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LEAD QUANTITIES IN YEW (*TAXUS BACCATA L.*) LEAVES IN THE GREATER ZAGREB AREA, CROATIA

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Hršak, V. & al.: **Lead quantities in Yew (*Taxus baccata* L.) leaves in the greater Zagreb area, Croatia. Nat. Croat. Vol. 2, No 2, 109-122, 1993, Zagreb.**

The concentration of lead in yew leaves (*Taxus baccata L.*) in urban areas (the city of Zagreb) was compared to that in non-urban woody areas (Mount Medvednica) between 3rd January and 15th February 1990. The samples differed significantly according to the lead concentration in the leaves with regard to traffic volume and road distances. The researched data established the following function of interdependence between the concentration of lead in yew leaves, and the distance from the main roads and traffic volume was established: $C_{Pb\text{-}inside}=f(L,G)$.

Key words: lead, leaves, yew, *Taxus baccata,* function, air pollution, traffic volume.

Hršak, V. & al.: **Količine olova u listovima tise (*Taxus baccata* L.) u okolici Zagreba, Hrvatska. Nat. Croat. Vol. 2, No 2, 109-122, 1993, Zagreb.**

Istraživana je koncentracija olova u listovima tise (*Taxus baccata* L.) komparativno u urbanoj i vanurbanoj šumovitoj zoni (grad Zagreb i planina Medvednica) u razdoblju između 3. siječnja i 15. veljače 1990. Uzorci su se međusobno signifikantno razlikovali u koncentraciji olova u listovima s obzirom na gustoću prometa i udaljenost od prometnica. Na temelju izmjerenih podataka ustanovljena je funkcija ovisnosti koncentracije olova u listovima tise o udaljenosti od prometnice i gustoći prometa $C_{Pb\text{-}inside}=f(L,G)$.

Ključne riječi: olovo, listovi, tisa, *Taxus baccata,* funkcija, onečišćenje zraka, gustoća prometa.

INTRODUCTION

Among the microelements whose quantity in ecosystems increases as a result of human activities and has an unfavourable impact on the environment lead occupies the dominant position. Therefore, research into lead pollution has been more intensive than research into all other microelements together (ORMROD 1988).

The main source of atmospheric lead pollution in soil is the burn-out of the lead additives in the petrol, metal industry, pesticides (lead arsenate), phosphate fertilisers, lead-based tints, etc. Approximately 75 percent of metal lead from petrol comes out with the exhaust gases in the form of aerosols into the atmosphere (SMITH 1971). This confirms the similarity of isotopic composition of aerosols containing lead and lead additives in petrol (CHOW & EARL 1970).

The emission of lead increases with the growth of traffic intensity and the utilised amounts of petrol. Thus, in Switzerland the lead emission increased from 150 tonnes to 1,300 to 1,400 tonnes annually from 1947 to 1969 (HÖGGER 1971).

The burn-out of lead additives caused significant pollution to roadside environments with great traffic volume (CANNON & BOWLES 1962, MOTTO et al. 1970, PAGE & GANJE 1970). The amount of lead on the surface of the roadside plants can be 5 to 200 times higher than that on locations away from the roads (SMITH 1976).

Recently there have been attempts to solve the problem of lead pollution by introducing unleaded petrol. Lead concentration measurements established a correlation between the increase of unleaded petrol and the drop of lead concentration in the air (ELSENREICH et al. 1986). Croatian petrols contain an average amount of 0.6 Pb/l, which is significantly higher than the leaded petrols in Europe where the average amount of petrol is 0.4g Pb/l (CEROVAC & DŽANIĆ 1986). In addition the amount of unleaded petrol in the total consumption of motor petrols has not reached the level of the developed European countries, despite the growing number of vehicles equipped for running on unleaded petrol. Due to all those reasons research into the lead quantities in the vicinity of roads in Croatia is always very interesting, especially since there is certain data on the Zagreb area from the earlier periods (JURČIĆ 1976).

AREA OF RESEARCH

The area researched includes the centre of the city of Zagreb (in latitude 45^0 44′ to 45^0 50′ N and in longitude 15^0 59 to 16^0 18′ E from Greenwich and at 120 meters altitude above sea level) where traffic is very extensive (Fig. 1), and yew (*Taxus boccata* L.) is widespread.

Two localities on Mount Medvednica were used for non-urban reference. They are localities 23 and 21-22 (**Fig. 1**) at distance of 1,000 and 800 metres from the roads, and 7 and 3 kilometres from the city.

The list of localities and their distances from the nearest road are shown individually in Table 1. The position of Mount Medvednica is north of Zagreb, its highest peak being

1035 meters. The phytogeographical position of Zagreb and Mount Medvednica is the Illyrian Province of the Euro- Siberian - North American Region.

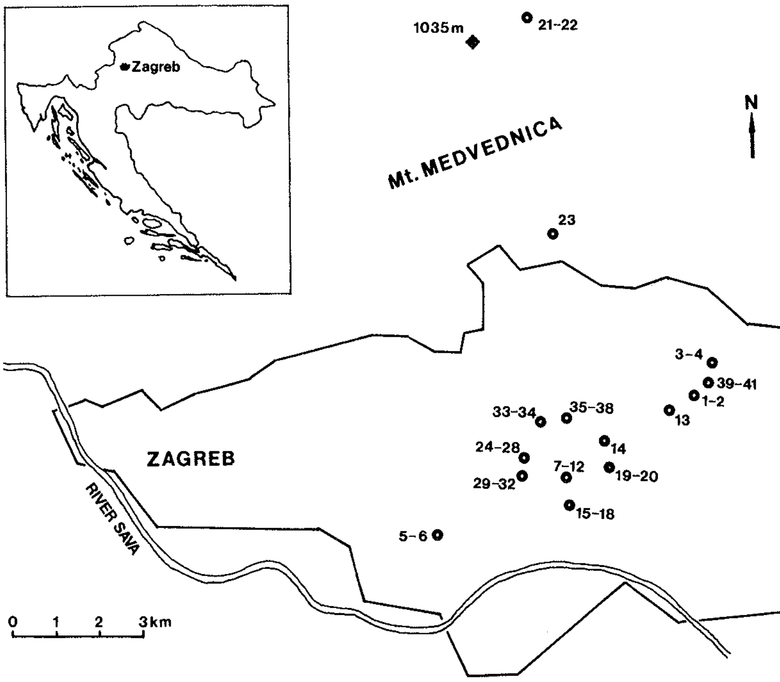

Fig. 1. - Research area.

MATERIALS AND METHODS

The lead quantity was researched in yew leaves (*Taxus baccata L.*) since it is an evergreen coniferous tree, and lead is accumulated all year round. In addition, due to its relative resistance to polluted atmosphere (LARCHER 1980) yew trees grow well in parks and other city grounds, and are also widespread in natural habitats in non-urban woody areas on Mount Medvednica at a distance from the roads but relatively close to Zagreb.

Yew leaves were taken at altitude 1.5 to 1.2 meters on the crown side facing the road in the winter period between 3rd January and 15th February 1990. Twigs up to two years old were picked, and the leaves were separated from the twigs in the laboratory. A half of the total amount of leaves of every sample was left at room temperature for a week and then dried in the drier at 105^0 C to achieve a constant weight (total of 4 hours). The dry samples were ground to powder in an electric grinder. The other half of every sample was rinsed in 200 millilitres of distilled water in a rotating shaker for three times during 15 minutes each time.

Two grams of each sample was digested by acid digestion in a mixture of sulphur, nitrogen and perchloric acid (ALLEN et al. 1974). The cooled solutions were filtered quantitatively through Whatman No. 44 into 50 millilitre volumetric flasks and then filled up to the volume.

The lead concentration in the solutions prepared in such a way was determined by atomic absorption spectrophotometer and was calculated in ppm dry matter.

The traffic volume (G) is shown in Pa units representing the number of cars per day expressed in personal car units (VESELINOVIĆ et al. 1989). Those values are demonstrated by index I to IV.

I ≤ 5000 Pa
II 5001 - 10000 Pa
III 10001 - 14000 Pa
IV > 14000 Pa

The distances of yew trees from which samples (L) were collected were classified into groups with indexes I to V.

I 0-15 m
II 16-30 m
III 31-80 m
IV 81-600 m
V > 600 m

The following basic statistical parameters were calculated for the lead concentrations measured (CPb) in yew leaves: minimal and maximal concentration, arithmetic mean (x) and standard error of arithmetic mean (Sx).

Two hypotheses were tested according to the Kruskal-Wallis test with $\alpha = 0.05$:

- hypothesis $H_0 1$: lead concentrations in yew leaves do not differ significantly dependent on the traffic volume, and

- hypothesis $H_0 2$: lead concentrations in leaves do not differ significantly dependent on the distance from the roads.

A third hypothesis $H_0 3$ was tested by Mann-Whitney test: lead concentration on leaf surface does not differ from the lead concentration inside leaves.

The ratio of lead quantity which can (Pb-outside) and of that which cannot be rinsed (Pb-inside) out was tested by linear regression (ROLF & SOKAL 1969, SOKAL & ROHLF 1980).

On the basis of the data measured the function $C_{Pb\text{-}inside}=f(L,G)$ was determined.

RESULTS

The results of the analysis of lead quantity in yew leaves (*Taxus baccata L.*) in the city of Zagreb and Mount Medvednica can be found on Table 1.

The highest total amount of lead in unrinsed leaves was 3.71 ppm, in locality 18 at 10 meter distance from a road with high traffic volume. The smallest amount of lead was 0.34 ppm in the samples taken in a non-urban area on Mount Medvednica (localities 21 and 22) at 1000 meter distance from the closest road with a low traffic volume. The mean value of the total amount of lead was 2.19 ± 0.06 ppm.

The greatest concentration of lead remaining in the leaves after rinsing ($C_{Pb\text{-}inside}$) was 2.32 ppm in locality 30, 5 meters from a road with very high traffic volume. The smallest lead concentration in rinsed leaves was 0.21 ppm and was also found in the non-urban zone on Mount Medvednica, at localities 21 and 22. Lead concentration mean value in the rinsed leaves was x=1.22 ± 0.08 ppm.

Table 1. List of locations and lead concentrations in yew leaves.

No	Locality	Dist. m	Dist. zone	Traff. vol.	CPb total	CPb inside
1	Maksimir	22	II	III	2.19	0.98
2	Maksimir	31	III	III	1.15	0.62
3	Vidikovac	500	IV	III	0.83	0.62
4	Vidikovac	480	IV	III	0.94	0.46
5	Tratinska	30	II	II	2.86	1.2
6	Tratinska	40	III	II	2.54	1.45
7	Stros. trg	35	III	III	1.79	1.03
8	Stros. trg	5	I	III	2.38	1.87
9	Stros. trg	15	I	III	3.44	1.78
10	Stros. trg	1	I	III	3.55	1.78
11	Stros. trg	1	I	II	2.86	1.28
12	Stros. trg	5	I	II	3.35	1.97
13	Maks./Bukov.	7	I	III	2.55	1.36
14	Vlaška	5	I	III	2.47	1.42
15	Av. Vukovar	60	III	IV	2.86	1.43

Continued

16	Av. Vukovar	70	III	IV	2.59	1.6
17	Av. Vuk./Mira.	35	III	IV	3.03	1.91
18	Av. Vuk./Lisin.	10	I	IV	3.71	2.01
19	Krešim. trg	12	I	III	1.43	0.77
20	Krešim. trg	2	I	II	2.64	1.91
21	Horv. stube	1000	V	I	0.34	0.21
22	Horv. stube	1000	V	I	0.34	0.3
23	Vila Rebar	800	V	I	0.47	0.43
24	Frankop.	2	I	IV		1.45
25	Dežel. prilaz	5	I	IV		1.24
26	Trg M. Tita	2	I	IV		1.3
27	Trg M. Tita	2	I	IV		1.3
28	Trg M. Tita	2	I	IV		1.35
29	Mažuran. trg	25	II	III		1.15
30	Kršnjavog	5	I	IV		2.32
31	Sav./Interkon.	15	I	IV		1.25
32	Klaićeva	20	II	III		0.83
33	Radićeva	15	I	II		0.64
34	Kaptol	2	I	II		1.15
35	Ribnjak	15	I	III		1.38
36	Ribnjak	5	I	III		2.02
37	Ribnjak	12	I	III		1.38
38	Ribnjak	25	II	III		0.78
39	Maksimir	72	III	III		0.78
40	Maksimir	85	IV	III		0.88
41	Maksimir	120	IV	III		0.47

In reference to $H_0 1$, the results of Kruskal-Wallis test showed that $H=13.447>\chi^2 {}_{.05}(3)=7.815$, which leads to the conclusion that the lead concentrations measured in the rinsed samples significantly differ depending on the traffic volume. Since $H=20.08>\chi^2.05(4)=9.488$ for H_02 we can definitely reject the hypothesis and conclude that the lead concentrations measured significantly differ depending on the distance from roads.

Faster increase of lead quantity can be identified inside leaves than on their surface. The results of Mann-Whitney test demonstrate that we certainly to accept hypothesis H_03 $Z_{(adj.)}=1.742<t_{.05(\infty)}=1.96$ and conclude that there is no significant difference in the lead concentrations inside leaves and those outside them.

Lead concentrations on leaf surface ($C_{Pb\text{-}outside}$) and inside the leaves, which cannot be removed by rinsing ($C_{Pb\text{-}inside}$), have a correlation coefficient r=0.723 and were fitted by linear regression

$C_{Pb\text{-}inside}=0.103 - 0.689G$ (Fig. 2).

Lead concentration depends on the distance from the road according to the following function $C_{Pb\text{-}inside}=1.771 - 0.437\log_{10}L$ (Fig. 3).

The dependence of lead concentration in leaves on traffic volume and road distance was fitted by function $C_{Pb\text{-}inside}=1.311 - 0.382\log_{10}L + 0.132G$ (Fig. 4).

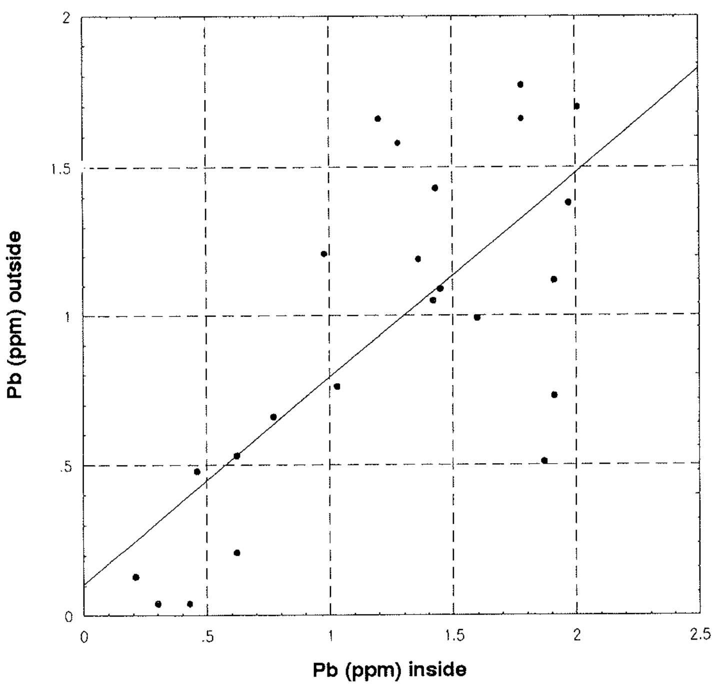

Fig. 2. - Scattergram of Pb-inside nad Pb-outside with regression line $C_{Pb\text{-}inside}=0.103 - 0.689G$.

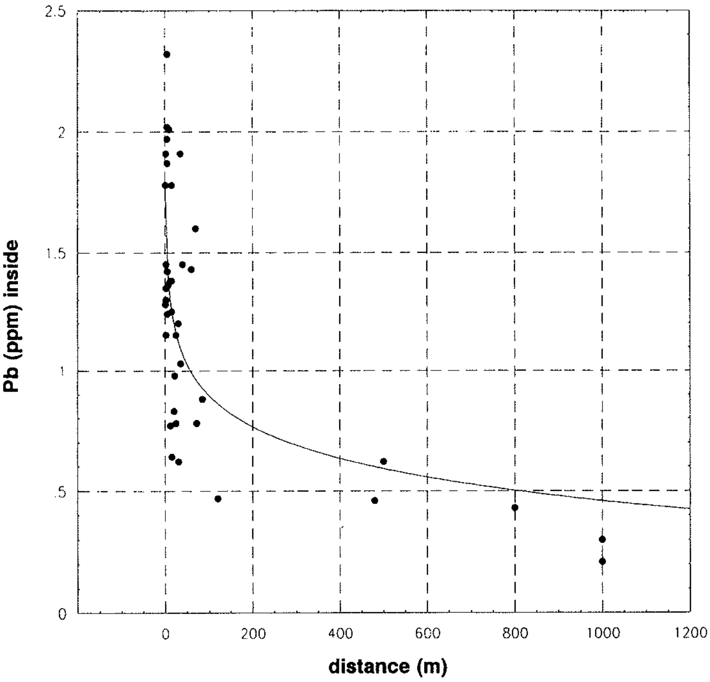

Fig. 3. - Scattergram of Pb-inside and distance from roads with fitted line $C_{Pb\text{-}inside}=1.771 - 0.437 \log_{10}L$.

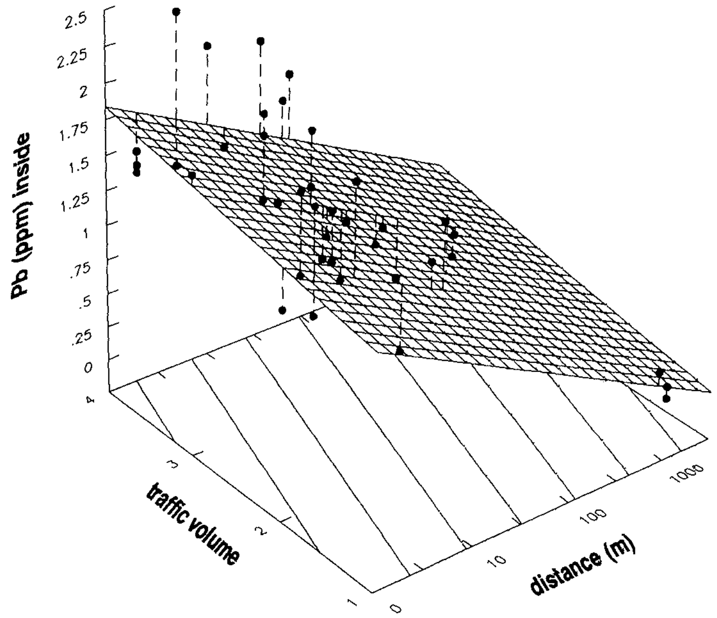

Fig. 4. - Fitted function $C_{Pb\text{-}inside}=1.311 - 0.382 \log_{10}L + 0.132G$ trough the data.

DISCUSSION

Research into numerous plant species that are exposed to the influence of exhaust pipe gases in different areas showed that great lead concentrations can result in some plants. Thus, the lead concentration in the leaves of plants growing in the vicinity of high traffic volume roads can be 5 to 200 times greater than the amount in parts far from roads (SMITH 1976). For example, in Frankfurt this amount was over 500 ppm in *Bryum argenteum* moss species, and about 300 ppm for caseweed *Capsella bursa-pastoris* while 180 ppm was calculated for *Poa annua* grass (LÖTSCHERT 1983). Some other grass species were established to contain as much as 300 ppm lead, while the natural amount of lead remote from the roads is only 5 to 10 ppm (KLOKE & RIEBARSCH 1964).

The highest concentrations of lead in plants can be identified in areas with the greatest traffic volume and the lowest air circulation (LÖTSCHERT 1983). Thus, lead concentration of an incredible 2,337 ppm was reached in moss at a spot with the lowest air circulation in Frankfurt due to a nearby building (LÖTSCHERT 1983).

In deciduous tree leaves in urban areas, the amount of lead varies between 280 and 430 ppm, while it can reach 790 ppm in the bushes in the middle of the roads (FIDORA 1972).

In yew tree leaves located at 8 meters from the road in New Haven, Connecticut (U.S.A.), SMITH (1972) measured a median lead concentration of 159 ± 21 ppm in unrinsed leaves, and 139 ± 20 ppm in rinsed leaves. The values were significantly higher than those stated by (LÖTSCHERT 1983) in reference to the yew tree in Frankfurt, where the highest value was 6.91 ppm. In comparison to those concentrations, the greatest lead concentration observed in Zagreb (3.71 ppm) was half as much as that in Frankfurt and substantially lower than in SMITH's research in the U.S.A.

This research also showed that the greatest lead concentration in Zagreb was about 11 times higher in comparison to the lowest one measured in non-urban area on Mount Medvednica. The results confirm earlier research (GOLDSMITH et al. 1970, SMITH 1976, WHEELER & ROLFE 1979) which determine the dependence of lead concentration in leaves on traffic volume.

It is also worth noticing that lead concentrations that occur in the leaves of deciduous trees and in the leaves of some other plants are regularly greater than with the yew or some other coniferous trees. That can be explained by the small surface of the leaves and by their low transpiration intensity (KELLER & PREIS 1967, KELLER & ZUBER 1970)

Lead is deposited in plants mostly as sediment on the leaf surface (SCHUCK & LOCKE 1970) which can be removed by more or less vigorous rinsing (KLOKE & RIEBARTSCH 1964, KELLER & PREIS 1967, SUCHODOLLER 1967, HEILENZ 1970). In our research we managed to remove an average of 43.4 percent of lead quantity by rinsing, while SMITH (1972) managed to remove only 18 percent of lead from the yew tree leaves. The amount of lead that can be removed from leaves by rinsing depends on the structure of the epidermis and on the form of lead deposited on the surface. Plants with bare leaves retain a smaller part of lead particles, so that only 11 percent of the lead amount can be rinsed from *Robinia pseudaccacia* (FIDORA 1972). Furry leafed plants and greater structure of the surface allow deposit of more lead on the surface. Thus, birch (*Betula pendula*) receives 39 percent of the total lead quantity deposit.

Lead is usually found on leaf surface in the form of a microcrystal or crystalline microaggregates with a diameter of 1 to 25µm (GRAHAM & KALMAN 1974). If insoluble lead salts are found on the leaf surface, they remain on the outside epidermis (SMITH 1971). If they are in a soluble form, such as PbClBr is, they can enter into the leaf through stomata /pores, while small quantities can penetrate through the cuticle (ARVIK & ZIMDAHL 1974).

Lead absorption through the root has almost no impact on the quantity in the leaves, since most of the lead is retained in the root (HAAR TER et al. 1969, SUCHODOLLER 1967, HEILENZ 1970, KELLER & ZUBER 1970, BROYER et al. 1972, KLOKE & RIEBARTSCH 1964).

Apart from traffic volume, lead concentration in leaves also depends on the distance from the roads. Inorganic lead particles most frequently have a diameter of 1 to 5 µm (SMITH 1971), so that the gravitational sedimentation causes high lead concentration in plants at a distance smaller than 35 meters. Lead concentration in plants is swiftly reduced with the increase of the distance, the drop being most frequently approximated with the distance exponential function (WARD et al. 1975). This model was generally accepted, until the model of the double exponential function was developed (WHEELER & ROLFE 1979) in which the first exponent is connected with the larger particles that are deposited up to 5 meters from the road and the other with the smaller particles that spread up to 100 meters from the road rim. On the basis of the data gathered in our research a function was developed that shows the dependence of lead concentration in leaves on the distance from the road (Fig. 3)

Local air circulation (LÖTSCHERT 1983) is also an important factor in the spreading of lead particles. Thus, local prevention of circulation caused by the vicinity of a building and connected with the great traffic volume resulted in absolutely the greatest lead concentration in plants, which was recorded in Frankfurt (LÖTSCHERT 1983). It was noted that lead concentration in the direction of a dominant wind can be about 5 percent (HAMPP 1973).

Our research was largely carried out in urban areas where the buildings and trees have greatly changed the natural air circulation, so that the local relations importantly affected the lead concentration in leaves (MÜLLER & MEURER 1989). Such relations, which are impossible to control, are certain to have made a significant impact on the great variability of the lead concentrations measured in the leaves.

CONCLUSIONS

Research into lead concentration in yew leaves (*Taxus baccata* L.) in the Zagreb city area and in the non-urban woody area on Mount Medvednica in the vicinity of Zagreb supports the conclusion that:

1. The amounts of lead in the leaves were greater in the city of Zagreb than on Mount Medvednica, the highest concentration in the centre being 11 times higher than the concentration in the non-urban woody area.

2. An average 43 percent of lead could be removed by rinsing the leaves in distilled water.

3. Lead concentration in yew leaves depended on traffic density and the distance from the road, resulting in the following dependence function:

$$C_{Pb\text{-}inside} = 0.311 - 0.382 \log_{10} L + 0.132G$$

4. The greatest concentration of lead in yew leaves was almost half as much as that in Frankfurt, and several times smaller than that found in Connecticut, U.S.A..

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SAŽETAK

Količine olova u listovima tise (*Taxus baccata* L.) u okolici Zagreba, Hrvatska

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Koncentracija olova u listovima tise (*Taxus baccata* L.) istraživana je komparativno između 3. siječnja i 15. veljače 1990. u urbanoj i izvanurbanoj sredini (grad Zagreb i planina Medvednica), pomoću atomske apsorpcione spektrofotometrije. Najveća koncentracija iznosila je 3.71 ppm i izmjerena je u središtu Zagreba u blizini prometnice s gustim prometom. Najmanja koncentracija iznosila je 0.21 ppm, a izmjerena je u izvanurbanom šumskom području na planini Medvednici. Koncentracije olova u listovima tise koje su izmjerene na području grada Zagreba bile su niže od koncentracija u drugim područjima. Pranjem u destiliranoj vodi moglo se ukloniti prosječno 43% količine olova. Statistički je utvrđeno da su se uzorci međusobno signifikantno razlikovali s obzirom na gustoću prometa i udaljenost od prometnice. Na temelju izmjerenih podataka ustanovljena je funkcija ovisnosti koncentracije olova u listovima tise o udaljenosti od prometnice i gustoće prometa $C_{Pb\text{-}inside}=1.311 - 0.382\log_{10}L + 0.132G$.