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Assessment of phosphorus fertilizer wastes production impact on subterranean water quality (Gomel chemical plant, Republic of Belarus)

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Abstract. ***Relevance.*** Phosphate raw materials processing at the Gomel chemical plant over a period of more than 50 years has led to accumulation of millions of tons of phosphogypsum dumps. They are a source of subterranean water contamination by sulfates, phosphates and other chemical compounds. Therefore, there is the need in current estimation of the scale and extent of aquifers contamination, taking into account the geological and hydrogeological conditions that influence the migration of contaminants. ***Aim.*** Assessment of hydrogeological parameters affecting distribution of contaminants in subterranean water and the degree of aquifers contamination. ***Objects.*** Aquifers and impermeable horizons. ***Methods.*** Methods for assessment of hydrogeological parameters, methods for determining the chemical composition of subterranean water. ***Results.*** The paper assesses a technogenic effect, produced by dumps of Gomel chemical plant. It reviews a subterranean water monitoring local network within the Gomel chemical plant influence area. The hydrogeological parameters of aquifers of the upper hydrodynamic zone are quantitatively assessed, given that this zone undergoes the strongest anthropogenic impact. The study of subterranean water quality ends up with three classes of contamination: high, moderate and insignificant. Major contaminants include sulphates, nitrogen ammonium, and phosphorus phosphate. The authors find a spatial pattern of subterranean water contaminants differentiation that tends to develop both horizontally and vertically. As a result of the study, areas and sources of high groundwater contamination are determined and mapped. A conclusion is drawn on the necessity of further control over the subterranean water conditions and contamination development in time and space.

Keywords: phosphogypsum dumps, local monitoring of subterranean water, aquifers, aquifer transmissivity, vertical permeability, contamination

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Оценка влияния отходов производства фосфорных удобрений на качество подземных вод (Гомельский химический завод, Республика Беларусь)

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Аннотация. ***Актуальность.*** Переработка фосфатного сырья на Гомельском химическом заводе за более чем 50-летний период времени привела к накоплению миллионов тонн отходов фосфогипса. Они являются источником загрязнения подземных вод сульфатами, фосфатами и другими химическими соединениями. Поэтому существует необходимость современной оценки масштаба и степени загрязнения водоносных горизонтов с учетом геологогидрогеологических условий, влияющих на миграцию загрязняющих веществ. ***Цель:*** оценка гидрогеологических параметров, влияющих на распространение загрязняющих веществ в подземных водах, и степени загрязнения водоносных горизонтов. ***Объекты:*** водоносные и водоупорные горизонты. ***Методы:*** методы оценки гидрогеологических параметров и методы определения химического состава подземных вод. ***Результаты.*** Представлена оценка техногенного влияния отходов производства Гомельского химического завода. На основании данных сети мониторинга подземных вод в зоне влияния Гомельского химического завода количественно определены гидрогеологические параметры водоносных горизонтов верхней гидродинамической зоны, которая наиболее подвержена антропогенному воздействию. В исследованных водоносных горизонтах выделено три уровня степени загрязнения: высокий, средний и низкий. Основными загрязнителями являются сульфаты, азот аммонийный и фосфаты. Установлена вертикальная и горизонтальная дифференциация загрязнения. Выполнено картографирование территории по площадному развитию загрязнения. Проведенное исследование показывает необходимость дальнейшего осуществления мониторинга для контроля распространения загрязнения в водоносных горизонтах.

Ключевые слова: отвалы фосфогипса, локальный мониторинг подземных вод, водоносные горизонты, водопроводимость, вертикальная проницаемость, загрязнение

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Introduction

High rates of development of various industries inevitably lead to formation and accumulation of industrial waste. One of the mass types of waste is waste from the chemical industry.

Phosphogypsum (PG) is a waste by-product from phosphate rock processing by the “wet acid method” of fertilizer and phosphoric acid production.

The current worldwide PG generation is about 300 million tons (Mt) per year. Only 15% of this amount is recycled as building materials, agricultural fertilizers or soil supplements. Mostly PG is stored in large dumps tens of meters height and cover as many as several square kilometers surface area [1–4].

Currently, approximately 4 billion tons of PG have been stored in dumps worldwide. The largest amount of PG have been stored in the USA at over 1.7 billion tons [2]. Other significant PG dumps are the ones in Brazil (150 Mt) [2, 3], Russia (140–200 Mt) [5], Spain (around 100–120 Mt) [3, 6], Ukraine (over 55 Mt) [7], Syria (30 Mt) [8], and Lithuania (15 Mt) [9].

PG is mainly composed by gypsum which content can reaches 94% and also contains impurities such as phosphate, fluoride, heavy metals and metalloids (As, Cr, Cd, Cu, Ni, Zn, etc.) [10], radionuclides (^{226}Ra, ^{210}Pb, etc.) [6, 11, 12] and rare earth elements (Sc, Y, lanthanides) [13–15].

The environmental concerns associated with phosphogypsum dumps, first of all include soil, ground and surface water contamination. One of the major source of environmental contamination can be the leachate generated by the wet stacking of PG or during precipitation.

Groundwater contamination may occur from process surface runoff from the PG dump or through the long-term downward leaching which occurs when rainwater infiltrates through the phosphogypsum dump [6].

The known cases of groundwater contamination, associated with PG dump, is fluoride, sulfate, calcium, phosphate, and trace elements or radionuclides in different countries, such as USA, Canada, Spain, and Tunisia [1, 16, 17].

The Gomel Chemical Plant (GCP), one of the largest chemical plants in Belarus, produces sulfuric and phosphoric acids, mineral salts (ammophos, superphosphate, and nitrogen-phosphorus-potassium fertilizers), aluminum fluoride, cryolite, etc. The amount of PG accumulated for more than 50 years is about 24 Mt [18], and its dumps occupy 0,91 km^2. Some of these dumps are more than 70 m high and can be either plateau-shaped or crest-shaped.

The PG at point contains up to 40% of moisture; its gypsum constituent comprises about 97%, while 3% are accounted for iron and aluminum phosphates, orthophosphoric acid, potassium and sodium fluorosilicates, and calcium fluorides. The dumpsite is a steady source of sulfates, nitrogen compounds, and phosphates that pollute surface water as well as subterranean water, and soil. Groundwater contamination represents the main problem, given that it adversely affects the water supply of the city of Gomel and neighbouring settlements.

The local monitoring of subterranean water in the vicinity of the GCP is part of a large-scale pollution monitoring system. Its main purpose is to control the fresh subterranean water conditions within the GCP influence area. The history of the subterranean water monitoring network in the GCP surrounding area is traceable over thirty years [19–22]. However, during certain periods, the observation was discontinued, and many of subterranean water observation sites have become technically unfit. Now, the monitoring data still requires its thorough analysis and interpretation, and the monitoring network itself needs optimization in accords with current requirements.

The aim of this study is to analyze the spread of technogenic contamination within subterranean water horizons at the GCP surrounding area.

Materials and methods

Subterranean water monitoring

As a rule, the highest contamination levels are observed in shallow aquifers.

Nowadays subterranean water monitoring local network in the GCP surrounding area is formed by 65 observation wells. Observation wells are the source of data about water regime (water levels, water chemical composition) of ground, submorainic and Paleogene aquifers. The stratification depth of aquifers varies from 2 to 35 m.

The research is based on geological and hydrogeological data of previous years and the results of field hydrogeological investigations carried out during the 2019–2021. They included estimation of hydrodynamics and hydrochemical parameters. For these purposes, a series of pumping-tests, water level measurements and water compositional analysis was carried out.

The data used for subterranean water quality assessment have been obtained as a result of the chemical analysis of water samples conducted either immediately at the sampling site and using portable devices (for temperature and pH) or laboratory methods in accordance with the state and international standards [23–28]. Water compositional analysis used several substances and properties as indicators of water contamination: sulphates (SO_4^{2-}), nitrogen ammonium (NH_4^+), phosphorus phosphate (PO_4^{3-}), total dissolved solids (TDS), temperature and acidity of water (pH).

Assessment of hydrogeological parameters

Assessment of aquifers' physical properties and hydrogeological parameters is based on the data of pumping tests and experimental studies.

In particular, for the pumping-test data analysis the authors used an equation by C.V. Theis and a method by C.E. Jacob. The modified Theis' equation gives the drawdown of water level at a radial distance at particular time (Eq. 1) [29]:

$$S = \frac{0.183Q}{KM} \lg \frac{2.25at}{r^2}, \qquad (1)$$

where S is the drawdown of water level at distance (r) at time (t) after the start of pumping; Q is the pumping rate, m^3/d; KM is the aquifer transmissivity (m^2/d); a is the pressure conductivity factor, m^2/d.

For data analysis by Jacob's method, the authors used a mode of time tracing of water level drawdown. The analysis involves matching a straight line to water-level displacement data as a function of the logarithm time from the beginning of pumping (Eq. 2):

$$S = f(\lg t). \qquad (2)$$

Impermeable or semi permeable layers play a significant role in protection of aquifers from technogenic contamination, which enters aquifers from the day surface. The hydrogeological parameters of impermeable layers are obtained from slug tests and hydrogeological modeling.

Vertical permeability of impermeable layers can be evaluated using vertical permeability coefficient (K_v) (Eq. 3):

$$K_v = \frac{k_0}{m_0}, \qquad (3)$$

where k_0 is the hydraulic conductivity coefficient (m/d); m_0 is the impermeable layer thickness (m).

Assessment of subterranean water contamination

Chemical compound was buildup as well as quality of subterranean water in natural conditions was investigated. There is a range of natural conditions that constitute the natural background. Subterranean water background components were used to assess the extent of technogenic impact on water quality.

In order to assess the extent of subterranean water contamination an index of water quality (K_c) was calculated (Eq. 4):

$$K_c = \frac{C_i}{C_f}, \qquad (4)$$

where C_i is the concentration of i-th substance, mg/dm^3; C_f is the background concentration of i-th substance, mg/dm^3.

Judging by K_c values thus obtained, there are three classes of subterranean water contamination:

Class 1: $K_c<3$ – insignificantly contaminated;
Class 2: $3<K_c<10$ – moderately contaminated;
Class 3: $K_c>10$ – highly contaminated.

Results and discussion

Aquifers characteristics and transmissivity mapping

The study area is part of Belarusian Polesye, a region with typical lowland landscapes, such as alluvial terraced, secondary water-glacial, lacustrine-boggy, and floodplain landscapes. The bedrock consists of many layers formed over a lengthy period. Glacial processes have had a deep impact on the regional geology. The most recently formed sediments are mostly constituted by morainic formations, covered with fluvioglacial and alluvial deposits.

As it was mentioned above, the paper describes three aquifers under the technogenic impact. Judging by the geological structure and hydrodinamic conditions, these aquifers are confined in Quaternary and Palaeogene deposits and divided by two regional impermeable layers. These aquifers are hydraulically connected and form a wide, single subterranean water system of the upper hydrodynamic zone.

The most common features of the hydrogeological regime are well correlated with the climatic conditions. More specifically, the hydrogeological regime of the territory is characterized by lower subterranean water levels in summer and autumn, while higher levels are registered in spring due to snowmelt floods and in autumn because of atmospheric precipitation. Generally, subterranean water table lowers from June to the end of January, and then slowly rises until mid-April, followed with a decrease until late May.

During the study period, subterranean water level changes from 0.81 m above grade to 7.44 m below grade. The closest to the surface subterranean water table position was found in the north-western part of the submorainic aquifer, while its deepest position was registered in the south-west of the Paleogene aquifer.

A subterranean water temperature has shown some seasonal trends. The minimum temperature ranges from 6 to 10°C in spring; thermal peaks oscillate between 11 and 13°C in August and September.

The natural subterranean water tends to have hydrocarbonate magnesium-calcium or calcium-magnesium compound; and it is mostly fresh, with mineralization ranging from 300 to 500 mg/dm^3.

The aquifer transmissivity changes at short distances due to sedimentation peculiarities and complex geological structure (down to 50 m) (Fig. 1).

Groundwater aquifer is located within middle to upper Pleistocene, Holocene and modern technogenic deposits and is 2 meters deep. It consists of various lithological types of deposits (sand, sandy loam, loam) and contains nonartesian as well as local subartesian waters. The water table is situated from 0.23 to 2.67 m below grade.

The aquifer principal feed source is atmospheric precipitation, while its secondary feed sources include leakage from underlying confined aquifers as well as seepages of the liquid phase from the PG dumps. Groundwater is drained by adjacent watercourses and ponds.

Groundwater flow has north-west and south-west gradient. Aquifer transmissivity changes from 0.1 to 5.0 m^2/d, and beneath the PG dumps it ranges from 0.5 to 4.0 m^2/d (Fig. 1).

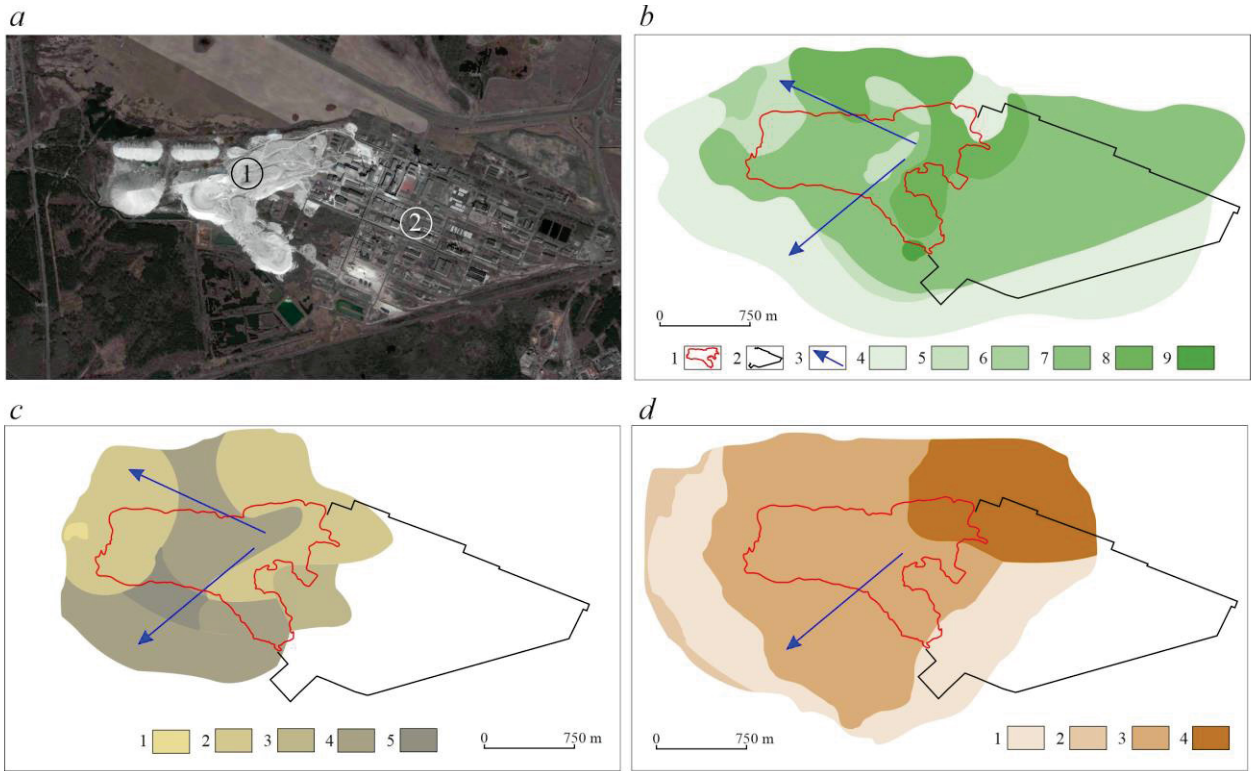

Fig. 1. *Aquifers transmissivity: a) satellite image of the study area: 1 – PG dumpsite; 2 – industrial site area; b) groundwater aquifer: 1 – PG dumpsite; 2 – industrial site area; 3 – groundwater flow; transmissivity (m^2/d): 4 – 0.1; 5 – 0.5; 6 – 1.0; 7 – 2.0; 8 – 4.3; 9 – 5.0; c) submorainic aquifer, transmissivity (m^2/d): 1 – 5.0; 2 – 30–50; 3 – 60–80; 4 – 100–130; 5 – 150; d) Paleogene aquifer, transmissivity (m^2/d): 1 – 100; 2 – 130; 3 – 200; 4 – 230*

Рис. 1. *Водопроводимость: a) спутниковый снимок: 1 – отвалы фосфогипса; 2 – промышленная площадка; b) грунтовый водоносный горизонт: 1 – отвалы фосфогипса; 2 – промышленная площадка; 3 – направление потока грунтовых вод; водопроводимость ($м^2/сут$): 4 – 0,1; 5 – 0,5; 6 – 1,0; 7 – 2,0; 8 – 4,3; 9 – 5,0; c) подморенный водоносный горизонт, водопроводимость ($м^2/сут$): 1 – 5,0; 2 – 30–50; 3 – 60–80; 4 – 100–130; 5 – 150; d) палеогеновый водоносный горизонт, водопроводимость ($м^2/сут$): 1 – 100; 2 – 130; 3 – 200; 4 – 230*

Submorainic aquifer is connected with fluvioglacial deposits of lower to middle Pleistocene and contains artesian waters. The aquifer consists of fine and medium-grained sands – sometimes argillaceous, with average thickness of 9 m. The depth of this aquifer occurrence is usually about 13–15 m. Piezometric levels range from 0.81 m above grade to 5.8 m below grade. The aquifer is recharged by rain and melt water at watersheds. Recharge rates are rising within hydrogeological openings in morainic deposits. Given the position of the piezometric surface, it can be concluded that there are two flow directions, to include south-east/north-west and north-east/south-west. Rivers and drainage canals act as this aquifer drains.

The aquifer transmissivity changes over a wide interval (from 5 to 150 m^2/d); and beneath the PG dumps, it varies from 30 to 100 m^2/d (Fig. 1).

Paleogene aquifer is confined within inequigranular, mostly fine-grained, containing artesian water sands, with their thickness of about 24 m. The aquifer top depth may reach 30–35 m. The position of piezometric levels range between 0,73 m above grade to 7,44 m below grade.

The aquifer is recharged by the leakage from upper aquifers, and its discharge occurs in the nearest watercourses. Confined groundwater flow is directed southward. Transmissivity of aquifer is variable, depending on the sorting of aquifer materials, but is generally high and ranges from 100 to 230 m^2/d, and beneath the PG dumps it varies from 200 to 230 m^2/d (Fig. 1).

Aquifers are divided by two regional impermeable horizons:

1. *Morainic impermeable horizon* consists of clay 9 m thick, and separates groundwater from submorainic aquifers.
2. *Paleogene silts* are 4 m thick and separate submorainic from Paleogene aquifer.

Vertical permeability of these horizons is very changeable, and hydrogeological openings facilitate the reach of aquifers by contaminants (Fig. 2).

Thus established, spatial variations of transmissivity and vertical permeability make various conditions the reach of aquifers by contaminants.

Assessment of spatial differentiation of contamination in aquifers

Calculated indexes of water quality (Eq. 4) clearly indicate the extent of subterranean water contamination. The background concentration of sulphates and phosphorus phosphate in subterranean water are relatively low (Table). The background concentrations of the analyzed substances were determined based on data from background wells located in the southeast direction outside the influence zone of the pollution source at a distance of 1000 m, upstream of the groundwater flow from the source of contamination. The increase of concentrations in monitoring wells is a warning sign of subterranean water chemical contamination. The difference between indexes and background content of ions in water reflects the extent of technogenic impact on background hydrochemical parameters. The degree of groundwater contamination is determined by the sum of three investigated substances (Fig. 3).

The highest concentrations of contaminants in the influence zone of the GCP are as follows: for sulphates – 1826–2880 mg/dm^3, for phosphorus phosphate – 304–585 mg/dm^3 and for nitrogen ammonium – up to 24 mg/dm^3. Total dissolved solids ranges from 3920 to 4955 mg/dm^3 [20, 30, 31].

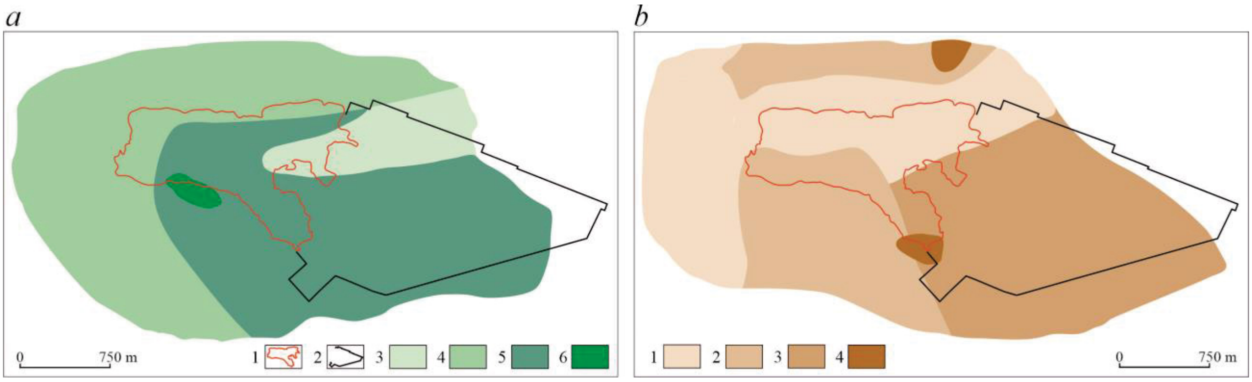

Fig. 2. *Vertical permeability of impermeable horizons: a) Morainic impermeable horizon: 1 – PG dumpsite; 2 – industrial site area; vertical permeability (day⁻¹): 3 – 0.00014–0.00016; 4 – 0.00031–0.00069; 5 – 0.0014–0.0036; 6 – 1.0 (hidrogeological openings); b) Paleogene silts, vertical permeability (day⁻¹): 1 – 0.00001–0.00008; 2 – 0.00012–0.00013; 3 – 0.00033; 4 – 1.0 (hidrogeological openings)*

Рис. 2. *Вертикальная проницаемость водоупорных отложений: а) моренные отложения: 1 – отвалы фосфогипса; 2 – промышленная площадка; вертикальная проницаемость (сут⁻¹): 3 – 0,00014–0,00016; 4 – 0,00031–0,00069; 5 – 0,0014–0,0036; 6 – 1,0 (гидрогеологические окна); b) алевриты палеогена, вертикальная проницаемость (сут⁻¹): 1 – 0,00001–0,00008; 2 – 0,00012–0,00013; 3 – 0,00033; 4 – 1,0 (гидрогеологические окна)*

Groundwater aquifer. High contamination level (K_c>10) of groundwater is found in wells sited next to the PG dumps, and as deep as the groundwater flow gradient. Most frequent contaminants are sulphates, with their concentration 100 times higher (K_c=15–170) than the background value (Fig. 3, Table). This testifies about high migration directly from the dumps of SO_4^{2-}-ion and its massive percolation in the groundwater aquifer. It is especially true for phosphorus phosphate (K_c=18–30000), and nitrogen ammonium (K_c=14–9000).

Table. *Average background concentrations of several chemical substances and properties of subterranean water for 2019–2021*

Таблица. *Средние фоновые концентрации некоторых химических соединений и свойства подземных вод за 2019–2021 гг.*

Aquifer Водоносный горизонт	pH	TDS/Минерализация	SO_4^{2-}	NH_4^+	PO_4^{3-}
		mg/dm³ мг/дм³			
Ground Грунтовый	8,1	290	17,0	1,3	0,009
Submorainic Подморенный	7,4	612	264,0	1,2	0,01
Paleogen Палеогеновый	7,6	172	11,4	1,2	0,07

Apart from sulphates, another basic contaminant is phosphorus phosphate. Its maximum concentrations are found next to the dumps, while they decrease 500 times and more at some distance from the dumps. The low migration mobility of phosphorus phosphate is remarkable. It is caused by absorption by both plant rootage and organic matter, and is due to chemical reactions that fix it in the illuvial horizon.

Submorainic aquifer. The high-level contamination of the submorainic aquifer is attributable to abnormally high level of sulphates (K_c=11–95) and nitrogen ammonium (K_c=12–250). Contaminants are found in wells situated next to the PG dumps (Fig. 3).

The contamination level in the submorainic aquifer is much lower and spatially differentiated than in the groundwater aquifer. It is associated with spatial variability of thickness and mineralogical heterogeneity of the morainic horizon as well as changeable permeability. High transmissivity within the submorainic aquifer leads to lateral migration of contaminants and their distribution along subterranean water flow paths.

Paleogene aquifer. The main contaminant of the aquifer is SO_4^{2-}-ion (K_c=10–70). Contamination is found in wells situated next to some preexisting (for 30 years) dumps characterized by long-term contamination. Decreasing downwards contamination is caused by high vertical hydraulic conductivity. Furthermore, contamination by SO_4^{2-}-ion (K_c=4–6) is found in wells situated along the subterranean water flow and testifies on high lateral migration of this ion.

Thus, as it is prognosed, the highest level and density of contamination is found in the groundwater aquifer and is caused by insufficient insulation of the groundwater aquifer from percolation of contaminants from the ground surface (Fig. 4). In both submorainic and Paleogene aquifers, contamination is reduced considerably.

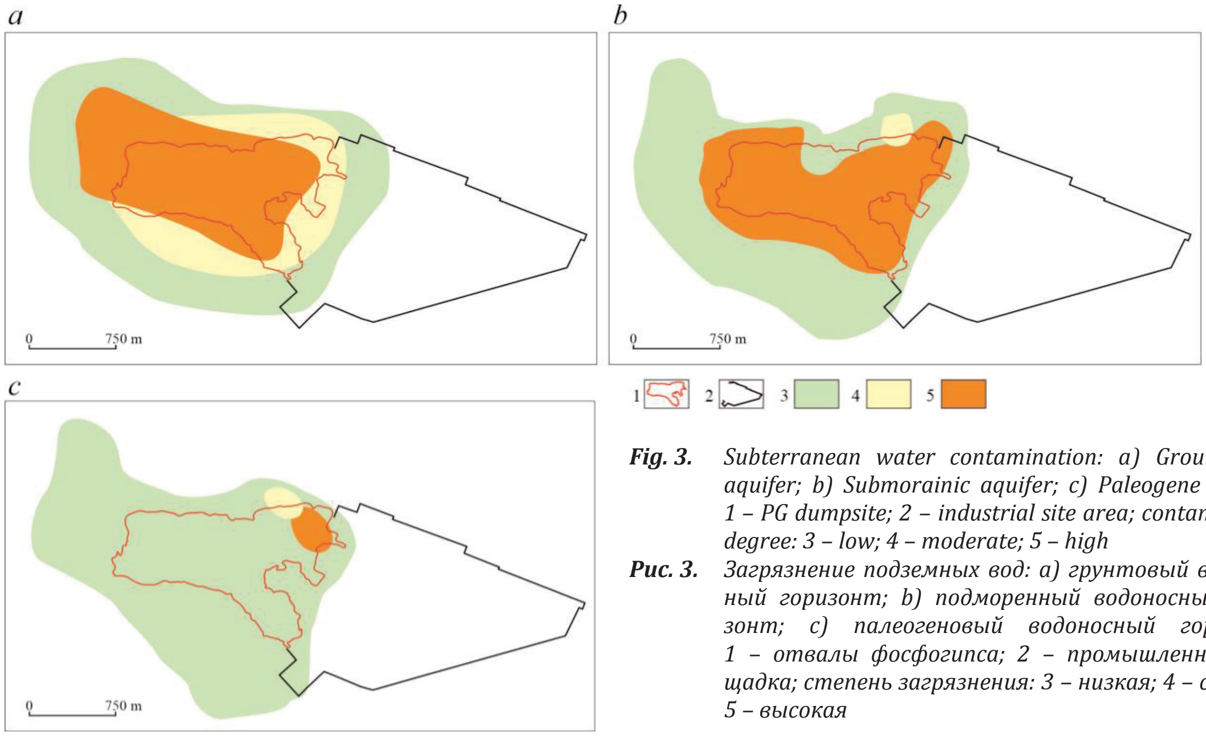

Fig. 3. *Subterranean water contamination: a) Groundwater aquifer; b) Submorainic aquifer; c) Paleogene aquifer; 1 – PG dumpsite; 2 – industrial site area; contamination degree: 3 – low; 4 – moderate; 5 – high*

Рис. 3. *Загрязнение подземных вод: a) грунтовый водоносный горизонт; b) подморенный водоносный горизонт; c) палеогеновый водоносный горизонт; 1 – отвалы фосфогипса; 2 – промышленная площадка; степень загрязнения: 3 – низкая; 4 – средняя; 5 – высокая*

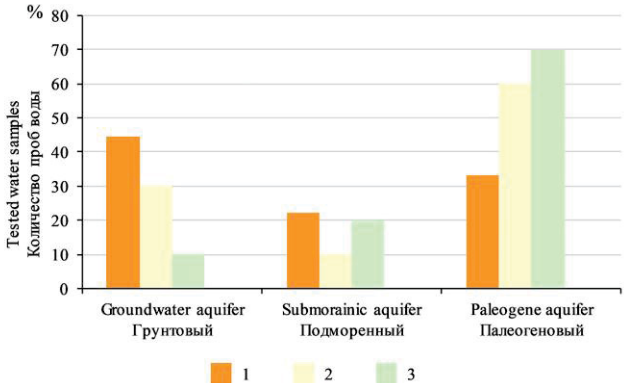

Fig. 4. *Water quality in observation wells: degree of contamination: 1 – high; 2 – moderate; 3 – low*

Рис. 4. *Качество воды в наблюдательных скважинах: степень загрязнения: 1 – высокая; 2 – средняя; 3 – низкая*

Contamination has local distribution next to the PG dumps. A tendency of either contamination increase with time or its shifting to peripheral part is not revealed. A periodical seasonal fluctuation of concentrations of some contaminated substances is established.

Conclusions

Research results point to the presence of numerous subterranean water areas with varying contamination levels that take place in and at some distance from the industrial site of the Gomel Chemical Plant.

Contamination of the aquifers is caused by the arrival of contaminants as a result of surface and subsurface runoffs and groundwater swell.

The main contaminant of the aquifers is SO_4^{2-}-ion. However, the extremely high-level contamination of aquifers is formed by nitrogen ammonium and phosphorus phosphate.

The groundwater aquifer is most severely contaminated, and the amount of some of its components is hundreds to thousands times higher than the background concentrations. Significant contamination levels for the submorainic aquifer are also established. The chemical composition of the Paleogene aquifer does not show any serious variability, except for some extra sulphates and phosphorus phosphate.

Migration of contaminants depends on hydrogeological conditions and physical as well as chemical properties of contaminants themselves.

As a rule, extremely high-level contamination is found next to the relatively new (less than 15 years) dumps in the site western and eastern parts. Concentrations of contaminants in subterranean water decrease at some distance from the dumps or next to the old dumps.

The influence of the PG on water quality remains significant but is localized within the dumpsite area.

Our research shows that it is the necessary to further monitor water conditions in the area of the GCP, because some additional analysis and information are needed to make for decisions concerning this territory.

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