

POTENTIAL EFFECTS OF THE CONTAMINATED GROUNDWATER ON HUMAN HEALTH IN SZEGED, SE HUNGARY

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Abstract: The aim of this study is to present the groundwater contamination in Szeged for seven components (NO_3^- , NH_4^+ , PO_4^{3-} , Zn, Cu, Pb and Cd) by investigation of twenty groundwater monitoring wells and define human-health risk of irrigation with the contaminated water. According to the results, Zn content is above the limit value (defined by Hungarian joint decree) in the majority of the water samples, often with extremely high concentrations. Pb and Cd extend the limit values in all the samples (the highest Pb concentration (138.3 $\mu\text{g/l}$) is 14 times higher). Cu and NO_3^- have not exceeded the limits anywhere. NH_4^+ is ten times higher values than the limit one in two wells (10741 $\mu\text{g/l}$ and 12549 $\mu\text{g/l}$). In the case of PO_4^{3-} one well contains triply higher concentration (1505 $\mu\text{g/l}$) than limit value. The most contaminated well is located in the downtown next to the river Tisza, however extremely high metal concentrations were also detected in peripheral urban areas, where inhabitants traditionally irrigate with groundwater. The risk of irrigation with groundwater was modeled by RISC 4.0 in the case of two heavily contaminated gardens with detached houses. Based on the total health risk quotient (HRQ) results, the first well for children and adults is 1.1 and 0.5, respectively; therefore the risk is high for children but moderate for the adults. In the case of the other well modeled the total HRQ belongs to the moderate risk category with 0.85 (for children) and 0.35 (for adults) values. Examining the metals, the highest HRQ can be observed in Zn (0.1 – 0.8) and Cd (0.1 – 0.45), whereas ones of Pb and Cu are negligible. Vegetable consumption of exposure pathways can be risky for children and adults, while the hazard of the other exposure ways (dermal contact, inhalation of volatiles, ingestion) is not so significant.

Keywords: groundwater; metal contamination; human health risk; water quality

1. INTRODUCTION

Nowadays the less and less proportion of subsurface water has met the daily requirements owing to the significant contamination in the last. Therefore, identification of unknown groundwater contamination sources is a crucial problem in groundwater management (Ayvaz, 2010).

Sources of pollution may be either diffuse or localized. The important question is in both cases whether contaminants are transported as conservative components, or to what extent they will be retarded by geochemical processes within the aquifer. Due to the long residence of groundwater in the invisible subsurface environment, pollution may make their effects felt first in many years (Appelo & Postma, 1999).

In accordance with the above facts, it is relevant that different types of contaminants in the

groundwater should be investigating and monitoring. The EU Water Framework Directive draws attention to this problem and distinguishes two types of monitoring objectives for groundwater quality: (1) to present an overview of groundwater chemical status and (2) to detect the presence of long-term anthropogenically induced upward trends in the concentration of pollutants (EU Directive, 2000). In our study, the contamination of groundwater in Szeged is investigated based on above two objectives.

Since mainly anthropogenic effects cause the water pollution so competent authorities has to monitor and regulate degree of the groundwater contamination. Hungary is not one of the mostly contaminated countries, but we have to give heed to groundwater quality since many people utilize groundwater for irrigation (Szabó et al., 2007). Based on these facts, regulations have been enacted in order to control the concentration of toxic

materials in the groundwater. In Hungary, a joint decree (6/2009. (IV. 14) KvVM-EüM-FVM collective decree) has been implemented to protect the quality of groundwater.

Unfortunately, the amount of toxic materials in the groundwater in Szeged has exceeded the limit values in many cases as a result of which human health risk can be significant. However, wide-ranging network of groundwater monitoring wells is available in Szeged, still few investigations have been realized regarding relation between groundwater metal contamination and human health risk so far. After these facts, the aim of the present study is to assess the condition of groundwater in Szeged and its outskirts with special regard to cadmium (Cd), copper (Cu), zinc (Zn), lead (Pb), orthophosphate (PO_4^{3-}), nitrate (NO_3^-) and ammonium (NH_4^+) contamination.

The evaluation of human health risk of groundwater in Szeged has never been carried out up to now. Consequently, human health risk of the irrigation with the groundwater in mainly the outskirts of Szeged is also investigated. It is very essential question what degree of the contaminants has effects on human organism considering the exposure pathways (e.g. contact with water, ingestion, inhalation, metal accumulation in plants etc.)

2. DESCRIPTION OF THE STUDY AREA

Szeged is a major city in South East Hungary with an elevation of 84 m a.s.l. The surface of the area is largely affected by the fluvial systems of the rivers Tisza and Maros, which are composed of active and inactive channels (Marosi & Somogyi, 1990). The Pliocene sequences of considerable thickness are overlain by fluvial deposits of Pleistocene and Holocene age of several hundred meters. The climate is warm and dry with an annual mean temperature ranging between 10.5 and 10.6 °C. After the Great Flood of 1879, two major flood protection systems were devised: one relying on the newly constructed ring of dams embracing the inner core areas and the other based on the elevation of the original surface via significant infilling of low-lying areas (Andó, 1979). Both the intensive artificial infill following the Great Flood and other anthropogenic activities (e.g. frequent filling, mixing, accumulation of rubbles and household wastes in depressions etc.) owing to expansion of urban functions have been defining the physical and chemical properties of the soils here. Consequently, original soil types can hardly be found in the city (Puskás & Farsang, 2009).

The groundwater in Szeged stores at 1-3 m, so mainly the geological structure of this depth is relevant

from the viewpoint of groundwater investigation. Silt, clayey silt, silty clay extended lacustrine clay can be observed in the allochthonous loessy sediment of infusion loess dominating on the right bank of Tisza. These loose sediments perfectly indicate the natural sedimentary deposition in the sample area significantly influenced by different watercourses. Here the groundwater is generally unconfined. Fluvial clay, fine and coarse silt, lacustrine clay on floodplains and wetlands formed by mainly river Maros are typical of the right bank of Tisza. Thus, here the groundwater is confined (Kaszab, 1987).

During our study, quality of 20 wells selected from groundwater monitoring network in Szeged (introduced between 1975 and 1984) were investigated. In the course of well selection, we were about to cover complete area of the city (Fig. 1).

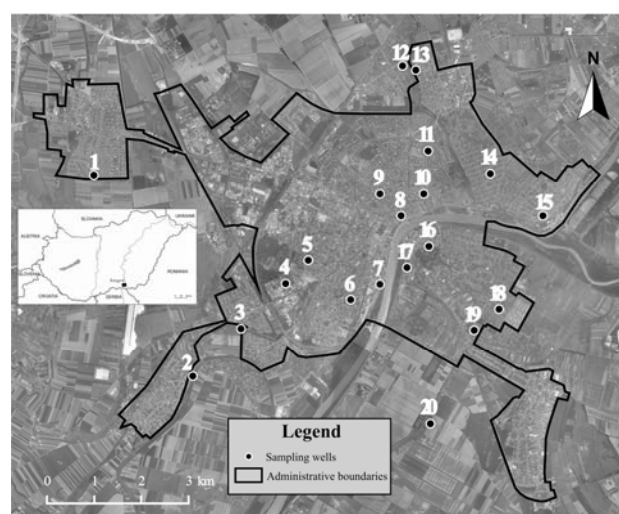


Figure 1. The location of the sampling wells on the orthophoto of Szeged

3. MATERIALS AND METHODS

The samples were taken (in May, 2008) after pumping based on operative Hungarian Standard, then were stored in totally filled up, cooled plastic bottles to avoid contact with air. The water level, depth, temperature and pH of the groundwater were measured on the premises. Furthermore, some more data (e.g. parameters of wells, weather conditions, depth of the sampling, and height of water column, GPS coordinates and land use) were put down in the field report (Table 1).

In the laboratory, NO_3^- , NH_4^+ , PO_4^{3-} (flow injection analysis, FIAstar 5000), Cd, Zn, Pb and Cu (AAS type Perkin Elmer 3110) were measured. The maps were compiled by ArcView GIS 3.2 The risk estimation was processed by RISC 4.0 to prepare transport modeling and risk analysis for human health in the case of contaminated area.

Table 1. Characteristics of the sample sites

Well number	Coordinates (WGS84)		Groundwater depth (cm)	Well bottom (cm)	Water column (cm)	Land use
1	46°15'53"	20°04'04"	277	445	168	rural residential area
2	46°13'40"	20°05'45"	332	688	356	rural residential area
3	46°14'14"	20°06'35"	210	471	261	garden suburb
4	46°14'46"	20°07'21"	233	446	213	inner residential area
5	46°15'02"	20°07'45"	262	591	329	inner residential area
6	46°14'33"	20°08'28"	206	551	345	inner residential area
7	46°14'44"	20°08'58"	448	552	104	downtown
8	46°15'33"	20°09'21"	260	781	521	downtown
9	46°15'49"	20°08'60"	156	516	360	inner residential area
10	46°15'49"	20°09'45"	185	756	571	inner residential area
11	46°16'19"	20°09'50"	368	514	146	housing estates
12	46°17'20"	20°09'27"	219	2000	1781	week-end houses
13	46°17'20"	20°09'28"	348	4990	4642	week-end houses
14	46°16'03"	20°10'53"	163	785	622	rural residential area
15	46°15'32"	20°11'47"	83	609	526	rural residential area
16	46°15'11"	20°09'49"	324	550	226	brown-stone district
17	46°14'56"	20°09'27"	605	779	174	housing estates
18	46°14'25"	20°11'00"	198	721	523	week-end houses
19	46°14'10"	20°10'35"	292	569	277	garden suburb
20	46°13'04"	20°09'48"	335	545	210	rural residential area

RISC 4.0 based on risk estimation guidelines of US EPA is suitable for estimation of human health risk of the environmental elements (soil, water, air) and assessment of remediation limit values; evaluation of ecological effects of sediment and surface water, preparation of simple transport models as well as management and storage of environmental data and limit values (Spence & Walden, 2001).

The risk of irrigation with groundwater was predicted in the areas with detached houses of Szeged since this activity is general in this zone. As there have not been exact data about the quantity of the uncooked vegetable consumption till now, so we plan to interview the inhabitants during a comprehensive survey. However, it is possible that the illegal water withdrawal from groundwater is typical of Szeged, too (Csáki et al., 2006).

4. RESULTS

4.1. Evaluation of the contamination in the groundwater

The concentrations of the studied components were compared with limit values in the joint decree (6/2009. (IV. 14) collective decree) (Fig. 2). According to the results (Table 2) PO_4^{3-} and NH_4^+ exceed the limit value in more wells, whereas NO_3^- has not been over this value anywhere. One well (No. 7) contains triply higher PO_4^{3-} concentration than limit value (500 $\mu\text{g/l}$)

but PO_4^{3-} content of four more wells locating in or nearby downtown approaches the limit value. NH_4^+ exceeds the limit value (500 $\mu\text{g/l}$) in the case of 6 wells; however, water of two wells on the right riverbank comprises outstanding high (more than tenfold of the limit value) NH_4^+ values. NO_3^- has not been beyond the limit value (50 000 $\mu\text{g/l}$) in the case of the studied 20 wells, either. The highest concentration (15 204 $\mu\text{g/l}$) has not approached the limit value at all. Therefore, it can be claimed that there is not nitrate contamination in the groundwater of Szeged.

Metals are well-known to be persistent but not PBTs (Persistent, Bioaccumulative, Toxic) in every case as many factors affect whether exposure to a metal poses problems (Hill, 2007). All the studied metals are PBT: Cu, Zn are essential, whereas Cd, Pb are not essential metals but the former ones can also be toxic in too high concentration. Zn content is above the limit value (200 $\mu\text{g/l}$) by the 70% of the water samples, often with extremely high values. Cd exceeds the limit value (5 $\mu\text{g/l}$) in all the wells, eight ones of which have twice as high value as the limit one. The highest concentration (No. 15) has been detected in the groundwater of gardens around detached houses in the outskirts. Consequently, it is very significant problem since Cd as a little amount is very toxic for the human organism. Pb exceeds the limit value (10 $\mu\text{g/l}$) in all the samples, which indicates remarkable Pb contamination all over the city. The highest value, which is 14 times higher than

the limit value, can be detected in a well in the outskirt (No. 20). Furthermore, Pb value of twelve

times higher than the limit value can be noticed in two more wells (No. 18, 20).

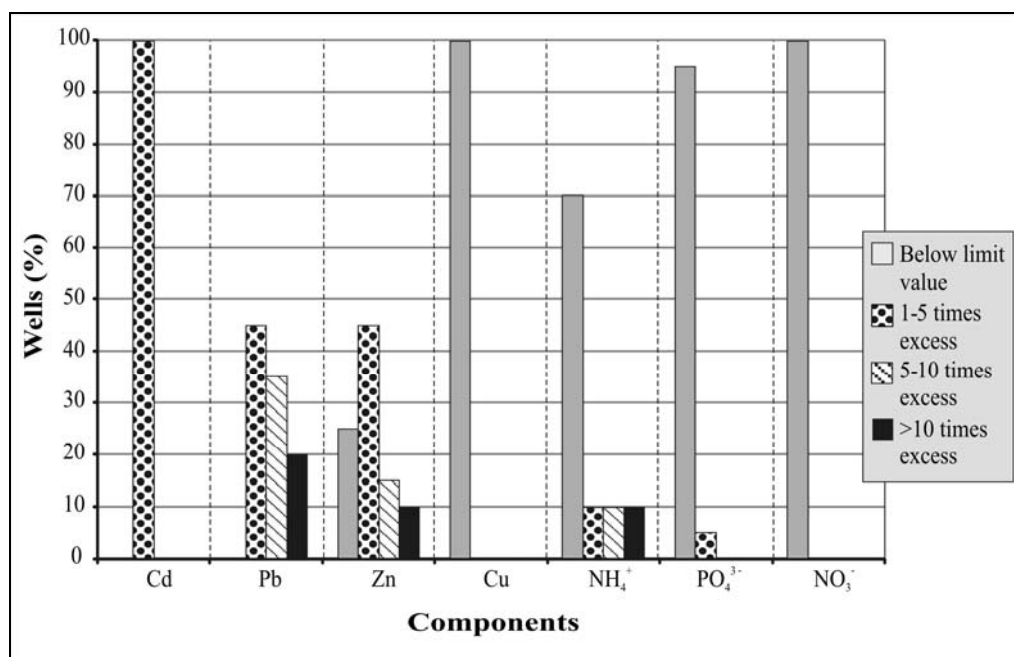


Figure 2. The percentage of the exceeded level compared to the contamination limit value determined in the joint degree, based on concentration of the collected water samples

Table 2. The concentrations of the measured elements

Well number	Concentration (µg/l)						
	Cu	Pb	Zn	Cd	NO ₃ ⁻	NH ₄ ⁺	PO ₄ ³⁻
1	42	25.1	284.4	8.8	9508	< DL	14
2	5.4	62.1	186.4	12	9538	111	18
3	8.8	43.1	473.9	11.5	5176	< DL	17
4	84.2	70.4	1510	8.3	12404	< DL	27
5	34.1	101	578.1	12.1	14831	< DL	45
6	20.5	15.2	441	10.8	5773	70	11
7	131.2	71.1	668.4	11.4	15	1070	1505
8	8.9	22.4	71.6	6.4	1645	< DL	64
9	71	96.2	1026	10.6	10814	< DL	353
10	40.4	37.1	88.7	8	15204	90	124
11	42.1	45.4	1189	8.5	11759	< DL	19
12	2.8	18.3	107.4	6.9	< DL	5041	15
13	6.7	20.1	623.1	6.6	139	865	28
14	48.4	69.2	706.4	7.6	9961	< DL	19
15	37.2	68.6	880.6	13.6	13673	< DL	19
16	42.7	120.2	2176	7.2	14989	< DL	32
17	170.2	61.9	502.9	12	48	10741	108
18	18.1	121.6	36.6	9.6	< DL	6072	22
19	9.3	11.7	143.8	6.6	< DL	12549	17
20	57.3	138.3	2252	8.4	349	311	66
Limit value*	200	10	200	5	50000	500	500

*according to the joint degree No. 6/2009 (IV. 14.)

The most Pb contaminated wells are situated at right riverbank with gardens so irrigation with groundwater is very hazardous regarding the health risk. Cu values are not higher than the limit value (200µg/l) in all the samples, thus, there is not Cu contamination.

4.2. Human health risk of the contaminated groundwater

Human health risk was processed considering four the most frequent metals (Cd, Pb, Zn and Cu) in the urban area, which can cause relevant water quality deterioration (Szalai et al., 2004).

Two wells (No. 15, 20) with high level of contamination, which are situated in the gardens around detached and week-end houses (Fig. 3), were selected for risk estimation where the irrigation with groundwater is probable. The human health risk quotient (HRQ) was determined for two receptors: average child and adult. The risk was modeled for four exposure pathways (ingestion, inhalation of volatiles, dermal contact and vegetable ingestion) concerning irrigation of groundwater. The HRQ is given during risk estimation. This value is ratio of the average daily intake (ADI) and tolerable daily intake (TDI) of the contaminant. If the HRQ is larger than 1, the health risk is unambiguously high (Table 3).

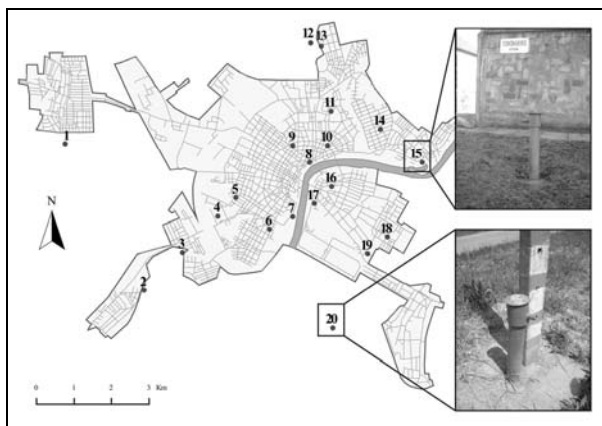


Figure 3. The location of the wells in human health risk modeling

Based on HRQs of metals of two wells, the results are as follows: regarding the well No. 20, the HRQ of Cd is 0.3 and 0.1 in the case of children and adults, respectively (Fig. 4 A). Consequently, it can be established that the risk is moderate. However, Cd in low concentration is very toxic for the human organism so you have to pay more attention to the moderate HRQ. In accordance with our model, human risk of Cu and Pb is negligible. The HRQ of Zn is 0.8 and 0.3 for children and adults, respectively. Thus, human risk is moderate in the case of this

element. However, the moderate risk is very close to 1 in the case of children. Despite Zn values 10 times exceeding the limit value and its relatively high mobilization ability, the HRQ is not as high as you would presume. The total HRQ for children and adults is 1.1 and 0.5, respectively; therefore the risk is high for children but moderate for the adults.

Table 3. Qualification of the health risk quotient (Dankó et al., 2004)

Health risk quotient (HRQ)	Qualification
< 0.01	negligible
0.01 - 0.1	low
0.1 - 1	moderate
1 - 10	high
> 10	very high

In accordance with vegetable consumption of exposure pathways, it can be stated some vegetables can uptake Cd and Zn due to relatively high mobilization ability of these elements. Thus, this pathway can be risky for children and adults; the hazard of the other exposure ways (dermal contact, inhalation of volatiles, ingestion) is negligible. Therefore, contaminant accumulation in the plants during irrigation with groundwater contaminated by human activities can increase the human health risk (Hinwood et al., 2006).

The results of the other well (No. 15) (Fig. 4 B) supports the above statements, namely high metal concentration in the groundwater can be hazardous for the health. Cd and Zn out of measured metals were estimated as the ones of the moderate risk. The HRQ of Cd exceeds 0.5, 0.2 in the case of children and adults, respectively. This quotient of Zn is 0.3 and 0.1. The total HRQ belongs to the moderate risk category with 0.85 and 0.35 values. Parallel to the sample No. 20, the vegetable consumption of the exposure pathways implies the highest risk. If we investigate the risk by components, it is relevant that Zn and Cd is the most dangerous both for children and adults.

5. DISCUSSION

During our study, it is obvious that the groundwater in Szeged is considerably contaminated due to NH_4^+ , Pb, Zn, Cd concentrations exceeding the limit value (Fig. 5). The PO_4^{3-} concentration, potentially originating from the chemicals used during industrial water treatment and detergents in the urban watercourses, has not exceeded limit value except for one well (No. 7) in the downtown. There are some relatively high NO_3^- concentrations citywide but heavy NO_3^- contamination cannot be detected in Szeged.

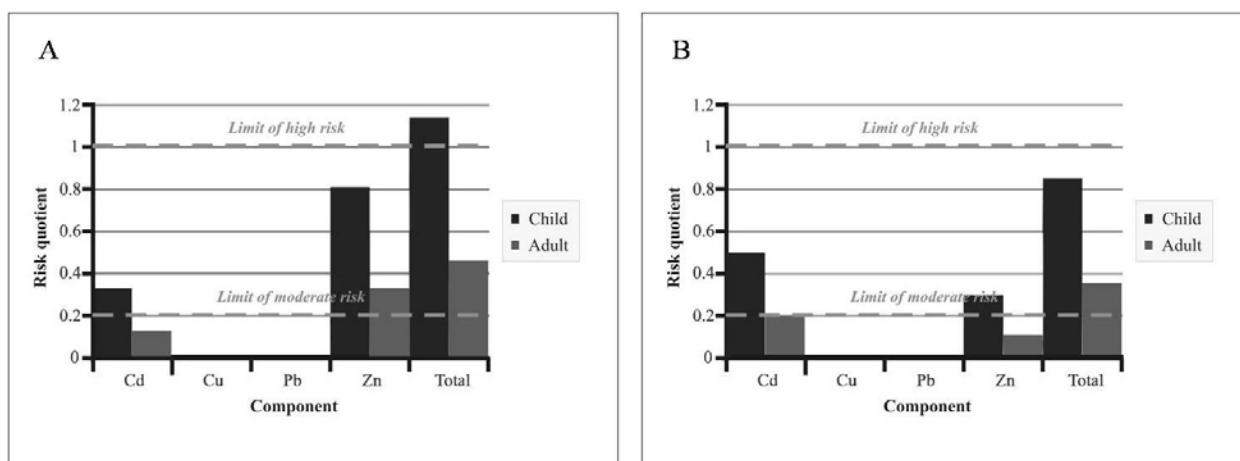


Figure 4. The human health risk of each metal for average child and adult at sample No. 20 (A) and No. 15 (B)

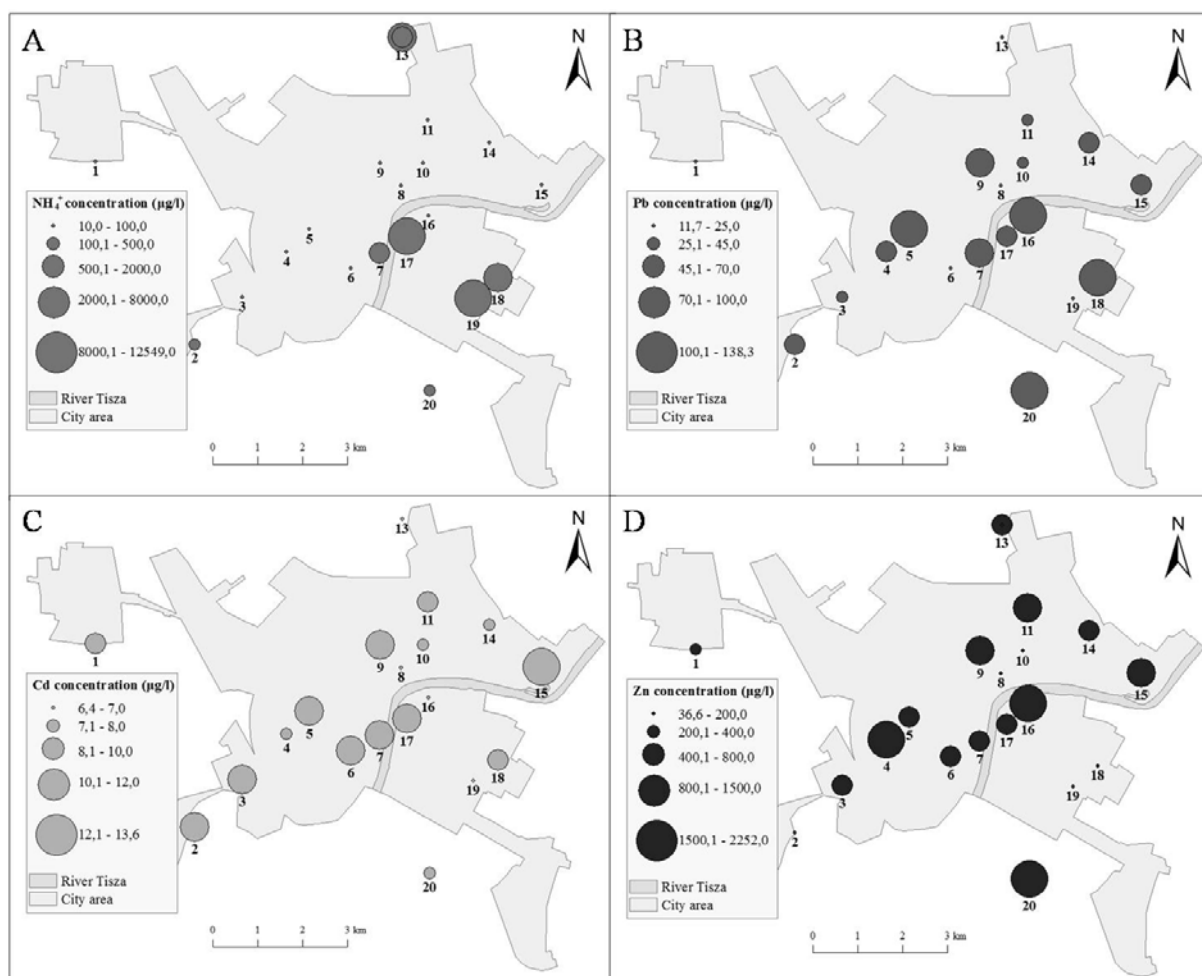


Figure 5. Horizontal variations of NH_4^+ (A), Pb (B), Cd (C) and Zn (D) contents of wells in Szeged

On the contrary, the amount of NH_4^+ as important indicator of the organic pollution is disquietingly high all over the city. The centre of the most significant NH_4^+ contamination can be observed in western part of Szeged (No. 17, 18, 19) (Fig. 5 A). The mentioned nitrogen forms probable arise from dissolution of solid wastes, ammonium nitrate fertilizers, pesticides, herbicides (Barótfi 2000). Pb

(Fig. 5 B) and Cd concentrations (Fig. 5 C), the highest contaminated points locating outside downtown, are higher than the limit value in all the water samples, which can be very dangerous for the human organisms.

Cu has not exceeded the limit at all, whereas the Zn is above the limit value in the most samples (Fig. 5 D). The water of two wells (No. 16, 20)

contains highly toxic Zn concentration (more than tenfold of the limit value). The latter wells are situated in the outskirts with gardens around detached houses where it is likely to irrigate from groundwater. The distribution in the city of Pb and Zn values is similar as the most contaminated samples originate from the same wells (No. 9, 16, 20).

The high concentration of the metals in the groundwater can be derived from old water pipelines (especially ones made of copper), industrial activities, heavy traffic, unsuitable treatment and disposal of the sewage and wastes, even from waste thermal water contaminated with Cd and Pb (Balog et al., 2011). Concerning this high contamination, it can be noted that the depth of the groundwater table exerts significant influence on the degree of contamination (Szabó et al., 2010). As a rule, the concentrations of dissolved substances decrease with depth (Jalgaonkar, 2008). In the wells the average depth of the groundwater table was 2.7 m. Therefore, this fact could also take an effect on high concentration of the measured elements.

The main aim of the human health risk's modeling is to draw the attention of the population to harmful effects of the contaminated groundwater on their health and increased hazard for the cancers. According to HRQ values predicted using the model, it can be established the heaviest risk of the exposure pathways is human consumption of the uncooked vegetables irrigated with groundwater. Consequently, these metals may accumulate to toxic concentration levels in the plants (Andráš et al., 2007). The degree of the risk can increase if the inhabitants consume exclusively the vegetables produced originating from the contaminated area. In accordance with results of Cd and Zn modeling process, it is relevant that HRQ values of both ones are the greatest. There is not risk considering the Pb in the city; however, it can be detrimental for the health if Pb from the traffic deposits with the dust on the foliage.

With regard to the foregoing facts, HRQs of each metal are moderate or negligible: however, total HRQ of all the metals can be significant, so it poses potential health hazard.

6. CONCLUSION

In our research the water quality of twenty wells were studied for typical impurities in the groundwater in urban area, Szeged. In accordance with quantitative and qualitative evaluation of results as well as estimation of human health risk of the most contaminated wells, the conclusions can be summarized as follows:

- the groundwater in Szeged is contaminated

with several pollutants, including toxic heavy metals;

- the most contaminated well (No. 7) located in the yard of the clinic centre of the downtown;

- extremely high metal concentrations were also detected in peripheral urban areas, with irrigation;

- irrigation with the contaminated groundwater may be risky for human health;

- Cd, Zn of metals and vegetable consumption out of the exposure pathways are the most detrimental to the human organism;

- these harmful substances can take negative effects especially on children since the concentration inert for the adults might be very noxious for children.

At the beginning of our study, groundwater in Szeged was not supposed to be so significantly contaminated and dangerous for health of the inhabitants living here. Consequently, it is very essential that persons exposed to contaminants should be informed of potential risks in plain language. Our investigation confirms significance of groundwater monitoring in the urban area and comprehensive survey methods of the contamination effects. In the light of the above, increasing the number of samples and components to be measured, the investigation of the groundwater in Szeged is being carried out in order to reveal the details of the contamination and its effects.

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