

Determination of soil and groundwater contamination resulting from the hydrocarbon exploitation activity

Original scientific paper



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Abstract

In order to create the basis for systematic environmental monitoring and eventual remediation procedures, at sites potentially contaminated by different activities, qualitative and quantitative determination of soil and groundwater contamination status must be carried out. Although European Union has not regulated the area of soil protection by a separate directive so far, the requirements for protection of this environmental component are set in other regulations, in particular the *Directive 2004/35/EC on environmental liability with regard to the prevention and remedying of environmental damage* and the *Directive 2010/75/EU on industrial emissions*. Mentioned acts clearly define the activities which can cause environmental damage and therefore are responsible for prevention measures application and remediation. Given that Croatian regulation prescribes only the methodology for monitoring soil quality of agricultural land, underground contamination analysis described in this paper represents an example of pollution risk assessment at sites that do not meet the agricultural land standards. The selected site is connected with oil and gas production activities for many years. On the basis of conclusions of field research carried out by authorized company, involving soil and water sampling and laboratory analyses, and in order to help to obtain a clearer picture of pedosphere status and to make the conclusions regarding further monitoring, the maps of spatial distribution of pollutants are done in this paper.

Keywords: soil and underground water contamination, hydrocarbon exploitation, Nearest Neighbour method.

1. Introduction

Soil is the surface part of the Earth's crust, located between the lithosphere and the atmosphere. From the constructional point of view, it is about a natural mixture of mineral grains, i.e. unconsolidated set of solid particles of mineral and organic origin (**Maksimović, 2005**). Furthermore, the agronomic aspect defines the soil as a thin layer of lithosphere that supplies the plants with water and other essential substances, while pedochemistry considers the soil as a constituent of solid particles of organic and inorganic origin, water, air, and microorganisms, derived from the source rocks under the influence of pedogenic factors, through a series of processes.

Existence of numerous definitions indicates multifunctional character of soil. In addition to its primary role, i.e. the production of organic matter in the photosynthesis process, there are some other important roles, such as the ecological-regulatory role, since the soil is a natural purifier and buffer due to filtering precipitation water, and binding pollutants by colloidal complex. Soil significantly affects the content and total amount of CO₂ in the atmosphere, and has the spatial role, representing the basis for urban expansion and integral element in the landscape design (**Bašić, 1994**).

The soil solid phase may refer to organic and mineral fraction. The organic fraction consists of roots and humic compounds (dead plant material in decomposition stages) which is important in the electrostatic adsorption of cations, water bonding, and the coherence of soil particles, i.e. it represents the main reactive phase for organic compounds (e.g. pesticides, oil). The soil mineral fraction contains quartz, other oxides, and clay minerals. The soil microbial biomass consist of many microorganisms which participate in oxidising plant and animal residues what results with generation of plants nutrients (**Bobić, 2005**).

1.1. Environmental protection as a legal requirement

Soil is considered almost non-renewable natural resource, due to limited natural distribution, but also due to the fact that for generation of 1 cm of this resource more than a thousand years are needed. However, despite the fact that environmental protection has become an integral part of sustainable development of European society, and therefore it is present in the businesses that have some impact on the environment, systematic soil protection in the European Union is still missing.

Although some EU member states (e.g. Germany, Denmark, the Netherlands) have clearly set the standards for soil protection, most of the countries still have not. Namely, the *Thematic Strategy on Soil Protection in the EU* was adopted in 2006, but the adoption of soil protection related directive, which proposal is repeatedly considered, has not been achieved yet. Thus, certain provisions regarding soil management and protection can be found within the legal acts regulating other areas, in particular the *Directive 2004/35 /EC on environmental liability with regard to the prevention and remedying of environmental damage* and the *Directive 2010/75/EU on industrial emissions*, but also the *Directive 2008/98/EC on waste*, the *Directive 2000/60/EC on water* and the whole set of directives regulating dangerous substances in groundwater and the environment.

Mentioned EU regulation has been transposed into the legal order of the Republic of Croatia in a way that the *Environmental Protection Act (OG 80/13, 153/13, 78/15)* obliges legal entities (environmental polluters) to apply prevention measures, remediation activities, and environmental monitoring. Then, the *Regulation on Environmental Permit (OG 8/14, 5/18)* establishes industrial activities in Croatia that could cause soil, air and water pollution. Integrated pollution protection conditions are set, and for the purpose of carrying out remediation, a baseline study before the start of operation and after facility closure has to be done.

However, in Croatia, as it is the case in most of the EU countries, there is no statutory obligation to identify contaminated and potentially contaminated sites out of the system of environmental permits. As far as pollutants and threshold limits are concerned, Croatian regulation considers only the category of agricultural land through the *Agricultural Land Act (OG 20/18)* and the *Ordinance on the Protection of Agricultural Land from Pollution of Harmful Substances (OG 9/14)*. Therefore it is quite impossible to do systematically monitoring of the polluted land used for other purposes.

According to the *Agricultural Land Act (OG 20/18)* soil degradation means following: degradation in intensive production (physical, chemical and biological characteristics) (a), contamination of harmful substances and organisms (heavy metals, potentially toxic elements, pesticides, organic pollutants and pathogenic organisms) (b), displacement (water and wind erosion, crop rotation, storage space, waste or other soil cover) (c), and transformation (urban areas, industrial and energy facilities, roads, hydro accumulation and exploitation) (d). The *Ordinance on the Protection of Agricultural Land from Pollution Damage (OG 9/14)* defines sources of pollution as follows: industrial production and services, industrial waste, urban waste, oil industry, mining, power plants, warehouses, military activity, transportation, incidents.

In Croatia, in the period from 2006 to 2008, a project entitled *Development of the Croatian Soil Monitoring Program with pilot project (LIFE05 TCY / CRO / 000105)* was carried out by the *Croatian Environment Agency* and *Faculty of Agriculture, University of Zagreb*. The project resulted in the *Program of Permanent Monitoring of Polluted Soils in Croatia (AZO, 2008)* which predicted polluted areas permanent soil monitoring for the sites selected according to the site activity. The program stated the need for the adoption of the *Regulation on the permanent monitoring of the Croatian soil*, but so far only the *Ordinance on the methodology for monitoring the state of agricultural land (OG 43/14)* has been in force.

Croatian water quality standards are defined by the *Regulation on Water Quality Standards (OG 73/13, 151/14, 78/15, 61/16)*, while the quality of waste water is given through the *Ordinance on Limits of Waste Water Emissions (OG 80/13, 43/14, 27/15, 3/16)*.

1.2. Hydrocarbon contamination

According to the *European Environment Agency (EEA)*, the EU has identified about 2.5 million sites potentially contaminated by various activities, such as mineral resources exploitation, electricity generation, waste management, transportation, military activity etc. Remediation necessity has been assigned for 14% of identified locations (<https://www.eea.europa.eu/highlights/soil-contamination-widespread-in-europe>).

Hydrocarbon contamination can be observed as Petroleum Hydrocarbons (PHC) and Total Petroleum Hydrocarbons (TPH). While the PHCs originate from crude oil, the TPHs refer to all hydrocarbons in the soil that can originate from crude oil and all derivatives.

Soil contamination with crude oil depends on crude oil composition. Crude oil consists of various hydrocarbons: saturated hydrocarbons, including alkanes (paraffins) and cycloalkanes (cycloparaffins or naphthenes) and aromatic hydrocarbons. One-ring aromatic compounds are referred to as BTEX (benzene, toluene, ethylbenzene and xylene). Alkanes are present at high concentrations, while naphthenes and aromatics are less represented. In the composition of the oil there are also non-hydrogen components: sulphur compounds (hydrogen sulphide, mercaptan (thiols), sulphides, disulphides, and thiophene), nitrogen and oxygen compounds, as well as heavy metals (V, Ni, Fe, Mo, Cu, Si, Al, Zn) (Wang et al., 2017).

Hydrocarbons penetrate rapidly into the ground, forming an impermeable film on the surface that prevents the flow of water and the normal exchange of gases with the atmosphere or with the gaseous phase of the soil, leading to the drying of the plant roots. By increasing the hydrocarbon content in the soil, the microbiological property is changed. Increased number of anaerobic bacteria leads to soil redox potential decreasing and gradual reduction of certain compounds (iron, manganese, sulphur, etc.), and imbalance in C:N ratio causing plant nutrition disturbance (Kisić et al 2009).

However, substances accumulated in the soil are exposed to biodegradation, transformation and synthesis into new compounds (finally to CO₂ and water). In this way all organic pollutants, such as *Polycyclic aromatic hydrocarbons* (PAHs), pesticides and petrochemicals can be eliminated (Bašić, 2015). Biodegradation in soil depends on different factors, such as: lack of nutrients (lack of C, N, P, K, S and trace elements can decrease pollutant degradation), aerobic/anaerobic conditions (for most of pollutants aerobic conditions are required), water content (certain moisture level is needed for biodegradation), soil texture and structure, pH (acidic conditions are not suitable for most bacteria), temperature, other toxic compounds (their presence can inhibit the activity of the microorganisms), and solubility (soluble pollutant is available to the microflora) (Bobić, 2005).

It is indisputable that the soil plays important role of pollutants collector, but if their presence reach certain level, soil becomes pollution emission source, threatening water and all types of ecosystems.

2. Investigation on site pollution

Underground pollution investigation was done by authorized company during the years 2014, 2015 and 2016. Such investigation is usually conducted through information gathering, data analysis, and reporting. Preliminary research is the first phase aiming at determination of pollution risk by identifying potential sources of contamination and environmental receptors (soil, groundwater). The second phase refers to detailed investigation performed in case that preliminary investigation indicated some contamination. It includes sampling and analysis of soil and groundwater. To get a clear picture of pollution level it is important to know the history of site using, as well as topographic, geological and hydrogeological conditions.

The selected site, which can serve as an example of potential pollution investigation, is used for the temporary disposal and regeneration of technological fluids generated in technological processes such as drilling, overhaul activities, oil and gas production and transportation. The fluid is supplied by tankers and temporarily disposed into facility provided for this purpose. After gravity separation the lighter hydrocarbons are returned to the process (into dehydration), while the residual (dense) phase is submitted to an authorized legal entity or washed with warm water and dehydrating agents, and after passing the solidification process (mixing with sand, lime and various absorbers), as a stable phase, handed over to authorized company.

The facility in a form of pit (5000 m³) is located on a gentle sloping ground. Its northern part is slightly buried, while the southern part is located at the ground level. Around the facility, an impermeable, clayey-dusty embankment is constructed. The bottom of the pit is constructed as a combination of sealing foil and gravel layer, thus achieving isolation from the environment. The soil is composed of low permeable (10⁻⁷ - 10⁻⁸ m/s) dusty-clayey-sandy material, with vertical and lateral variation of individual shares in composition.

Site investigation, carried out in 2014, included ground penetration at 6 locations surrounding the facility (P1, P-2, P-3, P-4, P-5 and P-6 wells, well depth was 10 m, drill hole diameter was 125 mm), and installation of permanent piezometers for monitoring purposes (at P-1, P -3, P-5 and P-6). Soil sampling was done by taking 1 sample per meter of drilling. In case of observed contamination it was necessary to take another sample on the same meter. Groundwater samples were taken out when entering the groundwater layer. Laboratory analyses, which included the parameters, as shown in **Table 1**, were done by accredited laboratory.

Table 1: Soil and groundwater analysis parameters

Soil analysis	Groundwater analysis
<ul style="list-style-type: none"> • TPH • PHC • Total BTEX • Individual BTEX • Heavy metals (Pb, Hg, Cd, Cr, Ni, Zn, Cu) 	<ul style="list-style-type: none"> • pH • Colour, smell • Biochemical Oxygen Demand (BOD) • Chemical Oxygen Demand (COD) • Heavy metals (Cr, Cd, Hg, Pb) • PHC • Total BTEX • Individual BTEX

The results of laboratory analyses were compared with maximum permissible concentrations for individual pollutants (thresholds limits). Since, as stated before, the Croatian national regulation set groundwater contamination criteria only for agricultural land, the guidelines of the Dutch *Ministry of Housing, Spatial Planning and the Environment (Soil Remediation Circular 2009)* were applied. Depending on the spatial use of the soil, the guidelines prescribe the optimum concentrations level (concentration of certain pollutants which has minor environmental impacts) and intervention concentration level (a value indicating the need for rehabilitation).

The *Regulation on Water Quality Standards (OG 73/13, 151/14, 78/15, 61/16)* provides permitted concentrations of certain pollutants (Pb: 10 µg/l, Cd: 5 µg/l, Hg: 1 µg/l). However, the regulation does not provide values for Cr, so the generally accepted "Dutch standards" are also used to determine the level of groundwater contamination with Cr and petroleum hydrocarbon compounds. The analysis of the results included the criteria set out in the *Ordinance on Limits of Wastewater Emissions (OG 80/13, 43/14, 27/15, 3/16)*.

3. Testing results

Laboratory analyses carried out in 2014 showed concentrations of all examined parameters under permissible level, while increased groundwater content of heavy metals (Pb, Cd and Cr) was detected at four piezometers (P-1, P-3, P-5 and P-6). The analyses showed no increases in TPH and PHC concentrations.

The piezometers with increased concentrations of pollutants (P-1, P-3, P-5 and P-6) were included in the monitoring during 2015 and 2016 by testing the concentration of pollutants in groundwater samples during one hydrological year, i.e. considering the high and low water table (**Table 2, Figures 1-4**).

Table 2: Determined concentrations of certain pollutants in groundwater

Piezometers	Year of sampling	Cr	Cd	Hg	Pb	PHC	Benzene	Toluene	Ethylbenzene	Xylene	BTEX
		(µg/l)									
P-1	2015	9.6	<2.0	<0.2	<4.0	5.45	<2.0	<2.0	<2.0	<2.0	<2.0
	2016	7.8	<2.0	<0.2	<4.0	<2.0	<2.0	<2.0	<2.0	<2.0	<2.0
P-3	2015	6.9	<2.0	<0.2	<4.0	15.9	<2.0	<2.0	<2.0	<2.0	<2.0
	2016	<1.0	<2.0	<0.2	<4.0	<2.0	<2.0	<2.0	<2.0	<2.0	<2.0
P-5	2015	5.1	<2.0	<0.2	<4.0	25.2	<2.0	<2.0	<2.0	<2.0	<2.0
	2016	<1.0	<2.0	<0.2	<4.0	<2.0	<2.0	<2.0	<2.0	<2.0	<2.0
P-6	2015	16.0	<2.0	<0.2	4.3	15.6	<2.0	<2.0	<2.0	<2.0	<2.0
	2016	10.2	<2.0	<0.2	<4.0	<2.0	<2.0	<2.0	<2.0	<2.0	<2.0
Optimum concentrations		1.0	5.0*	1.0*	10.0*	50.0	0.2	7.0	4.0	0.2	<2.0
Intervention concentration		30				600.0	30.0	1000.0	150.0	70.0	

* According to Regulation on Water Quality Standards

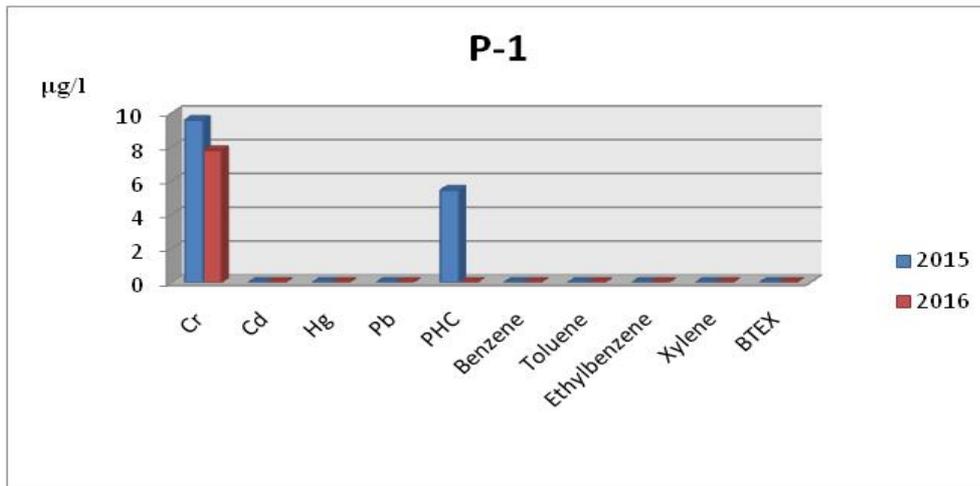


Figure 1: Concentration of groundwater pollutants measured at P-1

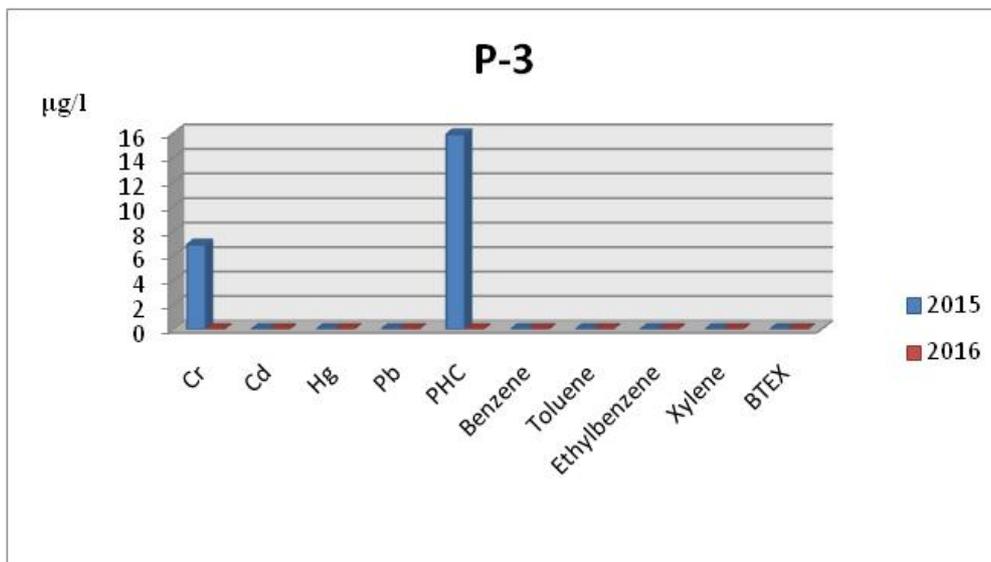


Figure 2: Concentration of groundwater pollutants measured at P-3

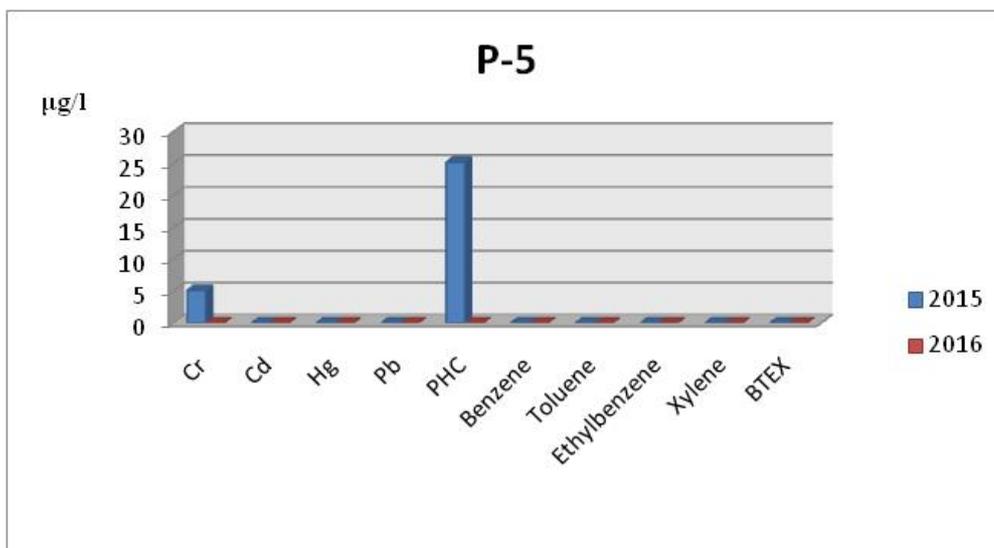


Figure 3: Concentration of groundwater pollutants measured at P-5

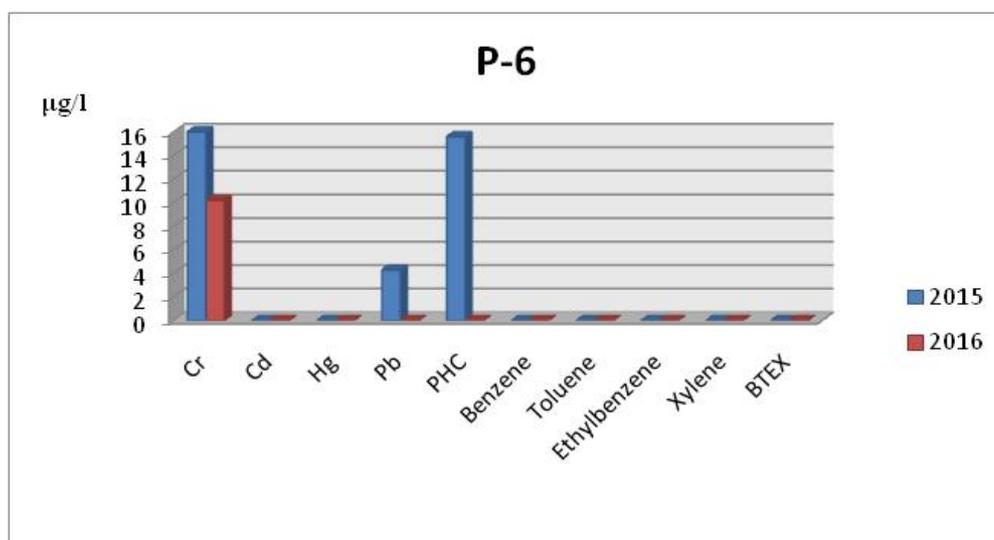


Figure 4: Concentration of groundwater pollutants measured at P-6

The laboratory analyses show an increase in Cr concentrations in 2015 at all piezometers, while in 2016 the increase in chromium concentrations was detected only at P-1 and P-6. However, determined concentrations are in the range between the optimal and the intervention values. The presence of increased hydrocarbon concentration is not recorded during the monitoring in 2015 and 2016.

5. Site maps of pollutants distribution using nearest neighbour technique

In order to show the distribution of pollutants in the underground, the maps were created using the Nearest Neighbours method. It is about a very simple mathematical method of estimation constant value within a polygon, which is recommended in case of a small number of data. If some data is missing, the gaps can be supplemented by this method in a relatively effective manner. The whole field is covered with a series of polygons. For the entire surface a value is assigned which is equal to the point value at the polygon centre. Although the map obtained by this method is not highly reliable, it provides an approximate distribution of the parameters, and based on it, one can conclude where to expand measurements network. The distribution maps of Cr concentrations measured at site in the groundwater in 2015 and 2016 were made using the Nearest Neighbour technique (Figure 5).

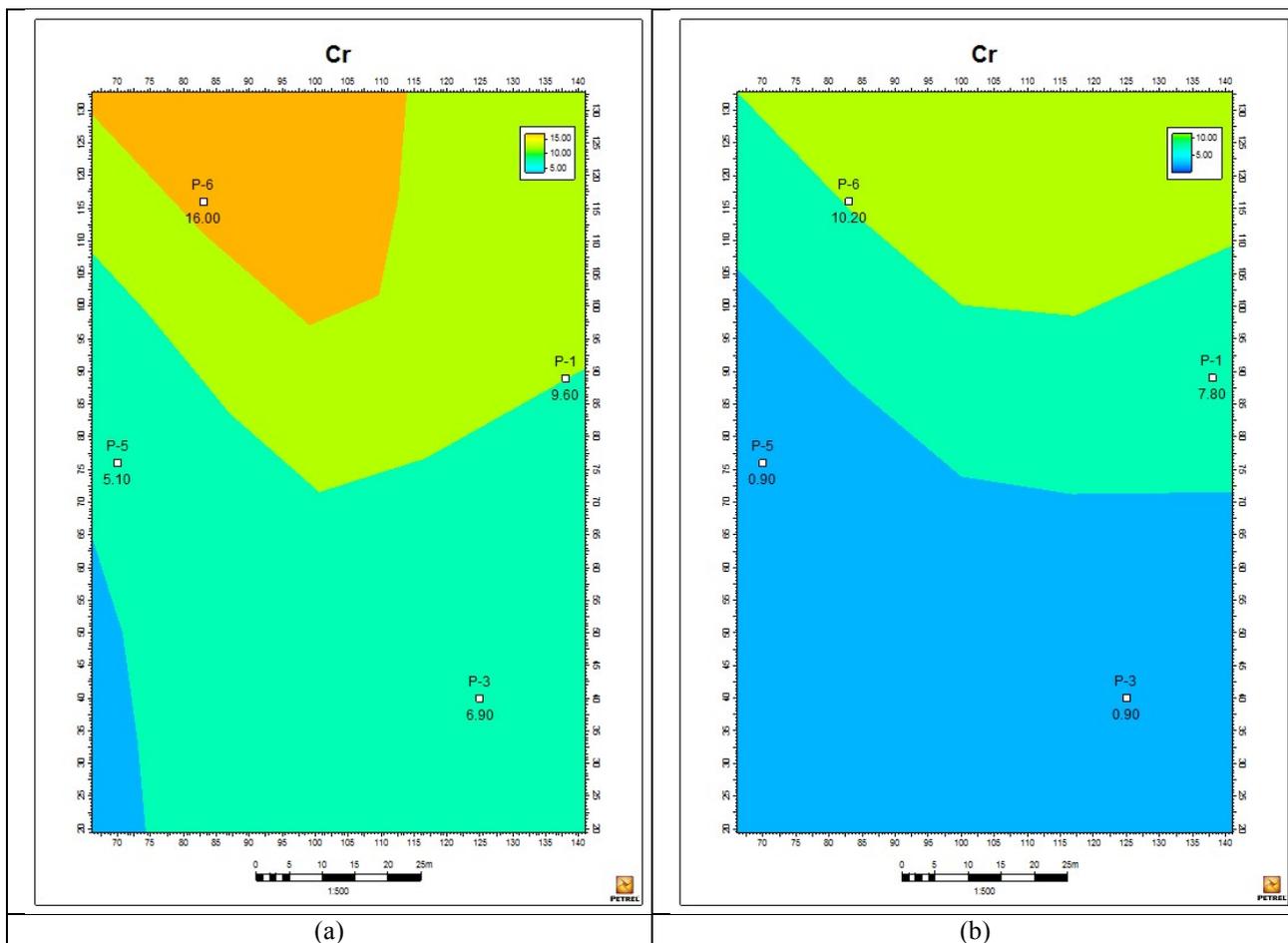


Figure 5: Site map of distribution of Cr in groundwater obtained by Nearest Neighbours technique. According to concentrations measured in 2015 (a) and According to concentrations measured in 2016 (b)

The maps indicate a situation where the highest measured values appear in the northern part of the facility. Taking into account the concentrations measured in 2015, it is evident that the concentration of pollutant in the entire area is decreased in 2016.

5. Conclusion

Based on the conducted research done by authorized company the site is characterized as a location of low pollution risk since:

- the research did not show any increased concentrations of pollutants in soil layers;
- determined presence of increased concentration of Cr in groundwater is below the intervention values;
- the characteristics of the ground soil, which consists of low permeable dusty clayey to dusty sandy material, as well as hydrogeological characteristics of the site and the low mobility of heavy metals, reduce the possibility of significant pollutant spreading.

The investigated site was chosen as a potentially contaminated due to ongoing activities of oil and gas exploitation. Taking into consideration that the research did not show any presence of higher concentrations of petroleum hydrocarbons in the soil and groundwater, it is possible to assume that the registered increased concentrations of Cr are not the consequence of technological fluid regeneration. Since the site is still active, it is recommended to monitor the groundwater quality, including petroleum hydrocarbons content, in the future.

Since the site does not have contamination baseline state measured before start of operation, by additional investigation soil and groundwater contamination status in the immediate vicinity of the facility could be determined.

Collecting data on incidents that might be associated with the high content of pollutants in the groundwater would help also in obtaining a clear picture of the state.

The scope of the investigation work was not sufficient to determine groundwater contamination borders and contamination sources, therefore, performed investigation can be marked as a preliminary investigation phase. The same could be obtained by additional research which would include extended sampling network (detailed investigation). Due to the lack of data pollution mapping was limited to simple mapping technique (nearest neighbour technique). In case of more data, some advanced and more reliable geostatistical method (e.g. Kriging) could be used.

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Extended abstract in Croatian

Utvrđivanje stanja onečišćenosti tla i podzemne vode nastalog kao posljedica aktivnosti eksploatacije ugljikovodika

U svrhu stvaranja osnove za sustavno praćenje stanja okoliša i eventualnih postupaka sanacije, na lokacijama potencijalno onečišćenim različitim gospodarskim aktivnostima, potrebno je provesti kvalitativno i kvantitativno određivanje stanja onečišćenosti tla i podzemne vode.

Prema Europskoj agenciji za zaštitu okoliša (*European Environment Agency*, EEA) u Europi je identificirano čak 2,5 milijuna lokacija potencijalno onečišćenih uslijed obavljanja različitih djelatnosti, kao što su eksploatacija mineralnih sirovina, proizvodnja električne energije, gospodarenje otpadom, transport, vojna djelatnost i dr. Za 14 % identificiranih lokacija pretpostavilo se o potrebi provedbe remedijacije (<https://www.eea.europa.eu/highlights/soil-contamination-widespread-in-europe>).

Premda Europska unija još uvijek nije regulirala područje zaštite tla zasebnom direktivom, zahtjevi za zaštitom ove okolišne sastavnice proizlaze iz drugih regulativa, prije svega *Direktive 2004/35/EZ o odgovornosti za okoliš u pogledu sprječavanja i otklanjanja štete na okolišu* i *Direktive 2010/75/EU o industrijskim emisijama*. Direktivama su jasno definirane djelatnosti koje mogu uzrokovati štetu u okolišu i koje stoga imaju odgovornost u smislu sprječavanja nastanka štete i sanacije okoliša kojemu je nanosena šteta.

Ispitivanje onečišćenosti podzemlja provodi se kroz skupljanje informacija, analizu podataka i sastavljanje izvješća. Preliminarna istraživanja predstavljaju prvu fazu istraživanja, a odnose se na određivanje rizika onečišćenja identifikacijom potencijalnih izvora onečišćenja i okolišnih receptora (tlo, podzemna voda). Drugu fazu ispitivanja čini detaljno ispitivanje onečišćenja lokacije, koje se provodi u slučaju da se preliminarnim istraživanjima utvrdi prisutnost onečišćenja, a obuhvaća uzorkovanje i analizu tla i podzemne vode. Za dobivanje jasne slike stanja lokacije potrebno je poznavanje povijesti korištenja lokacije te topografskih, geoloških i hidrogeoloških uvjeta.

Lokacija koja je uzeta za primjer istraživanja potencijalne onečišćenosti koristi se za potrebe privremenog odlaganja i regeneracije tekućih tehnoloških fluida nastalih u tehnološkom procesu bušenja, remonta, pridobivanja i transporta nafte i plina te drugog materijala nastalog u izvanrednim procesima pridobivanja nafte i plina na eksploatacijskim poljima.

S obzirom da nacionalna regulativa propisuje jedino metodologiju za praćenje stanja poljoprivrednog zemljišta, analiza onečišćenosti podzemlja, opisana u ovom radu, predstavlja primjer procjene rizika onečišćenosti na lokacijama koje ne zadovoljavaju standarde kakvoće poljoprivrednog zemljišta. Što se tiče definiranja onečišćenosti podzemne vode, pravni okvir predstavljaju *Uredba o standardu kakvoće voda (NN 73/13, 151/14, 78/15, 61/16)* i *Pravilnik o graničnim vrijednostima emisija otpadnih voda (NN 80/13, 43/14, 27/15, 3/16)*. Sijedom navedenog, kod definiranja statusa onečišćenosti podzemlja korištena je postojeća nacionalna regulativa, ali i općeprihvaćeni nizozemski standardi u slučaju kada granične vrijednosti nisu definirane nacionalnom regulativom.

Na temelju zaključaka terenskih istraživanja obavljenih od strane ovlaštene kompanije, koja su uključila uzorkovanje i analizu tla i vode, a u svrhu dobivanja jasnije slike stanja pedosfere na lokaciji, u okviru ovog rada izrađene su karte prostorne razdiobe onečišćujućih tvari korištenjem metode najbližeg susjedstva.

Ključne riječi: onečišćenje tla i podzemne vode, eksploatacija ugljikovodika, metoda najbližeg susjedstva.

Authors contribution

Karolina Novak Mavar (PhD, Assistant Professor, Oil Engineering) provided major part of theoretical background, provided distribution maps and conclusions. **Ivana Kapustić Pavić** (Postdoctoral researcher, HSE specialist) provided the charts and some parts of theoretical background.