Assessment of Aquifers' Vulnerability Degree in the Event of Accidental Pollution with Petroleum Products

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Abstract

Adequate management and protection of underground water resources is a desideratum of the current period of evolution as consumption, requirements and degree of impurity of fresh water is increasing with the development of human society. Also, the limited nature of continental fresh water resources requires additional protection measures for current resources, so the understanding and objective assessment of aquifers' vulnerability degree in case of accidental pollution, represents the basis of environmental policies developed for the purpose of sustainable development of human communities. The study of the vulnerability degree of environmental factors and ecosystems that directly or indirectly interact with groundwater resources was carried out by analyzing groundwater quality indicators (BTEX) based on the vulnerability intervals established according to national legislation in force. The aquifers' vulnerability degree the was evaluated for 3 possible scenarios of accidental pollution with petroleum products for a well field located in the south of Romania. The effects of BTEX compounds on ecosystems that directly or indirectly interact with groundwater were evaluated according to synergistic effects of xenobiotics, manifested at ecological niche level. The aim of the vulnerability degree study is for it to function as an analytical tool to identify areas vulnerable to pollution phenomenon and to develop the best measures to limit the phenomenon of contaminants' transfer to other underground water bodies. Aquifers and ecosystems' degree of vulnerability to external pressures is an integrated component of the environmental or ecotoxicological risk assessment process associated with the phenomenon of accidental pollution that may have effects on the quality of water stored in aquifers. Results of the conducted research are of interest for engineers and researchers who study the hydrological and hydrodynamic phenomena of underground water in order to develop and apply the best measures to reduce the impact caused by economic activities.

Keywords: aquifers, vulnerability degree, accidental pollution, petroleum products

Introduction

Research indicates that groundwater undergoes a process of mineral dissolution from rocks during internal circulation, resulting in the presence of mineral ions such as Na^+, Mg^{2+}, Ca^{2+}, and K^+. Consequently, groundwater exhibits a high degree of mineralization and is generally devoid of pathogens, eliminating the need for primary water purification in most cases. While short periods of drought and limited availability of surface water have minimal impact on groundwater supply, there is a significant risk of chemical pollution in groundwater sources and aquifers due to human activities in southern Romania, such as leaks and infiltrations. Chemical contamination renders groundwater unsuitable for domestic and industrial use, making the purification process challenging and expensive. The contamination of groundwater with oil products is primarily caused by unregulated exploitation of deposits, accidental leaks, or improper handling of crude oil extraction and transportation systems. Oil products spilled onto the ground infiltrate the soil, migrate through interstitial pores of rocks in the unsaturated zone, and may eventually contaminate water reserves stored in aquifers, depending on the degree of pollution. The accidental release of BTEX organic compounds into the environment leads to various partitioning and degradation processes that disproportionately affect certain ecological niches, depending on the effects of the chemical substances involved. Organisms inhabiting soils and sediments are in close proximity to these niches and, consequently, to the contaminants present in their surroundings. Evaluating exposure levels in sediments and soils is challenging and complex, as it requires specific assumptions and assessments limited to certain groups of chemical substances. Benzene, known for its high volatility, plays a crucial role in the transport and partitioning of this chemical in the environment. It readily volatilizes from surface water and returns to the atmosphere [1]. Benzene released onto soil surfaces volatilizes into the atmosphere, enters surface water through runoff, and leaches into groundwater. Benzene exhibits high mobility in soil and easily leaches into groundwater, as indicated by its soil organic carbon sorption coefficient (K_{oc}) range of 60-83 [2, 3]. Adsorption studies on aquifer solids reveal that benzene tends to adsorb to these solids. The rates of adsorption and desorption of benzene by dry soil grains show that achieving equilibrium requires hours, with adsorption occurring much faster than desorption [4]. The overall persistence of benzene in sandy soils is determined by the rate of volatilization and leaching [5]. Studies demonstrate that barley plants can uptake benzene from soil, with bioconcentration factors (BCFs) decreasing over time due to growth dilution [6, 7]. Air-to-leaf transfer is considered the major pathway for vegetative contamination by benzene, with estimates suggesting that exposed food crops consumed by humans and used as forage by animals contain approximately 587 ng/kg of benzene [8].The majority of this contamination (81%) occurs through air-to-leaf transfer, while root uptake accounts for the remaining 19% [8]. Benzene can also accumulate in the leaves and fruits of plants. After exposure to benzene-rich environments, blackberries and apples contained measurable amounts of benzene, exceeding the atmospheric partitioning coefficient of benzene [9]. At extremely high concentrations, such as those resulting from a petroleum spill, benzene (along with other petroleum compounds) can be toxic to microorganisms, leading to slow degradation rates compared to lower initial concentrations. However, under specific conditions, such as in acidic groundwater, microbial degradation of benzene can occur within a matter of days [10, 11].

The aerobic degradation of benzene can be influenced by the presence of other aromatic hydrocarbons. When a bacterial culture is grown with aromatic hydrocarbons along with nitrogen-, sulphur-, and oxygen-containing aromatic compounds, the efficiency of benzene degradation decreases compared to a culture grown with aromatic hydrocarbons alone. The presence of pyrrole strongly inhibits benzene degradation. Benzene degradation rates are high when toluene and xylene are present [12]. The bioconcentration potential of benzene in aquatic organisms and some plants is generally low to moderate [13, 14], but high in certain plants [15]. The accumulation of benzene on vegetation primarily occurs through air-to-leaf transfer. Bioaccumulation of benzene in the food chain is negligible, and the absorption of benzene by plant roots is insignificant. Although exposure to benzene through food ingestion is believed to be minimal, standardized methods for assessing benzene contamination in the environment and associated risks of exposure are needed. Benzene has been detected in human body fluids and tissues such as blood, urine, and fat [16, 17]. Biological monitoring studies have been conducted on the general population [18]. Information on baseline levels of benzene in breath for both smokers and non-smokers is available [19], as well as baseline blood levels and urinary metabolite levels in unexposed individuals. It would be helpful to have information on exposure levels for populations living near hazardous waste sites in order to estimate their level of exposure. The primary metabolite of benzene is phenol, which is excreted in urine as glucuronide and sulfate conjugates. Normal baseline levels of urinary phenolic metabolites in humans typically range from 2 to 18 mg/L. [20]

Materials and methods

In order to assess aquifers' vulnerability degree in case of accidental pollution with oil products, three potential scenarios of soil and water pollution in the unsaturated zone were developed, the dispersion of BTEX compounds in the unsaturated zone being modelled with the UNsat application using the SESOIL module (Seasonal Transport and Fate of Volatile Organic Contaminants in the Unsaturated Zone Model), which models transport and behaviour of pollutants taking into account adsorption, volatilization, biodegradation, cation exchange and hydrolysis. Thus, we have three sub-models that simulate water infiltration into soil, contaminant transport and erosion. The SESOIL model proves to be valuable in scenarios where the contaminant exhibits low mobility, is present in low concentrations, or possesses low toxicity. If the contaminant has low mobility, then it will not reach the top of the saturated zone (groundwater) even under long simulations (decades). If the contaminant is present in low concentrations or has low toxicity, then it may reach groundwater but concentration in leachate at the bottom of the unsaturated zone is not sufficient to create an impact on groundwater at levels that exceed limits set by standards of quality. Under these conditions, the existing soil contamination is not a danger for groundwater pollution. Alternatively, the model can be used to calculate acceptable soil decontamination limits. Thus, if the modelling indicates that present soil contamination is not a risk to groundwater, or if a site is remediated to site-specific criteria using this model, no further action is required. Mathematical modelling of the unsaturated zone and assessment of BTEX compounds' dispersion degree aims to predict the concentration of pollutant present in the aquifer after a period of 20 years from a potential soil pollution event.

Assessment of aquifers' vulnerability degree represents an important component in estimating ecotoxicological and environmental risks which consist of extensive development of terms characterizing 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 approach utilized involves categorizing characteristic terms into matrices associated with events, environmental risk, and the impact on soil and ecosystems. The quantification of environmental and ecotoxicological risk, based on the impact terms, is 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.

Thus, the estimation of the degree of vulnerability of aquifers to change and/or the estimation of the term V/C from equation 1 was carried out on vulnerability intervals established according to the national legislation in force and the degree of vulnerability of ecosystems to change was carried out through theoretical documentation and experimental of the toxicological effects manifested by xenobiotics on terrestrial and aquatic ecosystems.

Due to the complexity of the phenomena of predicting vulnerabilities and quantifying the eco-toxicological risk on biocenosis, only the eco-toxicological analysis of benzene is studied for this part of the methodology. The study of benzene is based on the specific physical and chemical properties of the compound, the high percentage of TPH, the mobility and the much higher degree of toxicity in relation to the other analysed substances.

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Results and discussion

Vulnerability of environmental factors represents the way in which certain parameters of environmental components are affected by certain xenobiotics. This particular case is focused both on a study of vulnerability degree of underground water and on studying ecosystems' degree of vulnerability to change by identifying the eco-toxicological effects through which water, used according to certain scenarios, can influence other environmental components or even humans.

Evaluation of groundwater vulnerability degree

For the analysis of groundwater vulnerability degree, the potential aquifer contamination values resulting from computer simulations were directly included in the aggregated vulnerability intervals established according to reference values of pollutants, regulated by Decision no. 53 of January 29, 2009 for the approval of the National Groundwater Protection Plan against pollution and deterioration - with subsequent amendments and additions.

This practice reduces the third term (V/C) from equation (1) for calculating the eco-toxicological risk to a vulnerability term compared to national legislation in force, for which it is assumed that when establishing these limits, cumulative ecological - toxicological effects of substances were also taken into account.

Aggregated vulnerability intervals for groundwater, that characterize the action of each xenobiote assumed to be able to modify the qualitative state of these waters, are established in table 1.

Tab. 1. Groundwater vulnerability intervals.

Quality indicator	MU	Vulnerability to change		
		Low	Average	High
Benzene	μg/l	$C \leq 10$	$10 < C < 50$	$C > 50$
Etil-benzene	μg/l	$C \leq 100$	$100 < C < 1000$	$C > 1000$
Toluene	μg/l	$C \leq 30$	$30 < C < 300$	$C > 300$
Xylenes (sum)	μg/l	$C \leq 50$	$50 < C < 500$	$C > 500$

For the studied area, the analysis of vulnerability to change by estimating the concentration of xenobiotics in groundwater was carried out based on output data resulting from computer simulations for the 3 scenarios. Aggregation of values in the intervals of vulnerability to change (table 2) was carried out based on the most unfavorable scenario, which assumes 1 month of continuous pollution with oil products and accepting maximum concentrations for interpretation.

Tab. 2. Groundwater vulnerability intervals in the studied area

Quality indicator (groundwater)	Vulnerability	Level	Vulnerability to change		
			Scenario		
			1	2	3
Benzene	Low	1	0,37	1,1	2,25
Etil-benzene	Low	1	$2,27*10^{-4}$	$2,19*10^{-3}$	$1,23*10^{-2}$
Toluene	Low	1	$6,07*10^{-4}$	$6,13*10^{-3}$	$3,13*10^{-2}$
Xylenes	Low	1	1,35	4,46	9,54

Following the simulations of BTEX compounds (Figure 1) in the unsaturated zone and the framing of expected xenobiotic concentrations in classes of vulnerability to change, it was found that there is a small possibility of aquifer contamination, because of low contaminant concentrations that may appear in the aquifer after a long period of time.

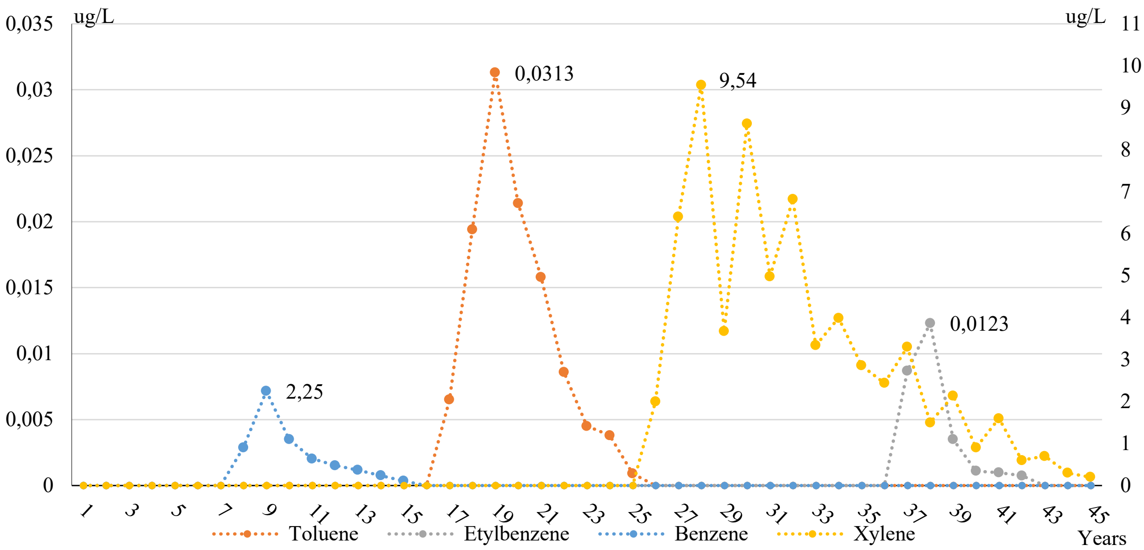

Fig. 1. Evolution of BTEX compounds in the unsaturated zone

The maximum results were processed to estimate possible effects on ecosystems, ensure safety standards, protect local communities and from the perspective of sustainable development of sustainable communities, based on objectives of sustainable development established within the 2030 Agenda. Which stipulates the protection and restoration of ecosystems related to water, wetlands, rivers, aquifers.

Evaluation of vulnerabilities and subsequently of risk, based on concentration limits regulated by in force legislation, does not constitute a genuine scientific foundation for estimating the risk of adverse effects on biocenoses, in this case it is recommended to expand the complexity of the study on direct study of effects and eco-toxicological mechanisms exerted on ecosystems.

Assessing the vulnerability degree of terrestrial and aquatic ecosystems

Table 3 in the EPA research presents findings on benzene toxicity thresholds for flora and fauna in soil ecosystems, examining their sensitivities, chronic toxicity, and acute toxicity at varying benzene concentrations. The data enhances understanding of benzene's potential adverse effects on soil-dwelling organisms, aiding ecological risk assessment and the development of mitigation strategies to protect ecosystem health and integrity.

Tab. 3. 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

The establishment of vulnerability classes in the eco-toxicological assessment plays a crucial role in understanding the potential impacts of benzene on species residing in soil ecosystems. These vulnerability classes are developed by evaluating the effects induced by benzene on various organisms, microorganisms, and the intricate root networks of vegetation present in the soil. For chronic and acute toxicity induced by benzene, medium and high vulnerability classes are assigned. This implies that certain species within the soil ecosystem are more susceptible to the adverse effects of benzene exposure over extended periods or at higher concentrations. These species may experience detrimental consequences, such as reduced growth, impaired reproduction, or even mortality, as a result of benzene's toxic properties. On the other hand, organisms with a lower sensitivity threshold are categorized into the low vulnerability class, indicating a comparatively higher tolerance to benzene's toxic effects.

Table 4, based on studies conducted by the US Environmental Protection Agency, and provides valuable insights into the toxicity levels associated with benzene exposure. It offers a comprehensive understanding of the concentrations at which aquatic flora and fauna, reliant on water for their survival and development, exhibit sensitivities or experience chronic and acute toxicities. This information aids in assessing the potential risks posed by benzene to aquatic ecosystems and assists in establishing appropriate protective measures and regulations.

By considering vulnerability classes and toxicity levels, the eco-toxicological assessment offers valuable guidance for monitoring and managing benzene contamination in soil and water environments. It helps ensure the protection and preservation of ecosystems and supports decision-making processes aimed at minimizing the potential adverse effects of benzene on both ecological and human health.

Tab. 4. 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

The establishment of vulnerability classes for species in aquatic environments plays a crucial role in understanding the potential impacts of benzene on the flora and fauna that depend on or inhabit water ecosystems. These vulnerability classes are determined by evaluating the effects induced by benzene on various organisms and plant species present in aquatic environments. For chronic and acute toxicity induced by benzene, medium and high vulnerability classes are assigned. This indicates that certain species in the aquatic ecosystem are more susceptible to the adverse effects of benzene exposure over prolonged periods or at higher concentrations. These species may experience detrimental consequences, such as impaired growth, reproductive problems, or even mortality, as a result of benzene's toxic properties. On the other hand, organisms with a lower sensitivity threshold are categorized into the low vulnerability class, indicating a relatively higher tolerance to the toxic effects of benzene.

To estimate vulnerabilities as part of the ecotoxicological risk assessment for the studied area, it is necessary to consider the transfer routes of pollutants to local organisms or populations. In this case, three pollution scenarios involving THP (Total Hydrocarbon Petroleum) contamination were examined. The presence of benzene in the surface area affected the root systems of fruit trees (apple - Malus pumila) and cereal crops (barley - Hordeum vulgare) that are irrigated with water extracted from boreholes. Furthermore, the water extracted from the boreholes is utilized in local aquaculture, specifically for breeding carp (Cyprinus carpio), and subsequently discharged into rivers. The benzene contamination levels were determined based on maximum concentrations extrapolated from computer simulations representing continuous pollution.

Analysing the vulnerability of species to changes in correlation with their adaptation capacity to benzene concentrations in the soil (as presented in Table 5), it is evident that species with exposed root systems have a medium vulnerability to the pollutant concentrations. On the other hand, species residing in pools fed by the supply drill exhibit a low vulnerability to benzene.

This assessment of species vulnerability and adaptation capacity provides valuable information for understanding the potential effects of benzene contamination on different organisms within the studied area. By considering these vulnerabilities, appropriate measures can be implemented to mitigate risks and protect these species and their respective aquatic habitats. It aids in guiding conservation efforts, management strategies, and regulatory measures to safeguard the integrity and health of aquatic ecosystems impacted by benzene contamination.

Tab. 5. Species' vulnerability degree in the studied area

Identified species	Species' vulnerability			Adaptability
	Simulated scenario			
	1	2	3	
Malus pumila	12 mg/kg s.u	30 mg/kg s.u	60 mg/kg s.u	High
Hordeum vulgare	12 mg/kg s.u	30 mg/kg s.u	60 mg/kg s.u	High
Cyprinus carpio	0,37 µg/l	1,1 µg/l	2,25 µg/l	High

Adaptability of species to benzene was based on conclusions of eco-toxicological studies carried out by the USA EPA on biogenesis. Thus, for fruit trees it was considered a high adaptation capacity because eco-toxicological studies emphasize a greater bioaccumulation of benzene in leaves than in fruits, and in the case of fish populations, the body metabolizes and transforms benzene concentrations into phenols (metabolites).

Conclusion

In terms of mobile BTEX (benzene, toluene, ethylbenzene, and xylene) contaminants migrating through the unsaturated zone to the aquifer, simulations indicate that peak concentrations do not exceed the limits established by legislation. When modelling the distribution of mobile BTEX-type contaminants associated with crude oil, it has been determined that a spill of crude oil on the ground, up to concentrations of approximately 1% (10000 mg/kg su) in crude oil, does not pose a risk to underground water through surface infiltration in the ford area. To assess the potential bioaccumulation of BTEX contaminants in identified species, linear extrapolation of accumulated amounts was performed based on conclusions from eco-toxicological studies conducted by the Environmental Protection Agency (EPA). These studies indicate a very low level of bioaccumulation, particularly in plant species through their root systems. It is important to note that the EPA studies on plants might have different pollutant behaviour compared to the specific case described in the research being conducted.

The amount of benzene metabolized through food ingestion is minimal compared to the body's metabolic capacity. Consequently, benzene does not possess the ability to bioaccumulate significantly in the human body, particularly during long-term exposure. Therefore, when considering vulnerability and adaptation indices, human organisms exhibit high adaptability to benzene and low vulnerability.

This information underscores the relatively lower risk associated with benzene bioaccumulation in both environmental ecosystems and the human body. It provides valuable insights for assessing the potential impacts and designing appropriate risk management strategies to ensure the protection of both ecological and human health.

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