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Impact of closed Kairiai landfill on the Ginkūnai Pond, Lithuania

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Abstract

The main goal of this study was to investigate the impact of the closed Kairiai municipal landfill located near Šiauliai City on the Ginkūnai Pond, into which the leachate from the landfill is released. The main aim of our study was to determine the phytotoxic impact of landfill leachate, water and bottom sediments of the Ginkūnai Pond and Švedė Creek, as well as drainage channel through which the leachate is discharged into the pond. The main characteristics of physico-chemical analysis of the leachate, water and bottom sediments of ecosystem tested were assessed. The amount of chlorophyll "a" and dominant groups of phototrophic organisms were also investigated. The tests were performed during 2012 and 2013. Phytotoxicity tests were carried out using seed germination and root growth test of *Lepidium sativum* and relative growth rate (RGR), biomass, and amount of total chlorophyll of *Lemna minor*. The leachate was very toxic to both tested plants. The water of the Ginkūnai Pond was moderately toxic for root growth of *L. sativum* and in many cases non-toxic or slightly toxic to *L. minor*. Investigations of the impact of Kairiai landfill on the Ginkūnai Pond confirmed the expected existence of the gradient of the usually measured physic-chemical parameters in water of the receiving pond. A strong trend of the decrease of chlorophyll "a" content across the pond was revealed. However, the data of phytotoxicity tests did not show similar tendency. Phytotoxicity testing and physico-chemical investigation showed that the closed Kairiai landfill is still remains a serious source of long-lasting permanent pollution, which affects the neighboring water bodies.

Keywords: landfill; leachate; test-organisms; phytotoxicity.

1. Introduction

The use of municipal solid waste landfills is the most widely utilized method of solid waste disposal around the world due to its economic advantages [1]. Since 2009 all municipal, commercial and mixed industrial waste of Lithuania are deposed in 11 landfills. However, about 800 municipal landfills of different size, age and state were in Lithuania in 2006 [2]. Most of them are closed now but their impact on the environment is unknown. The main scope of this study was to assess the impact of the closed Kairiai municipal landfill on the neighboring aquatic ecosystem. This landfill was established in 1960, and about 2 million tons of different waste are believed to be buried in an 8 ha area [3]. According to the age and state, Kairiai landfill can be attributed to the methanogenic phase of decomposition [4]. The most severe impact of landfills on the environment is probably exerted through the groundwater pollution [5-7]. However, the aftermath of the accidental spill in Kairiai landfill in 2002, when about 44 000 tons of leachate was discharged into drainage channel and then into the Ginkūnai Pond, neighboring pastures and arable land, was rather serious [8-9]. Leachates are defined as aqueous effluent generated as a consequence of rainwater percolation through wastes, biochemical processes in waste's cells and the inherent water content of wastes themselves [10]. Landfill leachate is a dark, highly polluted liquid of variable composition. It contains organic matter (biodegradable, but also refractory to biodegradation), where humic-type constituents consist an important group, as well as ammonia-nitrogen, heavy metals, chlorinated organic and inorganic salts [11]. Leachate composition depends on the characteristics of waste deposed, environmental conditions, landfill operations and on the dynamics of the decomposition processes that takes place in the landfill [7, 10, 12, 13]. The leachates compounds could be assimilated by any surviving aquatic species, could pass through the food chain, and could bioaccumulate by long-term

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exposure [14, 15]. Therefore, toxicity of landfill leachate must be investigated by biological methods and not just described using data of physico-chemical analysis. Numerous of investigations of the effects of landfill leachate on test-organisms are performed [13, 15–17]. However, the number of studies on the effects of landfill on surrounding ecosystem is limited [18, 19]. The leachates entering the water bodies are becoming ones of the main factors of pollution of the aquatic environment, since they change the water and sediment chemical composition, disturb biological balance of the self-cleaning processes, which can lead to unpredictable environmental changes in the ecosystem [19, 20]. In addition the landfills could remain a permanent pollution sources for long periods of time after the termination of their operation [16]. Therefore, the establishment and evaluation of the ecological significance of the outgoing pollution from closed landfills becomes an important subject. For this purpose physico-chemical and biological-toxicological parameters should be assessed.

The main aim of our study was: to determine the phytotoxic impact of landfill leachate, water and bottom sediments of the Ginkūnai Pond and Švedė Creek, as well as drainage channel through which the leachate is discharged into the pond (1); to compare their phytotoxicity in 2012 and after the accidental leachate spill in 2002 (2); to carry out hydrochemical and hydrophysical analysis of the water samples (3); to predict possible consequences of the persistent pollutant migration on the ecotoxicological state of aquatic ecosystem (4).

2. Materials and methods

2.1. Sample collection

Kairiai landfill is located 5 km east of Šiauliai City (55° 55' 42.7", 23° 23' 42.81", WGS). The landfill began operation in 1960 and was closed in 2007. The large scale municipal, household and industrial waste from bicycle, television factories, leather processing, furniture and food industry wastes containing toxic substances have been deposited in it. The landfill is still continue to seep leachate, which is channeled into two isolated holding reservoirs, maintained under open-air conditions, and from time to time is transported to treatment plants. The aquatic ecosystem incorporated in the landfill area consists of the nameless drainage channel surrounding the landfill which for the 1.5-km flows into the Ginkūnai Pond (of 1.1 km^2 area), and in turn the Švedė Creek flows out of the pond. Overall, the six sampling stations (No. 0, 1, 2, 3, 4, 5) moving away from the leachate holding reservoirs were set at the distance of about 10, 800, 1300, 2200, 2900 and 3200 meters, respectively in the drainage channel, the pond and the creek along the water flow direction (Fig. 1).

2.2. Sampling and water chemistry analysis

All water and bottom sediments samples were collected in June, August, and October, 2012 and in July, 2013.

The following physical-chemical characteristics of water sample were established: general – dissolved O_2, pH, salinity, conductivity, permanganate number (mg O/l), total hardness, alkalinity, cations – Na^+, K^+ Ca^{2+}, Mg^{2+}, NH_4^+, anions – Cl^-, SO_4^{2-}, HCO_3^-, CO_3^{2-}, NO_2^-, NO_3^-, as well as total priority heavy metal (Cu, Zn, Ni, Cr, Pb, Cd, Hg) concentrations were determined according to standardized procedures (ISO: 10304; 9963-1; 14911; 10523; 8467; 27888; 15586:2003; 1483:2000).

2.3. Spectral fluorometry analysis

The amount of chlorophyll "a" and yellow substances, as well as dominant groups of phototrophic organisms were determined using fluorometer AlgaeLabAnalyser (bbe Moldaenke, Germany).

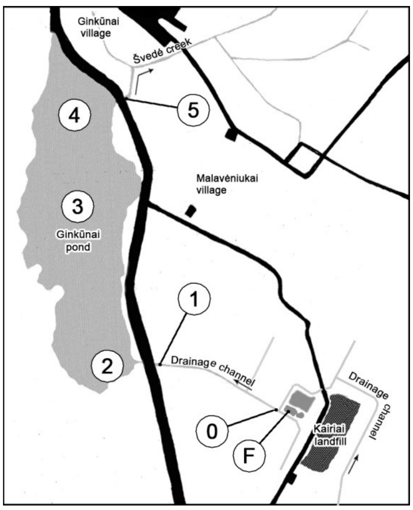


Fig. 1. The scheme of the study area and sampling stations: landfill leachate reservoir (F), drainage channel (station No. 0 and 1), the Ginkūnai Pond (station No. 2, 3, and 4) and Švedė Creek flowing out of the pond (station No. 5)

2.4. Bioassays

The phytotoxicity tests of the leachate, water and bottom sediments of the Ginkūnai Pond were carried out using 2-day seed germination and root growth test of *Lepidium sativum* L. and relative growth rate (RGR), biomass, and amount of total chlorophyll of *Lemna minor* L in 7-days experiment. Tests with *L. sativum* were carried out following a modified method suggested by I. Magone [21, 22] and tests with *L. minor* was performed according to the OECD guidelines for the testing of chemicals [23, 24].

***Lepidium sativum* L.** 9 ml of distilled water (as control) or testing sample solution was pipetted onto three layers of filter paper fitted into a 9-cm glass Petri dish. Twenty-five healthy looking and of similar size seeds of *L. sativum* were distributed evenly on filter paper. The Petri dishes were placed in the dark at 25±1°C for 48 hours. Afterwards the seed germination and root length of seedlings were measured. Germination power of seeds and length of *L. sativum* roots in distilled water were 90.7±2.5% and 20.2±1.9 mm, respectively. The experimental set of each testing scheme involved 5 control dishes and 5 replicates for each tested sample.

***Lemna minor* L.** Plants were collected from the Turniškiai Creek in Verkiai regional park (Vilnius, Lithuania) and identified as *Lemna minor* L. species by specialists of Nature Research Centre (Vilnius, Lithuania). Colonies consisting of 2 to 4 visible fronds were transferred from inoculum culture into the test vessels filled with 150 ml of tested sample. Each test vessel contained total of 10 or 20 (if the amount of chlorophyll was measured) fronds. Steinberg medium was used as control. The vessels were placed in 16/8 h day/night cycle at 25±1°C for 7 days. Frond numbers appearing normal or abnormal were determined at the beginning of the test, at least once every 3 days during the exposure period and at test termination. The RGR, biomass and changes in plant development, e.g. in frond size, appearance, indication of necrosis, chlorosis or gibbosity, colony break-up or loss of buoyancy were registered. The experimental set of each testing scheme involved 3 control vessels and 3 replicates for each tested sample.

2.5. Chlorophyll measurement

Chlorophyll “a” and “b” was determined in 80% acetone extracts of 0.2-g aliquots of fresh plants of *L. minor* as described by Su *et al.* [24], measured spectrophotometrically (Libra S32 PC Biochrom, Germany) and total chlorophyll amount was calculated by the equations proposed by Wellburn [25].

Content of chlorophyll “a” in water samples collected in 2012 was indicated spectrophotometrically in 90% acetone extracts and the concentrations were calculated by SCOR-UNESCO [26] suggested formula. Content of chlorophyll “a” in water samples collected in 2013 was measured using fluorometer AlgaeLabAnalyser (bbe Moldaenke, Germany).

2.6. Toxicity assessment

The EC50 values (i.e., leachate concentrations that induce 50% growth inhibition of *L. sativum* and *L. minor* as a percentage of the control, in 2- and 7-day experiments, respectively) were estimated by linear regression analysis of root length and RGR, and the logarithm of leachate concentration in folds.

According to the modified scale of Wang [27], which is based on inhibition of root growth of *L. sativum* and RGR of *L. minor* of 100–60%, 61–40%, 41–20% and lowers than 19%, the toxic impact was classified as very strong, strong, moderate and slight, respectively. The sample was non-toxic if the inhibition of growth did not statistically differ from the control. The toxicological classification, based on the toxic unit ranges, was established as proposed by Lapa *et al.* [28] and Wilke *et al.* [29]: highly toxic (TU > 100), very toxic (10 < TU < 100), toxic (1 < TU < 10) and no toxic (TU < 1).

2.7. Statistical analysis

Differences between the control and experimental groups were analyzed using one-way variance analysis (ANOVA). The ANOVA compares the variance between the groups with the residual variance using F-test. All statistical analysis were carried out using the program STATISTICA 7.0 (StatSoft Inc., Tulsa, Oklahoma, USA), at the significance level of $p < 0.05$.

3. Results and discussion

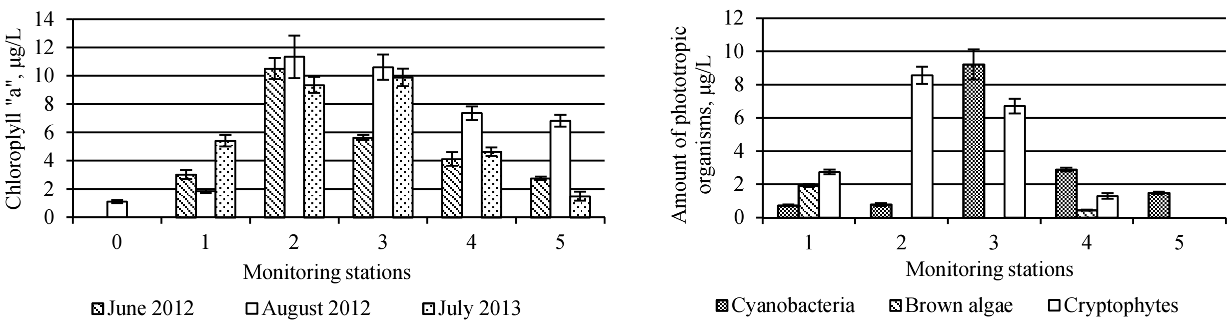
Chemical analysis of landfill leachate indicated that permanganate index was 234 mg/L, conductivity was 9.76 mS cm^{-1}, salinity – 5.4 (‰), pH – 8.45, and concentrations of O_2 and NH_4^+ were 1.05 mg/L and 100 mg/L, respectively. The concentrations of anions and cations were as follows: Cl^- (1889 mg/L), SO_4^{2-} (60.4 mg/L), HCO_3^- (1930 mg/L), CO_3^{2-} (8.71 mg/L), NO_2^- (2.89 mg/L), NO_3^- (<0.050 mg/L), Na^+ (1196 mg/L), K^+ (582 mg/L), Ca^{2+} (90.9 mg/L), Mg^{2+} (103 mg/L), NH_4^+ (100 mg/L). Comparatively high heavy metal concentrations were found in Kairiai landfill leachate (Cu – 2, Zn – 76, Ni – 100, Cr – 620, Pb – 3 and Hg – 0.2 mg/L, respectively) and were close to those median commonly occurring in landfill leachates [30]. However, chromium concentration was significantly higher, apparently, due to the fact that Kairiai landfill has been deposed with a lot of chromium leather processing industrial wastes for many years. In general, the physico-chemical results obtained for leachate in our study are in close agreement with those published by other authors [13, 16], especially in the cases of pH, conductivity, chlorides and ammonium nitrogen.

Analytical data showed that there was a significant variation of a number of physico-chemical parameters among the station water samples. As it was expected, chemical analysis of water in drainage channel and the Ginkūnai Pond showed the existence of the gradient of various physico-chemical parameters (e.g. salinity decreased from 0.5 to 0.0 (‰); concentration of NO_3^- and NH_4^+ decreased from 47.4 to 3.01 mg/L and from 5.75 to 0.08 mg/L, respectively). However, concentrations of most heavy metals remained unchanged across the pond, except of Cr and Ni, concentrations which decreased from 4 to less than 1 and from 4 to less than 2 µg/L. In accordance with the Lithuanian law [31], the Ginkūnai Pond was attributed to the water bodies of moderate ecological state (the average amount of total N was about 2.0 mg/L and the average amount of total P was about 0.073 mg/L) in 2012 [32].

The data obtained here coincide the statement that because of the high concentration of many pollutants, municipal landfill leachates are considered to be ones of the types of wastewaters with the highest environmental impact.

The coloured dissolved organic matter (CDOM) is described as part of dissolved organic matter which consists of products of plants and animals decomposition [33] and soil particles entering in the water [34]. It was found that the amount of CDOM in water of the Ginkūnai Pond fluctuated from 4.22 (station No. 2) to 5.56 µg/L (station No. 3). Average value of CDOM reached 5.1±0.47 µg/L. The amount of CDOM in other tested lakes of Lithuania varies from 1.32 µg/L (Lake Balsiai, Vilnius district) to 8.61 µg/L (Lake Gauštvinis, Kelmė district). In natural waters CDOM plays an important role: due to strong absorption of CDOM in the UV portion of the spectrum, it protects phytoplankton, macroalgae and other biota from damaging UV medium wave radiation [35]. However, the increased level of CDOM can reduce the amount and quality of photosynthetically active radiation to phytoplankton and other primary producers [36, 37].

Concentration of chlorophyll "a" in natural water well reflects the amount of nutrients, especially P and N, in water. With the increasing amount of P and N in water, the growth of phytoplankton and chlorophyll "a" concentration also increases. According to J. Kavalauskienė [38] chlorophyll "a" concentration in water of the Ginkūnai Pond in the summer of 1988–1990 fluctuated from 5.19 to 34.54 mg/m^3. She stated that concentration of this pigment in eutrophic water bodies of Lithuania fluctuated from 0.141 to 45.30 mg/m^3 (average value 7.52–21.6 mg/m^3). Our measurements showed that chlorophyll "a" concentration in water of the Ginkūnai Pond ranged from 7.05 to 11.34 µg/L in the summer of 2012 and from 4.64 to 9.89 µg/L in the summer of 2013 (Fig. 2). Average value of chlorophyll "a" concentration reached 9.76±2.12 µg/L in summer of 2012 and 7.96±2.89 µg/L in the summer of 2013. It was found that the content of chlorophyll "a" in tested water samples of the drainage channel (Station No. 1) was higher in June (3.03 µg/L) than in August of 2012 (1.84 µg/L) (Fig. 2). A strong trend of the decrease of chlorophyll "a" content across the pond was determined in 2012 and 2013 (r =0.945 and r =0.929, p < 0.05, respectively). The highest value of chlorophyll "a" was recorded in a part of the pond directly affected by landfill leachate, and the lowest value was identified in the water of Švedė Creek.



(a) (b)

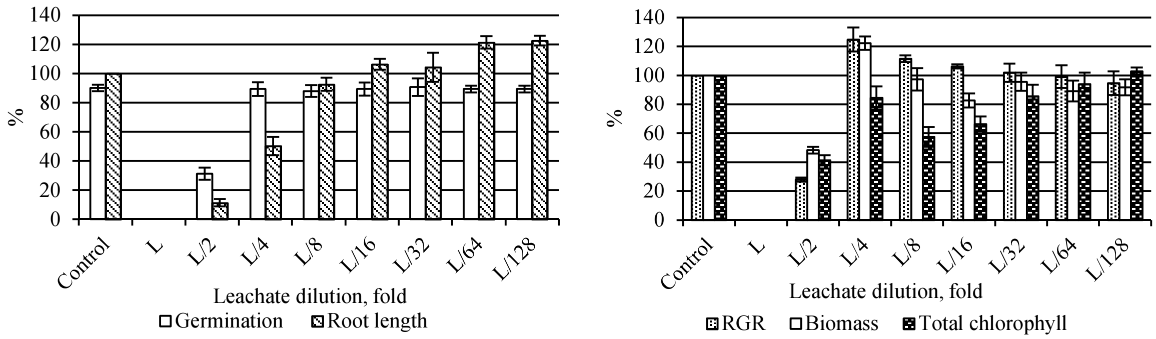
Fig. 2. Chlorophyll „a" concentration (µg/L) (a) and amount of phototrophic organisms (µg/L) (b) in water of drainage channel (station No. 0 and 1), the Ginkūnai Pond (station No. 2, 3, and 4), and Švedė Creek flowing out of the pond (station No. 5) in June and August of 2012 and in July of 2013

Analysis of the amount of phototrophic organisms groups in the Ginkūnai Pond using fluorometer AlgaeLabAnalyser (bbe Moldaenke), showed that in the summer of 2013 the dominant groups in the water of this water body were cyanobacteria and cryptophytes (Fig. 2). The highest concentration of cyanobacteria (9.22 µg/L) and cryptophytes (8.56 µg/L) was recorded in the water of station No. 3 and station No. 2, respectively (Fig. 2). Green algae were not recorded in any of the tested water samples and brown algae were found only in the water of the station No. 4 and in drainage channel (station No. 1) (Fig. 2). According to J. Kavaliauskienė [38] in the summer of 1990 cyanobacteria were the dominant phototrophic organisms in the Ginkūnai Pond, but a rather high concentration of green algae was also determined. The author stated that the abundance of cyanobacteria in eutrophicated water bodies of Lithuania is conditioned not only by high concentrations of P, but also by N, particularly organic, inflow, which is typical for the Ginkūnai Pond [38].

Investigation of the phytotoxicity of the leachate showed that it was very toxic to both tested plants, because seeds of *L. sativum* did not germinate and plants of *L. minor* died on the third day of the exposure (Fig. 3). Seed germination was also affected in the diluted leachate by 2-fold, but in higher dilutions significant ($p < 0.05$) effect was not observed (Fig. 3). It was found that significant difference, in comparison with the control, of root length was registered when leachate was diluted 2, 4, 64 and 128-fold (Fig. 3). Regarding to the toxicity of the leachate in *Alium cepa* Bortolotto *et al.* [39] and Klauck *et al.* [13] observed that leachate caused a significant inhibition of root growth beginning at concentration of 40% and 5%, respectively, and at 100% concentration the observed relative inhibition was 87% and 83%, respectively. Investigation of the toxicity of the leachate generated from Xingou Municipal Landfill in China, performed by Li with co-authors [40] indicated that the lower leachate concentrations stimulated the germination, growth and cell division, and did not induce obvious increase in micronucleus (MN) frequency in root tips of *Triticum aestivum* (wheat), while the higher concentrations inhibited these processes. Genotoxicity of the leachate samples in *A. cepa* also has been found in the studies of Klauck et al. [13] and Kwasniewska et al. [41].

The least RGR of *L. minor* was calculated in the diluted leachate by 2-fold (Fig. 3). When leachate was diluted from 4 to 8-fold the RGR was statistically significant ($p < 0.05$) higher than that in the control, and in other cases the RGR value did not differed from the control (Fig. 3). Very similar effects were observed in the cases of biomass (Fig. 3). In all cases the content of total chlorophyll was significantly ($p < 0.05$) smaller than that in control (Fig. 3). The decrease of chlorophyll was observed visually, because the plants were brighter in comparison with control. The colony break-up or loss of buoyancy was not fixed.

The EC50 of *L. sativum* and *L. minor* were calculated when leachate was diluted by 3.9 and 2.9-fold, respectively, as well as TU was 26 and 35, respectively. According to the toxicological classification proposed by Lapa *et al.* [28] and Wilke *et al.* [29] the leachate tested was very toxic to *L. sativum* and *L. minor* ($10<TU<100$). It should be noted that in 2002 EC50 value for root growth of *L. sativum* was determined when leachate was diluted 7.9-fold [9].



(a) (b)

Fig. 3. Impact of the leachate (L) of Kairiai landfill on seed germination and root length of *L. sativum* (a) and RGR, biomass and total chlorophyll of *L. minor* (b) in 2012

The tests performed in June and August 2012 showed that the water of drainage channel (station No. 0 and 1) and the Ginkūnai Pond was non-toxic for germination of *L. sativum* seeds. However, it was moderately toxic for root growth (Table 1) (data of root growth of *L. sativum* in June of 2012 not shown in Table 1, because there were no statistically significant ($p < 0.05$) differences between root growth in water collected in June and August). Among all tested water samples only the water of the station No. 5 was moderately toxic to *L. minor* (Table 1). All other samples were weakly toxic or non-toxic. However, the water from drainage channel (station No. 0 and 1) stimulated the growth of *L. minor* (Table 1). It could be related to a rather high concentration of biogens in this water. Toxic impact of the water from the mentioned-above drainage channel after accidental spill in 2002 was very strong to *L. sativum*, although water of the Ginkūnai Pond was moderately toxic [9].

Table 1. Toxic impact of water of drainage channel (station No. 0 and 1), the Ginkūnai Pond (station No. 2, 3, and 4), and Švedė Creek (station No. 5) on *L. sativum* and *L. minor* in August, 2012

Station	Root length, %±SD, of *L. sativum*	Toxic impact, according to inhibition of root length of *L. sativum*	Relative growth rate (RGR), %±SD, of *L. minor*	Toxic impact, according to RGR of *L. minor*
0	*62.3 ± 1.3	Moderate	*126.8 ± 5.15	Non-toxic
1	*59.9 ± 0.9	Moderate	*116.6 ± 4.68	Non-toxic
2	*63.8 ± 0.8	Moderate	103.6± 7.02	Non-toxic
3	*70.3 ± 0.7	Moderate	92.7 ± 9.3	Non-toxic
4	*65.4 ± 0.9	Moderate	*89.1 ± 5.15	Weak
5	*67.4 ± 0.5	Moderate	*68.5 ± 7.01	Moderate
Control	100	Non-toxic	100	Non-toxic

Asterisk (*) denotes the value significantly differed from control ($p < 0.05$).

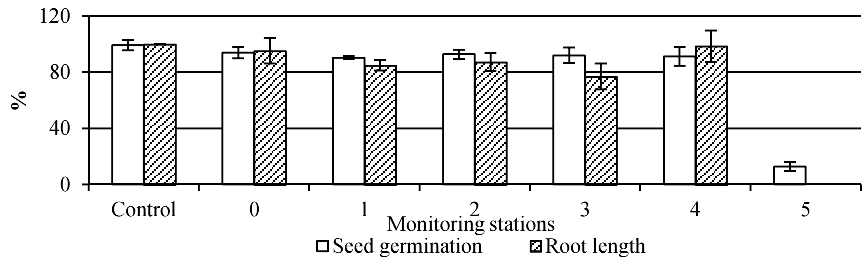


Fig. 4. Impact of bottom sediments of drainage channel (station No. 0 and 1), the Ginkūnai Pond (station No. 2, 3, and 4), and Švedė Creek flowing out of the pond (station No. 5) on seed germination and root growth of *L. sativum* in June, 2012

Ivestigations of the phytotoxicity of bottom sediments of drainage channel, the Ginkūnai Pond, and Švedė Creek was also performed. It is important that bottom sediments at the source of Švedė creek were very toxic to *L. sativum* in both 2002 [9] and 2012 (Fig. 4). Other samples tested of the bottom sediments were non-toxic (station No. 0 and 4), slightly toxic (station No. 1 and 2) or moderately toxic (station No. 3) to this plant in 2012 (Fig. 4), while in 2002 they all were moderately toxic to *L. sativum* [9].

In general, the data did not confirm the existence of a strict relationship between integral toxicity level of test water and the distance from the leachate holding reservoir which was obtained in the investigations of Svecevičius et al. [19] of the toxicity of the Ginkūnai Pond water on medicinal leech (*Hirudo verbana*), daphnia (*Daphnia magna*) and rainbow trout (*Oncorhynchus mykiss*). On the contrary, the effect of the increase of the toxicity of water with the increasing distance from the source of pollution was observed in the case of *L. minor* (Table 1). In addition, the strongest toxicity among all bottom sediments tested was also established in bottom sediments of Švedė Creek (station No. 5) source (Fig. 4). However, a strong trend towards the decrease of chlorophyll "a" content and amount of phototrophic organisms across the pond was recorded (Fig. 2). All that approved the statement that the use of battery of test-organisms is essential for reliable assessment of the effect of landfill leachates on the neighboring water bodies, because various organisms respond differently to landfill leachate [9, 13, 16, 18, 19, 42]. Consequently, the battery consists of test-organisms of different phylogenetic level, hydrochemical measurements together with parameters such as content of chlorophyll "a", amount of phototrophic organisms and CDOM could show real picture of the ecological state of polluted water bodies.

4. Conclusion

Phytotoxicity testing and physico-chemical investigation showed that the closed Kairiai landfill remains a serious source of permanent pollution, which affects the neighbouring water bodies. The landfill leachate is still extremely toxic wastewater, which falls from the holding reservoirs to the drainage channel and further to the Ginkūnai Pond and Švedė Creek. Therefore, the leachate becomes the primary and secondary source of pollution, what could cause irreversible changes in the aquatic ecosystem. Physico-chemical investigation and bioassay testing could enable to extrapolate obtained experimental data to environment in order to predict possible migration, distribution and accumulation of pollutants and possible consequences of their effects on the ecological state of aquatic ecosystem.

Acknowledgements

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