were taken from different locations with different degrees of metal pollution (urban, industrial, highway, and control sites). Then, the concentrations of lead (Pb), zinc (Zn), copper (Cu), nickel (Ni), and chromium (Cr) were measured using a flame atomic absorption spectrophotometer. The results of this study showed that the highest and lowest metal concentrations were found in the heavy traffic sites and the control site, respectively. Lead content was found to be the highest in high traffic density areas. The industrial part of the city was characterized by high Zn, Cr, and Ni contents. Variation in heavy metal concentrations between sites was observed and attributed to differences in traffic density and anthropogenic activities. The research also confirms the suitability of *Pinus eldarica* Medw barks as a suitable bio-indicator of aerial fallout of heavy metals.

Keywords: Air pollution; Bio-indicator; Heavy metals; Pine bark; Tehran

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INTRODUCTION

Heavy metals are natural components of the environment, but they are of concern lately because they are being added to soil, water, and air in increasing amounts. This is because of the rapid growth of population, increased urbanization, expansion of industrial activities, and more (Aksoy et al. 2000a). Most of the heavy metals are essential elements to living organisms, but their excessive amounts are generally harmful to plants and animals; the poison of heavy metals depends a great deal on their chemical form, concentrations, residence time, etc. (WHO 1972; Schubrek 1973; Mielk and Reagan 1988). As a result, heavy metal pollution now poses a serious threat to the earth's atmosphere by putting the natural environment, which forms the life support system of our planet, gravely at risk. Traffic emissions on roads are the main cause of heavy metal accumulation in the surrounding environment and plant species (Ward et al. 1974; Grodzinka 1977; Momani et al. 2000; Scerbo et al. 2002).

Biological monitors and vegetation have been used to measure the level of atmospheric trace metal concentration (Onianwa et al. 1986; Onsanaya et al. 1993; Wolterbeek et al. 1996; Celik et al. 2005). These monitors are applied as the cheapest and simplest indicator for monitoring the trace metal concentrations in the atmosphere. Numerous different bio-indicators have been used in monitoring air pollution, such as mosses, lichens, vascular plants, woody plants, etc. Both the broad-leaved and coniferous tree barks have been used in studies of air pollution (Löstschert and Köhm 1978; Grodzinka 1982; Lippo et al. 1995; Huhn et al. 1995; Adeniji 1996). The most economical and reasonable method for monitoring heavy metal levels in the atmosphere is by using vegetation such as Scot pine (Yilmaz and Zengen 2003), acacia (Aksoy et al. 2000a), and other plants (Aksoy et al. 2000b). Other organisms such as fishes (Rashed 2001) have also been used for biomonitoring.

Tree barks have not been shown to sensitively collect heavy metals and other pollutants, unlike mosses that take their nutrients from rainwater. However, tree bark can be used an indicator for various other pollutants based on electrical conductivity, pH, sulphur, nitrogen, and heavy metals. Thus they are suitable indicators in urban and industrial areas, where other bio-indicators are infrequent (Lostschert and Kohm 1978; Grill et al. 1981; Kreiner and Hartel 1986; Santamaria and Martin 1997). Also, in some studies only pine species were investigated to decide whether pine species can be used as a biomonitor for the determination of heavy metals. Results of such studies showed that the barks of the pine trees are good adsorbents of airborne pollutants, including anthropogenic heavy metals. Among the studied barks of pine tree are Turkish red pine (*Pinus brutia* Ten.) (Dogan et al. 2007); Italian stone pine (*Pinus pinea* L.) (Oliva and Mingorance 2006); Austrian pine (*Pinus nigra* Arnold.) (Coskun 2006); Masson pine (*Pinus massoniana* Lamb.) (Kuang et al. 2007), and Scots pine (*Pinus sylvestris* L.) (Laaksovirta et al. 1976; Huhn et al 1995; Lippo et al. 1995; Poikolainen 1997; Harju et al. 2002; Mattsson et al. 2005; Poykio et al. 2005; Samecka-Cymerman et al. 2006; Dogan et al. 2010).

The aim of this study was to investigate and assess heavy metal pollution in the atmosphere of Tehran city using pine tree (*Pinus eldarica* Medw.) barks as a bio-indicator. The result could be used as preliminary baseline data for trace elements concentrations in the ecosystems for future assessment and monitoring.

EXPERIMENTAL

Study Area

Tehran, the capital city of Iran, has an elevation of around 1400 m above sea level at latitude (35˚50´N) and longitude (51˚37´E). The average annual precipitation in the investigated area is 246 mm/y. Minimum temperature is 4.5 ˚C in January and maximum temperature is 31.5 ˚C in August. Relative humidity during daytime is relatively low, ranging from 28% in June to 64% in December. The city suffers from high traffic density caused by vehicles. The average number of vehicle movements per hour in urban, industrial, highway, and control sites of the study area are 360, 315, 1400, and <50, respectively.

Sampling and Analysis

The bark samples were collected from old trees (about 20 to 25 years old) situated along the sides of the main ways, at the end of summer season of 2007. The total number of collected samples was 36, distributed as follows: 10 samples were collected from urban, industrial, and highway sites, and 6 samples were collected from control sites. Control samples were taken from an area 20 km away from Tehran and any known source of contamination. Based on documents from the Tehran municipality, pine treesare widespread in comparison with the other tree species. Samples weighing around 100 g of the outer 5 mm of the bark at 2 m above the ground level were removed using astainless steel knife. The bark used in the study had a rough and hard surface.

In the laboratory the samples were carefully washed three times with distilled water to remove adhering particles (Babaoğlu et al. 2004) and dried in an oven at 105 ˚C for 48 h. The dried samples were ground, then homogenized by sieving them through a 2-mm plastic sieve to remove large particles.

Analysis of heavy metals was carried out by digesting 1 g from each pre-washed and dried bark sample with 10 mL of 50% HNO_3 solution, then leaving the mixture overnight. The digested samples were ultrasonicated for 1 h and heated in a test tube heater for another 1 h at 90 ˚C (Hewitt and Candy 1996). The final extracts were filtered into 25-mL polyethylene volumetric flasks through 45-μm filters, and then diluted to the mark with 1% (v/v) HNO_3 solution. Heavy metals concentrations were measured by flame atomic absorption spectrophotometer, Perkin-Elmer AAS analysis 300 model, with three replicates. Used metal standards were from Merck, Germany.

Electrical conductivity and pH values of bark samples were determined according to an earlier developed method of Hartel and Grill (1972) and Staxang (1969) and applied by Poikolainen (1997): 1.5 g of bark was mixed with 15-mL deionized water and left for 24 h. Electrical conductivity and pH of the extracted bark samples were determined usinga WTW conductivity meter (LF 320) and WTW 525 pH-meter, respectively.

Analysis of variance (ANOVA) was used to compare the significant difference in the mean concentration of heavy metals between the sampling sites. F is a parameter in the level of 5%. Pearson's correlation coefficient was used to measure the degree of correlation between logarithms of the metal concentrations. The ANOVA test and Pearson's correlation coefficient were performed using the SPSS statistical program. Also, three replications were considered for all tests.

RESULTS AND DISCUSSION

Heavy metal concentrations in the sampled pine barks are shown in Table 1. The pH values exhibited the approximately the same relative deviation for each type of sample site. The lowest values were found in the samples collected from the control site (5.18) and the highest value (5.97) was found in samples from the highway site. High electrical conductivity (EC) values (1529 μS cm^{-1}) were found in industrial sites, while the lowest value of EC was found in the control site (974 μS cm^{-1}). The results indicate that the highest and the lowest metal concentrations were found in the heavy traffic sites and the control site, respectively. The mean metal concentration values were in the following order: Pb > Zn > Ni > Cu > Cr. The lead, nickel, and copper contents were found at high concentrations in the highway sites, whereas industrial areas contained high concentrations of zinc and chromium.

Table 1. Heavy Metal Concentration Values for 36 PineTree Barks Samples Collected from Different Sites in Tehran City (mean value ± S.D.)

Parameters	Urban	Highway	Industrial	control
pH	5.31±0.3	5.97±0.5	5.45±0.4	5.18±0.3
EC (μS cm^{-1})	1165±38.2	1325±42.6	1529±48.4	974±25.5
Pb (ppm)	59.7±3.1	87.22±4.1	63.9±3.6	19.74±1.8
Zn (ppm)	20.24±1.1	25.88±1.4	33.82±1.8	2.14±0.2
Cu (ppm)	11.10±0.6	21.60±1.3	14.84±0.8	3.5±0.09
Ni (ppm)	14.22±0.5	23.38±1.1	18.48±0.7	2.60±0.07
Cr (ppm)	2.85±0.2	4.41±0.3	5.55±0.6	0.54±0.01

The correlations among metal contents in samples collected in Tehran are shown in Table 2. There are many highly significant correlation coefficients between heavy metals in all sampling sites, such as Pb vs. Cu, Ni, and Zn (r = 0.85, 0.82 and 0.79, respectively), Zn vs. Ni, and Cu (r = 0.80, and 0.77, respectively), and also Cu vs. Ni (r = 0.77). This indicated that the origin of metal in the investigated area is related to heavy traffic, industrial activities, and street dust emission. The correlation between Cr vs. Pb, Cu, Zn, and Ni were not statistically significant, which is attributed to the low Cr concentration.

Table 2. Correlation of Metal Concentration in Pine Trees (n=36)

	Pb	Zn	Cu	Ni
Pb				
Zn	0.79			
Cu	0.85	0.77		
Ni	0.82	0.80	0.77	
Cr	0.29	0.13	0.22	0.12

The analysis of variance (ANOVA) of heavy metal concentrations between the sampling sites is shown in Table 3. The results indicated significant differences in Pb, Cu, and Ni concentrations in pine trees samples collected from different sites, whereas no significant differences were found for the rest of elements. This can be attributed to different anthropogenic activities between the sites.

The Pb levels were the lowest at the control site (19.74 ppm) and the highest at highway sites (87.22 ppm), which have higher traffic density. The chemical form of lead is of critical importance, since this is a factor in movements into plants, translocation, and the toxic effectiveness of lead within the plant. Lead pollution on a local scale is caused by emissions from motor vehicles using leaded gasoline (Viard et al. 2004; Yilmaz and Zengin 2003). Allen (1989) considered a much lower value of 3 ppm as a normal natural level for plants. The close relationship between lead concentrations and traffic intensity has been demonstrated in detail by many authors (Gromov and Emelina 1994; Li et al. 2001; Viard et al. 2004). In this research there was a linear correlation between high Pb level and heavy traffic at Tehran city.

Table 3. Statistical Analysis of Variance (ANOVA) for Heavy Metals between all Sites

Parameter	Sum of squares between groups	df	Mean square between groups	Sum of squares within groups	df	Mean square within groups	F	Observed α
pH	1.81	3	0.60	6.74	32	0.21	2.85	0.22
EC	576289.41	3	192096.47	4229793	32	132181.03	1.45	0.46
Pb	16865.75	3	5621.91	24066.45	32	752.08	7.47	0.015*
Zn	824.25	3	274.75	3134.06	32	97.93	2.80	0.72
Cu	454.10	3	151.36	741.29	32	23.16	6.53	0.032*
Ni	322.58	3	107.52	721.65	32	22.55	4..76	0.024*
Cr	109.24	3	36.41	461.89	32	14.43	2.52	0.66

α, significance level.
*Significant difference between the samples (P-value<0.05).

The degree of metals content in the pine barks was found to be proportional to industrialand human activities, as well as urbanization. High metal concentrations in plants are found in industrial sites and urban and highway roadsides due to the anthropogenic activities in addition to the traffic density (Celik et al. 2005).

Zinc is an essential element in all organisms and considered to bean important factor in the biosynthesis of enzymes, auxins, and some proteins. Plants with symptoms of Zn deficiency experience a retarded elongation of cells. A critical toxic level of Zn in the leaves is about 100 ppm in dry plant matter (Allen et al. 1974; Yilmaz and Zengin 2003). High contents of zinc in leaves and plant roots may cause the loss of food production, and low levels in plants may cause deformation of leaves (Bucher and Schenk 2000; Celik et al. 2005). The level of zinc in bark samples decreases with decreased traffic density (Table 1).

Copper is a minor trace metal, with 70% copper in leaves contained in the chloroplast of land plants (Wilkinson 1994). It is an important constituent of many enzymes involved in oxidation-reduction reactions (Celik et al. 2005; Raven and Johnson 1986). Results indicated that the lowest mean value of copper (3.5 ppm) was found in samples collected from the control site, but the highest mean copper value (21.60 ppm) was found in samples collected from a highway that has heavy traffic.

The highest mean value of nickel was found in samples collected from the highway sites (23.38 ppm), whereas the lowest mean value was determined in a control site (2.60 ppm). This high concentration is attributed to emissions from motor-vehicles that use nickel gasoline and by abrasion and corrosion of nickel from vehicle parts (Al-Shayeb and Seaward 2001).

The level of chromium in the study area was generally low (Table 1). Cr is a toxic, non-essential element for plants (Shankeret al. 2005). Effects of Cr on plants are symptoms of chlorosis on leaves and decrease of root growth (Yağdıet al. 2000). In the study area, chromium pollution caused by engine and body erosion of automobiles, by extensive road marking using yellow lead chromate paint, and by some industrial activities (Al-Shayeb et al. 1995).

CONCLUSIONS

The results of this study shows that pine tree (*Pinuseldarica*Medw.) barks were found to be a good bio-indicator for air pollution, a result that agrees with other studies of pine tree barks (Dogan et al. 2007; Oliva and Mingorance 2006; Coskun 2006; Kuang et al. 2007; Laaksovirta et al. 1976; Huhn et al 1995; Lippo et al. 1995; Poikolainen 1997; Harju et al. 2002; Mattsson et al. 2005; Poykio et al. 2005; Samecka-Cymerman et al. 2006; Dogan et al. 2010). The mean concentrations (C) of the studied metals were ordered as follows: $C_{Pb}>C_{Zn}>C_{Ni}>C_{Cu}>C_{Cr}$. The mean values of metal concentrations were lower at the control site compared to all other sites. The variation in heavy metal concentrations between the studied locations is due to heavy traffic load and anthropogenic activities. Furthermore, industrial and traffic emission are the main source of metal pollution in the atmosphere of Tehran, although tree bark was washed to remove suspended dust particulates. No significant variations were found in pH values between the sites, which was attributed to a buffering effect of carbonate in the atmosphere. Electrical conductivity was high in all sites, which may be attributed to the high rate of dry deposition as the study area.

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