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Loading and dilution: arsenic, sodium and nutrients in a section of the River Tisza, Hungary

Abstract: We aimed to reveal the risk of arsenic in a Hungarian river (the Tisza) at the mouth of a polluted canal. Four sampling sites were involved in this work and samples were collected on a weekly basis for arsenic and sodium, and on a monthly basis for nutrients. Significant differences were found concerning each studied component between the sampling locations of the River Tisza. Statistical analysis also revealed that the values of the upper and lower river tracts did not differ significantly. Thus, water carried by the canal is being diluted before it reaches the farthest sampling location.

1 Introduction

Water found in natural conditions is never available in a chemically pure state. As an excellent solvent, its composition is determined primarily by its origin, and the exact type and nature of water can be established based on this information. There are certain components whose concentrations would exceed hygienic limits even under natural conditions [1, 2]; arsenic is one such element. Arsenic is proven to have carcinogenic effects both in its organic and inorganic forms [3, 4].

Arsenic is toxic, and small concentrations in drinking water can pose considerable risk for living organisms. In water, arsenic is mostly present in its inorganic form, either as arsenate [As(V)] or arsenite [As(III)], depending on the oxidative/reductive nature of the environment [5, 6]. Ground water with high arsenic content is usually slightly alkaline (pH= 8-9). In highly oxygenated waters, the As(III) →As(V) transformation usually occurs; however, this process would take several months. On the other hand, the reverse reduction results from bacterial processes [7]. The arsenic content of ground waters may be elevated due to mining in the vicinity, but it often appears in ground water and stratum water under natural conditions. Arsenic is relatively rare in its elemental form, and its compounds are formed with oxygen, iron, chlorine and/or sulphur [8, 9]. Arsenic compounds are constant companions of pyrite and other sulphide minerals [8]. In topsoil, the average concentration of arsenic is 1.5–2 ppm; it mainly occurs in its inorganic form [10]. Our drinking water predominantly contains its inorganic form, whereas in our food arsenic can be found both in organic and inorganic forms. In most cases arsenic enters the human organism through these two factors (water and food).

The largest arsenic poisoning in history was in Bangladesh, where residential water supply was contaminated by arsenic. This contamination was of natural origin caused by inorganic arsenic [11]. The southern part of Hungary has similar problems, but the problem was recognized very early [12, 13]. Although our study aims to reveal the risks of arsenic in surface water, ground water can have impact on it. There are several areas where the lower soil stratum contains high arsenic concentrations, due to its geological origin. If this leached into ground water, stratum or surface watercourses, the level of natural background contamination could be increased.

Water quality issues occurring in the study area's catchment are of geological origin (high arsenic contents), and also the results of the anthropogenic effects (such as communal and food industry waste waters, contaminated rainwater, livestock plants, improper agricultural cultivation, as well as water leached from illegal waste depositions). Such loads mean a taxing increase in the nutrient-

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and organic matter-contents of the surface watercourses [14, 15].

We aimed to study the concentration of arsenic and other components, their temporal changes. We also study whether upstream arsenic concentrations would affect concentrations measured in samples from the River Tisza. Furthermore, we analyzed possible correlations between arsenic concentrations and water volume, and between arsenic concentrations and other contaminants.

2 Experimental method

2.1 Study area

In the study area, arsenic is present as a natural contaminant. Canal Lónyay was created in the 19th century in order to drain and exsiccate areas without any outlet. The first measurements of arsenic concentration took place in the mid-1980s. Based on the experiences of the Southern Hungarian Plain, at first such measurements were carried out only in samples taken from the deeper wells of the regional waterworks. Regarding the origins of the arsenic content, several theories exist in the literature; the most plausible of them is that it originates from the Transylvanian Ore Mountains. Due to oxidative erosion, the primary magmatic and post-magmatic arsenic sulphide and arsenide ores of the Transylvanian Ore Mountains would become water-soluble arsenates. In this form, the mobility of arsenic may increase significantly [16–20]. Following transportation, it would precipitate in the euxinic basin's anoxic environment. Its accumulation is promoted by iron and organic substances, because ferric hydroxide would adsorb and bind arsenic from the arsenate solutions [21, 22]. High Arsenic concentration of natural origin can be measured in peak levels in the Nyírség region (Kótaj, Nagykálló, Máriapócs, Hodász), as well as in the environment of Nagyecsed (located at the border of the Nyírség and the Szatmár-Bereg Plain) [23, 24]. The bed of the canal, meandering among the sand-hills of the Nyírség microregion, is lower in many places than the surrounding ground water level. Because of the movement of ground water and the effect of the canal, arsenic is leached out of the deposits, and thus, the level of background contamination increases naturally. The luxuriant growth of vegetation at certain places of the river bed would indicate reprehensible water quality.

The river (Tisza) and canal (Lónyay) have significantly different discharge. While the mean discharge of Tisza is about 400-450 m^3s^{-1}, the Lónyay flows in a relatively drought environment, its discharge is hardly 5 m^3s^{-1}.

2.2 Sampling locations

We designated three sampling spots along the River Tisza, namely: (T1) the ferry of Tiszabercel (568.6 rkm); (T2) 360 m downstream from where the Canal Lónyay flows into the Tisza (558.9 rkm), and (T3) at the border of Szabolcs village (555.0 rkm), about 4.5 km downstream from where the canal meets the river. In case of the Canal Lónyay the samples were taken at the bridgehead of the public road bridge of Gávavencsellő (L1). Sampling was carried out at 20 cm depth. The source data set is continuous, with the exception of the winter-time icing. In the course of wandering all over the location, the preliminary assumption – i.e. that the area of investigation has no surface inflow other than the main canal – was confirmed. Nutrients were measured on a monthly basis, whereas the measurements of arsenic and sodium concentrations and water conductivity were performed on a weekly basis. The catchment of the canal is located entirely in Hungary. Thus, it is the first potential source of contamination that may introduce contaminant loads into the River Tisza solely from Hungarian sources (Figure 1). This was important because during the measurements in this section we were able to investigate exclusively the effects of the Canal Lónyay.

2.3 Analysis of arsenic

During sample preparation 0.01 L concentrated hydrochloric acid (HCl) of analytical grade was added to each 0.02 L filtered water sample [1]. The analysis was performed in the NanoFood Laboratory of the University of Debrecen, by a "PS Analytical Millennium Merlin" instrument. In the water samples only two inorganic arsenic components were present, and the pH-dependence of the process enabled us to determine both the arsenite and arsenate concentrations, even without applying any preliminary separation technique methods. Because of the added hydrochloric acid, the pH of the water samples dropped below 2.0, thus, we were able to determine the joint concentration of arsenite and arsenate.

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1 MSZ EN ISO 11969, Water quality. Determination of arsenic. Atomic absorption spectrometric method (hydride technique).
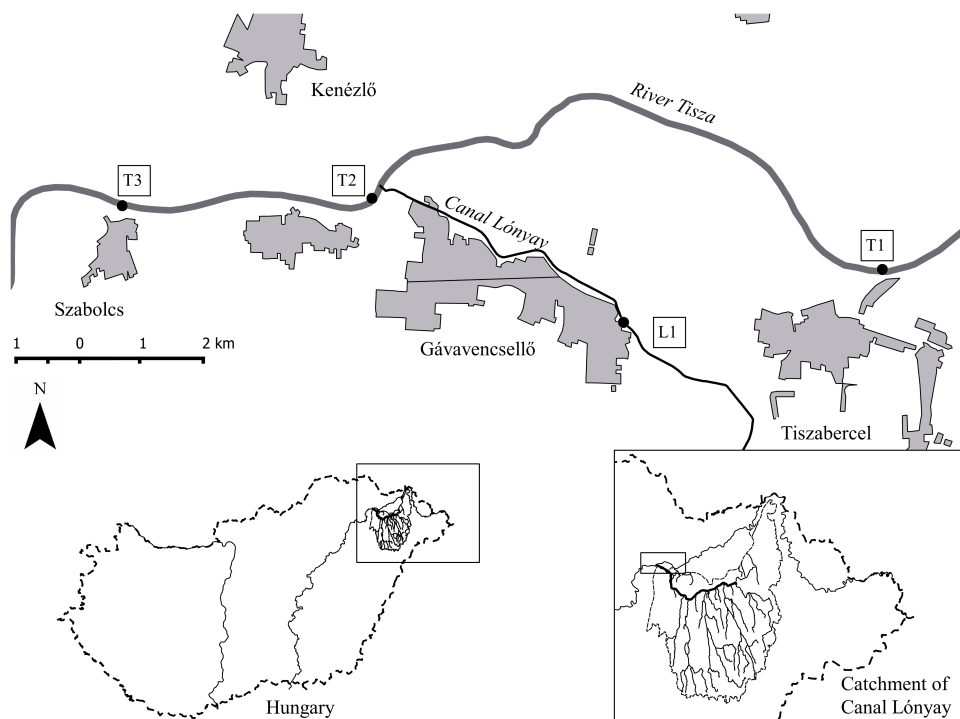
Figure 1: Location of the Canal Lónyay with its catchment, and the sampling sites.

2.4 Determination of other parameters

Because the area is intensively cultivated, and communal sewage is introduced into surface water via a number of ineffective sewage-treatment plants, nutrients (NO_3^-, NH_4^+, PO_4^{3-}) concentration, and electrical conductivity were also investigated. The concentration of these variables may be higher in Lónyay than in Tisza. Thus, the study of these parameters may be important in the aspect of contamination. Sodium is not typical in this region; thus its presence indicates anthropogenic input [25]. The specific electrical conductivity and the pH were measured in situ, while the NO_3^{-2}, NH_4^{+3}, PO_4^{3-4} contents were determined in the laboratory of the Department of Physical Geography and Geoinformatics of the University of Debrecen.

2.5 Statistical evaluation

The variables were checked with the Shapiro-Wilk test, and were found to follow a normal distribution. Accordingly, we applied variance analysis (ANOVA) for hypothesis testing. Homogeneity of variance was controlled with the Levene test. Data were compared by sampling sites and seasons, differences between the groups were estimated by the ANOVA, and the Tukey test was applied as post hoc testing. Correlation between concentrations and water level and discharge were identified with Pearson correlation coefficient; furthermore, we applied curve fitting using scatterplot diagrams. For the evaluation SPSS 19 for WINDOWS and the Microsoft Excel 2007 software were used.

3 Results

3.1 Analysis of arsenic

In the Canal Lónyay (L1) the arsenic concentration fluctuated between 4–45 $\mu\,l^{-1}$ (with an average 16.7± 9.56$\mu\,l^{-1}$) during the study period. Between September 2010 and March 2011 the measured values – due to the higher volume and frequency of rainfall – varied between 5–20 $\mu\,l^{-1}$ (Figure 2). From April 2011; however, the changes would

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2   MSZ 12750-18:1974 Testing of surface water. Determination of nitrate ion.

3   MSZ 260-9:1988 Wastewaters analysis. Determination of ammonium content.

4   MSZ 12750-17:1974 Testing of surface waters. Determination of forms of phosphorus.
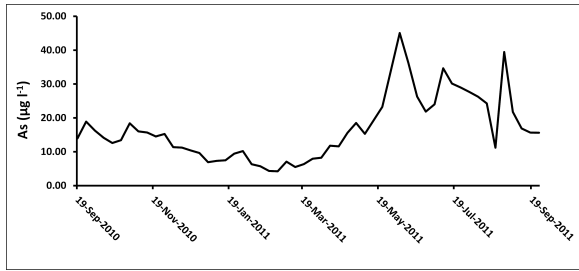
Figure 2: Changes of arsenic concentration in the Canal Lónyay during a one-year time period (19.09.2010 and 19.09.2011).
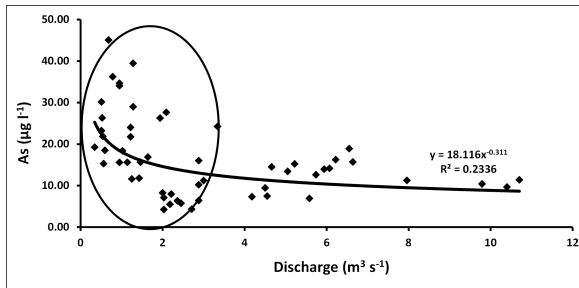

Figure 3: Connection between arsenic concentration and discharge at the L1 site, Canal Lónyay (circle highlights the relation between lower discharge and higher As-concentrations).

Figure 4: Quantitative changes of arsenic concentration along the Tisza River.

Figure 5: Connection between arsenic concentration and discharge at the T1 site, River Tisza (circle highlights the relation between lower discharge and higher As-concentrations).

become striking. Due to lower rainfall, we observed larger fluctuations and an increase of concentrations. The increase was not significant, which means that no contamination of anthropogenic origin occurred in the studied period.

By plotting the measured values against discharge, a characteristic power type relation could be established. As an effect of the increasing rainfall, the surface outflow increased, and the concentration (due to dilution) decreased (Figure 3). A large proportion (63%) of the data fluctuated in the 0.5–3.0 $m^3 s^{-1}$ range.

In the River Tisza, arsenic concentration values were much lower, compared to those measured in the canal. The reason for this is that the Tisza River's average discharge was about 400 times higher than that of the main canal. The arsenic concentrations of the individual locations along the River Tisza were as follows: T1: 0.09–1.68 μl^{-1} (average 0.58 ± 0.47 μl^{-1}); and T3: 0.06–3.12 μl^{-1} (average 0.73 ± 0.60 μl^{-1}). The highest variances in the results were found at T2. In that case, the values varied in the 0.15–4.26 μl^{-1} range, with an average 1.16 ± 1.10 μl^{-1} (Figure 4). The reason for this is that the Canal Lónyay meets the river nearby, and a minor mixing and slow thinning occurs. The river advances more than 4 km until the chosen sampling point of Szabolcs (T3), which is enough for a
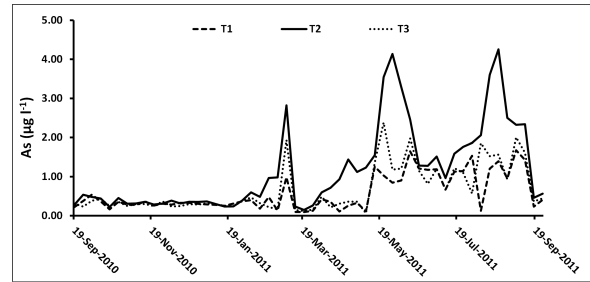
complete mixing. As such, the values at T3 fluctuate similarly to those of the sampling point at T1 [26].

Plotting the concentration values of the sampling points along the Tisza River against the water output (Figures 5, 6, 7), also reveals a power-type correlation. A significant proportion (74%) of the data fluctuated within the 100–400 $m^3 s^{-1}$ range.

3.2 Seasonal changes of arsenic at the individual sampling locations

Spring and summer concentration values showed greater fluctuations. Due to the lower temperatures in winter and autumn, as well as the cold, the measured arsenic concentrations decreased. In the warmer months – with the beginning of the vegetative period and concomitantly, the initiation of soil cultivation, and because of the increased rainfall – the rate of leaching increased. As an effect of the higher amount of precipitation the ground water rose, and leaching increased, too. As a result, both the concentrations and their variance increased (Figure 8a). Consequently, we found that in every season the concentrations measured at T2 were higher than those at the other two locations (T1 and T3) on the River Tisza (Figure 8b).
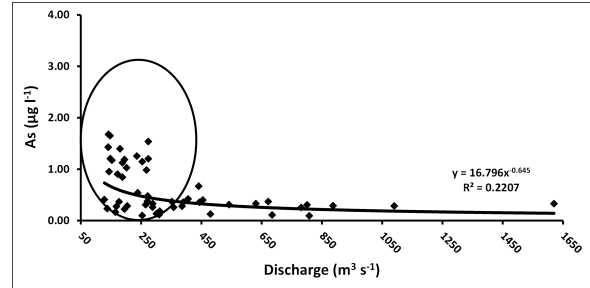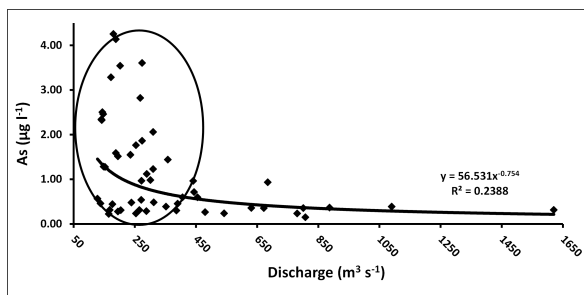
Figure 6: Connection between arsenic concentration and discharge at the T2 site, River Tisza (circle highlights the relation between lower discharge and higher As-concentrations).

Figure 7: Connection between arsenic concentration and discharge at the T3 site, River Tisza (circle highlights the relation between lower discharge and higher As-concentrations).
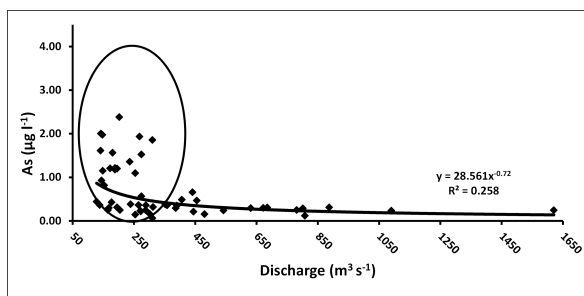
3.3 Variance of ammonium, nitrate, orto-phosphate and sodium

By plotting the distribution of ammonium (Figure 9a), nitrate (Figure 9b), orto-phosphate (Figure 9c), and sodium (Figure 9d) concentrations on box-plot diagrams, similar fluctuations can be found to those of arsenic (Figures 2, 4). These two parameters also showed that in the rainier period (September 2010 – March 2011) the fluctuations were more moderate, and the measured concentrations were lower, compared to those of the period with less rainfall. In the relatively arid period the fluctuation rate increased. Peak concentration levels were reached during the arid period. With the start of spring vegetation, and the more intensive use of artificial fertilizers that comes with it, the elements of the vegetative nutrient turnover show a large leap in their concentrations [27]. The highest level of increase was found in case of orto-phosphate (Table reft:1).

3.4 Effects of sampling date and location

The Mann-Whitney test revealed that, in terms of arsenic concentration, the samples of the Canal Lónyay (L1) were significantly different (p< 0.05) from those of the Tisza

River. If only the sampling points of the River Tisza are taken into consideration, the results showed that the post-confluence section differs significantly from the results of the two other samples of the Tisza. This difference proves the modifying effect of the canal. In terms of seasonal fluctuation, the vernal months differ significantly only from those of summer and winter, whereas the summer months differed from all others (Table 2). Arsenic concentrations at T1 and T2, and at T2-T3 were different (p< 0.05; Table 3).

Further connections were sought between the measured values of arsenic, sodium and conductivity. To that end, a correlation analysis was also performed (Table 4).

The weakest connection was found between the L1–T3 pair, which clearly showed that the two sampling locations were different. The strongest correlation was found between the concentration values at the T2-T3 sites (Table 5).

The correlation analysis was completed for each individual parameter as well, regardless of the sampling locations (Table 6). In this case, strong correlations were found between several components. The strongest of these turned out to be between arsenic and orto-phosphate. The arsenic-sodium, arsenic-conductivity and sodium-conductivity correlations also proved to be quite strong.

4 Discussion

Dilution of the measured chemical parameters can be traced with the recorded data of the sampling sites (Table 1). The Tisza has 80 times the discharge of the Canal Lónyay, but the canal is a relatively polluted watercourse. As such, the canal caused significant differences in each parameter. At the confluence, concentrations of EC, As, $Na+$, NO_3^-, NH_4^+, PO_4^{3-} were significantly higher than at sampling locations upstream and downstream of the confluence (Table 3). This confirms that contamination of anthropogenic origin can be detected around the confluence. However, the effects are only local and negligible, because – due to the dilution and mixing – the rate of such changes would be subdued by the time we reach the farther sampling location.

Regarding vegetative nutrients, the water of the canal was about 5–8 times more contaminated than the River Tisza. This is the result of several factors: (1) The area is influenced by intensive agricultural activity and as an result of intensive use of artificial fertilizers, more mobile elements are washed by rain into the surface waters; thereby increasing background contamination [28–33]. (2) Sewage-treatment plants along the canals and the Tisza
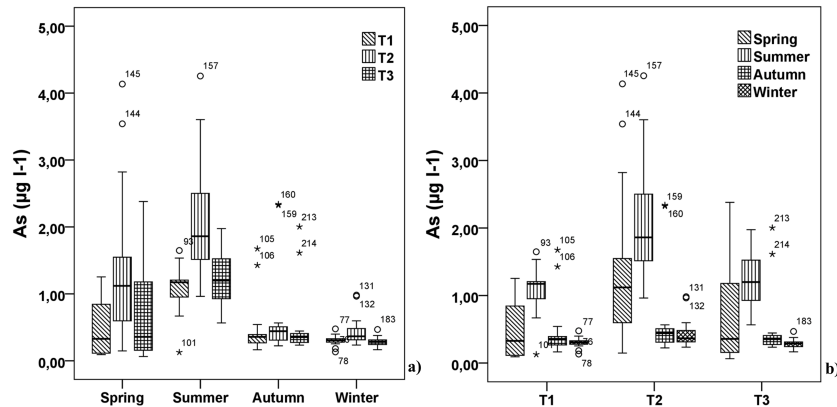
Figure 8: Concentration changes of arsenic concentration by seasons (a), and sampling sites (b). (Diagram boxes represented the interquartile range with lower quartile, median upper quartile and whiskers showed the 1.5 time interquartile range; o: outlier data; : extreme data).
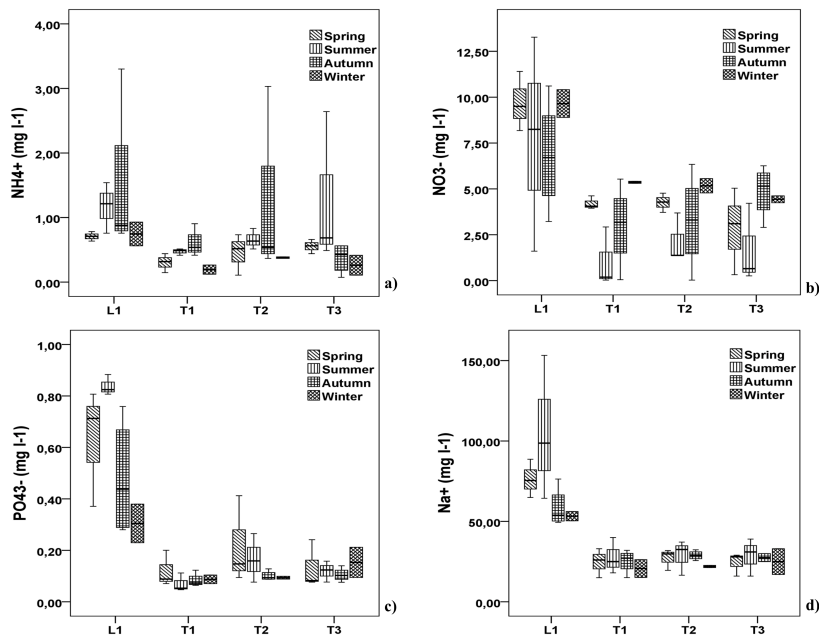
Figure 9: Seasonal fluctuation of the values of water chemical parameters at the individual sampling sites (diagram boxes represented the interquartile range with lower quartile, median upper quartile and whiskers showed the 1.5 time interquartile range).

Table 1: Statistical values of water-chemical parameters (mean ± SD) at the studied sampling locations (EC: electric conductivity; L1 and T1-T3: sampling sites, see Figure 1).

	L1	T1	T2	T3
NO_{3-} (mg l^{-1})	8.2 ± 3.5	3.0 ± 2.1	3.3 ± 2.1	3.3 ± 2.1
NH^{4+} (mg l^{-1})	1.1 ± 0.8	0.4 ± 0.2	0.7 ± 0.8	0.6 ± 0.7
PO^{43-} (mg l^{-1})	0.6 ± 0.2	0.1 ± 0.04	0.2 ± 0.1	0.1 ± 0.1
Na^+ (mg l^{-1})	75.4 ± 27.8	22.5 ± 7.6	25.4 ± 8.5	22.7 ± 7.4
EC (μS cm^{-1})	969.1 ± 104.5	395.3 ± 76.4	427.7 ± 72.9	388.7 ± 77.1

Table 2: Result of variance analysis of arsenic and sodium by seasons (**$p < 0.05$**).

	Arsenic		Sodium	
	Mean difference	P value	Mean difference	P value
Spring-summer	-0.63	**0.001**	0.74	0.973
Spring-autumn	0.32	0.196	-1.21	0.882
Summer-winter	1.08	**0.001**	4.93	**0.022**
Autumn-summer	-0.95	**0.001**	1.95	0.637
Autumn-winter	0.13	0.846	6.88	0
Winter-spring	-0.45	**0.037**	-5.66	**0.006**

Table 3: Variance analysis of arsenic by sampling sites (**$p < 0.05$**).

	Mean difference	P value
T1-T2	-0.58	**0.001**
T1-T3	-0.14	0.621
T2-T3	0.44	**0.014**

are aging (e.g. in Gávavencsellő) and overloaded. Concentration changes of sodium were informative of both the increased water usage, and also, poor effectiveness or faultiness of the treatment process[5]. (3) Due to the geological structure and history of the microregion where the study area is situated, several thermal spas were established, and, since the thermal water contains other components as well, their waste water increases the concentration of sodium indirectly (through the groundwater) or directly (poured into the canal)[5]. (4) Arsenic-rich deposited sediments – from the Transylvanian Ore Mountains – are constantly flushed and leached by the water of the canal, as a result of which the natural arsenic contamination of the surface water increases [24].

Several Indian and Chinese studies have been published which also demonstrated that in areas where the arsenic concentrations are high in the soil, and leaching is considerable (due to rainfall), surface waters are loaded with more contamination [34, 35]. The same conclusion can be drawn in the case of the catchment of the Canal Lónyay. The bed of the canal is often deeper than the actual underground water-level, so there is direct interaction between surface and ground water.
The actual concentrations of contaminants are highly influenced by the quantity of rainfall in the area. In the present study, during the wetter period we measured lower

arsenic concentrations than in the comparatively arid period. As soon as the amount of rainfall decreased, and its temporal and spatial distribution became irregular, the concentrations – and simultaneously, the rate of fluctuation – increased. From this phenomenon we can conclude that higher concentrations were a result of slower mixing and thinning. The more the rainfall, the bigger the discharge, and the stronger the rate of dilution. This is clearly shown in Figure 3, which shows that the majority of contaminants were present at around 0.2–4 m^3s^{-1} water flow values. Based on the determination coefficient, it was established that there is no significant correlation between the water flow and measured concentration values. The reason for ability to affect the values measured at the River Tisza is its high concentration.

Regarding seasonal fluctuations, we can conclude that the summer months differed significantly from all other months (Table 2). Temporary, sudden, and high volume downpours can explain the existence of such difference. it Islam [36] and his colleagues came to a similar conclusion during their research in Bangladesh. They proved their theory based on results of samples taken from groundwater wells, surface water, and soils. Regarding Hungary, *Rowland et al.* [37] reached similar conclusions: they collected both surface and ground water samples from several Hungarian and Romanian locations, and they determined the origin of the arsenic, and also, the geochemical changes it undergoes. During the tests they measured concentrations between 0.5–240 μl^{-1}.

We also established that sodium, and (of the studied elements of the vegetative nutrient turnover) ortophosphate showed a close correlation with arsenic. Arsenic is present in the highest amounts in sulphate and phosphatic ores, which can be found in the Transylvanian Ore Mountains [18, 34, 38, 39]. This proves the geological origin, as well as the close connection between the two components. This hypothesis has been reinforced by the results of the research of *Romić* [40] and colleagues, conducted in Osijek for several years. Our Southern neighbors

5 VKKI and FETIVIZIG., A Víz Keretirányelv hazai megvalósítása: 2-3 Lónyay-főcsatorna vízgyűjtő. Vízgyűjtő-gazdálkodási terv. Nyíregyháza, Hungary, 2010.

Table 4: Correlations between parameters measured at the individual sampling locations (EC: electric conductivity; DC: discharge; L1 and T1-T3: sampling sites, see Figure 1).

	L1			T2		
	As	**EC**	**Na$^+$**	**As**	**EC**	**Na$^+$**
EC	0.3292			-0.0122		
Na$^+$	0.5951	0.5821		0.2235	0.3861	
DC	-0.4125	-0.4158	-0.5486	-0.3939	-0.2375	-0.5605
	T1			**T3**		
	As	**EC**	**Na$^+$**	**As**	**EC**	**Na$^+$**
As						
EC	0.0645			0.08747		
Na$^+$	0.3804	0.6348		0.3338	0.5672	
DC	-0.4019	-0.3558	-0.5269	-0.3967	-0.3207	-0.5483

Table 5: Spearman correlation coefficients between arsenic concentrations measured at the individual sampling locations (L1 and T1-T3: sampling sites, see Figure 1).

	L1	**T1**	**T2**
T1	0.5998		
T2	0.5917	0.7215	
T3	0.5482	0.7912	0.8278

Table 6: Spearman correlation coefficients between each water-chemical parameters (significant correlations were highlighted with bold letters, p< 0.05).

	NO$_3^-$	**PO$_4^{3-}$**	**NH$_4^+$**	**Na$^+$**	**EC**
As	0.515	**0.915**	0.357	**0.899**	**0.832**
NO$_3^-$	-	0.528	-0.124	0.559	0.659
PO$_4^{3-}$		-	0.336	**0.843**	**0.801**
NH$_4^+$			-	0.333	0.233
Na$^+$				-	**0.854**

face a similar problem. However, the arsenic concentrations measured in their area is many times higher than those at our sampling locations[6] [37, 41–45].

5 Conclusions

Based on the results of our measurements, we found the following outcomes:

With respect to all studied components – the water of the main canal is about 5–8 times more contaminated than the Tisza River. The main factors are the intensive agricultural activity and the overloaded sewage-treatment plants that release treated water into the canal and Tisza. The Canal Lónyay has direct connection with ground water; as a result, both runoff and leaching from soil can increase the quantity of contaminants in surface water.

Strong correlation was revealed between the amount of precipitation and the concentration of arsenic, sodium and nutrients. Increasing discharge also decreased concentrations of other studied elements.

Significant differences were found in concentration values and general water quality between the river tracts upstream and downstream of the confluence. The statistical analysis also revealed that the values of the upper and lower river tracts did not differ significantly, i.e. the water carried by the canal is being diluted and mixed before it reaches the farthest sampling location, and does not affect the recipient river's water quality significantly. Nevertheless, the sample location directly downstreat of the confluence differed significantly from the two other sampling sites on the Tisza. This difference conclusively proves the modifying effect of the canal.

With this study we aimed to prove that arsenic, as a potential contaminant, is a serious issue not only in the areas of the Far East, but also here, in Hungary. However, in our study, the measured arsenic concentrations were found to be significantly lower, when compared to other regions. Our results also demonstrated that low arsenic concentrations do not considerably affect fauna, or the health of the people living along the banks of the river. We also established that such a small quantity (in the $\mu \, l^{-1}$ range) can modify the values measured in the Tisza River only to a small extent, and even that is only of minor and local significance.

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6 Upper-Tisza Environment and Water Authority., Gulács-Jánd protected areas report. FETIVIZIG, Nyíregyháza, Hungary, 2007.

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