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Elemental concentrations in deposited dust on leaves along an urbanization gradient☆

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HIGHLIGHTS

- Dust is used as indicators of the accumulation of inorganic pollutants.
- Scanning EM was used to explore the morphological structure of leaves.
- Amount of dust deposited of leaves correlated with trichomes' density.
- *A. negundo*, *C. occidentalis* and *Q. robur* are suitable to indicate air contaminants.
- *A. negundo* and *C. occidentalis* are suitable to decrease the amount of dust in air.

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ABSTRACT

Environmental health is an essential component of the quality of life in modern societies. Monitoring of environmental quality and the assessment of environmental risks are often species based on the elemental concentration of deposited dust. Our result suggested that stomata size and distribution were the most important factors influencing the accumulation of air contaminants in leaves. We found that the leaves' surfaces of *Acer negundo* and *Celtis occidentalis* were covered by a large number of trichomes, and these species have proven to be suitable biomonitors for atmospheric pollution difficult; these can be overcome using bioindicator species. Leaves of *Padus serotina*, *Acer campestre*, *A. negundo*, *Quercus robur* and *C. occidentalis* were used to assess the amount of deposited dust and the concentration of contaminants in deposited dust in and around the city of Debrecen, Hungary. Samples were collected from an urban, suburban and rural area along an urbanization gradient. The concentrations of Ba, Cu, Fe, Mn, Ni, Pb, S, Sr and Zn were determined in deposited dust using ICP–OES. Scanning electron microscopy (SEM) was used to explore the morphological structure and dust absorbing capacity of leaves. We found significant differences in dust deposition among species, and dust deposition correlated with trichomes' density. Principal component analysis (PCA) also showed a total separation of tree.

1. Introduction

The urban environment is a dominant habitat for humans (Duzgoren-Aydin et al., 2006). The rapid urbanization has resulted in the emission of several pollutants in the urban environment (Ahmed and Ishiga, 2006). Pollutants may enter to the urban environment from the atmosphere as gases, particles or aerosols, or by evaporation of liquids, from water by coevaporation of dissolved solvents and erosion of soil by wind (Ordónez et al., 2003). Many studies have focused on the elemental concentration of heavy metals and other contaminants (Al-Khashman, 2004; Apeagyei et al., 2011). Urbanization results in the deposition of pollutants and other toxic substances causing the degradation of environmental conditions (Duzgoren-Aydin et al., 2006).

Leaves are sensitive and highly exposed to air pollution (Prusty et al., 2005). Thus, in many studies tree leaves were used as indicators to assess the quality of air in urban environments (Aksoy et al., 2000; Al-Khlaifat and Al-Khashman, 2007; Olowoyo et al., 2010). Leaves can trap various airborne particles such as trace elements, pollens, spores and salts. Thus, they are good accumulators of atmospheric

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☆ Stomata size and distribution are important factors influencing the accumulation of air contaminants in leaves.

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contaminants (Lohr and Pearson-Mims, 1996). The capacity of leaves as dust trap depends on such factors as surface geometry, phyllotaxy, epidermal and cuticular features, leaf pubescence, and height and canopy of trees (Singh et al., 2005). Leaves collect dust on their leaf surface and trichomes (Lohr and Pearson-Mims, 1996). They could absorb gaseous pollutants via their stomata which interact between plants and their environment (Morani et al., 2011; Kardel et al., 2010); in this aspect leaves are active air contaminant collectors. On the other hand, leaves collect and deposit dust via their trichomes; thus they are also passive dust traps (Schulze and Hall, 1982; Pallardy, 2008).

In air pollution studies the leaf surface with trichomes, the size of stomata and stomatal density are the most important factors (Abbruzzese et al., 2009). Brighigna et al. (2002) studied peculiar peltate trichomes in *Tillandsia* species. These trichomes are qualified to catch aerosols and these increase the plain-tair interferences. Their studies also demonstrated that leaves by their trichomes are suitable for air pollution monitoring (Brighigna et al., 2002). Dust trap and air contaminant accumulation capability are highly dependent on the morphological and anatomical parameters of leaves. This aspect is demonstrated in an earlier study with the biomonitoring of epiphyte species of the genus *Tillandsia* (Brighigna et al., 2002). Therefore, the purpose of this study was to analyze the amount of dust and the elemental concentration of air contaminants in the deposited dust (Ba, Cu, Fe, Mn, Ni, Pb, S, Sr and Zn) along an urbanization gradient. By scanning electron microscopy (SEM) analysis we explored the interaction between morphological and anatomical parameters as well as the dust trapping and air contaminant accumulation capability of leaves. Our hypothesis was that the species best catch dust on its leaves and they could be useful biomonitors of atmospheric particulate matter.

2. Material and methods

2.1. Sampling sites and species

Sampling areas were in and around the city of Debrecen (Hungary). Debrecen is at a height of 120 m above the sea level on a nearly flat terrain of the Great Hungarian Plain (Kircsi and Szegedi, 2003). Mixed meteorological effects are typical in Debrecen because the city is located near to the border of different climatic zones. The prevailing wind direction is northwestern in Debrecen which could bring aerosol in the form of interregional transport (Dobos et al., 2009).

Along an urbanization gradient three sampling areas were chosen representing a decreasing level of urbanization (urban, suburban and rural). The urban sampling area (48°33′N 21°37′E) was located in a forested park of the city with heavy traffic around. The suburban area (47°33′N 21°36′E) was in a forested suburban area with lower traffic and other anthropogenic activities. The rural area (47°35′N 21°36′E) was in a protected forest (Nagyerdő Forest Reserve Area) where the traffic and visitor pressure were low which resulted in a more natural and undisturbed character of this area. Formerly, all sampling areas were covered by oak forest (Convollario–Quercetum roburis) (Török and Tóthmérész, 2004).

Within each sampling area four sites were selected, and 3 tree individuals were chosen randomly in each site. Ten leaves were collected from each tree individual from 1.5 m high during August 2010. In each site there were two pooled samples which contained 30 leaves each, collected from 3 tree individuals; i.e. 10 leave samples were collected from one tree individual. Thus, altogether we collected $2 * 30 = 60$ leaves from each sampling site. Leaves of *Padus serotina*, *Acer campestre*, *Acer negundo*, *Quercus robur* and *Celtis occidentalis* were collected. Samples were collected in plastic bags and they were stored at +4 °C in the dark for analysis.

2.2. Sample preparation

We used a flat bed scanner to determine the surface area of leaves. The deposited dust was washed down from leaves by deionized water. The leaves were put into a 500 mL plastic box and 250 mL of deionized water was added, then samples were shaken for 10 min. The dust containing suspension was filtered through a 150 μm sieve. The leaves were washed with 50 mL deionized water again, then filtered and added to the samples. This 300 mL of dust containing suspension was transferred into a microwave oven, where its volume was reduced to 20–30 mL. After the suspension was transferred into 50 mL glass beakers, the rest of the water was evaporated at 105 °C. The beakers were reweighted to determine the dry weight of deposited dust. Samples were prepared for analysis in the same vessels. They were digested using 5 mL of 65% (m/m) nitric acid and 2 mL of 30% (m/m) hydrogen-peroxide at 80 °C for 4 h. Digested samples were diluted to 10 mL using 1% (m/m) nitric acid and then the samples were diluted to 25 mL with deionized water (Simon et al., 2011). During digestion 10 blanks were used.

2.3. Determination of Ba, Cu, Fe, Mn, Ni, Pb, S, Sr and Zn in deposited dust

Inductively coupled plasma–optical emission spectrometry (ICP–OES) with IRIS Intrepid II XSP instrument was used during the elemental analysis. We used a six-point calibration procedure with multi-element calibration solution (Merk ICP multi-element standard solution IV).

2.4. Scanning electron microscopy

Scanning electron microscopy (SEM) was used to investigate the surface of leaves as well as the size and density of the stomata of the species. Sections (5×7 mm^2) were cut from leaves, and dehydrated in ethanol solutions of 30%, 50%, 70%, 90% and 100% for 10 min each. The sections were gold coated using an SEM coating unit PS3 evaporator. The samples were examined with an S-4300 CFE Hitachi SEM at 5.0 kV accelerating voltage.

2.5. Statistical analysis

Calculations were performed using the SPSS/PC+ and Canoco for Windows 4.5 statistical software packages. The normal distribution was tested with a Shapiro–Wilk test. The homogeneity of variances was tested with Levene's test. Principal Component Analysis (PCA) was used to display the effect of tree species and urbanization on the elemental concentration of foliage dust. The amount of dust on the surface of leaves, stomata size of leaves and the elemental concentration of foliage dust of the studied areas were compared by a two-way ANOVA where one factor was the studied sites and the other factor was the tree species. As post hoc test LSD Multiple Comparison test was used to explore the significant differences.

3. Results

3.1. Amount of deposited dust on leaves

The effect of urbanization and tree species on the amount of deposited dust on the leaves was analyzed by a two way ANOVA. The tree species had a significant effect on the amount of dust ($F_{14,45} = 64.661$, $p < 0.001$). The degree of urbanization had no significant effect on the amount of deposited dust ($F_{14,45} = 4.591$, $p = 0.068$) while the interaction of the urbanization and species was significant ($F_{14,45} = 3.786$, $p < 0.01$). The amount of deposited dust was the highest in all areas for the *A. negundo* and *C. occidentalis*. The amount of deposited dust was the lowest in all areas for *P. serotina, A. campestre* and *Q. robur*. There was no significant difference in the amount of deposited dust on the surface of *A. negundo* and *C. occidentalis* ($p > 0.05$). There was no significant difference in the amount of deposited dust among *P. serotina, A. campestre*, and *Q. robur* in the rural and suburban areas ($p > 0.05$). In the urban area a significant difference was found in the amount of deposited dust between these species ($p < 0.05$). We found no significant differences along the urbanization gradient (rural, suburban and urban areas) by LSD test (Fig. 1).

3.2. The concentration of air contaminants in deposited dust on leaves

PCA showed a total separation of tree species based on the concentration of air contaminants in the deposited foliage dust (Fig. 2). The first component (PC1) contributed to 80.2% of the total variance, while the second one (PC2) contributed 7.7% of the total variance. There were two groups of elements separated along the first axis (PC1). The first group of elements included the following elements: Sr, Ba, Cu, Ni, Pb and Zn. The second group was comprised by the following elements: S, Mn and Fe. Species and species–urbanization interaction affected significantly the concentration of Ba, Cu, Ni, Pb and Sr in the deposited dust ($p < 0.05$). The Fe and Mn concentrations changed significantly by species, urbanization and species–urbanization interaction ($p < 0.05$) (see Supplementary Data 1). The species and urbanization influenced significantly the concentration of S ($p < 0.01$). There was a significant difference in the Zn concentration ($p < 0.001$) between the species (see Supplementary Data 1). In the case of *P. serotina* a significantly higher Cu and Fe concentration was found in the deposited dust in the urban area than in the suburban and rural areas. The Pb concentration was not different between the urban and rural areas; there was a significant difference only in the case of the suburban area (Table 1). In the rural area a significantly lower S concentration was found than in the suburban and urban areas. In the case of deposited dust of *A. campestre* the Mn and Ni concentrations was significantly higher in the suburban area than in the urban and rural areas. There was no significant difference in Ni concentration between the suburban and rural areas ($p > 0.05$). In the suburban area the Mn concentration was also the highest in the deposited dust of *Q. robur* but there were no differences between the suburban and rural areas. In the deposited dust of *A. negundo* the Ba and Sr concentrations were the highest in the urban area (Table 1). The Mn and Ni concentrations were the highest in the rural area in the deposited dust of *C. occidentalis.*

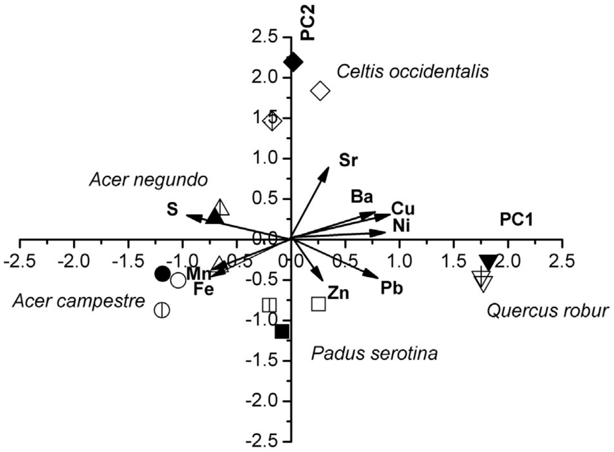

Fig. 2. Principal component biplot of element concentrations of deposited dust of species and study areas. Notations: *Padus serotina*: □ — rural area, ◫ — suburban area, ■ — urban area; *Acer campestre*: ○ — rural area, ⦶ — suburban area, ● — urban area; *Quercus robur*: △ — rural area, ⍋ — suburban area, ▲ — urban area; *Acer negundo*: ▽ — rural area, ⍒ — suburban area, ▼ — urban area; and *Celtis occidentalis*: ◊ — rural area, ⟠ — suburban area, ◆ — urban area.

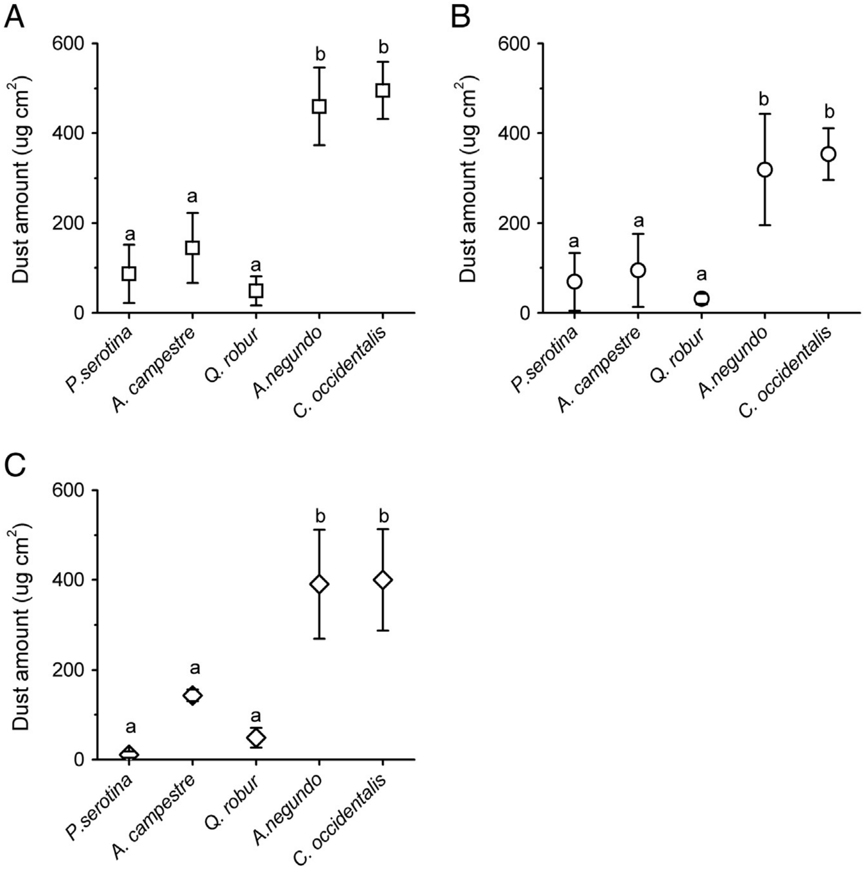

Fig. 1. Amount of deposited dust (mean ± SD) on leaves' surface of the studied species along the urbanization gradient. Notations: A = rural area, B = suburban area, C = urban area. Different letters indicate significant differences ($p < 0.05$).

Table 1
Concentration of air contaminants (mean ± SE, mg kg^{-1}) in deposited dust of tree species according to the studied areas. Different letters indicate significant differences ($p < 0.05$). Notations: "a" is always lower than "b" and "b" is always lower than "c".

		Air contaminants								
Species	Area	Ba	Cu	Fe	Mn	Ni	Pb	S	Sr	Zn
P. serotina	Rural	38 ± 9	17 ± 3^a	388 ± 145^a	609 ± 277	25 ± 5	17 ± 8^a	1223 ± 255^a	43 ± 6	91 ± 12
	Suburban	41 ± 12	22 ± 6^a	339 ± 97^a	1100 ± 166	22 ± 6	6 ± 2^{ab}	2188 ± 689^b	37 ± 9	112 ± 42
	Urban	96 ± 25	70 ± 23^b	2009 ± 718^b	828 ± 275	120 ± 94	43 ± 16^b	6400 ± 2138^b	145 ± 84	385 ± 138
A. campestre	Rural	15 ± 3	7 ± 1	286 ± 36	718 ± 111^a	3 ± 1^a	3 ± 1	5699 ± 1647	28 ± 3	54 ± 4
	Suburban	35 ± 8	15 ± 3	492 ± 114	2470 ± 496^c	7 ± 2^b	11 ± 4	12079 ± 1770	29 ± 2	119 ± 29
	Urban	16 ± 2	7 ± 1	289 ± 32	224 ± 20^b	1 ± 1^b	5 ± 1	10012 ± 3278	23 ± 7	79 ± 10
Q. robur	Rural	65 ± 16	27 ± 6	2439 ± 856	1274 ± 396^b	37 ± 20	27 ± 6	3060 ± 880	57 ± 18	159 ± 24
	Suburban	42 ± 14	31 ± 7	915 ± 128	1686 ± 155^b	19 ± 2	14 ± 6	4298 ± 1112	22 ± 7	138 ± 38
	Urban	44 ± 13	30 ± 7	1472 ± 184	348 ± 90^a	13 ± 1	26 ± 5	3632 ± 737	18 ± 8	137 ± 41
A. negundo	Rural	8 ± 2^a	7 ± 3	123 ± 15	219 ± 15	2 ± 1	2 ± 1	2081 ± 402	16 ± 4^a	30 ± 4
	Suburban	24 ± 6^a	10 ± 2	243 ± 77	248 ± 44	3 ± 1	3 ± 1	3582 ± 1038	45 ± 8^b	42 ± 10
	Urban	33 ± 11^b	7 ± 1	225 ± 40	656 ± 268	1 ± 1	1 ± 1	1574 ± 119	60 ± 12^b	35 ± 8
C. occidentalis	Rural	20 ± 2	13 ± 3	42 ± 20	165 ± 54^b	14 ± 4^b	1 ± 1	1564 ± 442	70 ± 9	27 ± 7
	Suburban	17 ± 6	18 ± 4	95 ± 14	71 ± 16^{ab}	4 ± 1^a	1 ± 1	2311 ± 279	59 ± 14	38 ± 5
	Urban	23 ± 3	16 ± 4	92 ± 23	46 ± 15^a	4 ± 1^a	1 ± 1	1985 ± 271	138 ± 24	30 ± 3

3.3. Structure of leaves' surface, stomata density and stomata size

Scanning electron microscopy (SEM) images showed that there were no trichomes in the epidermis of *P. serotina* and *Q. robur* (Fig. 3A and C). Occasionally, there were trichomes in the epidermis of *A. campestre* and *A. negundo*. The occasional occurrences of trichomes facilitate the adsorption of dust particles (see Fig. 3B and D). In the case of *C. occidentalis* a large number of trichomes were found in the epidermis. Moreover, the sizes of trichomes were extremely diverse (Fig. 3E).

The highest stomata density was found in *A. negundo* (757 ± 26 stomata mm^{-2}, N = 3) (Fig. 4D). Lower stomata density was found on the leaves of *A. campestre* (568 ± 47 stomata mm^{-2}, N = 3) (Fig. 4B), and *C. occidentalis* (511 ± 12 stomata mm^{-2}, N = 3) (Fig. 4E) than on the leaves of *A. negundo*. The lowest stomata density was found on the leaves of *Q. robur* (338 ± 65 stomata mm^{-2}, N = 3) (Fig. 4C). The lowest stomata density occurred on the leaves of *P. serotina* (237 ± 33 stomata mm^{-2}, N = 3) (Fig. 4A).

Significant differences were found among the stomata size ($F_{4,234} = 111.155$, $p < 0.001$). The highest stomata size was found on leaves of *Q. robur* (22.6 ± 0.4 μm). On the leaves of *P. serotina* significantly smaller stomata (19.9 ± 0.5 μm) occurred than on the leaves of *Q. robur*, but higher than on the leaves of other species ($p < 0.001$). There was no significant difference among the stomata size of *C. occidentalis* (13.3 ± 0.2 μm) and the other species. There were significant differences in the stomata size between *A. campestre* (14.5 ± 0.4 μm) and *A. negundo* (16.5 ± 0.6 μm) ($p < 0.05$).

4. Discussion

The amount of deposited dust is widely used in environmental pollution studies (Prusty et al., 2005; Freer-Smith et al., 2005). Prusty et al. (2005) observed an increased amount of deposited dust around sites with elevated traffic flow. A significant difference was found in the overall loaded amount of deposited dust on leaves between an urban park and a pasture land in London, where the particle uptake was higher at the urban site than at the rural area (Freer-Smith et al., 2005). They reported a significant effect of species and sites on the amount of deposited dust (Freer-Smith et al., 2005). Contrasted with their findings in our study no significant difference occurred in the amount of deposited dust along the urbanization gradient. This is caused by the meteorological and landscape characteristics of Debrecen city. The city is in the bottom of a basin, so the dust and air contaminants can accumulate in this city in high ratio. In the past 20 years the increasing traffic and industrial activities present higher air pollution (Kertész et al., 2008). Singh and Jothi (1999) and Garg et al. (2000) have also reported that the amount of deposited dust is highly dependent on the characteristic features of leaf surfaces. Similarly to our study Wang et al. (2006) investigated the correlation between the morphology and anatomy of leaf surface of plant species and the density of settling particles. They found that larger micro-roughness of a leaf surface captured higher amount of deposited dust. We found the highest amount of particles on the leaf surface of *C. occidentalis* and *A. negundo* in each investigated areas as well as these species showed the largest trichome density according to the SEM analysis. These two species contributed most effectively to the removal of urban deposited dust particles from the air. The lowest particle density occurred in the case of *P. serotina* and *Q. robur* that also showed a good correlation with the scanning electron-microscopic findings, since their surface was found to be the smoothest with the lack of trichomes. Tomašević and Aničić (2011) and Prusty et al. (2005) also explained the differences in the amount of deposited dust by the different epidermal characteristics and leaf structures.

In deposited dust samples among air contaminants the Pb concentration was lower in our study than in other findings (Al-Khlaifat and Al-Khashman, 2007; Apeagyei et al., 2011; Duong and Lee, 2011). There was high traffic in these areas. Lead is added to gasoline as organic tetraalkyl lead additives, so the main source of Pb is leaded gasoline

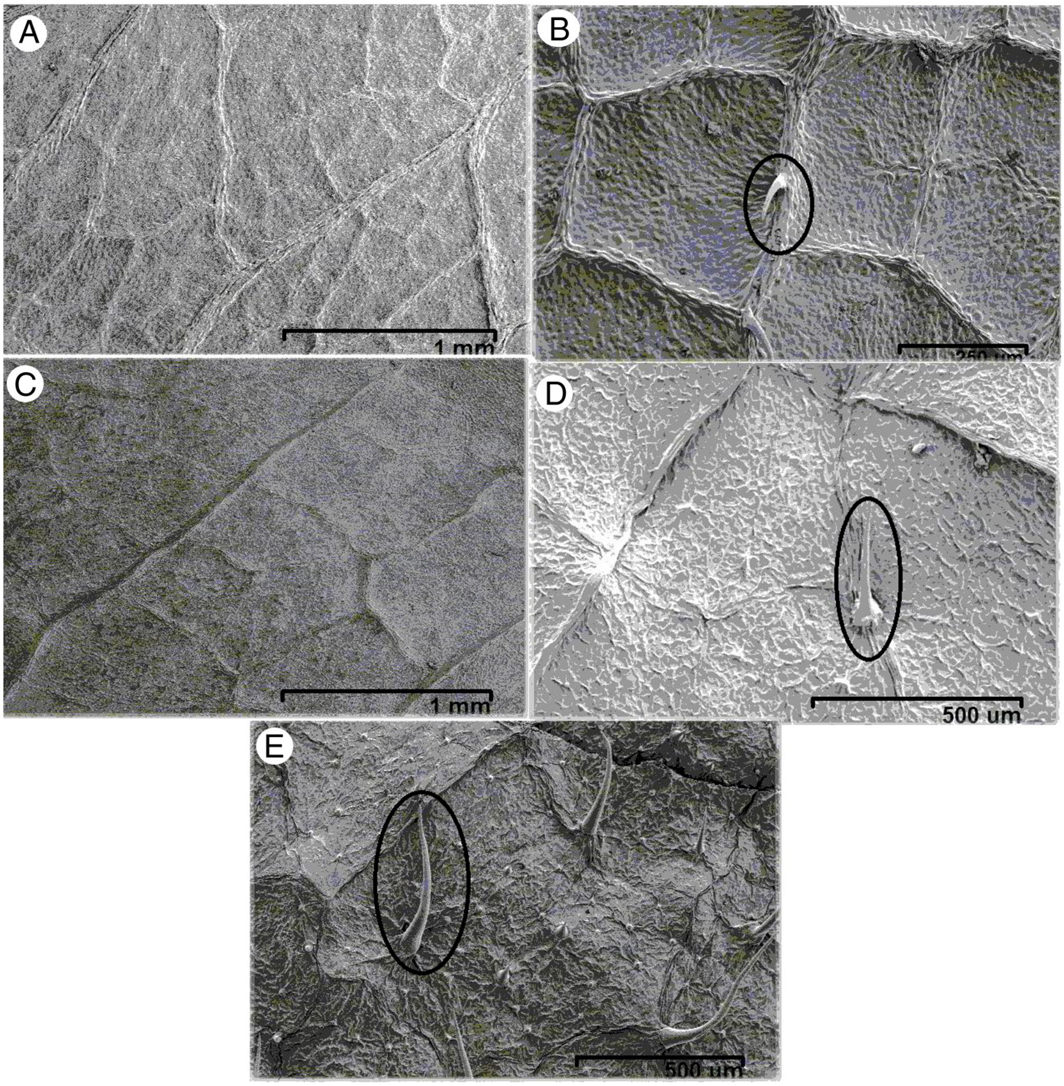

Fig. 3. SEM image of leaf surface of *Padus serotina* (A), *Acer campestre* (B), *Quercus robur* (C), *Acer negundo* (D) and *Celtis occidentalis* (E). Notations: The circle marks the location of trichomes.

(Al-Khlaifat and Al-Khashman, 2007; Thomas, 1995). Before April 1999 cars had used leaded petrol in Hungary. The lead concentration may derive from leaded petrol in environment because of lead remains in the soil for centuries (Szegedi, 2007). In spite of the removal of leaded petrol diffuse emissions (wear and tear of brake pads of cars) may cause Pb, Cu and Zn pollution (Al-Khlaifat and Al-Khashman, 2007).

Earlier reports also founded a higher Cu concentration in high traffic area than in our study (Apeagyei et al., 2011; Duong and Lee, 2011). We found a higher Cu concentration in the deposited dust of *P. serotina* and *Q. robur* in the urban area than results in an earlier study (Al-Khlaifat and Al-Khashman, 2007). The source of Cu was ascribed to corrosion of the metallic parts of cars as engineer wear, thrust bearing and brush wear (Meza-Figueroa et al., 2007). Similar to the Cu the Zn originated from car components such as tire abrasion, so tire treads and tire dust contain a large amount of Zn (Meza-Figueroa et al., 2007). In our study we found similar results of Zn concentration in deposited dust than in earlier studies (Apeagyei et al., 2011; Al-Khlaifat and Al-Khashman, 2007; Duong and Lee, 2011). The Ni concentration was higher in our study in deposited dust of *P. serotina* from urban area and in deposited dust of *Q. robur* than in earlier studies (Al-Khlaifat and Al-Khashman, 2007; Duong and Lee, 2011). Pacyna et al. (2007) reported that in the urban areas Ni mostly originated from the fossil fuel combustion stationary sources. The corrosion of cars and chrome plating of motor vehicle parts may also result in high concentration of Ni in dust in an urban environment (Christoforidis and Stamatis, 2009). Besides the Cu, the brake housing dust and crushed brake pads contain a high concentration of Fe (Adachi and Tainoshob, 2004; Schauer et al., 2006). In our study the Fe concentration was higher in the deposited dust than in the study of Al-Khlaifat and Al-Khashman (2007) but lower than in an earlier study (Apeagyei et al., 2011).

Our study indicated that the morphology and anatomy of leaves were important when they were used as environmental-quality indicator. The surface of leaves, and the size and density of trichomes were important in the dust deposition. Our study demonstrated that on the leave's surface of *P. serotina* and *Q. robur* the trichome density was low, so the dust absorbing capability of these species was also low. We found the highest trichome density on *C. occidentalis* leaves, and this resulted in a high dust absorbing capability. We also demonstrated that stomata size and density also play an important role in the accumulation of air contaminants in leaves. Tomašević and Aničić (2011) observed that most of the fine particles were deposited near the stomata, around and over, by which the physiological characteristics of leaves can be affected. Similar to their results we also demonstrated that the high size of stomata and/or high density of stomata may result in a high concentration of air contaminants in contrast with the small stomata size.

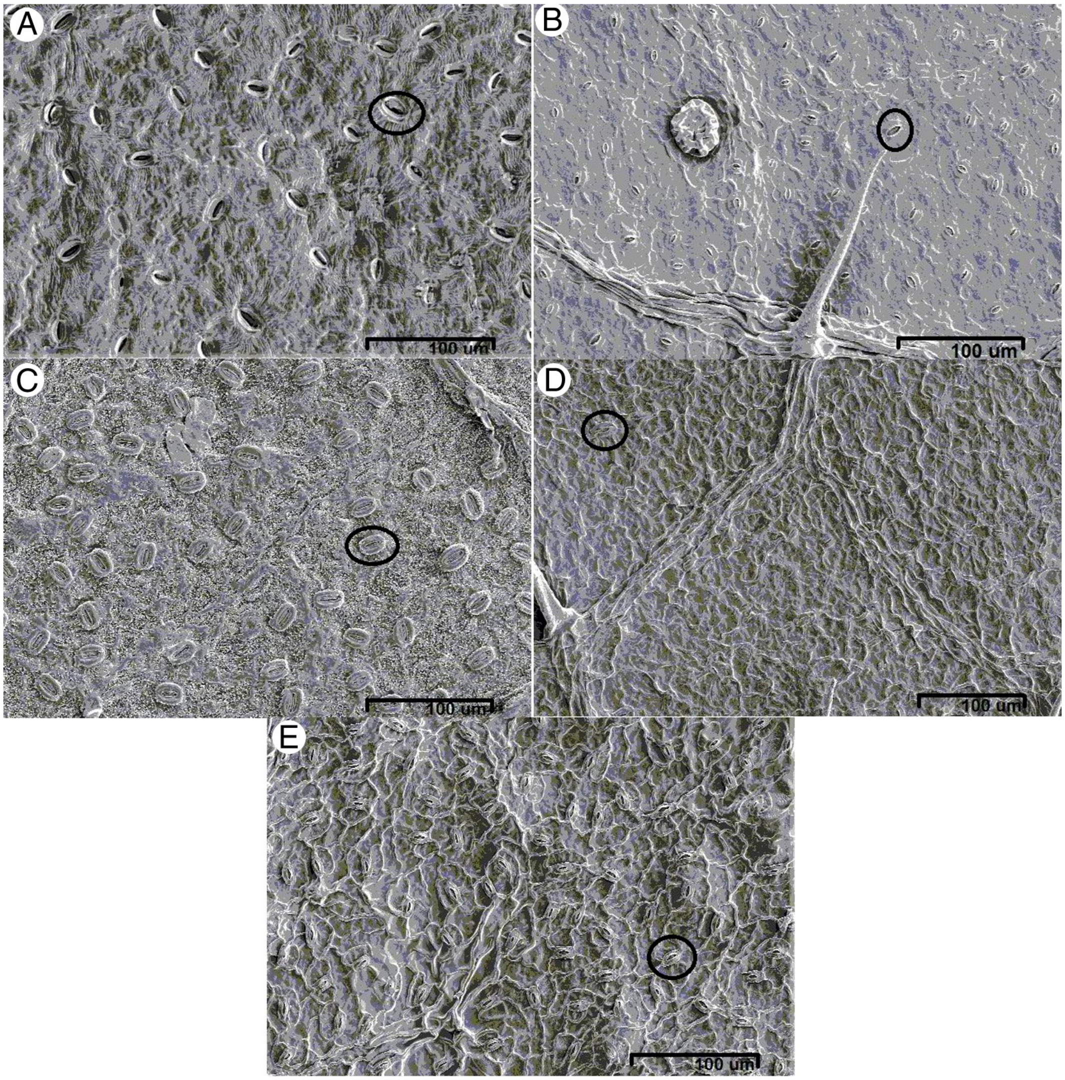

Fig. 4. Adaxial surface of with *Padus serotina* (A), *Acer campestre* (B), *Quercus robur* (C), *Acer negundo* (D) and *Celtis occidentalis* (E). Notations: The circle marks the location of stomata.

5. Conclusions

Our study demonstrated that *A.negundo, C. occidentalis* and *Q. robur* were useful biological indicators because of their large stomata size and high stomata density We found that the leaves' surface of *A. negundo* and *C. occidentalis* is covered by a large number of trichomes, which made them especially suitable to decrease the level of air pollution. Our results show that the density of trichome, stomata size and density were important in dust deposition. Leaves with a high trichome density can deposit a larger amount of dust, so these tree species may decrease the amount of dust from air in urban and contaminated areas.

Supplementary data to this article can be found online at http://dx.doi.org/10.1016/j.scitotenv.2014.05.028.

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