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Mu'taz M. Al-Alawi[1]
Mufeed I. Batarseh[1]
Hebe Carreras[2]
Mahmoud Alawi[3]
Anwar Jiries[1]
Susanne M. Charlesworth[4]

[1]Prince Faisal Center for Dead Sea, Environmental and Energy Research, Mu'tah University, Al-Karak, Jordan.

[2]University of Córdoba, FCEFyN, Córdoba, Argentina.

[3]Chemistry Department, University of Jordan, Amman, Jordan.

[4]Geography, Environment and Disaster Management, Coventry University, Coventry, UK.

Research Article

Aleppo Pine Bark as a Biomonitor of Atmospheric Pollution in the Arid Environment of Jordan

Dedicated to Prof. Dr. mult. Dr. h. c. Müfit Bahadir on the occasion of his 60th birthday

Monitoring of atmospheric pollution using Aleppo bark as a bioindicator was carried out in the industrial area surrounding the Al-Hussein thermal power station and the oil refinery at Al-Hashimyeh town, Jordan. The concentrations of heavy metals (copper, lead, cadmium, manganese, cobalt, nickel, zinc, iron, and chromium) were analyzed in bark samples collected from the study area during July 2004. The results showed that high levels of heavy metals were found in tree bark samples retrieved from all studied sites compared with the remote reference site. This is, essentially, due to the fact that the oil refinery and the thermal power plant still use low-quality fuel oil from the by-products of oil refining. Automobile emissions are another source of pollution since the study area is located along a major heavy-traffic highway. It was found that the area around the study sites (Al-Hashimyeh town, Zarqa) is polluted with high levels of heavy metals. Pine bark was found to be a suitable bioindicator of aerial fallout of heavy metals in arid regions.

Keywords: Atmosphereic Pollution; Biomonitor; Heavy metals; Oil refinery; Power plant; Tree bark

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1 Introduction

The emission of heavy metals to the environment is a serious problem which is increasing worldwide due to the rapid growth of population, increasing combustion of fossil fuels, and the expansion of industrial activities [1]. Heavy metals are mainly emitted in association with particulate material, hence the distance they are transported, compared to gaseous pollutants, is relatively short, but depends on factors connected with the production plant, such as the height of the stack, emission levels, and the size of the emitted particles [2]. The heavy fuel oil used in Jordan for cement processes contains vanadium, nickel and molybdenum, with average concentration of 50, 20, and 20 mg/L, respectively [3].

Biomonitoring may be defined as the "use of living organisms (or their parts) to ascertain the concentration of elements of interest in the given places" [4]. Heavy metals reach the bark primarily by direct deposition or via throughfall. Losses of heavy metals from the bark may occur by stem flow wash-off and by radial transport from bark to xylem [5].

Air pollution can be determined by measuring concentrations of toxic elements in dust [6], rainfall [7], dew [8], gaseous components [9], and soil [10]. However, it can be also determined by means of bioindicators, which are simple, rapid and inexpensive techniques which require no particular equipment [11]. Numerous different bioindicators can be used for monitoring pollutants, such as tree bark [12, 13], mosses [14], lichens [15], leaves [16], annual rings [17], bark pockets [18], peat [14], and invertebrates [19]. Mosses are suitable for biomonitoring of heavy metals pollution, however they is not widespread in Jordan, so Aleppo tree bark was chosen as it is widespread in Mediterranean regions [20].

The physiological function of bark is to protect the tree from mechanical injury, damaging agents and excessive evaporation [21]. However, because the bark is rough with broad, flat scales, it traps a variety of pollutants and thus has been used as a bioindicator of heavy metals [22], acid gases [23], ammonia emission [24], organic pollutants [25] and radioisotopes [26]. Tree bark is an effective bioindicator because it remains in place for an extended period of time, it is easily accessible, and sampling does not damage the tree [27]. Many different factors affect the content of heavy metals in tree bark, such as the concentration of heavy metals in the air, bark properties, climatic factors, and the mode of tree branching. Air pollutants do not accumulate in bark through simple absorption, since bark has a cation exchange system analogous to that of soils [28]. The accumulation process of the outermost bark shells is not influenced by variation in soil parameters and its elemental content [29]. Tree bark has been used to some extent to study traffic pollution and in industrial surveys [30, 31].

The present work is aimed to assess the atmospheric pollution caused by emission from the Al-Hussein Thermal Power Station and the Jordanian Oil Refinery located near Al-Hashimyeh town, Jordan, through determination of the heavy metal concentrations of Aleppo pine bark samples collected from the vicinity. The results of this work can be used as baseline data for current and future environmental impact assessments.

Correspondence: M. I. Batarseh, Prince Faisal Center for Dead Sea, Environmental and Energy Research, Mu'tah University, P.O. Box 3, 61710, Al-Karak, Jordan.
E-mail: Batarseh@mutah.edu.jo

2 Materials and Methods

2.1 Study Area

The study area is located in a flat area in the central part of Jordan at Al-Hashimyeh town, near the Zarqa governorate, and has the following coordinates: latitude 32°06′54.49° N, longitude 36°07′25.17° E, and 580 m elevation (see Fig. 1a). The area is characterized by arid climatic conditions; rainfall occurs only in the winter season that extends from November to April, and has an average annual precipitation of 182 mm. The mean temperatures are around 25 to 28°C for summer and 8 to 10°C for winter [32]. Figure 1b shows the dominant wind direction of the area to be northwesterly.

The air quality in the Al-Hashimyeh area is affected by the presence of many industries, which include the Jordanian Petroleum Refinery and a major power plant station called the Al-Hussein Thermal Power Station. Furthermore, a large wastewater treatment plant, a solid waste landfill and several light industries are located in the vicinity. The Al-Hashimyeh area is also crossed by a major highway connecting the largest three cities in Jordan: Amman, Zarqa and Mafraq (see Fig. 1b).

2.2 Sample Collection

Four sampling sites were chosen for this study, located to the north (A), southeast (B), east (C), and west (D) of the Al-Hussein Thermal Power Station (see Fig. 1b). For comparison purposes, samples were collected from a reference site that was located 5 km away from any anthropogenic activity. All samples were collected at the same time in order to get comparable results.

Bark samples were collected from Aleppo pine tree trunks (*Pinus halepensis*). The trees were of a similar age (15 to 20 years) and the outer 5 mm of the bark at 1.5 to 2 m above ground level was removed using a stainless steel knife and stored in sealed brown envelopes. Sampling was conducted during the dry season (July 2004), since rain could affect the concentrations of heavy metals in the samples.

2.3 Determination of Heavy Metals in Tree Bark

The bark samples were oven dried at 105°C for 24 h until the dry weight was constant. The dried samples were then ground and passed through a 0.2 mm plastic sieve. They were then wet digested

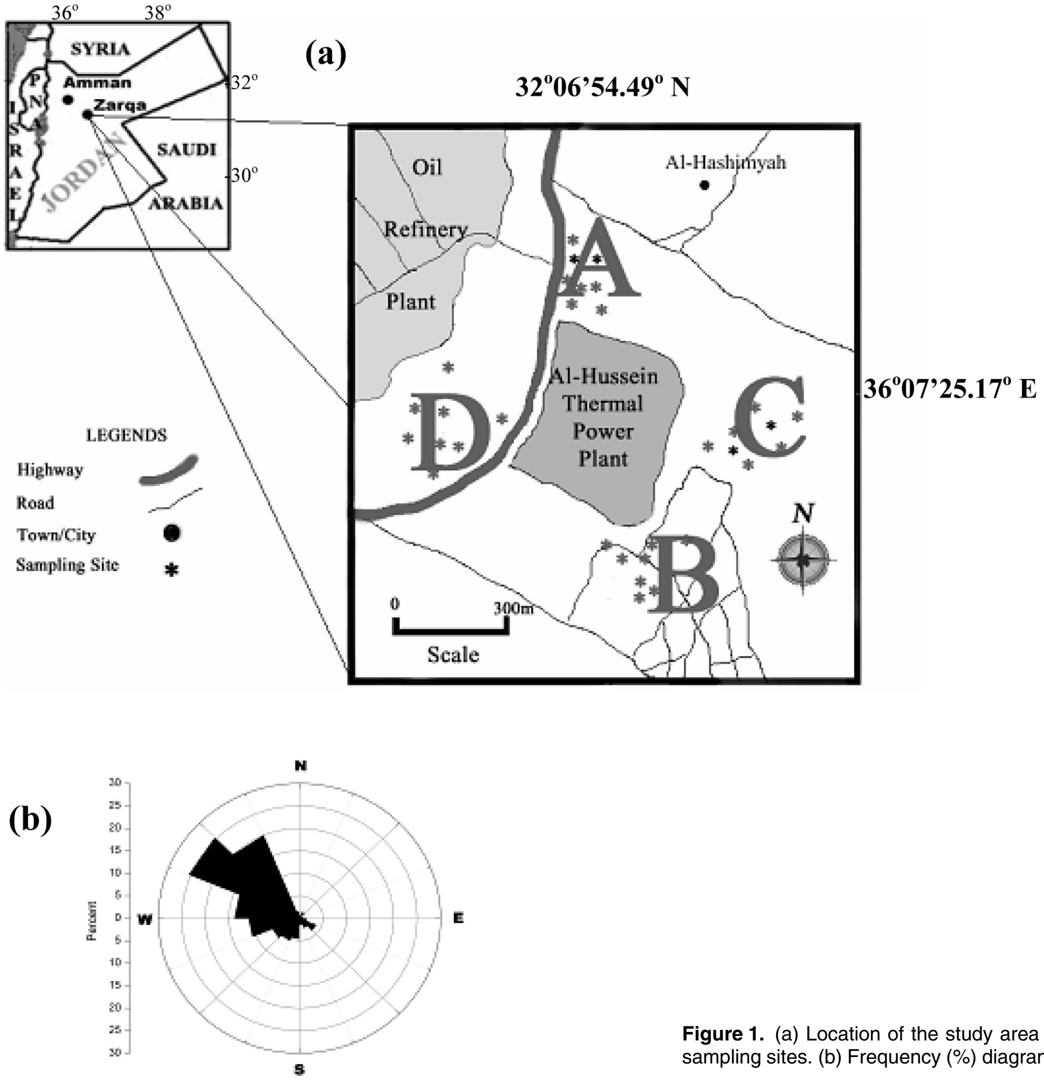

Figure 1. (a) Location of the study area in central Jordan and individual sampling sites. (b) Frequency (%) diagram of wind direction.

Table 1. Mean concentration (μg/g DW + standard deviation) of heavy metals in Aleppo bark samples for the different sampling sites (A, B, C, and D) around the power plant and reference site, also the results of ANOVA analysis distinguished by letters (a, b, c, and d).

Element	**Sampling sites**					
	A	**B**	**C**	**D**	**Reference**	**ANOVA*** **(*p*-value)**
Cu	134.89 c ± 40.15	120.04 c ± 49.33	**64.60 b ± 4.74**	**32.90 ab ± 2.19**	**4.45 a ± 0.31**	< 0.0001
Pb	**15.70 c ± 2.35**	21.35 d ± 3.31	**11.63 b ± 0.34**	19.76 d ± 3.20	**2.14 a ± 0.14**	< 0.0001
Cd	**1.88 d ± 0.15**	1.17 c ± 0.20	**0.92 b ± 0.12**	1.14 c ± 0.19	**0.17 a ± 0.02**	< 0.0001
Cr	**12.38 c ± 1.72**	8.15 b ± 1.17	9.61 b ± 1.22	8.43 b ± 2.36	**0.88 a ± 0.15**	< 0.0001
Ni	**39.72 d ± 2.71**	**25.31 bc ± 10.68**	**33.23 cd ± 8.97**	**22.95 b ± 3.93**	**0.56 a ± 0.09**	< 0.0001
Mn	**42.77 bc ± 3.61**	49.62 c ± 12.26	**35.49 b ± 2.19**	45.93 c ± 9.06	**13.42 a ± 0.19**	< 0.0001
Zn	**83.28 bc ± 7.50**	**99.14 cd ± 23.87**	**66.37 b ± 7.09**	**106.19 d ± 19.43**	**12.46 a ± 0.27**	< 0.0001
Fe	**2546.2 cd ± 17.30**	**3030.3 d ± 565.55**	**1147.4 b ± 445.88**	**2353.0 c ± 530.27**	**118.36 a ± 5.83**	< 0.0001
Co	**5.79 d ± 0.14**	4.03 c ± 0.93	4.43 c ± 0.29	**3.30 b ± 0.55**	**0.27 a ± 0.03**	< 0.0001

DW: Dry Weight.

* The values located on same row and followed by the same letter(s) do not differ significantly ($P < 0.05$), however values that have statistically significant variations were distinguished in bold font.

with an ultra-pure $HNO_3/HClO_4$ acid mixture obtained from Merck using a heating block digestion unit [33]. The final solution was filtered into a 25 mL volumetric flask through a 45 μm filter paper and diluted to the mark with ultra-pure water (resistivity 18.2 MΩ cm), obtained from a Milli-Q water purification system (Millipore Corp., USA). Ultra-pure water was used for all dilutions and sample preparation.

The methods that were used for the determination of Cu, Pb, Cd, Mn, Co, Ni, Zn, Fe, and Cr in bark were validated by the analysis of Standard Reference Materials (SRM). GBW07604 (poplar leaves) obtained from the Community Bureau of Reference were digested using the same procedure followed for the samples and analyzed along with the samples. High recoveries were obtained for all elements measured for GBW07604 (95.3%, 93.3%, 95.4%, 97.9%, 94.1%, 96%, 95%, 96.2%, and 94.4% for Pb, Cd, Zn, Cu, Mn, Co, Ni, Fe, and Cr, respectively), thereby confirming the validity of the method for the determination of these heavy metals.

A flame atomic absorption spectrophotometer model Analyst 300 equipped with graphite furnace model HG800 was used for the heavy metal analysis (PerkinElmer, Germany). Stock standard solutions containing 1000 mg/L of the analytes were obtained from Merck and used for the preparation of working standards.

2.4 Statistical Analysis

The heavy metal content from tree bark samples was analyzed with multivariate techniques (cluster analysis and principal coordinate analysis), followed by analysis of variance (ANOVA) and post-hoc comparisons using the least significant difference (LSD) test. A *P*-value of < 0.05 was considered as a significant difference. The statistical analysis was carried out using SPSS software (version 10).

3 Results and Discussion

3.1 Heavy Metals Content

The concentrations of heavy metals in bark samples of Aleppo pine from the study area and the reference site are presented in Tab. 1. The lowest concentrations of all heavy metals were observed for the reference site. This result indicates that the areas surrounding the Al-Hussein Thermal Power Plant and the Jordan Petroleum Refinery are highly polluted with heavy metals. This can be related to the fact that both the oil refinery and the power plant are still burning large quantities of low quality fuel oil without adequate control of the release of these pollutants. It was estimated that the Jordan Petroleum Refinery consumes about 147 000 t/year of fuel oil [34], whereas the Al-Hussein Thermal Power Station consumes about 518 400 t/year of heavy fuel and 2681 m^3/year of diesel oil to generate electricity [35]. Another probable source of heavy metals could be vehicle exhausts, since the study area is located along a busy major highway.

It is a well known fact that alkyl lead compounds, e. g. tetraethyl lead [$Pb(C_2H_5)_4$], were added to petrol as antiknock additives for boosting the octane rating of fuel. Consequently, the combustion of leaded fuel leads to the release of Pb in the form of particulates through vehicular exhausts into the roadside environment [36]. Considering that leaded fuel is still predominantly used in automobiles in Jordan, its combustion is the main source of Pb in this country. In the present survey, the highest content of Pb was found in bark samples retrieved from site B, followed by site D, which suggests contributions from vehicular emissions, the oil refinery and the power station. This has also been observed in Argentina, where high contents of Pb measured in biomonitors was attributed to the employment of leaded fuel by vehicles in the past decade [37].

It has been stated that the main origin of Mn, together with Fe and Al, in aeolian dust samples, is from naturally weathered materials [38]. However, the recent use of Mn-containing additives as a substitute for Pb may eventually result in automotive Mn emissions [39]. As a result, the sampling site with the highest Pb and Mn content was site B, which receives emissions from the two previous sources as well as from the highway traffic, depending on the prevailing wind direction (see Fig. 1b).

Since there is no major industry in the study area, such as smelting plants, it can be assumed that the primary sources of Zn and Cd are probably the attrition of motor vehicle tire rubber by the rough road surface [40]. Cadmium and zinc are also found in lubricating oils as a component of many additives, such as the antioxidant zinc dithiophosphates [41]. Table 1 shows that the highest concentration of Cd is found at site A and the highest for Zn at site D, both adjacent to the main highway.

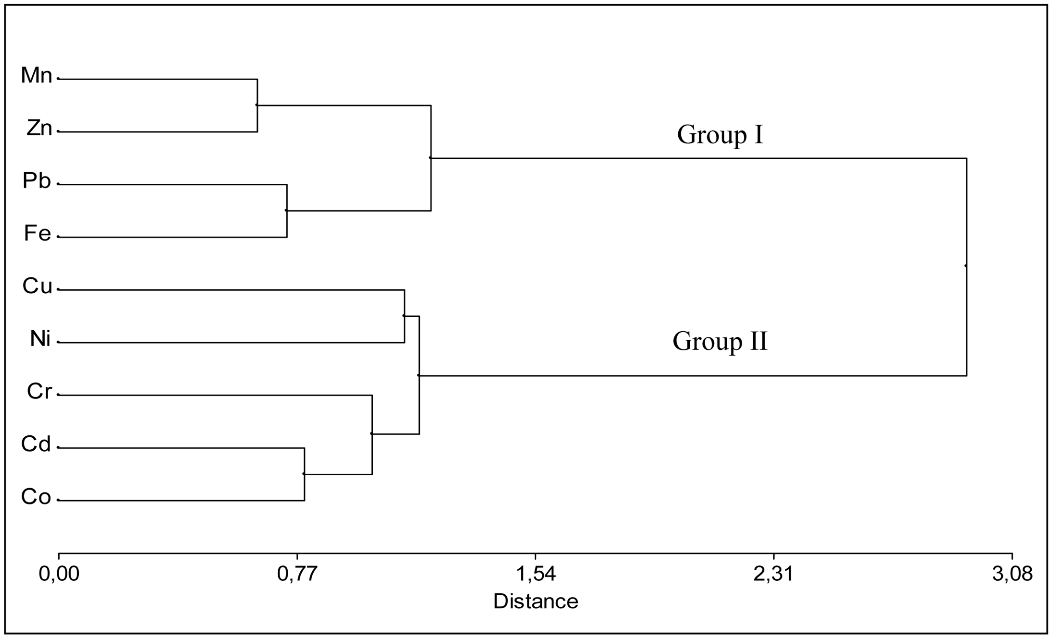

Figure 2. Results of hierarchical cluster analysis (dendrogram) of heavy metal concentrations in Aleppo bark.

The probable source of Cu in bark samples is combustion of fuel as well as vehicle engine wear [41, 42]. The highest Cu concentration was found at sites A and B, which could be related to the prevailing wind direction transporting particulate-associated contaminants from the highway and road vehicles.

The major source of Ni in urban environments is from the combustion of heavy fuel oil, which contains nickel concentrations up to 20 mg/kg [3]. Other elements, such as Co, V, Fe, and Cr are also emitted along with Ni during oil combustion [43]. In the present study, the highest concentration of Ni was found at site A.

Comparing the mean values of heavy metals measured in the present study with those reported previously by El-Hassan et al. [44] from Amman City using Cyprus (*Cupressus semervirens*) as a bioindicator, metal concentrations were mostly lower in Amman City. Lead was the exception to this trend, being lower in the present study. In spite of the continuing use of leaded fuel in Jordan, this result could indicate a possible reduction in the addition of Pb to fuel. A reduction in environmental Pb levels has taken place in many different countries prompted by a combination of the banning of the use of leaded fuel and the supply of unleaded fuel [45].

3.2 Hierarchical Cluster Analysis

In order to determine potential groupings of heavy metals, an explorative hierarchical cluster analysis was carried out according to Manta et al. [46]. The results obtained (see Fig. 2) allowed the elements to be divided into two groups: Mn, Zn, Pb and Fe constitute Group I, while Group II includes Co and Cd (closely associated), Cr, Ni, and Cu.

The first group includes those elements probably related to vehicle emissions, such as Pb and Mn which are employed as additives of fuel and Zn that may be derived from lubricating oil [47]. It is known that diesel fuels and lubricating oils contain Ca, Mg, Fe and Zn-based additives [48]. The results obtained are also in line with the finding of Wang et al. [49], who reported that emissions from the exhausts of buses are dominated by elements such as Ca, Mg, Fe, and Zn.

The second group is defined mainly by elements related to industrial production where heavy fuel oil is usually employed, with high contents of Ni. Similarly, in a survey made in Finland, the effect of industrial activity was characterized by Cu and Ni concentrations in mosses [50].

3.3 Multivariate Analysis

A principal coordinate analysis was performed to compare the heavy metal content of Aleppo barks among the sampling points (see Fig. 3). This analysis clearly revealed that tree bark retrieved from the sampling points in the study area had higher heavy metal contents than bark retrieved from the reference area. It is also demonstrated that bark from sampling points B and D had similar heavy metal contents. This result indicates that both sites are affected by the same emission sources; probably due to their location in the direction of prevailing winds.

In order to reveal the heavy metals in association with the groupings found in the previous analysis, an analysis of variance (one-way ANOVA) was performed among these groups as shown in Tab. 1. It was observed that bark retrieved from site A had significantly higher concentrations of Co, Cd, Cu, Cr, and Ni, while bark from sites B and D had significantly higher concentrations of Fe, Mn, Pb, and Zn. Including the results obtained from cluster analysis, it can be concluded that sites B and D are probably mainly affected by vehicular emissions, while sites A and C are more affected by industrial emissions.

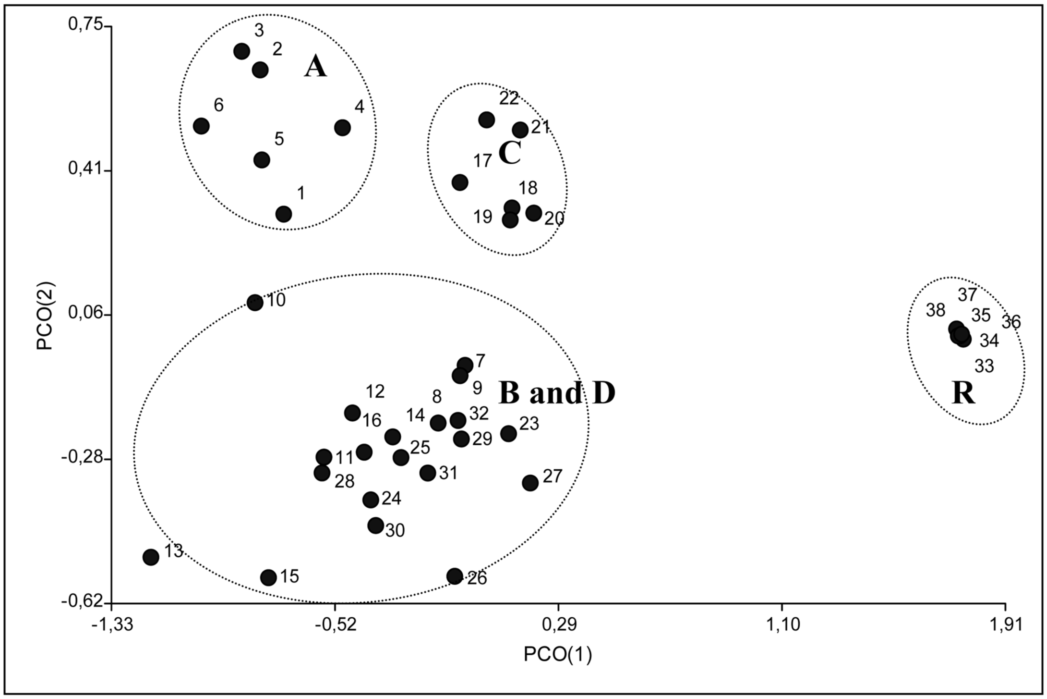

Figure 3. Spatial distribution of the sampling points where Aleppo bark was retrieved based on principal coordinate analysis.

4 Conclusions

The results obtained from the present study suggest that the area surrounding the Al-Hussein Power Plant is highly polluted with heavy metals which are attributed to the low-quality heavy fuel oil employed in the oil refinery and the power plant as well as vehicle emissions. Moreover, the analysis of the heavy metal content of Aleppo barks allows distinguishing areas with the same pattern of particles deposited. Thus, the areas located to the south and west from the power plant are the ones more affected by vehicular emissions while the areas located to the north and east are probably more affected by industrial emissions. These results are probably related to the direction of prevailing winds.

A general increase in the concentration of heavy metals was observed in the sampling area compared with a previous survey made in the same area. However, a general reduction in the level of Pb was found, which can be attributed to a reduction in the level of Pb added to fuels.

Taking into account the results obtained in the present study, tree bark samples can be proposed as efficient biomonitors for heavy metals in arid regions, where other suitable biomonitors do not usually occur.

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