

Assessment of risk of groundwater quality deterioration within Siversky Donets river basin

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Abstract. An adaptation of the national water resources management system in accordance with the requirements of European legislation creates the legislative basis for reforms implementation in the field of monitoring and water use. The basin management principle started to be applied, according to which surface and groundwater arrays are the water

resources management units. The preliminary groundwater array status assessment (both quantitative and qualitative) is a necessary procedure that enforces the development of appropriate monitoring program and measures elaboration in order to improve groundwater ecological status. This study tested a methodology of groundwater deterioration risk assessment as a tool for previous groundwater array cological status estimation. The research provides an approbation of the methodology in relation to groundwater arrays identified and delineated within Siversky Donets river basin (that covers Kharkiv, Donetsk and Lugansk regions). Surface water and groundwater are affected by significant anthropogenic pressures in form of pollution from point sources of heavy industry facilities. A risk model comprises groundwater vulnerability map and simulated model of anthropogenic pressure magnitude distribution reflecting the impact extent of the main sources of groundwater pollution. Vulnerability map was developed using the tool of input factors weight index estimation. Authors considered the following factors as determining - soils characteristic, aeration zone characteristics, geological environment of groundwater arrays of Cenozoic-Mesozoic group. The pollution load index was calculated. Input data for calculation are concentrations of hazardous substances (metals, semimetals, halogens and nitrates and phenol compounds) measured in groundwater samples during the 2017 monitoring year period. The results of the value interpolation of calculated pollution load index reproduce the focal (point) nature of groundwater pollution and indicates the significant groundwater pollution of Quaternary and Upper Cretaceous, both Carboniferous aquifers and corresponding groundwater arrays. A logical matrix is created on the basis of a combination of pressure magnitudes and vulnerability classes. The area of each class of risk is calculated within groundwater arrays with zonal statistic technique. Consequently, each groundwater array is assigned with preliminary estimated risk category. Created model enables to perform previous groundwater array status assessment. The proposed model expected to be more useful after the data on pollution from diffuse sources obtaining and its validation after the first stage of surveillance monitoring realization.

Keywords: groundwater body, ecological status, vulnerability, pollution load index, risk of pollution

Оцінка екологічного ризику погіршення якісного стану підземних вод у межах басейну р. Сіверський Донець

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Анотація. Адаптація національної системи управління водними ресурсами відповідно до вимог європейського законодавства створює основу для впровадження реформ у сфері моніторингу та водокористування. Здійснено перехід до басейнового принципу управління, згідно із яким, одиницями управління водними ресурсами є масиви поверхневих та підземних вод. Попередня оцінка статусу (якісного та кількісного) масиву підземних вод є необхідною процедурою перед розробкою та впровадженням моніторингу та заходів щодо покращення екологічного статусу підземних вод. У якості інструменту попередньої оцінки екологічного статусу масивів підземних вод запропоновано методику оцінки екологічного ризику погіршення якісного стану підземних вод. Оцінку здійснено для масивів підземних вод, виділених у межах річкового басейну Сіверського Дінця. З метою створення оціночно-ризикової моделі побудовано картографічну модель уразливості підземних вод до забруднення та створено модель розподілу амплітуди антропогенних навантажень, що відображає міру впливу основних об'єктів забруднення підземних вод. Модель уразливості побудовано із використанням індексної оцінки вхідних критеріїв, серед яких використано – характеристику ґрунтового покриву, зони аерації, геологічного середовища масивів підземних вод кайнозойськомезозойської групи. Здійснено розрахунок індексу навантаження від забруднення. У якості вхідних даних для розрахунку використано концентрації небезпечних компонентів І та ІІ класу небезпеки (металів, напівметалів, халькогенів), виявлені у пробах води. Результати інтерполяції розрахованих значень індексу навантаження від забруднення відтворюють вогнищеву (точкову) картину поширення забруднення у підземних водах алювіальних четвертинних та верхньокрейдових водоносних горизонтів у межах досліджуваної території. Створено логічну матрицю на основі поєднання амплітуд антропогенного тиску від забруднення із класами уразливості підземних вод. Створена модель оцінки екологічного ризику дозволяє попередньо віднести кожний масив підземних вод до відповідного класу ризику досягнення екологічних цілей.

Ключові слова: масив підземних вод, екологічний статус, уразливість, індекс навантаження від забруднення, екологічний ризик забруднення

Introduction. The process of Ukrainian legislative framework transition to the European-oriented direction of development creates necessary background for current and further reforming of all spheres of human activity. In particular, since the Association Agreement document between Ukraine and EU countries was signed and ratified in 2014, significant change has occurred to environmental management and protection policy. Series of legislative acts, documents and acquis that regulate the basic principles of water resources management in accordance with the Water Framework Directive 2000/60 (European Commission, 2000), have been developed and implemented.

The Law of Ukraine (dated October, 2016) No. 1641 - VIII "On Amendments to Certain Legislative Acts of Ukraine on Implementation of Integrated Approaches in the Management of Water Resources Based on the Basin Principle" introduces the legal basis and sets new prerequisites for water management system reforming by cancellation the territorial principle and adoption the river basin management, where the river basin serves as a management unit for all water resources, including groundwater. As a result of mentioned law implementation the Water Code of Ukraine (1995), was amended. There have been important developments associated with new water management units' definitions - surface and groundwater bodies, as well as water resources management core provision according to the ecological objectives, required and established by European legislation, good ecological status (both quantitative and qualitative) achievement for all water bodies (surface and groundwater).

The new groundwater resources management concept (in conformity with updated water legislation) focuses on the cycle of strategies and measures adoption, launched from the groundwater body identification and delineation procedure and expired by groundwater body status assessment (good or bad) and further appropriate measures development, aimed in groundwater body state restoration, if necessary. It is clear, that the status assessment reliability substantially depends on the appropriately and optimally designed monitoring program (Shestopalov, 2016; Davybida, 2018).

However, it is also important to recognize the role of the risk assessment phase of ecological objectives achievement failure in groundwater management structure, including the data and information preparation in order to develop monitoring network and monitoring program. In accordance with the new Procedure of state water monitoring (The order of Cabinet of Ministers of Ukraine, dated 19.09.2018, No. 758) statements, the development of a monitoring program is preceded by a procedure of identification of anthropogenic impact that can influence on groundwater quantitative and qualitative status. Actually, following the European experience - first cycle of surveillance monitoring focuses on actual targets emerged from preliminary performed risk assessment (European Commission, 2003). To date, however, the methodology of groundwater risk assessment that meets the requirements of European water legislation as well as reformed Ukrainian environmental legislation, has not been developed (Dovhanenko, 2017). Therefore, it should be expected that he first stage of groundwater state monitoring realization, in accordance with the reformed procedure, which will be organized in coming years for the Dnipro River basin, the Siversky Donets River Basin and the Dniester River basin, will probably produce a lot of methodological uncertainties while groundwater bodies status assignment.

Some concepts and methodological bases for environmental risk assessment, presented in the article and proposed by the authors, were carried out within the framework of the project "Assistance to the Ministry of Ecology and Natural Resources of Ukraine in improving the mechanisms of environmental monitoring" of the OSCE in Ukraine (Denisov, 2018, Ulytsky, 2018).

Material and Methods. *Study Site Description.* The research provides an approbation of the methodology for groundwater pollution risk assessment in groundwater bodies, delineated within Siversky Donets river basin. The river basin territory (comprising a part of Kharkiv and Donetsk regions, and Luhansk region

entirely) is characterized by rich fuel and energy, mineral and raw material resources base, and, as a result, by high concentrations of facilities of heavy industry sector. The man-made impact in study region is the highest compared to the other regions of Ukraine, so the objects of critical infrastructure creates the environmental risk. During the entire period of human activity, surface and groundwater, increasingly, were exposed to impacts and negative effects.

The study proposes to estimate preliminary groundwater bodies status in terms of qualitative status criteria.

Groundwater bodies within Siversky Donets river basin area were identified and delineated following the requirements specified by the "Methodology for surface and groundwater arrays identification" (The Order of the MENR, 2019): groundwater body should be identified as a part of aquifer if it contains significant volume of water enabling to supply drinking water abstraction average rates in 10 m³ per day; as a part boundary settings, including hydrogeological and geological natural boundaries.

Three-dimensional delineation preformed based on three geological structural floors understanding. So, identified groundwater bodies refer to Cenozoic, Mesozoic and Paleozoic aquifers systems.

Thus, as a result of hydrogeological conditions analysis and mentioned above criteria application, groundwater bodies within Siversky Donets catchment area were identified and delineated (Table 1).

The analysis of groundwater bodies' delineation outcomes within site area demonstrates that the largest number of groundwater bodies was identified and delineated in Cretaceous and Paleogene-Neogene system aquifers and aquifer complexes. Such an uneven division is based on exploitation value and significance of mentioned aquifers in sense of drinking water supply. Accordingly, these groundwater bodies require application of appropriate ecological objectives with increased demands. The poor quality sta-

Table 1. Groundwater bodies (GWB) identified and delineated within Siversky Donets river basin catchment area

GWB name	Aquifer media	Number of GWB	
GWBs in alluvial quaternary deposits (a,adH, $a^{1.5}P_{II.}$ III, $a^{6.10}$, laP_{I})	Irregular coarse sands with clay layers, sandy loams	8	
GWBs in alluvial deposits of Pliocene terraces (aN_2)	Browish gray, gray and yellow clayey irregular coarse sands	1	
GWBs in Paleogene and Neogene formations (\mathbf{P}_{2} ₃ +N ₁ , \mathbf{P}_2 kv- \mathbf{P}_3 hr+N ₁ , \mathbf{P}_2 kn-bč, N ₁ pn)	Irregular coarse sands, sandstones, loams	5	
GWBs in Cretaceous system deposits $(K_2, K_{1-2}s)$	Marls, sandstones, irregular coarse sands, chalk	12	
GWBs in Jurassic system deposits and complexes $(J_3, J_{3km}, J_{3ox}, J_2)$	Sands, sandstones, limestones	2	
GWBs in Triassic system deposits $(T_3, T_{1-2sr}, T_{1dr})$	Irregular coarse sands, sandstones	2	
GWBs in Permian system deposits (P_1)	Sandstones with layers of mudstones, aleurolites, lime- stones and dolomites	1	
GWBs in sandy-clayey deposits of carboniferous system (C_1-C_3)	Sandstones with layers of mudstones, aleurolites and thin layers of limestones and coals	3	
	Total	34	

of aquifer contoured by contamination borders, detected with previous monitoring data; and/or as a part of aquifer contoured by the poor quality groundwater flow boundaries in case it causes or can provoke significant deterioration of surface water and terrestrial ecosystems or underlying aquifers.

Spatial boundaries (horizontal dimension) of groundwater bodies respond to groundwater flow

tus of groundwater in Quaternary and Carboniferous aquifers, in turn, needs aquifers to be delineated into greater number of groundwater bodies, assuming and taking in the mind the necessity for further developing of operational monitoring program and measures in order to improve groundwater quality.

Anthropogenic pressure on groundwater quality within the Siversky Donets river basin is carried out by point and diffuse sources. Main source of **diffuse** pollution is agriculture (in form of agricultural wastes) and urban land use (in the form of urban drainage from surface, including runoff and snowmelt). Groundwater quality deterioration happens because of contamination by nitrogen compounds - NO₃ and NH₄. Chemical composition data analysis for the 2017 monitoring year period demonstrated significant excess of ammonium TV (2,6 mg·L⁻¹) in groundwater of Quaternary and Upper Cretaceous aquifers located on the territory of Lugansk region (Krasna river, Bila river, Derkul river).

The screening procedure for relevant anthropogenic pressures and impacts on groundwater quality within basin area indicated significant role of point sources. Screening sources of anthropogenic loading on groundwater status within the basin indicates the significant impact of **point** sources of pollution. Powerful petrochemical, metallurgical, machinebuilding, facilities, as well as coal industry objects are concentrated on the territory of Siversky Donets river basin. So, the largest amount (compared to the whole territory of Ukraine) of sludge collectors and tailing ponds, industrial discharges tanks, dumping ground for solid household waste and rubbish heaps are situated here. Polluted wastewater produced by the coal, chemical and petrochemical industry, iron and steel industry, as well as household wastewater, according to preliminary estimates, are the main source of regional pollution not only of surface water but also of groundwater due to contaminants transport by filtration.

Particular attention should be paid to the impact analysis of the objects of high ecological hazard and critical infrastructure - coal mines – and it influence on groundwater quality and associated ecosystems status. In the current circumstances there has been a massive mines flooding. In future, it can probably cause such negative processes as land flooding, subsidence, as well as groundwater chemical state deterioration within territories adjacent to mine workings.

The risk assessment of groundwater quality deterioration aims to establish causal links between certain anthropogenic pressures (that can takes the form of pollution load) and corresponding impact on the environment and human health. For groundwater, the ecological risk assessment procedure should be based on analysis of the pollution sources, pollution pathways susceptibility (in form of groundwater vulnerability) regarding to aquifer and groundwater as a receptor.

In fact, the method of groundwater vulnerability estimation, as European (Voudouris, 2018) and nation-

al experience has shown (Koshljakov, 2014; Levonjuk, 2018), is limited to two possible approaches:

- direct estimation based on groundwater bodies chemical status monitoring, as well as on certain contaminants' residence time calculation while reaching aquifer, taking into account the protective properties of waterbearing and low-permeable geological complexes and also physic-chemical contaminants properties.
- indirect estimation based on the pollution load calculation combining aquifer vulnerability assessment.

In this study, the groundwater pollution risk assessment is based on a combination of cartographic models of vulnerability and the magnitude of anthropogenic pressure (Kozłowski, 2019):

Risk = Vulnerability + Pressure Load Magnitude

Groundwater vulnerability assessment

The pathway susceptibility can be defined with the same characteristics as aquifer vulnerability – the sensitivity of groundwater system to anthropogenic loads. At the same time, vulnerability value is inversely proportional to the value of groundwater protection level, which demonstrates the lithologic-filtration protective ability of the geological entire settings.

The approach of groundwater vulnerability cartographic model obtaining is a reflection of the DRASTIC model, built on the input factors index estimation method as a tool (Jang, 2017). The DRASTIC method, developed by experts from the US Environmental Protection Agency (Aller, 1987), has been widely used in Europe for recent years as a tool for groundwater vulnerability to pollution mapping. Groundwater vulnerability map is the result of overlay analysis of the layers characterizing input factors' values distribution (1) and the further division of the resulting surface of obtained total vulnerability index into classes. Each layer is divided into classes according to the rule of natural breakdown, a weight coefficient is given to each class.

$$DRASTIC Index = D_r \times D_w + R_r \times R_w + A_r \times A_w + S_r \times S_w + T_r \times T_w + I_r \times I_w + C_r \times C_w, \qquad (1),$$

where *DRASTIC Index* – the resulting vulnerability map (computed surface of the total vulnerability index distribution); D - layer of the depth to groundwater level value distribution; R - layer of the recharge value distribution;

A – layer, that demonstrates the aquifer media characteristics' distribution (sands, limestones, etc.); S – layer, that demonstrates soil types distribution; T - layer of the relief slope value distribution; I – layer, that demonstrates vadoze zone characteristics' distribution; C – layer of hydraulic conductivity value distribution; r - parameter class; w - is the weighting coefficient for each parameter.

Pollution load index and pressure load magnitude calculationz

Taking into account that the main source of groundwater pollution within the Siversky Donets river basin are industrial facilities it was decided to perform the pollution load estimation basically on data analysis comprising concentration estimation of components of the 1st and 2^d hazard classes in groundwater samples. The list of components includes

- metals (Be, Cd, Hg, Co, Ni, Cu, Zn, Pb, Mo, Sr, Cr, Li), Chalcogen (Se), Halogens (Br), Semimetals (B). The analysis showed that in the vast majority of water samples the content of harmful components (each individually) does not exceed the maximum permissible concentrations determined by the sanitary norms for drinking water.

The analysis showed that in majority of water samples, the content of harmful components (individually for each component) does not exceed the TV concentrations determined by the sanitary norms for drinking water in Ukraine (Derivative Sanitary Norms and Rules document 2.2.4-171-10). Instead, in some water samples there is a wide range of components of the 1st and 2^d hazard classes, although sometimes they are contained at low concentrations. Therefore, in order to assess the groundwater resistance in certain groups of chemical elements accumulation (Sobhanardakani, 2016; Bhutiani, 2017), the pollution load index (2) was used:

$$PLI = (CF_i \times CF_{i+1} \times \dots CF_n)^{1/n}, \qquad (2),$$

where *PLI* – pollution load index;

 CF_i – index of contamination by a certain substance;

n – the amount of hazardous substances identified in the water sample.

Index of contamination by a certain substance, identified in the water sample, is calculated by the equation (3):

$$CF_i = \frac{CA}{CN} - 1, \tag{3},$$

where CA – the estimated value of the hazardous substance;

CN – standard value of hazardous substance in drinking water sample (TV established by national standards).

Even if concentrations of hazardous substances in groundwater sample do not exceed the established TV, the value of pollution load index, which is always > 0, matches the presence of dangerous compounds with an appropriate distribution of concentrations.

In order to calculate the pollution load index, indicators of the maximum allowable concentrations (TV analogue in Europe) determined by the sanitary standards for drinking water (Table 2) were used.

Table 2. TV of compounds, used for PLI calculation

 under the study

Substance	TV, mcg·L ⁻¹		
Br	200		
В	500		
Cd	1		
Zn	1000		
Ni	20		
As	10		
Pb	10		
Li	30		
Cu	1000		

In the course of the study, chemical composition data of water samples of the Mesozoic-Cenozoic groundwater aquifers of 2017 year were processed. Further, Kriging interpolation method was applied to each section of well-grouping with calculated pollution load index in order to obtain a simulation of index distribution. Accounting for the study specification, this method is more appropriate, since applying kriging assumes that the distance between the reference points reflects the spatial correlation that can be used to explain the change on the surface. The resulting interpolation surfaces are combined into one for further analysis.

Results and discussion. Based on the DRASTIC index tool applying, a map of groundwater vulnerability within first from the surface unconfined and partly confined aquifers was exposed. As an input criterion for vulnerability model constructing the preliminarily prepared reclassified surfaces (layers) of factors were used. Input layers describe slopes of relief, soil permeability (expressed by reclassified layer of soils mechanical composition), the distribution of rainfall values, recharge zones characteristics, vadoze zone thickness values' distribution. The assignment of weight coefficient to each of the input factors was based on the analysis of the hydrogeological and geological settings of the study area (Table 3).

Input factor	Weight		
Slope	1		
Soils mechanical composition	2		
Rainfall	4		
Recharge zones	2		
Vadoze zone thikness	5		

 Table 3. Input factors for vulnerability model building

The resulting vulnerability model built for groundwater in Cenozoic-Mesozoic aquifers is divided into 5 classes: of very low, low, medium, high and extremely high vulnerability (Fig. 1).

As a result of the application of equation (2), the pollution load index for groundwater in the Cenozoic

and Mesozoic sediments was calculated (Table 4).

Additionally, pollution load indexes from phenols and nitrates in Cenozoic aquifers (and corresponding groundwater bodies) were analyzed, calculated and mapped as a separate layer.

As a result of pollution load index calculating and the corresponding cartographic surfaces modeling, we found out that groundwater contamination has a focal nature and very accurately reflects the behavior of receptors in the system "shallow groundwaterpartly confined groundwater". For this reason, as well as due to the lack of input monitoring information on the groundwater chemical composition, it was decided to use the resulting surface of the pollution load index distribution as the basis for characterizing the magnitude of anthropogenic pressure (Table 5).

Table 4. Concentration values of substances (in $mcg \cdot L^{-1}$) identified in water samples from Cretaceous aquifers within Siversy Donets river basin and calculated value of pollution load index

Sample No.	Br	В	Cd	Zn	Ni	As	Pb	Li	Cu	PLI
1	300	-	-	-	-	-	-	-	-	1.5
2	-	-	2	-	-	-	-	-	-	2.0
3	0.95	0.40	-	-	-	-	-	-	-	0.0019
4	0.95	0.28	-	-	-	-	-	-	-	0.0016
5	0.63	0.40	-	-	-	-	-	-	-	0.0025
6	-	-	-	0.05	0.01	-	0.02	-	-	0.0036
7	1.37	0.37	-	-	-	-	-	-	-	0.002251
8	0.21	0.28	-	-	-	-	-	-	-	0.000767
9	0.20	0.36	-	-	-	-	-	-	-	0.000849
10	0.20	0.50	-	-	-	-	-	-	-	0.001
11	0.20	0.36	-	-	-	-	-	-	-	0.000849
12	0.20	0.19	-	-	-	-	-	-	-	0.000616
13	0.20	0.10	-	-	-	-	-	-	-	0.000447
14	0.42	0.28	-	-	-	-	-	-	-	0.001084
15	0.42	0.24	-	-	-	-	-	-	-	0.001004
16	0.84	0.90	-	-	-	-	-	-	-	0.00275
17	0.20	0.44	-	25	-	-	-	0.008	-	0.001556
19	2.53	0.56	-	-	-	-	-	-	-	0.002381
20	0.63	0.85	-	-	-	-	-	-	-	0.01464
22	-	-	-	0.012	-	-	-	-	-	0.000012
23	0.20	0.05	-	7	-	-	-	0.016	-	0.000782
25	0.32	0.40	-	23	-	-	-	0.016	-	0.001991
28	0.20	0.86	-	20	-	-	0.005	-	-	0.001547
29	-	-	-	0.413	-	-	-	-	-	0.000413
33	410	200	-	-	-	-	-	-	-	0.905539
40	-	-	-	0.06	-	-	-	-	-	0.00006
41	-	-	-	10	-	-	-	-	-	0.01
43	-	-	-	0.04	-	-	-	-	-	0.00004
44	3.38	0.32	-	-	-	-	-	-	-	0.003289
45	1.27	0.19	-	-	-	-	-	-	-	0.001553
47	200	100	-	-	-	-	-	-	-	0.4472
48	0.001	-	-	-	-	0,005	0.01	-	0.005	0.0000059



Fig. 1. Mapping model of Cenozoic-Mesozoic aquifers vulnerability within Siversy Donets river basin

A range of calculated PLI values	Pressure Magnitude		
0-0.2	Very low		
0.2-0.8	Low		
0.8-1	Middle		
1-3	High		
3-7	Extremely high		

Table 5. Anthropogenic pressure magnitude classes andcorresponding ranges of pollution load index values

In order to build a model of risk assessment, the logical matrix of pollution risk classes developed based on vulnerability classes and anthropogenic load magnitudes combination (Table 6). As a result of zonal statistics application, the area of each class of risk is calculated within groundwater bodies. A preliminary assessment of the groundwater body status is carried out on the basis of the predominant risk class according to the area criteria. **Conclusion.** The risk of groundwater chemical status deterioration assessment referring to the new objects of water management system – groundwater bodies – was performed for the first time. It reflects the development of current negative phenomena affecting the groundwater status within the studied region. Validation of the performed estimation and calibration of the input parameters of the estimationrisk model seems to be feasible after the first stage of surveillance monitoring (that meets the requirements

Table 6. The matrix	for groundwate	r pollution risk	assessment
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Pressure Magnitude	Vulnerability						
	Extremely High	High	Middle	Low	Very Low		
Extremely High	EH	EH	Н	Н	М		
High	EH	Н	Н	М	L		
Middle	Н	Н	М	М	L		
Low	М	М	М	L	L		
Very Low	L	L	L	L	VL		

of updated State Water Monitoring Procedure) and obtained data analysis.

The calculated model, proposed under the study, and its application enables to make a preliminary pollution risk assessment for each groundwater body and, accordingly, to adjust the monitoring program. It has been established that the highest risk level (in grades "high" and "extremely high") is set for groundwater bodies in Quaternary and Upper Cretaceous aquifer systems, identified within Siversky Donets river basin catchment area. Also, according to the results of risk assessment, the poor quality status of groundwater bodies in Carboniferous aquifers was identified. However, the further groundwater vulnerability Carboniferous aquifer system model within elaboration is a subject of detailed and in-depth research.

The proposed methodology for risk assessment is based on the groundwater vulnerability model combining the direct building anthropogenic magnitude calculation. pressure Groundwater vulnerability model consists of input parameters weight index estimation and adopted for applying at following stages of groundwater resources management cycle as a screening tool for operational assessment of relative magnitude of other types of anthropogenic pressures (for example, from diffuse sources of groundwater pollution with nutrients, ammonium compounds and nitrate pollution from agricultural sources). Methodology can also be used for preliminary groundwater bodies' status assessment. The possibility of the integrated consideration of proposed vulnerability model with direct characteristics of contamination behavior (migration properties) released from diffuse sources requires further detail investigations.

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