

ORGANIC POLLUTANTS IN GROUNDWATER
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Abstract: Oil derivatives are commonly used and they play a key role in the economy. They are used in many industries. Such big amounts of oil derivatives products generate vast quantity of pollution. Those pollutants can get into the ground and water beyond any control during catastrophes or due to inadequately managed waste and storage. The aim of the paper was to determine the level of oil derivatives pollution in the groundwater on the area of a former airbase, where between 1950 and 1990 the Soviet Army stationed. Analysis was carried out on groundwater samples from three piezometers placed on the area of the former airbase. In the samples some parameters were determined, i.e. temperature, reaction, electrolytic conductivity, the depth of groundwater surface, the content of aliphatic hydrocarbons, monoaromatic and polycyclic aromatic hydrocarbons. Determined amount of dissolved hydrocarbons was large what proves unsatisfactory effectiveness of previous rehabilitation processes.

INTRODUCTION

Just a few hundred years ago nearly whole energy used by human beings came from renewable sources, mainly from biomass. More and more commonly used petroleum and its products as fuel to internal-combustion engines caused increasing threat of polluting environment by oil derivatives. Groundwater and ground pollution is not as spectacular as surface water pollution and it appears mostly locally [20]. It mostly appears on petrol stations, railroad sidings and petrol handling stations as well as during accidental collisions causing not only emission of VOCs (mainly benzene, toluene, ethylbenzene, m+p-xylene, and 1,2,3-trimethylbenzene) to the air but also penetration of oil derivatives into the ground [22]. These pollutions result from human business activity and degrade the environment as well as negatively influence life forms, including people. Catastrophes in overland and sea transportation are especially dangerous because they cause considerable pollution on small and bigger areas having negative influence not only on ecosystems but on economy far beyond local scale [11]. Of distinct characteristics are permanent pollutions in the case of which no rescue operations and consequently rehabilitation processes to restore original state of the environment are carried out [20]. The example of such long-term processes are places, where oil derivatives pollutants get into the environment without any control. The areas of airbases, especially those used by units of the Soviet Army

are such examples. Essential activities necessary to restore environment to its original state include pumping out organic phase floating on the water surface known as Light Non-Aqueous Phase Liquid (LNAPL) using automated skimmers. Common methods of eliminating compounds diffused in water phase are microbiological methods, injecting oxygen or rinsing out polluted soil by detergents [3, 8, 9, 10, 16, 21].

MATERIALS AND METHODS

Characteristics of the research scope

The airbase, whose area was examined, is a place that was taken over by the armed forces of the Soviet Union after the second world war from German air force. During the post-war time the place was considerably rebuilt and modernised by the units of the Soviet Union Army. In 1991 the civil authorities took over the complex together with its technical development. To the north-east, at a distance of about 1 km from the boundary of the airbase, there is a town of Skarbimierz, while villages Małujowice and Pepice are situated south-west of the airbase. The nearest buildings are placed 0.5 km from its boundaries. Additionally, in Małujowice and Skarbimierz there are potable water intakes.

Taking samples

Samples were collected twice: in the autumn (21.11.2009) and in the spring (22.04.2010). The research included groundwater from three piezometers located in the southern part of the complex (near Pepice village) in direct vicinity of places where planes were refuelled. All piezometers were between bunkers, in which Soviet Union's planes were stationed. In the site of samples collection hydrogeological whistle was used to check the depth of occurring groundwater table, taking into consideration LNAPL layer lying on the surface. Furthermore, LNAPL phase thickness was determined using glass calibrated pipe sampler. Before taking water samples to check physical and chemical properties, the water from the piezometer was pumped out by an electric pump until stable parameters (pH and electrolytic conductivity (EC)) were established. In the taken samples the primary parameters (i.e. temperature, colour, smell, pH, and EC) were determined. Next, water was taken to 1 dm³ dark glass bottles. Such samples were preserved by dichloromethane (DCM) to determine hydrocarbons. At the same time a second series of samples was preserved by hydrochloric acid 1:1 to check the amount of oil derivatives.

Water quality parameters

The samples preserved with DCM underwent liquid-liquid extraction using dichloromethane and careful shaking for 15 minutes. When the water and organic layers were separated, the effluent was taken to volumetric flasks and dried using anhydrous sulphate sodium (Na₂SO₄ analytically pure). The content of aliphatic and monoaromatic hydrocarbons was determined by gas chromatograph VARIAN CP – 3800 with flame ionisation detector. In the research capillary column VF5-ms 30 m long, ID 0.25 mm and if 0.25 μm was used. In the given extracts, the authors determined: aliphatic hydrocarbons with C₈-C₂₂ long chain, monoaromatic (benzene, toluene, o-xylene, m+p-xylene, ethylbenzene, styrene, isopropylbenzene, 1,2,3-trimethylbenzene, tertbutylbenzene and 1,2,4-trimethylbenzene, secbutylbenzene, 4-isopropylbenzene, and n-butylbenzene) and polycyclic aromatic hydrocarbons (16 compounds according to US EPA). The temperatures of heating

zones of the chromatograph for determining monoaromatic hydrocarbons and aliphatic hydrocarbons were: injector – 250°C, detector – 300°C. The furnace temperature program during determining monoaromatic hydrocarbons began at 30°C and it was stabilised for 2 minutes, then it was heated up to 50°C at a rate of 4°C per minute, and subsequently to 150°C at a rate of 5°C per minute. In order to check aliphatic hydrocarbons the initial temperature 30°C was maintained for 2 minutes, then the furnace was heated up to 50°C at a rate of 4°C per minute and then to 280°C at a rate of 6°C per minute. To analyse PAH the injector was heated up to 300°C, while detector FID to 310°C. Temperature programme of the chromatograph began with maintaining the furnace temperature of 100°C for 2 minutes and then heating up the temperature to 300°C at a rate of 4°C per minute and maintaining this temperature at the end of the programme for 10 minutes. During all analyses the flow of carrier gas (He) through the column was 1 cm³ per minute. Total content of soluble organic substance was determined as ether extract by gravimetric method.

RESULTS AND DISCUSSION

The primary source of pollution on the area of the airbase was fuel warehouse along with pipelines, groundwater drainage system and waste stockpile. On the examined area waste was disposed in unsealed hollows in the ground, which after filling-up were covered with a layer of soil. Leaky fuel pipelines and leaky fuel tanks constituted another crucial problem. A long time of using those installations caused degradation of soil and groundwater, especially in the area of aircrafts' refuelling and petrol stations.

The reports presented after research had been carried out by Military University of Technology and State Inspectorate for Environmental Protection show in detail the state of pollution caused by oil derivatives on the airbase. According to their results in the groundwater on the area of the airbase contains, among others, increased concentrations of lead, chrome, copper, zinc, cadmium, arsenic, and oil derivatives compounds [21]. During the field investigation it was found out that groundwater is constantly exposed to oil derivatives which float on the water table in the form of LNAPL with diversified thickness. Important issue is the depth of groundwater table. The experiments performed show that it is shallow water, and, as such, it is exposed in a considerable degree to a direct influence of pollution left on the ground (Tab. 1). The deepest groundwater level was noted near piezometers no. 3. Water there was at the depth of 194.5 cm ugl (underground water level). The shallowest groundwater level was noted in piezometers 1 where its value was 149.2 cm ugl. During the experiment carried out in spring a significant raise in groundwater level was noted. It was, in all probability, caused by a significant thickness of snow cover and the lack of proper maintenance of existing dewatering ditch system in the area. During the spring, the deepest groundwater level was noted in the piezometer no. 2, at the depth of 147 cm ugl, where at the same time the smallest changeability of that parameter in both checked seasons was noted. The shallowest occurring groundwater level was noted in piezometer no. 3, which was 66 cm ugl. Such fluctuation of water surface can increase solubility of organic pollutants in groundwater [14]. All water samples taken are characterised by intensive greenish colour, especially the samples taken from piezometers no. 1 and 2. In piezometer no. 3 the colour was less intensive. Its colour points to the content of organic remains of mainly plant origin. Piezometers, from which the samples were taken, had no covers protecting them from pollution getting inside. Piezometers

no. 1 and 2 were placed in the area surrounded by trees and bushes and that is why inside their inlets there were large amounts of dry leaves. Piezometer no. 3 was surrounded only by annual plants, hence surface pollution of underground water table was negligible.

Table 1. Main parameters of investigated groundwaters

Piezometer	Watertable [cm ugl]		LNAPL phase thickness [mm]		Temperature [°C]		Reaction (pH)		EC [$\mu\text{S}/\text{cm}$]	
	A	S	A	S	A	S	A	S	A	S
1	149.2	77	< 2	2	13.5	9.2	5.97	6.15	689	785
2	173	147	< 2	3	13.5	8.9	5.99	6.15	556	396
3	194.5	66	13	15	13.0	10.0	5.86	6.03	470	406

ugl – underground water level, A – autumn water samples; S – spring water samples

All tested waters were characterised by a slight acid reaction which allows to qualify them to the 1st purity class (Tab. 1). At the same time, temperature fluctuations noted in both seasons (3.0–4.6°C) point to a direct relation between the tested waters and external factors. The odour of samples was described as specific (characteristic for oil derivatives) with insensitivity level 4. LNAPL phase thickness appearing in tested piezometers was not high and it reached a maximum of 13 mm. The content of monoaromatic hydrocarbons in the tested waters was very high (Tab. 2) especially in the case of benzene and toluene.

Table 2. Aromatic hydrocarbons content in investigated samples [$\mu\text{g}/\text{dm}^3$]

Piezometer	1		2		3	
	A	S	A	S	A	S
Autumn/Spring						
Benzene	5790	1799	15757	1958	11164	5944
Toluene	27.25	131.25	56.58	397.33	50.73	286.03
Ethylbenzene	50.59	66.38	25.09	51.10	19.75	97.14
m+p xylene	51.34	67.42	50.99	41.77	9.41	3.76
o-xylene	< 0.01	2.53	7.40	12.78	1.69	2.02
Styrene	5.77	9.74	6.88	610.97	5.07	11.48
Isopropylbenzene	11.50	16.16	35.66	82.07	9.37	10.15
n-propylbenzene	24.21	15.53	37.89	82.06	19.62	40.23
1,3,5-trimethylbenzene	27.87	52.71	47.36	210.89	14.99	37.33
Tertbutylbenzene + 1,2,4-trimethylbenzene	43.23	67.62	64.36	420.14	20.02	20.52
sec-butylbenzene	9.26	7.08	19.62	28.59	6.82	3.79
4isopropylbenzene	86.87	151.26	210.11	10.97	8.12	23.23
n-butylbenzene	30.35	56.40	72.04	194.81	26.82	49.44
Σ	6158.2	2443.1	16391.0	4101.5	11356.4	6529.1

A – autumn water samples; S – spring water samples

Their high concentrations (exceeding respectively 15.7 and 0.39 mg/dm³) can be explained by high solubility of these compounds in water. Comparable and even higher exceeding of allowable concentrations of these compounds was noted in the 1990s on the area of petrol station [1, 2]. Increased amounts (exceeding 100 µg/dm³) were noted as well in the case of styrene, 1,3,5-trimethylbenzene, 4-isopropylbenzene, n-butylbenzene and tert-butylbenzene with 1,2,4-trimethylbenzene. High amounts of compounds belonging to benzotriazoles penetrating to the ground and groundwater cause negative changes in micro-organisms' activity, and biodegradation of those compounds may proceed very slowly. It depends, however, on additives used in some fuel types [5, 6]. Nonetheless, specialised micro-organisms in laboratory conditions can effectively degrade even high concentrations of BTEX exceeding 1.1 g/dm³ [13]. Field investigation show that ethylbenzene, toluene, and o-xylene are the most prone to degradation, however, lack of oxygen can drastically slow down biodegradation of even these compounds [18, 19]. Recorded in this research concentrations of BTEX, especially of benzene, categorize the water tested as unclassified.

Table 3. Aliphatic hydrocarbons content in investigated samples [µg/dm³]

Piezometer	1		2		3	
	A	S	A	S	A	S
Octane	14.02	21.23	22.17	35.84	10.47	29.47
Decane	56.66	31.02	9.51	26.47	4.54	17.45
Dodecane	29.37	22.50	144.3	45.25	132.6	78.61
Tetradecane	18.59	15.12	80.99	15.71	17.06	17.95
Heksadecane	6.64	5.24	24.45	6.13	2.87	5.45
Oktadecane	3.25	2.15	4.35	1.18	2.01	2.62
Eikosane	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
Docosane	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01

A – autumn water samples; S – spring water samples

In the case of leaky fuel tanks, n-alkanes are noted in groundwater. The most often observed are compound ranks from C14 to C34 [4]. Due to significant hydrophobicity of those compounds, they often create emulsion and single compounds concentrations can exceed even 1000 µg/dm³. In soils exposed to oil derivatives over 16 mg/kg single alkanes with total content of hydrocarbons in ranks C6-C35 exceeding 120 g/kg d.m. were determined [7, 17]. Such strong groundwater pollution eliminate them from use in households, what is more, they pose a direct threat to human and animal health [15]. In tested samples high concentrations of aliphatic hydrocarbons with shorter chains were noted (exceeding 100 µg/dm³ in the case of dodecane). There were much less heavier hydrocarbons and no eicosane and docosane were observed.

In the tested groundwater samples high concentrations of polycyclic aromatic hydrocarbons were noted (Tab. 4). Such phenomenon is characteristic for water polluted with organic compounds easily soluble in water (e.g. benzene) acting as a solvent. Taking into consideration recorded concentrations exceeding acceptable PAH standards it can

be noticed that water samples from both autumn and summer tests belong to 5th class of groundwater quality. Such high content of PAH can be a result of original fuel content or leaching of these compounds from the soil during the percolation of aviation fuel to groundwater.

Table 4. Polycyclic aromatic hydrocarbons content in investigated samples [$\mu\text{g}/\text{dm}^3$]

Piezometer	1		2		3	
	A	S	A	S	A	S
Naphthalene	45.9	48.03	37.1	77.02	39.9	65.98
Acenaphthylene	0.54	0.40	0.71	0.66	0.50	0.78
Acenaphthene	0.69	0.65	0.96	0.88	0.87	0.95
Fluorene	1.24	1.10	1.57	1.24	1.70	1.82
Phenanthene	0.36	0.31	0.43	0.20	0.31	0.50
Anthracene	0.13	0.09	0.15	0.13	0.12	0.16
Fluoranthene	0.28	0.45	0.56	0.86	0.25	0.37
Pyrene	0.13	0.16	0.16	0.16	0.16	0.12
Benzo(a)anthracene	0.08	0.12	0.12	0.14	0.04	0.06
Chrysene	0.27	0.40	0.28	0.32	0.34	0.43
Benzo(b)fluoranthene	0.09	0.04	0.09	0.13	0.09	0.08
Benzo(k)fluoranthene	1.63	3.13	1.23	4.38	1.03	1.26
Benzo(a)pyrene	0.39	0.20	0.24	0.38	0.29	0.21
Indeno(1,2,3-cd)pyrene	0.94	0.57	1.80	1.41	1.85	1.29
Dibenzo(a,h)antracen	0.74	0.22	0.95	0.68	0.58	0.64
Benzo(g,h,i)perylene	1.17	0.79	1.61	1.53	0.69	0.63
Σ	54.6	56.7	48.0	90.1	48.7	75.3

A – autumn water samples; S – spring water samples

Studies of other authors also indicate that hydrocarbons content in the ground can be reduced by sowing polluted area with plants, and then performing e.g. mycorrhization which stimulates the growth of flora decomposing hydrocarbons [12, 20]. Similar actions carried out on the tested field could improve effectiveness of removal of LNAPL layer appearing on water table. Groundwater polluted with hydrocarbons migrates to surface freshwater (Peńicki stream) through trench system in the airbase. This can cause a significant expansion of the area affected by such compounds on the environment, what is emphasized by Batlle-Aguilar *et al.* 2009 [1].

CONCLUSIONS

For over forty years of operation of the airbase in Skarbimierz by the soldiers of the Soviet Army there has been significant environmental degradation in the area. Incompe-

tent handling of oil derivatives materials, corrosion and thus the leakage of installations and human negligence led to environmental disaster in the discussed area, the effects of which are still present. Rehabilitation of the area, carried out since 1995, has produced tangible results in a decrease of oil derivatives pollution – thickness of LNAPL phase. However, lack of continuity of these activities resulted in slowing down the process of removing pollutants from the tested area and thus stopping them from spreading. It has been observed that oil derivatives are constantly present in the groundwater in dissolved form or LNAPL phase, creating a patch with thickness up to 13 mm. It still poses a real threat to nearby farmlands.

The studies, in most determinations showed a similar level of pollution with polycyclic aromatic hydrocarbons and monoaromatic hydrocarbons in every tested piezometers on the area of the airbase. Comparison of the results showed only understandable slight variability of parameters between the autumn 2009 and the spring 2010. Taking into consideration the amounts of all tested pollutions and comparing them with boundary values of particular classes of groundwater quality (defined by the minister of environment's order of 23rd July 2008 on the criteria and methods of evaluation of groundwater), water in the area can be qualified to 5th purity class. Potential threat posed by the pollution of the airbase area is the possibility of migration of oil derivatives with the water flow to places from which drinking water is taken for people and animals and to the Pempicki stream whose drainage area covers the polluted land. Main activities in the examined area should be aimed at most effective removal of LNAPL layer from groundwaters, what can improve their quality and the quality of the whole ecosystem.

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ZANIECZYSZCZENIA ORGANICZNE W WODACH GRUNTOWYCH NA TERENIE BYŁEJ BAZY LOTNICZEJ

Produkty ropopochodne są szeroko rozpowszechnione i odgrywają kluczową rolę w gospodarce człowieka. Znajdują one zastosowanie w wielu gałęziach przemysłu. Tak duże ilości produktów ropopochodnych generują znaczne ilości odpadów. Odpady te mogą przedostawać się do środowiska gruntowo-wodnego w sposób niekontrolowany podczas różnego rodzaju katastrof czy niewłaściwie prowadzonej gospodarki odpadami, w tym składowania. Celem badań było określenie stopnia zanieczyszczenia substancjami ropopochodnymi wód podziemnych na terenie byłego lotniska wojskowego, na którym w latach 1950–1990 stacjonowały jednostki Armii Radzieckiej (JAR). Analizie poddano próbki wód podziemnych pochodzących z trzech piezometrów znajdujących się na terenie byłej bazy lotniczej. W pobranych próbkach określono takie parametry jak: temperatura, odczyn, przewodność elektrolityczna właściwa, głębokość zalegania wód gruntowych, zawartość węglowodorów alifatycznych, monoaromatycznych oraz wielopierścieniowych węglowodorów aromatycznych. Oznaczona ilość rozpuszczonych węglowodorów była wysoka, co wskazuje na niezadowalającą efektywność przeprowadzonych wcześniej procesów rekultywacyjnych.