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# Pollution of Small Kaunas Rivers with Biogenic Compounds and Heavy Metals

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The pollution of surface water bodies with biogenic compounds and heavy metals is a serious global concern for environment as well as for human health. Therefore, the aim of this study was to investigate the pollution of secondary rivers of Nemunas, flowing in Kaunas city, with respect to concentrations of biogenic nitric and phosphorus compounds as well as heavy metals in water and bottom sediments of the rivers. The samples of water and bottom sediments were taken in the influxes of Sėmena, Girstupis, Marvelė, Vėžpienis and Veršvas. The concentrations of inorganic nitric compounds (ammonium, nitrites and nitrates), and phosphates as well as heavy metals (Zn and Cu) in water and bottom sediments were investigated. Sėmena was found to be the cleanest river, as its ecological status class varied from high (with respect to phosphates) to moderate (with respect to ammonium). Girstupis was highly polluted with phosphates; however, it showed a relatively good ecological status class with respect to nitric compounds. High concentration of phosphates was also found in Veršvas; however, contrary to Girstupis, this river contained exceptionally high concentration of reduced nitric compounds (ammonium). Vėžpienis and Marvelė were the most polluted rivers, with most of the nitric and phosphorus compounds exceeding the limit values set in the wastewater management regulations. These rivers were attributed to the poor or bad ecological status class with respect to ammonium and the good or moderate ecological status class with respect to nitrates and/or phosphates. The concentrations of Zn were relatively low in the investigated Kaunas rivers; however, the concentration of Cu was found to be almost 3-times higher than the limit value in Sėmena, Girstupis and Veršvas.

**Keywords:** urban river pollution, heavy metals, nitric compounds, phosphates.

## Introduction

Urban river pollution is typically understood as a problem of the modern age. Scientists investigate two main aspects of river pollution: how by-products of industrial and agricultural run-off contaminate waterways, prompting legislative responses (Oosthoek, 2002; Massard-Guilbaud, 2004); and how domestic wastewater disposal creates hygiene concerns and difficulties with providing clean drinking water (Barles and Lestel, 2007; Closmann, 2007).

Pollutant discharge causes widespread organic and toxic pollution, as well as excess of biogenic nitric and phosphorus compounds, leading to eutrophication and severe ecological destruction (Miao et al., 2012). Biogeochemical cycles of phosphorus and nitrogen, which are important biogenic elements that determine the productivity and structural as well as functional organization of ecosystems, are currently undergoing significant anthropogenic transformations. One of the significant types of anthropogenic interference in the circulation of biogenic elements is the use of mineral fertilizers, in particular the ones containing nitrogen and phosphorus (Копла-Дикс and Старвинская, 1993). Another source of anthropogenic N and P compounds (and phosphates) is a discharge of partly treated wastewater (Chesterikoff et al., 1992; Sitonytė and Kerienė, 2010; Vrzel et al., 2016). The process of eutrophication of land-based reservoirs was ranked first in terms of the degree of danger of a global anthropogenic impact on the environment at the 22nd Session of UNEP as early as in 1984. Eutrophication of water bodies is becoming one of the most urgent problems of surface water nowadays (Wang et al., 2012).

Toxic pollution of water bodies is determined by the presence of toxic compounds, in particular, heavy metal compounds. Heavy metals, such as arsenic, cadmium, chromium, copper, lead, mercury, nickel and zinc, polluting and accumulating in the water bodies present a rapidly growing global concern (Ахметшиева, 2015). These elements can be found in the environment (water reservoirs, the atmosphere and soil) in excess due to various anthropogenic activities. It is also important to note the natural sources of heavy metal pollution. These include weathering and erosion of all types of rocks (igneous, sedimentary and metamorphic), leading to liberation of various heavy metals, its transport and redistribution in the environment (Carpenter, 1998).

The pollution of surface water with biogenic compounds and heavy metals is a serious global concern for environment, as well as for human health. Overabundance of these elements poses severe health risks for humans, and for other life forms through bioaccumulation through food chains. Pollution of small urban water bodies is a widespread problem (Paul and Meyer, 2001; Ellis and Butler, 2015; Revitt and Ellis, 2016); however, there is a lack of data describing the quality and the specific pollution sources of freshwaters in urban areas (McGoff et al., 2017). Therefore, the aim of this study was to investigate the pollution of secondary rivers of Nemunas, flowing in Kaunas city, with respect to concentrations of biogenic nitric and phosphorus compounds as well as heavy metals in water and bottom sediments of the rivers.

## Methods

### Research object and sampling

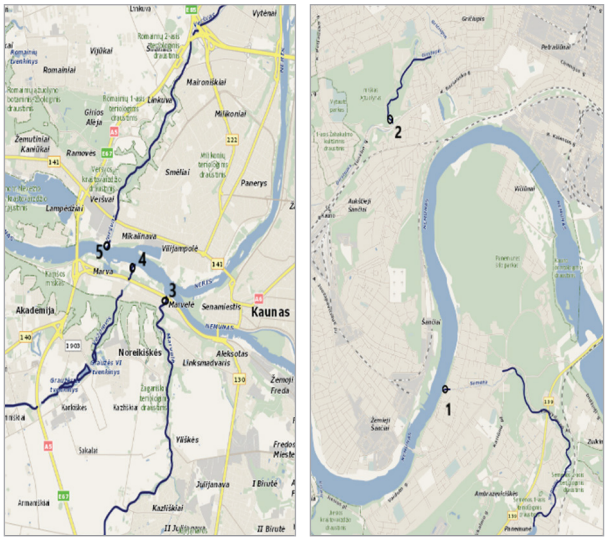
The following investigated rivers are the secondary rivers of Nemunas, the biggest river in Lithuania (arranged in downstream order): Sėmena (left inflow), Girstupis (right inflow), Marvelė and Vėžpienis (left inflows) and Veršvas (right inflow) (Fig. 1) All these rivers are relatively small, belonging to the 1st type (catchment area <100 km$^2$) (Ministry of Environment, 2007) and flow through different regions of Kaunas city. The samples of water and bottom sediments were taken in December 2017 and January 2018 in the influxes of the investigated rivers (except Girstupis, where samples were taken immediately before the underground channelized part of the river) (Fig. 1).

Water quality of the investigated rivers was assessed on the basis of EU and Lithuanian legislation. The rivers were assessed to one of the five ecological

status classes (high, good, moderate, poor and bad) according to concentrations of nutrients (ammonium, nitrates and phosphates) and heavy metal micronutrients (Zn and Cu) (Table 1) (Ministry of Environment, 2007). Assessment of water quality according to the ecological status classes requires at least 4 seasonal measurements per year. This study, however, is only the beginning of longer investigation, revealing the fact of pollution of small Kaunas rivers and the need of continuous survey of water quality in it. In order to compare the water quality of the investigated streams with other bigger Lithuanian rivers, the concentrations of nitric compounds and phosphates were compared with the limit values set in the wastewater management regulations of Lithuania (Ministry of Environment, 2006). Although the investigated rivers are not official sewage receivers, some of them (Sėmena, Girstupis, Veršvas and Marvelė) are used for collecting surface run-off. Moreover, some collectors of surface run-off are also used for collecting the sewage

**Fig. 1**

The sampling places in the secondary rivers of Nemunas in Kaunas (1 – Sėmena, 2 – Girstupis, 3 – Marvelė, 4 – Vėžpienis, 5 – Veršvas)




from individual residential houses (Municipality Administration of Kaunas City, 2010).

The water samples were stored at –30°C before analysis. The samples of bottom sediments were dried in electric oven 3 days at 80°C, sieved through a 2-mm sieve, grounded with a mortar and stored at room temperature before analysis.

## Chemical Analysis

### Determination of concentrations of phosphates and nitric compounds

Concentrations of phosphates and inorganic nitric compounds – ammonium, nitrites and nitrates – were investigated spectrophotometrically according to Bartoshova et al. (2012) with minor corrections. The concentration of ammonium was detected by reaction of ammonium ion with Nessler. Water sample was mixed with Segnet salt and Nessler reagent, light absorption was measured at 400 nm. The concentration of nitrites was determined by reaction with sulphanilamide. Colour development was measured at 540 nm. For determination of the concentration of nitrates, a diluted water sample was mixed with sodium salicylate and evaporated in a sand bath at 100°C. Light absorption was measured after precipitates' reaction with sulphuric acid and alkaline solution of tartrate at 420 nm. The concentration of phosphates was determined according to the production of a blue complex phosphorus and molybdenum compound after reaction with antimony salt and reduction with ascorbic acid. Light absorption was measured at 670 nm.

### Determination of concentration of heavy metals

The mineralization of the sediment sample was performed according to the manufacturer's instructions. The sample was mixed with the following acids: concentrated acetic acid, hydrochloric acid and hydrofluoric acid, as well as 5% (w/v) boric acid. The mixture was digested using High Performance Digestion System Ethos One (Milestone Helping Chemists, JAV) until full mineralization of the sample. The concentration of Zn and Cu in the diluted acidic solution of the sample was detected with atomic absorption spectrometer AA-6800 (Shimadzu, Japan).

## Statistical analysis

All measurements were performed in five replicates. The data were analyzed using STATISTICA 8 software and Student's t test at $p < 0.05$.

## Results and Discussion

### Concentration of biogenic N and P compounds

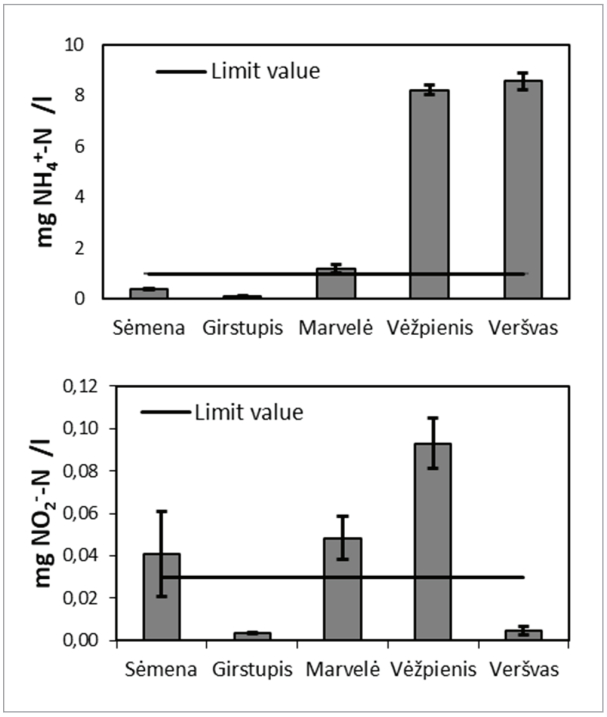
The concentration of biogenic nitric compounds in the secondary rivers of Nemunas is shown in Fig. 2. The investigated rivers contained relatively high concentrations of ammonium, nitrites and/or nitrates, as in many cases the concentrations of these compounds were close to or even exceeded the limit values, set in the wastewater management regulations (Ministry of Environment, 2006). The highest concentrations of ammonium were found in Vėžpienis and Veršvas (8.2 mg/L and 8.6 mg/lL respectively), exceeding the limit value of 1 mg/L by approximately 8.3 times. Such high concentrations are rarely determined even in urban rivers. For example, a similar level of $NH_4^+$-N was detected only in the surface water outfalls, polluted by sewage in the suburbs of London (Ellis and Butler, 2015). Attention should be paid to the fact that the lowest concentration of $NH_4^+$-N (0.12 mg/L) was detected in Girstupis. This fact evidences the ecological role of Mickevičius valley for self-purification of this river before entering the underground channel and the main River Nemunas, as the park is located in the middle part of Girstupis, above the underground channelized part and the water sampling point of this river. According to ammonium concentration, Girstupis was attributed to the rivers of the good ecological status class, Sėmena to moderate, Marvelė to poor, Vėžpienis and Veršvas to the bad ecological status class (Ministry of Environment, 2007).

The highest concentration of nitrites was detected in Vėžpienis river (0.093 mg N/L), exceeding the limit value (0.03 mg N/L) approximately by 3-times ($p < 0.05$). A high level of $NO_2^-$-N was also observed in Sėmena and Marvelė, whereas other investigated rivers contained a relatively low level of this N compound (Fig. 2).

The highest concentration of nitrates was found in Marvelė (3.8 mg N/L). The concentration of nitrates was lower than the limit value (2.3 mg N/L) in other rivers of Kaunas, fluctuating from 0.4 mg N/L in Veršvas to 1.9 mg N/L in Vėžpienis. In the latter case the concentration of nitrates was close to the limit value, set in the wastewater management regulations

**Fig. 2**

The concentration of ammonium and nitrites in the secondary rivers of Nemunas in Kaunas city

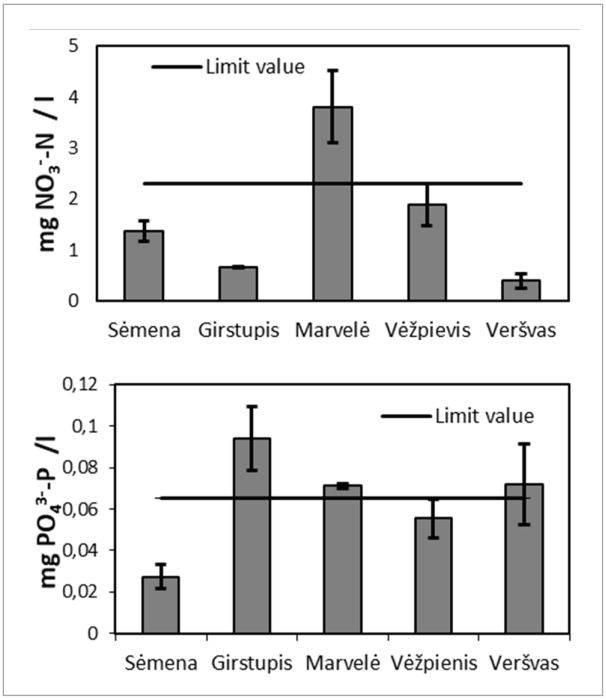



(Ministry of Environment, 2006) and did not differentiate from it statistically significantly. A similar level of $NO_3^-$-N was also detected in urban streams in London (McGoff et al., 2017) and in Spanish rivers with low agricultural intensity in catchments areas (Moreno et al., 2006). The ecological status classes of the investigated rivers with respect to the concentration of nitrates were as follows: Girstupis and Veršvas – high, Sėmena and Vėžpienis – good, Marvelė – moderate (Ministry of Environment, 2007) (Fig. 3).

Our study revealed that all the investigated rivers, except Sėmena, were polluted with phosphates. The concentrations of $PO_4^{3+}$-P were higher than the limit value (0.0653 mg P/L) in Girstupis and Marvelė ($p < 0.05$), whereas the concentrations of phosphates in Vėžpienis and Veršvas did not differ from it statistically significantly. According to the concentrations of phosphates, Sėmena should be attributed to the rivers of the high ecological status class, Vėžpienis, Veršvas and Marvelė to the good ecological status class, and Girstupis to the moderate ecological status class

**Fig. 3**

The concentration of phosphorus and nitrates in the secondary rivers of Nemunas in Kaunas city




(Ministry of Environment, 2007) (Fig. 3). On the other hand, the concentrations of $PO_4^{3+}$-P in both Girstupis and Marvelė were above the limits, set for receiving waters in the wastewater management regulations (Ministry of Environment, 2006). High P concentrations correlate with urban use in catchment areas, industry and households being as the main sources of pollution (Moreno et al., 2006). Moreover, according to the Municipality Administration of Kaunas City, Kaunas district Aleksotas and the City centre, which are drained by Marvelė and Girstupis, respectively, still have mixed sewerage, with some household sewage incorporated to surface run-off sewers (Municipality Administration of Kaunas City, 2010). Such sewerage might be the major reason of the relatively high concentrations of $PO_4^{3+}$-P in these rivers.

The concentration of ammonium and nitrates is climate-dependent and tends to be higher in the cold season, when consumption by plants and microorganisms is terminated and leaching from soil is the most intensive (Brett et al., 2005; Sitonytė and Kerienė, 2010). Therefore, a relatively high level of these biogenic N compounds in the investigated rivers of Kaunas could be at least partly determined by the seasonal variation and is expected to be lower in spring and summer. On the other hand, concentration of phosphates follows the different pattern and reaches the highest level in summer (Brett et al., 2005; Sitonytė and Kerienė, 2010). Therefore, the results of the earlier studies suggest that an average annual level of $PO_4^3$-P in the investigated rivers of Kaunas might be even higher than determined during this survey. Since this study revealed that small Kaunas rivers are considerably polluted with biogenic compounds, further investigations, including at least 4 seasonal measurements annually, are needed in the future.

In summary of the results of the concentrations of biogenic N and P compounds in the secondary rivers of Nemunas, Sėmena should be distinguished as a relatively unpolluted river, as its ecological status class varied from high (with respect to phosphates) to moderate (with respect to ammonium). The catchment area of this river is mostly situated in wooded areas with low population density, leading to relatively low water pollution. Girstupis was highly polluted with phosphates; however, its ecological status class with respect to nitric compounds was good or even high. In contrast, Veršvas was highly polluted with reduced nitrogen (ammonium), indicating the pollution with sewage (Chesterifoff et al., 1992; Revitt and Ellis, 2016; Česonienė et al., 2017). Vėžpienis and Marvelė were found to be among the most polluted rivers in Kaunas, with most of the N and P compounds exceeding the limit values in the influxes of these rivers. Both rivers flow through urbanized suburb areas of Kaunas, some sections of them are channelized or dammed, limiting self-purification and increasing the risk of pollution with untreated or partly purified wastewater. Household sewage, entering the streams both accidentally or deliberately might be the major reason of their pollution. This presumption is based on the fact that about 10% of inhabitants of Kaunas city still do not have connections to municipal sewerage system (Municipality Administration of Kaunas City, 2010). This leads to surface water sewers misconnections (illicit discharges of wastewater into surface water

sewerage), which is the widespread problem even in developed countries, such as the United States and the United Kingdom (Ellis and Butler, 2015).

## Heavy metals in water and bottom sediments

The concentrations of heavy metals copper (Cu) and zinc (Zn) were investigated in the water and sediments of secondary rivers of Nemunas, in Kaunas city (Table 2). Our study has shown that concentration of Zn in water was relatively low and did not exceed the limit value 100 µg/L (Ministry of Environment 2006). The average Zn concentration in Sėmena and Vėžpienis was 15.5 µg/L, whereas Zn concentration was below the detection limit in Girstupis, Marvelė and Veršvas. In contrast to zinc, the level of Cu in Sėmena, Girstupis and Veršvas exceeded the limit value of 10 µg/L (Ministry of Environment, 2006) almost 3 times. Cu concentration in the bottom sediments of the investigated rivers was also higher than Zn. The highest concentration was found in Girstupis (20 µg/g), whereas in other rivers it varied from 3.7 µg/g (in Marvelė) to 7 µg/g (in Vėžpienis). The rivers are attributed to the good or moderate ecological status class with respect to pollution with Zn or Cu, if concentration of these heavy metals is lower or higher than 20 µg/L or 5 µg/L, respectively (Table 1) (Ministry of Environment, 2007). Therefore, all the investigated rivers (except Marvelė) were in the moderate ecological status class according to the concentration of Cu. In contrast, good water quality with respect to Zn was characteristic of the investigated streams (Table 2). Cu and Zn are among the most prevalent heavy metals in urban rivers, entering the water bodies mostly via non-point sources, such as surface run-off from roads and parking lots (Paul and Meyer, 2001). Our study showed that the bottom sediments of the investigated rivers contained a relatively low amount of Zn and Cu (with exception of Cu in the bottom sediments of Girstupis) (Table 2). A relatively high concentration of Cu in Girstupis sediments might be explained by lower water velocity and a higher amount of silt in the bottom of this stream, as fine particles and organic matter have the highest metal adsorption potential (Paul and Meyer, 2001). The research conducted by Žaltauskaitė (2013) demonstrated much higher concentrations of Zn, Cu, Ni and Pb in the rivers of Kaunas (Sėmena,

**Table 2**

The concentration of heavy metals (HM) in water and the bottom sediments of the investigated rivers

| River | HM in water µg/L | | HM in bottom sediments µg/g | |
|---|---|---|---|---|
| | Zn | Cu | Zn | Cu |
| 1 | 2 | 3 | 4 | 5 |
| Sėmena | 16 | 27 | n.d. | 2.94 |
| Girstupis | n.d. | 29 | n.d. | 20.06 |
| Marvelė | n.d. | 2 | 0.221 | 3.69 |
| Veršvas | n.d. | 28 | 3.977 | 2.99 |
| Vėžpienis | 15 | 7 | 0.693 | 6.99 |
| Limit value | 100 | 10 | | |

**Table 1**

The ecological status classes of the rivers according to the physico-chemical quality parameters (Ministry of Environment, 2007

| Physico-chemical quality parameter | Ecological status class | | | | |
|---|---|---|---|---|---|
| | High | Good | Moderate | Poor | Bad |
| 1 | 2 | 3 | 4 | 5 | 6 |
| $NO_3^-$-N, mg/L | < 1.30 | 1.30–2.30 | 2.31–4.50 | 4.51–10.00 | > 10.00 |
| $NH_4^+$-N, mg/L | < 0.10 | 0.10–0.20 | 0.21–0.60 | 0.61–1.50 | > 1.50 |
| $PO_4^{3+}$-P, mg/L | < 0.05 | 0.05–0.09 | 0.091–0.18 | 0.181–0.04 | > 0.40 |
| Zn, µg/L | | ≤ 20 | > 20 | | |
| Cu, µg/L | | ≤ 5 | > 5 | | |

Vėžpienis, Girstupis, Marvelė), varying in the range of 10–65 µg/g. These findings emphasize the necessity of further investigations of Kaunas streams.

## Conclusions

Small secondary rivers of Nemunas in Kaunas city were differently polluted with phosphates and biogenic nitric compounds. Sėmena was found to be the cleanest river, as its ecological status class varied from high (with respect to phosphates) to moderate (with respect to ammonium). Girstupis was highly polluted with phosphates; however, it showed the good ecological status class with respect to nitric compounds. High concentration of phosphates was also found in Veršvas; however, contrary to Girstupis, this river contained an exceptionally high concentration of reduced nitric compounds (ammonium). Vėžpienis and Marvelė were the most polluted rivers, with most of the nitric and phosphorus compounds exceeding the limit values set for receiving waters in the wastewater management regulations. These rivers were attributed to the poor or bad ecological status class with respect to ammonium and to the good or moderate ecological status class with respect to nitrates and/or phosphates. The concentrations of Zn were relatively low in the investigated Kaunas rivers; however, the concentration of Cu was found to be almost 3-times higher than the limit value in Sėmena, Girstupis and Veršvas.

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## Kauno upelių užterštumas biogeniniais junginiais ir sunkiaisiais metalais

Gauta:
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Paviršinių vandens telkinių užterštumas biogeniniais junginiais ir sunkiaisiais metalais yra svarbi pasaulinė aplinkos ir žmogaus sveikatos problema. Dėl šios priežasties šio tyrimo tikslas buvo ištirti Nemuno intakų, tekančių per Kauno miestą, užterštumą biogeniniais azoto ir fosforo junginiais bei sunkiaisiais metalais. Vandens ir dugno substrato mėginiai buvo paimti Sėmenos, Girstupio, Marvelės, Vėžpienio ir Veršvo upelių žiotyse. Buvo įvertintos neorganinių azoto junginių (amonio, nitritų ir nitratų), fosfatų ir sunkiųjų metalų (Zn ir Cu) koncentracijos vandenyje ir dugno nuosėdose. Nustatyta, kad Sėmena yra švariausias iš tirtų upelių, jo ekologinės būklės klasė svyravo tarp labai geros (pagal fosfatų koncentraciją) ir vidutinės (pagal amonio koncentraciją). Girstupis buvo labai užterštas fosfatais, tačiau jo ekologinės būklės klasė buvo gana aukšta remiantis azoto junginių koncentracijomis. Didelė fosfatų koncentracija taip pat buvo nustatyta ir Veršvo upelyje, tačiau, priešingai nei Girstupyje, šio upelio vandenyje taip pat buvo ir didelės redukuotų azoto junginių (amonio) koncentracijos. Tyrimas parodė, kad Vėžpievis ir Marvelė yra labiausiai užteršti upeliai, daugelis N ir P junginių viršijo leistinas koncentracijas, nustatytas nuotekų tvarkymo reglamente. Šie upeliai buvo priskirti blogos arba labai blogos ekologinės būklės klasei pagal amonio koncentracijas ir vidutinės arba geros ekologinės būklės klasei pagal nitratų bei fosfatų koncentracijas. Cinko koncentracija tirtuose Kauno upeliuose buvo palyginti maža, tačiau vario koncentracija Sėmenos, Girstupio ir Veršvo upeliuose viršijo ribinę vertę apie 3 kartus.

**Raktiniai žodžiai:** miesto upių tarša, sunkieji metalai, azoto junginiai, fosfatai.