

# Environmental and ecotoxicological risk assessment of pollution with light crude oil for an oil exploitation field

Alexandru Florin Simion <sup>1\*</sup>, Angelica Nicoleta Găman <sup>1</sup>, George Artur Găman <sup>1</sup>, Ionuț Drăgoi <sup>2</sup> and Cătălina Ghiță <sup>2</sup>

<sup>1</sup>National Institute for Research and Development in Mine Safety and Protection to Explosion – INSEMEX Petroșani, 32-34 G-ral Vasile Milea Street, Petroșani 332047, Romania

<sup>2</sup>Institute for Research and Technological Design - ICPT Câmpina - 29 Bvd Culturii Street, Câmpina Town, Code 105600, Prahova, Romania

**Abstract.** Global development of transportation dependent on internal combustion engines, involves major increases in the consumption of fossil fuels obtained by extracting crude oil from depths by means of wells and refining it. In this context, protection of environment and groundwater quality is an increasingly difficult objective to maintain, requiring modern methods to address possible negative effects on the environment. The current research assesses the degree of eco-toxicological and environmental risk for an oil exploitation area in case of an accidental pollution scenario with crude oil. The entire risk spectrum was analysed through a set of qualitative and quantitative risk assessment tools to highlight and quantify the most important effects caused by petroleum products on biotope and biocenoses. The primary aim of the research is to identify vulnerable environmental reservoirs for accidental oil pollution and to establish the best tools to quantify the environmental and ecotoxicological risk of groundwater contamination. The results obtained from the area showed a low to medium risk of contamination of the saturated and unsaturated area with crude oil expressed as TPH (total petroleum hydrocarbons) and a low risk of contamination with volatile organic compounds type BTEX associated contamination of biocenoses that can directly or indirectly interact with potentially polluted areas.

## 1 Introduction

Pollution of soils and groundwater with petroleum products or hydrocarbons from crude oil generates major effects on local flora and fauna and once discharged into soil, they enter the food chain and tend to bioaccumulate. In case of land polluted with crude oil extraction products, on which pollutant removal techniques have been applied (drainage, combustion, decomposition with microorganisms), both land surface and groundwater

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\* Corresponding author: [alexandru.simion@insemex.ro](mailto:alexandru.simion@insemex.ro)

remain unrecoverable for a long time. Ecotoxicity of substances present in waters and soils is one of the major concerns with special implications for the quality of human life and aquatic, terrestrial and riparian terrestrial ecosystems, because many substances from extractive industries have a high potential to affect biotic and abiotic balances [1, 2].

The mechanism of accumulation or bioaccumulation of pollutants in ecosystems of the aquatic environment as well as of ecosystems that interact discontinuously with it, is studied in relation to mechanisms and processes at the interface between ecosystem components [3]. From this point of view, special attention must be paid to the interfaces: sediments - water, water - organisms and water - air. Evaluation of the ecotoxicological risk in the studied area, requires complex investigations directed towards soil, subsoil and groundwater. These investigations may contain information on identifying and characterizing pollutants, determining their concentration in the environment, locating potentially affected areas and defining the distribution and dispersion gradient, determining possible sources and causes of pollution, characterizing the physico-chemical and hydrogeological conditions of the site, environmental vulnerability, etc.

The current article is a part of the results of the scientific research project "Assessment and forecasting of pollution caused by accidental spillage in the oil extraction industry, on soil and groundwater" carried out in collaboration with S.C. OMV Petrom S.A. Upstream Division - Development Engineering Institute for Research and Technological Design (ICPT) Câmpina. Research documentation started from the sources of information held by ICPT Câmpina regarding both history of the area, the monitored pollutant concentrations, followed by modelling of the extent and magnitude of pollutant concentrations in soil and groundwater.

## 2 Materials and methods

Documentation studies of the assessment area, evaluation of impact extension and of the mechanisms of accumulation or bioaccumulation of pollutants in terrestrial and aquatic ecosystems were performed through detailed research on local and regional climate, geology, soil and subsoil, groundwater and surface water resources and terrestrial and aquatic ecosystems. Also, anthropogenic activities that can generate major effects on ecosystems, possible sources of pollution and the impact of pollutants on the environment have been identified.

The environmental and ecotoxicological risk assessment was performed by a complex method for assessing the eco-toxicological risk consist in the extensive development of terms characterising an event (probability and severity), as follows:

- Probability is defined as an amount of pollutant (xenobiotic) with a certain degree of toxicity that can influence a certain environmental component.
- Severity is defined as the ratio between the vulnerability of a species or environmental indicators to a xenobiotic and the ability of the species to adapt (neutralize) to that xenobiotic.

The method used classifies characteristic terms in the form of matrices associated with events, environmental risk and impact on soil and ecosystems. Quantification of environmental and ecotoxicological risk based on the terms of impact was performed using equation 1.

$$R=f(T \times E \times \frac{V}{C}) \quad (1)$$

Where:

R – Eco-toxicological risk;

T – Type of event;

- E – Risk components;
- V – Vulnerability of the identified species in an ecosystem;
- C – Ability of the species to adapt to a particular substance.

In the case of the studied field of exploitation, hazard hierarchy was set aside because in a preliminary stage of the research we established, along with our partners, that the risk assessment methodology should be applied on an accidental oil leakage caused by the damage of a crude oil pipeline.

### 3 Result and discussion

Quantification of groundwater contamination risk in the studied field, based on the scenario of accidental spill of very light oil (instantaneous or for one month length) involves understanding the mechanism of physical adsorption of crude oil, stored in the upper soil horizons and the study of oil dispersion in the area unsaturated to the aquifer.

For the established scenario, the dominant soil profile of area was modelled, dispersions of the compounds of aromatic origin (Benzene, Toluene, Ethyl-benzene and the sum of Xylenes) from very light streams in concentrations of 2000 (scenario 1), 5000 (scenario 2), 10000 (scenario 3) mg/kg dry matter being simulated in the UNsat application.

Quantification of pollutant concentration discharged (determined) in the upper soil horizon in impact units (levels) was performed according to reference values for traces of chemical elements in soils, according to order no. 756 of 03/11/1997 for the approval of the regulation on assessment of environmental pollution with subsequent amendments and completions [4].

Depending on reference values regulated by the national legislation (Romanian), the intervals of impact (Table 1) units were established as follows:

- **Low impact** is represented by pollutant concentrations below the normal values or the alert threshold for sensitive uses;
- **Moderate impact** is represented by pollutant concentrations situated between the alert threshold and the intervention threshold for sensitive uses (alert threshold of less sensitive uses);
- **High impact** is represented by pollutant concentrations in soil, that exceed the intervention threshold for less sensitive uses;

**Table 1.** Reference values of soil pollutants

Indicator	Unit of measurement	Normal values	Sensitive uses		Less sensitive uses	
			Alert threshold	Intervention threshold	Alert threshold	Intervention threshold
Benzene	mg/kg dry unit	<0,01	0,25	0,50	0,50	2
Etyl-benzene		<0,05	5	10	10	50
Toluene		<0,05	15	30	30	100
Xylene		<0,05	7,5	15	15	25

The toxicity potential of aromatic origin compounds (Benzene, Toluene, Ethyl-benzene, Xylene) has been correlated, based on in vitro studies conducted by the United States Environmental Protection Agency (EPA) on animals (rats and mice) and on sensitive and less sensitive groups of humans [5, 6] and quantification of toxicity potential in impact units (levels) was performed by correspondence: low toxicity - low impact, average toxicity - moderate impact, and high toxicity - high impact.

The three simulation scenarios were developed on total quantities of petroleum hydrocarbons from which the BTEX contents were extrapolated, according to the characteristics specified in the case of very light crude oil, as follows:

- Benzene = 0,6% gr;
- Toluene = 1,7% gr;
- Ethyl benzene = 0,2% gr;
- Xylenes = 3, 4% gr.

To estimate the size of impact on soil and terrestrial ecosystems that grow and live in that habitat, BTEX concentrations extrapolated from the total oil hydrocarbon content were compared with soil reference values for sensitive and less sensitive uses (Table 2).

**Table 2.** Events correspondence matrix

No.	Event [T]	Pollutant concentration [mg/kg dry matter]			Pollutant toxicity
		Scenario			
		1	2	3	
1.	Accidental spillage THP (Benzene)	12	30	60	Moderate
2.	Accidental spillage THP (Ethyl benzene)	4	10	20	Moderate
3.	Accidental spillage THP (Toluene)	34	85	170	Low
4.	Accidental spillage THP (Xylenes)	68	170	340	Moderate

The estimated impact on the environment for the 3 scenarios of accidental spills was determined by correlating the concentration of pollutant in soil (per BTEX element) with the toxicity degree of each chemical compound discharged into the environment (Table 3).

**Table 3.** Level of risk and impact on soil organisms

No.	Event [T]	Level			Impact		
		Scenario			Scenario		
		1	2	3	1	2	3
1.	Accidental spillage THP (Benzene)	2.5	2.5	2.5	Moderate to high		
2.	Accidental spillage THP (Ethyl benzene)	1.5	1.5	2	Low to moderate	Low to moderate	Moderate
3.	Accidental spillage THP (Toluene)	1.5	1.5	2	Low to moderate	Low to moderate	Moderate
4.	Accidental spillage THP (Xylenes)	2.5	2.5	2.5	Moderate to high		

The extent of the impact generated by the leakage of petroleum hydrocarbons (very light crude oil) into soils, according to the scenarios, achieved by aggregating the concentration and toxicity indices of BTEX compounds from soils is:

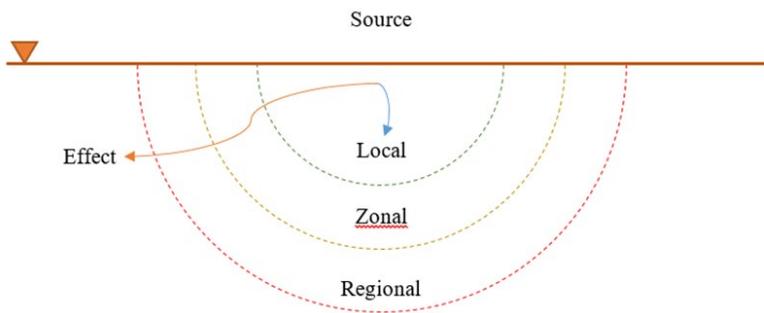
- Moderate to high in the case of benzene and xylenes in THP in all simulated scenarios;
- Moderate in the case of toluene and ethyl benzene in THP for scenario 3;
- Reduced to moderate for toluene and ethyl benzene in THP for scenarios 1 and 2.

This first assessment estimates only a physical dimension of the impact generated by the substance (compound) itself, without taking into account the physical and chemical

characteristics of the soil in which the xenobiotic is discharged, or other specific features of the studied area (orography, unsaturated area thickness, direction groundwater flow etc.).

### 3.1. Risk components

The effects of pollutants on the environment are diverse, depending on pollutants' propagation (dispersion) through that environment. The purpose of identifying the environmental components that are affected by a pollutant is to study how pollutants are dispersed through that environment, in order to obtain an extension of pollutants' concentration in time and space. Following the accidental discharge of xenobiotics, the ecosystems in the studied area are exposed to pollutants' action, at various levels that vary considerably: local, zonal and regional exposure (Fig.1). This action varies depending on where the accidental discharge occurs, because it may have a local expansion, in the unsaturated area, but may also have a regional expansion, in surface waters, if the polluted area (or source) is close to a water course.



**Fig. 1.** Levels at which effects occur in groundwater and surface water

Thus, on a site where several potential sources of contamination are identified, the risk may be varied, depending on the area where the contamination occurs. These sources may be represented by national and natural parks, protected natural areas, archaeological sites, species of special scientific and ecological importance, etc.

In addition to the influences exerted by the place of accidental pollution, pollutant expansion scenarios also largely depend on characteristics of the environment in which the pollutant is dispersed (transfer path) to the source of interest (target) and how the substance is bio transformed, when passing through the unsaturated area. [7].

The risk assessment methodology was applied for a simplified accident with light crude oil, being taken into account the transfer of pollutant in soil and groundwater, thus were considered for the studied crude oil field 3 evolution scenarios of BTEX organic compounds associated with TPH from light crude oil. Based on the lithological and stratigraphical model of the field was chosen a migration profile made of sandy clay (of 10 m average thickness) in order to establish the area of influence for each BTEX compound studied.

The simulation of Benzene, Toluene, Ethyl-benzene and Xylene from light crude oil using the UNSat application was performed depending on the maximum migration time of compounds, through the unsaturated area. Thus, for: Benzene - 20 years, Toluene - 30 years, Ethyl-benzene - 45 years and for Xylenes - 35 years. Computer simulations performed along with the project collaborators, show a spatial and temporal extension that is quantified as areas of influence (Table 4) for the crude oil field. Computer simulations took into account the physical and chemical properties of the unsaturated area, the rainfall regime, the solubility

of pollutants in water, the physical and chemical characteristics of pollutants and the rates of liquid phase biodegradation for aerobic conditions.

Computer simulations of the migration of BTEX-type organic compounds from the composition of a light crude oil, at both instantaneous and continuous pollution (for one month), that can reach the saturated area (groundwater), show the following temporal extension of pollutants through the unsaturated zone:

- Benzene - 9 years;
- Ethyl benzene - 37 years;
- Toluene - 19 years;
- Xylenes - 27 years.

**Table 4.** Risk components for the studied area

No.	Environmental factor [E]	Expansion	Level	Areas of influence [m <sup>2</sup> substance/year]		
				Scenario		
				1	2	3
1.	Soil (BTEX)	Local	1	1	1	1
2.	Groundwater (Unsaturated area) Benzene	Zonal	2	1,1	1,1	1,1
3.	Groundwater (Unsaturated area) Ethyl-benzene	Local	1	0,27	0,27	0,27
4.	Groundwater (Unsaturated area) Toluene	Local	1	0,53	0,53	0,53
5.	Groundwater (Unsaturated area) Xylenes	Local	1	0,37	0,37	0,37

BTEX organic compounds areas of influence that affects the soil is considered to be local, because in the scenarios developed, pollution was considered to be point-like, a unit areas of 1 m<sup>2</sup> being used for computer simulations of pollutant dispersion. At this scale, pollutant emissions were considered coming from a single point source and the environmental objectives that need to be protected are considered being in the source's vicinity.

This pollution scenario, as well as research objectives, sights the risk of groundwater contamination, therefore the actual spread of pollutant in soil was partially neglected because it was tracked the contaminant migration through unsaturated zone. If a thorough analysis of soil pollution is desired, soil contamination duration/exposure time to certain xenobiotics shall be taken into account.

Groundwater's areas of influence was calculated according to the migration rate (years) of BTEX organic compounds in the aquifer from computational simulation performed with UNSat software, relative to the average thickness of the unsaturated area in the studied area. Thus, the simulation of the pollutants in the unsaturated area with the UNSat software were performed by taking into account the time scale, frequency and duration of pollutants' discharge into soil and multiplication time of organisms and microorganisms were taken into account. Based on the results of the simulations for each BTEX compound was established extension of the pollution intervals being established, as like:

- At local level -  $A_i \leq 1 \text{ m}^2 \text{ substance / year}$ ;
- At zonal level -  $1 \geq A_i \geq 2 \text{ m}^2 \text{ substance / year}$ ;
- At regional level -  $2 \geq A_i \geq 3 \text{ m}^2 \text{ substance / year}$ .

Multiplication times are reduced for organisms and microorganisms living in soil, so pollutants' emission can cover a considerable part of their life cycle. Also, organisms exposed to such concentrations during such an episode are continuously affected, and exposure times can be compared to levels with no lethal effect (NLE) extrapolated from chronic toxicity data. The use of the regional scale is recommended only if the point source of pollutants cannot be individualized, because a substance whose concentration is diffused locally by cumulative and dissolution effects of substances contained by the mineral matrix, will have higher concentrations at regional level.

### 3.2. Benzene toxicity level

In order to assess the risk from the environmental and ecotoxicological point of view must be chosen first what chemical component is harmful to the ecosystems and human body. Thus, in this case was chosen benzene because the evolution in soil and unsaturated zone is faster than other studied xenobiotic and the toxicological effect in nature are more harmful to the environment than Xylene, Toluene and Ethyl-benzene.

According to studies conducted by the US Environmental Protection Agency, flora and fauna living and growing in soils may be sensitive, present chronic and acute toxicities to certain concentrations of benzene, as shown in table 7.

**Table 7.** Level of toxicity factor for benzene concentrations in soils

Level of toxicity	Eco-toxicological indicators	Benzene concentration
Species Sensitivity Data	PNEC (mg/ kg soil dw)	4.8
Chronic Toxicity	NOEC (mg/kg soil dw)	0 ÷ 63
	LOEL (mg/kg soil dw)	97 ÷ 172
Acute toxicity	LC 50 (mg/kg soil dw)	199
	EC 50 (mg/kg soil dw)	130.2

In the general methodology of eco-toxicological risk assessment, these toxicity levels are correlated on vulnerability classes for species present in a certain environment (soil). Vulnerability classes of species living in soils are developed according to the effects induced on organisms, microorganisms and roots of vegetation present in soil. Thus, for chronic and acute toxicity induced by benzene, the moderate and high vulnerability classes were established and the low vulnerability class was set as the sensitivity limit of organisms.

Toxicity levels (Table 8), according to studies conducted by the US Environmental Protection Agency, flora and fauna which are living and growing in water, may present sensitivities, chronic and acute toxicities at certain concentrations of benzene, after a certain exposure time.

**Table 8.** Level of toxicity factor for benzene concentrations in water

Level of toxicity	Eco-toxicological indicators	Benzene concentration			
		Toxicity to fish		Toxicity to invertebrates	
		Short term	Long term	Short term	Long term
Species Sensitivity Data	PNEC (mg/ L)	N/A	N/A	N/A	N/A
Chronic Toxicity	NOEC (mg/ L)	N/A	0.8	N/A	3
	LOEL (mg/ L)	N/A	1.6	N/A	N/A
Acute toxicity	LC 10 (mg/ L)	N/A	0.8	10	3
	EC 10 (mg/ L)	N/A	0.8	N/A	3
	LC 50 (mg/ L)	5.3	X	10	N/A

Vulnerability classes of species living in the aquatic environment are established according to the effects induced on fauna living in water or using the water, so the vulnerability classes were established on the degree of toxicity induced by benzene on the ecosystems. Thus for acute toxicity is appointed to highly vulnerable to the ecosystems, chronic toxicity for moderate class and the class of low vulnerability was set as the sensitivity limit of organisms.

It is considered that following the 3 scenarios of petroleum hydrocarbons pollution, the surface polluted with benzene might interfere with the root systems of fruit trees (apple - *Malus pumila*) and that of a cereal crop (barley - *Hordeum vulgare*) artificially watered with water extracted from drillings. The water extracted from drillings is also used in local aquaculture (carp breeding - *Cyprinus carpio*) and later discharged into rivers. The benzene contamination scenario from light crude oil is determined by the maximum concentrations extrapolated from computer simulations for continuous pollution. Analysis of the vulnerability of species to change, correlated with the ability of the species to adapt to concentrations of benzene discharged into the soil (Table 9), shows moderate vulnerabilities in case of root system's exposure to soil pollutant concentrations and low vulnerability for species living in basins fed from supply drillings, used by local aquaculture.

In order to estimate the vulnerabilities, as a part of the eco-toxicological risk for the crude oil field, it is necessary to define some routes for the transfer of pollutants to local organisms or populations.

**Table 9.** Vulnerability degree for species in the studied area

Identified species	Vulnerability of species			Adaptation capacity
	Scenario			
	1	2	3	
<i>Malus pumila</i>	12 mg/kg s.u	30 mg/kg s.u	60 mg/kg s.u	High
<i>Hordeum vulgare</i>	12 mg/kg s.u	30 mg/kg s.u	60 mg/kg s.u	High
<i>Cyprinus carpio</i>	0,37 µg/l	1,1 µg/l	2,25 µg/l	High

The ability of species to adapt to benzene was based on the findings of the EPA USA's eco-toxicological studies on biocenoses [8]. Thus, the adaptability of fruit trees was considered to be high because eco-toxicological studies highlight a higher bioaccumulation of benzene in leaves than in fruits, and in fish populations the concentrations of benzene are metabolized and transformed into phenols (metabolites) by their bodies [9], [10]. In this scenario, the human communities represent the last link in the food chain, which actively has the opportunity to consume organisms (*Malus pumila*, *Hordeum vulgare*, *Cyprinus carpio*) that have indirectly bio accumulated amounts of benzene, that they pass on in the food chain as human exposure concentrations (Table 10).

**Table 10.** Bioaccumulation of benzene in the food chain

Identified species	Vulnerability of species		
	Scenario		
	1	2	3
<i>Malus pumila</i>	157.09 ng/g	392.72 ng/g	785.45 ng/g
<i>Hordeum vulgare</i>	276.11 ng/g	689.94 ng/g	1379.88 ng/g
<i>Cyprinus carpio</i>	N/A	N/A	N/A

The bio accumulated quantities in the identified species were linearly extrapolated from the conclusions of eco-toxicological studies (EPA) showing a very small bioaccumulation in the case of plants, through the root system. It is worth mentioning that EPA studies in plants had a different way of pollutant's action than the particular case described in the methodology.

The amounts of benzene metabolized in food are very small compared to the metabolic capacity of the human body, therefore in the case of long-term exposure, benzene does not have the capacity to bio accumulate in the human body [11, 12]. Thus, for ranking in vulnerability and adaptation indices, human organisms have a high capacity to adapt to benzene and a low vulnerability.

### 3.3. Quantification of environment and ecosystem's eco toxicological risk

Risk definition, as the significance of the risk and all the assumptions on which its assessment is based, the uncertainties and scientific reasoning necessary to identify the risk, assess the effects or toxicity of substances as well as the exposure. Eco-toxicological risk quantification in the studied area, for the 3 scenarios of accidental pollution of petroleum hydrocarbons (THP) in soil, implies the aggregation of indices (T, E, V, C) from equation 3.1 and the estimation of pollution degree based on the obtained risk level.

*Environmental eco-toxicological risk* is the estimation of the incidence and severity of effects that occur in an environmental component, due to exposure, or by estimating adverse effects that could be caused by a predicted exposure to a particular chemical (or group of chemicals).

The risk of groundwater contamination in the studied area (Table 11), quantified by extrapolating the limits regulated by the Romanian legislation in force for groundwater and soils, into risk components established by the eco-toxicological risk calculation *methodology*, is shown below.

**Table 11.** Risk levels of groundwater contamination

Environment factors	Xenobiotic	Eco-toxicological risk		
		Scenario 1	Scenario 2	Scenario 3
Groundwater	Benzene	5 (high)	5 (high)	5 (high)
	Ethyl-benzene	1.5 (low)	1.5 (low)	2 (low)
	Toluene	1.5 (low)	1.5 (low)	2 (low)
	Xylenes	2.5 (low)	2.5 (low)	2.5 (low)

The analysis shows that each xenobiotic, in correspondence with the Romanian legislation in force, results in a high level of risk for groundwater contamination with benzene and a low level of risk for ethyl-benzene, toluene and xylenes.

Eco-toxicological risk to ecosystems represents the estimation of the incidence and severity of effects, occurring in a population (human, flora, fauna), caused by exposure to a substance or by estimating adverse effects that could be caused by a predicted exposure to a particular chemical (or group of chemicals).

The risk of bioaccumulation and transfer of contaminants (Table 12) in the body of ecosystems which are living in the studied area, is quantified by extrapolating the limits regulated by Romanian legislation in force for soils and correlating them with toxicity levels (bioaccumulation) of substances in eco--toxicological risk components.

**Table 12.** Risk levels of bioaccumulation and transfer to ecosystems

Xenobiotic	Species	Eco-toxicological risk		
		Scenario 1	Scenario 2	Scenario 3
Benzene	<i>Malus pumila</i>	3.33 (low)	3.33 (low)	3.33 (low)
	<i>Hordeum vulgare</i>	3.33 (low)	3.33 (low)	3.33 (low)
	<i>Cyprinus carpio</i>	1.66 (low)	1.66 (low)	1.66 (low)
	<i>Homo sapiens sapiens</i>	1.66 (low)	1.66 (low)	1.66 (low)

The eco-toxicological analysis of benzene in the food chain, by correspondence with its dispersion in soil, indicates a low level of risk for bioaccumulation and transfer of

contaminants in the food chain for the 3 scenarios of accidental spill of petroleum products in soil.

## Conclusions

The research carried out for the 3 scenarios in the crude oil exploitation field, resulting a low risk of bioaccumulation and transfer to ecosystems in case of accidental crude oil pollution and a low risk of groundwater contamination in case of accidental pollution by ethyl-benzene, toluene and xylene was assessed for all 3 scenarios.

The risk of bioaccumulation and transfer in the studied ecosystems (*Malus pumila*, *Hordeum vulgare*, *Cyprinus carpio*, *Homo sapiens sapiens*) is low, because the toxicity effects of these volatile organic compounds are manifested in the air through the air-blood interface. On short term, human body is not decisively affected by ingestion of food in which these volatile organic compounds have bioaccumulated or by consumption of contaminated water, because these compounds are not metabolized by the human body, being eliminated from the body in a reduced time through the urinary tract. In the case of potential pollution of the crude oil exploitation site, for which the level of groundwater contamination risk could be moderate and high, it is recommended to apply risk mitigation measures, which may include the addition of substances that may neutralize the crude oil content of soil or replacing hazardous chemicals with safer alternatives, if other substances with a high potential for groundwater contamination are identified.

If a drainage of oil-bearing compounds into the soil takes place on the site of the studied field, it is recommended to use the best techniques for immediate remediation of soil. It is recommended to practice soil remediation techniques because at a secondary event on a site already loaded with residual historical pollution, synergistic effects on the environment and ecosystems will be multiple. Adverse effects on ecosystems are very difficult to predict and so is the distribution of risk within the affected ecological niche, so the only possible thing is to obtain an order of magnitude of relative risk based on a general and simplified model, which can compare either one chemical with another, or groups of chemicals.

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