PROGNOSTICATION OF GROUNDWATER CONTAMINATION CAUSED BY OLD SANITARY LANDFILLS – PART I. MODELS OF THE MASS TRANSPORT OF POLLUTANTS THROUGH THE UNSATURATED ZONE

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ABSTRACT. Sanitary landfills are one of the most common sources of environmental contamination that have accompanied humans from ancient times to the present day. An important task in the complex assessment of their impact is to acquire long-term prognostications of groundwater contamination by landfill leachate. A general approach is proposed to resolve it, involving the consecutive development of mathematical models of the mass transport of pollutants in the unsaturated and saturated zones. Its applicability is illustrated by a specific example for the region of the old sanitary landfill near Haskovo, Bulgaria. The VS2DI computer program was applied in order to develop two-dimensional models of the conditions for passage of liquid emissions from the landfill body through the bottom isolating layer and the unsaturated zone. These models were used for investigating the behavior of highly mobile and less mobile pollutants on the example of chloride and ammonium ions (Cl^- and NH_4^+). The scheme of convective-diffusion mass transport is employed, taking into account reversible elimination (sorption-desorption), mechanical dispersion and mixing. Based on the obtained model solutions, the impact during the period of operation of the landfill was reconstructed. In addition, a medium-term forecast of the extent and degree of contamination in the unsaturated zone is developed and the concentration of pollutants in the percolation flux towards the saturated zone is estimated.

Keywords: groundwater contamination, unsaturated zone, sanitary landfills, solute transport models

ПРОГНОЗИРАНЕ НА ЗАМЪРСЯВАНЕТО НА ПОДЗЕМНИТЕ ВОДИ ОТ СТАРИ ДЕПА ЗА БИТОВИ ОТПАДЪЦИ – ЧАСТ I. МОДЕЛИ НА ДВИЖЕНИЕТО НА ЗАМЪРСИТЕЛИ ПРЕЗ НЕНАСИТЕНАТА ЗОНА

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РЕЗЮМЕ. Депата за битови отпадъци са едни от най-разпространените източници на замърсяване на околната среда, които съпътстват човека от древността до наши дни. Важна задача при комплексната оценка на тяхното въздействие е изготвянето на дългосрочни прогнози за възможно замърсяване на подземните води със сметищен инфилтрат. За нейното решаване е предложен общ подход, включващ последователно разработване на математически модели на движението на замърсители в ненаситена и водонаситена среда. Неговата приложимост е илюстрирана с конкретен пример за района на старото депо за отпадъци на гр. Хасково, България. Посредством компютърна програма VS2DI са съставени двумерни модели на условията за преминаване на течни емисии от сметищното тяло през защитния екран и зоната на аерация, с които е изследвано поведението на силно подвижни и слабо подвижни замърсители по примера на хлоридните и амониеви йони (Cl^- и NH_4^+). Използвана е схемата на конвективно-дифузионен пренос на вещество, с отчитане на обратимото елиминиране (сорбция-десорбция), механичната дисперсия и смесването. На базата на моделните решения е направена реконструкция на въздействието в периода на експлоатация на депото, а след неговото закриване и средносрочна прогноза за обхвата и степента на замърсяване в ненаситената зона и концентрацията на замърсители в подхранващия подземните води инфилтрационен поток.

Ключови думи: замърсяване на подземните води, ненаситена зона, депа за битови отпадъци, миграционни модели

Introduction

Sanitary landfills as a major source of contamination are an important and very relevant topic in the discussions on the protection of global water resources. The study and the search for solutions to reduce their harmful effects is a worldwide problem considering their wide distribution, enduring action and the serious damage they cause to all elements of the environment – air, water and soil (Christensen et al., 1989; Montgomery, 2019; Vaverková, 2019). Part of the complex assessment of the problem is the development of long-term prognostications for the extent, degree and period of groundwater contamination caused by old sanitary landfills. Our experience in this sphere shows that the most effective tools for making these forecasts are the mathematical models of the mass transport of pollutants in the unsaturated and saturated zones. The applicability of the proposed general approach will be demonstrated by an example for the region of the old sanitary landfill near Haskovo, Southern Bulgaria. The two-dimensional models of the conditions for passage of liquid emissions from the landfill body through the bottom isolation layer and the unsaturated zone, presented in this first part of the article, are developed using the computer program VS2DI (Healy, 1990). The acquired model solutions were used for investigating the fate and transport of highly mobile and less mobile pollutants on the example of chloride and ammonium ions (Cl^- and NH_4^+) during the operation of the landfill and in the period after its closure.

General approach for modeling of groundwater contamination caused by sanitary landfills

Selection of key pollutants

The contamination models usually do not include all the pollutants contained in the landfill leachate, as this is usually not necessary and the additional costs required cannot be compensated by a higher quality of the end result. For that reason, from the many pollutants in the contamination source (emitter), two or more key pollutants are selected, with which different conservative (critical) scenarios are reproduced in the models. The main selection criteria are the pollutant concentration in the emitter; the pollutant mobility in the unsaturated and saturated zones; the pollutant ability to affect the geochemical barriers; its harmful effects on human health and the environment. The key pollutants must represent the maximum extent of subsurface contamination and the most intense (toxic) pollution around the source. From this point of view, highly mobile, with low sorption potential, pollutants (chlorides, sulphates, some organic compounds such as TCE, PCE, etc.) and less mobile, with high sorption potential, contaminants (ammonium ions, heavy metals, etc.) are an adequate selection (Fatta et al., 1999; Kjeldsen et al., 2002; Christensen et al., 1989).

Contaminant migration field and its components

The contaminant migration field by definition includes this part of the subsurface in which the pollutants from the landfill advance to the first recipient – the nearest river or surface water body (Stoyanov, 2019). Pollution models should cover the three main components of this field: (1) engineering barriers at the landfill bottom – isolating layers; (2) unsaturated zone – from the last engineering barrier to the groundwater level; (3) saturated zone – potentially endangered aquifers or aquifer complexes. Formally, the engineering barriers are part of the unsaturated zone.

Modelling techniques

The starting positions in the proposed general concept for modeling of groundwater contamination caused by old sanitary landfills are: (1) The mass-transport of key pollutants through the unsaturated zone and the engineering barriers (if any) is simulated with 2D models, and their subsequent spreading in groundwater – with 3D models. The obtained two-dimensional solutions are used as input data in the 3D models. (*Remark: The development of complex 3D models, including all components of the contaminant migration field, is difficult to apply due to the fact that computer simulation in case of long-term predictions an extremely time-consuming process. At the same time, the separate simulation of the processes in the unsaturated and saturated zones does not a priori reduce the solutions accuracy, on the contrary – it improves the stability of the mathematical models and makes the prognostications more detailed.*) (2) The 2D model area covers sections along lines passing through the central part of the landfill body in the direction of the underground flow, and the 3D one – the subsurface from the emitter to the first recipient. (3) The models are deterministic. The hydrogeological units and the engineering barriers are simulated as two-dimensional or three-dimensional objects according to their main characteristics: geometric (thickness, length, extension), physical (density, porosity), hydrodynamic (hydraulic conductivity) and mass transport ones (distribution, dispersion and molecular diffusion coefficients). In the 2D models are additionally set parameters specific for the unsaturated zone: natural moisture content, coefficient of moisture conductivity, etc. (4) The groundwater recharge by infiltration and percolation is set in accordance with the climatic, geologic and technogenic conditions. (5) The mechanism of the mass transport of key pollutants is in its full form: convective and diffusion transport, accompanied by mechanical dispersion, reversible elimination (sorption-desorption), irreversible elimination (radioactive decay and biodegradation) and mixing. (6) The flow through the engineering barriers and the input concentrations of the studied key pollutants are set as variables taking into account the specific technogenic conditions. (7) The models simulate the fate and transport of selected key pollutants (chlorides, sulfates, ammonium ions, heavy metals, etc.) and the target is to achieve long-term predictions – forecast period of up to 100 years. (8) The results of the model studies are presented as vertical and horizontal sections of the concentration field at different points in time.

Modelling tools

Standard computer programs are used, in whose algorithms are implemented numerical solutions of the equations of moisture and mass transport in the unsaturated zone and the flow and mass transport equations in a water-saturated environment. The two-dimensional models of contaminant transport across the engineering barriers and the unsaturated zone are compiled using the VS2DTI package from the computer program VS2DI (Healy, 1990, Hsieh et al., 2000). The three-dimensional models of the pollutant fate and transport in the saturated zone are developed using the computer programs Modflow and MT3D-MS (Anderson et al., 2015; McDonald and Harbaugh, 1988; Stoyanov, 2019; Zheng and Bennet, 2002).

Characteristics of the study area

The applicability of the presented general approach will be illustrated with models of groundwater contamination caused by the old sanitary landfill near the town of Haskovo (Fig. 1). The studied region is situated in the northern periphery of the Haskovo valley in the catchment area of the Gidikli dere River, a right tributary of the Banska River. The geological section is dominated by Paleogene volcano-sedimentary rocks (latites and pyroclastic rocks), partially covered by a 1-2 m thick clay layer. Down to a depth of 60-70 m the rocks are fractured, differently weathered, in places decomposed to clay.

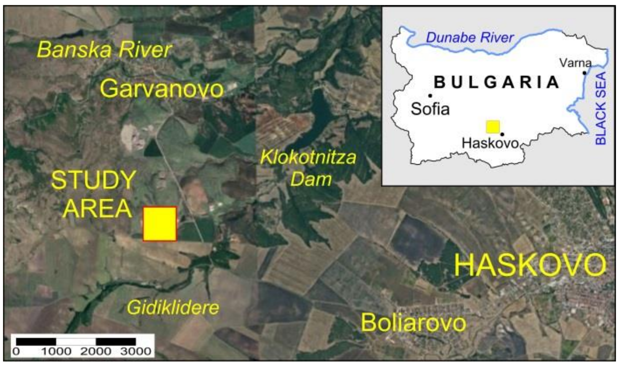

Fig. 1. Location of study area

The main hydrogeological unit in the region is the Paleogene aquifer complex, which is very heterogeneous and is characterized by relatively low water permeability. Based on the degree of secondary alteration, several hydrogeological units of lower rank are separated in it. The hydraulic conductivity most often varies between 0,005 and 1,5 m/d. The aquifer complex is unconfined and vulnerable to surface pollutants. The groundwater recharge occurs chiefly from infiltration of rainfall and snowmelt. The general direction of groundwater flow is to the southeast – towards Gidikli dere River. The average hydraulic gradient is 0,02, about 0,012 in the sanitary landfill area. The modulus of groundwater discharge is less than 0,1 (l/s)/кm², i.e. the groundwater resources are insignificant. The sanitary landfill is located in the upper part of a ravine, which in the period 1997/2015 was gradually filled with waste materials. The engineering barrier designed to provide leachate control is a layer of compacted clay laid down under landfill cell.

Key pollutants for the study area are chloride and ammonium ions (Cl^- and NH_4^+). They are selected on the basis of continuous monitoring of landfill leachate and groundwater chemical composition.

Methodology. Development of models of pollutant transport in the unsaturated zone

The two-dimensional models M2D-CL and M2D-NH4, designed for studying the behavior of the key pollutants Cl^- and NH_4^+ in the unsaturated zone, were compiled with the program VS2DI. The model area covers the section underneath the landfill cell along profile I-I, passing lengthwise the axis of the ravine (Fig. 2).

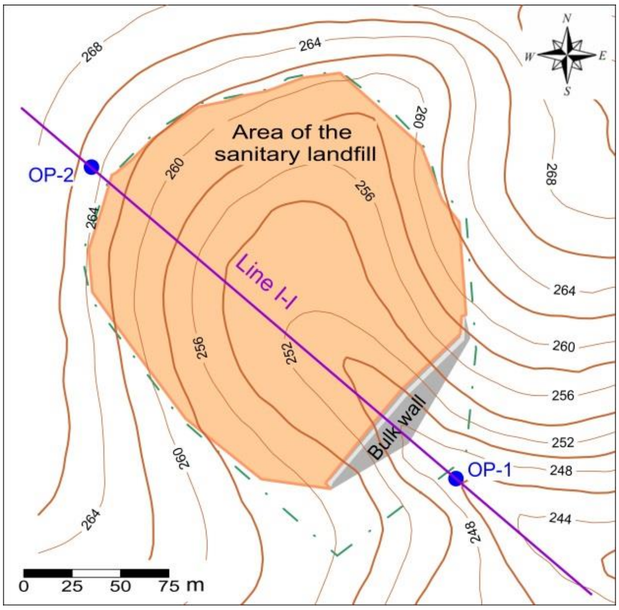

Fig. 2. Boundaries of the landfill as of 2015. Topographic map of the landfill area

In the section, down to a depth of 90 m, are separated two hydrogeological units, which together with the engineering barrier (Table 1) are set in the models as zones with corresponding hydrogeological parameters - total porosity n, bulk density ρ_d, hydraulic conductivity k, longitudinal dispersion α_L, diffusion coefficient D_M and distribution (partition) coefficient K_D (Fig. 3 and Table 2). The van Genuchten function is used to model the relations between pressure head, moisture content, and relative hydraulic conductivity. The time of the computer simulation, including the period of landfill operation 1998/2015 and the period after its cessation until 2025, is divided into 27 stress periods, each lasting 1 year.

Table 1. *Mass transport field components, low-rank hydrogeological units and model zones*

Component	Hydrogeological unit	Model zone
Engineering barrier	Layer of compacted clay	MZ 1
Unsaturated zone	Layer of low permeability (diluvial clay)	MZ 2
	Zone of low permeability (rock complex)	MZ 3

Table 2. *Hydrogeological parameters for the model zones*

Hydrogeological parameter	Model zone		
	MZ 1	MZ 2	MZ 3
n, dimensionless	0,32	0,35	0,36
ρ_d, g/cm³	1,61	1,74	1,78
k, m/d	2,6.10⁻⁵	0,05	0,25
α_L, m	0,55	2,50	10,00
D_M, m²/d	5,0.10⁻⁴	4,5.10⁻⁴	4,0.10⁻⁴
K_D for Cl^-, cm³/g	0,70	0,50	0,20
K_D for NH_4^+, cm³/g	4,50	1,50	1,10

Remark: The values of n, ρ_d, k, α_L, as well as the values of K_D for Cl^- are determined according to data from slug tests and tracer tests conducted by the authors (Stoyanov et al., 2010; Stoyanov, 2010), and the values of D_M and K_D for NH_4^+ are determined from literature data for a similar soils (Spitz and Moreno, 1996).

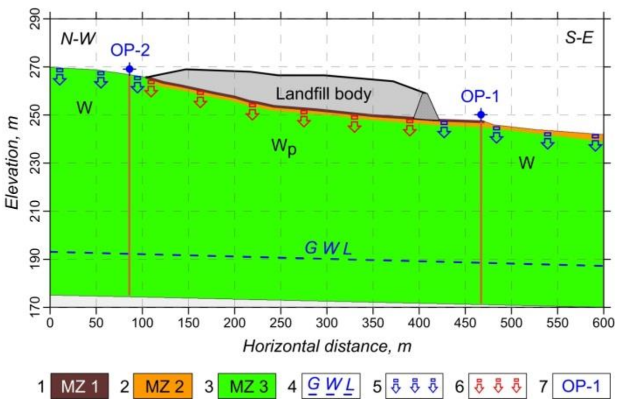

Fig. 3. Conceptual framework of the two-dimensional models
1, 2 and 3 – Boundaries of the model zones, 4 – Groundwater level; 5 – Rainfall infiltration; 6 – Leachate infiltration; 7 – Monitoring observation points.

The groundwater level is set as specified total head at a depth of 65-80 m (Fig. 3). The landfill body is set as a line source of pollution, the length of which increases according to the rate of its expansion (Fig. 4). The leachate is simulated as specified flux characterized by the calculated for each stress period average values of the infiltration rate W_p and the concentrations of Cl^- and NH_4^+ in them – C_{Cl} and C_{NH4} (Table 3). The values of W_p are presumed on the basis of the landfill water balances for the respective time intervals, determined by the average annual values of precipitation, air temperature and evaporation provided by Hydro-meteorological observation station Haskovo (Table 4), taking into account the water permeability of the surface layer (Bredenkamp, 1990).

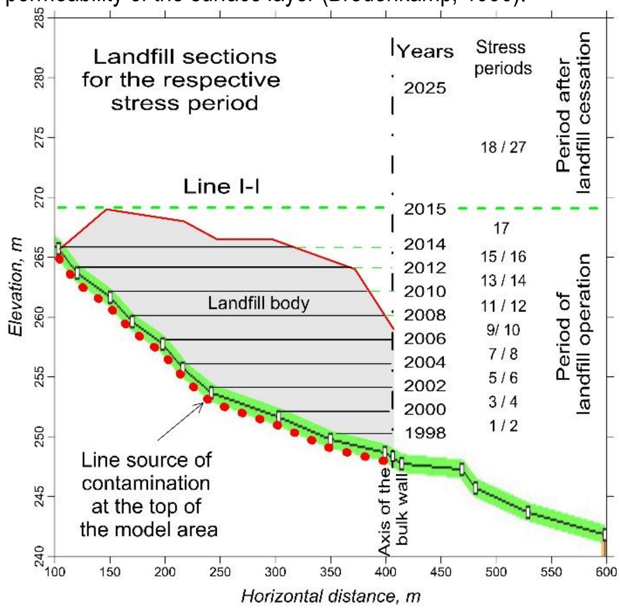

Fig. 4. Scheme of waste disposal along profile I-I. Length of the line source of contamination at the top boundary of the two-dimensional models in the different stress periods.

Table 3. *Average values of W_p, C_{Cl} u C_{NH4} for the landfill leachate*

Time interval	Stress period	W_p, m/d	C_{Cl} mg/l	C_{NH4} mg/l
1998-2006	1/8	$4,6.10^{-4}$	1850	700
2007	9	$5,9.10^{-4}$	1850	700
2008	10	$6,3.10^{-5}$	1850	700
2009	11	$3,5.10^{-4}$	1850	515
2010	12	$6,2.10^{-4}$	1850	460
2011	13	$3,2.10^{-4}$	2000	350
2012	14	$8,3.10^{-4}$	1300	85
2013	15	$3,0.10^{-4}$	1860	700
2014	16	$1,2.10^{-3}$	1820	85
2015	17	$3,7.10^{-4}$	600	450
2016-2025	18/27	$5,0.10^{-4}$	1400	120

Table 4. *Average annual values of precipitation P, air temperature T and evaporation E*

Observation period	Precipitation P, mm	Air temperature T, ^{0}C	Evaporation E, mm
1956-2006	668,0	12,5	501,0
2007	790,1	14,1	574,1
2008	372,1	13.6	349,0
2009	621,1	13,2	491,7
2010	785,1	13,4	557,8
2011	580,4	12,4	462,0
2012	892,0	13,4	588,8
2013	597,4	13,6	486,4
2014	1059,9	13,7	634,6
2015	647,0	14,1	517,0
2007-2015	724,0	13,5	538,7

Remark: Data provided by Hydro-meteorological observation station Haskovo

The values of C_{Cl} and C_{NH4} are deduced according to data from the monitoring of landfill leachate chemical composition. The groundwater recharge from rainfall is set along the entire length of the profile with an average for the period infiltration rate W = $2,0.10^{-5}$ m/d and zero concentrations of Cl^- and NH_4^+. The models are calibrated to the registered concentration values in the monitoring points by varying the values of k, D_M, α_L and W_p.

Results and discussion

The results of the computer simulations of the mass transport of highly mobile and less mobile pollutants through the unsaturated zone and their subsequent passage into the saturated zone are illustrated with the model solutions presented in Fig. 5 and Fig. 6. They display the extent and degree of subsurface and groundwater contamination in different periods of the landfill operation (2005, 2010 and 2015) and demonstrate the acquired forecast for the development of these negative processes by 2025.

During landfill operation, the following more important patterns in the behavior of the key pollutants used in the simulations are observed: *Chloride ions* are highly mobile, so the boundaries of the region, contaminated by them, are dynamic and quickly cover large subsurface areas. By 2012 they reach the groundwater level beneath the oldest part of the sanitary landfill. Here the unsaturated zone is the most polluted, and the Cl^- concentration down to a depth of 25-30 m is around and above 1000 mg/l. These high values are an indirect sign for the presence of some non-sorbent organic compounds (TCE, PCE, phenol, etc.) part of which are very toxic. The Cl^- concentration in the contaminated infiltration flux that enters groundwater is in the range of 200-250 mg/l. At the end of the operation period, the highly mobile pollutants spread out over a distance of 130-150 m from landfill's southeastern border. *Ammonium ions* have high sorption potential and are principally retained by the engineering barrier and the diluvial clays. They migrate very slowly and contaminate more limited subsurface parts. The most polluted by them areas in the unsaturated zone are down to a depth of 20-25 m beneath the old part of the landfill and down to 10-15 m under the new parts. In these zones the NH_4^+ concentration is 150-200 mg/l, and in depth it decreases very rapidly. At the end of the operation period, the front of the zone of NH_4^+ contamination reaches a depth of 45-50 m underneath the old part of the landfill and laterally spreads out over a distance of 10 m from its southeastern border. In a very limited area, small quantities of ammonium ions infiltrate to the groundwater level, affecting a minor part of the saturated zone, where their concentration is not more than 2-3 mg/l. Due to their very low mobility, NH_4^+ ions have only indicative value for localizing the most intensive pollution. Their presence in the already contaminated areas is associated with the manifestation of many other pollutants including heavy metals, organic compounds, etc.

After the cessation of the sanitary landfill operation, the medium-term forecast for the fate and transport of pollutants in the unsaturated zone and groundwater, based on the model solutions – Fig. 5-D and Fig. 6-D, in short is the following. The concentrations of the various components in the composition of the leachate generated by the old landfill will gradually decrease. The concentration of key pollutants in the liquid emissions infiltrating beneath the landfill will also decrease.

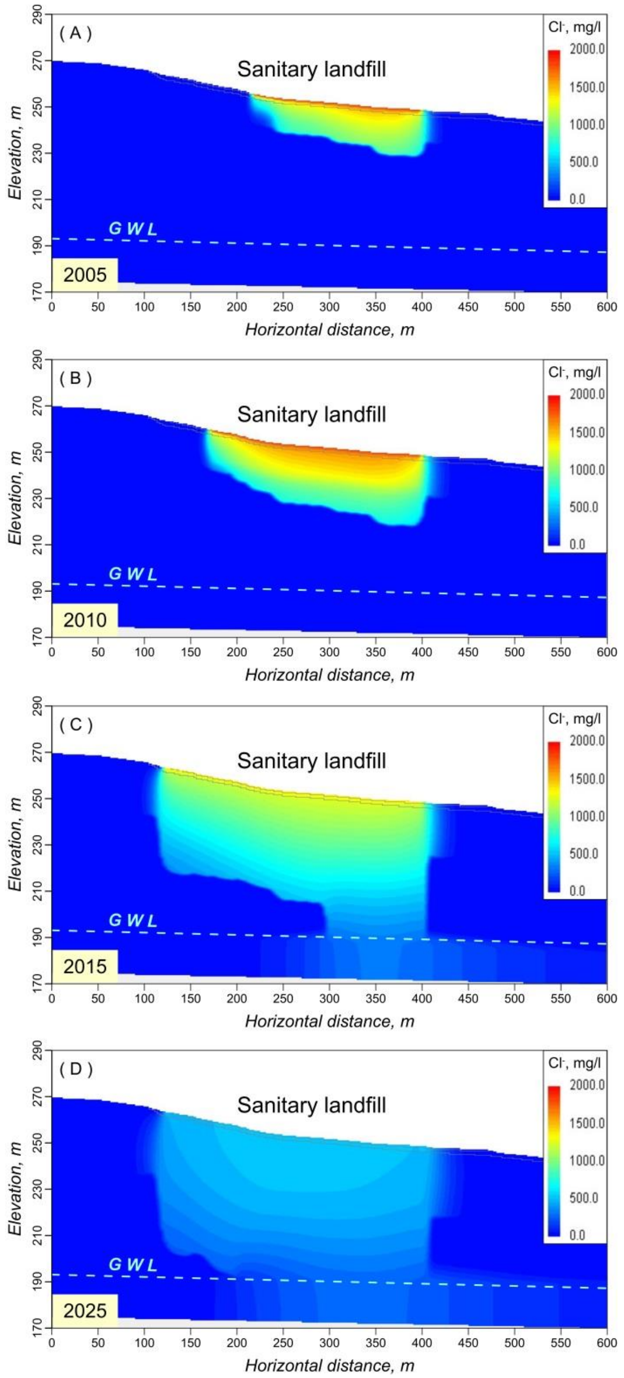

Fig. 5. Model M2D-CL. Distribution of highly mobile pollutants on the example of Cl^-.
(A), (B) and (C) – model solutions for different periods of landfill operation, respectively at 2005, 2010 and 2015, (D) – model solution after landfill cessation, prognosis for 2025.

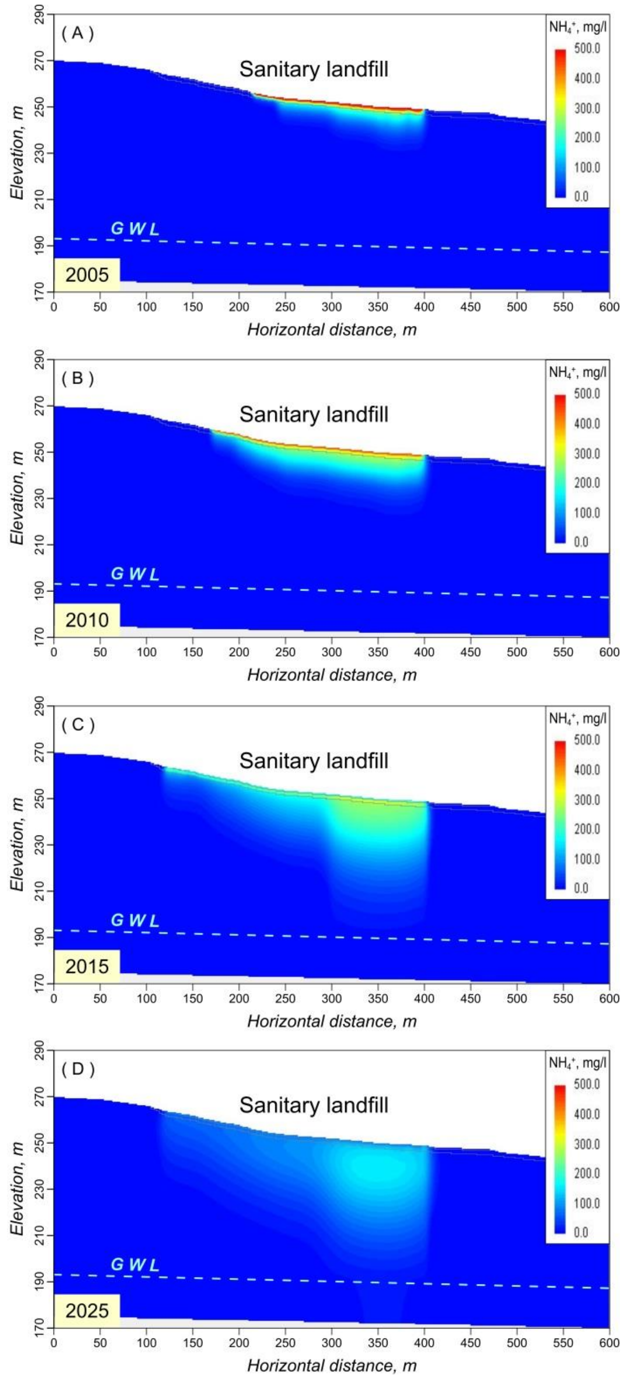

Fig. 6. Model M2D-NH4. Distribution of less mobile pollutants on the example of NH_4^+.
(A), (B) and (C) – model solutions for different periods of landfill operation, respectively at 2005, 2010 and 2015, (D) – model solution after landfill cessation, prognosis for 2025.

The spatial boundaries of the polluted zones, although slower, will continue to expand, while the concentrations in the most contaminated parts of these areas will progressively decline. At the end of the forecast period by 2025, the Cl^- concentration in the most polluted parts of the unsaturated zone will be between 350 and 650 mg/l, and the NH_4^+ concentration will not be higher than 100-180 mg/l. At the same time, the contaminated infiltration flux will continue to enter groundwater through larger areas. The Cl^- concentration in it will be between 200 and 350 mg/l, and the NH_4^+ concentration will not exceed 7-8 mg/l. The groundwater contamination by highly mobile pollutants will expand to about 150-200 m southeast of the landfill, and the contamination by less mobile pollutants – to no more than 50-60 m.

The performed model studies of the fate and transport of pollutants in the unsaturated zone show that the low water permeability characteristics of the rock massif and the covering diluvial clays are a good natural barrier that minimize the leachate infiltration beneath the bottom of the sanitary landfill. The layer of compacted clay (the engineering barrier) laid down under landfill cell further limits the possibilities for environmental pollution.

Conclusion

Using two-dimensional models, a computer simulation of the mass transport of selected key pollutants through the unsaturated zone is performed for the region of the old sanitary landfill near Haskovo. The concentrations of contaminants in the percolation flux towards the saturated zone are estimated for the period of operation, as well as for ten years after landfill cessation. The obtained solutions show that the landfill operation is accompanied by a local and low level subsurface contamination caused by the leachate passage through the compacted clay layer laid down under landfill cell. According to the developed prognostic models, assuming that the old sanitary landfill continues to emit pollutants, although with lower concentrations, the polluted zones will continue to expand their spread with gradually declining concentrations in the most contaminated parts of these areas. The obtained solutions for the transit time of pollutants to groundwater level and their concentration in the contaminated percolation flux towards the saturated zone are presented in the second part of the article, where they are used as input data in the three-dimensional models.

The developed two-dimensional models provide an opportunity to quantify the effectiveness of engineering barriers that restrict the free flow of leachate from old landfills, as well as to identify measures for improving their performance. The obtained results are of practical importance as they provide a prognostic evaluation of the possible ecological consequences in the studied area. The presented methodological approach can be successfully implemented in solving other similar hydrogeological problems.

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