PROCEEDINGS OF THE LATVIAN ACADEMY OF SCIENCES. Section B, Vol. 69 (2015), No. 3 (696), pp. 87–97.
DOI: 10.1515/prolas-2015-0013

CHEMICAL COMPOSITION OF SCOTS PINE BARK AS A BIOINDICATOR OF ENVIRONMENTAL QUALITY IN RIGA, LATVIA

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Communicated by Pēteris Zālītis

The chemical composition of pine bark was used for the determination of environmental quality in Rīga. Bark samples were collected at 54 sites in Rīga differing in vegetation and building area characteristics (urban forests, parks and cemeteries, dwelling houses, and streets and railway zones) and at 52 sites from rural areas in Latvia (background level). Concentrations of Ca, Mg, Na, K, Fe, Mn, Zn, Ni, Cu, Cd, Pb, and pH was determined in the samples. The results showed significantly higher concentrations of chemical elements in pine bark collected in Rīga, compared to background levels: Fe and Cu concentrations three times higher, and other elements — up to 1.8 times. The highest element concentrations in Rīga were near streets (Sarkandaugava, Jaunmīlgrāvis, Daugavgrīva, Čiekurkalns, Imanta area, etc.). Element concentration and pH was significantly dependent on the distance from the closest street. The lowest element concentrations were found in urban forests (Jugla, Biķernieki, Beberbeķi) and parks and cemeteries (I Forest Cemetery, Jaunciema Cemetery). Mežaparks (forest), Sarkandaugava, Jaunmīlgrāvis, and Vecmīlgrāvis together formed the most polluted area in Rīga due to the close location near and downwind from the harbour.

Key words: Pinus sylvestris *L., bark, urban environment, chemical elements, pollution.*

INTRODUCTION

Atmospheric pollution is one of the major issues in the urban environment worldwide due to various anthropogenic activities, e.g., industry, vehicular traffic, etc. Chemical element concentrations in tree bark have frequently been successfully used to determine environmental quality and pollution levels, as well as transport of local pollution (Lötschert, 1983; Carneiro *et al*., 2011; Kord and Kord, 2011; Sawidis *et al*., 2011; Guéguen *et al*., 2012). Tree bark has structural porosity, and thereby retains pollutants for a long time without influencing the health status of trees (Berlizov *et al*., 2007; Sawidis *et al*., 2011). Thereby the transported chemical elements in the environment can accumulate in tree bark. Wide studies on bark chemical composition have been carried out during the second part of the 20th century, mainly in the vicinity of factories and in the urban environment (Grodzinska, 1971; Härtel and Grill, 1972; Westman, 1974; Löschert and Köhm, 1977; Grodzinska, 1978; 1982; Härtel, 1982; Grodzinska, 1984; Heniņa and Laiviņš, 1995). Studies on chemical composition of bark as an informative indicator of environmental status were also carried out in the 21st century (Harju *et al*., 2002; Saarela *et al*., 2005; Samecka-Cymerman *et al*., 2006; Kord and Kord, 2011; Sawidis *et al*., 2011; Rutkovska, 2014).

Scots pine (*Pinus sylvestris*) is one of the most frequent used model trees for assessment (Poykio *et al*., 2005; Saarela *et al*., 2005; Samecka-Cymerman *et al*., 2006; Marmor and Randlane, 2007; Melece *et al*., 2011). Its bark surface is very porous and the absence of metabolic processes in dead outer bark layer where airborne pollutants mainly accumulate makes it almost inert for organic and inorganic substances (Schulz *et al*., 1999; Harju *et al*., 2002).

In Latvia, regular studies on the chemical composition of pine bark in the urban environment started at the beginning of the 1990s of the 20th century in Jūrmala. The research results revealed that the chemical composition of pine bark could be used to describe the impact zone of industrial objects, private houses and streets in the city (Nikodemus *et al*., 1993). In subsequent years, studies of chemical composition of pine bark in the largest cities in Latvia (Daugavpils and the Mežaparks area in Rīga) showed that the typical chemical elements in the urban environment were zinc, copper, iron, and nickel, the associations of these elements with pollution sources (factories, harbour complex, transport nodes, etc.) were established, zoning was delineated based on the pollution levels (Dombrovska, 2011; Rutkovska, 2014). In the rural environment, a neutral bark reaction, as well as higher magnesium and iron concentration were observed in pine bark growing in the close vicinity of roads (Melece *et al*., 2011).

It is important to assess environmental quality, as the amounts and composition of industrial and transport emissions have changed during the last decades due to different environmental and EU regulations etc. In this study the chemical composition of pine bark was used as a bioindicator for the analysis of current environment quality in Rīga and Latvia. The objectives were: 1) to determine levels of macronutrients and heavy metals in Rīga; and 2) to evaluate the local and regional differences in the environmental quality of the city and countryside (background, rural environment).

MATERIALS AND METHODS

Study area. The study was conducted in Rīga (the capital of Latvia), which is situated near the Baltic Sea at the southern part of the Gulf of Rīga in the boreo-nemoral zone. The climate in Rīga is moderately warm and humid, the average annual precipitation is 708 mm, the average temperature in January is –3.0 ^{O}C, but in July +17.7 ^{O}C (Lizuma, 2000). Traffic intensity has increased in Rīga during the last decades, causing unsatisfactory low air quality (Anonymous, 2010).

Field work. To characterise environmental quality, pine bark samples were collected in 54 study sites in the Rīga area, and in 52 sites located in the rural environment or countryside forests in Latvia (background level or unpolluted area) in December 2013 and January 2014 (Fig. 1, Table 1).

In each study site three replicate pine bark samples (1 mm outer bark) were collected from about 5–10 pine trees around their stems at 1.5 m height in each urban sampling site, and from 10–15 pine trees in each rural (forest) sampling site. The study sites were classified in four groups according to their visual evaluation, the dominant vegetation type and build-up area nature: the study sites in the compact forest area (24 sites or 44%), parks and cemeteries (7 sites or 13%), dwelling house yards (10 sites or 19%) and near streets and railways (13 sites or 24%). Study sites were located 40–500 m from streets in urban forests, 50–310 m in parks and cemeteries, 15–160 m in housing areas, 2–40 m in street and railway zones, and least 200 m from roads in rural forest sites. Each bark sample was placed in a plastic bag and immediately transported to the laboratory.

The location of study sites in Rīga was irregular due to concentration of pine forest in the city periphery. In rural areas the study sites were selected in forests at least 150–200 m from roads and 1 km from buildings. The average distance bertween the study sites was 50–60 km and study sites were located throughout Latvia. To assess impact of pollution

Fig. 1. Location of research sites of pine bark in Rīga (A) and Latvia (B).

from Rīga on suburb areas, five sites were selected in the area near the Rīga border (Priedaine, Dārziņi, Dreiliņi, Berģi, Carnikava).

Laboratory analysis. The pine bark samples were dried at +60 ^{O}C, ground and dry-ashed in concentrated HNO_3 vapour and re-dissolved in HCl solution (HCl – distilled water mixture 3:100) (Rinkis *et al*., 1987). The levels of Ca, Mg, Fe, Cu, Zn, and Mn were determined by atomic absorption spectrophotometer (AAS) *AAnalyst 700* (PerkinElmer, Singapore), acetylene-air flame, Pb, Ni, and Cd determined using a graphite furnace (Page *et al*., 1982; Anonymous, 2000). K and Na were determined with a flame photometer JENWAY PFPJ (Jenway Ltd, Gransmore Green, Felsted Dunmow, Essex, UK). Pine bark pH was determined in 1 M KCl (1 g pine bark : 20 ml 1 M KCl) using a pH-meter Sartorius *PB-20* (Sartorius AG, Goettingen, Germany) (Rinkis *et al.,* 1987). Analytical replication was performed three times.

Statistical analysis and cartographical material. The statistical analysis of the results was conducted using

Table 1

STUDY SITES IN RĪGA

Site	Site name	Habitat/ land use type	Site	Site name	Habitat/ land use type
1	Juglas forest	A/a*	28	Ziepniekkalns/ Medēmu bog	A/a
2	Biķernieku forest	A/a	29	Ziepniekkalns, Ozolciema/ Kazdangas Str.	B/a
3	Mežciems	A/a	30	Katlakalns	A/a
4	Sports complex in Biķernieku forest	B/d	31	Āgenskalna Pines, Āgenskalna Str.	C/c
5	Āgenskalns	C/d	32	Āgenskalns, Jaunatnes Garden	C/c
6	Bastejkalns	D/b	33	Jaunmīlgrāvis, Tvaika Str.	C/d
7	Berģi	B/d	34	Vecmīlgrāvis, Emmas Str.	C/c
8	Juglas Paper-Mill village	C (d)	35	Vecmīlgrāvis, park of Ziemeļblāzma	D/b
9	Brekši	A/a	36	Vecmīlgrāvja Cemetery	E/b
10	Markusa Cemetery	E/b	37	Rīnūži, Thermal Central	C/d
11	I Meža Cemetery	E/b	38	Mangaļu peninsula	A/a
12	Sarkandaugavas Cemetery	E/b	39	Čiekurkalns, Ezermalas Str.	C/d
13	Mežaparks, Catholic Church	B/a	40	Mežaparks, Visbijas Prosp.	C/c
14	Vakarbuļļi, forest dunes	A/a	41	Mežaparks, Janševska Prosp.	A/a
15	Rītabuļļi	B/a	42	Aplokciems, forest dunes	B/a
16	Garden of Bolderājas library	C/c	43	Vecāķi, forest dunes	A/a
17	Daugavgrīva, Ship-yard	C/d	44	Jaunciema Cemetery	E/b
18	Kleistu forest, dunes	A/a	45	Ozolkalni	A/a
19	Lāčupe	B/a	46	Dreiliņi, Baltinavas Str.	B/c
20	Imanta, Kurzemes Prosp.	C/d	47	Šmerlis	A/a
21	Imanta, Rigondas Str.	C/c	48	Čiekurkalns, Lake Bābelītes, forest dunes	A/a
22	Guberņciems, Bolderājas station	B/d	49	Sarkandaugava, Viestura Prosp. 69	C/d
23	Bolderājas New Cemetery/ gravel-pit	A/a	50	Sarkandaugava, Viestura Prosp. 73	C/c
24	Aniņmuižas Park	C/c	51	Sarkandaugava, Viestura Prosp. 61/71	C/d
25	Beberbeķu Nature Park (northern area)	A/a	52	Maršalkas	A/a
26	Beberbeķu Nature Park (southern area)	A/a	53	Rumbula	B/a
27	Mārupe, Kantora Str.	C/c	54	Rumbula, Maskavas Str.	C/d

* A – forest, B – light forest, C – group of pines, D – park, E – cemeteries; a – forest, b – cemeteries and parks, c – dwelling-house area, d – streets and railway zones. Str. – street, Prosp. – prospectus

Microsoft Excel 2013, RStudio, and *PC-ORD* software for multivariate analysis of ecological data. Means and standard errors (SE) were calculated. Correlation coefficients (Pearson) were classified as follows: $r < 0.5$ – weak correlation, $0.5 < r < 0.7$ – medium correlation and $r > 0.7$ – high correlation. The Student's t-test (Two-Sample Assuming Unequal Variances) was used for testing the differences between urban and rural sites, and between other groups of sites. Coefficient of variation (S) of element concentrations was calculated. To evaluate effect of auto transport emissions and traffic along streets in the urban environment, relationships distance to street and elements concetrations were calculated (logarithmic regression $y = b_0 + b_1 ln x_i$). To assess relationships between the element concentrations and pH of pine bark, principal component analysis (PCA) was conducted using PC-ORD Version 5 (McCune and Mefford, 1999).

To prepare the cartographical material the software Arc View 9.2 was used.

RESULTS

Concentration of chemical elements in Rīga and Latvia. Concentrations of most elements and their variance were higher in pine bark collected in Rīga in comparison those from other regions (Fig. 2). The largest differences between samples collected in Rīga and Latvia were found for concentrations of Fe and Cu. No significant differences were found Mn concentration. The pH of pine bark was slightly higher in Rīga compared with other regions. However, the highest pine bark pH and Mg concentration were found in site No. 6 (Tīreļi), located in a rural forest in Latvia.

Chemical composition diversity in the urban environment. In general, the chemical composition of pine bark in the urban area of Rīga varied between sampled groups of pines, particularly between urban forests, cemeteries and parks, dwelling houses, and street and railway zones (Table 2). The largest differences were found between urban forests and streets and railway zones in concentrations of elements and pH of pine bark. The highest variance was found for Na in samples of pine bark from streets and railway zones. The highest coefficient of variation in other sample groups were found for the heavy metals Fe (urban forests) and Cu (cemeteries, parks and dwelling houses).

Anomalies of chemical elements in urban environment. The highest concentrations for most of the studied chemical elements (Ca, Mg, Na, K, Fe, Ni, Zn, and Cu) occurred in samples collected along streets and railways in Rīga (Table 2, Fig. 3). Maximum Fe and Cu concentration was found in the Jaunmīlgrāvis (site 33) between Tvaika Street and the railway, K and Mg in Daugavgrīva (site 17) near Flotes Street, Ca near Ezermalas Street (site 39) close to the thermoelectrical power station (TEC 1), Na in a greenery midstreet strip located in Kurzemes prospect (site 20), Ni near the Bolderājas railway station (site 22), Zn in Āgenskalns near Baložu Street (near trolleybus station) (site

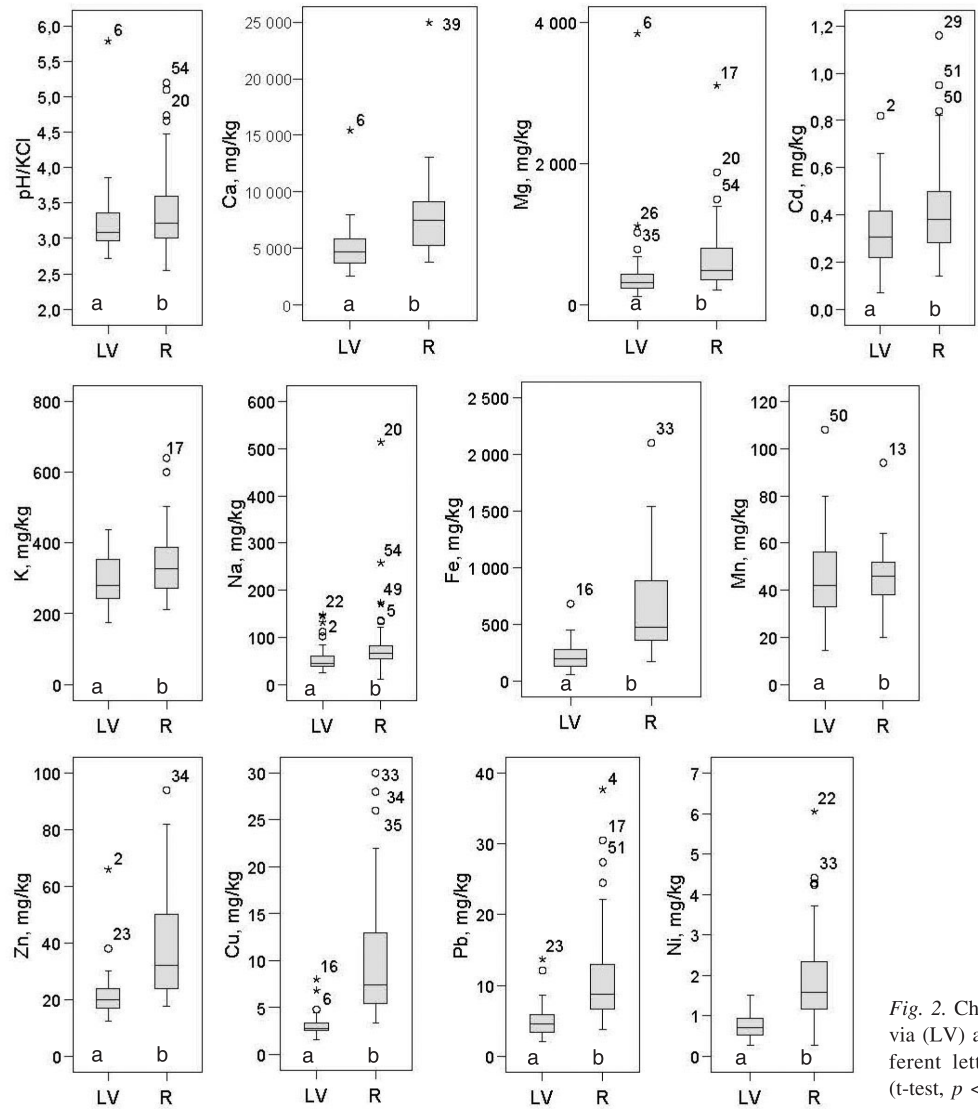

Fig. 2. Chemical composition of pine barks in Latvia (LV) and Rīga (R). (Means annotated with different letters (a, b) were significantly different (t-test, $p < 0.05$)).

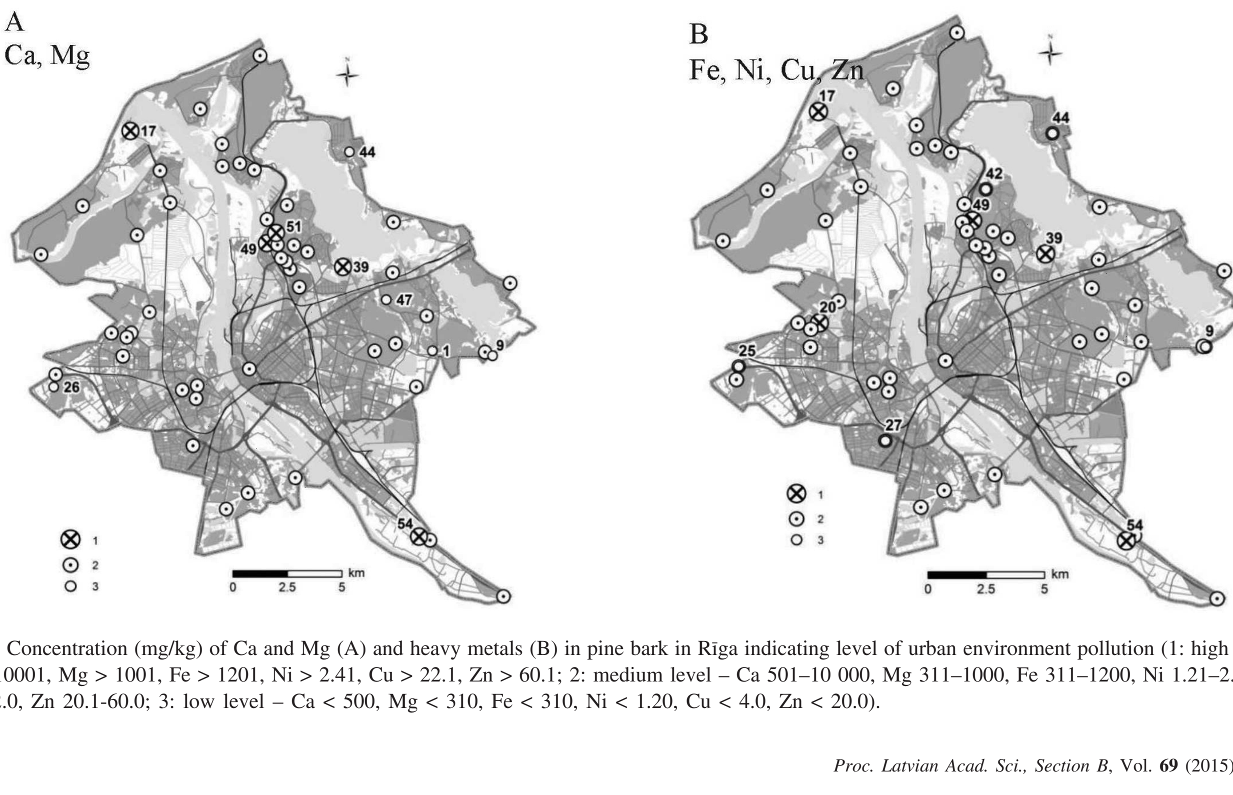

Fig. 3. Concentration (mg/kg) of Ca and Mg (A) and heavy metals (B) in pine bark in Rīga indicating level of urban environment pollution (1: high level – Ca > 10001, Mg > 1001, Fe > 1201, Ni > 2.41, Cu > 22.1, Zn > 60.1; 2: medium level – Ca 501–10 000, Mg 311–1000, Fe 311–1200, Ni 1.21–2.40, Cu 4.1–22.0, Zn 20.1-60.0; 3: low level – Ca < 500, Mg < 310, Fe < 310, Ni < 1.20, Cu < 4.0, Zn < 20.0).

Table 2

CHEMICAL COMPOSITION OF PINE BARK IN RELATION TO LAND USE IN RĪGA

Element/ parameter	Urban Forests	Cemeteries and parks	Dwelling-house area	Streets and railway zones
pH, mean ±SE	3.10 ± 0.07 a	3.27 ± 0.16 a	3.30 ± 0.09 ab	4.05 ± 0.21 c
Range	2.55 – 4.33	2.64 – 3.90	2.80 – 3.73	2.95 – 5.20
S%	12.3	13.0	8.8	18.6
mg/kg				
Ca, mean ±SE	^6282 ± 407 a	8155 ± 897 b*	7636 ± 612 b	10372 ± 1396 b*c
Range	3768 – 10800	4800 – 12040	3960 – 10200	4560 – 24960
S%	31.8	29.1	25.4	48.5
Mg, mean ±SE	448.3 ± 50.2 a	517.1 ± 101.8 a*	560.8 ± 36.5 ab*	1179.5 ± 200.1 c
Range	236 – 1212	220 – 948	320 – 696	320 – 3104
S%	54.9	52.1	20.6	61.2
K, mean ±SE	289.8 ± 10.9 a	349.1 ± 28.5 b	421.2 ± 16.4 c	398.8 ± 36.2 bc
Range	212 – 394	258 – 494	358 – 504	236 – 640
S%	18.4	21.6	12.3	32.8
Na, mean ±SE	60.7 ± 3.6 a	55.4 ± 9.1 a	88.2 ± 7.2 b	147.9 ± 34.7 c
Range	30 – 114	12 – 82	62 – 122	54 – 514
S%	28.9	43.8	26.0	85.2
Fe, mean ±SE	^477.3 ± 55.8 a	631.7 ± 88.8 b	587.4 ± 121.8 ab	1050.8 ± 149.9 c
Range	212 – 1380	368 – 1020	170 – 1240	270 – 2100
S%	57.3	37.2	65.6	51.4
Mn, mean ±SE	45.2 ± 3.0 a	46.0 ± 5.1 a	40.0 ± 3.3 a	47.2 ±3.3 a
Range	20 – 94	22 – 60	24 – 52	22 – 64
S%	32.3	29.2	26.0	25.0
Zn, mean ±SE	^27.98 ± 1.85 a	40.00 ± 5.49 b	39.60 ± 6.77 b	54.86 ±5.81 c
Range	17.8 – 50.0	18.0 – 58.0	20.0 – 94.0	19.2 – 82.0
S%	32.3	36.3	54.1	38.2
Cu, mean ±SE	^6.81 ± 0.61 a	12.11 ± 2.70 b*	9.82 ± 2.32 b	14.94 ± 2.14 b*c
Range	3.6 – 14.8	6.2 – 26.0	3.4 – 28.0	4.2 – 30.0
S%	44.1	59.0	74.6	51.7
Pb, mean ±SE	^8.74 ± 0.73 a	9.87 ± 1.18 a	8.85 ± 1.13 a	18.46 ± 2.74 b
Range	4.8 – 19.3	5.5 – 14.8	3.8 – 15.6	5.7 – 37.7
S%	40.8	31.6	40.5	53.5
Ni, mean ±SE	^1.43 ± 0.14 a	1.91 ± 0.22 b	1.67 ± 0.37 ab	2.97 ± 0.42 c
Range	0.60 – 3.72	0.93 – 2.55	0.27 – 4.28	0.67 – 6.05
S%	47.6	30.8	70.7	50.4
Cd, mean ±SE	^0.43 ± 0.04 a	0.42 ± 0.05 a	0.40 ± 0.07 b	0.43 ± 0.07 a
Range	0.19 – 1.16	0.18 – 0.52	0.14 – 0.84	0.15 – 0.95
S%	49.7	31.9	55.5	57.8

SE – standard error. Means annotated with different letters (a, b, c) were significantly different (t-test, $p < 0.05$), *specified differences in the 3rd or 4th column from previous equal; ^ - significantly higher compared with rural Latvia (background level) in Fig. 2; S – coefficient of variation

5), and Pb in Biķernieki forest in the narrow zone between Biķernieku Street and a motor road covered with asphalt in the Biķernieku Sports Centre (site 4).

The highest Mn and Cd concentrations occurred in forests: Mn in the Mežaparks forest area (site 41), and in sites located near Mežaparks – I Meža and Sarkandaugavas Cemeteries (sites 11 and 12). The maximum Cd concentration was found in a pine forest of Rīga (site 29).

Sorting by the five highest concentrations in Rīga (12 elements × five highest = 60 cases), in 47 cases (78% of the total) the highest values occurred near streets and railways. The most polluted forests were in Sarkandaugava along Viestura Prospect (site 49) and near the railway (site 51), in Daugavgrīva along Flotes Street (site 17), in Jaunmīlgrāvis between Tvaika Street and the railway (site 33), and in Imanta along Kurzemes Prospect (site 20).

In 10% of cases high element concentrations was found in yards of dwelling houses: in Sarkandaugava (site 50) and in Vecmīlgrāvis (site 34). Both these study sites were located 100–140 m from streets and railways with intensive traffic. However, in 8% cases, high element concentrations was found in forest areas. The most polluted forests were in Mežaparks (sites 13 and 41), Ziepniekkalns (site 29), and Lāčupe (site 19). Only in 4% of cases, high concentrations were found in parks and cemeteries: Ziemeļblāzmas Park and I Meža Cemetery (sites 35 and 11).

The lowest metal concentrations and highest pH in Rīga were in forest areas, parks, cemeteries, and between houses. The lowest Fe, Cu, and Pb concentrations were found in the Mārupe area between summer cottages near the river Mārupīte located in the southern part of Rīga (site 27). The lowest K and Zn concentrations occurred in Biķernieki cemetery near Juglas forest (site 1), Ni and Cd between dwelling houses in Imanta district in a site exposed to wind opened to winds (site 21). The minimum concentrations of the other element studied were found in the following sites: Na in Bastejkalns (hillock) located in Rīga's centre (site 6), Mg in Jaunciema Cemetery (site 44), Ca in Mežciems forest (site 3), Mn in Beberbeķi Nature Park (site 25). The most acid pine bark occurred in Mežaparks near the residential area of Aplokciems (site 42).

Sorting by the lowest five concentrations of elements,the most unpolluted sites were located in forest areas — 70% or 42 of 60 cases, e.g. in Jugla (sites 1, 3, 9), Beberbeķi (site 26) and Vakarbuļļi (site 14). 17% or 10 cases were found in residential areas of Mārupe (site 27) and in Imanta in an area with medium tall (five floor) buildings (site 21). 10% or 6 cases were in parks and cemeteries, e.g. Bastejkalns, (site 6) and Jaunciema Cemetery (site 44). Low chemical element concentrations were found only in 3% or 2 cases along streets and railways — Biķernieku Street (site 4) and Baložu Street (site 5) in Āgenskalns.

Correlations and associations of chemical parameters. To identify associations or clusters of chemical elements in bark from urban (Rīga) and rural forests, correlations between the element concentrations were calculated (Tables 3

Table 3

PERSON'S CORRELATION COEFFICIENTS BETWEEN PARAMETERS DETERMINED IN PINE BARK SAMPLES IN RĪGA

Parameter	pH	Ca	Mg	K	Na	Fe	Mn	Zn	Cu	Pb	Ni	Cd
pH	1.00											
Ca	0.62**	1.00										
Mg	**0.89****	0.54**	1.00									
K	0.50**	0.37**	0.62**	1.00								
Na	0.61**	0.43**	0.59**	0.51**	1.00							
Fe	0.64**	0.59**	0.63**	0.47**	0.44**	1.00						
Mn	-0.03	0.28*	-0.01	-0.08	-0.04	0.23	1.00					
Zn	**0.72****	0.63**	0.64**	0.57**	0.48**	**0.76****	0.05	1.00				
Cu	0.58**	0.48**	0.55**	0.50**	0.32*	**0.83****	0.08	**0.89****	1.00			
Pb	0.63**	0.29*	0.64**	0.33*	0.40**	0.56**	0.06	0.52**	0.54**	1.00		
Ni	0.58**	0.57**	0.53**	0.31*	0.41**	**0.76****	0.20	**0.75****	0.68**	0.43**	1.00	
Cd	0.01	0.39**	-0.01	0.04	0.01	0.34*	0.56**	0.30*	0.34*	0.09	0.25	1.00

n = 52; significance of correlation coefficients: * $p < 0.05$; ** $p < 0.01$; in bold – the highest correlations

Table 4

PERSON'S CORRELATION COEFFICIENTS BETWEEN PARAMETERS DETERMINED IN SAMPLES OF PINE BARK COLLECTED IN LATVIA AS BACKGROUND LEVEL

Parameter	pH	Ca	Mg	K	Na	Fe	Mn	Zn	Cu	Pb	Ni	Cd
pH	1.00											
Ca	**0.83****	1.00										
Mg	**0.92****	**0.79****	1.00									
K	0.07	0.09	-0.02	1.00								
Na	0.25	0.24	0.15	0.09	1.00							
Fe	0.50**	0.41**	0.41**	0.28*	0.29*	1.00						
Mn	-0.17	-0.26	-0.05	-0.23	-0.14	-0.32*	1.00					
Zn	0.24	0.12	0.18	0.05	0.48**	0.36**	-0.09	1.00				
Cu	0.38**	0.30*	0.33*	0.24	0.18	**0.76****	-0.11	0.41**	1.00			
Pb	0.10	0.03	0.08	0.04	0.35*	0.49**	-0.14	0.69**	0.45**	1.00		
Ni	0.31*	0.18	0.21	0.21	0.41**	0.70**	-0.12	0.45**	0.58**	0.64**	1.00	
Cd	0.14	0.14	0.16	-0.04	0.27*	0.09	0.25	0.58**	0.33*	0.41**	0.10	1.00

n = 54; significance of correlation coefficients: * $p < 0.05$; ** $p < 0.01$; in bold – the highest correlations and 4). The results showed two main associations of elements in pine bark of Rīga, and two in rural areas. The first association was stated between Fe, Ni, Cu, and Zn, where the highest correlation was found between Cu and Zn, as well as Cu and Fe ($r \geq 0.83$). Another association in Rīga was Mg, Zn and pH ($r \geq 0.72$). In samples collected from the rural environment, one association was pH, Ca and Mg, and the second — Fe with Ni and Cu ($r \geq 0.70$). The correlation of Fe, Ni, and Cu was weak, compared to element associations of Rīga. Generally, 78.8% of the calculated correlation coefficients were positive and significant in Rīga (65.2% – $p < 0.01$, 13.6% – $p < 0.05$). For rural samples, only 45.5% of the correlation coefficients were significant (31.8% – $p < 0.01$, 13.6% – $p < 0.05$), and one was negative (Fe-Mn).

Relation with distance from street. Higher concentrations of elements occurred in study sites located in Rīga close to streets with more intensive traffic and along railways (Table 5). Concentrations of nine of the elements (Ca, Mg, Na, K, Fe, Ni, Zn, Cu, and Pb) were significantly related to distance from a street ($p < 0.05$). Location near street particularly affected concentration of Mg, Ca, K, and Na, and pH in the pine bark. The heavy metals Cu, Fe, Ni, Pb, Zn had weaker correlation with distance from the street, compared with macroelements (Mg, Ca, K). The closest correlation was found for Zn and Fe.

Principal component analysis. Based on chemical composition, the PCA ordination separated pine bark from Rīga and from rural forest (Fig. 4). The first two components explained 67.18% of the total variance for the PCA for Rīga (Fig. 4A) and 55.21% of the total variance for the PCA of pine bark collected outside Rīga (Fig. 4B). The most important factors in the PCA of rural pine forest bark were Ca, Mg, Fe, Ni, Cu, Zn, and Pb with eigenvector values ranging from 0.618 (pH) to 0.821 (Fe). In the PCA of pine bark collected in Rīga eigenvector values for the variables were between 0.634 (Na) and 0.875 (Fe). In both PCA ordinations, 3 PCA components could be explained by element concentrations and pH: in Rīga 1) Fe, Cu, Zn, Ni, Ca; 2) pH, Mg, Na, K, Pb; and 3) Mn, Cd; and in rural areas 1) Fe, Cu, Zn, Ni, Pb; 2) pH, Mg, Ca; and 3) Mn. The study sites with higher levels of elements and pH for both PCA ordinations of Rīga and Latvia were located on the left side of the ordination, and lower concentrations (less polluted) on the right side.

Table 5

PARAMETERS OF THE REGRESSION EQUATION ($y = b_1 \ln(x_i) + b_o$) BETWEEN DISTANCE FROM STREET AND ELEMENT CONCENTRATION IN PINE BARK

Element	Regression coefficients		*p*-value	F-statistic	Standard error
	b_0	$b_1(\ln x_i)$			
Na	**210.38***	**-29.79**	.001	30.48	57.59
Ca	**12857.6**	**-1219.30**	.001	20.20	2895.00
Mg	**1588.45**	**-223.51**	.001	40.38	375.40
K	**495.28**	**-35.22**	.001	20.65	82.71
Mn	**46.93**	-0.51	0.681	0.17	13.09
Fe	**1271.17**	**-147.21**	.001	17.66	373.90
Ni	**3.41**	**-0.36**	.001	13.02	1.06
Cu	**16.90**	**-1.65**	0.006	8.07	6.19
Zn	**64.04**	**-6.19**	.001	15.85	16.60
Cd	**0.36**	0.02	0.414	0.68	0.21
Pb	**20.65**	**-2.25**	.001	15.28	6.14
pH	**4.51**	**-0.27**	.001	35.75	0.48

*bold – statistically significant coefficients

DISCUSSION

Concentration of chemical elements in Rīga and rural forests in Latvia and chemical anomalies. Our study revealed several differences in the chemical composition of pine bark between samples collected in Rīga and rural forests in Latvia. In general, the average concentrations of most of the elements, except Fe, Mn, Pb, in pine bark collected outside Rīga were at the same levels as those described by Melece *et al.* (2011) for the drainage basin of

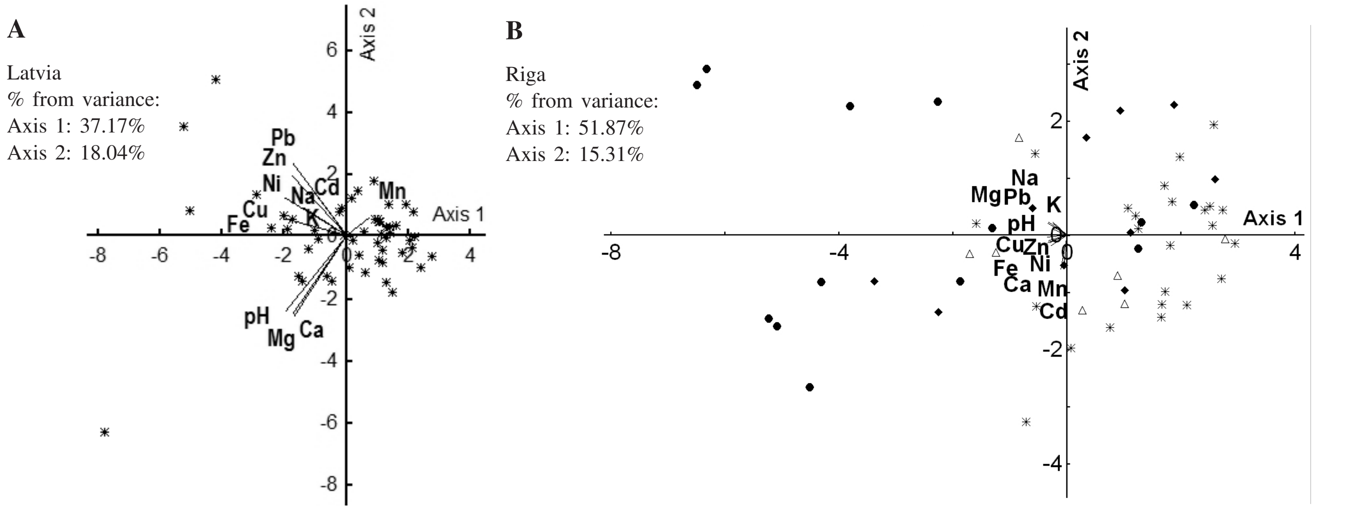

Fig. 4. Principal component analysis of chemical element results and pH of pine bark collected in rural forests in Latvia (A) and different land use types in Rīga (B).

Lake Engure. Mean Fe, Mn and Pb concentrations were slightly higher than those of pine bark collected in the drainage basin of Lake Engure (Fe 184.0 mg/kg, Mn 31.0 mg/kg, Pb 7.50 mg/kg). This might be due to soil factors (soil chemical composition and soil reaction) affecting element uptake by tree roots, or due to accumulation of dust particles from soil directly on tree bark. The chemical composition of soils in Latvia is more heterogeneous than within the drainage basin of Lake Engure. Soil type, soil pollution, and microclimatic conditions may differ between sampling sites and thus affect metal accumulation (Sawidis *et al*., 2011). Compared with concentrations in pine bark in Jūrmala region 20 years ago (Nikodemus *et al*., 1993), the concentrations of Ca, Zn, and Cd were similar, Mg and Mn — up to two times higher, and Fe, Pb, Cu, and Ni — 2–5 times lower, than in the rural forests in Latvia, 2013–2014. The higher pollution level in Jūrmala region might be explained by industrial activity and intensive transport during the Soviet Union period.

The concentrations of elements in pine bark samples collected in Rīga were generally higher than those collected outside Rīga. The effect of the urban environment on tree bark concentrations has been previously demonstrated (El-Hasan *et al*., 2002; Rossini Oliva and Mingorance, 2006; Kord and Kord, 2011; etc.), indicating its usefullness as a bioindicator. The largest differences between urban and rural environments (>2 times) were found for concentrations of Fe, Cu, Pb, and Ni. These differences can be explained by longer accumulation of pollutants in both soil and pine bark in the urban environment, especially for Pb. The main source of Pb in the environment of Latvia was intensive use of leaded petrol until the end of 20th century. Pb forms insoluble compounds in urban soil, which are not leached from the upper soil layer, which usually is the source of dust pollution and forms the uptake pool by plants. Our previous investigations showed elevated concentrations of Pb in street and park soils in Rīga even more than 10 years after lack of use of Pb in petrol (Cekstere and Osvalde, 2013). An especially high concentration of Cu and Fe in bark was found in site 33 (Jaunmīlgrāvis), located between a street and railway. This can be explained by wearing of train wires and rail and the accumulation of dust in the surrounding environment. Pine bark collected in rural and urban Latvia differed in ratio of concentrations of Fe and K. In rural areas the relative concentration of K, a plant macronutrient, was higher compared to the level of Fe, and in urban areas it was lower.

Surprisingly, there were no significant differences for the level of Mn between urban and rural environments, nor between different land use types in Rīga. Previous studies on pine bark in the Mežaparks area in Rīga (Dombrovska, 2011), an in central Rīga (Cekstere and Osvalde, 2013) showed elevated levels of Mn due vehicle transport. For example, in the world and probably also in Latvia, the use of fuel with antiknock agents containing Mn for motor vehicles can be one of the pollution sources of Mn (Zayed *et al*., 1999). However, our study did not show this tendency, possibly due to differences in the location and number of study sites.

A recent study on urban environment quality using pine bark was conducted by Rutkovska (2014) in Daugavpils, the second largest city in Latvia. The results showed that in the largest cities in Latvia — Rīga and Daugavpils — only the levels of Mn and Pb were similar between cities. For the majority of the studied elements (Ca, K, Zn, Fe, Cd, Na, Mg, and Cu), 1.3 to 3.1 times higher concentrations were found in Rīga. This suggested greater pollutant levels in Rīga. As an exception, the concentration of Ni was 1.8 times higher in Daugavpils. According to Rutkovska (2014), in Daugavpils, the metal concentrations in the pine bark were seriously affected by the Northern industrial zone. Studies in Finland (Saarela *et al*., 2005) have also shown significantly increased concentrations of several heavy metals in tree bark, e.g., Fe, Zn, Cu, Pb, and Ni, in the vicinity of a Cu-Ni smelter, e.g., mean Cu – 89 mg/kg, Fe – 147 mg/kg, Ni – 18 mg/kg, Pb – 9.1 mg/kg, and Zn – 43.4 mg/kg.

The pH of pine bark was slightly higher in Rīga compared to that of rural forests in Latvia. However, the highest pine bark pH, as well as of Mg, was found in site 6 (Tīreļi), located in a rural forest in Latvia. This can be explained by the type of road cover (dolomite stones), as sediment dust and aerosols near the roads contain Ca and Mg. These elements can accumulate in pine bark, leading to a higher pH. In Rīga, the correlation of bark pH with Ca concentration was weaker or medium.

Very acid pine bark was also found in several sites located in different areas in Rīga. The most acid pine bark (pH 2.55) was observed in Mežaparks near the residential area of Aplokciems (site 42). The pH of pine bark in rural Latvia and other countries (Samecka-Cymerman *et al.,* 2006; Melece *et al*., 2011; Rutkovska, 2014) was observed to be lower than in Rīga. During the last decades, industrial SO_2 emissions have decreased in Europe, and thus have decreased effect on bark pH (Oulehle *et al*., 2006; Jankovska *et al*., 2008). Aplokciems (site 42) is located near a harbour of Rīga, where coal is loaded in the open on cargo ships and other industrial activities are intensive. Thereby harbour operation can affect air quality in the surrounding areas of Sarkandaugava, Mežaparks, Vecmīlgrāvis and other areas in the direction of SW winds (dominant in Latvia) from the port.

Almost twice more significant correlations between chemical elements occurred for urban pine bark, compare to rural pine bark, indicating increased human effect in Rīga. Positive correlations between heavy metals have also been found in bark from pine (*Pinus eldarica* Medw) and cypress tree (*Cupressus semervirens* L.) in urban and industrial areas of Tehran (Iran) and Amman (Jordan), correspondingly (El-Hasan *et al*., 2002; Kord and, Kord, 2011). A significant positive, medium close correlation between Ca-Mg and Mg-K in pine bark from in Rīga and also Daugavpils (Rutkovska, 2014) suggested effect of road dust. The significant correlations between elements in pine bark in Rīga suggest a common source of origin for the metals. Accumulation of Ni, Zn, Cd, and Pb in the environment is ofter associated with traffic through the combustion of fossil fuel, use of anticorrosive materials and colours, and wear of asphalt, tyre rubber and steel surface (Narodoslawsky and Obernberger, 1996; Ozaki *et al.*, 2004; Fontenele *et al.*, 2010; Guéguen *et al.*, 2011).

Chemical composition diversity in the urban environment and PCA ordination results. The most significant differences in the chemical composition of pine bark between groups of trees and land use types was found for urban forests and street and railway zones. The highest values of elements showed that many of the polluted sites in Rīga were located on the right and left sides of the River Daugava. This could be explained by accumulation of chemical elements from local pollution sources, e.g., intensive traffic flow, the Rīga's harbour, industrial enterprises, thermal power stations, etc. It is in good agreement with studies from other cities. For example, the area of the industrial part of Amman City (Jordan) has high and variable Pb, Zn, Mn, Cr, Ni, and Co concentrations due to different intensity and types of anthropogenic activities, mainly, transport and industry (El-Hasan *et al*., 2002).

In Rīga the highest K concentrations were found in the sites located in areas of dwelling houses and near streets and railway zone. In Daugavpils (Rutkovska 2014), the highest K concentrations in *Pinus sylvestris* bark were found near a summer cottage area and in a cemetery. This suggests that K accumulation in the urban environment might be an indicator of gardening, fertilizer application and transportation, as well as KCl use as de-icing salt during winter.

The pH of pine bark collected from areas with different land use types in Rīga showed significant differences. Our findings differ from Tehran (Iran), where no significant variations were found in pH of *Pinus eldarica* Medw. bark between different land use types (Kord and Kord, 2011), as well as *Cupressus semervirens* L. bark in Amman (Tehran) (El-Hasan *et al*., 2002), which was attributed to a buffering effect of carbonate in the atmosphere originated from soil of area. In Tallinn (Estonia), traffic was observed to affect pH of *Pinus sylvestris* bark: in control plots mean pH (0.1 M KCl) was 3.0, while near roadways it was subneutral or up to 5.7 due to the dust pollution, which was a side effect of motor traffic, caused by wearing out of tires and asphalt, burning of fuel, and use of sand on roads in winter. Traffic can create air turbulence increasing air-borne road and soil dust (Anonymous, 2006; Marmor and Randlane, 2007). Various investigations in Latvia, e.g., Jūrmala (Nikodemus *et al*., 1993; Heniņa and Laiviņš, 1995), Rīga — Mežaparks (Dombrovska, 2011), Daugavpils (Rutkovska, 2014) and Finland – Harjavalt (Saarela *et al*., 2005) have found a higher concentration of Ca in *Pinus sylvestris* bark near roads with intensive traffic, and near industrial objects.

Yards in Rīga are enclosed between houses and thereby are mainly weakly ventilated. This promotes accumulation of various pollutants. Unfortunately, in such comparatively polluted yards, play grounds and recreation zones for inhabitants have been established, e.g., in Vecmīlgrāvis (site 34) and Sarkandaugava (site 50).

The PCA of chemical composition data also showed that the less polluted study sites were mainly located in areas of urban forests, cemeteries and parks, and around dwelling houses. The results of our study showed that wind was an important factor influencing urban air pollution distribution in Rīga. The most unpolluted sites, e.g., urban forest in Beberbeķi (site 26), Vakarbuļļi (site 14), residential area in Bieriņi (site 27) and Imanta (site 21), were mainly located in the periphery in the western and north western parts of Rīga, where air masses arise from unpolluted south-western and western rural and less urbanised areas, and from the Gulf of Riga. The most polluted study sites indicated in the PCA ordination (Tīreļi, Grobiņa, Dārziņi, Īvande) were located on the left side of the ordination space. These polluted areas were located in the direction of dominant winds (to the East) close to urban areas or about 2–3 km from reinforced concrete constructions. In both PCA ordinations (urban and rural areas) there were two main components affecting distribution of sites. The first main component was associated with heavy metal concentrations in pine bark and the second with dust pollution. However there were differences between the PCA ordinations due to different anthropogenic activities, which promoted formation, distribution and accumulation of air pollution in Rīga.

Relation with distance from street. Location near street significantly affected concentrations of several elements in pine bark in Rīga. The element concentration in pine bark was lower with decreased trafic intensity. At distances greater than 10 m from roads, the Na and Mg concentrations in pine bark were about 32% lower, Fe – 27%, Ca and Zn – 22%, and K – 16%. Accumulation of Mg, Ca, and K in pine bark occurred due to dust and aerosol pollution from roads, resulting in more neutral bark pH near streets. This is in good agreement with a study carried out in the rural environment by Melece *et al.* (2011), which showed a neutral bark reaction, as well as higher Mg and Fe concentrations, in pine bark close to roads. The distance from street in Rīga significantly affected Na concentration in pine bark due to application of de-icing salts on roads during winter season to prevent ice formation and to improve driving conditions (Čekstere *et al*., 2007; Cekstere and Osvalde, 2013).

In general, the results revealed significantly higher concentrations of the majority of the studied chemical elements and a higher coefficient of variation of concentrations in samples of pine bark collected in urban Rīga, in comparison with samples collected in other pine forests in Latvia. The largest differences were found for concentrations of Fe and Cu. The chemical composition of pine bark in the urban area of Rīga varied between sample groups according to type of land use: urban forests, cemeteries and parks, dwelling houses, as well as streets and railway zones. The largest differences were found between concentrations of chemical elements and pH of pine bark of urban forests and streets and railway zones. The lowest metal concentrations and more acid pine bark in Rīga were in forest areas, parks, cemeteries, and between houses. Two main associations between the chemical parameters in the pine bark collected in Rīga were observed. The first closest association was between Fe, Ni, Cu, and Zn, and the second between Mg, Zn and pH. Concentrations of nine elements (Ca, Mg, Na, K, Fe, Ni, Zn, Cu, and Pb) in pine bark collected in Rīga significantly depended on distance from street (p 0.05), and particularly regarding Mg, Ca, K, and Na, as well as pH. In urban and rural environments, the chemical composition of pine bark was mainly affected by three factor groups (PCA components). In Rīga: 1) Fe, Cu, Zn, Ni, Ca; 2) pH, Mg, Na, K, Pb; and 3) Mn, Cd; and in Latvia – 1) Fe, Cu, Zn, Ni, Pb; 2) pH, Mg, Ca; and 3) Mn.

ACKNOWLEDGEMENTS

The study was financially supported by the European Social Fund, project No. 2013/0060/1DP/1.1.1.2.0/13/APIA/VIAA/041.

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Received 24 March 2015

PRIEŽU MIZU ĶĪMISKAIS SASTĀVS KĀ VIDES STĀVOKĻA BIOINDIKATORS RĪGĀ (LATVIJA)

Vides stāvokļa novērtēšanai Rīgā kā bioindikators izmantots priedes mizas ķīmiskais sastāvs. Paraugi tika ievākti 54 vietās pilsētā atkarībā no veģetācijas un apbūves īpatnībām: urbānie meži, parki un kapsētas, dzīvojamās mājas un ielu malas un dzelzceļa zonas. Fona līmeņa raksturošanai izvēlētas 52 vietas Latvijā ārpilsētas mežos. Visos paraugos noteikta Ca, Mg, Na, K, Fe, Mn, Zn, Ni, Cu, Cd, Pb koncentrācija un pH. Rezultāti parādīja, ka Rīgā, salīdzinot ar fona līmeni, bija lielāka visu priedes mizā analizēto ķīmisko elementu vidējā koncentrācija. Fe un Cu koncentrācija pilsētvidē bija trīs reizes, bet pārējo ķīmisko elementu koncentrācija — līdz 1,8 reizes lielāka, salīdzinot ar lauku vidi. Pilsētvidē lielākās ķīmisko elementu koncentrācijas konstatētas gar brauktuvēm (Sarkandaugavā, Jaunmīlgrāvī, Daugavgrīvā, Čiekurkalnā, Imantā un citur). Konstatēts, ka ķīmisko elementu koncentrācijas būtiski izmainās, attālinoties no brauktuves. Viszemākās ķīmisko elementu koncentrācijas priedes mizā bija pilsētmežos (Jugla, Biķernieki, Beberbeķi), kā arī parkos un kapsētās (I Meža kapi, Jaunciema kapi). Tomēr Mežaparks (mežs) līdz ar Sarkandaugavu, Jaunmīlgrāvi un Vecmīlgrāvi bija stiprāk piesārņotais Rīgas rajons, jo tas atrodas ostas kompleksa un valdošo rietumu vēju tiešā ietekmes zonā.