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Front cover photography: Agriculture plays an important role in both protecting and developing farmland and is an important factor facilitating development of other sectors (photograph: Matej Lipar).
Fotografija na naslovnici: Kmetijstvo ima pomembno vlogo pri varovanju in razvoju kmetijskih zemljišč in je pomemben dejavnik tudi pri razvoju drugih sektorjev (fotografija: Matej Lipar).

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NITROGEN AND PHOSPHORUS POLLUTION IN GORIČKO NATURE PARK

Darijo Ilić, Jože Panjan

DARIJO ILIĆ

Ledava River at Domajinci.

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Nitrogen and Phosphorus Pollution in Goričko Nature Park

ABSTRACT: This article deals with the impact of diffuse and point sources of nitrogen and phosphorus pollution on the environment in Goričko Nature Park. The park was divided into three parts: the Ledava, Big Krka (*Velika Krka*), and Kobilje Creek (*Kobiljski potok*) basins, which were then compared. The surface waters were monitored and their chemical composition was examined. All three areas are characterized by elevated levels of nitrogen and phosphorus compounds in the water. Nitrogen and phosphorus pollution results from unregulated manure pits on livestock farms, unregulated sewage systems, and runoff of nitrogen and phosphorus compounds from farmland.

KEY WORDS: geography, nature protection, pollution, nitrogen, phosphorus, Goričko Nature Park, Slovenia

Onesnaženje z dušikom in fosforjem v Krajinskem parku Goričko

POVZETEK: Članek obravnava vpliv razpršenih in točkovnih virov na obremenjevanje okolja z dušikom in fosforjem na območju Krajinskega parka Goričko. Krajinski park Goričko smo razdelili na tri dele: porečja Ledave, Velike Krke in Kobiljskega potoka ter med območji izvedli primerjalno analizo. Z monitoringom površinskih tekočih voda smo preučili njihovo kemijsko stanje. Za vsa tri območja so značilne povišane koncentracije dušikovih in fosforjevih spojin v vodi. Onesnaženje z dušikovimi in fosforjevimi spojinami, je posledica neurejenih gnojnih jam na živinorejskih obratih, neurejena kanalizacijska infrastruktura in izgube dušikovih in fosforjevih spojin s kmetijskih zemljišč.

KLJUČNE BESEDE: geografija, varstvo okolja, onesnaženje, dušik, fosfor, Krajinski park Goričko, Slovenija

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1 Introduction

Factors that alter the chemical, biological, physical, and hydromorphological properties of water are those that pollute water and thus have an impact on its condition. Nutrient sources of water contamination (primarily nitrogen and phosphorus) can be divided into point and diffuse sources (Novotny 1988). The point sources of one or more pollutants can be defined and illustrated as points on the map from which pollution spreads into the surrounding areas; the impact decreases with distance. Point sources include industrial and domestic wastewater, direct discharges from livestock farms, and so on. Diffuse pollution sources, which cannot be defined as a single point but rather originate from a specific area, include settlements, agriculture, and traffic infrastructure. Diffuse pollution is the leading form of pollution that is difficult to control (Novotny 2003; De Wit and Behrendt 1999). Intensive agriculture is especially problematic in this regard, because increased fertilizer use and intensive livestock farming increase nutrient inputs.

Healthy drinking water (ground or surface) is recognized as one of the fundamental environmental problems. Water is a partially renewable resource, but excessive contamination, especially by inorganic matter, can turn it into a health risk. In order to prevent this type of contamination as much as possible, water pollution sources must be determined to the greatest possible extent. This is an especially big challenge in the case of diffuse water pollution because the sources must be defined locally. An expressly local approach is required because each area has its own special features.

Nitrogen is an important element of the global ecosystem and a component of many organic and inorganic substances (Williams 2001). Water contains low levels of nitrogen in the form of organic or inorganic compounds (Ibanez et al. 2007). The most important inorganic forms of nitrogen include ammonium (as the ammonium ion NH_4^+ and ammonia or NH_3, which are in balance in a water solution; they have an oxidation number of –3), nitrate (NO_3^- with an oxidation number of +5), and nitrite (NO_2^- with an oxidation number of +3). These ionic forms play an important role in the nitrogen cycle.

After nitrogen, phosphorus is the second most important element in primary production (Green et al. 2007) and it is the most important nutrient to cause the eutrophication of fresh water (Lemmunyon and Daniel 1998), which stimulates algal growth, decreases dissolved oxygen levels, and reduces water transparency (Wood 1998). Excess phosphorus in water from both point and diffuse sources can result in increased primary production and eutrophication, with the potential for seasonal toxic algal blooms, which can have a major negative impact on global water quality (Worsfold 2005). The majority of phosphorus is washed from farmland into surface waters, whereas only small amounts are washed into the groundwater (Bryant 2004). Phosphorus losses from farmland amount to 0.97–1.85 kg/ha a year (Baker 1984). Phosphorus losses from farmland in Goričko Nature Park can be up to 8.2 kg/ha (Karta presežkov fosforja 2006) as a result of surface runoff (Karta površinskega odtoka 2003).

2 Methods

Water quality in Goričko Nature Park was monitored through field measurements and laboratory analyses. Field research included measurements of water and air, pH, electrical conductivity, redox potential, turbidity, and oxygen. Sampling was carried out in line with the Slovenian Standard (Kakovost vode … 2007). Eleven sites were used for sampling, which was carried out once a month from May 28th, 2008 to May 20th, 2009. Discharge was measured using the float method (Brilly 1992), and chemical parameters (i. e., ammonium, nitrate, nitrite, total nitrogen, orthophosphate, total phosphorus, potassium, chemical oxygen demand (COD), five-day biochemical oxygen demand (BOD_5), and undissolved matter) were determined using standard methods (Eaton et al. 1995).

2.1 Description of the study area

Goričko Nature Park is located in the extreme north-east of Slovenia. Most of the area (96%) is a Natura 2000 site (Uredba … 2004). This is a hilly area with an average elevation between 300 and 350 m above sea level and intermittent valleys at an elevation of 220–260 m (Digitalni … 2001).

Acid to neutral soil developed on noncarbonate bedrock (Internet 3). Average annual air temperature is 9.7 °C and the average annual precipitation is 761.8 mm (Meteorološki podatki … 2009). According to

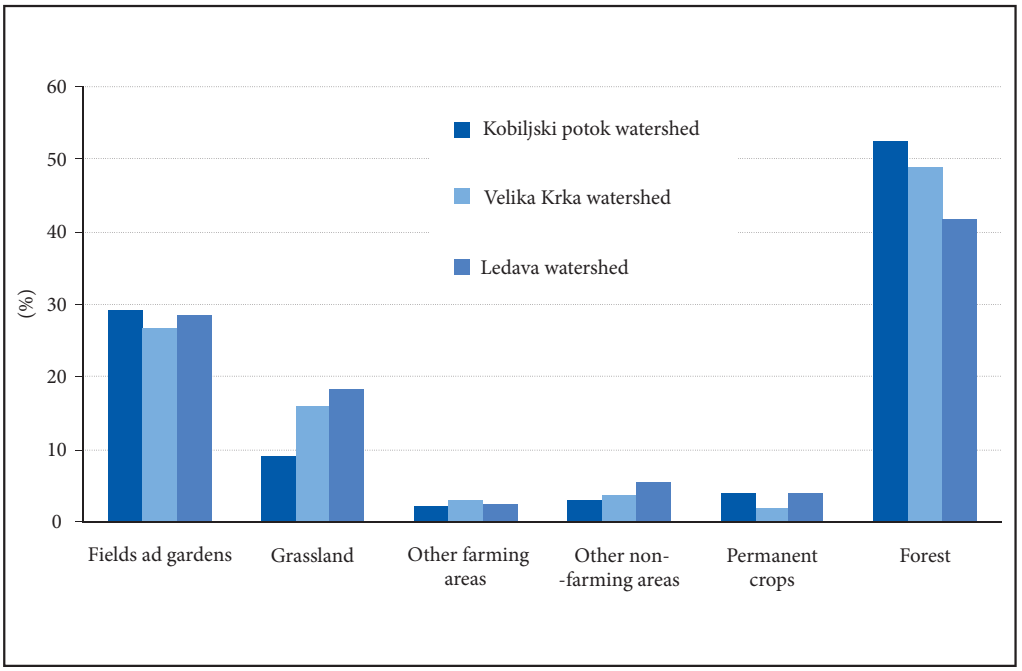

Figure 1: Land use by basin in Goričko Nature Park.

Gabrovec and Kastelec (1998), the annual quasi-global radiation energy (the sum of direct and diffuse solar radiation) on inclined surfaces ranges from 3,300 MJ/m^2 at lower elevations of the central and eastern parts of Goričko Nature Park to around 4,000–4,800 MJ/m^2 in the remaining parts.

Goričko Nature Park was divided into three parts: the Ledava, Big Krka (*Velika Krka*), and Kobilje Creek (*Kobiljski potok*) basins.

The Ledava Basin (Internet 1) covers 21.4 km^2 (46.3%) of Goričko Nature Park, the Big Krka Basin covers 14.6 km^2 (31.6%), and the Kobilje Creek Basin covers 7.9 km^2 (17%). The total length of all watercourses in the nature park is 664 km: 309 km in the Ledava Basin, 95 km in the Big Krka Basin, and the rest in the Kobilje Creek Basin. These three basins cover a total of nearly 95% of the area of Goričko Nature Park.

There are differences (Internet 4) in the land use structure (Figure 1) between individual basins. The share of forest increases from the west; it accounts for 42% of land use in the Ledava Basin, 49% in the Big Krka Basin, and 52% in the Kobilje Creek Basin in the east. The Kobilje Creek Basin has a significantly smaller share of grassland (9%) compared to the Big Krka and Ledava basins, where the percentages are 16% and 18%, respectively. There are no significant differences between the basins in other land use categories.

3 Results

Pollution sources are studied in relation to discharge. In watercourses with predominantly diffuse pollution sources, pollution increases with discharge (Novotny 1988). The opposite is typical of point pollution sources, where pollutant concentration decreases with increased discharge.

3.1 Nitrogen compounds

The presence of ammonium nitrogen (NH^{4+}) in river water is the result of faecal pollution (with people and livestock farming being its main sources; Ibanez et al. 2007). The recommended ammonium levels of 0.04 mg/l

Table 1: Correlation coefficients at sampling sites

r^2	Ledava Nuskova	Ledava Domajinci	Bodonci Creek	Bokrači Creek	Adrijanci Creek	Dolenci Creek	Big Krka	Small Krka	Kobilje Creek	Bukovnica Creek	Bogojina Creek
[WaterT]/[AirT]	0.92	0.88	0.92	0.93	0.93	0.85	0.87	0.94	0.94	0.92	0.91
[WaterT]/[Oxygen concentration]	–0.50	–0.84	–0.89	–0.87	–0.80	–0.76	–0.42	–0.88	–0.07	–0.82	–0.84
[Nitrite]/[Ammonium]	0.27	–0.27	–0.17	0.03	–0.11	0.00	0.33	0.31	0.76	0.02	0.46
[Turbidity]/[Discharge]	0.02	–0.05	0.55	0.89	0.77	0.61	0.47	0.75	0.37	0.48	0.86
[Turbidity]/[Undissolved matter]	0.94	0.93	0.95	0.88	0.99	1.00	0.35	0.10	0.87	0.90	1.00
[Undissolved matter]/[Discharge]	–0.01	–0.15	0.75	0.93	0.77	0.62	0.14	0.36	0.33	0.75	0.87
[Orthophosphate]/[Undissolved matter]	0.79	–0.06	0.70	0.91	0.87	0.47	0.15	0.25	0.22	0.16	0.90
[Total phosphorus]/[Undissolved matter]	0.95	0.73	0.91	0.87	0.99	0.58	0.53	0.39	0.81	0.58	0.98
[Nitrate]/[Discharge]	0.95	0.47	0.67	0.81	0.59	0.57	0.31	0.91	0.45	0.27	0.76

specified in the Decree on the Quality of Surface Waters for the Life of Freshwater Fish Species for Salmonid Waters (Uredba … 2002) are regularly exceeded at all the sampling sites. It can be established that the average ammonium levels at the Ledava Nuskova sampling site before its inflow into Lake Ledava are lower than at the Ledava Domajinci sampling site after its outflow from Lake Ledava. On the other hand, the mean nitrate levels (NO^{3-}) before Lake Ledava are higher than after Lake Ledava. Hence it can be concluded that denitrification is taking place in the predominantly anaerobic conditions in the reservoir Lake Ledava. Because the standard deviation for nitrate at the Ledava Domajinci sampling site is typically smaller than at the Ledava Nuskova sampling site, this seems to be an ongoing process.

Nitrate is a soluble form of nitrogen that usually seeps into groundwater quickly and is then released into the river water as base runoff. Table 1 shows the correlation coefficients for [NITRATE]/[DISCHARGE], $r^2 > 0.4$. It can be concluded that this results from the nitrate being washed from farmland due to poor soil permeability. The nitrate levels do not exceed the limits specified in the Rules on Drinking Water, but the levels measured in the surface watercourses are nonetheless high. A seasonal impact of nitrate being washed from farmland can also be observed, with excesses after spring or fall fertilization, depending on precipitation.

Based on what is known about the nitrogen cycle, nitrites result from nitrification processes. Compared to the levels recommended in the Decree on the Quality of Surface Waters for the Life of Freshwater Fish Species for Salmonid Waters (Uredba … 2002), these levels are elevated, and they are constant. Some sampling sites (Kobilje Creek, Bogojina Creek, Small Krka, and Big Krka) have a high correlation for [NITRITE]/[AMMONIUM], $r^2 > 70$, which may be due to nearby settlements, unregulated sewage systems, and livestock farming. Such correlations were not established at sampling sites for which the impact of settlements is smaller.

Pollution by total nitrogen compounds is presented in Figures 2 to 4. The pollution curves indicate a significant impact of diffuse sources on the watercourses in Goričko Nature Park. The impact is less pronounced at the Ledava Domajinci sampling site, which is most likely due to the influence of Lake Ledava, in which chemical processes and accumulation take place. This impact is also less pronounced at the Bukovnica Creek sampling site, which is probably due to the creek's lower flow rate, which is regulated by the artificial reservoir Lake Bukovnica. This is a tourist area, where a large number of visitors can influence the current conditions in the watercourse.

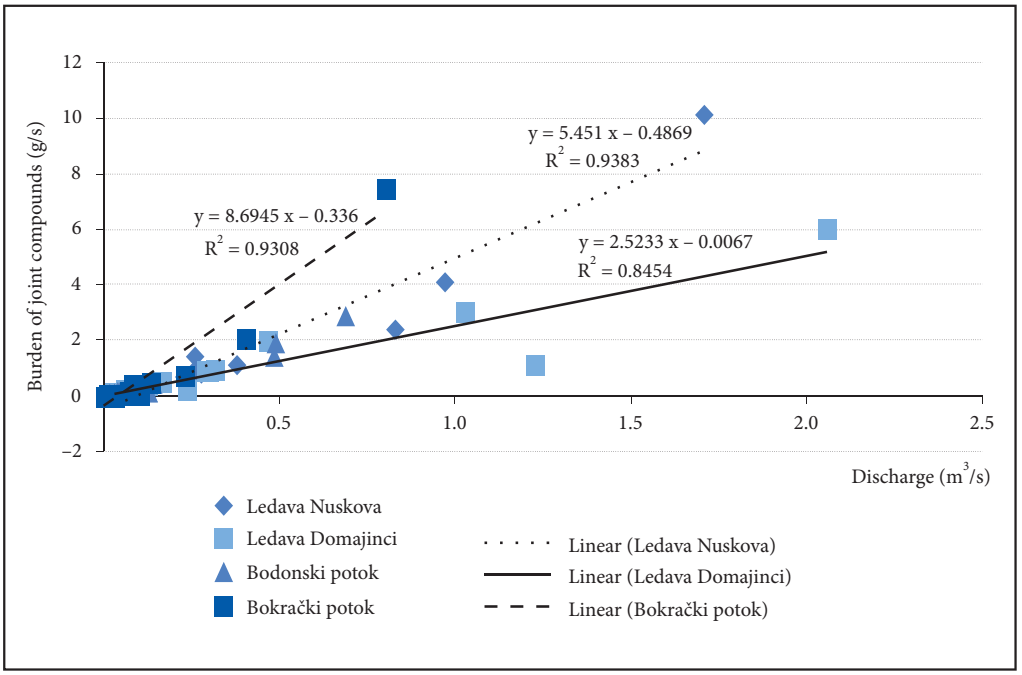

Figure 2: Total nitrogen pollution in the Ledava Basin.

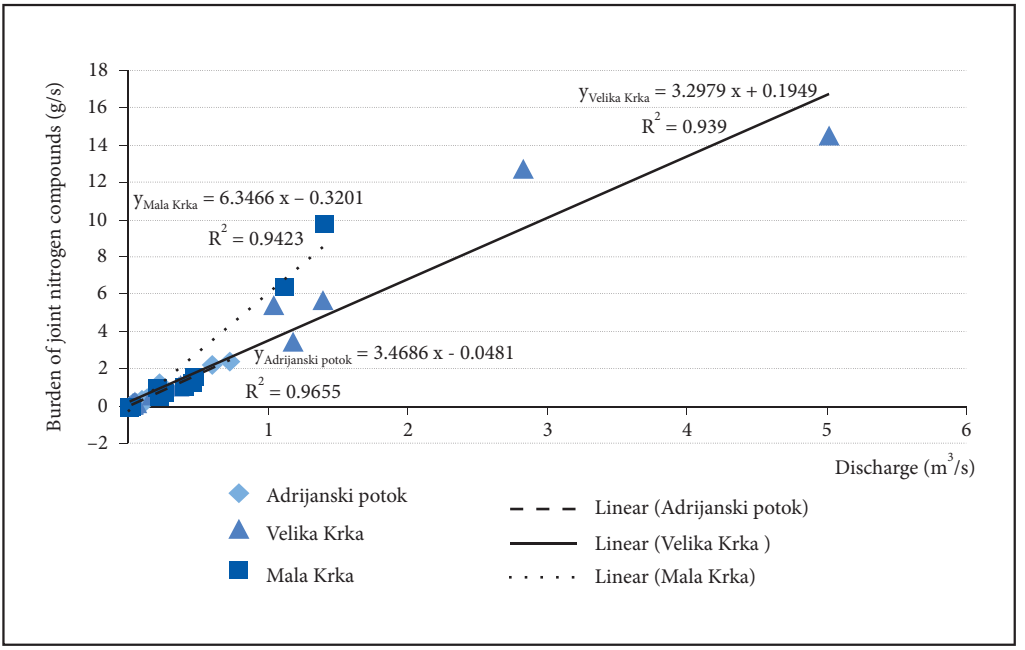

Figure 3: Total nitrogen pollution in the Big Krka Basin.

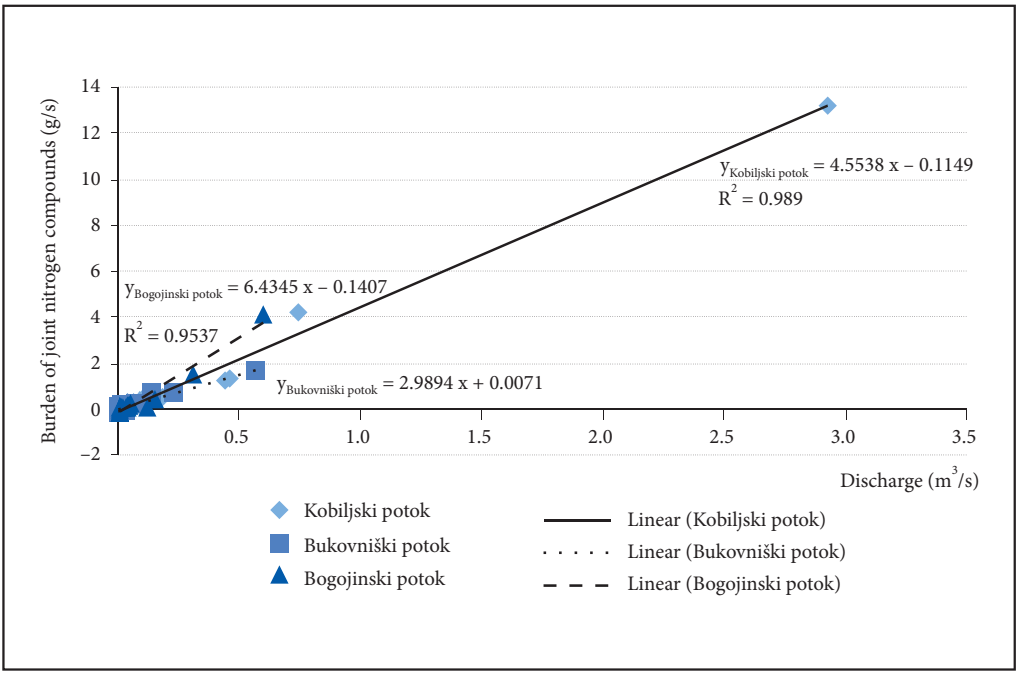

Figure 4: Total nitrogen pollution in the Kobilje Creek Basin.

3.2 Phosphorus compounds

The moderate correlation of [TOTAL PHOSPORUS]/[UNDISSOLVED MATTER], $r^2 > 0.5$ stands out. When water erodes the soil, the phosphorus bound in the soil particles is washed into watercourses (the impact of diffuse source pollution; Pierzynsky et al. 1994). During periods of low discharge, total phosphorus concentrations are typically smaller, but constant, and are the result of unregulated sewage systems and manure pits on farms (the impact of point source pollution by total phosphorus). Just like with total phosphorus, the correlation for [ORTHOPHOSPHATE]/[UNDISSOLVED MATTER] is also high: $r^2 > 0.7$ is typical of the Ledava Basin, except at the Ledava Domajinci sampling site, where there is hardly any correlation. It can be concluded that this is due to the accumulation of orthophosphate in Lake Ledava, which is a hypertrophic lake according to the OECD criteria (Poročilo … 2007).

Pollution by total phosphorus compounds is shown in Figures 5 to 7. The impact of point sources on pollution in the Ledava Basin is small, but constant. Based on these results it can be concluded that unregulated sewage systems and farming (livestock breeding) contribute to both point and diffuse source pollution.

4 Discussion

Extensive surface water quality monitoring was performed in Goričko Nature Park in order to determine the level of nitrogen and phosphorus pollution and other accompanying parameters. The study area was divided into three subareas or third-order basins: the Ledava, Big Krka, and Kobilje Creek basins. Measurements were taken once a month over the course of 1.5 years. Pollution in relation to discharge was calculated for each sampling site. The results show that the nutrient release dynamics in Goričko Nature Park are in high correlation with precipitation events. Similar dynamics have also been established for the Krka River Basin (Drolc 1998) and the Padež Basin (Rusjan 2008). The chemical composition of the waters included in the study is poor at all sampling sites. During the study the total nitrogen concentrations were high and fairly stable. Ammonium and nitrite (as an intermediate product of nitrification) stand out more than nitrogen.

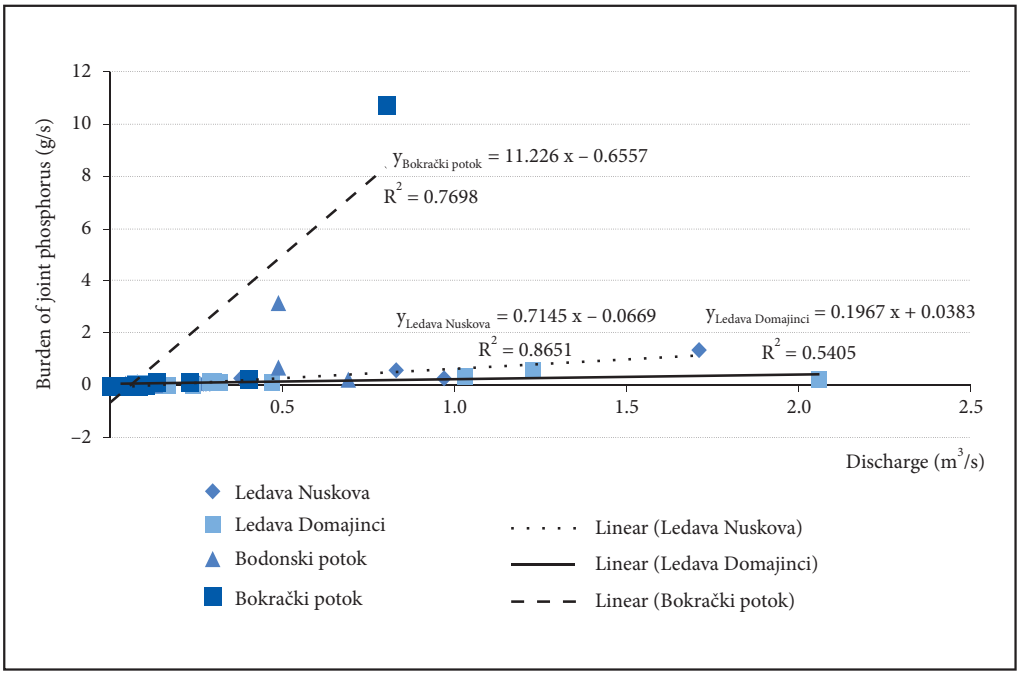

Figure 5: Pollution by total phosphorus compounds in the Ledava Basin.

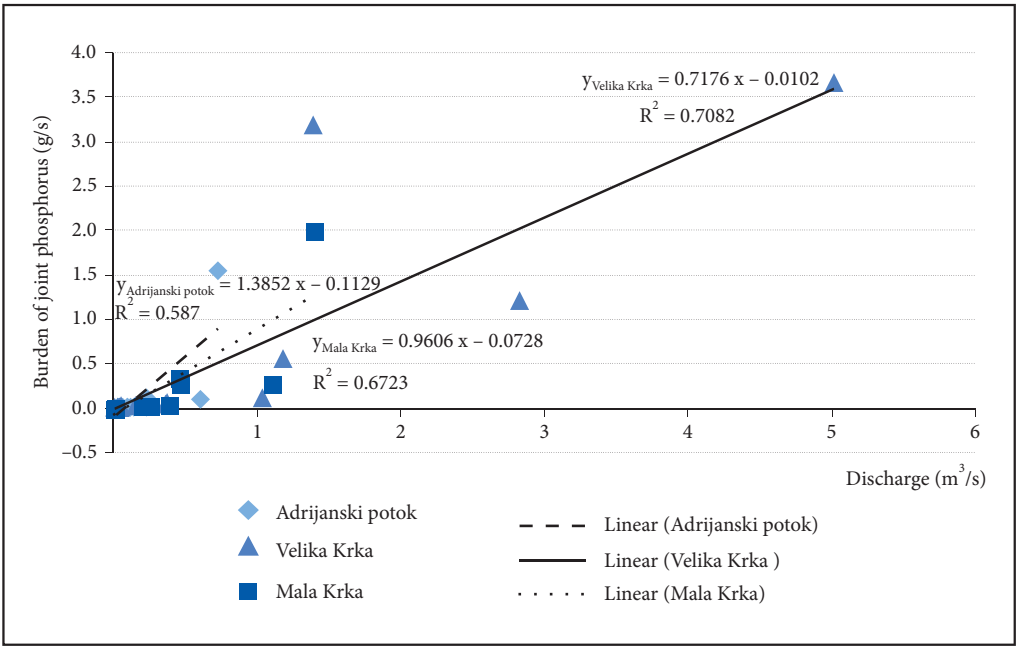

Figure 6: Pollution by total phosphorus compounds in the Big Krka Basin.

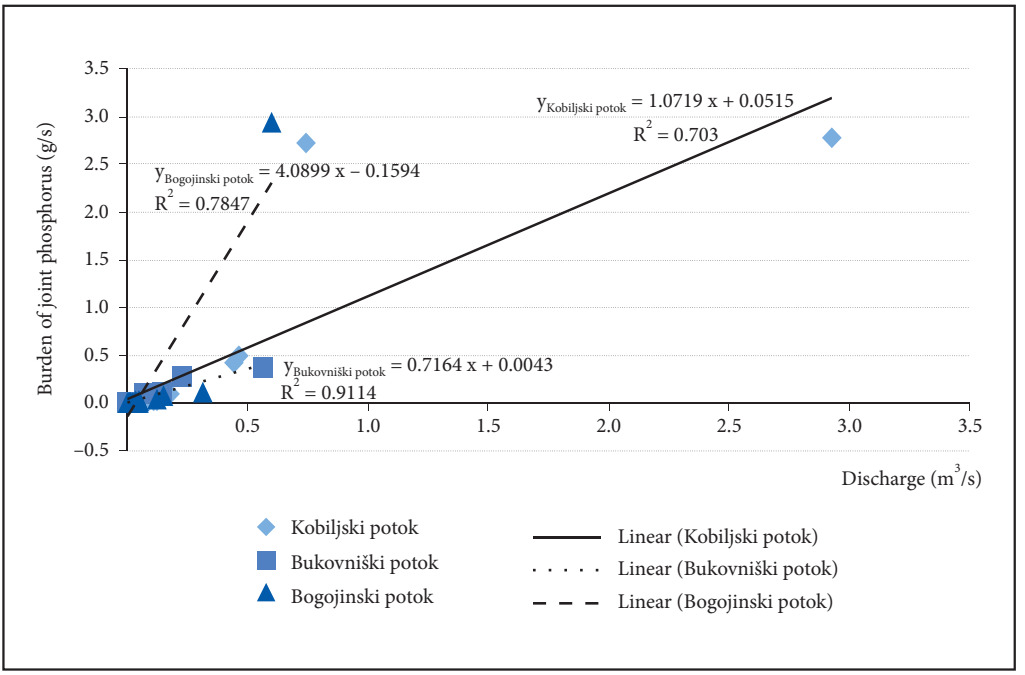

Figure 7: Pollution by total phosphorus compounds in the Kobilje Creek Basin.

It can be concluded that unregulated manure pits on livestock farms are the main reason for the high levels of ammonium release. In a study conducted by Lapajne (2006), no final conclusions were reached on the high ammonium levels in the Ledava Valley, but the researchers did conclude that they were due to emissions from the livestock farms or wastewater from unregulated sewage systems. The main reasons for the high levels of nitrogen compounds lie in the chemical composition of nitrogen and the nitrogen releases from intensively farmed land (Eickhout et al. 2006), as well as the low effectiveness of nitrogen fertilizers (Strebel et al. 1989). Beaudoin et al. (2005) established that the levels of nitrates washed from farmland depend primarily on the type of soil: low levels were determined in deep, poorly permeable clay soil and the highest levels were found in shallow, permeable sandy soil. Ju et al. (2006) established a high correlation between the intensity of farming and nitrate levels in groundwater. Another important factor in nutrient release is the natural conditions that affect land use and the use structure of farmland.

The total phosphorus concentration is highly correlated with precipitation events, because it binds to suspended particles and erodes into the drains. Likewise, the excessive levels of phosphorus compounds in the Ledava River also result from farm runoff via precipitation (Lapajne 2006). Sharpley et al. (1999) also determined that the concentration of phosphorus compounds increases with precipitation, and Hanrahan et al. (2003) concluded that the majority of phosphorus transfer takes place periods of intense precipitation.

Measures for reducing nutrient pollution in rivers should focus on decreasing the nutrient concentrations at the outflows through tertiary treatment at treatment plants and significant reduction of inputs from agriculture. Drolc and Končan (2002) believe that by implementing all of these water management measures, the total phosphorus emissions in river basins could decrease by 40%. In order to reduce the nutrient release caused by diffuse pollution, certain measures have been proposed (Internet 2) to increase fertilizer effectiveness and hence decrease erosion from farmland (Komac and Zorn 2005; Zorn 2009), and to promote unconventional farming in environmentally more sensitive regions. Some measures for decreasing diffuse sources of pollution do not comply with agricultural practice and economics. Tertiary treatment of phosphorus and nitrogen at large treatment plants can also help reduce water pollution. Despite the work carried out by large treatment plants, minor point sources of pollution still remain a problem. In order to solve it effectively, tertiary treatment at small treatment plants should be introduced. These treatment plants release water into small, environmentally more sensitive creeks in the countryside (Wheater and Daldorf 2003), where tertiary treatment could help reduce the pollution of river basins.

5 Conclusion

The acquired data show that rivers have only moderate thermal potential and that weather has the most significant impact on watercourse conditions. This is primarily reflected in the high correlations between water and air temperature. Subsequently, the thermal potential of watercourses has a strong impact on the concentration of dissolved oxygen in water. A comparative analysis was conducted using the data collected from eleven sampling sites used to monitor the surface waters in Goričko Nature Park. Because the study area was divided into three parts, the data obtained were also compared by river basin. All three basins are characterized by increased concentrations of nitrogen and phosphorus compounds, which resulted in poor chemical composition of surface waters at all sampling sites.

The results of this study show that the watercourses in the entire study area are polluted by nitrogen and phosphorus compounds. A trend of significant nitrate increase can be observed in the Mura Basin and the watersheds of Adriatic rivers, and orthophosphate pollution is increasing as well (Internet 5).

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