

POLLUTION OF ARTESIAN WELLS IN THE URBAN AREAS OF KRAKOW, EUROPE

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ABSTRACT

Artesian wells are sources of water from the late Pleistocene era. They are in common use in Krakow, in southern Poland, where five years ago limited studies were carried out, the results of which disputed the potability of water from some of the wells. This paper presents complex studies carried out at the end of 2012 concerning the pollution and composition of water samples from nine artesian wells. We studied smell and taste, color, turbidity and conductivity, pH levels, hardness, oxygen concentration and demand, total organic content, compound concentrations (NH_4^+ , NO_2^- , NO_3^- , Br^- , Cl^- , benzene, ethylbenzene, xylene, toluene), element concentrations (Al, Ca, Cd, Cu, Fe, Hg, Mg, Mn, Pb, Zn) and the quantity of bacteria. In only one well was water fit for human consumption. In other cases, numerous parameters were above the permissible limits. Most often, the parameters of color, turbidity and concentrations of Fe and NH_4^+ exceeded the acceptable levels. In one well the concentration of benzene also exceeded the maximum permissible level. The significance of the differences in the levels between wells shows that aquifers mix either very slowly or not at all.

KEYWORDS:

water; groundwater; artesian wells; pollution; metals

INTRODUCTION

Artesian water is a type of groundwater which lies at a great depth between two impervious layers of rock. It is generally believed that this type of water is healthy and should form a part of our daily diet [1, 2, 3]. Krakow has artesian wells commonly in public use [4]. The springs date back about 10 000 years to the late Pleistocene era. The ceiling of aquifer rocks lies at a depth of between 54 m and 68 m below a thick layer of insulating clay. The performance of individual wells varies between 300 and 1500 liters per hour, giving a total of about 84,000 liters per day, for all the spa wells in Krakow [5, 6].

The exploitation of groundwater for human

consumption means we must evaluate the organoleptic properties, as along with physical, chemical and microbiological properties that inform us about the quality of water intended for drinking [7]. In 2008 the Polish Sanitary Service checked the quality of water in artesian wells in Krakow. Those studies revealed that the water was unfit for human consumption in a number of wells with respect to selected parameters [8]. However, people still use all the wells. No complete analysis of water for physical, chemical and biological parameters was carried out for the Krakow wells and such data remain unavailable in the literature.

We aimed to evaluate the water quality in nine commonly used artesian wells in Krakow. We measured organoleptic parameters (directly through the senses), physical and chemical parameters (with ions, metals and organic compounds) and microbiological parameters. We compared the data with permissible limits and with the previous data available. Our studies addressed the question whether the artesian wells in the industrialized city of Krakow are a safe source for drinking water.

MATERIAL AND METHODS

Samples for this study were collected on 2nd December 2012 between 5 p.m. and 8 p.m. in Krakow, which is located in the valley of the Vistula at the interface of three regional tectonic units: Monocline Silesian-Krakow, Miechów Hutch and Carpathian Foredeep (FIGURE 1). The meteorological conditions prior to and during the collection (from 30th November) were monitored by the ICM service [9].

Water from nine artesian wells (Table 1, FIGURE 2) was collected in plastic bottles (3 x 500 mL). The collected samples were examined in the laboratory of the Institute of Biology (Pedagogical University of Cracow) for smell and taste, color, turbidity, conductivity (SEC), pH levels, hardness (sum of Ca and Mg), concentration of dissolved oxygen, biochemical oxygen demand (BOD_5), concentrations of the compounds NH_4^+ , NO_2^- , NO_3^- , Br^- , Cl^- , concentration of the elements Al, Ca, Cd, Cu, Fe, Hg, Mg, Mn, Pb, Zn and quantity of bacteria

TABLE 1
Physicochemical characteristics of the samples studied (respectively mg/L, NTU and $\mu\text{S}/\text{cm}$) with maximum permissible levels (or permissible range for pH) according to Polish law [10]

Well no.	Localization	GPS data	Color	Turbidity	Conductivity	pH
1	Osiedle Tysiaclecia	50.092N	20	26.40	1927	7.48
2	Osiedle Piastow	50.101N	5	4.70	1486	8.23
3	Osiedle Bohaterow	50.098N	20	36.10	1649	7.84
4	Osiedle Dywizjonu 303	50.085N	5	0.08	1215	7.18
5	Ulica Majora	50.091N	20	4.29	1454	7.78
6	Ulica Podchorazych	50.076N	25	38.30	1034	8.00
7	Plac Inwalidow	50.069N	15	16.20	1104	7.64
8	Plac Sikorskiego	50.061N	10	7.22	1388	8.25
9	Ulica Kosciuszki	50.054N	20	28.20	1298	8.01
Mean			15.6	17.943	1395,0	7.82
SD			7.3	14.662	277.5	0.350
Permis. levels			15	1.00	2500	6.5-9.5

(coliform bacteria, *Escherichia coli* and intestinal enterococci, all results given as CFU: colony forming unit per 100 mL of water).



FIGURE 1

Krakow (dark dot) – sampling area in southern Poland (dark grey), Central-Eastern Europe

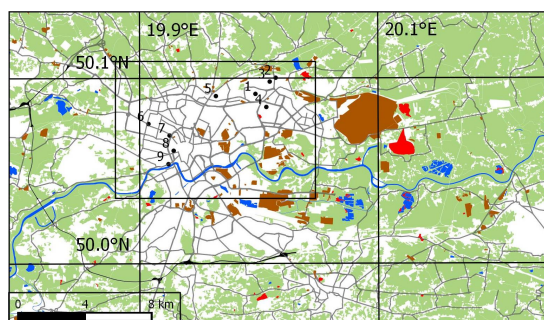


FIGURE 2

The distribution of artesian wells studied (1 to 9) in Krakow. Green represents natural areas (parks, forest, bushes), blue represents water areas (rivers, ponds), white represents residential areas, retail outlets and recreational amenities, brown represents industrial areas, red represents mineral extraction and dump sites.

Concentrations of organic compounds (BTEX: benzene, ethylbenzene, toluene, xylene) and the total organic content were examined in the laboratory of the Department of Environmental Management and Protection (AGH University of Science and Technology). Most measurements were taken in accordance with international norms and procedures used in commercial laboratories (Table 2). Taste and smell were studied immediately the samples were taken; concentrations of metals were measured within two weeks of the samples having been taken; and all other measurements were taken within 24 hours of the samples' being taken. The analyses of metals (with the exception of aluminum) were carried out in accordance with internal procedures. Samples for that protocol were stabilized with 1 mL of nitric acid (65% Ultranal®, POCH) per 500 mL of water. All samples were initially studied by means of flame atomic absorption spectrometry (AAAnalyst 200, PerkinElmer). If the concentration was below the limit of quantification, the sample was re-analyzed with electrothermal AAS (AAAnalyst 800, PerkinElmer).

The BTEX concentrations were analyzed with a gas chromatograph equipped with the Trace Ultra mass spectrometer detector and DSQ-II (Thermo Electron Corporation), with helium as a carrier gas. The RxiTM-1ms capillary column (Restek) was used (100% polydimethylsiloxane; film thickness 0.25 μm ; column length 30 m; column diameter 0.25 mm). The BETX were extracted by means of the Head-Space method and determined on GC-MS according to PN-ISO 11423-1. The samples were incubated in 90°C for 40 minutes; the column was heated from 31°C to 200°C at a temperature rate of 16°C/min.

The bromide concentration was determined by spectrophotometry with chloramine T as an oxidizing agent, and phenol red as an indicator. The bromide content was measured in a spectrophotometer Aurius UV-VIS 2021 Cecil Instruments Company.

In all the analyses each sample was analyzed twice per analyte and the mean value was used as the final result. If the RSD level between replicates was higher than 15%, the sample was re-analyzed. Levels of all the parameters lower than LoQ were treated as 0. After each ten instrumental measurements the control sample (with a known concentration or parameter value) was analyzed. All the recoveries were positive and satisfactory (100±10%).

All the maps included in the manuscript (with the exception of FIGURE 1) were prepared with QGIS 2.2 software with implemented co-ordinate system EPSG:2180.

RESULTS

On the day the samples were taken and for two days before both the daytime and nighttime

temperatures were stable (up to 2°C and down to -1°C respectively). Precipitation was low and the atmospheric pressure was stable and close to normal (Table 3).

In the samples from the artesian wells only one sample had an acceptable taste and smell (well no. 4). Among the physical and chemical characteristics of the water studied turbidity revealed the highest variation (Table 1). The color of the well samples varied between 5 and 25. Only four had a color value lower than or equal to 15. The mean pH value for all the water samples was 7.82 and individual values varied from 7.18 to 8.25 (Table 1).

Calcium was a dominant cation in all the wells studied. Magnesium concentrations were a little lower with a maximum of 93 mg/L in well sample no. 7 (Table 4). The highest level was found in well no. 4. Zinc was the next highest concentration with a mean of 19.0 µg/L.

TABLE 2
Summary of the procedures used in the studies with limits of quantification (LoQ)

Parameter	Standards	LoQ
color (visual examination)	PN-EN ISO 7887:2011	5 mg/L Pt
turbidity (turbidity meter)	PN-EN ISO 7027:2003	0.05 NTU
electrical conductivity (electrochemical method)	EN 27888:1993	88 µS/cm
pH (electrochemical method)	PN-90 C-04540/01	-
smell (organoleptic method)	PN-EN 1622	-
taste (organoleptic method)	PN-EN 1622	-
aluminum (UV-VIS spectrometric method)	PN-92/C-04605:02	40 µg/L
calcium (F-AAS method)	internal procedure	30 µg/L
cadmium (ET-AAS method)	internal procedure	0.057 µg/L
copper (F-AAS method)	internal procedure	27 µg/L
iron (F-AAS method)	internal procedure	415 µg/L
mercury (CV-AAS method)	internal procedure	4 µg/L
magnesium (F-AAS method)	internal procedure	30 µg/L
manganese (F-AAS method)	internal procedure	30 µg/L
lead (ET-AAS method)	internal procedure	1.478 µg/L
zinc (F-AAS method)	internal procedure	24 µg/L
the sum of calcium and magnesium (EDTA titrimetric method)	PN-ISO 6059	10 mg/L
ammonium ion (UV-VIS spectrometric method)	PN-ISO 7150-1:2002	0.070 mg/L
nitrate (UV-VIS method with sodium salicylate)	PN-82/C-04576-08	1.00 mg/L
nitrite (molecular absorption spectrometric method)	PN-EN 26777:1999	0.030 mg/L
bromide (UV-VIS method with chloramine T)	internal procedure	0.1 mg/L
chloride (silver nitrate titration with chromate indicator – Mohr's method)	PN-ISO 9297:1994	5.0 mg/L
dissolved oxygen in water (electrochemical probe method)	EN 25814	0.01 mg/L
biochemical oxygen demand after 5 days (BOD ₅)	EN 1899-2	0.01 mg/L
benzene (GC-MS method)	PN-ISO 11423-1	0.01 µg/L
toluene (GC-MS method)	PN-ISO 11423-1	0.01 µg/L
ethylbenzene (GC-MS method)	PN-ISO 11423-1	0.01 µg/L
xylene (GC-MS method; as the sum of o-, m- and p- isomers)	PN-ISO 11423-1	0.01 µg/L
TOC (GC-MS method)	PN-EN 1484	0.3 mg/L
coliform bacteria (membrane filtration method)	PN-ISO 9308-1	1 CFU/100 mL*
Escherichia coli (membrane filtration method)	PN-ISO 9308-1	1 CFU/100 mL*
intestinal enterococci (membrane filtration method)	ISO 7899-2:2000	1 CFU/100 mL*

*CFU – colony forming unit.

TABLE 1
Summary of meteorological conditions in days just before the sampling [9]

Date	Average day temp. [°C]	Average night temp. [°C]	Precipitation [mm]	Atmospheric pressure [hPa]	wind [m/s] (direction)
30.11.2012	2	-1	1 (rain with snow)	1005	2 (→)
1.12.2012	1	-5	0	1017	2 (→)
2.12.2012	2	-3	1 (snow)	1012	3 (changeable)

Iron, manganese and cadmium were found in significantly lower concentrations (Table 4). Aluminum, copper, mercury and lead generally did not occur in the artesian wells studied.

Chlorides predominated over other anions in all the samples studied (up to 186 mg/L). The concentration of nitrates was significantly lower and was found in only two wells (no. 4 and no. 5). The ammonium ion concentration was significantly higher than that of nitrites and was found in the lowest concentrations among all the anions studied. The variation in the concentration of oxygen was small and the mean was 5.908 mg/L. BOD₅ was characterized by a higher variation, the mean of

which was 2.238 mg/L (Table 4).

All the well waters examined contained the BTEX organic compounds studied. Apart from the sample of well no. 1, which contained 1.039 µg/L of benzene, concentrations of this compound did not exceed 0.610 µg/L. Among other BTEXs studied, xylene was found in the highest concentrations, 0.974 µg/L. The range of the total organic content was the highest among all the organic parameters examined (Table 5).

Apart from one of the samples (from well no. 2, where one CFU of intestinal enterococci occurred), no bacteria (none of the bacteria groups studied) were found in the samples studied.

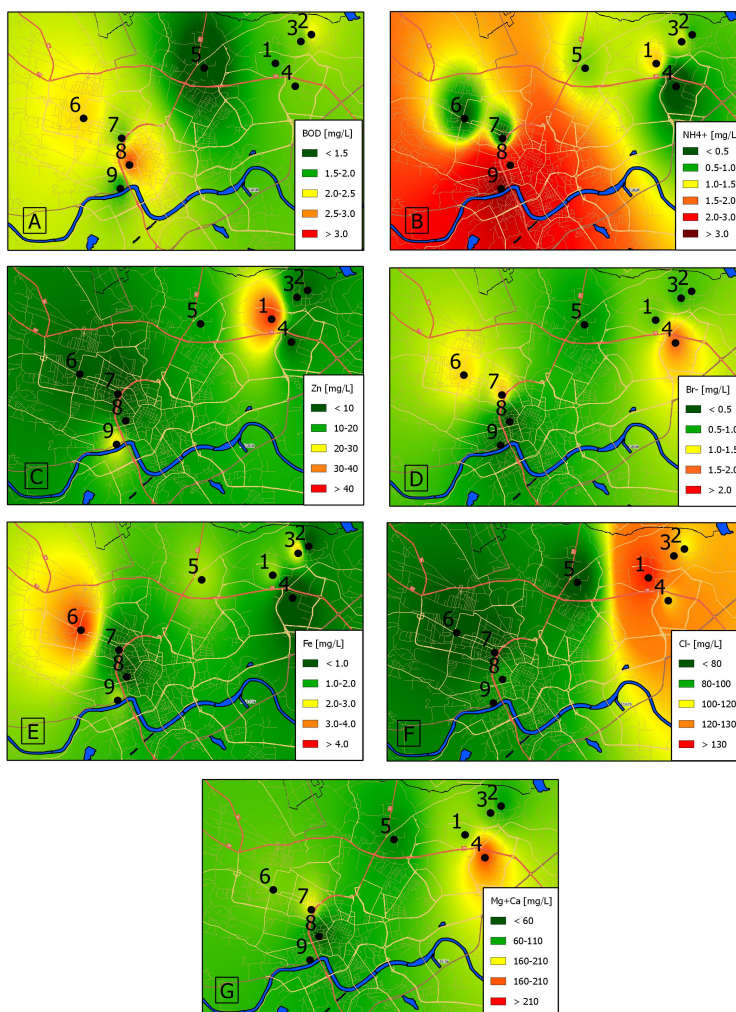


FIGURE 3

Particular variation among wells in biological oxygen demand (BOD₅), and concentrations of NH₄⁺, Fe, Zn, Mg+Ca, Br⁻ and Cl⁻. The maps correspond to the rectangle indicated in FIGURE 2.

TABLE 4
Concentrations of elements and ions in samples studied [mg/L] with maximum permissible levels according to Polish law [10] * values given in µg/L; parameters in bold presented in details in FIGURE 3.

Well no.	Al ⁺	Ca	Cd ⁺	Cu	Fe	Hg ⁺	Mg	Mn	Pb ⁺	Zn	NH ₄ ⁺	NO ₂ ⁻	NO ₃ ⁻	Br ⁻	Cl ⁻	O ₂	BOD ₅
1	0	71	0.025	0.00	2.57	0	66	0.00	0	52	1.68	0.14	0	1.17	186	6.90	2.08
2	0	43	0.029	0.00	0.80	0	62	0.53	0	12	1.06	0.03	0	1.15	120	5.63	2.50
3	0	59	0.000	0.00	3.27	0	66	0.08	0	14	1.44	0.01	0	1.05	128	5.67	2.19
4	0	180	0.036	0.00	0.00	0	35	0.00	0	12	0.00	0	30.45	2.02	123	7.62	2.22
5	0	43	0.000	0.00	2.66	0	48	0.00	0	19	1.24	0.01	0.43	0.86	76	5.70	1.11
6	0	57	0.035	0.00	4.60	0	76	0.04	0	11	0.50	0	0	1.62	75	5.43	2.65
7	0	67	0.011	0.00	1.24	0	93	0.02	0	6	0.37	0	0	1.67	74	5.39	2.18
8	0	21	0.036	0.00	0.84	0	34	0.02	0	12	2.72	0	0	0.49	99	5.44	3.24
9	0	47	0.022	0.00	2.65	0	70	0.04	0	31	8.60	0	0	0.69	94	5.39	1.97
Mean	0.0	65.3	0.0216	0.000	2.069	0.0	60.9	0.080	0.000	19.0	1.957	0.021	3.431	1.193	108.3	5.908	2.238
SD	0.0	45.7	0.0146	0.000	1.452	0.0	19.2	0.170	0.000	14.2	2.618	0.046	10.133	1.983	36.0	0.797	0.572
Permis. levels	200	-	5	2.0	0.200	1	30-125	0.050	10	-	0.50	0.50	50.00	-	250	-	-

TABLE 5
Concentrations of organic compounds in samples studied [µg/L] with maximum permissible levels according to official Polish regulations [10]

Well no.	Benzene	Ethylbenzene	Toluene	Xylene	TOC*
1	1.039	0.274	0.284	0.880	0.728
2	0.184	0.234	0.442	0.974	0.722
3	0.195	0.134	0.242	0.279	0.001
4	0.305	0.158	0.306	0.381	0.585
5	0.298	0.200	0.378	0.416	0.879
6	0.365	0.074	0.277	0.260	1.031
7	0.171	0.110	0.265	0.193	0.698
8	0.604	0.111	0.229	0.288	0.496
9	0.609	0.062	0.143	0.244	0.101
Mean	0.4189	0.1509	0.2851	0.4349	0.5824
SD	0.2856	0.0724	0.0862	0.2879	0.3392
Permis. levels	1.0	-	-	-	5.0

* values given in mg/L.

DISCUSSION

We found great variation in the physical and chemical levels among well waters from different parts of Krakow. Only the pH level was characterized by a narrow range of differences. In all the water samples calcium and chloride were the dominant ions. Concentrations of toxic metals, such as Cd, Hg and Pb, were low. We also noted the quantifiable concentrations of BTEX compounds. Microbiological parameters showed that no bacteria were found in the wells examined (with the exception of one colony of enterococci in well no. 2).

The sampling method allowed the collection of representative samples of water from each well. The year of the study was relatively representative regarding hydro-meteorological conditions insofar as neither substantial floods nor droughts were noted, so the hydrological characteristics of wells were time representative. The selection of sampling

points was governed by the necessity of gathering information about the pollution and condition of the artesian wells in Krakow. As may be seen (FIGURE 2) wells N°s 6, 7, 8 and 9 are in a residential area which also includes retail and leisure outlets. Wells N°s 1, 2, 3, 4 and 5 are located in the northern and north-eastern parts of Krakow, and are therefore closer to its industrial area, part of which is the Nowa Huta industrial region. Some of these wells are located near industrial plants and landfills that may cause pollution of surface waters and therefore pose a risk to groundwater.

Alpine tectonic movements in the Krakow area caused the creation of numerous tectonic ridges and trenches. This stimulated the formation of a multitude of aquifers: Quaternary, Tertiary, Cretaceous and Jurassic [5]. The anthropogenic impact on the upper aquifer is significant and connected to the centuries-old settlement in the city. Jurassic and Cretaceous layers, where water might

remain for thousands of years, are less susceptible to contamination. Jurassic layers form no single aquifer, but are cut by faults creating many separate hydrostructures. Due to the configuration of layers, the mirror bodies are at different heights, which hinders the flow of water between neighboring hydro-structures, as well as with surface water [11]. This may be the reason for the significant variation in numerous parameters of the water samples studied (FIGURE 3, Tables 2 to 5).

A common feature in all the wells studied was the presence of water in the Jurassic and Cretaceous layer. This layer is rich in limestone which makes its water fairly hard with high concentrations of calcium and magnesium (Table 4 and Figure 3G). The high mineralization of waters from the Jurassic formations in the Krakow area may also be caused by confined groundwater tributaries in the fracture zone [12]. A comparison of the current data with the results of research carried out in the early 1990s showed that calcium and magnesium concentrations have not changed significantly over the last twenty years [5]. Interestingly, even though the calcium concentrations decreased by a few percent, the magnesium concentrations increased, maintaining water hardness at a similar level.

Ammonium compounds may naturally occur under anaerobic conditions [13]. The ammonium ion content in the wells examined may be the result of continuous contamination of the water intakes with organic compounds (Figure 3B). Only well no. 4 contained ammonium within the permitted levels (Table 4). The data from previous years also showed that ammonia concentrations often exceed acceptable levels, particularly in well no. 9, where even today the highest levels were noted (8.60 mg/L NH_4^+). This was most probably an inorganic type of ammonia related to the origin of the Miocen formations [5, 14]. The high content in well no. 9 may additionally be caused by the accumulation of ammonia nitrogen in the silt layer on the banks of the Vistula, and subsequent slow infiltration into groundwater [15].

The worldwide average bromide concentration in water ranges from trace quantities to 0.5 mg/L. These compounds are considered of minor toxicity and their maximum acceptable daily intake is 24 mg for an adult [16]. The regulation regarding the quality of water fit for consumption does not define the maximum bromide concentration levels.

Increased human interference in the natural environment has resulted in higher levels of groundwater pollution by organic substances. Most frequently these pollutants are gasoline and diesel oils, and various derivatives of petroleum [17]. Their influence on water quality is extremely damaging; they also have a toxic and mutagenic effect. The regulations on potable water quality give maximum concentration levels for benzene, but none of the other organic compounds examined in the present

study. The substances beyond the reach of legislation (ethylbenzene, xylenes and toluene) occur in the samples at levels of 0.062 to 0.974 $\mu\text{g/L}$. Benzene, however, exceeds this range and in sample no. 1 it reached a concentration of 1.039 $\mu\text{g/L}$. This level exceeds the permissible levels for drinking water and thus makes the water unfit for human consumption [10].

Biochemical oxygen demand (BOD) is an indicator of the content of the biodegradable organic matter. Similarly to O_2 concentrations, there is no acceptable level of BOD for groundwater, and it does not itself specify whether underground water is contaminated. It rather indicates the general water quality. According to the US EPA procedures, evaluation of biological contamination of groundwater may be carried out by comparison of the basic parameters of water quality, such as turbidity, temperature and pH in groundwater, and surface water or rain. If a positive correlation occurs, contamination is suspected. These criteria may also apply to the presence of pathogenic organisms [18]. In accordance with the official regulations on the requirements for the quality of surface water used for human consumption (there are no regulations for groundwater at present), the well water examined, except for well no. 8, qualify as class A1 [19]. This means that their level of pollution by organic matter is minimal.

In eight of wells studied, the iron concentrations exceeded the permitted level of 0.2 mg/L. Only the water from well no. 4 contained no iron (FIGURE 3E and Table 4). The increase in mean iron concentrations over the past 20 years is clearly noticeable [5]. It is also worth emphasizing that the permitted level has changed over the years from 0.5 to 0.2 mg/L [20]. As far as copper, aluminum, mercury and lead ions are concerned, the levels found placed below the limit of method quantification. Cadmium concentrations ranged from the minimal quantification limit to 0.036 $\mu\text{g/L}$. All these concentrations fell within the potable water norms. Similarly, no other levels researched (NO_2^- , NO_3^- , Cl^-) exceeded the acceptable levels (Table 4). Apart from well no. 4, the concentrations of nitrates and nitrites are low. The concentration of chlorides in all the samples is significantly higher. This is particularly true for wells N^os 1, 2, 3 and 4, where the content of Cl^- ranged from 120 to 186 mg/L. This may be caused by the high mineralization of the water due to having spent so long in the rock layers or by the anthropogenic impacts of the centuries-old city [10]. Only the concentration of manganese in well no. 2 significantly exceeded the acceptable level. Levels such as Zn have no acceptable level set, but in comparison with the average concentration in groundwater in Poland (15 $\mu\text{g/L}$), concentrations in the wells examined are similar [20].

Only the water from well no. 2 contained one CFU of intestinal enterococci, which made it unfit

for human consumption. This result suggests possible fecal pollution of the water. In 2008 no excessive microbiological levels were noted, so the quality of water in the above-mentioned well must have deteriorated. Generally, however, water from the wells examined is safe when it comes to microbiological parameters.

CONCLUSIONS

The current condition of artesian waters in Krakow is vastly different from their state in the 1990s, when the wells were famous for their bacteriological cleanliness and lack of nitrogen compounds or any other substances indicative of external interference [5]. In 2008 the Polish Sanitary Service examined only the basic physico-chemical and microbiological aspects of Krakow's artesian wells, and listed the waters either fit or unfit for drinking [8]. According to those results well N^os 1 and 4 were fit for human consumption. Since then it transpires that only well no. 4 has retained the good quality of its water regarding fitness for human consumption. All other wells are unfit for human consumption at least on the basis of turbidity. The above-mentioned well no. 1 additionally fails to meet the requirements regarding various levels: the water consumption criteria have not changed diametrically since 2008, so the water quality in this well must have deteriorated substantially. Despite the fact that most of the wells examined give water unfit for human consumption, the water is consumed by humans, which may put the consumer at risk. People generally believe that boiling the water makes it fit for human consumption. This is certainly effective for the microbiological pollution, but in the case of artesian wells, the unfitness for consumption is chemical in nature, so boiling does not improve the water quality. Health considerations of the population determine that the above-mentioned wells should be adequately signposted with information regarding the waters' unfitness for human consumption, and a thorough inquiry into the sources of the pollution should be carried out. It is difficult to determine which levels should be particularly closely monitored and studied in the future. The higher concentration of benzene, however, is alarming. The presence of pathogenic bacteria, which clearly indicates anthropogenic pollution of deep water, should additionally be verified.

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