PROGNOSTICATION OF GROUNDWATER CONTAMINATION CAUSED BY OLD SANITARY LANDFILLS – PART II. MODELS OF THE MASS TRANSPORT OF POLLUTANTS IN THE SATURATED ZONE

Nikolay Stoyanov [1], Stefan Dimovski [1]

[1] University of Mining and Geology "St. Ivan Rilski", 1700 Sofia; nts@mgu.bg; dimovski@mgu.bg

ABSTRACT. In Part I were described a general approach for prognostications of groundwater contamination caused by old sanitary landfills, as well as the developed models of the contamination in the unsaturated zone on the example of the old landfill in Haskovo, Bulgaria. Part II presents the composed, according to general approach, three-dimensional models of the conditions for mass transport of pollutants in the saturated zone through which a long-term prediction of groundwater contamination for a period of 100 years is acquired. A numerical three-dimensional flow model for the vulnerable to pollution aquifer complex was compiled using the Modflow computer program. Based on the flow model and taking into account the two-dimensional model solutions, applying computer program MT3D-MS, two 3D mass transport models were developed, which simulate the behavior of highly mobile and less mobile pollutants on the example of Cl- and NH_4^+. For the modeling of mass transport in the three-dimensional models, as in the two-dimensional models, the convective-diffusion scheme and the accompanying processes of reversible elimination, mechanical dispersion and mixing were used. The first mass transport model estimates the maximum spread of the groundwater contaminated area, and the second model determines the boundaries of the critical high contamination area, in which many and often toxic pollutants are expected to be present.

Keywords: groundwater contamination, saturated zone, sanitary landfills, flow models, mass transport models

ПРОГНОЗИРАНЕ НА ЗАМЪРСЯВАНЕТО НА ПОДЗЕМНИТЕ ВОДИ ОТ СТАРИ ДЕПА ЗА БИТОВИ ОТПАДЪЦИ – ЧАСТ II. МОДЕЛИ НА РАЗПРОСТРАНЕНИЕТО НА ЗАМЪРСИТЕЛИТЕ ВЪВ ВОДОНАСИТЕНАТА ЗОНА

Николай Стоянов, Стефан Димовски

Минно-геоложки университет "Св. Иван Рилски", 1700 София

РЕЗЮМЕ. В Част I бяха описани един общ подход за прогнозиране замърсяването на подземните води от стари депа за отпадъци и модели на замърсяването в ненаситената зона по примера на старото депо на гр. Хасково, България. Част II представя съставените по общ подход тримерни модели на условията за движение на замърсители във водоноситената зона, посредством които е направена дългосрочна прогноза за замърсяването на подземните води за период от 100 години. Посредством компютърна програма Modflow е съставен филтрационен тримерен модел на застрашения от замърсяване водоносен комплекс. На базата на филтрационния модел и при отчитане на двумерните моделни решения с компютърна програма MT3D-MS са съставени два миграционни тримерни модела, с които е симулирано поведението на силно подвижните и слабоподвижните замърсители по примера на Cl^- и NH_4^+. За моделиране на миграцията в тримерните модели, както в двумерните, е използвана конвективно-дифузионна схема и съпътстващите я процеси на обратимо елиминиране, механична дисперсия и смесване. С първия миграционен модел е направена прогноза за максималния обхват на зоната със замърсени подземни води, а с втория модел са определени границите на зоната с критично високо замърсяване, в която се очаква да присъстват много на брой и често токсични замърсители.

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

Introduction

Following global trends, various software products are used in Bulgaria to model the fate and transport of various pollutants in the saturated zone – Modflow, MT3D-MS, Feflow, Hidrus, etc. (Benderev et al., 2015; Gerginov et al., 2017; Stoyanov and Dimovski, 2004; Stoyanov, 2007, 2012; Stoyanov et al., 2015, 2018, 2019). In the first part is presented the concept of one very effective approach for quantitative assessment and prediction of groundwater contamination caused by old sanitary landfills. A computer simulation of the mass transport of leachate pollutants through the engineering barriers and the unsaturated zone for the period of operation, as well as for ten years after the cessation of the sanitary landfill nearby Haskovo is performed implementing 2D models. These models are used for studying the behavior of highly mobile and less mobile pollutants on the example of chloride and ammonium ions. On this basis, an evaluation and a medium-term forecast for the transit time of pollutants to groundwater level and their concentration in the contaminated percolation flux towards the saturated zone are made. Three-dimensional flow and mass transport models of the fate and behavior of pollutants in the Paleogene aquifer are developed in order to predict the following development of the processes of groundwater contamination for a longer period (100 years) after the landfill cessation. The three-dimensional models are compiled using computer programs Modflow and MT3D-MS. The obtained solutions from the 2D models are employed as input data. Applying 3D mass transport models, a computer simulation of the distribution of Cl^- and NH_4^+ is developed. Subsequently, a long-term forecast for the size and degree of groundwater contamination in the Paleogene aquifer complex, caused by the old sanitary landfill near Haskovo, is acquired.

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

The mathematical model studies for assessment and prognostication of contamination processes in the saturated zone include two main problems – the hydrodynamic field and the mass transport one (Anderson et al., 2015; Stoyanov, 2019). Standard hydrogeological software is used to develop 3D numerical models. To solve the first problem, a flow model (FM3D) of the Paleogene aquifer in the area of the studied site is compiled using the computer program Modflow (McDonald and Harbaugh, 1988). The second problem is solved with mass transport models (M3D-CL M3D-NH4) of the conditions for distribution of Cl^- and NH_4^+. These models are developed implementing the basic flow model and applying the computer program MT3D-MS (Zheng and Bennet, 2002).

Conceptual model of the pollution source

The source of contamination in the saturated zone is the percolation flux entering through the entire area of the projection of the landfill on the groundwater table, marking the boundary between the unsaturated and the saturated zone. The conservative scenario accepts the simplified assumption that the infiltration rate over the entire source area is constant and equal to the rate set in the two-dimensional models for the leachate coming from the landfill. The pollutants concentration in the source, in this case in the percolation flux, depends on the age of the deposited waste materials, the leachate infiltration rate beneath landfill, the depth towards groundwater table, the water permeability and the retention capacity of the unsaturated zone. According to monitoring data, the Cl^- concentration in the "fresh" leachate varies from 1300 to 2000 mg/l, having an average value of 1700 mg/l, and the NH_4^+ concentration – from 85 to 700 mg/l, having an average value of 450 mg/l. For a short period, however, these values decrease exponentially by one or two orders of magnitude and remain relatively constant for decades. At the same time, the leachate infiltration rate is high (1.10^{-4} m/d), the depth towards groundwater table is great (65-80 m), and the conditions for mass transport of pollutants beneath the old and the new sectors of the sanitary landfill are different. Under these conditions and based on the results of the 2D models in the source range, three zones with different recharge mode and intensity of the percolation flux are separated. Their areas coincide with the projections of the landfill's oldest sector – Zone 1, old sector – Zone 2 and new sector – Zone 3. The recharge mode and the concentrations of Cl^- and NH_4^+ in the percolation flux in each zone are presented by the functions C_{CL} = f (t) and C_{NH4} = f (t) – Fig. 1 and Fig. 2.

Basic flow model

The basic flow model FM3D is a three-dimensional simulation of the hydrodynamic field structure. The model area covers part of the Paleogene aquifer to a depth of 100-150 m, falling within the boundaries of a catchment area of 2,4 km^2 that includes the old landfill region and the potential recipients. Two low-rank hydrogeological units are separated in the section (Table 1), which are set as model layers with their corresponding geometry and values of hydraulic conductivity k (Fig. 3 and Table 2). The relief of their bottom and top surfaces is in accordance with the terrain and the extension of the hydrogeological low-rank units. The regional flow is modeled according to the General Head Boundary (GHB) scheme, with the general direction of groundwater being southeast at a mean hydraulic gradient of 0,02. The recharge from infiltration and percolation is set with the Recharge software package, as the accepted initial value for the rate of recharge from infiltration is 2.10^{-5} m/d, and for the rate of contaminated percolation flux – 1.10^{-4} m/d. The basic flow model FM3D is calibrated to the water levels at the monitoring observation points by varying the values of the heads set at the external boundaries, the infiltration rate and the hydraulic conductivity in model layer ML 1.

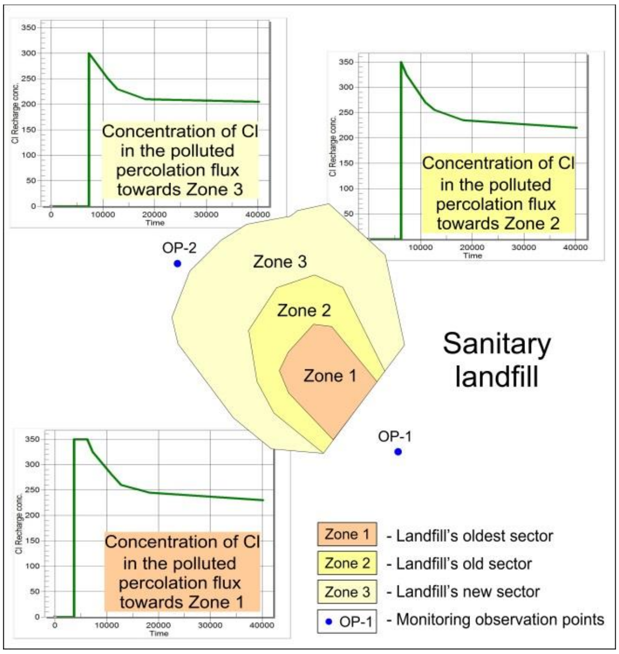

Fig. 1. Concentration of Cl^- in the pollution source

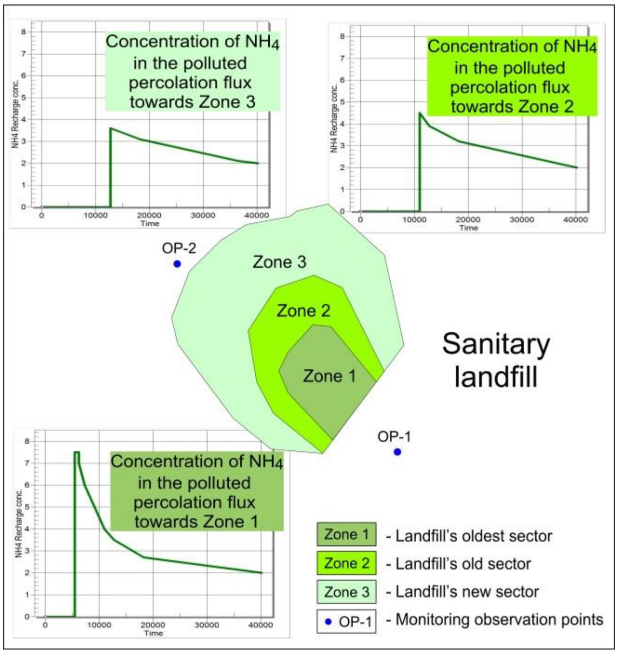

Fig. 2. Concentration of NH_4^+ in the pollution source

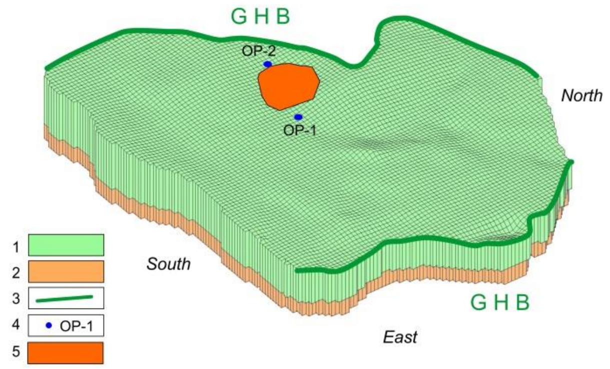

Fig. 3. Grid frame, model layers and boundary conditions
1 – Model layer ML 1; 2 – Model layer ML 2; 3 – General Head Boundary (GHB); 4 – Pollution source; 5 – Monitoring observation points.

Table 1. *Hydrogeological units. Model layers.*

Hydrogeological unit		Model layers
Paleogene aquifer complex	Upper zone of low permeability	ML 1
	Bottom zone of very low permeability	ML 2

Table 2. *Hydrogeological parameters for the model layers – total porosity n, bulk density ρ_d, hydraulic conductivity k, longitudinal dispersivity α_L, diffusion coefficient D_M and distribution (partition) coefficient K_D for Cl^- and NH_4^+*

Hydrogeological parameter	Model layer	
	ML 1	ML 2
n, dimensionless	0,31	0,26
ρ_d, g/cm^3	1,80	2,07
k, m/d	0,10	0,03
α_L, m	12,00	5,50
D_M, m^2/d	4,0.10^{-4}	2,5.10^{-4}
K_D за Cl^-, cm^3/g	0,20	1,10
K_D за NH_4^+, cm^3/g	0,50	2,30

Remark: The values of n, ρ_d, k, α_L, and K_D for Cl^- in model layer ML 1 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 n, ρ_d, k, α_L, and K_D for Cl^- in ML 2, as well as the values of D_M and K_D for NH_4^+ in both model layers are determined from literature data for a similar soils (Spitz and Moreno, 1996).

Mass transport models

The mass transport models M3D-CL and M3D-NH4 are three-dimensional computer simulations of the fate and transport of the key pollutants Cl^- and NH_4^+. Both models are based on the spatial distribution of hydraulic heads, gradients and groundwater flow velocities obtained with the FM3D model. The calculation scheme includes the processes of convective transport, molecular diffusion, mechanical dispersion, reversible elimination and mixing. The accepted for the respective model layers values of the hydrogeological parameters total porosity n, bulk density ρ_d, hydraulic conductivity k, longitudinal dispersivity α_L, diffusion coefficient D_M and distribution (partition) coefficient K_D for Cl^- and NH_4^+ are presented in Table 2. The horizontal and vertical components of transverse dispersivity are determined by the ratio $\alpha_L = 10\alpha_{TH} = 100\alpha_{TV}$. The initial concentrations of Cl^- and NH_4^+ in the groundwater, in the precipitation, and in the groundwater inflow along the NW boundary, are set as zero in both models. The source of pollution is simulated with three model zones, delineating the boundaries of the separated three sectors characterized by different recharge mode and intensity of the percolation flux. In the Recharge software package, the concentrations of Cl^- and NH_4^+ in the percolation flux in each zone are set as variables by the pre-defined functions for all sectors C_{CL} = f (t) and C_{NH4} = f (t). The mass transport models are calibrated to the concentrations of Cl^- and NH_4^+ at the monitoring observation points OP-1 and OP-2 by varying the values of D_M and α_L.

Results and discussion

Structure of the hydrodynamic field

The hydrodynamic field structure, obtained with the basic flow model FM3D, is illustrated in Fig. 4. The correspondence achieved during the calibration between the registered water levels in the monitoring boreholes and the model piezometry guarantees the reliability of the model and the stability of the predictive solutions.

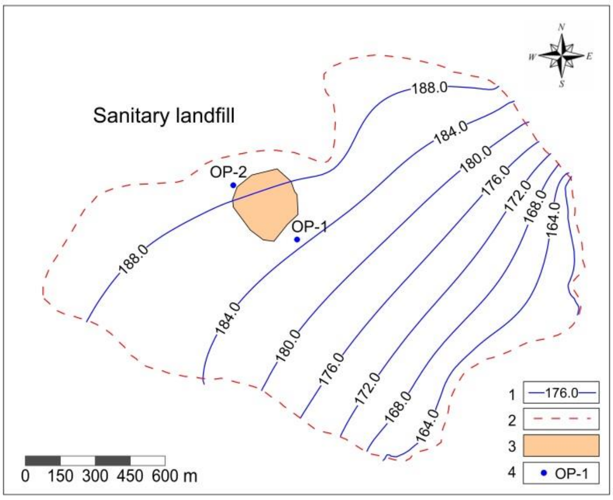

Fig. 4. Model FM3D. Structure of the hydrodynamic field
1 – Piezometric line; 2 – Limits of the model area (surface catchment area); 3 – Pollution source; 4 – Monitoring observation points.

The general direction of groundwater flow is to the southeast and the average hydraulic gradient in the region of the sanitary landfill and its adjacent areas is around 0.015. Such a structure of the hydrodynamic field, in combination with the low water permeability of the Paleogene complex, suggests that the pollutants that have entered the saturated zone will move slowly, following the direction of the simulated groundwater flow.

Groundwater budget

The groundwater budget for the studied part of the Paleogene aquifer complex (Table 3), composed with the FM3D model, shows that the landfill area has poor groundwater resources (about 150 m^3/d) that are practically of no economic importance. The main ingredient in these resources (over 62%) is the groundwater inflow along the NW boundary of the model, and the other significant element (about 34%) is the precipitation that percolates through the unsaturated zone to the water table. The landfill leachate contributes the remaining 4% of the inflow part of the groundwater budget. The outflow segment of the budget has only one ingredient and it is the groundwater outflow along the SE boundary of the model. The modulus of groundwater discharge is 0,7 (l/s)/km^2.

Table 3. *Model FM3D. Groundwater budget.*

INFLOW Q_i^{in} , m^3/d	
Groundwater inflow along the NW boundary of the model	92,45
Recharge from precipitation that percolates through the unsaturated zone to the water table	50,11
Landfill leachate	6,05
Total:	148,61
OUTFLOW Q_i^{out} , m^3/d	
Groundwater outflow along the SE boundary of the model	148,87
Total:	148,87
Overall error in the budget 0.2 %	

Pollutants transport in the saturated zone

The results of the computer simulations of pollutants fate and transport in the saturated zone, performed with model M3D-CL and model M3D-NH4, are illustrated with a series of maps of the distribution of chloride ions and ammonium ions presented in Fig. 5 and Fig. 6. They exhibit the prognostic impact of the old sanitary landfill on the Paleogene aquifer for 1, 5, 10, 50, and 100 year computational (forecast) periods after the start of the computer simulation, i.e. after the first "portions" of polluted percolation flux have reached the saturated zone. This moment is preceded by a period of 10-15 years during which the leachate that has passed through the engineering barrier percolates through the unsaturated zone to the water table.

The following conclusions can be drawn as a result of the analysis of the presented model solutions:

(1) The chief mechanism for movement of contaminants in the saturated zone is convective transport. The direction and rate of mass transport processes are controlled by the hydrodynamic field structure, respectively by the spatial distribution of hydraulic heads and gradients. The pollution front is moving to the southeast following the general direction of groundwater flow.

(2) The pollutant transport in groundwater is not only in lateral direction, but also in depth. The vertical mass transport is determined by the density differences between the contaminated waters and the fresh groundwater lying below them, i.e. by the concentration gradients. Reaching in depth to the undisturbed and practically impermeable parts of the rock massif, the pollutants move in the direction of underground flow, forming a characteristic "plume".

(3) Within the delineated boundaries of the contaminated area, the distinct types of pollutants show significant differences in their behavior (Fig. 5 and Fig. 6).

Chloride ions are conservative, characterized by a very low sorption potential, and move at a pace close to the flow rate of groundwater. They mark the maximum range of possible groundwater contamination. The plume will extend to the southeast with an average rate of 3 m/a. Long-term prognosis shows that in 100 years the pollution front will reach to no more than 300-350 m from the old landfill (Fig. 5). The most intensive contamination will be limited in the space beneath the sanitary landfill and in a narrow strip 70-80 m southeast of it, where the expected Cl^- concentration will be between 200 and 350 mg/l. Outside these limits, the level of pollution will be much lower. Chloride concentration in groundwater will most often be in the range of 30 to 200 mg/l, i.e. will not exceed the permissible limits of drinking water in Bulgaria, established by Ordinance No. 9/16.03.2001. This Regulation sets out the requirements for the quality of water intended for drinking and household purposes in order to protect human health against the negative impact of drinking water pollution.

Ammonium ions have high sorption potential, which is the reason for their very low mass transfer ability. Their movement in the Paleogene aquifer complex is slow, so the groundwater polluted by them is expected to occupy small areas. Calculations made with the M3D-NH4 model show that NH_4^+ transport in groundwater flow direction has an average rate of 1 m/a. Long-term prognosis shows that in 100 years the less mobile pollutants will reach to no more than 100-120 m from the old landfill (Fig. 6). The prevailing concentrations of ammonium ions will be low, most often in the range of 0,01 to 0,25 mg/l. Only beneath the landfill and in the strip 20-25 m from its borders the concentration will be higher, but will not exceed 2 mg/l.

The prognostic solutions show that as time progresses, within the expanding boundaries of the already contaminated zone and in the groundwater flow direction, there is a gradual decrease in the concentrations of Cl^- and NH_4^+. This is due to the decreasing concentration of pollutants in the leachate produced by the old sanitary landfill, as well as due to the processes of reversible elimination (sorption-desorption), hydrodynamic dispersion and mixing that are accompanying the convective transport. The low water permeability of the Paleogene rock massif in combination with the high sorption characteristics of the medium are the reasons for its very good self-purification abilities.

Conclusion

Using three-dimensional models, a computer simulation of the mass transport of selected key pollutants in the saturated zone is performed. The obtained prognostic solutions show that the groundwater contamination caused by the old sanitary landfill near the town of Haskovo, from a long-term point of view (100 years), will be quite limited in size and of very low intensity. It will affect only a minor part of the characterized by low water permeability Paleogene aquifer complex, whose groundwater resources are poor and of no economic importance.

The methodological approach, implemented in the both parts of the presented study, includes the complex application of standard software for development of 2D and 3D models of fate and transport of pollutants in the unsaturated and saturated zones. This general approach is very effective for acquiring quantitative assessments and long-term predictions about groundwater contamination caused by surface sources.

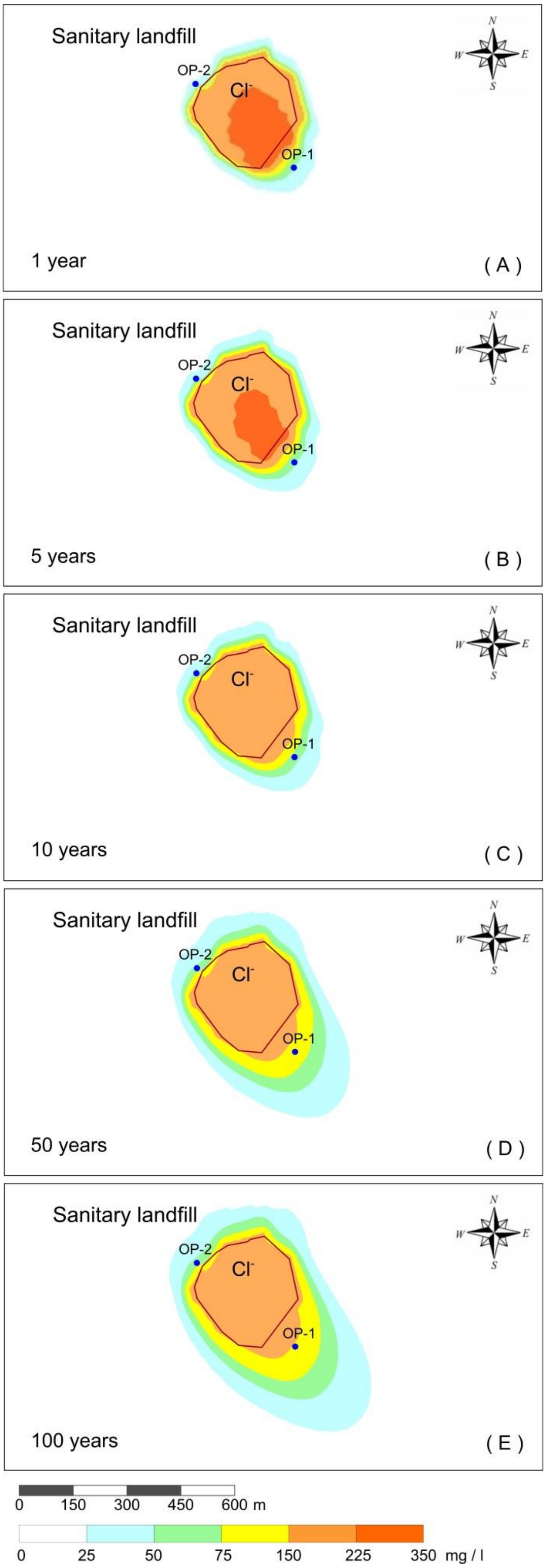

Fig. 5. Model M3D-CL. Distribution of highly mobile pollutants on the example of Cl^-. Model solutions for 1, 5, 10, 50, and 100 year forecast periods after the start of the computer simulation.

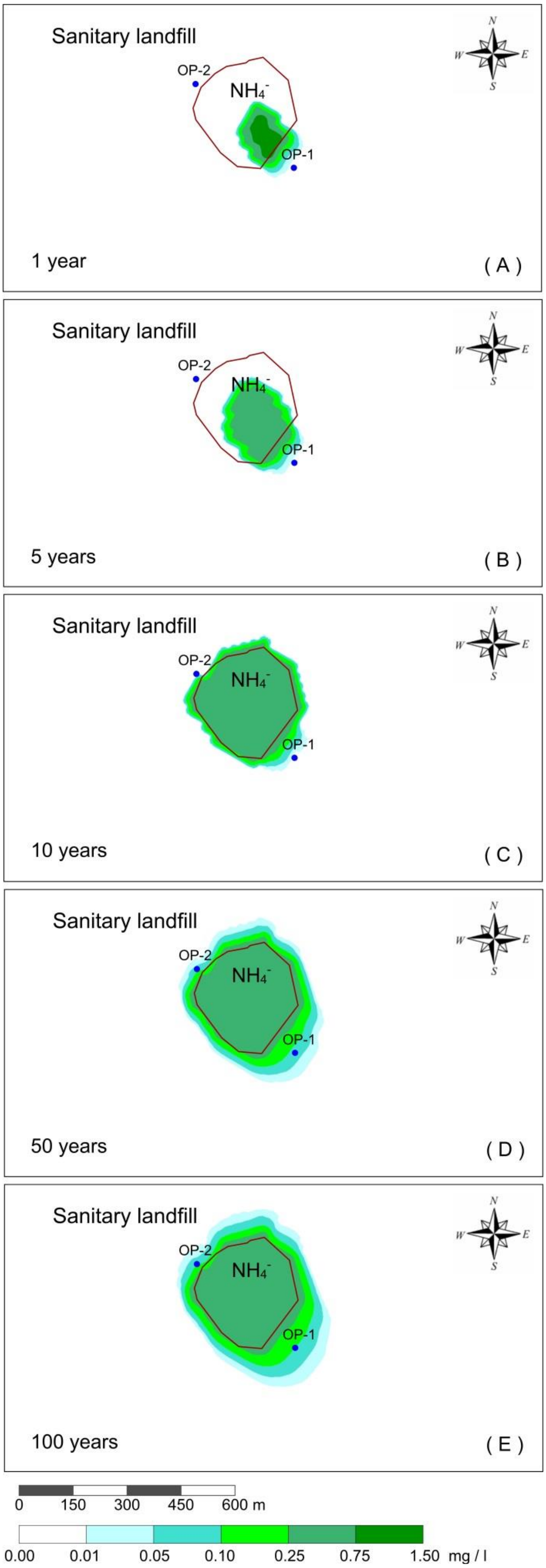

Fig. 6. Model M3D-NH4. Distribution of less mobile pollutants on the example of NH_4^+. Model solutions for 1, 5, 10, 50, and 100 year forecast periods after the start of the computer simulation.

Our experience shows that this methodology can be successfully applied in solving many similar environmental problems related to the evaluation of the impact on groundwater caused by of different technogenic sources of pollution. These are disposal facilities for different types of waste materials, tailings dams, chemical warehouses, hazardous industries, such as oil and gas production, hydrocarbon refining, nuclear power, manufacture of chemicals and pharmaceuticals, etc.

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