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Full Length Research Paper

Concentrations of heavy metals in soil and leaves of plant species *Paulownia elongata* S.Y.Hu and *Paulownia fortunei* Hemsl

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This paper sums up the results of the research on heavy metals contents (Pb, Ni, Fe, Zn and Mn) in soil and leaves of the plant species, *Paulownia elongata* S.Y. Hu and *Paulownia fortunei* Hemsl. at the plantation established on the eutric brown soil in Banat (Vojvodina). The plantation, which served as the control field is at the municipality of Bela Crkva, far away from the main traffic lines. Concentrations of analyzed heavy metals in the leaves of the tree species, *Paulownia* growing in urban and suburban conditions were compared with the concentration of polluters in the leaves of the tree species, *P. elongata* and *P. fortunei* in the experimental field in Bela Crkva.

Key words: Heavy metals, Pb, Ni, Fe, Zn, Mn, soil, plant species paulownia.

INTRODUCTION

Paulownia elongata S.Y. Hu and *Paulownia fortunei* Hemsl belong to the family of *Bignoniaceae*. Species of the genus, *Paulownia* are mostly grown on plantations. In agroforestry, there is usually interlinear planting with agricultural cultures.

The authors, Wang and Shogren (2003) described in their work the system of enriching the soil by cultivation of *Paulownia.* They point out that when *Paulownia* is grown with cereals, their different developmental period and root distribution result in more efficient use of water and other limited resources. The positive results of the usage of *Paulownia* in combination with the adaptation to the natural conditions could be very attractive to other countries. *Paulownia* also has a significant role in recultivation and protection of soil against erosion.

Plants can absorb heavy metals from soil as well as from air. The accumulation of heavy metals as well as metals predominantly toxic to plants in the soil can be a consequence of the natural lithogenic and pedogenic processes (Woolhouse, 1983), as well as anthropogenic factors which result in environmental pollution (Piperski and Radisic, 2003). A very important source of heavy metals and other pollutants of soil and plants is passenger traffic (Primault, 1958; Fidora, 1972; Johnson, 1980; Memmon et al., 2001; Stankovic, 2008 a, b).

Heavy metal uptake by plants is performed constantly during their vegetation period and during the year when they reach their highest value at the end of the vegetation period (Krstic et al., 2007; Stankovic, 2006). Also, there are a lot of references in literature that suggest influence of heavy metals on morphological, anatomical and physiological characteristics among other wooden species, such as poplars and willows (Nikolić, 2009). Species of the genus, *Paulownia* are, according to numerous literary data, characterized as efficient extractors of heavy metals (Shaw, 1990; Wang et al., 2009.)

MATERIALS AND METHODS

The research of the heavy metal content in the soil was performed at the plantation of *P. elongata* and *P. fortunei* near Bela Crkva in the region of Banat. The soil samples were taken from the pedological profiles on the fixed depths as follows: 0 - 10 cm, and 10 - 20 cm. The contents of "pseudototal" heavy metals (Pb, Ni, Fe, Zn and Mn) in the soil were determined by AAS-atomic absorption spectrometry method. The preparation of soil samples was performed in accordance with UNEP-UN/ECE procedure (UNEP-UN/

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Table 1. Sampling locations.

Label and Location description	Species	Sampling point
-K- Bela Crkva Plantation	**E1-** *Paulownia elongata* S.Y. Hu**	Top of the tree crown
	E2- *Paulownia elongata* S.Y. Hu**	Bottom of the tree crown
	F1- *Paulownia fortunei* Seem. Hemsl**	Top of the tree crown
	F2- *Paulownia fortunei* Seem. Hemsl**	Bottom of the tree crown
-1- Mali Mokri Lug	*Paulownia elongata* S.Y. Hu	In suburban conditions 10 m away from the main traffic line
-2- Mali Mokri Lug	*Paulownia elongata* S.Y. Hu	In suburban conditions 1 m away from the main traffic line
-3- Bulevar Kralja Aleksandra	*Paulownia elongata* S.Y. Hu	In urban conditions along the main traffic line
-4- Bulevar Kralja Aleksandra	*Paulownia elongata* S.Y. Hu	In urban conditions in the green zone between two lanes of the main traffic line

** E1 = *Paulownia elongata* S.Y. Hu - top of the tree crown, E2 = *Paulownia elongata* S.Y. Hu - bottom of the tree crown, F1 = *Paulownia fortunei* Seem. Hemsl - top of the tree crown, F2 = *Paulownia fortunei* Seem. Hemsl. - Bottom of the tree crown.

ECE, 1991). Methods used were 9190 SH and 9190 SA. The soil was treated by the mixture of HCl, HNO_3 and H_2O_2 in the ratio of 3:1:2.

The heavy metal contents (Pb, Ni, Fe, Zn and Mn) in the leaves of the tree species, *P. elongata* and *P. fortunei* were researched in five locations shown in Table 1.

On the location of Bela Crkva (control field), the samples were taken from the trees of two different species of *Paulownia*: *P. elongata* and *P. fortunei*, on two different sampling points on the trees (bottom and top of the tree crown).

The samples of analyzed leaves were collected before the end of the vegetation period. The samples were dried at room temperature without previous washing until they became airy dry mass. Airy dry leaves were then dried in dry-kiln at 105°C. The analysis of chemical composition of plants was performed in accordance with the standard procedure (APHA, 1995). The heavy metal concentrations were determined after dry burning at 450°C under the treatment by HCl. Out of the obtained master solutions there were measured the concentrations of Pb, Ni, Fe, Zn и Mn, AAS.

The measurement of the heavy metal concentrations in the leaves were performed by atomic absorption spectrometry method. The obtained data were processed by the statistics software Statgraph. The influence of the type and point of sampling on the concentrations of heavy metals in the leaves of the trees at Bela Crkva location was measured, using two factorial analyses of variance with interactions. There was analyzed the influence of two factors: type of the tree and point of sampling on the tree. Besides the influence of the factors and their treatment on the content of the elements (Pb, Ni, Fe, Zn and Mn), there were also analyzed the interactions among factors, especially interactions of level 2. The analysis of the influence of the location on the concentration of heavy metals in the leaves of *P. elongata* S.Y. Hu was performed by single factor analysis of variance.

The testing of the presence of significant differences between the two average values in both cases was determined by the Duncan's test for the level of significance at 95%.

The obtained results are shown in an adequate way in the form of a table and chart.

RESULTS AND DISCUSSION

The essential biogenetic elements also known as heavy metals, which are not necessary for physiological processes, are absorbed from soil. The availability of some element to plants depends on a number of factors, the most important being bioecological characteristics of plant species, concentration, chemical forms of appearance of elements in soil and eco-pedological conditions which determine mobility and availability of elements.

In the process of monitoring environmental pollution by heavy metals, more and more attention is paid to researching the heavy metals contents in plants.

Heavy metal contents in soil

Heavy metals in soil may originate from natural and anthropogenic sources. The anthropogenic sources may be numerous and the main form of accumulation of heavy metals in soil from anthropogenic sources is represented by dry and wet atmospheric deposition. Some heavy metals represent important biogenic elements which are in small quantities necessary to plants (Zn, Mn, Ni and Fe) while others are not essential biogenic elements (Pb).

Recording the deposition of a number of heavy metals from air into plants, Vukmirovic et al. (1997) established the dependence of pollution on traffic, weather conditions and vegetation period. Researching the concentration of heavy metals in soil in Hong Kong, Chen et al. (1997) concluded that urban environment is mostly polluted by lead while agricultural land and fruit gardens are polluted with cadmium, copper and zinc. They also concluded that

Table 2. Total heavy metal content in soil.

Location	Depth soil	Fe (µg/g)	Mn (µg/g)	Zn (µg/g)	Pb (µg/g)	Ni (µg/g)
Bela Crkva	0 -10 cm	23800	632.1	82.4	34.2	36.2
	10 - 20 cm	24233	625.3	79.3	33.0	36.3
Critical values *		100 - 100.000**	500-1.000***	60 - 150 (200)	25 - 100	10 - 85

* Multifunctional use possibilities (DeVries and Bakker, 1998); ** average values (Vanmechelen, 1997); *** average values (Adriano, 1986).

forest ecosystems are the least polluted.

This paper presents research on the contents of "pseudototal" forms of iron, manganese, zinc, lead and nickel in the surface layers of soil with 0-10 and 10-40 cm of euteric brown soil where plant species, *P. elongata* S.Y. Hu and *P. fortuneii* Hemsl are grown near Bela Crkva in Banat region. The research results are shown in Table 2.

The data shown in Table 2 demonstrate that there are no significant differences in the concentration of heavy metals among layers. The concentrations of iron are significantly higher in relation to other elements. In the soil layer of 0-10 cm, the concentration of iron amounts to 23,800 µg/g, and in the layer of 10 - 40 cm, it was 24,233 µg/g. Jones and Jelvis (1981) Vanmechelen et al. (1997) state that iron in the mineral of soil layers varies within a wide range of 100 to 100,000 µg/g.

Kadovic and Knezevic (2002) noted that iron concentration in the mineral of forest soils layer in Serbia amounts to a minimum value of 10,442µg/g, and a maximum value of 80µg/g. The recorded iron concentration values in the soil on the plantation of Paulownia species may be considered as "regular".

Regarding values of recorded content, manganese takes second place. In the layer from 0-10 cm, manganese concentration amounts to 632,1 µg/g; and in the layer from 10-40 cm, 625,3 µg/g. Adriano (1986) states the conclusions of Aubert and Pinta (1977) who believe that for most of soil types, regular content amounts to 500-1000 µg/g. Kastori et. al., (1991) believes that from the total quantity of manganese in soil only 0.1-1% is available to plants. Pendias and Pendias (1989) suggested concentrations of 1,500 mg/kg as a threshold where toxic symptoms of manganese appear. Therefore, the determined concentrations of manganese in the soil of Paulownia species plantation are below the critical load and plants' needs may be satisfied in accordance with the determined concentrations.

Zinc is a very important microelement. It plays a very catalytic role in enzyme reactions. Zinc content in soil types varies in a wide range. Adriano (1986) points out to an average range from 50-100 µg/g. Wietes (1966) suggested that the critical value for zinc in soil is 170 µg/g and Pendias and Pendias (1989) define lower "toxic" limit as 70 µg/g. The recorded zinc concentrations of 82.4 and 79.3 µg/g in the analyzed soil do not represent critical load. This is because high pH value limits its availability.

Lead content in soil also varies in a very wide range. Vanmechelen et al. (1997) states that the regular lead concentrations in mineral soils range from 10-100 µg/g and according to Pendias and Pendias (1989), the content of 100µg/g represents toxic value. The recorded lead concentrations in soil at Paulownia plantation amount to 34.2 and 33.0 µg/g and belong to the bottom level of critical load in relation to the possibility of multi-functional exploitation of soil defined by De Vries and Bakker (1998). The possibility of lead uptake from euteric brown soil is limited by chemical reactions and formation of heavily soluble compounds. This makes for no risk of toxicity.

In accordance with literature sources, nickel content in soil amounts to 5-100 µg/g. High nickel concentrations are contained in soil types formed on serpentinites and peridotites. According to Ubavic et al. (1993) soils on serpentinites contain up to 600 µg/g of nickel. Nickel concentrations on the soil where we established Paulownia plantation amount to cca 36 µg/g. This is significantly lower that the maximum allowed concentration, which amounts to 100µg/g.

It can be concluded that euteric brown soil on the location of Paulownia plantation provides favourable ecopedological conditions for growing the species of *P. elongata* S.Y.Hu and *P. fortuneii* Hemsl, and that certain contents of heavy metals in soil do not represent critical limitation for their growth.

As shown in Table 2, the analysis of soil for determining heavy metal concentrations showed that the content of all analyzed elements in soil is below critical limits. It may also be stated that the concentration for almost all elements is increased in the surface layer of the soil.

The analysis of variance for determining the differences in heavy metal concentrations in relation to the depth of the soil has shown that, at the probability level of 95% for nickel ($p = 0.6433 > 0.05$) and iron ($p = 0.1458 > 0.05$), there are no statistically significant differences between average values of concentration. That is according to the determined iron as well as nickel concentrations on both depths they represent a homogenous group.

On the other hand for lead ($p = 0.0011 < 0.05$), zinc ($p = 0.0008 < 0.05$) and manganese ($p = 0.0000 < 0.05$) where the level of probability is 95%, there are statistically significant differences between average concentration values. That is the average values of the concentration of lead, zinc and manganese on both

Table 3. Total heavy metal content in leaves.

Location	species	Fe (µg/g)	Mn (µg/g)	Zn (µg/g)	Pb (µg/g)	Ni (µg/g)
- K - Bela Crkva Plantation	E1 **	78.50	34.21	71.02	0.73	2.59
	E2 **	105.35	42.82	91.88	1.22	2.81
	F1 **	114.14	32.25	32.32	0.80	3.49
	F2 **	236.25	31.32	56.09	1.06	2.69
-1- Mali Mokri Lug	*Paulownia elongata* S.Y. Hu	138.49	13.16	23.14	1.07	3.23
-2- Mali Mokri Lug	*Paulownia elongata* S.Y. Hu	128.46	25.50	46.79	0.94	4.54
-3- Bulevar Kralja Aleksandra	*Paulownia elongata* S.Y. Hu	246.25	32.33	48.37	2.25	3.13
-4- Bulevar Kralja Aleksandra	*Paulownia elongata* S.Y. Hu	361.63	41.72	44.50	3.16	6.63
Average values. ECCE*		5 - 200	1 - 700	15 - 150	0.1 - 5	0.4 - 4

* Element Concentration Cadasters in Ecosystems (Licht and Markent, 1994).
** E1 = *Paulownia elongata* S.Y. Hu - top of the tree crown, E2 = *Paulownia elongata* S.Y. Hu - bottom of the tree crown, F1 = *Paulownia fortunei* Seem. Hemsl - top of the tree crown, F2 = *Paulownia fortunei* Seem. Hemsl. - Bottom of the tree crown.

depths do not represent a homogenous group.

Heavy metal contents in leaves

Phytoremediation of contaminated soil represents an effective method of decreasing the risk for human health as well as ecosystem. Besides chemical measurements, it is desirably to perform also biological assessment of ecological situation. For that purpose, biological indicators are used. The accumulation of heavy metals in plant tissue points out to a very important role of certain plant species as (bio) indicators of environmental pollution (Ten-Houten, 1983; Prasad and Freitas, 2003).

On the basis of obtained results (Table 3) on the analysis of the influence and interaction of analyzed species and sampling point on heavy metal concentrations in the tree of the plant species, *P. elongata* and *P. Fortuneii* from the control location, Bela Crkva it may be noted that:

- For the analyzed heavy metals (Fe, Ni, Mn, Zn) there are statistically significant differences in the average values of pollution of the species *P. elongata* and *P. Fortuneii* since the level of significance is $p = 0.0000 < 0.05$. However, there are no statistically significant differences in average values of lead pollution in leaves of the trees of *P. elongata* and *P. fortuneii*, since the level of significance is $p = 0.6326 > 0.05$.
- As for sampling point, the factor of influence, for all analyzed heavy metals (Pb, Fe, Ni, Mn, Zn), there are statistically significant differences in average concentrations in leaves from the bottom and the top of the crown bearing in mind that the level of significance for lead is $p = 0.0057 < 0.05$, and for other elements $p = 0.0000 < 0.05$.
- As for Fe, Ni, Mn and Zn with 95% certainty, we can claim that there is interaction between the factors A and B (species and sampling point) since the correlation coeficient amounts to $p = 0.0000 < 0.05$. It means that the influence of species on average pollution values is not independent of sampling point or rather that average pollution levels at the bottom and the top of the tree is largely affected by different plant species (*P. elongata* and *P. fortuneii*).

The situation is completely different with lead (Pb) since p = 0. 2796 > 0.05, therefore with 95% certainty we can claim that there is no interaction between the factors A and B (species and sampling point).

For the purpose of gaining the data on significance of pollution differences on all analyzed locations the data were processed by the analysis of variance and Duncan's test.

On the basis of the results of Duncan's test (Figures 1 - 5), it may be concluded that the concentration values of the analyzed elements, Pb, Ni, Fe, Zn и Mn are statistically significantly different by locations and range within A-E. The obtained results point out that heavy metal concentration adsorbed and absorbed in Paulownia leaves may be used as a very reliable indicator of chemical air and soil pollution.

The literature sources show that there is antagonism between iron and manganese or rather that the surplus of one element causes the reduction of the other and vice versa. The data in this research confirm the validity of this statement (Table 3). Plants on the locations with the greatest iron content have very little manganese.

According to average values of accumulation (Element Concentration Cadasters in Ecosystems, 1994), we can conclude that there are locations in the urban environment where iron concentration exceeds average values in plants. In location 3, there was 246.25 µg/g of iron, while the concentration of manganese on the same location was 32.33µg/g. In location 4, where iron concentration amounts to 361.63µg/g, manganese concentration amounts to 41.72 µg/g.

On all locations in urban conditions there were recorded higher iron concentrations. As a result, it may be concluded that besides other polluters, traffic has direct impact on concentration of this element in plants.

Observing manganese concentration in plants by

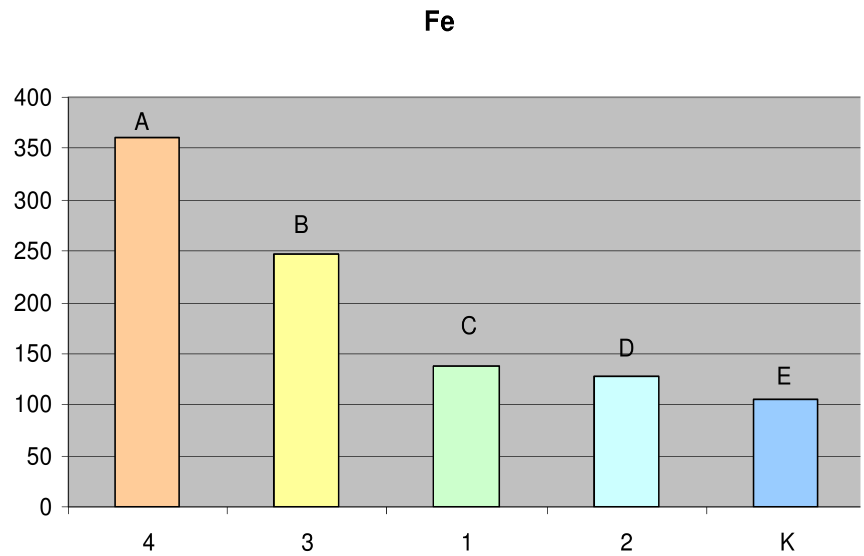

Figure 1. Iron content in *Paulownia elongata* on the analyzed locations. K = Bela Crkva Plantation; 1 = Mali Mokri Lug; 2 = Mali Mokri Lug, 3 = Bulevar Kralja Aleksandra; 4 = Bulevar Kralja Aleksandra.

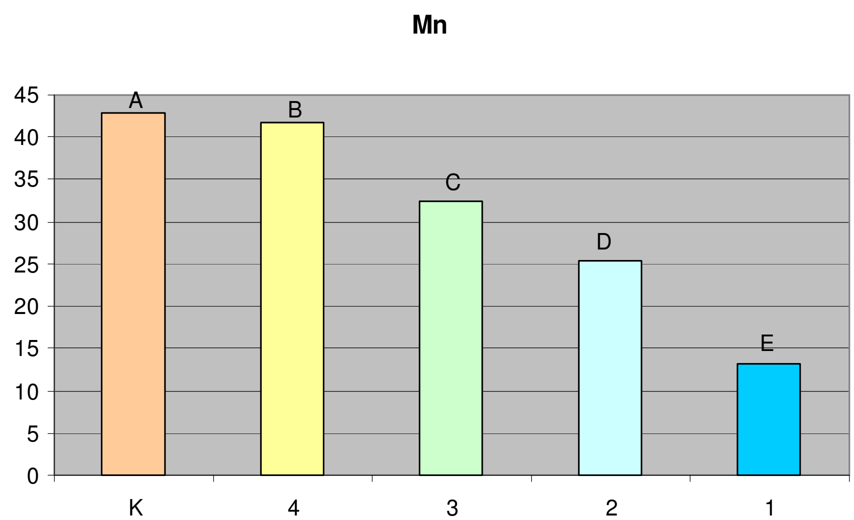

Figure 2. Manganese content in *Paulownia elongata* on the analyzed locations. K = Bela Crkva Plantation; 1 = Mali Mokri Lug; 2 = Mali Mokri Lug, 3 = Bulevar Kralja Aleksandra; 4 = Bulevar Kralja Aleksandra.

locations (Figure 2), it can be noted that the greatest manganese concentration is at the location in Bela Crkva where soil is also loaded with this element (Table 3).

Comparing plant species at the location in Bela Crkva, we can note that iron (Fe) concentration exceeds average values in *P. fortuneii* where it amounts to 236.25µg/g and 105.35µg/g in *P. elongate.* While manganese (Mn) values are 31.32µg/g in *P. fortuneii* and 42.82µg/g in *P. elongata*.

Bearing in mind the importance of zinc in plant nutrition, more significant attention is paid to its deficiency than its surplus by researchers in the world.

When talking about zinc, the most important data refer to the fact that plants in the experimental field are not loaded with pollution by this heavy metal.

Analyzing the obtained data in this research leads us to conclude that irrespective of plant species, higher zinc concentrations appear at the location of Bela Crkva in comparison to the other locations. This point out that pollution caused by this element does not only come from exhaust gases from traffic, but also to a large extent the presence of this heavy metal in soil.

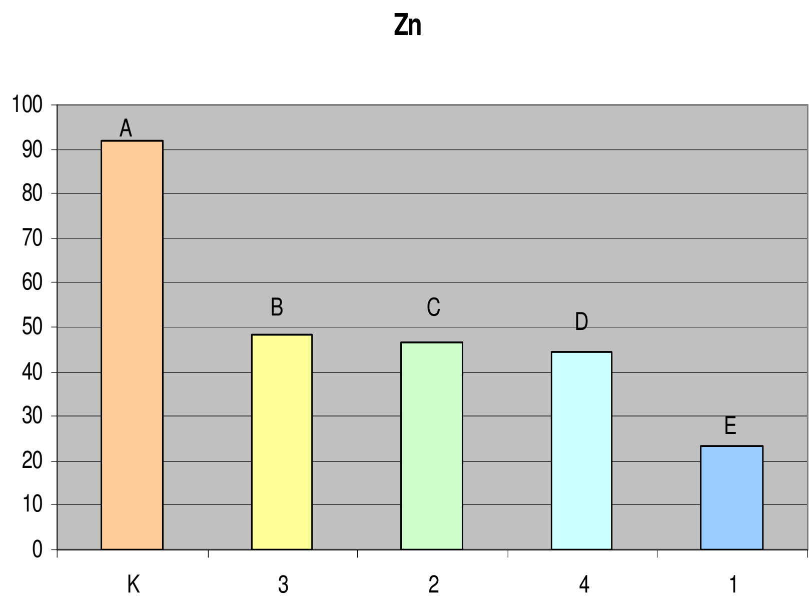

Figure 3. Zinc content in *Paulownia elongata* on the analyzed locations. K = Bela Crkva Plantation; 1 = Mali Mokri Lug; 2 = Mali Mokri Lug, 3 = Bulevar Kralja Aleksandra; 4 = Bulevar Kralja Aleksandra.

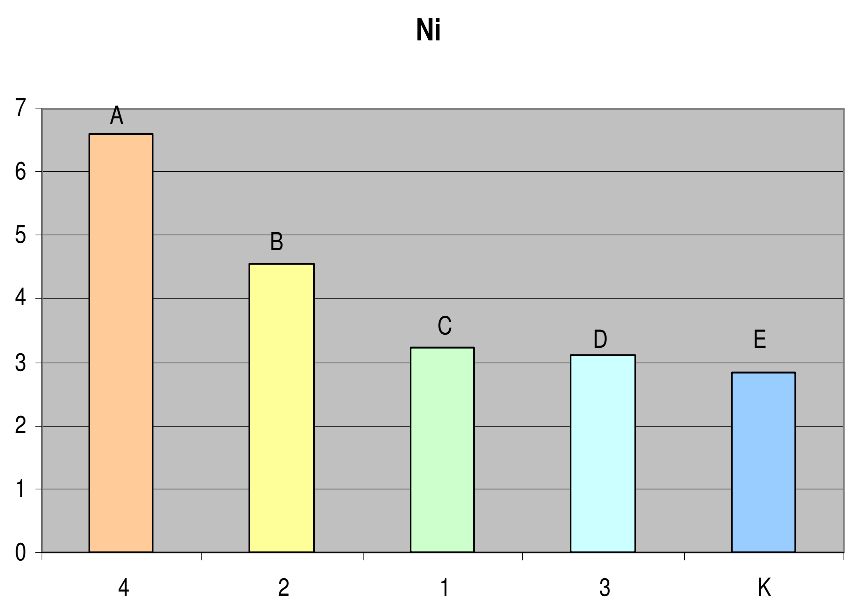

Figure 4. Nickel content in *Paulownia elongata* on the analyzed locations. K = Bela Crkva Plantation; 1 = Mali Mokri Lug; 2 = Mali Mokri Lug, 3 = Bulevar Kralja Aleksandra; 4 = Bulevar Kralja Aleksandra.

In our research, the range of zinc concentration values of all plant species on all analyzed locations ranged from 23.14 µg/g (at location 1) - 91.88 µg/g in *P. elongata* at the location in Bela Crkva.

The obtained data on lead concentrations in this research point out to anthropogenic origin of lead or rather that it originates from traffic. The highest concentrations (3.16µg/g) are at the location with the greatest traffic frequency, in trees which grow under urban conditions in the green zone between two lanes of the main traffic line at Bulevar Kralja Aleksandra in Belgrade (location 4).

An important difference obtained in pollutants concentrations from different locations of test-plants undoubtedly

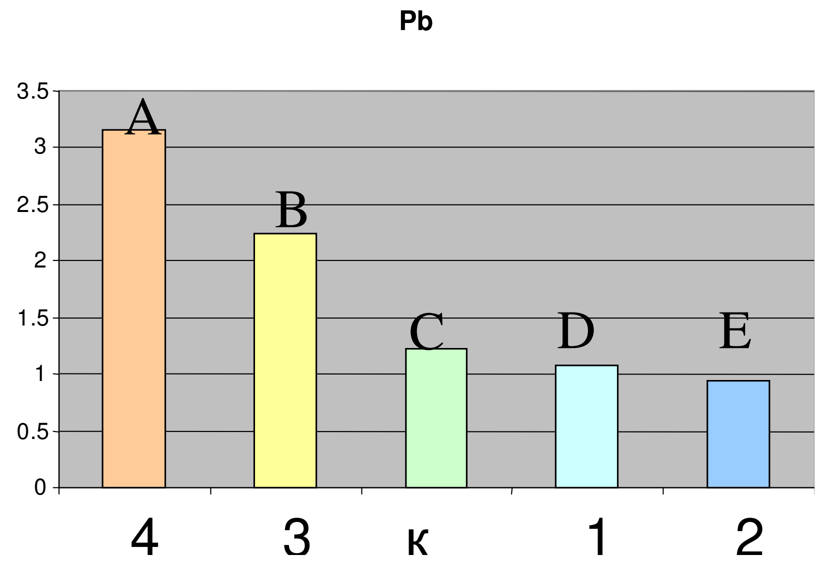

Figure 5. Lead content in *Paulownia elongata* on the analyzed locations. K = Bela Crkva Plantation; 1 = Mali Mokri Lug; 2 = Mali Mokri Lug, 3 = Bulevar Kralja Aleksandra; 4 = Bulevar Kralja Aleksandra.

points out that there was lead contamination in the regions of frequent traffic. Moreover, the presence of significantly higher lead concentrations in leaves of *P. elongata* in plants taken from the control location, Bela Crkva in comparison to that of the Mali, Mokri and Lug locations indicates the existence of chemical contamination that is primarily of air, and also of soil.

The origin of this pollution undoubtedly indicates that traffic is the main pollutant, and then higher concentration of soot and other particles in air on which lead is absorbed and later deposited on vegetation.

Comparing the two species, *P. elongata* and *P. fortuneu* both growing on the experimental field in Bela Crkva, it can be assumed that average concentrations of Ni are less in the leaves of the species, *P. elongata* than in the leaves of the species *P. Fortunei.* While with lead the situation is vice versa. The obtained differences in concentration are statistically significant only for Ni.

The average concentrations of Ni at 2.7µg/g in the experimental field in Bela Crkva are two times lesser than its concentrations in the extreme urban conditions with 6.63µg/g (sample 4). In the immediate vicinity of suburban traffic lines it amounts to 4.54µg/g (Sample 2). This is in line with the data obtained by Frey and Corn, (1967), who emphasizes that Ni appears in higher concentrations, especially in the vicinity of busy roads as a consequence of fuel consumption.

The results of this research (Chart 4) show that the main source of nickel on locations 1, 2, 3 and 4 in the urban environment are of anthropogenic origin, that is the origin of nickel in these locations undoubtedly points out to traffic emissions. The values at locations 2 and 4, where there is the greatest frequency of traffic range from 4.54-6.63µg/g.

This relation of the accumulation values for nickel by locations is similar to the accumulation of lead and it precisely shows that these two elements are directly influenced by traffic.

Conclusion

On the basis of the research results for the content of iron, manganese, zinc, lead and nickel in the soil and leaves of the plantation of *P. elongata* and *P. fortunei* near Bela Crkva as well as in the leaves of the trees under urban conditions at Bulevar Kralja Aleksandra in Belgrade and suburban conditions in Mali, Mokri and Lug, there can be drawn the following conclusions:

The determined heavy metal concentrations in the soil of the plantation near Bela Crkva do not represent critical load for the growth of the species, *Paulownia elongata* and *Paulownia* fortunei in these locations.

Comparing the two Paulownia species on the location of Bela Crkva, it can be concluded that according to the average heavy metal concentration, the species, *P. fortunei Hemsl* is hyperaccumulator of Fe and Ni in comparison to the species *P. elongata S.Y.Hu.* While comparing the concentrations of Mn and Zn, we have an opposite situation. For Pb there are no statistically significant differences in the average pollution.

The species, *P. elongata* excellently bears urban conditions so that it may be recommended for growing in parks, tree alleys and wind protection zones along urban and regional traffic lines. In addition, it is a very decorative species which is very prominent in blossoming period.

On the basis of Duncan's test, the concentration values of the analyzed elements per locations are statistically significantly different and range from A-E.

On all locations under urban conditions (where the traffic is the primary anthropogenic polluter), there were found higher concentrations of Fe, Ni and Pb on the basis of which one may note that in the analyzed area the anthropogenic influence is in the first place represented by the influence of emission from internal combustion engines.

The polluters concentrations and especially lead and nickel fall with the increase of the distance from the anthropogenic pollution source, that is the plants at the location of Bela Crkva have on average 50% less lead and nickel in comparison to the locations in urban conditions. In the location of Bela Crkva the leaves of the plants show the greatest quantities of Zn and Mn which undoubtedly points out to domination of emission of these elements from soil or rather that on this location prevails natural source of heavy metals emission.

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