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ECOMONITORING STUDIES OF GROUNDWATER AND SOIL FOR POLLUTION WITH OIL AND PETROLEUM PRODUCTS

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Abstract

The results of ecological monitoring studies of groundwater and soils in the region of the city of Plovdiv, East Aegean Basin, and Ecoregion 7 Eastern Balkans are presented. The studies were necessary as a result of the repair activities carried out on a damaged oil pipeline. The results obtained from the physicochemical monitoring of waters and soils are discussed (electrical conductivity, odour, turbidity, pH, colour, anthracene, aromatic hydrocarbons, acenaphthene, acenaphthylene, benzene, benz(a)pyrene, naphthalene, petroleum pyrene, polycyclic petroleum products, fluorene, fluoranthene, phenanthrene, chrysene), with the application of the existed standards. The exceedances of the monitored indicators of water quality for irrigation of crops, for the adjacent sources of surface water, and these oils are assessed. An ecological assessment of the studied groundwater and soils is presented. The health risks from using anthropogenically influenced groundwater and soils for plants, animals, and humans have been assessed. Measures to improve the ecological state have been identified.

Key words*: chemical state, ecological assessment, groundwater, health risk assessment, pollutions, soils.*

INTRODUCTION

Oil, or petroleum, is a mixture of gaseous, liquid and solid hydrocarbons of varying densities. Petroleum products are derived from crude oil (petroleum) (Walther & Otto, 2005). Oil spills result from the release of oil from tankers, oil platforms, drilling rigs and oil wells, as well as from refined petroleum products (gasoline, diesel) and their by-products (fuel oil). Spilled oil can affect animals and plants in two ways: directly through the oil spills and through the response to the clean-up process (Bautista & Rahman, 2016b; Sarbatly, 2016). There is no clear relationship between the amount of oil in the aquatic environment and the potential impact on biodiversity. A smaller spill in the wrong place or at the wrong season in a sensitive environment can be much more devastating than a larger spill at a different time of year, even in the same environment (Bautista & Rahman, 2016a). Animals can be poisoned or die from oil entering their lungs. Oil spills can degrade air quality (Middlebrook et al., 2011). The constituents of crude oil are mainly hydrocarbons, which contain toxic chemicals such as benzene, toluene, and polycyclic aromatic hydrocarbons (Tidwell et al., 2015). These chemicals have adverse effects when they enter the human body. They can be oxidized in the atmosphere and form fine particles (Li et al., 2013). Clean up and restoration also generates air pollutants, such as nitrogen oxides and ozone (Ehrenhauser et al., 2014; Nance et al., 2016). The impact of oil and oil products on aquatic systems can be expressed in the following directions: some substances dissolved in water are toxic, and their effect on organisms can be lethal; another part accumulates in the tissues and disrupts the physiological activity of organisms; leads to a change in the species composition of aquatic

communities because organisms adapt to oil pollution differently (Velev, 2015).

The study aims to present ecomonitoring results for contamination of anthracene, aromatic hydrocarbons, acenaphthene, acenaphthylene, benzene, benzo(α)pyrene, naphthalene, petroleum pyrene, polycyclic petroleum products, fluorene, fluoranthene, phenanthrene, chrysene, as well as the indicators electrical conductivity, odour, turbidity, pH, colour in groundwater and soil samples from the vicinity of the city of Plovdiv, Bulgaria, provoked by repair activities of a damaged oil pipeline.

MATERIALS AND METHODS

Twenty-three groundwater samples and four soil samples were tested near a damaged oil pipeline (Figure 1) during 2024. Groundwater samples were taken from a 9-10 m depth according to standard BSS ISO 5667-11:2011. Soil samples were taken from two depths: 0-10 cm and 10-20 cm, according to standard BSS ISO 18400-203:2020.

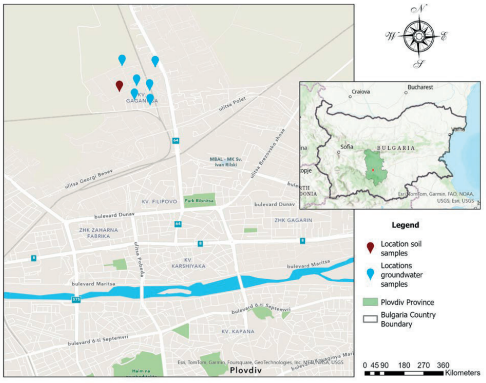

Figure 1. Spatial sampling locations

Chemical analyses were carried out in an accredited laboratory using approved standards: active reaction (pH) - BSS EN ISO 10523:2012; dissolved oxygen (mg/l) - BSS EN ISO 5814:2012; temperature (^{0}C) - BSS 17.1.4.01:1977; electrical conductivity (μS/cm) - BSS EN 27888:2000; petroleum products (μ/l) - BSS EN ISO 9377-2:2004; benzene (μ/l) - BSS EN ISO 15680:2004; benzo(α)pyrene, naphthalene, petroleum pyrene, polycyclic petroleum products, fluorene, fluoranthene, phenanthrene, chrysene - Internal laboratory methodology (ILM 1016/2010). Soil samples were tested for petroleum products according to BSS EN ISO 16703:2011. The results are statistically processed using MS Excel (Microsoft 2010).

RESULTS AND DISCUSSIONS

Chemical analyses were performed to determine the benzene, benzo(α)pyrene, petroleum products, anthracene, acenaphthene, acenaphthylene, naphthalene, pyrene, phenanthrene, fluoranthene, fluorene, chrysene content in the samples of groundwaters (Table 1).

Table 1. Contamination of the samples of groundwater

Pollutants	Mean ± SD (μg/l)
Benzene	79.17 ± 25.38
Benzo(α)pyrene	78.50 ± 6.36
Petroleum products	653.00 ± 386.35
Polycyclic aromatic hydrocarbons:	
Anthracene	0.22 ± 0.06
Acenaphthene	0.80 ± 0.44
Acenaphthylene	0.18 ± 0.07
Naphthalene	25.82 ± 22.24
Pyrene	0.19 ± 0.12
Phenanthrene	1.50 ± 1.73
Fluoranthene	0.17 ± 0.03
Fluorene	5.2 ± 1.19
Chrysene	0.11 ± 0.01

The ecological assessment of the impact of groundwater contaminated with oil and petroleum products on the health of humans, animals and plants is based on the provisions of the Water Directive (Directive 2000/60/EC), the Groundwater Directive (Directive 2006/118/EC), the Environmental Protection Act (EPA), the Water Law, the Health Law. The analysis of the results obtained from the study is based on specific standards regulated by Regulation No.1 of 10 October 2007 on the study, use and protection of groundwater; Regulation No.H-4 of 14 September 2012 on the characterization of surface waters; Regulation No.9 of 16 March 2001 on the quality of water intended for drinking and domestic purposes; Regulation No.8 of 27 May 2009 on the quality of waters for irrigation of crops; Regulation on environmental quality standards for priority substances and certain other pollutants (2010), etc.

Twenty-three groundwater samples were tested by Appendix No.1 of Regulation No.1 (2007)

for the study, use and protection of groundwater. The standards approved under the ordinance are: Benzene – 1.0 µg/l; Benzo(a)pyrene – 0.01 µg/l; Petroleum products – 50 µg/l; Polycyclic aromatic hydrocarbons – 0.10 µg/l.

According to Appendix No.1 to Regulation No.1 (2007) for the study, use and protection of groundwater, polycyclic hydrocarbons are defined as the sum of benzo(b) fluoranthene, benzo(k) fluoranthene, benzo(ghi)perylene and indeno(1,2,3-cd) pyrene, naphthalene, acenaphthene, fluorene, phenanthrene, acenaphthalene, anthracene, pyrene, fluoranthene, and chrysene are polycyclic aromatic hydrocarbons. Measurements on groundwater samples show contamination with petroleum products and related pollutants such as benzene and benzo(a)pyrene, representatives of polycyclic aromatic hydrocarbons.

The highest exceedances compared to the applicable standards were found for benzo(b)pyrene (7400-8300 times), followed by those for benzene (1.9-98 times) and petroleum products (1.7-19.9 times) (Table 2).

Table 2. Exceedance of standards under regulatory documents

Pollutants	Exceedances, Mean ± SD				
	1	2	3	4	5
Benzene	59.94 ± 41.56	55.35 ± 42.66	-	5.83 ± 10.65	1.78 ± 0.18
Benz(a)pyrene	7850 ± 636.39	7850 ± 636.39	-	289.21 ± 21.50	
Petroleum products	12.95 ± 7.63		1.12 - 3.32		
Polycyclic petroleum products				Anthracene 2.15 ± 0.64	

Legend: 1 - Regulation No.1(2007); 2 - Regulation No.9(2001); 3 - Regulation No.18(2009); 4 - Regulation (2010); 5 - Regulation H-4(2012).

The Health Law (2004) and Regulation No.9 (2001) on the quality of water intended for drinking and domestic purposes, promulgated to protect human health from the adverse effects of drinking water pollution, regulate requirements for the quality and safety of drinking water, as well as for improving access to water intended for drinking and domestic purposes. According to Regulation No.9, the maximum values for chemical water quality indicators for drinking and domestic water supply are (µg/l): benzene – 1.0; benzo(a)pyrene – 0.010; polycyclic aromatic hydrocarbons – 0.10 (as the sum of the concentrations of: benzo(b)fluoranthene; benzo(k)fluoranthene; benzo(ghi)perylene; indeno(1,2,3-cd)pyrene). Indicators with indicative significance are: pH ≥6.5≤9.5; Electrical conductivity – 2,000 µS.cm^{-1}; Odour – without significant fluctuations; Turbidity – without significant fluctuations; Colour – without significant fluctuations. The exceedances of the indicators from Regulation No. 9 are as follows: pH – 0.99 times; benzene – from 1.9 to 98 times; benzo(b)pyrene – from 7,400 to 97,400 times. A smell of petroleum products and a visible film were detected (Table 2). Based on these findings, the withdrawal of groundwater for drinking and domestic use is not permitted, regardless of whether wells are constructed for private or industrial purposes, as the studied groundwater poses a risk to human health if used for drinking or other domestic applications. At the same time, no pollutants were detected in the samples from the water supply network for drinking and domestic water supply.

Exceedances of the indicators in Regulation No.18 (2009) on the quality of water for irrigation of crops were established for petroleum products at the MPC of 0.3 mg/dm^3 (= 300 µg/l): from 1.12-3.32 times. Therefore, using water for irrigation of crops from the tested groundwater is unacceptable because it poses a risk to human health, cultivated crops, and animals. The Water Law carries out monitoring of irrigation water, Regulation No.5 (2007) on water monitoring, and Regulation No.18 (2009) on the quality of water for irrigation of crops.

The Regulation on Environmental Quality Standards for Priority Substances and Certain Other Pollutants establishes the environmental quality standards (EQS) for priority substances and certain other pollutants to achieve good chemical status of surface waters by the provisions and objectives of Chapter Ten, Section III of the Water Law (WL). According

to this regulation, the MPC (μg/l) in surface waters for some of the pollutants identified in groundwater from the studied sites are: benz(a)pyrene – 0.27; anthracene – 0.1; naphthalene – 130; fluoranthene – 0.12; benzene – 50. The tests carried out on groundwater samples showed excesses of benzene (from 1.5-30 times), benzo(α)pyrene (from 274-304.41 times), anthracene (from 1.7-2.6 times), and fluoranthene (1.42 times). Therefore, using tested groundwater is unacceptable because it poses a risk to humans, plants, and animals.

Regulation No.H-4 (2012) regulates the procedure and manner for the characterization, classification and presentation of the ecological status/potential of surface water bodies. According to this regulation, the SCC/MPC (μg/l) in surface waters, for comparison with some of the established pollutants in groundwater from the studied sites, are: acenaphthene – 3.8 SCC; 50 MAC; acenaphthylene – 0.64 SCC; fluorene – 3.1 SCC; phenanthrene – 1.3 SCC; pyrene – 0.012 SCC; chrysene – 0.02 SCC; benzene – 10 SCC; 50 MPC; Oil and oil products – without visible film on the surface and without odour. Exceedances of the standards set by Regulation H-4 were found in the tested groundwater samples for benzene (1.5-1.96 times), anthracene (1.7-2.6 times), and chrysene (5.5 times). The established exceedances and changes in the condition and quality of the analysed groundwater (smell of oil products; visible film) make the water abstraction and use of the tested groundwater inadmissible because they pose a risk to humans, animals and plants.

According to Regulation No.4 (2000) on the quality of waters for fish farming and shellfish breeding, the presence of oil products that form a film on the surface of the water or form deposits on the beds of river streams and lakes is not allowed, because they give a petroleum taste to fish and fish products and they have a harmful effect on fish. The deteriorated quality of groundwater affects the quality of surface waters when used to connect with surface waters. The tested soil samples did not show excesses in the oil products indicator. The reported amount is less than the detection limit according to the approved standard (below 15 $\mu g.kg^{-1}$ soil) and the requirements of Regulation No.3 (2008). Deterioration in the condition of terrestrial ecosystems is expected due to water abstraction and use of the studied groundwater, which is unacceptable because it poses a risk to the health of people, animals and plants.

The increase in oil production and its processing, its transportation through pipelines, accidental spills during its transportation, poor storage and manipulation lead to a number of environmental problems. Hydrocarbon pollution of the environment is a huge catastrophe for ecosystems because it leads to lethality and damage to flora and fauna for a long period. Oil and its derivatives penetrating the soil specifically impact the entire environment, the former communities and ecosystems. It has been established that the main reason for the delay in the development or death of plants is due to the deterioration of the water-physical properties of the soil, the blocking of nutrients (Crummer, 1964), the occurrence of oxygen starvation (Odu, 1972) and a complete disruption of biochemical processes (Gaydarova et al., 1990). The extracted groundwater contains oil and many salts (80-90%). Soil contamination occurs most often when corroded pipelines break, resulting oil and salt pollution. Drilling wells are located on vast areas, usually agricultural lands and pastures. The most intensive pollution is about 2.5 km from petrochemical and oil refining enterprises. With a distance of 6-10 km from the source, the content of pollutants decreases, but the range of soil pollution is not less than 20 km (Suleĭmanov et al., 1996).

Crop contamination occurs when groundwater containing petroleum products is used for irrigation. Soil contamination with petroleum products is not limited to the surface horizon. Infiltration is directly dependent on the current humidity. This rate decreases by 6 to 14 times at higher humidity compared to dry soil (Dimitrov et al., 1995). The properties of soils contaminated with petroleum and petroleum products change. Petroleum products are a permanent component of organic environmental pollution. Their presence in the soil significantly disrupts its water-physical, agrochemical and microbiological properties. The changes resulting from pollution with petroleum products are mainly associated with

the disruption of the water-air regime due to filling the pore spaces with petroleum products and the aggregation of soil particles. As a result, the oxidative conditions dominant before the contamination in the soil change to oxidative-reductive and reduction. The emergence of anaerobic conditions leads to the suppression of nitrification and the enhancement of ammonification. An accumulation of ammonia nitrogen of 20-40 $mg \cdot kg^{-1}$ occurs, and the content of nitrate nitrogen remains in traces. The amount of mobile phosphorus decreases (from 2 to 9 times), which is explained by the immobilization of inorganic phosphorus by microorganisms and phosphine formation. With very strong salinization, regardless of the pollution with petroleum products, the content of mobile phosphates can increase. Mineralization increases, and sulphate, chloride, calcium, magnesium, sodium, and potassium ions appear. The amount of trace elements increases - Mn (up to 4 times), Co, Mo, Cu, Zn, Pb, Ni (up to 2 times), the oxidation-reduction potential decreases (up to -340 mW) (Haziev et al., 1987; Gabbasova et al., 1997).

Oil and oil products have a toxic effect on soil microorganisms. Fungi are relatively resistant to pollution. Their resistance to higher concentrations of oil products and the increase in saprophytic fungi is related to the fact that the pH in some of the studied contaminated soils is in the acidic range, favouring fungi development. Fungi use the hydrocarbons as a source of "carbon" nutrition, and thanks to the high enzymatic activity, they decompose oil into low-molecular compounds quite quickly. Of the algae, the most resistant to oil pollution is blue-green algae. When components of oil pollutants penetrate the aquifer, specific microorganisms settle which are difficult to be purified. The number of one of the most important participants in soil-forming processes - the earthworm, decreases almost twice near the source of pollution (Molodova, 1978). Oil and petroleum products have a detrimental effect on the metabolism of plants, with some lighter fractions entering them directly (Markov, 2009).

CONCLUSIONS

The studied groundwater is significantly contaminated with petroleum products. Environmental monitoring studies show excess petroleum products and/or related indicators such as benzene, benzo(α)pyrene, and representatives of polycyclic aromatic hydrocarbons. According to environmental legislation, the excesses range from several times to tens and thousands of times as deviations from the standards. On this basis, water withdrawal and water use from the studied groundwater for personal or industrial purposes, drinking and domestic needs, water use for irrigation of crops and other needs is inadmissible because it poses a risk to people, plants and animals until their decontamination.

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