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Physicochemical characteristic and assessment of groundwater quality within the area of selected fuel stations in Opole

Groundwater quality monitoring is intended to provide information on chemical status of groundwater, tracking its changes and signalling environmental threats. Monitoring activities allow proper management of groundwater resources and adequate assessment of preventive measure effectiveness. The aim of the study is the analysis of groundwater quality status collected within the area of selected fuel distribution stations in the city of Opole. Samples were taken at monthly intervals during a four-month period. The content of petroleum substances, anions of: F^-, Cl^-, NO_2^-, NO_3^-, PO_4^{3-}, SO_4^{2-} and cations of: NH_4^+, Na^+, K^+, Mg^{2+} and Ca^{2+} was determined. Ion content tests were performed by using the ion chromatography method. Petroleum substances were determined as mineral oil index by gas chromatography method. Conditions for performing the chromatographic analysis are given in the research study. There were also examined the specific electrical conductivity, the pH value, "*in situ*" temperature, and water level in piezometers. Based on guidelines set out in the Regulation of the Minister of the Environment of 23 July 2008, groundwater quality classes were determined. Furthermore, research results were compared with results characterising groundwater collected from measurement point located in low-density housing on the out-skirts of the city of Opole. A significant impact on the degree of contamination of groundwater samples caused by location of measurement points was discovered. Samples collected within the area of 6 fuel stations, located in the city centre, indicated the highest level of pollution. The test results of six from fifteen tested physicochemical parameters exceeded limit standards determined for waters with good chemical status. In the test water samples, substantially increased conductivity, temperature, contents of chlorides and calcium ions were designated, which classify the water quality to IV class; as well as, the exceeded concentration of petroleum hydrocarbons and ammonium ion determined the level of water quality as of V class. Due to unsatisfactory research results of the analysed samples, further actions should be undertaken to improve the condition of groundwater in the analysed area. Among necessary actions to be undertaken are undoubtedly the elimination of leakage from tanks collecting fuel and checking of installations discharging wastewater.

Keywords: groundwater, groundwater quality classes, ion chromatography, petroleum hydrocarbons, water environmental pollution

Introduction

Water is a basic element of life, a fundamental component conditioning life on the Earth. This chemical compound is an ingredient of living organisms, as well as an essential element of inanimate nature. It is therefore an essential component of natural ecosystems and a factor shaping the climate. Contamination of water and its

shortages pose serious threat both to human health and the quality of life. Despite that, deficiency of water or water of a satisfactory quality adversely affects the aquatic and terrestrial environments, threatening the life of the flora and fauna inhabiting ecosystems.

Groundwater is a significant element of hydrological cycle and a valuable natural resource that ensures the basic source of water for farming, households and industrial use in many countries. Currently, it is one of the main sources of water for human consumption, which constitutes almost half of the quantity of drinking water available across the whole word [1].

The purity of groundwater can be treated by pesticides more and more commonly used in agriculture [2] or by sewage and industrial wastes. Despite that, the main sources of groundwater pollution having a major impact on the water quality arise from [3]:

– stations which are the main source of pollution with petroleum substances [4, 5];
– industrial companies, the activities of which are related to disposal of solid wastes, sewerage water disposals, emission of dusts and gases [6];
– opencast mining by removing the aquifer-bearing rock or soil, which leads to the risk of easier penetration of contamination [7];
– underground mining discharging salt mine waters that permeate into groundwater [8];
– transport industry, growth of automotive industry, expansion of communication networks and fuel distribution networks [9, 10];
– farming improperly utilizing fertilizers and pesticides, which leads to an inadequate management of wastes arising from animal husbandry and fodder productions [11-14].

Although the groundwater is more resistant to pollution than surface water, the prolonged and systematic release of pollutants into the soil or the process of washing out pollutants, for example from the area of leaking landfill sites, caused by rain waters, can lead to permanent contamination of groundwater. In particular, aquifers located within the urban and industrial areas, as well as areas of intensive agricultural production, are the ones that are mostly exposed to pollution.

Groundwater monitoring is an essential tool for evaluation of water quality status and for management of its resources. It provides current information on chemical composition of waters and their quality, as well as on components of the natural environment, being directly dependent on groundwater. The system scheduling monitoring of waters is one of the primary elements, based on which administrative decisions regarding the issues of water management and environmental protection are undertaken.

Poland's accession to the European Union and the implementation of European legislation led to key changes in management and protection of water resources [15]. Under Polish legislation, the limit values of physicochemical components contained in groundwater, according to the groundwater quality classes are standardized by the Regulation of the Minister of the Environment of 23 July 2008 on

the criteria and methods of groundwater assessment [16]. Selected limit values of physicochemical parameters entered by the Regulation are presented in Table 1.

Table 1. **Selected limit values of physicochemical elements of groundwater status in groundwater quality classes [16]**

Water quality indicator	Unit	Limit values in groundwater-quality classes I-V				
		I	II	III	IV	V
Temperature	°C	<10	12	16	25	>25
pH	–	6.5-9.5			<6.5 or >9.5	
The electrolytic conductivity at 20°C	µS/cm	700	2500	2500	3000	>3000
Petroleum hydrocarbons	mg/L	0.01	0.1	0.3	5	>5
Ammonium ion	$mgNH_4^+/L$	0.5	1.0	1.5	3	>3
Nitrates	$mgNO_3^-/L$	10	25	50	100	>100
Nitrites	$mgNO_2^-/L$	0.03	0.15	0.5	1	>1
Chlorides	$mgCl^-/L$	60	150	250	500	>500
Fluorides	mgF^-/L	0.5	1	1,5	2	>2
Phosphates	$mgPO_4^{3-}/L$	0.5	0.5	1	5	>5
Magnesium	$mgMg^{2+}/L$	30	50	100	150	>150
Potassium	mgK^+/L	10	10	15	20	>20
Sulphates	$mgSO_4^{2-}/L$	60	250	250	500	>500
Sodium	$mgNa^+/L$	60	200	200	300	>300
Calcium	$mgCa^{2+}/L$	50	100	200	300	>300

In Poland approximately 62% of water used for human activities is derived from groundwater intakes. The degradation degree of these waters depends on the amount of inflowing outbreaks of impurities of spatial character, from single point to larger area contamination.

Presence of the petroleum substances in the environment, in elevated concentrations, is a source of serious pollution and it poses a direct threat to all living organisms inhabiting the affected area. Petroleum hydrocarbons inhibit gas exchange, limit the access of light, reduce the dissolved oxygen concentration, degrade groundwater and surface water. These compounds have very toxic, mutagenic and carcinogenic effects on the organisms and disturb the homeostasis process. Because of these properties, each uncontrolled release of petroleum products is a very serious violation of hydrogeological environment. The migration of hydrocarbon compounds in the soil may also pose explosion and fire hazard. This is an especially dangerous phenomenon if point of contamination is situated nearby fuel tanks, residential buildings or transport routes.

Increased concentrations of inorganic ions being present in groundwater that is the source of drinking water can lead to numerous health problems of people consuming the contaminated water.

Exceeded concentrations of nitrates in drinking water can cause harmful interference to growth and can lead to increased need for vitamin A. Concentration of these ions at the level of 10 mg/L is particularly dangerous, while it causes cyanosis in infants. In turn, the increased content of nitrites in groundwater (above the level of 0.010 mg/L) can indicate contamination caused by animal remains. The outstanding harmfulness of nitrites arises from their tendency of forming nitro-compounds causing neoplastic changes. Chloride ions being present in large quantities in drinking water contribute to rise of hypertension and heart diseases. Concentration of fluoride maintained at the range between 3÷6 mg/L in drinking water causes fluorosis, leading to bone lesions and disabilities. Large amounts of fluorine accumulated in human body cause gastroenteritis, nephritis, liver damage, and myocardial injury. Exceeded levels of sulphates in drinking water cause bitter taste, and the presence of magnesium sulphates in drinking water causes diarrhoea.

1. Research area

Due to various geological and hydrological circumstances, the area of Opole is characterized by considerable soil typological diversity. In this area, they are generally rendzinas, alluvial soils, black earth and slightly occurring brown soils and podzolic soils. Within the city the largest areas are occupied by rendzinas and alluvial soils. Areas of petrol stations no. 1 and 4 are characterized by rendzinas, while in station no. 2, 3, 5 and 6 areas the predominant types of soils are alluvial soils [17]. Construction of rendzinas profile occurring in the analysed areas indicates that they are low permeability and low value of permeability coefficient soils ($k < 1.3$ mm/h). Because of their low permeability, they are classified as soils with a high possibility of surface runoff. Alluvial soils are characterized by a slightly higher permeability, but the permeability coefficient is also small ($1.3 < k < 3.8$ mm/h) compared to soil having good permeability ($k > 7.6$ mm/h) [18]. Analyzing the hydrogeological conditions, which characterize the desired area, it can be said that the underground water comes from deep bedrock layers defaulting under poorly permeable overburden which are insulation from direct surface contamination [19]. Flow of groundwater is compliant with the slope of the land and takes place in a north-westerly direction [20].

65 samples of groundwater collected from 20 measurement points, as piezometers located at six fuel stations in Opole, were subject of the research study. Each of the fuel stations was marked with subsequent number. Their approximate locations within the territory of the city of Opole indicates the map presented in Figure 1. In order to determine the impact of location on groundwater contamination degree, the obtained results were compared with parameters of water sample from outside highly urbanized area. A control sample was groundwater which was taken from

the measuring point located in the loose urban development on the outskirts of the city of Opole [21], where a large part of the soil is used for agriculture.

Fig. 1. Distribution of fuel distribution stations, where groundwater samples were collected; K - control sample

2. Methodology

Water samples were taken at monthly intervals for a period of four months. Selected physicochemical parameters were determined in the samples. Analysis of selected water quality parameters, such as the value of pH, conductivity, ion and petroleum hydrocarbons concentration were determined. Furthermore, the "*in situ*" temperature in samples and water level in piezometers were designated. These parameters were selected on the basis of the Regulation of the Minister of Environment of 23 July 2008 on the criteria and method of assessing the groundwater status [16], which compiles physicochemical elements limits of groundwater status in the individual quality classes.

Samples of groundwater were collected from measurement points that were piezometers, according to the PN-ISO 5667-11:2004 standard [22]. Water temperature measurement was performed under field conditions, while chemical designation was carried out under laboratory conditions, just after delivering the collected samples to the laboratory. After the temperature of samples were normalized to room values, their pH values were measured, according to the PN-EN ISO 10523:2012 standard [23] and the specific electrical conductivity in accordance to the PN-EN 27888:1999 standard [24].

In order to determine the cationic and anionic composition in the collected samples, the analysed waters were filtered through a syringe membrane filter with a pore size of 45 μm, and then, they were subject to chromatographic analysis using ion chromatograph with a high performance sequential suppression. The anions of: F^-, Cl^-, NO_2^-, NO_3^-, PO_4^{3-} and SO_4^{2-}, and cations of: NH_4^+, Na^+, K^+, Mg^{2+} and Ca^{2+} were designated in the water. Ions were determined according to the methodology described in applicable standards [25-26].

The content of petroleum substances was determined by using a gas chromatograph with flame-ionization detection (GC/FID), according to the PN-EN ISO 9377-2: 2003 [27].

Table 2 lists conditions under which the chromatography analysis was performed. Calibration curves were determined by measuring ion reference solutions prepared by the method of dilution from initial solutions with concentrations of 1000 ±2 mg/L. While a calibration curve for determining petroleum substances was prepared from the standard containing A and B pure mineral oils.

Table 2. **Standards for performing the chromatographic analysis**

Standards for conducting the analysis	Cations	Anions	Petroleum hydrocarbons
Column	Metrosep C4	Metrosep A Supp 5	ZB-5HT
Column dimensions	150 mm x 4 mm	150 mm x 4 mm	30m x 0.25mm x 0.25μm
Eluent	1.7 M HNO_3 + 0.7 M DPA	3.2 M Na_2CO_3 + 1.0 M $NaHCO_3$	–
Detection	conductometric	conductometric with suppression	flame-ionization (FID)
Acceptable pH range	2÷7	3÷12	–
Injection volume	10 mm^3	100 mm^3	1 μL
Pressure	~6.37 (max 20.0 MPa)	~7.0 MPa (max 15.0 MPa)	–
Temperature	25°C	25°C	Injector - 300°C Detector - 320°C
Analysis time	25 min	25 min	50 min
Carrier gas	–	–	Helium - purity 5.0

3. Results and discussion

The researched areas were rich in underground water. Average water level of all collections in particular fuel stations tested by piezometers ranged from 1.8 m for the 2 fuel station to 4.0 m below ground level in the case of 6 fuel station. The

temperature of groundwater was in the range between 13.5°C at the 1 fuel station to 17.7°C at the 2 fuel station.

Based on the results of laboratory tests, the petroleum substances content, "*in situ*" water temperature, the pH value, electrical conductivity and the content of individual ions, the water quality class was determined according to the Regulation of the Minister of Environment dated 23 July 2008 [16]. Provisions of the Regulation provide that good chemical status is indicated by groundwater of I-III classes, while waters with poor chemical status are classified to IV-V classes. The performed classification results with their average results of the tested parameters, being characteristic for each measurement point are presented in Figures 2-4. Values shown in graphs represent the arithmetic means of all measurement points, designated for every fuel station.

On the basis of data presented in Figure 2, it can be concluded that on the four of the six surveyed petrol stations, the limits of petroleum substances content, indicating their good chemical status, are exceeded. These values for the stations no. 1, 2 and 6 determine the IV class of groundwater quality, in turn, for station no. 5 reach a very high level and significantly exceed the limit values for which a V quality class is specified.
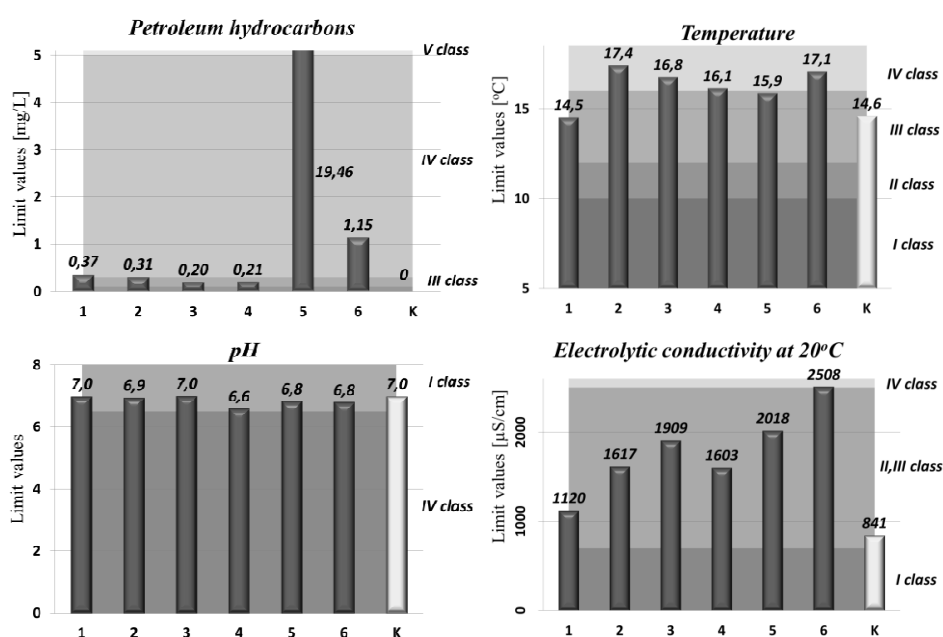

Fig. 2. Graphs classifying groundwater samples into particular water quality classes by measuring temperature, specific electrolytic conductivity and pH value, on the basis of [16]; K - control sample

Furthermore, it is important to mention the fact that in one of piezometers located within the fuel station no. 5, after filling sampling dipper, the presence of pure petroleum product was observed. In carrying out monthly monitoring the content of

this piezometer was not taken. The presence of oil substances in the piezometer was merely checked. The product appeared in each case at the measurement point.

Groundwater temperature values within the area of four from six fuel distribution stations classify the tested waters to IV class of water quality, placing them as waters with poor chemical status. These values are slightly increased, when compared to the temperature of the control sample.

In the case when electrical conductivity is adopted as a classification criterion, groundwater in the area of fuel station no. 6 is characterised by poor chemical condition. It should be noted that conductivity values in each case are significantly higher than the reference value in the control sample. This demonstrates the presence of high concentrations of ions in the samples. Whereas, the pH value, both in the control sample as well as in the samples collected from fuel stations, classify the test samples as waters with good chemical status (Fig. 2).
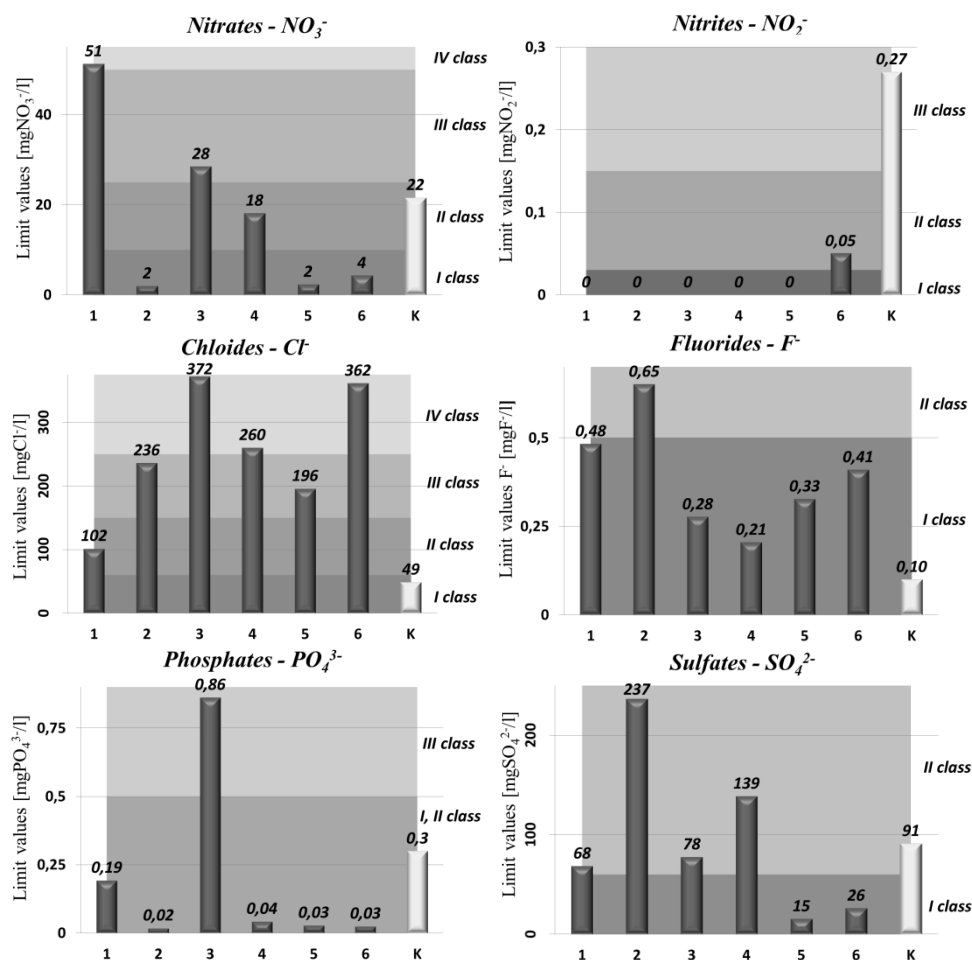

Fig. 3. Graphs with classification of groundwater samples into particular water quality classes by measuring the concentrations of anions, according to [16]; K - control sample
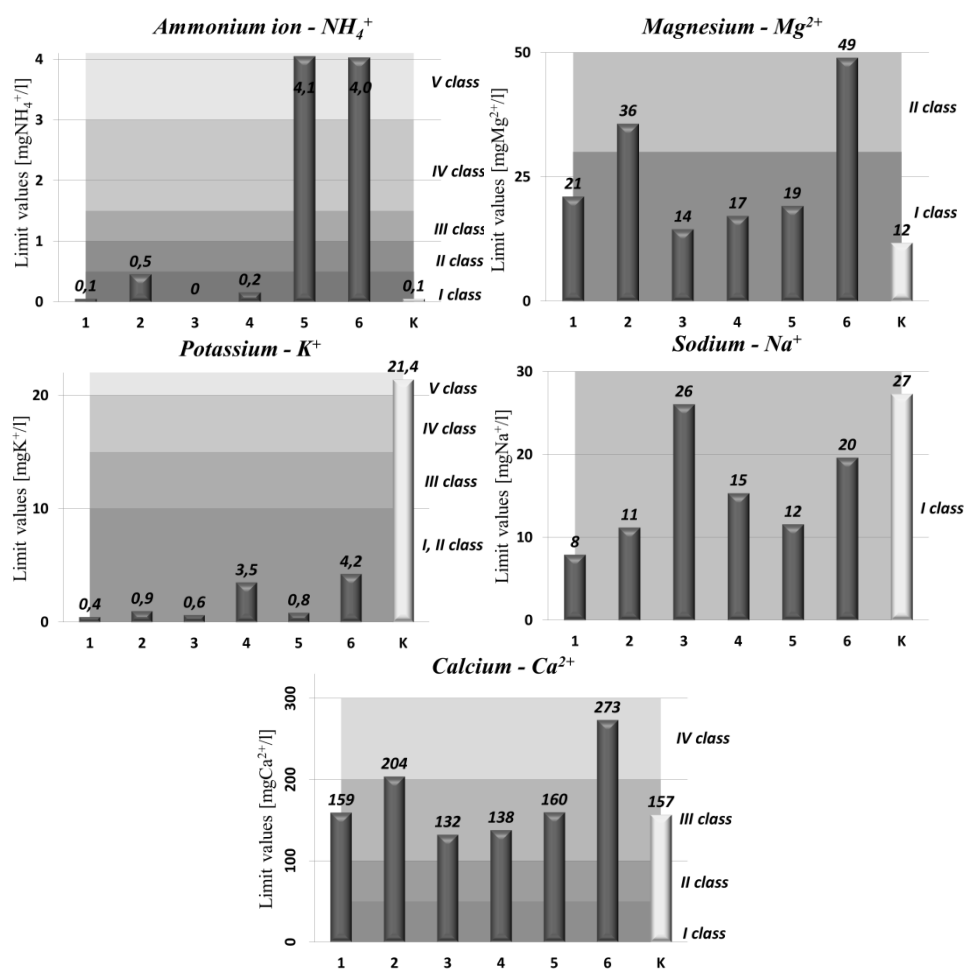
Fig. 4. Classification of groundwater samples into particular water quality classes by measuring the concentrations of cations, according to [16]; K - control sample

By analysing the cationic-anionic composition of groundwater, significant quality differentiation of these waters can be observed, which occurs within the territory of the city of Opole. Table 3 indicates the membership of groundwater samples to corresponding water quality classes, as well as it determines the overall chemical status of the analysed groundwater collected at each fuel station.

Fuel station no. 1 is characterized by an above-average content of nitrates in water samples. In accordance with the Regulation [16], concentration of nitrates is one of the physicochemical parameters, for which it is not allowed to exceed the limit value when determining the groundwater quality class at the measurement point. Such parameters are designated in the Regulation with „H" symbol. On the basis of provisions stated in the Regulation, groundwater from the area of 1 fuel station should be classified to IV quality class, so as waters characterized by poor chemical status. The dominance of nitrate nitrogen over other nitrogen compounds

is a sign of a significant and lasting for a certain period of time contamination, which can be caused by infiltration of municipal and industrial wastewater.

In the groundwater of the area of 3 and 6 fuel stations, very high concentrations of chlorides were designated, by which waters from such area can be considered as waters with poor chemical status. The presence of chlorides observed in large quantities can suggest on contamination caused by wastewater which seeps into groundwater.

In turn, due to the fact that the threshold of chlorine concentration is minimally exceeded at the station no. 4, which classifies the groundwater from the area of this station as waters with poor chemical status, and slightly increased value of temperature, it is allowed to classify the analysed water to III quality class, simultaneously recognizing its chemical status as good.

In samples of groundwater collected within the area of the station, raised concentration of fluorides was recorded, when comparing it to the control sample. The cause for this increased concentration of these ions can result from leakage of municipal wastes into groundwater. Large amounts of fluorides can be infiltrated into groundwater also together with air pollutants in the gas state or in the form of dust [28].

Table 3. **Classification of tested groundwater according to corresponding water quality classes and the assessment of their chemical state based on [16]**

Measurement point	pH	Conductivity, $\mu S/cm$	T, °C	Petroleum hydrocarbons [H] mg/L	Cations, mg/L					Anions, mg/L						Groundwater chemical status
					NH_4^+	Mg^{2+}	K^+	Na^+	Ca^+	NO_3^{-} [H]	NO_2^{-} [H]	Cl^-	F^- [H]	PO_4^{3-}	SO_4^{2-}	
1	I	II	III	IV	I	I	I	I	III	IV	I	II	I	I	II	–
2	I	II	IV	IV	I	II	I	I	IV	I	I	III	II	I	II	–
3	I	II	IV	III	I	I	I	I	III	III	I	IV	I	III	II	–
4	I	II	IV	III	I	I	I	I	III	II	I	IV	I	I	II	–
5	I	II	III	V	V	I	I	I	IV	I	I	IV	I	I	I	–
6	I	IV	IV	IV	V	II	I	I	IV	I	III	IV	I	I	I	–
K	I	II	III	I	I	I	V	I	III	II	III	I	I	I	II	–

[H] - physicochemical element for which it is not allowed to exceed the limit values when determining the groundwater quality classes at the measurement point

„–" - groundwater chemical status assessed as poor

K - control sample

All tested groundwater samples demonstrated good chemical status, regarding the content of phosphates and sulphates, being classified to I, II or III water quality class.

Very high content values of ammonium ion measured in waters from the areas of stations no. 5 and 6 classify them to the lowest water quality class, thus determining their chemical status as poor. High concentrations of ammonia nitrogen at a minimal contents of nitrates and nitrites demonstrate recent contamination caused by sewage water.

Groundwater within the area of fuel station no. 6 indicates exceeded limit values of calcium ion concentration, which classify them into the water quality class IV with poor chemical status.

Concentrations of magnesium, potassium and sodium at each measurement point were within the limits, representing good chemical status of waters.

In turn, high concentration of potassium ions in the control sample which classify the water to V quality class, can be caused by the fact that the land on which the measuring point was located is of agricultural use. Potassium fertilizers used in farming are the main source causing his pollution.

Among all of the tested groundwater samples, it turned out that water collected within the area of stations no. 5 and 6, located in strict centre of the city, demonstrated the highest pollution. Urban areas are characterized by a significant accumulation of pollutants in the form of particulate matters from fuel combustion processes, abrasion of asphalt surfaces and processes causing building destructions. All contaminants are washed and rinsed by rainwater and they infiltrate into the groundwater.

Conclusions

In order to assess the impact of fuel station activities on the chemical status of groundwater, a laboratory analyses of 65 groundwater samples collected from 20 measurement points taken at monthly intervals during a four-month period were performed. The samples were collected from piezometers being used as measurement points, placed on areas of petrol stations in the territory of the city of Opole.

In the groundwater samples following were tested: petroleum hydrocarbons, specific electrical conductance, the pH value, "*in situ*" temperature of the collected samples, and water level in piezometers. Furthermore, the research tests covered determination of ionic composition regarding anions: F^-, Cl^-, NO_2^-, NO_3^-, PO_4^{3-}, and SO_4^{2-}, as well as cations: NH_4^+, Na^+, K^+, Mg^{2+}, and Ca^{2+}. The obtained results were compared with limit values covered under the Regulation of the Minister of Environment dated 23 July 2008 [16]. Based on the Regulation, groundwater quality classes were determined in the collected groundwater samples.

Within the area of selected fuel stations, extremely large groundwater contamination was demonstrated. Exceeded results of groundwater quality tests - when compared to indices given by regulations - can indicate unsatisfactory technical

condition of these facilities, especially in relation to the high content of petroleum substances. At the area of station no. 5, in addition to high concentrations of petroleum hydrocarbons, the presence of petroleum product in one of the piezometers was observed in each collection. The occurrence of pure petroleum product in a piezometer indicates a very high groundwater pollution and this may indicate leaking underground fuel tanks.

The pollution scale demonstrates the need for further research on monitoring the groundwater status. Based on the research results, it can be concluded that activities of fuel stations and the location of measurement points in the center of urban location, are a potential cause of groundwater contamination.

It should be noted that detected groundwater pollutions may have various origins.

Due to the nature of the studied objects, it can be concluded that their activities contribute to the pollution of petroleum substances. The presence of groundwater contaminations at the individual measuring points can be also determined by the groundwater runoff, resulting from the slope of the terrain. A significant impact on the exceeded limits occurrence can have structure of the bedrock. The probability of contaminations infiltration at stations no. 1 and 4 can be excluded because of the poorly permeable rendzinas properties.

Due to the type of soil present at the stations no. 2, 3 and 5, 6, groundwater contamination could result from infiltration of surface contaminants.

Due to unsatisfactory research results of the analysed samples, further actions should be undertaken to improve the condition of groundwater in the analysed area. Among necessary actions to be undertaken are undoubtedly the elimination of leakage from tanks collecting fuel and checking of installations discharging wastewater.

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Charakterystyka fizykochemiczna i ocena jakości wód podziemnych w rejonie wybranych stacji paliw miasta Opola

Monitoring jakości wód podziemnych ma na celu dostarczenie informacji o stanie chemicznym wód podziemnych, śledzenie jego zmian oraz sygnalizację zagrożeń ekologicznych. Działania monitoringujące umożliwiają właściwe zarządzanie zasobami wód podziemnych oraz odpowiednią ocenę efektywności działań zapobiegawczych. Celem pracy jest analiza stanu jakościowego wód podziemnych pobranych z piezometrów znajdujących się na obszarze wybranych stacji paliw na terenie miasta Opola. Próbki wód pobierano w odstępach comiesięcznych przez okres czterech miesięcy. Oznaczono w nich zawartość substancji ropopochodnych, anionów: F^-, Cl^-, NO_2^-, NO_3^-, PO_4^{3-} i SO_4^{2-} oraz kationów: NH_4^+, Na^+, K^+, Mg^{2+} i Ca^{2+}. Badania zawartości jonów wykonano z wykorzystaniem metody chromatografii jonowej. Substancje ropopochodne oznaczono jako indeks oleju mineralnego metodą chromatografii gazowej. W pracy podano warunki prowadzenia analizy chromatograficznej. Zbadano również przewodność elektryczną właściwą oraz wartość pH, temperaturę „*in situ*" oraz poziom lustra wody w piezometrach. Opierając się na wytycznych określonych w Rozporządzeniu Ministra Środowiska z dnia 23 lipca 2008 r., określono klasy jakości badanych wód podziemnych. Wyniki badań zostały porównane z parametrami charakteryzującymi wodę podziemną z punktu pomiarowego znajdującego się w luźnej zabudowie miejskiej na obrzeżach miasta Opola. Stwierdzono znaczny wpływ umiejscowienia punktów pomiarowych na stopień zanieczyszczenia próbek wód podziemnych. Największym zanieczyszczeniem charakteryzowały się próbki pobrane z obszaru stacji 6 zlokalizowanej w ścisłym centrum miasta. Wyniki badań przekroczyły normy graniczne odpowiadające wodom o dobrym stanie chemicznym w przypadku sześciu z piętnastu badanych parametrów fizykochemicznych. W badanych próbkach wód odnotowano znaczne podwyższone wartości przewodności, temperatury oraz zawartości chlorków i jonów wapnia klasyfikujące je do IV klasy jakości, jak i przekroczone wartości stężeń substancji ropopochodnych i jonu amonowego na poziomie odpowiadającym V klasie jakości. W związku z niezadowalającymi wynikami badań analizowanych próbek należy podjąć działania mające na celu poprawę stanu wód podziemnych na badanym obszarze. Do działań koniecznych należy niewątpliwie likwidacja nieszczelnych zbiorników paliwowych, kontrola instalacji odprowadzania ścieków.

Słowa kluczowe: wody podziemne, klasy jakości wody podziemnej, chromatografia jonowa, substancje ropopochodne, zanieczyszczenie środowiska wodnego