IMPACT OF PETROLEUM INDUSTRY IN THE PHYSICO-CHEMICAL CHARACTERISTICS OF NATURAL WATERS FROM KUÇOVA REGION (ALBANIA)

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Abstract. Kuçova region lies on Miocene and Oligocene flysch deposits. This region includes some structural units of the subzone syncline of Permetit, subzone anticline of Berati and the extreme east of the Adriatic depression. The petroleum exploration has detailed geological setting, mainly of the Messinian-Pliocene section, flysch and carbonate deposition of eroded anticline of Kuçova. The hydrographic network is represented by the Devoll river, artificial lakes in the hilly area and a dense network of canals that discharge to the river. In the region of Kuçova, there are four water bearing complexes where the main one is of Quaternary deposits. Hydrogeological interpretations of Kuçova region are based on two results: those carried out earlier and those carried on recently in the context of studies on environmental rehabilitation. Water that comes from the separation of crude oil in the separators has a high content of total dissolved solids (TDS up to 100 g/l). Surface waters and those of the Quaternary deposits are somewhat polluted by the petroleum industry and urban emissions, particularly in the area of the former petroleum refinery. The TDS goes up to 6,363 mg/l, while that of natural water sources and of canals varies in the range 790 mg/l ÷ 1,217 mg/l. The analysis emphasized that natural waters (not those of oil traps) have been polluted by the petroleum industry. Based on the physico-chemical characteristics of natural waters, we came to the conclusion that some water sources (Spring and P-1) should not be used for domestic purposes. The obtained results serve local government structures for the management of hydrological systems, water bearing complexes and rehabilitation of wastewater.

Keywords: Quaternary water reservoir**,** Water reservoir where oil circulation predominates, Total dissolved solids, Total Petroleum Hydrocarbons.

Rezumat. Impactul industriei petroliere asupra caracteristicilor fizico-chimice ale apelor naturale din regiunea Kuçova (Albania). Regiunea Kuçova se suprapune depozitelor de fliş miocene și oligocene. Această regiune include unele unități structurale ale subzonelor sinclinalului Permetit, anticlinalului Berati și estului extrem al depresiunii Adriaticii. Exploatarea petrolului a permis redarea detaliilor geologice, în principal, din secțiunea Messinian-Pliocen, a depunerilor de fliş şi carbonat ale anticlinalului erodat de la Kuçova. Rețeaua hidrografică este reprezentată de râul Devoll, lacurile artificiale din zona de deal și de o rețea densă de canale de deversare. În regiunea Kuçova sunt patru complexe acvifere, principal fiind cel din depozitele cuaternare. Interpretările hidrogeologice ale regiunii Kuçova se bazează pe două rezultate: cele efectuate mai devreme și cele efectuate recent, în cadrul studiilor privind reabilitarea mediului. Apa care provine din separarea țițeiului prezintă o valoare ridicată a totalului solidelor dizolvate (TSD până la 100 g/l). Apele de suprafață și cele din depozitele cuaternare sunt poluate într-o oarecare măsură de industria petrolieră și de emisiilor urbane, în special în zona fostei rafinării de petrol. TSD ajunge la 6.363 mg/l, în timp ce pentru apa din sursele naturale și canale variază în intervalul 790 mg/l ÷ 1.217 mg/l. Analizele au evidenţiat faptul că apele naturale (nu cele provenite din separarea ţiţeiului) au fost poluate de industria petrolieră. În funcție de caracteristicile fizico-chimice ale apelor naturale, s-a ajuns la concluzia că unele surse de apă (izvor și P-1) nu ar trebui să fie utilizate de către populaţie. Rezultatele obţinute servesc autorităţilor locale pentru managementul sistemelor hidrologice complexe, a acviferelor și reabilitarea apelor uzate.

Cuvinte cheie: ape subterane cuaternare, rezervor de apă în cazul în care circulația uleiului este predominant, total solide dizolvate, hidrocarburi totale din petrol.

INTRODUCTION

Hydrogeological studies are performed for several reasons: water supply (Kuçova region has no natural water sources); in the framework of petroleum production; in the context of the preparation of the hydrogeological map of Albania; under the impact of petroleum industry pollution of natural waters and surface studies for the rehabilitation of the former petroleum refinery.

In the region of Kuçova, the main water-bearing (aquifer) complexes are those of the Messinian section and of the Quaternary sediments, which are used for public water supply and irrigation.

In the water-bearing complex of the Messinian deposits, three areas are separated, depending on the relations with: eroded limestone of the Kuçova anticline; oil traps; degree of surface water infiltration.

Quaternary deposits waters that lie on the surface of the former petroleum refinery are contaminated and cannot be used for public water supply.

Many canals cross the area of the Kuçova oilfield and flow into the river. Based on their analysis water pollution is emphasized. In conformity with the scale of the water pollution (contamination) (Na / Cl ratio) waters of oil traps and polluted waters are separated.

The implementation of the hydrogeological studies should be accompanied by geological and geomorphological features of the region.

GEOLOGICAL SETTING

Kuçova region lies in the south-eastern extremity of the Adriatic depression and Ionian area. The Ionian zone is represented by Kuçova eroded anticline and covered by the Adriatic depression (Fig. 1). The Oligocene flysch deposits in the east of the region are those of the east wing of the Kuçova anticline (BANDILLI et al., 2005).

Carbonate section (Cr_2-Pg_2) builds the Kuçova anticline. These deposits are represented by some lithological types of limestone.

Flysch deposits (Pg_3) are a combination of claystone, siltstone and sandstone layers. They have very low water capacity (PRIFTI et al., 2007).

Molasse deposits represented by those of the Messinian and Pliocene.

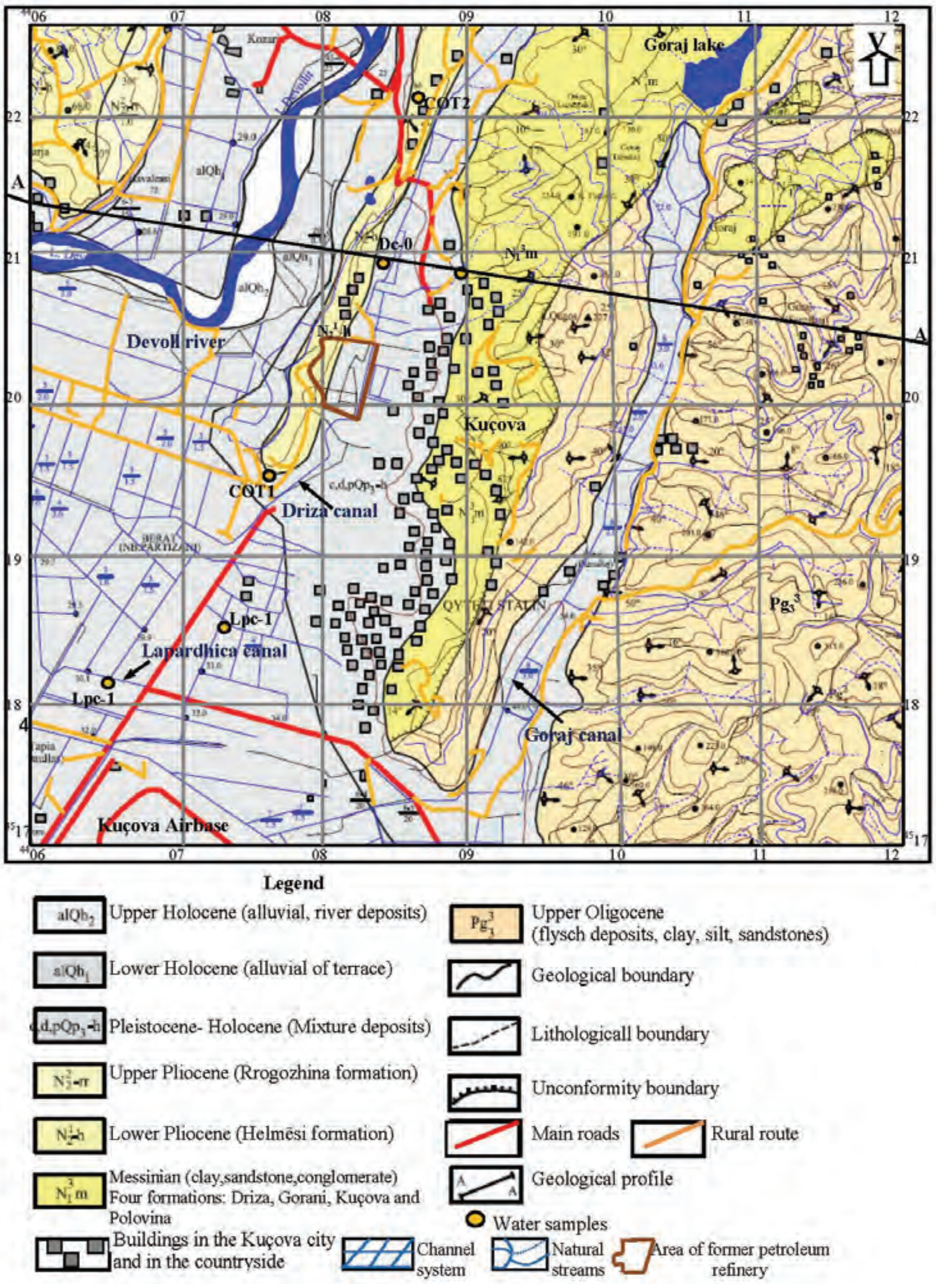

Figure 1. Geological map of Kuçova region (prepared by Prifti).

Messinian deposits (Messinian - N_1^3m) are exposed on the east of the Kuçova town and in area of town. To the west, they are covered by Quaternary and Pliocene deposits. They placed unconformity on flysch deposits. These sediments are divided into four formations: Driza, Gorani, Kuçova and Polovina. These formations are represented by a combination of clay, silt and sand packages (PRIFTI et al., 2007; PRIFTI et al., 2014).

Terrigenous deposits of the Pliocene (Pliocene - N2) are an integral part of the Adriatic depression. In these deposits, there are two lithostratigraphic units: the "Helmës" (mostly clay) and "Rrogozhine" (mainly sandstone) formations.

Geomorphological Conditions. Geomorphologically, Kuçova region lies in a hilly and flat area with local elevations not exceeding 227 m (east of Kuçova town).

Hydrological Conditions. The region is crossed by a system of artificial canals. Goraj canal starts from the artificial lake of Goraj, collects waters of others canals and, in west, it passes near the airbase and flows into the Osumi River.

Lapardhic canal collects the waters of the southern part of the Kuçova and southern sectors of the oilfield. This canal passes in the west and join Goraj canal.

Artificial canal named "Prroi i Drizes" (Driza canal), which flows through Driza valley. This canal collects several canals that pass through the valley and serve as sanitary sewerage. Driza canal goes westwards and, after about 3.5 km, it flows into the Osum River as its right-hand tributary.

The region is crossed by three valleys with N-S orientation. These valleys are (east to west): Goraj valley; Driza Valley and the Devoll River valley. Goraj Valley is separated from Driza Valley by a system of hills up to a height of 227 m and belongs to the Oligocene flysch deposits and those of the Messinian. The Valley of the Driza shared with the Devoll valley from a height of 25m belongs to the Pliocene deposits (Helmës formation).

The oilfield the Kuçova lies to the west of the Messinian transgression base and on the Pliocene, Quaternary deposits.

The premises of the former refinery lie in the floodplain of the Pliocene and Quaternary deposits. The ground is moderately sloping in the NNE – SSW direction.

Based on the performed analysis, on conducted studies and surveys in Kuçova area, four water-bearing complexes are distinguished (Fig. 2).

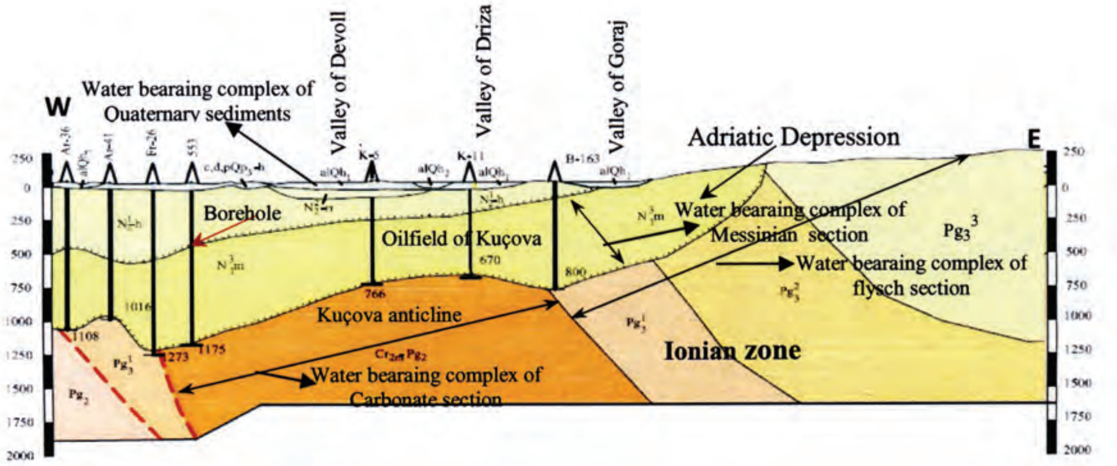

Figure 2. Water-bearing complex in Kuçova region.

The water-bearing complex of the carbonate reservoir. This complex is not exposed on the surface because it is covered by Messinian deposits. Structurally, it represents the eroded Kuçova anticline. Some wells penetrated this water-bearing complex providing water with TDS up to 20 g/l. TDS values increase with depth.

The water-bearing complex of the flysch deposits. This aquifer complex consists mainly of Oligocene flysch deposits nearly impermeable. They have a very low water-bearing capacity.

The water-bearing complex of the Miocene reservoir. In accordance with the distribution of the sandstone reservoir, three levels are distinguished.

a. The lower hydrogeological level includes the formation of Driza and bottom of the Gorani formation. This level is placed directly on the eroded surface and communicates with the water-bearing complex of carbonate. Waters are mixed and have low values of TDS (20 ÷ 40 g/l) and high potential energy, about the same as the complex of the carbonate aquifer.

b. The middle hydrogeological level includes the bottom water of oil traps. Within this level, there are met waters with high values of TDS (70 ÷ 100 g/l). Waters focus on two levels:

- In separate horizons;
- Bottom waters under the oil-water contact of the oil traps (TUSHE & SHTREPI. 1996; PRIFTI & BITRI. 2000).

Characteristic to this level is the fact that it does not communicate either with the limestone layer or the surface.

These are the waters of sedimentation and modified by the migration of petroleum and have high values of TDS.

c. The upper hydrogeological level is included in the deposits of the Polovina and Kuçova formations. This level is characterized by mixing with infiltration waters from the surface.

This level has water with low content of TDS that is used for public supply.

The first Quaternary pseudo water reservoir oil body (Fig. 3) has an episodic character similar with that in the western hill. During the subsurface exploration work, its thickness was verified at about 0.4 – 2.0 m. Lithologically, it is formed by anthropogenic fills (predominantly clayey to sandy loam, locally with gravel and remnants of building structures), less often autochthonous clayey hummus loams (URBÁNEK & BARTOŇ. 2009).

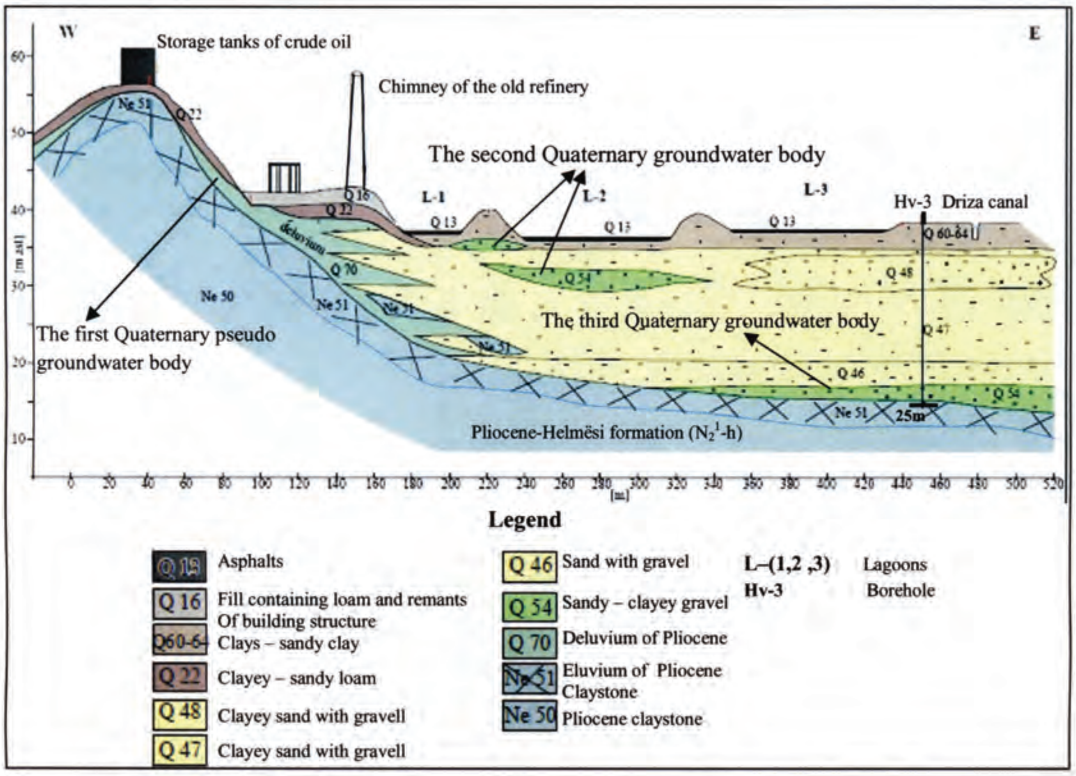

Figure 3. The Quaternary water reservoir oil body on former petroleum refinery.

The second Quaternary water reservoir oil body is mostly formed by sands and gravel. It was not documented along the whole width of the studied part of the floodplain, but only in several separated areas irregularly arranged over the floodplain area. This suggests the presence of a partial palaeo-canal in which gravels, sands and clays were sedimented in relationship with the dynamics of the stream.

The third Quaternary water reservoir oil body was encountered only by drilling work at the base of the Quaternary filling of the floodplain. Its thickness is about 1.5 to 7.0 m.

With respect to the results of drilling work and the finding that in the vicinity of the former petroleum refinery there is a number of private wells (one is P-1 well) encountering gravel layers at the similar depth, it can be assumed that these basal gravel layers have a considerable areal extent. These gravels are utilized for supplying industrial or drinking water to the local population.

MATERIALS AND METHODS

Water reservoir oil samples were collected from a total of 8 hydrogeological objects. Both the existing hydrogeological objects (St-1, St-7, COT1, COT2 wells, Table 1) and the newly installed hydrogeological objects (Hv-1, Hv-2, Hv-3, P-1, Table 2) were used for water reservoir oil sampling (Fig. 4).

Three samples of surface water from Driza canal running through the Driza valley were taken from newly established sampling points Dc-0, Dc-1 and Dc-2. Two water samples from the Lapardhica canal and one from a spring were taken.

Physical-chemical determinations of natural waters are conducted in two laboratories: in the laboratories of the company GEOtest Brno, a.s. (in the framework of an environmental remediation project); in the laboratories of the Albanian Geological Survey (Water sample taken in a public boreholes P-1 for the preparation of the paper).

In the sample of water taken from the well P-1, TPH was not defined but organic matter was. Definitions of the characteristics of natural waters are conducted in three different times. Initially, tests were carried out in the framework of hydrogeological studies to Kuçova oilfield, a few years ago are analysed in the framework of Environmental remediation project and an analysis was conducted in the framework of the preparation of the paper. Analytical determinations were performed in accordance with ISO European standard.

Table 1. Some result of chemical parameters water samples.

Parameters	**Units**	**Hv-1**	**Hv-2**	**Hv-3**	**DC-1**	**DC-2**	**Spring**	**St-1**	**St-7**	**P-1**
TPH	(mg/l)	0.06	0.07	0.84	<0.05	0.97	<0.05	<0.05	<0.05	11.9
pH		7.04	7.06	6.34	7.35	7.34	7.9	6.96	7.01	6.98
Hardness	(mmol/l)	5.03	5.08	38.94	4.46	4.84	1.24	7.42	5.98	31
Conductivity	µS/cm	1,013	1,015	5,210	1,010	1,056	930	1,362	1,219	1,223
TDS	(mg/l)	844	938	6363	891	864	868	1239	1081	981.23
COD	(mg O2/l)	2.49	3.57	9.79	6.21	15.71	1.86	2.49	3.88	0
Na	(mg/l)	43.6	52.7	355	70.8	62.4	207	59	71.6	50.04
K	(mg/l)	2.81	0.97	10	6.4	9.47	3.45	1.17	1.66	2.96
Ca	(mg/l)	125.1	88	364.5	107.8	116.3	10.1	196.9	87.1	139.56
Mg	(mg/l)	46.4	70.1	604	43	47.1	24	60.9	92.5	49.96
Mn	(mg/l)	0.7	1.51	9.57	0.17	0.41	**0**	<0.05	1	1.891
Fe	(mg/l)	5.3	7.52	17.88	0.31	0.35	0	<0.01	0.65	0.04
Cl	(mg/l)	112	61	47	88	78	74	108	128	89.44
SO4	(mg/l)	127	85	3447	74	73.5	1.5	236	40.4	108.2
NO2	(mg/l)	0.79	<0.01	<0.01	6.52	0.03	<0.01	<0.01	<0.01	0.01
NO3	(mg/l)	3.5	10.3	<3.0	11	<3	<3	7.3	<3	3.64
F	(mg/l)	0.43	0.52	0.46	0.34	0.58	0.37	0.33	0.7	0.44
HCO3	(mg/l)	375.9	560.8	1305.8	478.4	566.3	546.1	569.3	657.2	530.7
PO43	(mg/l)	<0.05	<0.005	.0.05	1.66	3.68	<0.005	<0.05	<0.05	<0.05
NH4	(mg/l)	<0.1	<0.1	1.48	2.31	6.77	1.03	.0.01	0.15	0.24

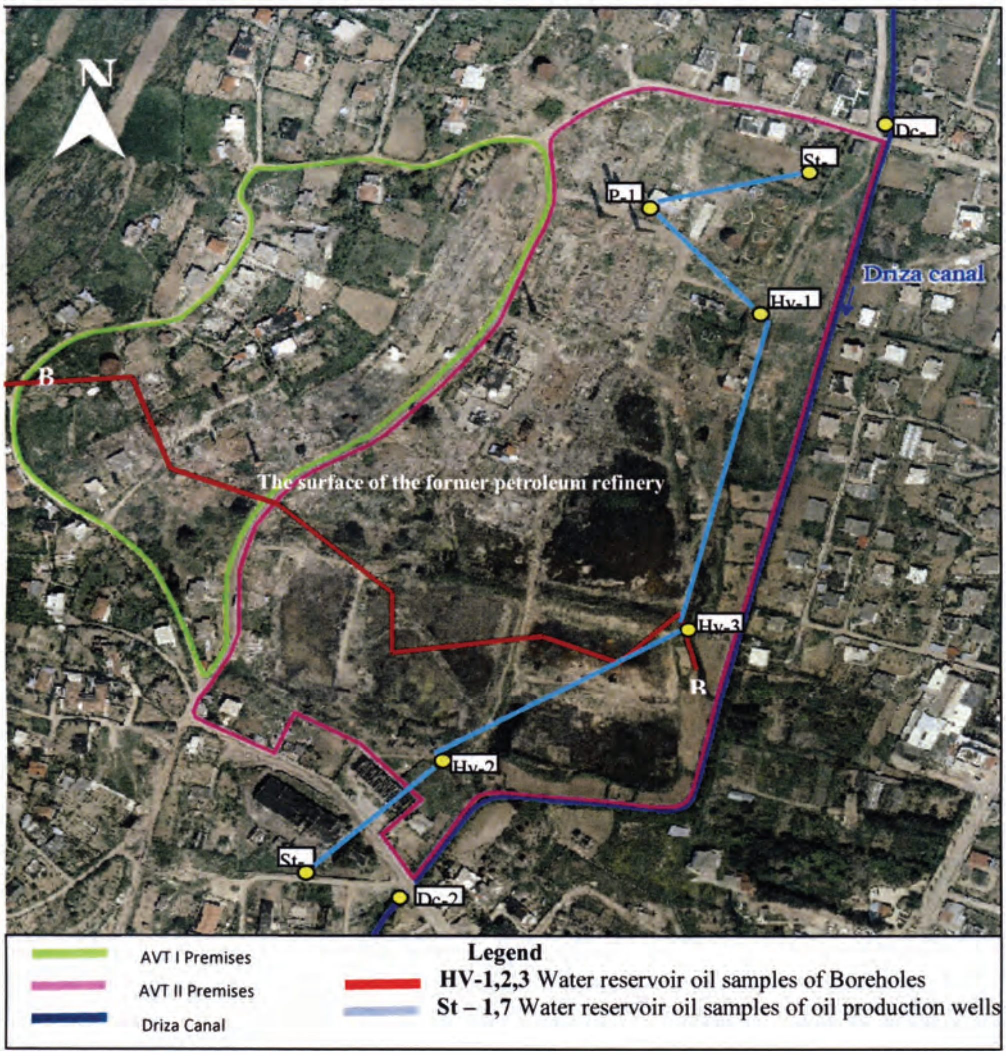

Figure 4. Water samples from the area of the former petroleum refinery.

Table 2. Physical-chemical parameters of the water samples in area of the former petroleum refinery.

Parameters	Units	Lpc-1	Lpc-2	DC-0	COT1	COT2
pH		7	8.3	8.5	6	6.5
Hardness	(mmol/l)		11.2	12	0	0
TDS	(mg/l)	790	1217	897	24300	91,400
Na + K	(mg/l)	169.74	264.96	125.1	7,033.4	27,588.5
Ca	(mg/l)	70	128	144	850	4,000
Mg	(mg/l)	6	57.6	57.6	990	2,400
Cl	(mg/l)	208.7	379.3	278.3	15,133.65	55,664
SO4	(mg/l)	0	19.2	0	120	1,200
CO3	(mg/l)	0	0	36	0	0
HCO3	(mg/l)	335.5	366	512.4	122	396.5
Iodine	(mg/l)	0	1.52	0.1	20.4	63.2
Bromine	(mg/l)	0	1	0.5	0	0
Boron	(mg/l)	0	0	0	0	0
NH4	(mg/l)	0	2.2	0	0	0

RESULTS AND DISCUSSIONS

The results of the physical-chemical analysis of water are given in two tables. The physical-chemical parameters, which showed distinctive changes within the obtained group of analyses, were plotted into figures, which correspond to an idealized section through the site of interest in the N-S direction. This profile was chosen so as to reveal potential differences in the hydrochemical indicators of the water reservoir oil across the floodplain. Figures show at first sight that most values of the selected parameters determined in a sample from spring are the lowest as compared to the other samples.

The values of the parameters from the spring can be considered as the background, because they lie outside of the former petroleum refinery, against the direction of water reservoir oil flow. In the group of boreholes St-1, Hv-1 and Hv-2, most of the parameters are comparable (Fig. 5).

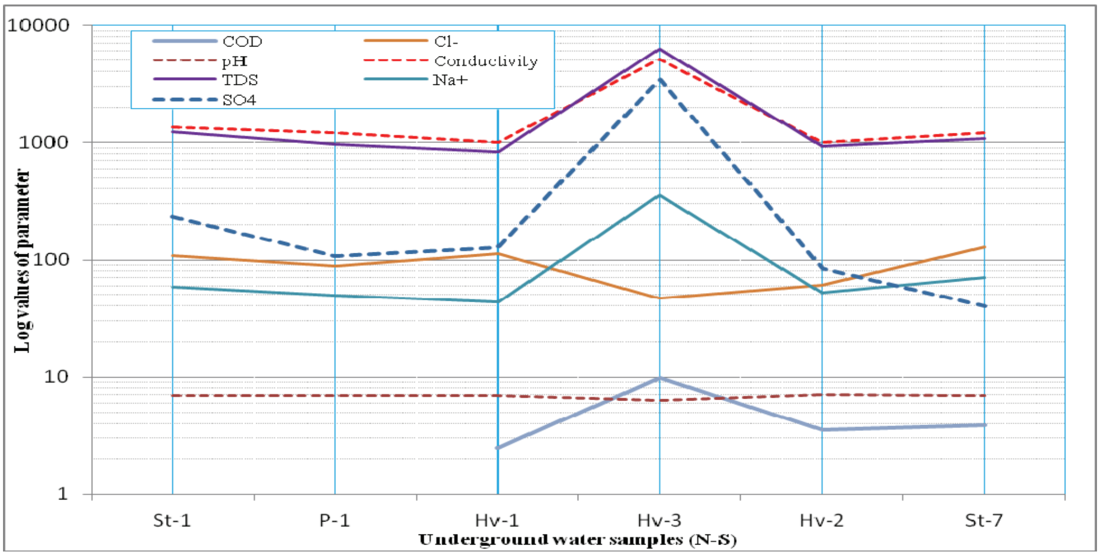

Figure 5. Correlation of water reservoir oil parameters in the area of the former petroleum refinery.

Borehole Hv-3 is different rather by increased values than by their different correlation (URBÁNEK & BARTOŇ, 2009). Another group of water samples with similar characteristics is that of COT1 and COT2, which originated in oil traps (PRIFTI & KURTI, 2000).

The results of the analyses of the water samples emphasized that the measured values of pH in stagnant surface waters were not influenced by the leachate of acid refining residues in a period of more intensive precipitation and smaller evaporation during different times. The measurement of pH in August, when temperatures reach a maximum of about 40°C and the area has lower precipitation amounts with intensive evaporation, proved that pH was markedly shifted into the acid area. The measured value proves a distinctive relationship between pH and the degree of weathering (URBÁNEK & BARTOŇ, 2009).

The variation of values of this parameter obviously relates to the intensity of sewage waters into Driza canal. The pH value is higher in a period of intensive inflows.

A water reservoir oil sample from a spring was taken for the purpose of verifying the quality of the water reservoir oil, in relation to the limits for drinking water because it is used for water supply.

Surface-water samples were taken from two canals: a canal running through Driza valley, above (DC-0, DC-1) and below the area of the former petroleum refinery (DC-2) and two samples were taken from Lapardhica canal (Lpc-1,2).

Based on two parameters such as TDS and Na/Cl ratio we can render the metamorphism degree of natural water or effects of metamorphic waters of oil traps (PRIFTI & KURTI, 2000).

High values of TDS and lower values of the Na /Cl (< 1) indicate metamorphic water. The figure shows clearly that the spring and Hv-3 borehole waters are not metamorphic. This phenomenon must be related to the substances that are deposited during the petroleum refinery process. The waters with higher values of TDS are waters of oil traps, which are metamorphic.

The high-risk contamination at the site is posed by the following contaminants:

In the unsaturated zone:

♦ PS (determined as TPH) which particularly include oil, asphalt, tars and coke oven residues; and

♦ Vanadium.

In the saturated zone:

♦ Free phase of PS (gasoline, Diesel oil, oil) on the water surface; and

♦ MBAS and vanadium.

Another risk factor is the occurrence of sulphuric acid in tars and the relating low pH values of potential leachates.

In the selected waste samples, the physical and chemical properties of the waste were recorded in relation to the possible handling of them and to the possibilities of its further treatment for the purpose of its disposal.

It seems clear that all other waters are influenced by metamorphic waters. They can be separated into two groups:

The first group - TDS fluctuates in the interval 790 mg/l ÷ l 938 mg/l and Na/Cl fluctuates in the interval 0.8 ÷ 0.86. In this group, there are included water samples of Dc-1, DC-2, Hv-2 and Lpc-1.

The second group - TDS ranges within the interval 844 mg/l ÷ 1,239 mg/l and Na/Cl fluctuates in the interval 0.38 ÷ 0.69. In this group, there are included water samples of Lpc-2, P-1, DC-0, Hv-1 and two wells of St-1, St-7.

The increased concentrations of NH4+ in the area of borehole Hv-3 could indicate a reduction medium or faecal contamination. The same result is for the water of the spring.

A high content of NH4 + was also registered in case of the two samples of Driza canal (Dc-1 = 2.31 mg/l; Dc-2 = 6.77 mg/l) and one of the Lapardhica canal (LCP 2 = 2.2 mg/l). This is the result of sanitary wastewater discharge and faecals. There are also filters from lagoons where acid substances are deposited during refinery process.

Mineralization is greatly increased in the area of hot spots of contamination around boreholes Hv-3. It is indicated by the increased content of cations, especially Na, Ca, Mg, K, Fe, Mn, and anions, especially sulfates, hydrogen carbonates and chlorides. The increased mineralization of the water reservoir oil is caused by the presence of pollutants (particularly HM), which due to their high sorption capacity, displace elements naturally occurring in soil (Ca, Mg, K, Na) and above all by the process of a long-lasting formation of leachates from dumped wastes and their concentration as a consequence of the limited permeability of the basement.

Total petroleum hydrocarbons (TPH) were determined in the water samples of Driza canal, water reservoir oil of boreholes in the area of the former petroleum refinery and the water of the spring.

High TPH values is registered by two water samples: Dc-2 (0.97 mg/l) and Hv-3 (0.84 mg/l), while the two water samples Hv-1 and Hv-2 have values of 0.06 mg/l and 0.07 mg/l. Other water samples have TPH <0.05 mg/l. In case of public borehole (P-1), TPH is not estimated but complete content of organic matter.

This phenomenon is caused particularly by the fact that there is a sewerage outlet in a section of Dc-1 to Dc-2, which discharges municipal wastewaters (so-called gray water) from an adjacent residential area and industrial wastewaters as well. Theoretically, changes in redox conditions could have an effect on this phenomenon, when the surface waters in the canal would be recharged by water reservoir oil from the former petroleum refinery with redox potential due to the hydraulic continuity between the Quaternary aquifer and the canal Driza (URBÁNEK & BARTOŇ, 2009).

The chemical analysis of the water reservoir oil sample from the spring for the content of TPH did not prove the presence of contamination by petroleum substances, even though the water was sensorially unsatisfactory. The water sample was clear without turbidity, without a free phase of PS, but had a strong smell of petroleum substances or other organic substances and had a soapy taste.

CONCLUSIONS

Natural water of the oil traps is metamorphic in compliance with two parameters such as TDS and the Na/Cl ratio.

The spring and Hv-3 borehole waters are not metamorphic (Na/Cl > 1), but may be influenced by sodium substances.

Others water samples are influenced by metamorphic waters of the oilfield or by technological waters.

Mineralization is greatly increased in the area of hot spots of contamination around boreholes Hv-3. The increased mineralization of the water reservoir oil is caused by the presence of pollutants.

The increased concentrations of NH4+ in water samples could indicate a reduction medium or faecal contamination.

High TPH (Total petroleum hydrocarbon) values is registered by two water samples: Dc-2 (0.97 mg/l) and Hv-3 (0.84 mg/l), while other water samples have TPH < 0.07 mg/l.

Driza canal is not considered as a threatened object due to the unproved hydraulic continuity between this canal and the area of the former petroleum refinery.

The chemical analysis of the spring water did not prove the presence of contamination by petroleum substances. The water sample was clear without a free phase of hydrocarbons, but had a strong smell of petroleum substances or other organic substances.

The waters of the spring and P-1 borehole must not be used by local residents.

In the region of Kucova, the main water-bearing (aquifer) complexes are those of the Messinian section and of the Quaternary sediments, which are used for public water supply and irrigation.

The middle hydrogeological level includes the bottom water of oil traps. Within this level, there are met waters with high values of TDS (70 ÷ 100 g/l).

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