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THE INFLUENCE OF CHEMICAL CHARACTERISTICS OF PRECIPITATION ON TREE HEALTH IN BANJICA FOREST (BELGRADE, SERBIA)

DOBRILA LUKIĆ[1*], D. KARADŽIĆ[2], M. RADOVANOVIĆ[3], M. MILENKOVIĆ[2], MIRJANA GAJIĆ[4], S. MILANOVIĆ[5] and JELENA KOVAČEVIC-MAJKIĆ[3]

[1*] *The Eighth Belgrade Gymnasium,* 11000 Belgrade, Serbia
[2] *Faculty of Forestry, University of Belgrade,* 11030 Belgrade, Serbia
[3] *Geographical Institute "Jovan Cvijić" SASA,* 11000 Belgrade, Serbia
[4] *Faculty of Geography, University of Belgrade,* 11000 Belgrade, Serbia
[5] *Institute of Forestry, University of Belgrade,* 11030 Belgrade, Serbia

Abstract - The most represented tree species in the Banjica Forest are *Acer negundo*, *Quercus robur*, *Acer pseudoplatanus*, *Populus nigra*, *Fraxinus pennsylvanica*, *Fraxinus ornus* and *Robinia pseudoacacia*. According to the ICP Forests combined assessment (degree of defoliation and decolorization), endangered species are *Populus nigra* (64.3% of heavily damaged trees), *Quercus robur* (45.5%), *Fraxinus pennsylvanica* (37.0%) and *Acer negundo* (26.6%), while the situation is much better for *Acer pseudoplatanus* and *Fraxinus ornus*. For *Robinia pseudoacacia,* 83% of trees are without decolorization, however, defoliation is established. In the period from April to October 2009, the average pH of rainwater was 5.46, and 5.18 in the period from November 2009 to March 2010. The concentration of SO_4^{2-} in the period from April to October 2009 amounted to an average of 24.21 mg/l, and 28.87 mg/l in the period from November 2009 to March 2010. The concentration of SO_4^{2-} and pH values is a possible explanation for the condition of the trees.

Key words: Rainwater, acid rain, pH, sulphate, Banjica Forest, Serbia

INTRODUCTION

"Banjica Forest" is located in Belgrade and is a natural memorial park (protected from 1993). The total area is 41.59 hectares (Management Unit "Banjica"), while the forest covers about 39 ha. The specificity of Banjica Forest is that it is completely surrounded by urban areas, and is also a habitat for about 70 species of birds. In 2005, signs of defoliation and decolorization were observed in a number of the trees in Banjica Forest. Conducted entomological and phytopathological analyses could not fully explain this phenomenon (Karadžić et al., 2007). Since the studied area is affected by a number of anthropogenic sources of pollution, it was presumed that this could be the framework for further research into the causes of the above-mentioned phenomenon. Therefore, research into the chemical characteristics of precipitation was undertaken. Primarily it was thought that the larger share of tree damage could be explained by so-called "acid rain" (pH below 5).

Bini and Bresolin (1998) emphasize the importance of anthropogenic components in the acidification of rainwater in northern Italy, where the average pH value is 5.2. Lara et al. (2001) found that anthropogenic activities (dust from the soil, burning of sugar cane and industrial emissions) affected the chemical composition of rainwater in the Piracicaba Basin in southeastern Brazil. The average pH value was 4.4 to 4.5, and the acidity of rain was significant at all tested points. However, Gregory et al. (1996) considered acidic pollutants to be only one of the causes of damage to forests, and their importance was greater in areas with strong local emissions. The author pointed out the importance of bad breeding measures and climate impacts. Cape (1993) states that the indirect influence of wet deposition on vegetation over processes in the soil is usual, but that leaves exposed to fog and clouds, which often contain much higher concentrations of pollutant ions than rain, can also be directly damaged. It has been established that there is a risk to vegetation in mountain areas, occurring near the base of the clouds, where the concentrations of ions are the highest.

The large number of trees with signs of damage, location of sites and the presence of pollution sources were taken into account in the case of Banjica Forest. The research objectives were: (i) to determine the degree of health deficiency of the most represented tree species on the investigated site, and (ii) to carry out chemical analysis of rainwater over a period of 12 months, and based on the results obtained, to assess the possibility of the impact of certain chemical characteristics on the forest trees.

MATERIALS AND METHODS

Methods based on visual assessment of the state of treetops were used to assess the health of the trees in Banjica Forest. In the studied area, 19 test areas were singled out, covering 15 m^2 each. All the trees were marked with numbers. Assessment of defoliation was carried out during June and July 2007, while the assessment of decolorization was performed in August of the same year. After that, in 2008 and 2009 a check of the state of trees was made, which showed that there were no changes. In this part of the research, methods in the ICP Forests program were used (Tables 1-3) (Innes, 1990). Assessments of defoliation and decolorization are shown as a percentage of the ideal treetop appearance for the observed species (Tables 1 and 2).

The examination also included the biological status of the trees in order to separate the impact of

Table 1. Assessment of defoliation

Assessment	Range	Described
0	0-10%	No defoliation
1	>10-25%	Weak defoliation
2	>25-60%	Moderate defoliation
3	>60-<100%	Severe defoliation
4	100%	Dried tree

Table 2. Assessment of decolorization

Assessment	Range	Described
0	0-10%	No decolorization
1	>10-25%	Weak decolorization
2	>25-60%	Moderate decolorization
3	>60%	Severe decolorization

Table 3. Combined assessment

Assessment	Range	Described
0	0-10%	No damage
1	>10-25%	Slightly damaged
2	>25-60%	Moderately damaged
3	>60-<100%	Severely damaged
4	100%	Dry

Table 4. Methods for determining ionic concentrations

ion	method	procedure
PO_4^{3-}	Standard colorimetry with ethanoyl chloride	Measuring was carried out at 690 nm, values were scored from calibration curve
SO_4^{2-}	Turbidimetric with barium chloride with standard curve	Measuring was carried out on spectrophotometer at 420 mμ
HCO_3^-	Volumetric with standard HCl solution with methyl orange as indicator	
NO_3^-	Method with brucine	Color intensity was measured by spectrophotometer at 410 mμ, and concentration was calculated from standard curve
NO_2^-	Standard with sulfanilic acid and α-naphthylamine	Color intensity was measured by colorimeter with the use of green filter, and concentration was being calculated from calibration curve
Cl^-	Volumetric according to Mohr	Titration was carried out by solution of silver nitrate with potassium chromate as indicator
$Mg^{2+} + Ca^{2+}$	Volumetric with complex III with indicator eriochrome black T	
NH^{4+}	Standard colorimetry with Nessler reagent	Concentration was being calculated from calibration curve
$Na^+ + K^+$	Calculation	All anions are calculated and expressed in milliequivalents, as well as cations Ca^{2+} and Mg^{2+}. The sum of all anions should be equal to the sum of all cations.

competition among trees from the influence of other factors on the appearance of the treetop.

Collecting samples of rainwater and snow water was done during the period 1 April 2009 – 31 March 2010. Analyses were undertaken at the Faculty of Chemistry in Belgrade. The pH and conductivity were examined, and methods for determining the concentrations of some ions in the samples are shown in the Table 4.

The results are shown by dates of sampling, as well as by periods: April-October (roughly coinciding with the vegetation period) and November-March (outside the vegetation period).

RESULTS AND DISCUSSION

Table 5 presents data on tree species, the presence of which is confirmed in the test areas.

The most represented species are *Acer negundo*, *Quercus robur, Acer pseudoplatanus*, *Populus nigra*, *Fraxinus pennsylvanica*, *Fraxinus ornus* and *Robinia pseudoacacia*. Other species comprise less than 5% of the share of the total number of labeled trees. Therefore, in this paper attention was focused primarily on the seven most represented species.

Data on the distribution of the seven most represented tree species according to the degree of defoliation (Table 6) indicate that the best situation is that for *Acer platanoides*, of which 85% did not exhibit defoliation. A good state was also recorded for *Acer pseudoplatanus*, 70% of which did not exhibit defoliation. However, there were no *Populus nigra* without defoliation: 57% have severe and 40% moderate defoliation. For *Robinia pseudoacacia*, most trees were with moderate (56%) and severe (22%) defoliation. *Quercus robur* is considered to be an endangered species, since 64% of the trees have moderate and 11% severe, defoliation. *Fraxinus pennsylvanica* had 63% with moderate defoliation, so that in this species, as in the past one, the transition of these trees to the group with severe defoliation can be expected. A similar, but somewhat more favourable situation was that of *Fraxinus ornus*, of which 46% of the trees have moderate defoliation.

Table 5. Number and presence of tree species in the test areas

Tree species	Number of analyzed trees	% of the total number of analyzed trees	Number of test areas in which species is present	% of the total number of test areas
Acer dasycarpum	7	2.1	2	10.5
Acer negundo	79	23.4	11	57.9
Acer platanoides	7	2.1	3	15.8
Acer pseudoplatanus	33	9.8	5	26.3
Aesculus hippocastanum	5	1.5	2	10.5
Ailantus glandulosa	6	1.8	2	10.5
Fraxinus excelsior	8	2.4	3	15.8
Fraxinus ornus	26	7.7	7	36.8
Fraxinus pennsylvanica	27	8.0	8	42.1
Gleditchia triacanthos	8	2.4	3	15.8
Juglans regia	8	2.4	5	26.3
Platanus orientalis	1	0.3	1	5.3
Populus nigra	28	8.3	5	26.3
Prunus sp.	3	0.9	3	15.8
Quercus robur	55	16.3	6	31.6
Robinia pseudoacacia	18	5.3	6	31.6
Sambucus nigra	5	1.5	3	15.8
Tilia sp.	6	1.8	3	15.8
Ulmus sp.	7	2.1	4	21.1
total	337	100.0		

Table 6. Distribution of trees by degrees of defoliation (%)

Tree species	Total number of investigated trees	Degrees of defoliation				
		0 %	1 %	2 %	3 %	4 %
Acer dasycarpum	7	14.3	28.6	42.9		14.3
Acer negundo	79	25.3	24.1	41.8	7.6	1.3
Acer platanoides	7	85.7		14.3		
Acer pseudoplatanus	33	69.7		24.2	3.0	3.0
Aesculus hippocastanum	5	100.0				
Ailantus glandulosa	6	16.7		66.7		16.7
Fraxinus excelsior	8		12.5	62.5	25.0	
Fraxinus ornus	26	23.1	23.1	46.2	7.7	
Fraxinus pennsylvanica	27	11.1	22.2	63.0	3.7	
Gleditchia triacantos	8	12.5		62.5	12.5	12.5
Juglans regia	8	25.0	37.5	37.5		
Platanus orientalis	1	100.0				
Populus nigra	28		3.6	39.3	57.1	
Prunus sp.	3			66.7		33.3
Quercus robur	55	5.5	14.5	63.6	10.9	5.5
Robinia pseudoacacia	18	5.6	11.1	55.6	22.2	5.6
Sambucus nigra	5	20.0		60.0	20.0	
Tilia sp.	6	100.0				
Ulmus sp.	7	42.9		28.6	28.6	

Table 7. Distribution of trees by degrees of decolorization (%)

Tree species	Total number of investigated trees	Degree of decolorization			
		0 %	1 %	2 %	3 %
Acer dasycarpum	7	85.7	14.3		
Acer negundo	79	30.4	17.7	40.5	11.4
Acer platanoides	7	57.1			42.9
Acer pseudoplatanus	33	90.9	6.1	3.0	
Aesculus hippocastanum	5				100.0
Ailantus glandulosa	6	100.0			
Fraxinus excelsior	8	87.5	12.5		
Fraxinus ornus	26	96.2	3.8		
Fraxinus pennsylvanica	27	33.3	14.8	48.1	3.7
Gleditchia triacantos	8	87.5		12.5	
Juglans regia	8	75.0	12.5	12.5	
Platanus orientalis	1				100.0
Populus nigra	28	32.1	10.7	57.1	
Prunus sp.	3	100.0			
Quercus robur	55	20.0	20.0	49.1	10.9
Robinia pseudoacacia	18	83.3	11.1	5.6	
Sambucus nigra	5	60.0		40.0	
Tilia sp.	6	50.0	16.7	33.3	
Ulmus sp.	7	100.0			

Table 8. Distribution of trees according to the degrees of combined assessment

Tree species	Total	Degrees of combined assessment				
		0 %	1 %	2 %	3 %	4 %
Acer dasycarpum	7	42.9	28.6	14.3		14.3
Acer negundo	79	16.5	24.1	31.6	26.6	1.3
Acer platanoides	7	42.9	14.3	42.9		
Acer pseudoplatanus	33	66.7	27.3	3.0		3.0
Aesculus hippocastanum	5			100.0		
Ailantus glandulosa	6	16.7	66.7			16.7
Fraxinus excelsior	8	12.5	62.5	12.5	12.5	
Fraxinus ornus	26	46.2	42.3	11.5		
Fraxinus pennsylvanica	27	14.8	29.6	18.5	37.0	
Gleditchia triacantos	8		75.0	12.5		12.5
Juglans regia	8	50.0	37.5	12.5		
Platanus orientalis	1			100.0		
Populus nigra	28	3.6	3.6	28.6	64.3	
Prunus sp.	3		66.7			33.3
Quercus robur	55	1.8	16.4	30.9	45.5	5.5
Robinia pseudoacacia	18	16.7	50.0	11.1	16.7	5.6
Sambucus nigra	5	20.0	40.0		40.0	
Tilia sp.	6	66.7	33.3			
Ulmus sp.	7	42.9	28.6	28.6		

Table 9. Mean daily air temperature, precipitation, pH, conductivity and ion content in mg/l by dates (April 2009 – March 2010)

Date of sample	Avg Tmp	RR	pH	Cond.	Cl^-	Ca^{2+}	Mg^{2+}	HCO_3^-	NH_4^+	NO_2^-	PO_4^{3-}	SO_4^{2-}	NO_{3-}	$Na^+ + K^+$
7/5/2009	17	0.4	5.3	68.5	0.16	0.25	0.2	0.4	0.3	0.07	0.003	25.41	11.76	37.28
23/5/2009	22.6	10.1	6.1	103.6	0.6	0.25	0.25	0.8	0.26	0.08	0.39	25.49	19.85	46.24
25/5/2009	23.5	7	5.61	67.6	0.53	0.3	0.22	0.8	0.16	0.07	0.27	23.52	0.73	25.33
28/5/2009	15	0.1	5.05	63.1	0.51	0.2	0.15	0.4	0.11	0.02	0	25.49	0	25.69
1/6/2009	16.6	5.5	4.89	44.8	0.4	0.22	0.13	0.8	0.63	0.02	0	23.52	0	24.1
2/6/2009	14	13.6	5.75	50.9	0.4	0.15	0.3	0.9	0.53	0.06	0	21.56	0	22.31
5/6/2009	18.4	2.6	5.53	39.6	0.64	0.1	0.1	0.8	0.55	0.02	0	25.49	0	26.73
20/6/2009	23	20.7	5.59	49	0.3	0.15	0.05	0.6	2.1	0.03	0.37	25.51	13.23	38.99
21/6/2009	14.5	4.2	5.31	47.9	0.32	0.12	0.03	0.8	1.03	0.07	0	25.49	0	26.46
25/6/2009	18.6	6.5	4.65	24.6	0.5	0.07	0.13	0.4	0	0.47	0	25.49	0	26.19
26/6/2009	18.8	18	5.4	22.2	0.28	0.1	0.07	0.6	0	0.03	0	25.49	0	26.2
28/6/2009	20.3	0	5.21	27.5	0.34	0.12	0.08	0.6	0	0.07	0	21.56	0	22.3
2/7/2009	23	0.9	5.87	33.9	0.36	0.07	0.08	0.4	0	1.43	0.72	23.52	8.08	32.21
17/7/2009	27	4.5	5.08	29.8	0.32	0.1	0.1	0.5	0.96	0.28	2.22	7.84	11.76	20.22
7/8/2009	22.2	21.8	5.25	42	0.3	0.07	0.08	0.6	1.03	0.08	0	25.49	11.75	37.99
13/8/2009	24.4	0.6	6.05	20.9	0.02	0.12	0.05	0.4	0	0.07	0	17.64	0	11.89
18/10/2009	6	1.8	5.56	53.6	0.28	0.1	0.1	0.3	1.97	0.32	0.16	21.56	0	21.94
2/12/2009	7.4	1.8	4.77	48	0.3	0.25	0.2	0.6	0	0.44	0	23.55	0	24
4/12/2009	7	0.2	5.21	37	0.23	0.22	0.2	0.8	0	0.02	0	24.59	0	25.2
17/12/2009	-3.2	3.4	5.1	39.76	0.38	0.19	0.14	0.6	0	0.66	0	21.44	0	21.85
18/12/2009	-6.2	6.6	4.78	44.34	0.33	0.12	0.03	0.6	0.12	0.01	0.5	22.52	5.1	28.4
21/12/2009	-2.8	0.1	5.51	36.7	0.4	0.12	0.03	0.6	0.23	0.07	0.6	22.52	11.32	34.54
8/1/2010	8.2	2.1	5.15	34.56	0.4	0.12	0.03	0.4	0.22	0.08	0.12	25.49	12.02	27.34
9/1/2010	10.7	0.1	5.12	47.87	0.21	0.06	0.07	0.5	0.99	0.05	0.33	21.43	7.9	29.91
27/1/2010	-5.9	1.5	5.65	56.7	0.24	0.05	0.06	0.4	0.78	0.04	0.37	22.5	11.72	34.75
28/1/2010	-6.6	5.3	5.22	65.45	0.18	0.2	0.15	0.9	0.98	0.03	0.1	25.41	8.9	32.05
6/2/2010	-0.6	2	5.31	44.32	0.4	0.3	0.22	0.4	1.2	0.07	0.02	23.55	15.4	39.25
13/2/2010	-1	18.5	5.21	41.23	0.25	0.3	0.22	0.6	1.88	0.3	0.003	19.89	12.4	32.89
24/2/2010	11.5	0.3	4.79	44.12	0.11	0.25	0.2	0.6	0.69	0.25	0.04	21.36	11.55	33.17
26/2/2010	9.7	8.6	5.23	22.4	0.19	0.15	0.04	0.5	0.58	0.04	0.07	22.51	11.3	34.31
27/2/2010	6.9	6.1	4.89	21.34	0.31	0.06	0.12	0.8	0.49	0.04	0.39	24.59	10.8	36.13
1/3/2010	9.1	0	5.12	33.1	0.32	0.05	0.05	0.4	0.87	0.06	0.34	20.81	9.6	30.41
4/3/2010	3.7	0	5.43	38.22	0.29	0.4	0.09	0.5	0.68	0.01	0.33	21.17	7.1	27.96
11/3/2010	1.1	9.2	5.65	37.1	0.26	0.03	0.2	0.2	0.81	0.02	0.32	23.55	4.66	28.44

Note: The data on the mean daily air temperature and precipitation are given according to the Republic Hydrometeorological Institute of Serbia. The precipitation value deviations occurred due to differences in the time of sampling and different locations.

Table 10. Average values of ions (mg/l) in rainwater for the periods April - October 2009 and November 2009 – March 2010

period	pH	Conductivity	Cl^-	Ca^{2+}	Mg^{2+}	HCO_3^-	NH_4^+	NO_2^-	PO_4^{3-}	SO_4^{2-}	NO_3^-	Na^++K^+
April October	5.46	46.44	0.02	0.14	0.12	0.59	0.56	0.18	0.24	24.21	4.53	27.76
November March	5.18	40.71	0.28	0.16	0.12	0.55	0.61	0.14	0.14	28.87	8.51	30.62

Regarding decolorization (Table 7), the most unfavorable situation was that for species with a large number of trees with moderate decolorization. These were *Populus nigra* (57%), *Quercus robur* (49%), *Fraxinus pennsylvanica* (48%) and *Acer negundo* (40%). A more favourable condition was that for species for which the majority of trees were without decolorization, namely *Fraxinus ornus* (96%), *Acer pseudoplatanus* (91%) and *Robinia pseudoacacia* (83%).

Combined assessment includes defoliation, which is permanent, and decolorization, which is more a result of the seasonal influences of biotic (phytopathogenic fungi and injurious insects) and abiotic factors (lack of moisture, extreme temperatures). According to the combined assessment (Table 8), endangered species are *Populus nigra* (64.3% of heavily damaged trees), *Quercus robur* (45.5%), *Fraxinus pennsylvanica* (37.0%) and *Acer negundo* (26.6%). The situation for the species *Acer pseudoplatanus* (66.7% of trees without damage) and *Fraxinus ornus* (46.2%) was much better. A significant share of trees without damage could be seen in the less represented species of *Tilia* spp. (66.7%) and *Juglans regia* (50%).

Regarding the test areas, it can be concluded that both defoliation and decolorization are fairly evenly represented throughout the studied area. Due to the cumulative effect of moderate defoliation and moderate decolorization, severe treetop damage was recorded in seven of the test areas.

Research results related to precipitation are shown in Tables 9 and 10.

In 6 cases out of 34, individual samples (Table 9) had a pH below 5. The lowest recorded value was 4.65 (25 June 2009), while the highest value of 6.1 was recorded on 23 May 2009. Extreme values appeared at an interval of 33 days.

In the studied area, the dominant anion is SO_4^{2-} (Tables 9 and 10). Türküm et al. (2008) found that significant amounts of SO_4^{2-} arrive to Turkey from northern Europe, Ukraine, Russia and some parts of the Balkans. Başak and Alagha (2004) state that the average pH of rainwater in the area of Istanbul is 4.81 and the main cause is sulfur emissions. Millet et al. (2001) found for Thann (Alsace, France) a pH in the range of 3.60 to 6.58, while in Tours (Indre et Loire, France) values exhibited a narrow range (5.49 – 7.01). In both cases, the dominant anion was SO_4^{2-}. Momin et al. (2005) found surprisingly high concentrations of SO_4^{2-} in a rural area of India during the monsoon season. Akoto et al. (2011) found that the pH of rainwater in a mining area in Ghana was between 4.0 and 5.6, where the main anions were SO_4^{2-} and Cl^-. Leal et al. (2004) studied the chemical composition of rainwater in Sao Paulo (Brazil), where the largest contribution to free acidity was from SO_4^{2-} (28.8%). Based on research in northeastern Uruguay, Zunckel et al. (2003) stated that the presence of NO_3^- and SO_4^{2-} in rainwater was characteristic for agricultural areas. The link between damage to treetops and SO_2 concentration was determined for spruce in northern Bohemia (Ardo et al., 2000), and for *Pinus brutia* and *Pinus nigra* in the area around Izmir (Turkey) (Kantarci, 2003).

High concentrations of Na^+ and K^+ were detected in the studied area, but the question arises as to their origin.

It can be seen in Table 10 that the reaction of atmospheric water in both tested periods was below average (pH = 5.7). Higher values than the average ones are stated by Topcu et al. (2002), who found that in the vicinity of Ankara (Turkey) the average pH value of rainwater was 6.3 – 7.0. Only about 4% of the rainwater samples had a pH below 5.0, while in only 15% the pH was lower than 5.6.

The slightly lower pH in the examined area during the winter period (Table 10) could be explained by anthropogenic influence, i.e., an increased consumption of fossil fuels. Sulfurous, sulfuric and hydrochloric acid, as well as the hydrolysis of ammonium sulfate, are important for the reduction of the pH of rainwater. The importance of sulfuric acid was confirmed by this research, given the high values obtained for the SO_4^{2-}.

Although the results obtained indicate the anthropogenic influence, it should be noted that the lowest pH (4.65) was found in early summer (25 June 2009), when the consumption of fossil fuels decreases and the number of people is lower compared to the rest of the year due to the beginning of the holiday season. Research results in other states also indicate the complexity of problems. Sanusi et al. (1996) investigated the chemical composition of rainwater in eastern France and found lower pH values (4.4) in rural compared to urban areas (Strasbourg 5.0 and Colmar 5.7); they explained this by the presence of $CaCO_3$ in the loess, which is the main constituent of the land in this area. Arsene et al. (2007) found that the average pH value of rainwater in northeastern Romania was 5.92 and that the main neutralizers were $CaCO_3$ and NH_3. Lara et al. (2010) found that the average pH value in Monterrey, an industrial centre in northeastern Mexico, was 6.58, which they explained by alkalization affected by high concentrations of Ca^{2+} and Mg^{2+}.

These results indicate the need to reduce emissions of pollutants. The example of Portugal should be kept in mind, where the reduction of SO_2 emission came from the reduced quantity of sulfur in fuels (Santos et al., 2011). One should also examine the possibility that rainwater is a source of nutrients for plants, which is indicated by the results obtained in the region of Rio Grande do Sul, Brazil (Calil et al., 2010).

The results presented in this paper highlight the complexity of the problem and pose the question of the origin of pollutants. Bearing in mind the results of Radovanović et al. (2003), Milovanović and Radovanović (2009) and Stanojević (2010), the impact of regional atmospheric circulation on this phenomenon should also be investigated. However, the impact of the chemical characteristics of precipitation in the area of Banjica Forest will only be fully understood if long-term research is continued.

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