

CONCENTRATION OF HEAVY METALS IN LEAVES OF LIME TREES (*TILIA SPP.*) IN THE URBAN AREAS OF TIRANA, CAPITAL OF ALBANIA

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Abstract

*Rapid urbanization leads to an increase in air pollution. The source of pollution that contributes mostly is the release of emissions from fossil fuels used in internal combustion engine vehicles. Vegetation can be used to mitigate urban pollution. Lime tree is a common urban tree, well adapted to the conditions of Tirana, the capital of Albania. Within this study, the concentration of the following heavy metals, Pb, Cd, Cr, Cu, Ni, Mn, Zn, Al, and Fe, have been measured in the leaves of three species of the Lime tree, and specifically *Tilia tomentosa* Moench, *Tilia cordata* Mill., and *Tilia platyphyllos* Scop. The leaf samples were collected in spring, summer, and autumn and in three stations with qualitatively evaluated different intensities of urban traffic (high, medium, low). Results show that the average concentration of heavy metals measured in the leaves of Lime tree followed the order: Fe>Al>Zn>Mn>Cu>Ni>Cr>Pb>Cd. Cr, Al, and Fe were measured at a higher concentration in the leaf samples collected in autumn, compared to the leaf samples collected in spring in the same stations. For Cu, it is exactly the opposite, meaning that the concentration was higher in the leaves collected in spring, compared to the ones collected in autumn in the same stations. Whereas, for the concentration of Pb, Cd, Ni, Mn, and Zn it was not noticed a straight seasonal trend. The average measured concentration of*

Cu, Cd, Ni, Cr, and Al, resulted in exceeding the recommended threshold of the World Health Organization, indicating pollution from these heavy metals.

Key words: *Urban Pollution, phyto-remediation, lime Tree, heavy metals, Tirana (Albania).*

Përmbledhje

Urbanizimi kontribuon ndjeshëm në ndotjen e ajrit, kryesisht përmes emetimeve nga karburantet që përdoren në automjetet me motor me djegie të brendshme. Ky studim analizon rolin që luan bimësia në reduktimin e ndotjes urbane, me fokus bimën e blirit, e cila është e përshtatur mirë ndaj kushteve urbane në Tiranë, kryeqytetin e Shqipërisë. Në këtë studim u matën përqëndrimet e disa metaleve të rënda: Pb, Cd, Cr, Cu, Ni, Mn, Zn, Al dhe Fe, në gjethet e tri lloje bliri: Tilia Tomentosa Moench, Tilia cordata Mill. dhe Tilia platyphyllos Scop. Mostrat e gjethëve u morën gjatë pranverës, verës dhe vjeshtës në tri stacione me dendësi të ndryshme të trafikut. Rezultatet treguan se përqëndrimi mesatar i metaleve të rënda në gjethet ndiqte renditjen: Fe > Al > Zn > Mn > Cu > Ni > Cr > Pb > Cd. Veçanërisht, përqëndrimet e Cr, Al dhe Fe ishin më të larta në mostrat e vjeshtës krahasuar me mostrat e pranverës, për të njëjtat stacione. E kundërta ndodh me përqëndrimin e Cu, i cili ishte më i madh në pranverë krahasuar me vjeshtën. Për Pb, Cd, Ni, Mn dhe Zn, nuk u vërejtën ndryshime sezonale të mëdha. Është thelbësore të theksohet se përqëndrimet mesatare të Cu, Cd, Ni, Cr dhe Al e tejkaluan pragun e rekomanduar nga Organizata Botërore e Shëndetësisë, duke treguar ndotje të madhe nga këto metale të rënda.

Fjalë kyçe: *Ndotje urbane, fitoremedimi, bliri, metalet e rënda, Tirana (Shqipëri).*

Introduction

Urbanization contributes to increased air pollution (Zhan et al., 2023), with the primary source being the emissions from fossil fuels released by internal combustion engine vehicles (Lu et al., 2020, Ghio et al. 2001). In recent research (Harvard University, 2021), it is evaluated that more than 8 million people died in 2018 because of fossil fuel pollution, meaning that air pollution

from burning fossil fuels like coal and diesel was responsible for about 1 in 5 deaths worldwide.

Tirana, the capital of Albania, has seen fast urban growth in the last 30 years, after the fall of communism. By the end of 1990s, Tirana's urbanized area covered 12 km², and its population was about 250,000 inhabitants (Dino, 2020). Nowadays, 1/3 of the population of Albania or around 1 million inhabitants live in Tirana (Institute of Statistics - INSTAT, 2023). The growth has expanded the size of the city and has highly increased the density of buildings and people per unit of surface, and also the number of vehicles per family. It is evaluated that a number of 848,127 vehicles circulate in Tirana within 24 hours (INSTAT, 2023). As a consequence, this has increased the air pollution and, in some areas, far exceeding the norms. In Albania, 68 of 100'000 persons die every year because of air pollution and only in Tirana air pollution is 60 % over the safe level (World Health Organization - WHO, 2019).

In order of mitigating air pollution, different methods could be implemented (Sofia et al., 2020). Phytoremediation has proved to be an efficient method (Yan et al., 2020). This is because trees are very efficient at trapping atmospheric particles as they have a special role in reducing the level of fine, "high risk" respirable particulates, which have the potential to cause serious human health problems (Ren et al., 2023; Beckett et al., 2000). The ten most important elements in atmospheric heavy metal pollution are in ranking: Fe, Al, Pb, Zn, Ti, Mn, Cu, V, Ni, Cr (Hoodaji et al., 2012). There is evidence that trees play an important role in ecosystems because they transfer elements from the abiotic environment to the biotic one and reduce human exposure to anthropogenic pollutants (Speak et al., 2012; Martínez-López et al., 2014). Plants can be used as bio-monitors of environmental pollution (Sawidis et al., 2011). Urban trees play an important role in the urban living space, by bringing social benefits, improving environmental quality (through filtering and taking up gases and particles (Beckett et al., 2000) and also contributing to local identity (McDonald et al., 2007; Nowak, 2007).

Tirana has a high diversity of plants and suitable climatic conditions. In the urban areas of Tirana, one of the most common planted trees is the Lime tree. The potential use of *Tilia spp.* as ornamental urban trees and as medicinal plants is well-known in all regions of Europe (Ivănescu et al., 1966; Şofletea

& Curtu, 2007; Soukand et al., 2013). *Tilia* species leaves have showed significant seasonal accumulation especially for Pb but also for Cr, Fe, Ni, Zn and Mn (Bargagli, 1998; Piczak et al., 2003; Tomašević et al., 2004b; Anicic et al., 2011). The difference in the leaf structure, morphology of the crown is an advantage for *Tilia* genus when compared with other popular urban trees (Nowak, 2006). It is obvious that the heavy metal accumulation in plants varies depending on species, tissues and metals, but the findings shows that *Tilia* spp. have many of the requirements of a good bioindicator for heavy metals accumulation. Sjöman & Oprea (2010) reported that *Tilia tomentosa* Moench and *Tilia cordata* Mill. show long-lasting health and hardiness in street environments of Northern Europe.

Tilia species are hermaphroditic and mostly large, deciduous trees, reaching typically 20 to 40 m tall, with oblique-cordate (heart-shaped) leaves 6 to 20 cm across. In “Flora of Albania”, *Tilia* genus is represented by three species: *Tilia tomentosa* Moench. (Silver linden), *Tilia cordata* Mill. (Small-leaved linden), and *Tilia platyphyllos* Scop. (Large-leaved linden) (Qosja et al., 1992). The three of them are part of this study.

The aim of this study was to evaluate the concentration of heavy metals in the leaves of *Tilia tomentosa* Moench, *Tilia cordata* Mill. and *Tilia platyphyllos* Scop., which grow in different urban traffic sites of Tirana city.

Material and methods

Aiming to evaluate the potential pollution of urban air, three sampling locations were selected, based on different traffic intensity. Accordingly, the first station represents an intersection, with 2-3 lanes in each side (qualitatively evaluated with the heaviest vehicle traffic pollution). This intersection is planted in both sides of the road with silver linden - *Tilia tomentosa* Moench. The second station, represents one of the biggest road intersections in the center of Tirana, with 3-4 lanes in each side (qualitatively evaluated with medium level of vehicle traffic pollution). In the middle of this intersection originates Lana River, and the both sides are planted with small-leaved linden - *Tilia cordata* Mill. and other vegetation. The third station is located in the biggest park of Tirana, a park only for the movement of pedestrians and not cars, but very close to the roads with heavy traffic (qualitatively evaluated with the lowest level of pollution). In this station the large – leave linden - *Tilia platyphyllos* Scop. was selected for the study. The map of sampling stations is

presented in Figure 1. The identification of *Tilia* species was done by consulting the second volume of “Flora of Albania” (Qosja et al., 1992). Collection of leaf samples was carried out during three seasons of year 2021, in spring (second week of May), in summer (second week of July) and in fall (second week of October).

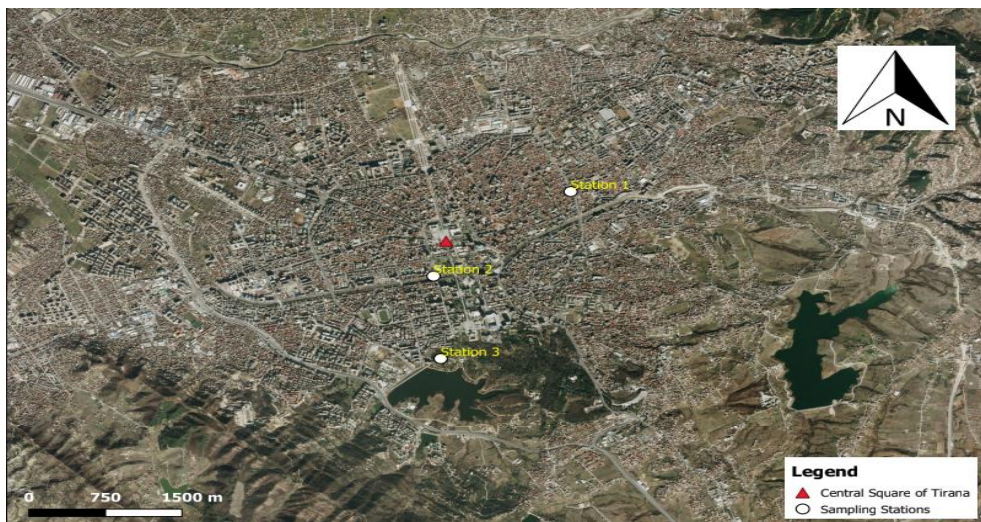


Figure 1: Sampling locations of Lime spp., tree leaves in Tirana city
(Station 1- *Tilia tomentosa* Moench, Station 2- *Tilia cordata* Mill.,
Station 3 - *Tilia platyphyllos* Scop.)

After collection, leaves were immediately placed in plastic bags and were imparted to the laboratory. Upon the arrival, leaves were washed with deionized water and were completely dried at $105\pm 5^{\circ}\text{C}$, then grinded, homogenized and placed in polypropylene bags with closure and accompanied with the corresponding labels.

Determination of metals concentration in selected samples first started by samples digesting in aqua regia ($3\text{HCl}:1\text{HNO}_3$) followed by the measurements of the diluted solutions using atomic absorption spectroscopy technique. Digestion was carried out in open glass vessels, where about 0.5 gram of each sample was treated with 10 ml of aqua regia and left overnight at room temperature covered with watch glasses. Then, the temperature was raised up to 200°C for three hours. After three hours, vessels were opened and solution was boiled until a wet salt was formed, cooled, diluted to 50 ml with deionized

water and then filtered (a micro-porous membrane with the pore size of 0.45 μm) into a PET bottle. Determination of metal concentration in solution continued using the atomic absorption spectroscopy technique with electrothermal atomization, Analytik Jena NovAA400.

Calculation of metal concentration was carried out by the calibration curves of each element while for quality control of results, blanks, replicates and certified reference materials were also measured in parallel with our samples. Statistical treatment of the obtained results was carried out by using MINITAB 19, EXCEL, etc.

Results and discussion

The concentrations of heavy metals in trees leaves and their descriptive analysis are given in Table 1 and Table 2.

Table 1. Concentration of heavy metal in lime trees (*Tilia spp.*) in mg/kg

Station	Species	Season	Pb	Cd	Cr	Cu	Al	Ni	Fe	Mn	Zn
S1	<i>Tilia tomentosa</i> Moench	Spring	1.34	0.19	1.08	31.74	250.0	9.53	409.9	29.24	42.03
	<i>Tilia tomentosa</i> Moench	Summer	1.28	0.15	1.68	15.27	355.1	14.93	569.3	65.68	77.49
	<i>Tilia tomentosa</i> Moench	Autumn	2.58	0.15	3.95	10.52	532.9	15.29	792.6	63.06	99.77
S2	<i>Tilia cordata</i> Mill.	Spring	1.29	0.18	1.22	17.46	236.2	9.99	466.8	49.33	54.57
	<i>Tilia cordata</i> Mill.	Summer	1.05	0.15	1.7	16.5	294.1	8.89	616.9	29.2	76.9
	<i>Tilia cordata</i> Mill.	Autumn	1.31	0.19	3.41	4.15	369.6	10.13	713.5	40.86	69.5
S3	<i>Tilia platyphyllos</i> Scop.	Spring	0.97	0.12	0.82	9.91	299.9	8.70	599.9	50.74	51.21
	<i>Tilia platyphyllos</i> Scop.	Summer	0.99	0.13	2.36	7.39	289.9	7.55	381.7	54.93	97.77
	<i>Tilia platyphyllos</i> Scop.	Autumn	0.7	0.15	2.74	3.17	326.9	8.08	604.3	61.07	35.95

WHO safe limits (2006)	2.00	0.02	1.3	10.00	200	10.00	na	300	150
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Table 2. Descriptive statistics of metals in leaves

	Pb	Cd	Cr	Cu	Al	Ni	Fe	Mn	Zn
Mean	1.28	0.16	2.11	12.90	328.3	10.34	572.8	49.35	67.24
Standard Error	0.18	0.01	0.36	2.91	29.43	0.94	45.04	4.58	7.66
Median	1.28	0.15	1.7	10.52	299.9	9.53	599.9	50.74	69.5
Standard Deviation	0.53	0.03	1.08	8.74	88.3	2.83	135.1	13.73	22.98
Minimum	0.7	0.12	0.82	3.17	236.2	7.55	381.7	29.2	35.95
Maximum	2.58	0.19	3.95	31.74	532.9	15.29	792.6	65.68	99.77
Count	9	9	9	9	9	9	9	9	9

Descriptive statistics results showed that the average concentration of metals in the leaves of selected trees followed the order: Fe>Al>Zn>Mn>Cu>Ni>Cr>Pb>Cd. Maximum concentrations of Fe (792.6 mg/kg), Al (532.9 mg/kg), Zn (99.8 mg/kg), Mn (65.68 mg/kg), Cu (31.74 mg/kg), Ni (15.29 mg/kg), Cr (3.95 mg/kg), Pb (2.58 mg/kg) and Cd (0.19 mg/kg) were observed in station 1 (represented by *Tilia tomentosa* Moench), mainly during autumn and spring. Except for Al and Mn for which the lowest concentration (250.0 mg/kg and 29.2 mg/kg) were observed respectively in stations station 1 (represented by *Tilia tomentosa* Moench) and station 2 (represented by *Tilia cordata* Mill.), all the other elements exhibited their lowest metal content in station 3 (represented by *Tilia platyphyllos* Scop.), mainly during summer and autumn.

In the following graph (Figure 2.), it has been shown the average concentration of heavy metals for the three seasons in each station.

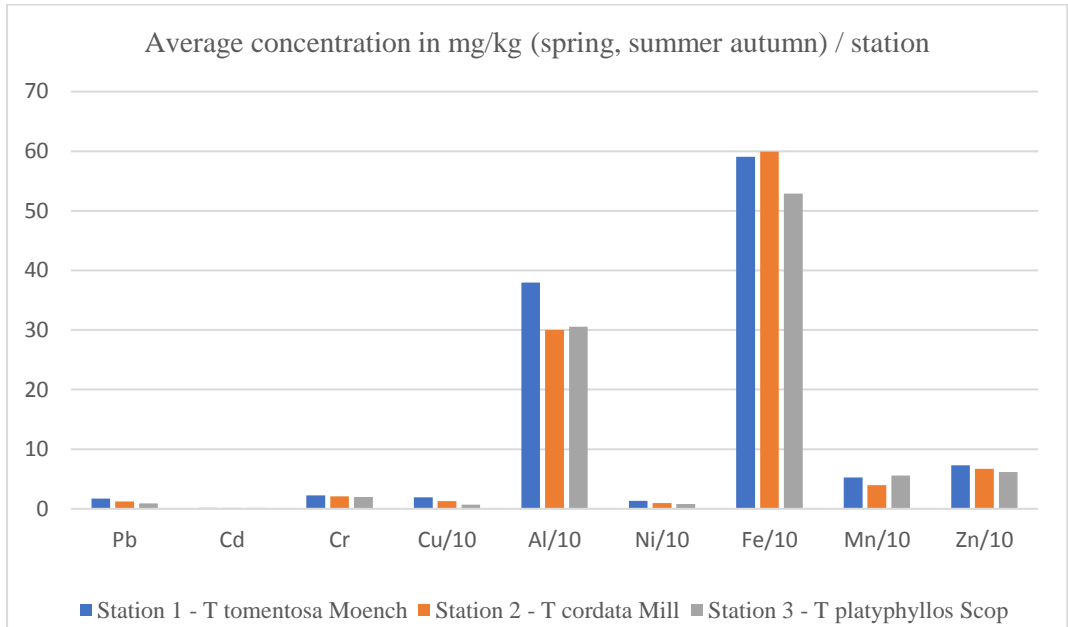


Figure 2: Average concentration of each element in all seasons / station

The highest average concentration of the following heavy metals Pb, Cd, Cr, Cu, Al, Ni and Zn was found in the leaves of *Tilia tomentosa* Moench, sampled in station 1, qualitatively evaluated with the heaviest vehicle traffic pollution. On the other hand, regarding Fe, the highest average concentration was measured in the leaves of *Tilia cordata* Mill. sampled in station 2, qualitatively evaluated with medium level of vehicle traffic pollution. In addition, for Mn, the highest average concentration was measured in the leaves of *Tilia platyphyllos* Scop., sampled in station 3, qualitatively evaluated with the lowest level of pollution.

Heavy metal concentration in plant leaves depends on many factors such as; metal concentrations in the soil, soil pH, solubility, soil type, organic matter content, cation exchange capacity, plant species, and plant age (Al-Heety et al., 2021). The interpretation of data related to accumulation of heavy metals from the air is difficult, even some metals could be used as micronutrients (Fe) or macronutrients (Cu, Zn) (Iordache, 2009). Iron (Fe), once deposited in the leaf tissue, is not readily re-translocated; hence, older leaves may have a relatively high concentration of Fe (Robb & Pierpoin, 1983), that explain the

reason why in the three stations chosen for this study, the concentration of Fe is very high during autumn season, when leaves have reached the maximum of their growth.

It is widely accepted that higher plants, including trees, take up elements mostly via the roots, although some uptake is considered possible through the leaves from atmospheric deposition (Shaid et al., 2017). The foliar uptake of elements and atmospheric origin has been clearly proven only for Pb (Hovmand et al., 2009). After the phasing out of leaded gasoline, lead (Pb) emissions to the atmosphere dramatically decreased, leading to low concentration of this element in dust particles. In our study, the average concentration of Pb has resulted almost lower than the limit value (2.0 mg/kg) in all samples, apart for *Tilia tomentosa* Moench, for which the autumn concentration of Pb is 2.58 mg/kg.

Zinc (Zn) as an essential micronutrient, it's involved in numerous aspects of cellular metabolism, such as influencing the activities of hydrogenase and carbonic anhydrase, stabilization of ribosomal fractions and synthesis of cytochrome (Tisdale et al., 1984; Classen et al., 2011). In the present study the concentration of Zn is inside the normal range (WHO, 2006) in the analyzed samples.

Copper (Cu) is considered as a micronutrient for plants and can accumulate considerable amounts under different natural and anthropogenic condition (Padmavathiamma & Li, 2007; Serbula et al., 2012). In our study, the concentration of Cu is higher than the safe limit (WHO, 2006) in station 1, represented by *Tilia tomentosa* Moench, during the three seasons (respectively 31.74 mg/kg, 15.27 mg/kg, 10.52 mg/kg) and in station 2, represented by *Tilia cordata* Mill., during spring (17.46 mg/kg) and summer (16.5 mg/kg). An excess of Cu in soil induces stress and causes injury to plants, thus playing a cytotoxic role. This leads to plant growth retardation and leaf chlorosis (Lewis et al., 2001).

Solid particles have a low concentration of cadmium (Cd) (Hu et al., 2014), which is the major reason why Cd concentrations in the plant samples are low. Typical concentrations of Cd in plants are less than 10 mg/kg (Tomasevic et al., 2004). In the present study, in all the samples, the concentration of Cd resulted to be very low and within the safe limits.

In this study all the samples analyzed have a higher concentration of chromium (Cr) than the accepted normal range, apart from the autumn season. Cr is usually known to have a low liability from soil to plants, but its content in plants is closely related to the concentration in soils. Based on the data published by Gjoka et al. (2011), the content of Cr in soils of Tirana city is 174.2 mg/kg, which is about twice of the value recommended by the WHO (1996).

High aluminum (Al) in plants is usually an indication of very low soil pH or poor soil aeration due to compaction or flooding (Ofoe et al., 2022). Concentration of Al resulted higher than the safe limits in all our samples, and even higher during autumn season, when the leaves are in the end of the life cycle.

Manganese (Mn) is an essential micronutrient with many functional roles in plant metabolism. Normal concentration of Mn in plants ranges between 20-300 mg/kg, while values below 15 mg/kg are considered deficient for plants (WHO, 1996). In the present study the concentration of Mn is inside the normal range for all the samples analyzed.

Nickel (Ni) is considered to be essential for plant growth at low concentration; however, Ni pollution is increasing in the environment, and therefore, it is important to understand its functional roles and toxic effects on plants (Liu et al., 2011). The concentration of nickel leaf samples was higher than the toxic concentration of 10 mg/kg, only in three samples, for *Tilia tomentosa* Moench (station 1), during spring and autumn, and for *Tilia cordata* Mill. (station 2), during autumn. In all the other samples the concentrations were within the normal values.

Conclusions

For the following heavy metals Pb, Cd, Cr, Cu, Ni, and Zn it is found a direct correlation between the average concentration on the leaves of the *Tilia* species and the level of vehicle traffic pollution. Meaning that the average concentration of these heavy metals was higher in the leaves of the *Tilia tomentosa* Moench plants located in a qualitatively evaluated higher vehicle pollution station.

An exception to this correlation was observed only for Al, Fe and Mn. Regarding Al, the average concentration measured in the leaves of *Tilia*

platyphyllos Scop. in station 3 (qualitatively evaluated with lowest vehicle traffic pollution) was slightly higher than the average concentration measured in the leaves of *Tilia cordata* Mill. located in station 2 (qualitatively evaluated with medium vehicle traffic pollution).

Regarding Fe, the average concentration measured in the leaves of *Tilia cordata* Mill. located in station 2 was higher compared with station 1 and 3. Whereas for Mn, the average concentration measured in the leaves of *Tilia platyphyllos* Scop. in station 3 was higher than the average concentration measured in the leaves of *Tilia tomentosa* Moench in station 1 and *Tilia cordata* Mill. sampled in station 2.

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