

Technical paper

Hydrological Effect on the Chemical Status of Groundwater

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Dedicated to the memory of professor Vojko Ozim

Abstract

Different Institutions carried out several monitorings of groundwater quality within the Ljubljana area. Our objectives were to collect data from different programmes and use them for modelling. The groundwater flow and mass transport models were constructed with the help of geohydrological and geophysical information using, Modflow software. By taking the hydrological characteristics of Ljubljansko polje into account, we developed spatial maps showing expected values for electrical conductivity, nitrate, atrazine, and chromium. We investigated areas strongly influenced by the River Sava, areas with high levels of vulnerability and numerous sampling places, and which are not representative of this area. We determined trends for nitrate, atrazine and chromium. Analyses over several years have indicated that sooner or later, the diversity of pollution in Ljubljansko polje could be measured at the Hrastje site.

Keywords: Groundwater, water quality, hydrology, Ljubljansko polje

1. Introduction

A fast growing economy and a more demanding population in Europe is leading to increased socio-economic pressures, thus causing large-scale land usage and landscape changes. This is putting increasing pressure on water resources and conflicts are arising regarding water distribution and of water quality degradation. Consequently, several modelling procedures have been developed to evaluate and predict water quality and quantity in regard to the affects of anthropogenic activities.¹⁻⁶

Ljubljana's drinking water supply comes from groundwater. This study is based on data collected within the framework of the following monitoring programmes:

- Groundwater quality monitoring for the period 1999 to 2004, Ministry of the Environment and Spatial Planning – Environmental Agency. Access to data was made possible by the Ministry of the Environment and Spatial Planning – Environmental Agency;

- Groundwater and surface water monitoring within the area of the Ljubljana Municipality for the period 1999 to 2004. Access to data was made possible by The Municipality of Ljubljana/Institute for Environmental Protection Ljubljana;
- Groundwater quality monitoring, Public Holding Vodovod – kanalizacija Ljubljana (VOKA), for the period 1999 to 2004. Access to data was made possible by VOKA Ljubljana.

Physical, chemical and hydrogeological studies were performed on groundwater gathered from twenty different sampling sites. Here, we present data collected within the time period 1999 to 2004 for the Ljubljansko polje area. Ljubljansko polje is one of the most important aquifers in Slovenia which is, at the same time, the source of drinking water for app. 300.000 citizens of Ljubljana and its outskirts. Groundwater used for public drinking water supply, gathers in the aquifer of Ljubljansko polje, which is composed of holocenic and pleistocenic alluvium.

2. Experimental

Standardized method was used for sampling.⁸ Water was collected in polyethylene and glass bottles 0.5 m below the surface. All glass and plastic ware used for sampling and analyses were rinsed with milli-Q water. A volume 3 times greater than the volume of the well was pumped out before sample was taken and placed in a polyethylene bottle.

Standardized analytical methods were used for the determination of 4 physico-chemical variables. GC/MS Hewlett Packard was used for determining any atrazine (internal method IM/GC-MSD/SOP 034) and ion chromatograph Dionex was used for determining any nitrates (ISO 10304-1:1993),⁹ while ICP-MS (Inductively Coupled Plasma Mass Spectrometry) Perkin – Elmer apparatus and a WTW conductivity meter were employed for Chromium (ISO 17294-2:2003)¹⁰ and electrical conductivity (EN 278888: 1993)¹¹ measurements, respectively. All reagents were analytically graded. The milli-Q system was used for water purification. Eight sites equipped with piezometers and twelve wells, used for different monitoring purposes, were selected on the Ljubljansko polje (Fig. 2).

The piezometers/wells were selected on the basis of mathematical groundwater flow modelling results, as presented in Fig. 3. Groundwater – flow modelling was performed using a ModFlow 4.1 pro,¹² a mathematical modelling tool developed from US Geological Survey, and is a very well known and accepted modelling tool for making

environmental, engineering and hydrogeological predictions of flow and mass transport. It can be clearly seen that we have two main inflows to the Ljubljansko polje aquifer. The first comes from the north-western area and has connections with the River Sava. The second comes from the south-western area and has connections with the hydrogeological system of Ljubljansko barje, which is a multilayer aquifer system made from pleistocene and holocene sediments. The main idea behind selected piezometers was, in fact, that they possess the ability to obtain data from their water samples which are representative for groundwater flow – dynamic and its influences on the chemical quality of the Ljubljansko polje aquifer's groundwater. Only on this basis we were able to make a prediction regarding groundwater quality and its development in the near future. The results of numerical modelling are presented in Fig. 3. The hydrological situation is of great importance, because any possible pollution spreading through the aquifer system of Ljubljansko polje is implicit. In fact this means that a general movement of pollution can be expected from the west to the east area of the Ljubljansko polje aquifer.

Different chemical parameters were selected for laboratory analyses in order to determine any correlations regarding those flow directions, predicted using numerical modelling with mass transport occurring in the aquifer body. It is also known that some pollution sources are present within the city area and that surface water from the River Sava is affecting the hydrodynamic situation in

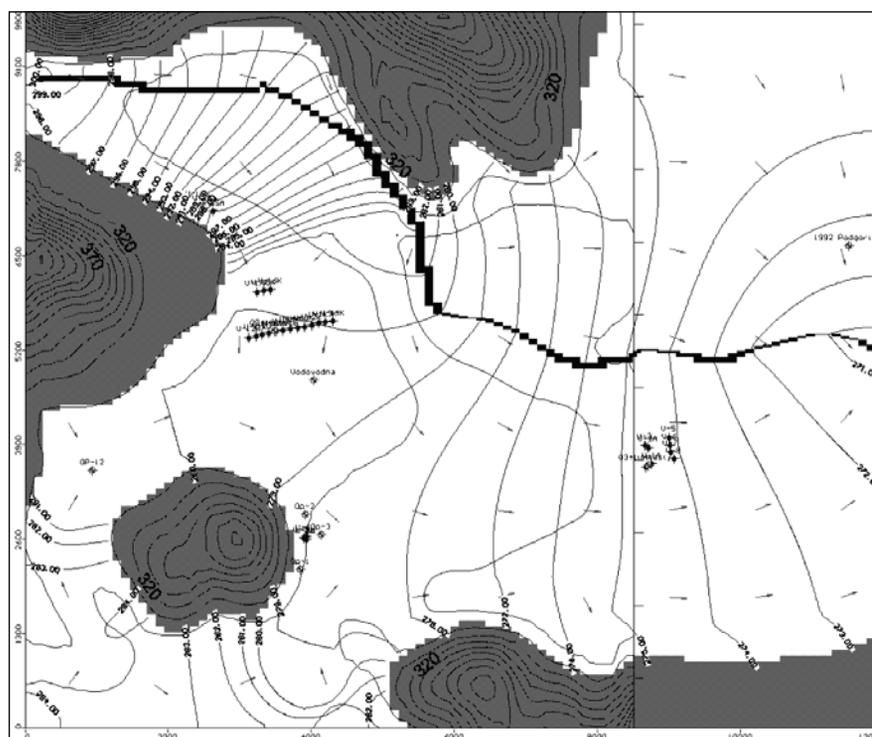


Figure 3: The direction of groundwater flow into the aquifer of Ljubljansko polje. (The scattering arrows to the south – west area are rather the result of hydrodynamic border set up than the real hydrological situation. North orientated)

the aquifer's body. By taking all the above mentioned into account, we can obtain a better understanding regarding the groundwater quality dynamics of the Ljubljansko polje aquifer. The results and their explanations are presented in the following Chapter.

2. 1. Data Analysis

The following parameters were taken into consideration for the estimation of Ljubljansko polje's groundwater quality:

- electrical conductivity,
- nitrates,
- atrazine,
- chromium.

Electrical conductivity was selected because it shows the general status of groundwater contamination, nitrates, and atrazine, since they are connected with agricultural usage and chromium because of a well-established pollution from the galvanic industry located in the Šiška area of Ljubljana.

3. Results and Discussion

Electrical conductivity (EC) estimates the total dissolved salts (TDS) amount, or the total amount of dissolved ions in the water. In the case of groundwater these are nitrates, hydrogen carbonates, phosphates, chlorides, and sulphates. EC is an indicative parameter from which different influences on groundwater can be inferred, such as the infiltration of surface or meteoric water, the influence

of a river or pollution, connected to human activity (i.e. street salting, the influence of wastewater). The average values for EC (N – number of samples taken = 474) of water samples in the area of Ljubljansko polje was $507 \mu\text{Scm}^{-1}$ (at 25 C). The results show that the EC value is between $273 \mu\text{Scm}^{-1}$ and $824 \mu\text{S cm}^{-1}$, Table 1.

In the area of Ljubljansko polje, values for EC up to $400 \mu\text{S cm}^{-1}$ (which is a consequence of the River Sava's inflow) prevail close to the River Sava (between BR12, Roje and Jarški prod). Values between $400 \mu\text{S cm}^{-1}$ and $600 \mu\text{S cm}^{-1}$ prevail in the area south of the River Sava (Kleče – Obrije – Hrastje). Figure 4 shows those areas of ground waters which come under the influence of the River Sava (blue-green fields) and those areas which come under the influence of groundwater from a south-westerly direction (yellow-red fields). Those measured values that exceed $600 \mu\text{S cm}^{-1}$ usually occur at sampling sites between Navje – Julon – Dekorativna (Fig. 4). Any increases in values can be the consequence (or are the only consequence) of pollution on the sampling site (exp. Stožice) or the improper construction of sampling sites (exp. Dekorativna). The Stožice sampling site lies within area which does not enjoy good vertical protection and, thus, the negative influences of pollutions on the surface are directly visible as increased values.

The Dekorativna sampling site lies in the part of Ljubljansko polje which enjoys good vertical protection. In spite of that, the values for EC and nitrate content are constantly increasing. On the basis of recent hydrogeological research, we know that there are several perched aquifers in the areas between Šentvid, Rožnik and Ljubljanski hrib, which do not enjoy good vertical protection (they can be polluted by sewage water). The sampling sites in this area are of an older execution and enable the flow of perched aquifers into the groundwater of the Ljubljansko polje aquifer. Because of this, we can not exclude the possibility that perched aquifers are the possible source of the pollution regarding the Ljubljansko polje aquifer.

Nitrate is an inherent component in all kinds of water, i.e. groundwaters and surface waters. Increased values of nitrates occur in those agricultural areas on which natural and artificial fertilizers are used. Furthermore, urban wastewaters and meteoric waters from agricultural areas that flow directly into the aquifer can also be looked upon as a consequence of increased nitrate values. The average measured nitrate value in the samples from the area of Ljubljansko polje (N – number of samples taken = 745) is $17 \text{ mg L}^{-1} \text{ NO}_3^-$ (Table 2). Results show that the nitrate values are between 4 and $41 \text{ mg L}^{-1} \text{ NO}_3^-$, measurement uncertainty is 12%. These values do not exceed the limit value, which is determined in the 'Rules on drinking water'.¹³

The maximal correlation coefficient of the data was found between measurements of nitrate content and electrical conductivity, Figure 5. Results are similar to those in a chemometric study.¹⁴

Table 1: The electrical conductivity of groundwater on some sampling places of Ljubljansko polje.

Sampling site	EC _{AVERAGE} ($\mu\text{S cm}^{-1}$)	EC _{MIN} ($\mu\text{S cm}^{-1}$)	EC _{MAX} ($\mu\text{S cm}^{-1}$)
Elok	459	406	490
Julon	581	510	646
Koto	525	459	620
Obrije	535	490	571
Zadobrova	524	474	550
Hrastje IA	568	496	610
Hrastje IV	601	590	610
Hrastje V–VIII	585	535	599
Hrastje ŠM	382	273	482
Kleče VIII A	448	352	492
Kleče XI	571	494	596
Kleče V–XIV	496	460	542
Stožice	463	362	690
Navje	554	515	620
Jarški prod III	493	446	530
Šentvid II A	501	433	600
Roje	383	317	458
Brod 11	422	383	480
Brod 12	392	366	457
Dekorativna	719	611	824

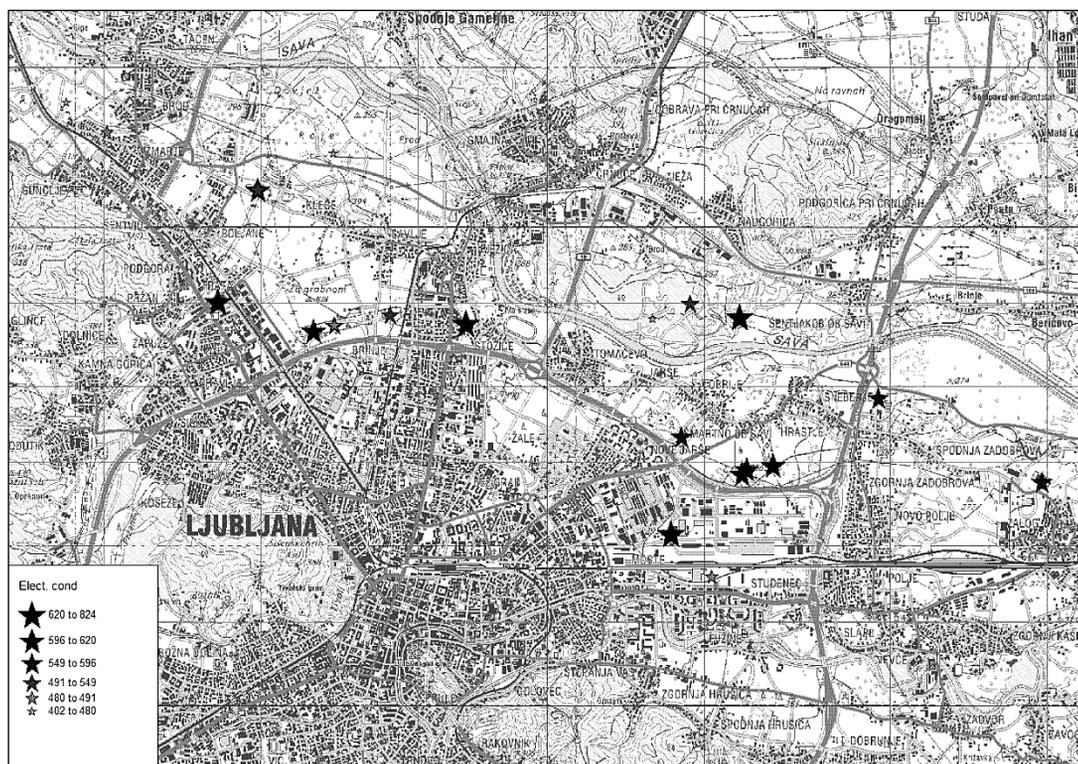


Figure 4: Distribution of electrical conductivity (in $\mu\text{S cm}^{-1}$) in the groundwater of Ljubljansko polje. (North orientated)

Table 2: The nitrate values in groundwater in the area of Ljubljansko polje

Sampling site	NO_3^- AVER. (mg L^{-1})	NO_3^- MIN (mg L^{-1})	NO_3^- MAX (mg L^{-1})
Elok	10	7.5	15.6
Julon	17.7	13.7	19.4
Koto	16	12	23
Obrije	18.2	10.2	23.5
Zadobrova	21.7	18.6	41.1
Hrastje IA	24	20	37
Hrastje IV	24.2	19.4	30.9
Hrastje V–VIII	24	22.1	27
Hrastje ŠM	9.1	3.8	13.1
Kleče VIII A	13	8	25
Kleče XI	25.2	15.5	39.8
Kleče V–XIV	16.3	12.8	27.8
Stožice	13	6	34
Navje	12.5	6.2	27.8
Jarški prod III	12.7	10.6	14.6
Šentvid II A	16.2	15	18.1
Roje	8.4	6.6	14.1
Brod 11	13.6	8.4	17.6
Brod 12	10	8	12.8
Dekorativna	32.6	29.6	34.6

As expected, the results from mathematical groundwater – flow modelling of electrical conductivity (Fig. 4) are similar to the results of the modelling of the nitrate values (Fig. 6).

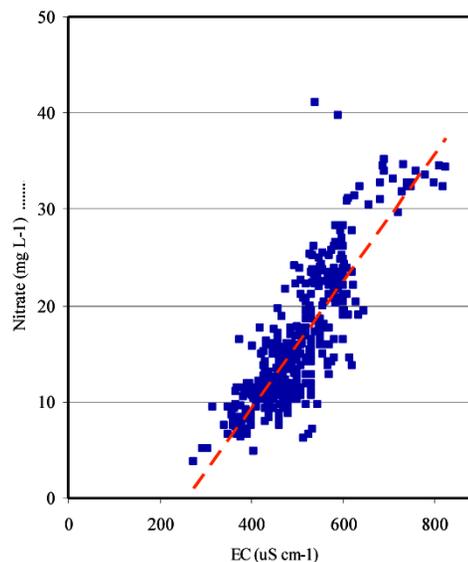


Figure 5: Correlation between nitrate content (NO_3^-) and electrical conductivity (EC)

In the area of Ljubljansko polje, the nitrate values up to $15 \text{ mg L}^{-1} \text{ NO}_3^-$ prevail close to the River Sava (between BR12, Roje and Jarški prod). Values between 15 mg L^{-1} and $25 \text{ mg L}^{-1} \text{ NO}_3^-$ prevail in the area south of the River Sava (Kleče – Obrije – Hrastje). Those measured values that exceed $25 \text{ mg L}^{-1} \text{ NO}_3^-$ which was the limit value (until 2005) as determined in ‘The Decree on the

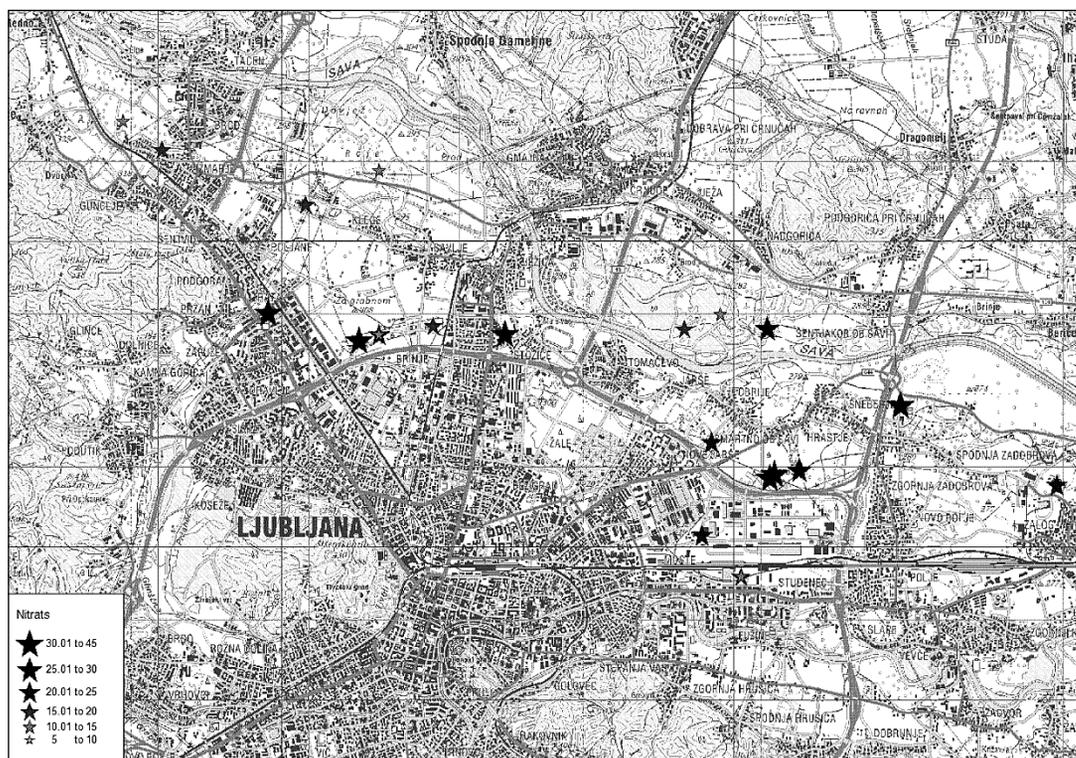


Figure 6: Maximum nitrate values on different sampling sites in the Ljubljansko polje aquifer. (North orientated)

quality of underground water',¹⁵ usually occur at the Hrastje V and Dekorativna sampling sites (Fig. 6).

The groundwater in the area of Ljubljansko polje contains different amounts of dissolved ions, which can also be said of nitrate. When considering the results of nitrate values, we can divide Ljubljansko polje in to three areas:

- area of constantly low nitrate values. This is the aquifer area, under the influence of the River Sava.

This same river influences the decrease of the nitrate value in the aquifer during the water supply, since the river herself has a relatively low amount of nitrate (less than $7 \text{ mg L}^{-1} \text{ NO}_3^-$);^{16–17}

- area with varying nitrate values, i.e. Zadobrova, Kleče, Stožice, Obrije and Navje sampling sites, which lie in the area of Ljubljansko polje and do not enjoy good vertical protection. This means that, in case of fertilization on the surface, nitrate

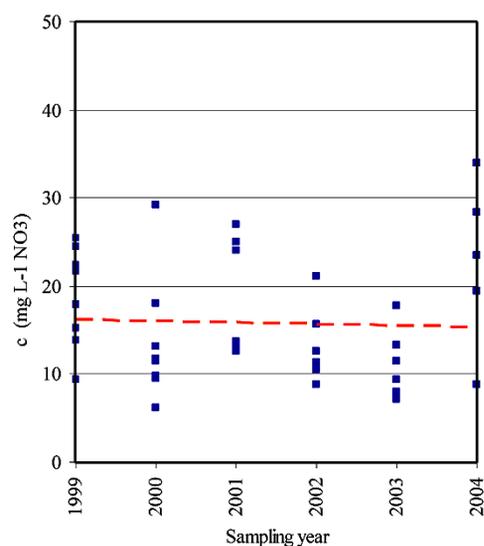


Figure 7a: Measurements of nitrate on Stožice from 1999 to 2004

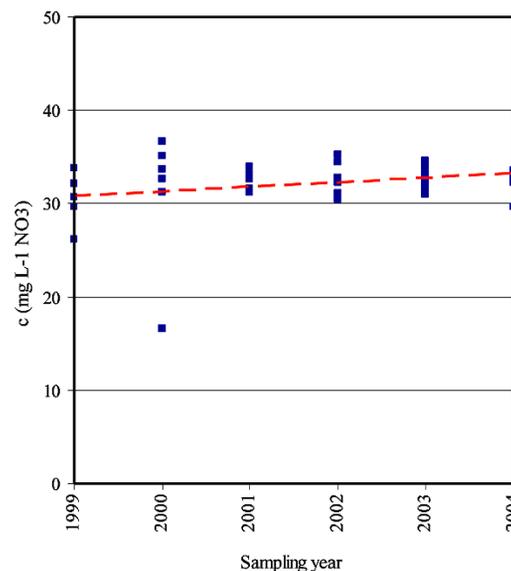


Figure 7b: Measurements of nitrate on Dekorativna from 1999 to 2004

easily leaks into the groundwater. Figure 7a represents nitrate concentrations at the Stožice sampling site. Concentrations here are variable.

- area with relatively high and constant nitrate values, i.e. the Dekorativna sampling site, Figure 7b. Values here are increasing, despite the fact that Dekorativna lies in an area with good vertical protection.

Moreover, the River Sava's influence can be noticed in the atrazine values. On those sampling sites closer to the river we measured lower atrazine values (or atrazine was undetected) than on other sites. The values for atrazine increase in the direction from the river towards the central area of the aquifer (especially in the Hrastje wells).

These conclusions are backed up the measured atrazine values for the Kleče and Hrastje wells, where the atrazine values in wells closer to the river are lower than those located more to the south.

As can be seen from table 3, the groundwater is polluted by atrazine predominantly in the area of Hrastje, measurement uncertainty is 15%.

Table 3: The atrazine values from 1999 to 2004

Sampling site	Atrazine _{AVER.} ($\mu\text{L cm}^{-1}$)	Atrazine _{MIN} ($\mu\text{S cm}^{-1}$)	Atrazine _{MAX} ($\mu\text{S cm}^{-1}$)
Elok	<0.03	<0.03	0.08
Julon	0.06	0.04	0.10
Koto	0.07	<0.03	0.18
Obrije	<0.03	<0.03	0.04
Zadobrova	0.07	0.05	0.08
Hrastje IA	0.18	0.10	0.39
Hrastje IV	0.18	0.15	0.23
Hrastje V – VIII	0.18	0.14	0.23
Hrastje ŠM	<0.03	<0.03	<0.03
Kleče VIII A	0.04	<0.03	0.11
Kleče XI	0.12	0.04	0.17
Kleče V – XIV	<0.03	<0.03	0.05
Stožice	<0.03	<0.03	0.07
Navje	<0.03	<0.03	0.05
Jarški prod III	<0.03	<0.03	<0.03
Šentvid II A	<0.03	<0.03	0.06
Roje	<0.03	<0.03	<0.03
Brod 11	<0.03	<0.03	0.04
Brod 12	<0.03	<0.03	0.04
Dekorativna	0.04	<0.03	0.13

Atrazine is highly persistent in soil. Chemical hydrolysis followed by degradation from soil microorganisms, accounts for most of the atrazine breakdown. Hydrolysis is rapid in acidic or basic environments but slower at neutral pHs. The addition of organic material increases the rate of hydrolysis. Atrazine can persist for longer than 1 year under dry or cold conditions. Furthermore, it is moderately to highly mobile in soils with low contents of clay or organic matter. Because it does not

adsorb strongly to soil particles and has a lengthy half-life (60 to >100 days), it has a high potential for groundwater contamination despite its moderate solubility in water. Atrazine is the most common pesticide found in both private and community wells. In soil, and also in water, atrazine breaks down in to two main products, namely desethyl atrazine and desisopropyl atrazine.^{18–19} From data trends between 1999 and 2004, the atrazine concentrations are on the decrease at all sampling sites. This is mainly because of the new legislation, adopted in 2003 and 2004 by of the Ministry of Environment and Spatial Planning, which does not allow the use of atrazine – based herbicides in those areas containing drinking water reserves.^{20–22}

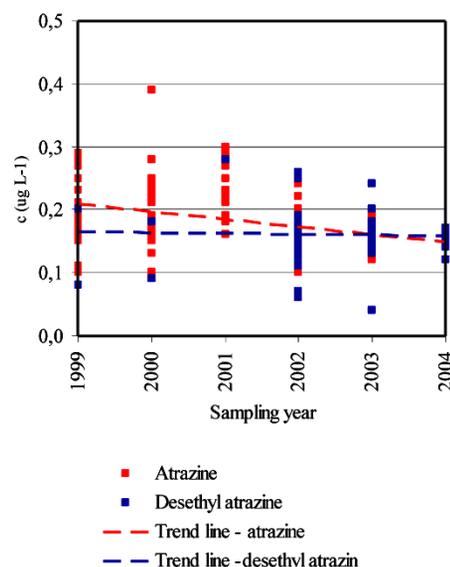


Figure 8a: Trends of atrazine from 1999 to 2004 on Hrastje IA

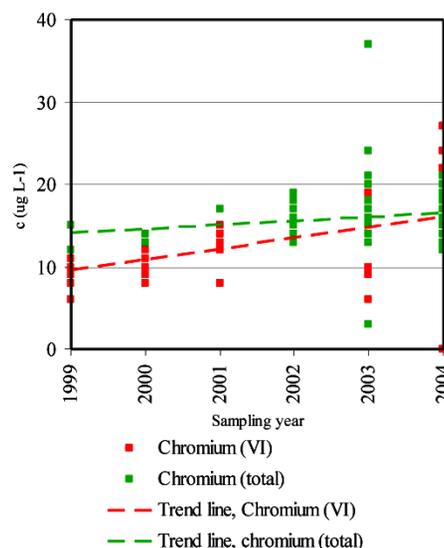


Figure 8b: Trends of chromium from 1999 to 2004 on Hrastje IA

Table 4: Values of chromium (total) in the area of Ljubljansko polje

Sampling sites	Cr(total) _{AVERAGE} ($\mu\text{g L}^{-1}$)	Cr (total) _{MIN} ($\mu\text{g L}^{-1}$)	Cr(total) _{MAKSIM} ($\mu\text{g L}^{-1}$)
Ljubljansko polje without Hrastje (N = 263)	4	<2	28
Hrastje (wells IA, IV and V–VIII), (N = 95)	16	3	37
Kleče (wells VIII A, XI), (N = 73)	5	1	26

Some wells in Hrastje are polluted by chromium, Table 4. This pollution is of great concern, since groundwater from these wells is used for public drinking water supply. The average measured values for chromium (total) in the groundwater from Hrastje area (N – number of samples taken = 95) is $16 \mu\text{g L}^{-1}$ Cr. Measurement uncertainty is 9%.

We took chromium into consideration, due to industrial pollution from chromium in the early 1980's regarding the Šiška area of Ljubljana. From experimental measurements it was determined, that the chromium which was initially spreading throughout the aquifer to the wells of Kleče has now moved down to the Hrastje wells. From the results of modelling, it can be seen that values for chromium increase with the general groundwater flow direction, i.e. northwest-southeast.

Figure 8b shows the higher polluted well in Hrastje. The amount of chromium in the wells of Kleče is app. 3 times lower than initial values. Further research in to the wells of Hrastje revealed that chromium (VI) is predominant.

Because of the above mentioned, some attention to this issue needs to be given by the Water Supply Company (VOKA), when managing the Hrastje wells.

4. Conclusions

The groundwater of Ljubljansko polje represents a valuable source of drinking water. Ljubljansko polje is also a densely populated area with an intricate traffic network, and industrial structure. Agricultural land still exists spreading throughout a greater part of Ljubljansko barje in the midst of all the above – mentioned.

Urbanized areas, industrial structures and agricultural land have had an influence on the groundwater pollution in the past but nothing beats the reckless and unprofessional overseeing of waste material and wastewaters.

The results of this study highlight the distinctive influence of the River Sava on conditions in the aquifer. They are reflected in the lower pollution and higher stability of those chemical measurements taken at sampling sites close to the River Sava. The area containing Šentvid, Rožnik and Grajski hrib is full of low permeable sediments. At some spots the thickness of clay layers exceeds 10 m (i.e. the groundwater is well-protected from the pollution). Nevertheless, at some sampling sites, higher values for electrical conductivity and pollutants were recorded

the source of which is due to actions on the surface. The crucial question is whether these results are the consequence of inappropriate constructions of sampling sites. Electrical conductivity is a parameter that does not attract enough attention, despite the fact that it is a quick, simple and inexpensive measurement. However, in spite of these facts, we do have a low number of EC measurements in regard to nitrate. At the same time, EC provides essential immediate answers about possible pollution.

A characteristic of the whole Ljubljansko polje aquifer is the presence of several pesticides, especially atrazine. The general trend is a decrease in pesticides present in groundwater. Figure 8a presents concentrations of atrazine and desethyl atrazine at Hrastje IA. Whilst atrazine concentrations are decreasing this is, however, not the case for those of desethyl atrazine. The values here remain relatively constant. Spatial analysis of data concerning pollution from atrazine shows that the wells of Kleče and Hrastje, located closer to the River Sava, are less polluted.

It can be seen, from the mathematical modelling of groundwater flow that the general flow goes from the northwest and southwest to the east along the river Sava. This general hydrological situation can also be clearly seen from the atrazine and chromium data. The data sets trends for chromium were higher at the beginning of sampling in the north-western area (Kleče) and then again in the later part of sampling at the Hrastje. The same general situation occurs when examining the atrazine data trends. It can also be seen that pollution is less at the northwest part of Ljubljansko polje than in the area around the Hrastje. These can be explained by the general hydrological situation in which the River Sava is recharging the Ljubljansko polje at the north western section (Kleče, Šentvid) and draining at the south eastern section (Hrastje). So the groundwater is moving in a general direction from Kleče down to Hrastje and transporting the chromium and atrazine.

It can be seen on the data sets used for analysis that the hydrological situation has a big impact on the chemical status of the groundwater. This is bearing in mind that the chemical status of groundwater always has to be explained by strongly considering any hydrological and hydrogeological situations. The same is also valid for those sampling sites where special attention has to be given on the hydrogeological characteristics of the surrounding area.

In the southeastern part of Ljubljansko polje, we can simultaneously find substances that are characteristic indicators of industrial pollution. Pollution from chromium

(total) and chromium (VI) is spreading from the area of Kleče towards the wells of Hrastje. Pollution by chromium (VI) is increasing, especially in the Hrastje area.

5. Acknowledgment

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22. Decree on the area of the Ljubljansko polje aquifer and its hydrographic reserves at risk from plant protection products and volatile chlorinated hydrocarbons (OJ RS, No.102/**2003**, 120/**2004**, 7/**2006**).

Povzetek

Na območju mesta Ljubljane se izvaja več monitoringov kakovosti podzemne vode, katere nosilec so različne institucije. Naš namen je bil zbrati fizikalno kemijske podatke iz različnih monitoringov ter te podatke uporabiti za modeliranje. Model toka podzemne vode in prenos snovi je izdelan s pomočjo hidrogeoloških in geofizičnih informacij z uporabo programa ModFlow. Z upoštevanjem hidroloških značilnosti Ljubljanskega polja, smo izdelali prostorske karte s predvidenimi vrednostmi za električno prevodnost, nitrata, atrazina in kroma. Določili smo območja, ki so pod znatnim vplivom reke Save, območja, kjer je visoka stopnja ranljivost vodonosnika, ter mesta vzorčenja, ki niso reprezentativna za to območje. Za nitrat, atrazin in krom smo ugotovili trende naraščanja oziroma upadanja. Analiza večletnih meritev je pokazala, da so različna onesaženja na Ljubljanskem polju slej kot prej merljiva na merilnem mestu Hrastje.