Institute of Geology at Tallinn University of Technology

IMPACT OF OIL SHALE ASH AND SEMI-COKE LANDFILLS ON THE WATER ENVIRONMENT

HYDROGEOLOGICAL MODELS OF THE OIL SHALE INDUSTRY AREA IN ESTONIA

Leo Vallner

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CONTENTS

INTRODUCTION

The world's largest industrially used oil shale basin is located in North-East Estonia. About 900 million tons of oil shale has been excavated since 1919. To date, about 80% of oil shale mined is used for generating electricity by incineration and the rest to produce shale oil by retorting. More than 300 million tons of the oil shale industry waste has been placed in 45 landfills.

In 2004 a risk-based environmental site assessment of North-East Estonian landfills was completed as a joint venture of the Norwegian Geotechnical Institute and the Norwegian Geological Survey with the Institute of Geology at Tallinn University of Technology, the Geological Survey of Estonia, and the Institute of Chemical Physics and Biophysics, Tallinn, Estonia (Sørlie J-E *et al.* 2004). The Kohtla-Järve semi-coke and ash landfill, the Kiviõli semi-coke landfills, and ash landfills of the Balti Power Plant (BPP) at Narva were selected for detailed research.

Investigations performed included drilling of 18 wells and their testing, sampling and analyses of soil, groundwater, and surface water, characterization of oil shale waste, inventory of landfills, classification of leachates, and ecotoxicological researches. Groundwater flow models of the semi-coke landfills at Kiviõli and Kohtla-Järve and of the ash landfill at Narva were completed.

It was established that most of the semi-coke wastes were chemically inert according to the EU standard (Decision on…2002; Directive of…1999), exclusive of As, Ba, Mo, Se, and Zn. However, the real leachate of semi-coke landfills, especially at Kohtla-Järve, contained several organic contaminants (BTEX, PAHs, phenols) classified as hazardous. The pH of leachate from fresh waste and the waste pore water was extremely high, reaching up to 12.9. The ecotoxicological testing demonstrated that the contaminant situation at the semi-coke landfills was not acceptable according to the EU landfill directive since the effluent leachate was toxic. Altogether, none of the landfills investigated met the EU environmental requirements (Directive of…1999).

It was concluded that a perfect monitoring of the groundwater quality and experimental testing of flow and transport properties of water bearing layers was necessary to clarify and interpret the contamination impact of landfills. It would be rendered possible to specify the subsurface area of contaminant plumes around landfills.

In 2008 a new joint venture called 'The sustainable groundwater monitoring system of East-Viru County, Estonia' was established with the Ministry of the Environment of Estonia, the University of Tartu, the Institute of Geology at Tallinn University of Technology, and the Geological Survey of Estonia as a Norwegian Financial Mechanism project 52/2006-EE0010. The main purpose of the venture was to motivate and elaborate the principles of an optimum groundwater monitoring system of East-Viru County.

In the framework of this project, the tasks posed to the Institute of Geology were as follows:

- to review the predictive groundwater modelling reports and approaches considering the East-Viru County;
- to establish a groundwater-modelling platform to elaborate an optimum monitoring system that supports an effective groundwater utilization and protection in the Ida-Viru County;
- to develop the groundwater flow and transport models of semi-coke and ash landfills to analyse the fate of the contaminants formed.

Within these tasks, intermediate research reports were completed including an analysis of the impact of the input data accuracy on the authenticity of simulation results (Vallner L 2009a, 2009b, 2008a). The present study supplements and elaborates the results of reports mentioned and profoundly treats the groundwater flow and transport in the areas of landfills by means of hydrogeological modelling.

Chapter 1 of the report presents a critical review of all groundwater modellings that have considered the problems of the East-Viru County until 2010. First, a short compendium is provided about contemporary principal standpoints of groundwater flow modelling. It is necessary for a better understanding of the principles, terms, and notifications performed further. A platform for the development of further groundwater modelling projects is proposed in Chapter 2. The groundwater flow and transport models compiled in the framework of the present study are described and their outputs analysed in Chapter 3. The need for subsequent investigations in connection with the remediation projects of landfills and development of the groundwater monitoring system is discussed in Chapter 4.

Unfortunately, professional English proofreading of this research has not been carried out.

1. REVIEW OF GROUNDWATER MODELLING REPORTS CONSIDERING THE IDA-VIRU COUNTY

1.1. Criteria for evaluation

The groundwater flow modellings except for one case have been carried out in the Ida-Viru County until present. Thereat variable computational codes were used to compile the models and to visualize the results of simulations. In this connection, the main concepts and principles of the contemporary groundwater modelling should be elucidated. It renders to assess to what extent the modelling projects completed match the contemporary state of the art (Anderson M, Woessner W 1992; Ashok K, Sophocleous M 2009; Bear J, Cheng A 2010; Fetter CW 1999, 1993; Kinzelbach W 1986; Zheng C 2011; Zheng C, Bennett GD 2002; Zheng C, Wang P 2003, 1998).

In the most popular MODFLOW modelling environment, the distribution of hydraulic head in a 3D porous domain containing groundwater of constant density is described by the governing partial-differential equation (McDonald M, Harbaugh A 1996)

$$(\partial/\partial x)\ (K_{xx}\partial h/\partial x) + (\partial/\partial y)\ (K_{yy}\partial h/\partial y) + (\partial/\partial z)\ (K_{zz}\partial h/\partial z) - W = S_s\ \partial h/\partial t \qquad (1.1.1)$$

where K_{xx}, K_{yy}, and K_{zz} are values of hydraulic conductivity along the x, y and z coordinate axes [LT^{-1}]; $h(x, y, z, t)$ is the potentiometric head [L]; $W(x, y, z, t)$ is the volumetric flux per unit volume and represents sources and/or sinks of water [T^{-1}]; S_s *is* the specific storage of the porous material [L^{-1}]; and t is time [T].

To solve equation (1.1.1) it is necessary to fix the 3D distribution of hydraulic head h at the moment $t = 0$ in the groundwater domain (the initial condition) and to pose the boundary conditions of this domain.

The initial condition is necessary for calculations describing transient time-dependent alterations of the hydraulic head caused by pumping or changing the intensity of the groundwater recharge. To restore initial distribution of head may be quite difficult for a multi-layered domain where a number of groundwater intakes have been active for a long time. However, it is possible to establish the initial head according to predevelopment conditions (for a historic time before pumping started). It would give an opportunity to check the development of groundwater drawdown from the starting moment of pumping until the latest available monitoring data. Achieving the best match between head data observed and calculated by correcting of conductivity properties of layers allows enhancement of the authenticity of the model for predictive calculations.

Three kinds of boundary conditions of a groundwater domain are mostly distinguished with some modifications. The first kind or Dirichlet type of boundary condition prescribes a value of head $h(x, y, z, t)$ on boundaries of the groundwater domain. The second kind or Neumann type of boundary condition specifies a given flux on the boundary. A special case of this condition is an impervious boundary where the flux is zero [$\partial h(x, y, z, t)/\partial n = 0$; n is a normal to the boundary]. If streamlines form boundaries of the modelled domain, they may also be treated as impervious boundaries. The third kind or mixed or Cauchy type of boundary condition expresses a linear combination of head and flux on the boundary of the model area $\{\partial h(x, y, z, t)/\partial n = = C[h_c - h(x, y, z, t)]\}$ where C is a constant and h_c is a constant head given.

Boundary conditions have a significant impact on head distribution in the groundwater domain modelled. On the other hand, the given boundary conditions always contain a portion of the subjectivism. Therefore, it is useful to shift the outer boundaries of the groundwater domain as distant as possible from the area of the main modelling interest. This way the disadvantageous influence of the possible inaccuracy of boundary conditions can be minimized.

A hydraulic interaction between a river and an underlying groundwater domain is mostly described by the Cauchy type of the boundary condition. In the MODFLOW modelling environment, it is expressed by the equation

$$Q(t) = C_r[(h_r - h_m(t)] \qquad (1.1.2)$$

where $Q(t)$ is the transient flux between the river and the groundwater system; C_r is the conductance of the riverbed; h_r is the river stage, and $h_m(t)$ is the head in a computational cell of the model directly underlying the riverbed.

The conductance C_r may be calculated from the length of the river L_r through a cell, the width of the river W_r in the cell, the vertical hydraulic conductivity of riverbed material K_r, and the thickness of the riverbed M using the formula

$$C_r = K_r L_r W_r M^{-1}. \quad (1.1.3)$$

In general, a direct estimation of the vertical hydraulic conductivity K_r of a bed separating the surface water body from an underlying groundwater system is a very complicated hydrogeological task. Usually, there is not enough field test data for correct calculations. Therefore, indirect methods should be used for solving the problem of an adequate estimation of boundary condition (1.1.2).

Surficial recharge to a groundwater system occurs commonly as a result of precipitation or thaw water percolating into the topmost layer. This process is usually called as infiltration. An opposite process of infiltration is evaporation including plant transpiration and direct evaporation from the groundwater table. The real amount of the surficial groundwater recharge called net infiltration is a sum of infiltration and evaporation. In the course of a hydrologic year, the value of net infiltration is mostly positive in Estonia. Net infiltration is a very important parameter for an authentic groundwater modelling. Due to its positive value, divides of the groundwater table have formed supporting the groundwater flow to river network. The net infiltration is a main component of the groundwater inflow at regional investigations of the groundwater budget. In the equation (1.1.1) net infiltration is given by the term $W(x, y, z, t)$.

A direct measuring of net infiltration by lysimeters is commonly possible only for restricted areas not exceeding few square kilometres. Net infiltration of greater areas can principally be determined by variable formulas elaborated on the basis of such indirect data as the air temperature, atmospheric humidity, wind speed, albedo, plant characteristics, etc. (Herrmann F *et al.* 2009; Scanlon B, Healy R, Cook P 2002; Williams J *et al.* 1998). Unfortunately, those kinds of simulations are often not reliable enough. Their results are mainly acceptable for the same domains which were used for deducing of empiric functions.

In Estonia, the net infiltration can most trustfully specify by means of profound water balance investigations. This conclusion rests on the conception that a great majority of the whole groundwater runoff discharges to river network. The river runoff is perfectly studied by statistic processing of long-term observation data of numerous gauging stations distributed quite evenly on the whole territory of Estonia (Resursy poverhnostnyh…1972). The portion of the groundwater runoff from the surface runoff may be separated by special calculations. This way the real groundwater recharge can be estimated via its discharge instrumentally checked in gauging stations. It gives a best possible credibility to the value of net infiltration calculated.

For groundwater flow modelling a complex of water bearing layers of a heterogenic domain must be divided into model layers and a great lot of computational cells (Trincchero P 2009). In some cases, the number of cells may reach several million. Hydrogeological parameters of the equation (1.1.1) must be given for every cell. A set of mutually connected and dependent modelling data contains often tens of millions items. This data set coupled with a computational code capable to solve numerically the equation (1.1.1) form a simulation model of the groundwater flow. The hydraulic head, the direction, the velocity, and the rate of the groundwater flow can be determined by running the simulation model for every point of the domain modelled, and for every moment of time considered.

The trustworthiness of a model is assessed by comparing of measured and calculated data. For that purpose, mostly the value of the hydraulic head estimated in a field observation point is compared with the head value simulated by modelling for the same point. If a too big deviation takes place then the model parameters should be corrected until the deviation will be in a range permitted. Such adjustment is called the model calibration. The points of the model calibration should be representative enough. It means that an adequate amount of calibration points should be placed in areas where the model is most sensible and where the main interests of modelling have been concentrated. The deviation permitted between measured and modelled data depends on tasks posed for modelling.

Usually, a model is considered a correct one if the value of the coefficient of correlation between the measured (X) and modelled (Y) data is more than 0.9. On the other hand, a 95% confidence interval allows visualizing a range of calculated values for each observed value. The goal of model calibration should be to have the 45 degree line where $X = Y$ fall within the 95% confidence interval lines. The results of calibration should be represented graphically or in a tabular form. The groundwater models not calibrated at all or calibrated poorly are untrustworthy.

The calibration process, besides of its checking and correcting functions can also be used for determination of model parameters which direct experimental estimating is very difficult or even impossible in practise. For instance, to establish the vertical hydraulic conductivity of medium or strong aquitards is extraordinary troublesome by pumping or other field tests. Therefore, the permeability of aquitards is usually insufficiently characterized in reports of hydrogeological investigations. However, using the trial and error method at calibration renders possible to determine the conductivity of every computational cell of model confining beds. The same way the net infiltration may be established analysing the impact its given values on groundwater discharge rates or heights of the groundwater table.

A correct estimation of natural and induced groundwater resources is very important for the determination of an optimum exploitation regime of an aquifer system (Bredehoeft J, Durbin T 2009; Custodio E 2002; Devlin J-F, Sophocleous M. 2005). By means of a subroutine of some advanced modelling codes, it is possible to compile detailed water budgets of a domain modelled and to quantify the flow components forming the rates of groundwater resources. However, despite virtual fitting of these flows, there is no guarantee that they match always adjacent groundwater balance systems. A discordance between neighbouring balance systems witnesses about an incorrect calculation of some flow components. This problem can be overcome by completing a hierarch water balance system where smaller systems interconnected frame into bigger ones and them in their turn suit for a general water budget unit. A regional water budget system containing subsystems reciprocally balanced minimize possible errors in establishing of groundwater flow rates.

The hydrogeological setting of Estonia is very appropriate to put together a hierarch water balance system mentioned above. The water bearing formation is surrounded by seas and big lakes from three sides here. A comparatively tense network of gauging stations on rivers controls the groundwater runoff. All this conduces to an adequate specification of boundary conditions for a correct determination of main components of the regional water budget. The subsystems of the groundwater balance may be composed on the basis of variable units of the hydrogeological stratigraphy.

1.2. Modelling projects

A number of groundwater modelling projects have been carried out considering the whole Ida-Viru County or its local objects including the border area of Russian Federation. A comparative evaluation of adequacy and authenticity of these models and results of modelling is presented below. Lay persons can obtain additional information about the hydrogeological situation of the study area from numerous sources published (Kattai V, Saadre T, Savitski L 2000; Raukas A, Teedumäe A 1997; Perens R 1998; Perens R, Vallner L 1997; Resursy poverhnostnyh…1972, Vallner L 1997a, 1997b, 1996a, 1995, 1994, 1980; Vallner L, Järvet A 1998; Vallner L, Savitskaja L 1997; Vallner L, Sepp K 1993; etc.).

The groundwater modelling projects are considered accordingly to their chronological sequence below. The name of every project translated into English and corresponding reference are presented at the beginning of every review. Projects are evaluated on the ground of above methodological principles. Thereat it is necessary to regard that electrical analogue computers were used for groundwater flow modelling until 1980.

<u>Impact of dewatering of the Ahtme mine on the groundwater table in the caption zone of the Vasavere groundwater intake (Vallner L, Riet K, Metslang T 1972).</u> The Kurtna Landscape Reserve (KLR) in the eastern portion of the Ida-Viru County includes 40 picturesque lakes amidst hillocks overlying the Vasavere ancient valley fulfilled by Quaternarian sands and gravels (Fig. 1.2.1). Some of these lakes are unique due to their ecological type as well as the rare species occurring there. Despite the high ecological and recreational value of the Kurtna landscape, a groundwater intake tapping Quaternarian deposits of the Vasavere valley was put into operation to supply additional water to Kohtla-Järve urban area in 1972. The maximum pumping rate planned for the intake by the geological survey of that time ranged 22,250 m^3 /day (Gontar' V 1965). Such abstraction intensity would induce a drawdown of the groundwater table ranging up to 15–20 m in the central area of the landscape reserve. It was proved by a 2D one-layer groundwater modelling (Vallner L, Riet K, Metslang T 1972) that from the total drawdown up to 10 m would be formed by dewatering of mines and opencast pits surrounding the landscape reserve.

In 1972–1987 at an abstraction rate from the Vasavere groundwater intake reaching only 6,000 m^3/day the water table of many lakes was lowered below their acceptable minimum level, and the unique hydrobiological lake associations and the amenity value of the Kurtna landscape were significantly deteriorated (Mäemets A 1987; Vallner L 1987). In this connection was declared by a professor of the Leningrad Mining Institute Norvatov Ju. (1988) that accordingly to a groundwater modelling carried out under his leadership the mine dewatering would practically not affect the ecological regime of the landscape reserve. This statement was obviously made to defend the Soviet oil shale industry from accusations of Estonian environmentalists.

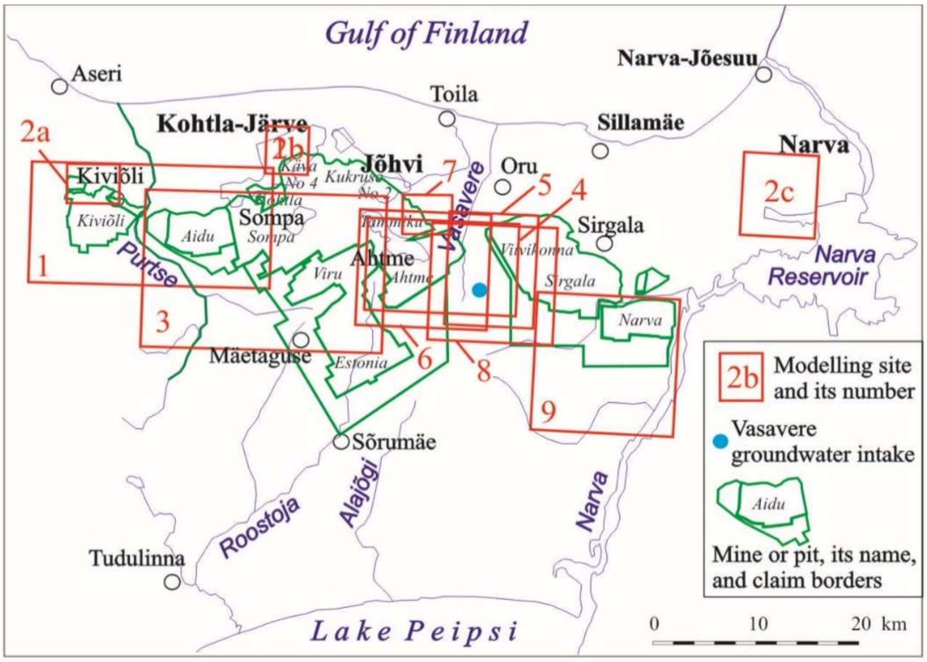

Fig. 1.2.1. Sites of groundwater modelling. Numbers indicate references to modelling projects. 1 – Savitski L., Savva V 2001b; 2 – Sørlie JE *et a*l. 2004 (a – Kiviõli, b – Kohtla-Järve, c-Narva); 3 – Perens R, Savva V, Boldõreva N 2006; 4 – Vallner L, Riet K, Metslang T 1972; 5 – Tamm I 2004, Nontechnical…1996; 6 – Savitski L, Savva V 2001c; 7 – Lind H 2005; 8 – Savitski L, Savva V 2005a; 9 – Savitski L, Savva V 2004.

Regional estimation of the safe yield of fresh groundwater of the Baltic artesian basin (Iodkazis V *et al.* 1977). The main goal of this investigation was to make a preliminary assessment of potential possibilities of groundwater abstraction in Estonia, Latvia, and Lithuania. In this connection, a concept of the safe yield of groundwater resources was implemented. The groundwater safe yield was considered to be an amount of groundwater which could be withdrawn from a certain aquifer at a given drawdown during 30 years without producing an unpermitted deterioration in the quality of the water pumped (Vallner L, Savitskaja L 1997c). To determine the safe yields a 2D flow modelling of main aquifers of the Baltic artesian basin was preformed. The cell size of the orthogonal modelling grid was mostly 20 km. Groundwater safe yields were calculated chiefly for towns correspondingly to Soviet hydrogeological prescriptions. Results of that basin-wide work are obsolete for time being but the initial data used (values of hydraulic conductivity, measured groundwater heads, etc.) are still usable for hydrogeological research of Ida-Viru County.

Hydrodynamic division of Estonian groundwater basin and its water budget (Vallner L 1980). A groundwater flow model of water bearing formation of Estonia and surrounding its shelf was completed by means of a digital computer. The groundwater system was divided into seven hydraulically interconnected model layers representing all main aquifers. The net infiltration given to the top of the model was determined by former investigations on the basis of the groundwater runoff measurements (Vallner L 1976, 1975; Vallner L, Metslang T 1970). Surface water bodies (river network, lakes, and sea) were hydraulically connected with the model. An optimum match between measured and modelled values of hydraulic heads and groundwater runoff was obtained by model calibration. As a result of 3D simulations carried out the water bearing formation was divided into 59 spatial hydrodynamic units. Their boundaries were determined by sharp contrasts of direction and velocity of groundwater fluxes.

For every hydrodynamic unit distinguished the inflow components (fluxes from above, below, the side, possible net infiltration, an induced flux from a surface water body), and outflow components (fluxes up, down, sideward, to the river network, the discharge to the sea, through springs, abstraction) were calculated. The data set obtained was equilibrated and adjusted by a solution of an algebraic system of balance equations. For every hydrodynamic unit distinguished the inflow components (fluxes from equations. These way 767 constituents of the basin wide groundwater flow were estimated. The velocities of water exchange between the balance units were established. It was proved that the amount of groundwater flow penetrating the upper zone of active water exchange exceeded by 10 times the flow rate in the underlying zone of the passive water exchange. The data got and concepts elaborated were used later for compiling of regional groundwater models.

Hydrogeological model of North-East Estonia (Vallner L 1996b). Since 1996 in Estonia groundwater flow modelling projects have mostly been carried out on the basis of the well-known digital code MODFOW. The first investigation of this kind enfolded an area covering 21,650 km^2 (Fig. 1.2.2). This domain included the territory of Estonia eastward from Kehra and northward from Põltsamaa together with a land strip of Russian Federation from the Narva River until the longitude of Kingisepp-town and the outcrop of the Cambrian-Vendian aquifer system on the bottom of the Gulf of Finland. The main units of the hydrogeological stratigraphy were represented by seven model layers (from top to bottom):

1) Quaternarian deposits, Devonian layers, and Silurian-Ordovician aquifer system as an integrated water bearing formation hydraulically densely interconnected;
2) Silurian-Ordovician basin-wide aquitard;
3) Ordovician-Cambrian aquifer system;
4) Lükati-Lontova basin-wide aquitard;
5) Voronka aquifer;
6) Cambrian-Vendian aquifer system in the western portion of the study area, Kotlin aquitard in the eastern area;
7) Cambrian-Vendian aquifer system in the western portion of the study area, Gdov aquifer in the eastern area.

The computational code Visual MODFLOW v. 2.1 was used for groundwater flow modelling. The Dirichlet and Neumann type of boundary conditions were mostly omitted to the outer boundaries of the model layers. The groundwater outflow from Ordovician layers along the Cliff (Klint) of North Estonia and the discharge of the Silurian-Ordovician aquifer system through numerous springs on the Pandivere Upland were modelled by the Cauchy type of boundary conditions (MODFLOW’s Drain

Package). The hydraulic interaction between the groundwater system and surface water bodies (streams, rivers, Lake of Peipsi, Gulf of Finland) was described by the boundary condition (1.1.2).

The surficial groundwater recharge into the top layer of the model was given as the net infiltration (total groundwater recharge minus evaporation from the zone of saturation or capillary fringe) differentiated on the study area. It was preliminarily calculated from the budget equation connecting the main components of the basin-wide groundwater flow (Vallner L 1997, 1980):

$$I = F + P + M - A \pm V \pm G \tag{1.2.1}$$

where I is the net infiltration; F is groundwater discharge (base flow) to river network; P is groundwater abstraction; M is the direct seepage of groundwater to the sea; A is the flux of surface water from river network into aquifers (induced recharge, mostly in vicinity of mines dewatered and locally in karst areas); V is the subsurface exchange of groundwater between the study area and surrounding region and G is the storage change.

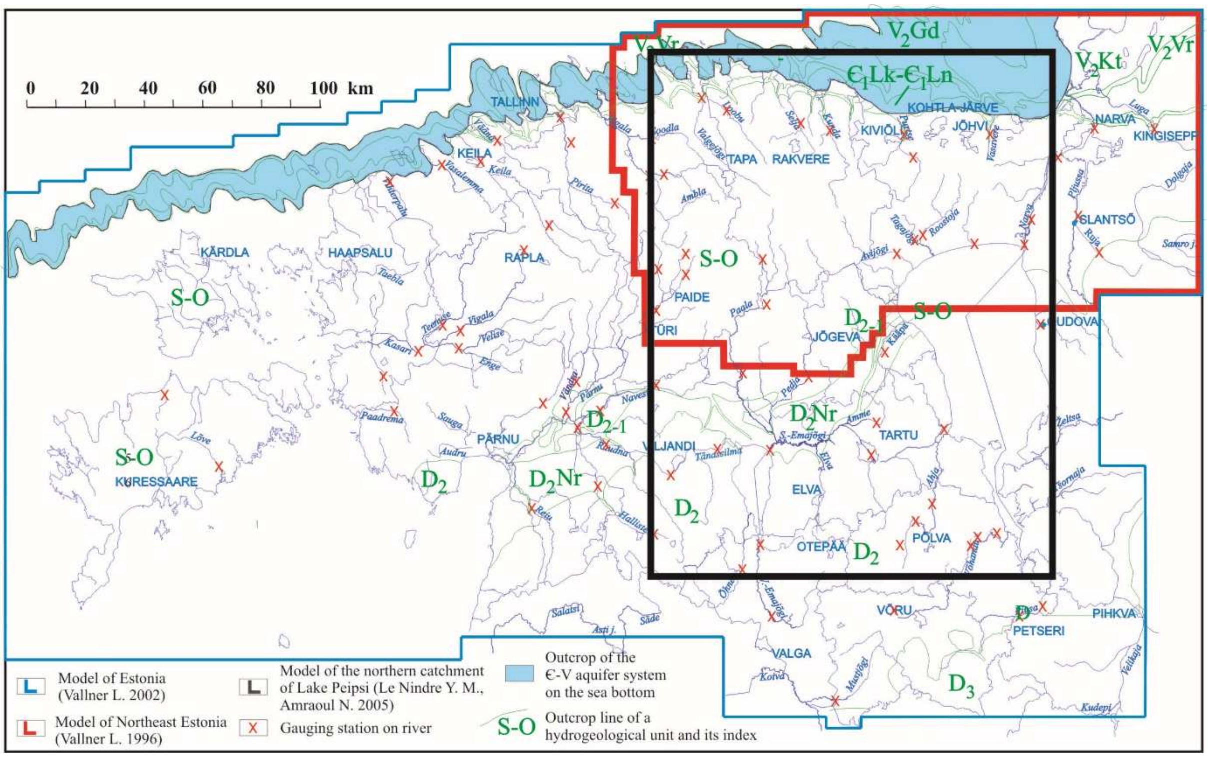

Fig. 1.2.2. Borders of regional groundwater models.

A detailed map of the groundwater runoff at the scale 1:200,000 compiled on the basis of many measurements of the low flow (Resursy poverhnostnyh…1972; Vallner L 1976, 1975; Vallner L, Metslang T 1970) was used to estimate the fluxes F and A for equation (1.2.1). The pumping data P were obtained from state institutions checking the groundwater use. The groundwater exchange with adjacent areas and most deep layers V, as well the direct subsurface flux to sea M (if it did exist) were calculated by Darcy's formula.

The riverbed conductance C_r needed for a correct determination of boundary condition (1.1.2) was calculated by the equation

$$C_r = F/(H - H_r) \tag{1.2.3}$$

where F is the groundwater discharge to river network under consideration; H is the average value of the groundwater head beneath the river and H_r is the mean river stage elevation.

The values of the parameter F were taken from the groundwater runoff map mentioned and the heads H were estimated on the basis of variable boring data. This way a trustworthy calculation of the riverbed conductance C_r was feasible. The conductance of bed material of the sea and Lake Peipsi was estimated the same way as for river network. Instead of the value of *F,* the direct seepage of groundwater to the sea or to the lake M was used in the equation (1.2.2). The value of M was calculated by the Darcy formula.

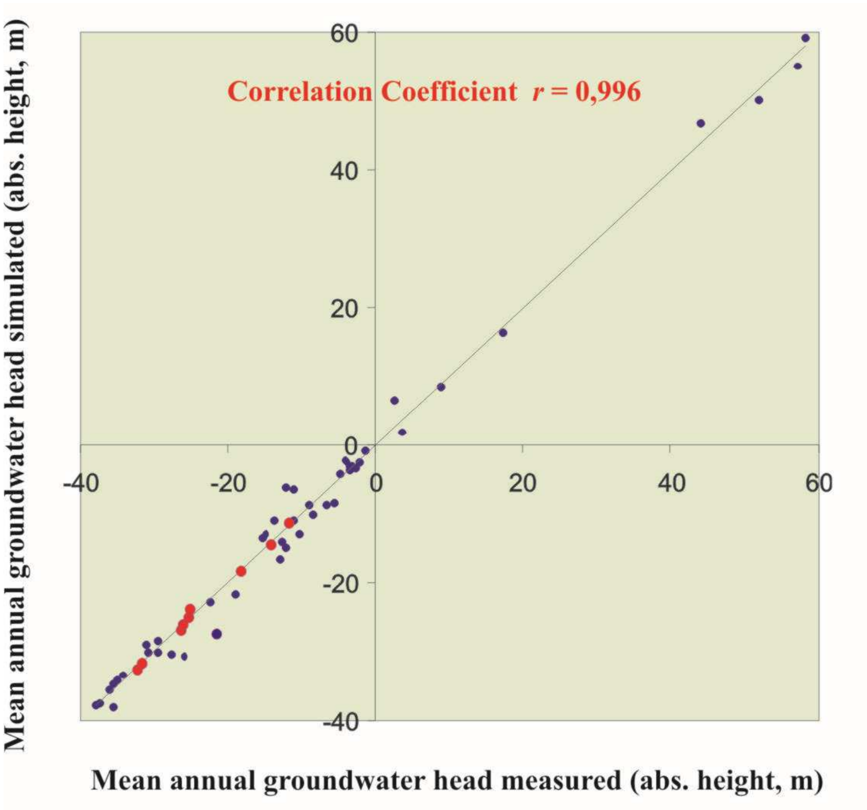

Fig. 1.2.3. Calibration graph of the regional model of Northeast Estonia. Calibration dates: blue dots – 1976, and 1995; red dots – 1998.

The model was calibrated against two different sets of calibration targets – one set representing the measured elevation of groundwater table and head averaged to 1976, 1995, and 1998, and another set corresponding to groundwater runoff to the river network in the same time. Using different kinds of calibration targets enhanced the trustworthiness of modelling results and especially the adequacy of budget calculations. Calibration was carried out using the trial-and-error adjustment for achieving the optimum match between simulated parameters and calibration targets. Simulated elevations of the groundwater table and heads were rechecked against field data until the correlation coefficient between computed and measured values was more than 0.99 (Fig. 1.2.3). The maximum difference allowed between measured and model-calculated rates of the base flow was ±10%. The results of model calibration were shown at observations points, which were presented on the maps included in the report.

The model described was henceforth used for solving of several sophisticated regional hydrogeological problems (Savitski L, Savva V 2005b, 2001a, 2000a, 2000b; Savitski L, Vallner L 1999a, 1999b).

Nontechnical summary of the environmental impact assessment Kurtna Lakes and groundwater protection. 1996. As it was mentioned above the water regime of the KLR suffered seriously from the groundwater overexploitation and dewatering of surrounding mines and pits. To find out an optimum solution for this problem an environmental impact assessment was undertaken by the Estonian Ministry of Environment using the methodological aid of the U. S. Environmental Protection Agency. Besides of other investigations a groundwater flow modelling which enfolded the water bearing formation of the KLR was carried out by the Maves Company (Keskkonnaekspertiis. Kurtna…1996; Nontechnical…1996). Sizes of the area modelled were 17 km × 14 km (Fig. 1.2.1). The model comprised four layers from above to bottom representing:

1) Quaternarian sand;
2) Quaternarian till and Devonian sandstones;
3) Ordovician limestones (O_2Id-Kk);
4) Ordovician clayey limestones and marls (O_2Uh).

The pre-processor ModelCad[386] coupled with the MODFLOW code was used for modelling. Net infiltration into the top of the model was neglected. The lakes were not considered as surface water bodies hydraulically connected with the topmost model layer. It was presumed that the water table of lakes corresponded to the groundwater table in localities of lakes. Along the outer borders of the domain modelled the boundary conditions were given by the constant heads or by no-flow cells. These boundary conditions reflected partially the height of the water table withdrawn in mines and pits.

A series of steady state simulations were completed by modifying of the minimum distance of mines and pits from the KLR. Thereat, the pumping rate of the Vasavere groundwater intake was changed from zero until 10,000 m^3/day. A number of scenarios were analysed to find out the most suitable ones at which the damage of lakes of the KLR would remain in acceptable ranges. It was concluded that the minimum distance between the KLR and claim borders of surrounding mines and pits must not be less than 2,000 m. If the mine faces will be moved to the distance of 1,000 m from the KLR then several lakes would be irreversible ruined. The pumping rate of the Vasavere groundwater intake must be reduced to the level of 4,000 m^3/day during the period 1996–2000. Later the Vasavere intake must be shut down completely. The Kurtna Lakes will perish gradually if the discharge rate of the Vasavere intake would be 10,000 m^3/day.

The ecological state of Kurtna Lakes was mostly good in 2006 because the pumping intensity of the Vasavere intake was decreased to an average value ranging 6,000 m^3/day in 1996–2006 (Perens R, Savva V 2007b, 2006; Savitski L, Savva V 2005a; Viru alamvesikonna…2006). During the same period on ⅓ of the perimeter of the KLR, the distance to mine faces was only 200–1000 m. Thus, the impact of mine dewatering and groundwater pumping from the Vasavere intake were significantly overestimated by modelling considered. This rough miscalculation was obviously made because of ignoring of the net infiltration. The existing data about the net infiltration (Vallner L 1987) were not accounted for modelling.

Estimation the safe yield of the Cambrium-Vendian aquifer system for Ida-Viru County (Savitski L, Vallner L 1999a). The hydrogeological model of North-East Estonia (Vallner L 1996b) described above was used for estimation the prognostic safe yield of the Voronka aquifer and Gdow aquifer in the Ida-Viru County (Fig. 1.2.2). Until 2004 the groundwater safe yield was defined as an amount of groundwater that could be withdrawn in a rational mode accordingly to a pumping schedule given without worsening the ecological situation (Keskkonna-alaste õigusaktide…1998). Thereat was required that the quality of water pumped must remain in the acceptable ranges during an abstraction period foreseen for an intake (usually 10,000 days). Specification of the groundwater safe yield did not explain what is 'the worsening of the ecological situation'. No restrictions were established on the lowering of the groundwater table or pressure in aquifers during the pumping.

The model of Northeast Estonia run in the transient state was over again profoundly calibrated against groundwater heads and river low runoff observed in 1995 and 1998. Discharge rates needed for consumers were omitted to groundwater intakes of the model. Other hydrogeological parameters remained the same as they were in the model completed in 1996. The size of rectangular modelling cells varied from 200 m to 1,000 m in the area of main research interest. In the marginal areas of the model, the size of cells was up to 4,000 m. On the basis of modelling simulations was accepted that until 2020 in Sillamäe the prognostic safe yield for the Voronka aquifer would be 23,700 m^3/day at the maximum drawdown reaching 61 m b.s.l. The prognostic safe yield of the Gdov aquifer accepted for the same period was 21,600 m^3/day at the drawdown up to 42 m b.s.l. in Kohtla-Järve.

Modelling showed that groundwater abstraction corresponding to maximum value of safe yields estimated would induce the intrusion of saline seawater into the Cambrian-Vendian aquifer. The amount of seawater encroaching into the Voronka aquifer will be 19,200 m^3/day, and into the Gdov aquifer – about 11,000 m^3/day.

Determination of the safe yield proved for groundwater intakes of Narva and Narva-Jõesuu (Savitski L, Vallner L 1999b). Correspondingly to a prescription of the Estonian Ministry of the Environment (Keskkonna-alaste õigusaktide…1998), the preliminary estimated prognostic safe yield must be specified by additional investigations and real pumping before its value could be used in groundwater intakes designs. Such maximum abstraction rate allowed by the Ministry of the Environment is called 'the safe yield proved' (in Estonian 'põhjavee tarbevaru') in contrary to the prognostic safe yield. The goal of the project under consideration was to determine the safe yield proved for groundwater intakes of Narva and Narva-Jõesuu. The model of North-East

Estonia (Fig. 1.2.2) well-calibrated at the previous investigation (Savitski L, Vallner L 1999a) and perfected by additional data of the Narva area was used for this purpose again. As a result of adjusting simulations, the safe yield proved was determined for the Voronka aquifer ranging 3,500 m^3/day for intakes in Narva and 2,500 m^3/day in Narva-Jõesuu.

Determination of the safe yield proved of the Cambrian-Vendian aquifer system for groundwater intakes of Jõhvi, Kohtla-Järve, Kohtla-Nõmme, and Kiviõli (Savitski L, Savva V 2000a). The model of North-East Estonia elaborated (Savitski L, Vallner L 1999a) was used to determine the safe yield proved for groundwater intakes of Jõhvi, Kohtla-Järve, Kohtla-Nõmme, and Kiviõli (Fig. 1.2.2). This model was made perfect by adding and adjusting of localities and exploitation characteristic of production wells. All production wells existing were incorporated into the model. Pumping rates sought by water consumers were given to production wells from 2000 until 2020. Transient simulations demonstrated that all safe yields proved were the same as prognostic safe yields calculated earlier (Savitski L, Vallner L 1999a) except the safe yield for Sompa. One production well was decided to switch off there, and it brought to a decreasing of the safe yield by 600 m^3/day in Sompa. The predicted drawdowns were also very close to drawdowns which would be caused by pumping out of prognostic safe yields.

Determination of the groundwater safe yield proved for Sillamäe (Savitski L, Savva V 2000b). The groundwater safe yield proved for Sillamäe was also determined by the model of North-East Estonia (Vallner L 1996b). For the calculations, all existing production wells in the area of Sillamäe were included in the model. Values of the hydraulic head and conductivity of Cambrian-Vendian aquifer system were somewhat corrected accordingly to additional data obtained. Discharge rates suitable for consumers were omitted to production wells from 2000 until 2020. The simulation showed that in the transient state the safe yield proved for the Voronka aquifer was 6,500 m^3/day, and for the Gdov aquifer – 500 m^3 /day, with the maximum drawdowns reaching 62 m b.s.l., and 22 m b.s.l., respectively. This result exactly concurred with the prognostic safe yield estimated before (Savitski L, Vallner L 1999a).

Estimation of the safe yield of the Ordovician-Cambrian aquifer system in Ida-Viru County until 2020 (Savitski L, Savva V 2001a). Based on the groundwater model of North-East Estonia (Vallner L 1996b) the safe yield of the Ordovician-Cambrian aquifer system was calculated for the Ida-Viru County (Fig. 1.2.1). The data of all 258 production wells tapping this aquifer system were input into the model. An additional model calibration was carried out against the hydraulic heads to seek an optimum match between the newest data observed and modelled. The cell size of the quadratic modelling grid was reduced to 200 m in the area between Sonda, Vasavere, North Estonian Cliff, and Iisaku to achieve a higher exactness of calculations. The safe yield proved totalling 1,450 m^3/day was determined for Kiviõli and Mäetaguse where groundwater abstraction exceeded 500 m^3/day. The prognostic safe yield, all together 4,370 m^3/day, was estimated for other settlements of Ida-Viru County. The safe yield was calculated for the period from 2000 until 2020. In Mäetaguse the potentiometric head will be lowered up to 12 m b.s.l. due to consumption of the whole safe yield.

Thus, the suitability, adequacy, and universality of the hydrogeological model of North-East Estonia were proved in five sophisticated research projects carried out in 1999–2001, where the model was successfully applied. Thereat, the set of modelling characteristic was supplemented and corrected by additional data, and the spatial particularity of the model was improved by the refining of the cell size of the computational grid up to 200 m. The model was satisfactorily verified by trying of variable data sets.

Forecasting of the hydrogeological situation in the area mined of the Estonian Oil Shale Deposit. 2nd stage: Closing the Aidu open pit (Savitski L, Savva V 2001b). The main goal of this project was to forecast the variation of the groundwater table in the Aidu opencast pit and adjoining Kohtla mine after closing the Aidu pit. It was also requested to estimate a pumping rate needful to keep the groundwater table at an optimal level in the Aidu pit closed. A groundwater model of the study area covering about 450 km^2 was completed (Fig. 1.2.1). The four model layers represented following hydrogeological units from above to bottom:

1) Keila-Kukruse aquifer;
2) The oil shale production seam of the Kukruse Stage;
3) Uhaku aquitard;
4) Lasnamäe-Kunda aquifer.

The cell size of the orthogonal modelling grid was 50–100 m. The modelling code used was the Groundwater Modeling System (GSM) v. 2.0. Unfortunately, the research report does not explain the boundary conditions, or the principles establishing the net infiltration. The lack of calibration data nor the simulation mode (steady-state or transient) aren’t explained. Nevertheless, it is noted that according to the modelling results the water table in the Aidu pit might be kept on the absolute height of 37 m if 60,000 m3/day of water was pumped out from there. In the given state, it is possible to hold the water table at an altitude of 40 m a.s.l. in the Kohtla mine with the condition that Aidu pit is connected to an outlet and Kohtla mine is submerged. On the other hand, the water table in the closed Aidu pit might be kept at a height of 42 m a.s.l. without any pumping, through an outlet directing water from the pit into the Ojamaa River. The Kohtla mine was closed in 2001 but an outlet was not built into the operating Aidu pit. Nevertheless, the amount of water pumped out from the Aidu pit for its dewatering was about 124,000 m^3/day in 2001 and 105,000 m^3/day in 2002 (Savitski L 2003). Thus, the real pumping rate exceeded the forecasted one by two times on average. The modelling inaccuracy cannot be explained due to the lack of important modelling characteristics elucidated in the project report

Forecasting of the hydrogeological situation in the area mined of the Estonian Oil Shale Deposit. 3rd stage: Closing the Ahtme mine (Savitski L, Savva V 2001c). The project report declared that a groundwater model was completed which included the KLR and eastern portions of the Tammiku mine, Viru mine, Ahtme mine, and Estonia mine in a rectangular area covering 156 km^2 (Fig. 1.2.1). The information about the model layers and their discretization is not clear. However, it seems that the water bearing Quaternarian deposits, local Ordovician aquifers, and aquitards were incorporated into the model. The principles of establishing of boundary conditions were not explained. The data about net infiltration, model calibration, verification, and simulation mode were not represented in the project report. Due to the lack of important modelling characteristics in the project report, it is not possible to evaluate the trustworthiness of the model under consideration.

The groundwater table stage was forecasted by modelling until the 10th year after the flooding of the Ahtme mine. It showed that the raising groundwater would start to move towards Vasavere groundwater intake in six years after closing down the Ahtme mine. The velocity of this movement was not estimated (for instance, by the MODPATH-calculations). The distance between the face of the polluted mine water and the Vasavere intake was 1.0–1.2 km in 2001 (Savitski L, Savva V 2001c).

Hydrogeological model of Estonia (Vallner L 2002). A 3D groundwater flow model was created on the basis of the code Visual MODFLOW in 2002 (Guiger N, Franz T 1996). Later this model was significantly perfected with the incorporation of supplementary data and it was successively converted to the codes Visual MODFLOW v. 4.0, v. 4.2, v. 4.3, and v. 4.5 (Visual MODFLOW…2010, 2006, 2004). The preliminary steady-state flow model was developed to a coupled transient flow and transport model (Marandi A, Vallner L 2010).

The model enfolds the whole territory of Estonia, the surrounding coastal sea, Lake Peipsi with the Lake Pihkva, border districts of Russian Federation, and Latvia, all together 88,032 km^2 (Fig. 1.2.2). The 20 model layers include all main aquifers and aquitards from the ground surface to as low as the impermeable portion of the crystalline basement. They represent the basin-wide hydrogeological units from top to bottom as follows:

1) Aquifer system of Quaternarian deposits;
2) Upper-Devonian aquifer;
3) Middle-Devonian aquifer system;
4) Narva regional aquitard;
5) Middle-Lower Devonian aquifer system;
6) Upper-Silurian-Upper-Ordovician aquifer system;
7) Vormsi aquitard;
8) Nabala-Rakvere aquifer;
9) Oandu aquitard;
10) Keila-Kukruse aquifer;
11) Commercial oil shale bed;
12) Uhaku aquitard;
13) Lasnamäe-Kunda aquifer;
14) Silurian-Ordovician regional aquitard;
15) Ordovician-Cambrian aquifer system;
16) Lükati-Lontova regional aquitard;
17) Voronka aquifer;
18) Kotlin aquitard;
19) Gdov aquitard;
20) Proterozoic aquifer system.

The study area was initially covered with a virtual rectangular grid at a spacing from 1,000 to 4,000 m for finite-difference discretization. The top boundary of the model coincides with the ground surface or bottom of the river network, lakes, and seas. A virtual surface lying at a depth of 100 m beneath the upper surface of the crystalline basement acts as a supposed impermeable bottom boundary of the model. The thickness of all water-bearing formation modelled varies from 100–150 m in the north to 600–900 m along the southern border of the area.

Principles of establishing of boundary conditions, initial condition, net infiltration, and other model parameters were the same as at compiling of the model of North-East Estonia (Vallner L 1996b) described above. All data of the model of North-East Estonia were completely turned to account for creating of the northeast portion of the model of whole Estonia.

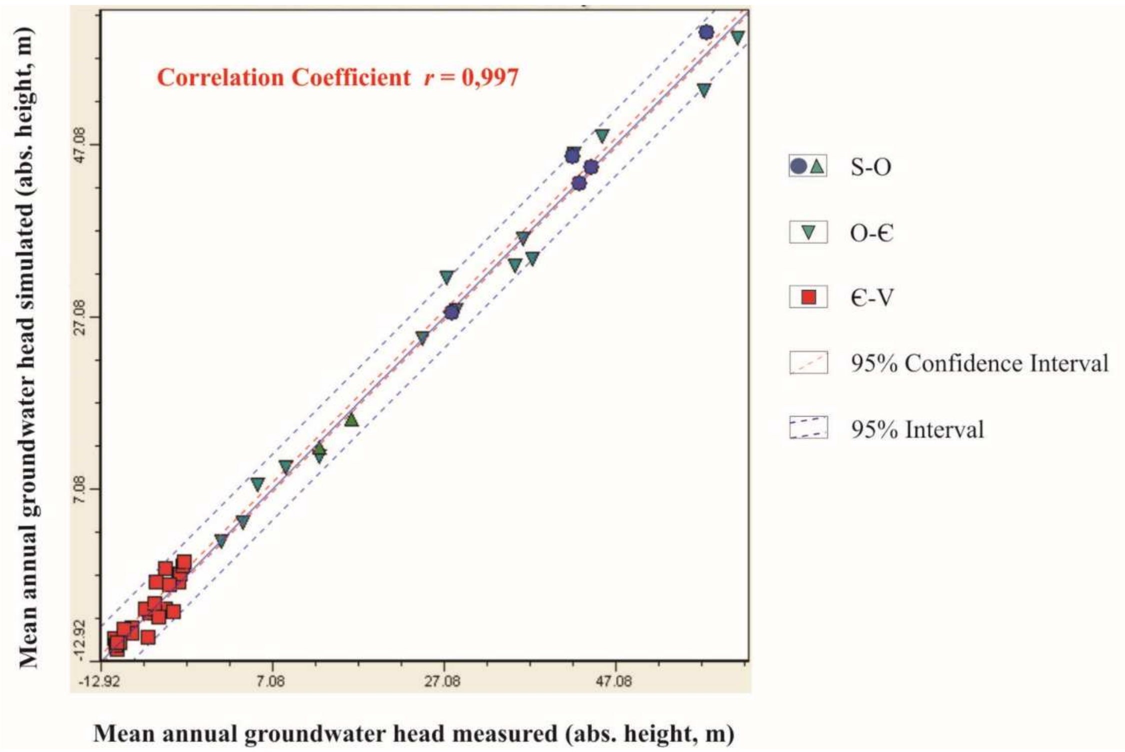

Fig. 1.2.4. Calibration graph of the regional model of Estonia. Observation points of deep aquifers of Northern Estonia are presented.

Basin-wide distribution of the groundwater head in all aquifers and aquitards of the model was determined by several series of steady state and transient simulations solving the equation (1.1.1) by numerical methods of the MODFLOW-code. The steady-state model was preliminarily calibrated against the sets of observed elevation of the groundwater table and head in the study area in September 1976, and against the measured rates of the base flow at stream gauging stations at the same time. The synchronous pumping data were also incorporated into the model for its calibration. This calibration term was fixed because only one set of profound hydrogeological maps existed where heights of the groundwater table and heads of all hydrogeological units modelled were modified to a united date – September 1976 (Vares H, Viigand A 1979). The total groundwater abstraction reaching 603,000 m^3 per day in the study area in 1976 was close to the mean abstraction during the last four decades. This made it possible to prevent the eventual unfavourable impact of extreme abstractions on the exactness of calibration.

All groundwater intakes were deactivated in the calibrated steady-state model and a corresponding distribution of heads $h(x, y, z)$ was simulated. This 3D head distribution was considered as an initial condition *h*(*x, y, z,* 0) for the transient model. After that, pumping schedules were omitted from groundwater intakes for transient simulations. Mean annual elevations of the groundwater table and heads estimated for 1976, 1990, 1995, and 1998 in the observation network were used as targets for transient calibration. In general, the correlation coefficient between the measured and simulated values was reached 0.99 (Fig. 1.2.4).

Using the perfectly calibrated model, the 3D distribution of the hydraulic head of every hydrogeological unit for predevelopment conditions was simulated first time in Estonia. It gave a possibility to assess the quantitative degree of the man-made impact on the groundwater environment. The vertical hydraulic conductance of basin-wide aquitards (Middle-Lover Devonian Narva, Silurian-Ordovician, Cambrian Lükati-Lontova, and Vendian Kotlin) was specified by the trial-and-error adjustment of hydrogeological parameters at the calibration. All these determinations included also the Ida-Viru County.

The regional hydrogeological model of Estonia (RHME) is suitable for the simulation of variable steady-state and transient 3D groundwater flow, transport, and particle tracking problems. It has been used for profound investigation of groundwater resources and completing detailed water budgets of Northern Estonia (Bruin de EFLM *et al*. 2006; Gavrilova O, Vilu R, Vallner L 2010; Vallner L 2008; Gavrilova O *et a*l. 2005; Perens R. *et a*l. 2006; Sørlie JE *et a*l. 2004). Thereat, the transboundary interactions between Estonia and adjacent states might be studied (Fig. 1.2.6).

The basin-wide model can be very useful in the research of many local problems, too. For that purpose, the spacing of the virtual computational grid of the regional model can be refined to the 100 m or even less, and the data of the 'big' model checked by calibration can be used for a new local model (Marandi A, Vallner L 2010). The flow lines simulated by the basin-wide model should serve as the boundaries of the local model, with the Cauchy non-flow conditions along them. The local model can contain fewer layers than the basin-wide one, but in this case, initial conditions of the local model should be calculated by basin-wide modelling. All these methods facilitate the completing of the local model and enhance its authenticity.

Hydrogeological model of Estonia and its applications (Vallner L 2003). The principles of creating of the basin-wide groundwater model completed in 2002 (Fig. 1.2.2) and recommendations for its application were presented in this publication. The distribution of hydraulic head and groundwater flow velocity in the Cambrian-Vendian aquifer system, and its water budget was simulated for the both, predevelopment era and for 1976.

Hydrogeological modelling of the Usnova real estate and of the northeast portion of the Puhatu Nature Protection Area (Savitski L, Savva V 2004). The main goal of the project was to estimate how the dewatering of the Narva pit and a new pit planned on the Usnova real estate would influence the groundwater table in the Puhatu Nature Protection Area (PNPA). For that, the northern portion of the PNPA, the adjacent Usnova real estate, and the southern portion of the Narva open pit was included into a rectangular modelling area covering about 300 km^2 at the Narva River (Fig. 1.2.1). The model completed comprised five layers from top to bottom as follows:

1) peat;
2) glaciolacustrine sand;
3) varved clay;
4) Devonian marl with seams of dolomite;
5) Ordovician limestone and oil shale.

The total thickness of the water bearing formation modelled was about 40–60 m. The code GMS was used. The spacing of the orthogonal computational grid was 250 m. The data about of boundary conditions, net infiltration, modelling technology, and calibration were not presented in the report. In result of modelling was estimated that the water table might be lowered up to 2–3 m on an area covering only 1–2 km^2 at the northern border of the PNPA due to dewatering of the Usnova pit planned. It is difficult to assess the trustworthiness of this prediction since the modelling characteristics were not sufficiently explained in the project report.

Impact of expansion of the Viivikonna department (Sirgala II) on the water table of Kurtna Lakes (Tamm I 2004). In a former research project, the Maves company stated that based on modelling results, the distance between the KLR and adjacent pits must not be less than 2,000 m (Non-technical…1996). Now, this conclusion was overthrown by means of a new modelling (Fig. 1.1.1). For that purpose, very similar models to the model used earlier (Kurtna piirkonna…1996) were completed. A virtual vertical impermeable barrier was designed between the northeast portion of the KLR and the Viivikonna department of the Narva opencast pit. It was proved by simulations that the pit of the Viivikonna department dewatered could be shifted toward the KLR up to the distance of 100 m. In that case, at an abstraction rate from the Vasavere groundwater intake equal to 4,260 m^3/day, the water table of Kurtna Lakes would be lowered up to 0.21 m, only. It is not possible to check the results of modelling because the data about net infiltration and the vertical conductance of lake beds have not been presented in the project report.

Estonia, the oil shale industry. Risk based environmental site assessment of landfills. (Sørlie J-E *et al*. 2004). The semi-coke landfills in Kiviõli and Kohtla-Järve, and the ash landfill at Narva were chosen as typical demonstration sites for the profound research and environmental risk assessment. In addition to other investigations, the groundwater flow and contaminant transport were studied by hydrogeological modelling.
Three detached rectangular modelling sites ranging from 17 to 49 km^2 covered the landfills mentioned (Fig. 1.1.1). The completed models contained four or five model layers representing following hydrogeological units from top to bottom:

1) The semi-coke in Kiviõli and Kohtla-Järve, and ash at the Narva inside borderlines of landfills; outside of landfills – Quaternary deposits (predominantly peat, glaciolacustrine sand, varved clay in places, and till).
2) The lowermost seams of semi-coke compressed inside of landfills in Kiviõli and Kohtla-Järve; outside of landfills – the lower portion of Quaternary deposits and uppermost seams of the Ordovician carbonate bedrock. The confined Ordovician aquifer system in the Narva site.
3) The water bearing Ordovician carbonate bedrock from the Kukruse Stage to the Uhaku Stage in Kiviõli and from the Uhaku Stage to the Kunda Stage in Kohtla-Järve. The Silurian-Ordovician basin-wide aquitard on the Narva site.

4) The lower portion of the confined Silurian-Ordovician aquifer system in Kiviõli; the regional Silurian-Ordovician aquitard in Kohtla-Järve. The Ordovician-Cambrian aquifer system on the Narva site.
5) The Silurian-Ordovician basin-wide aquitard in Kiviõli, the Ordovician-Cambrian aquifer system in Kohtla-Järve. The Narva model did not comprise the 5th model layer.

The 3D models were constructed using an orthogonal uniform grid with an initial spacing of 125 m on the horizontal plane. The Visual MODFLOW v. 3.1 code was applied for modelling. The hydraulic conductivity of layers was specified using the data and experience get by previous regional hydrogeological modellings (Savitski L, Vallner L 1999a, 1999b; Vallner L 2002, 1996b). In each study area, two slug tests were made to determine the conductivity of semi-coke or ash. The net infiltration into the landfills was estimated by trial-and-error adjustment at the calibration of models. The groundwater recharge rate and the riverbed conductance outside of landfills were determined on the basis of principles discussed above in describing the model of Northeast Estonia (Vallner L 1996b).

The steady-state flow and transport simulations demonstrated that a considerable part of the landfill leachate enters the groundwater flow as a diffusive loss in Kiviõli. About a half of this diffuse loss intrudes into groundwater and another half into surface water channels in Kohtla-Järve. For that reason, a plume of contaminated groundwater is spreading beneath and around of landfills in the Ordovician bedrock. The transport of phenols was simulated by K. Rudolph-Lund based on an assumption that their half-life is 7 days. It was concluded that the spatial disposition of the contaminant plum of phenols is more or less stable because of biodegradation and natural attenuation of contaminants. Unfortunately, the boundary conditions were not specified in the description of the phenols transport model used for simulations.

According to the simulation, the initial movement of leachate intruding into the carbonate bedrock beneath the Narva ash plateaus is radial. Afterwards, the flux of leachate bends upwards and discharges in channels and ponds surrounding the ash landfills. Some portion of the leachate encroaches into the Narva Reservoir. Compared to the Kiviõli and Kohtla-Järve semi-coke landfills, the Narva ash landfills storing the power plants ash, are less harmful to the environment, since the organic pollutants are burnt up at the combustion process of oil shale.

Hydrogeological modelling of the northern catchments of Peipsi Lake in Estonia. (Le Nindre Y, Amraoul N 2005). The available project report manifested an intension to compile a groundwater model of northern catchments of Peipsi Lake on the territory of Estonia. The modelling area distinguished was rectangular and covered about 32,000 km^2 (Fig. 1.2.2). The western border of the area coincided with the longitude of Türi, the eastern one attained to the Narva Reservoir (the Narva town itself did not belong to the study area). The southern border was congruent with the latitude of Tõrva and the outcrop of the Cambrian-Vendian aquifer system on the bottom of the Gulf of Finland was included into the borders of the model in the north. The performers of the project were the French Geological Survey (BRGM) and Geological Survey of Estonia. The code MARTHE of the BRGM was used for groundwater modelling.

The water bearing formation modelled included 11 layers from top to bottom (the terminology of the project authors is followed):

1) Quaternary aquifer;
2) Middle Devonian aquifer;
3) Narva (aquifer in the upper part and aquitard in the lower part);
4) Pärnu aquifer (Devonian);
5) Ordo-Silurian aquifer system;
6) Ordovician aquitard;
7) Cambro-Ordovician aquifer;
8) Lontova aquitard;
9) Voronka aquifer;
10) Kotlin aquitard;
11) Gdov aquifer.

The determination of the net infiltration was not explained in the project report. For characterizing of hydraulic interconnection between aquifers and rivers it was assumed that the river stage was 1 meter below the local topographic surface, the riverbed was 2 m below the river stage, its hydraulic conductivity was 1 m/day, and its thickness was 0.3 m. The boundary conditions of the Cambrian-Ordovician aquifer system and of the Cambrian-Vendian aquifer system were not identified in 2005. The completion of the model ceased and no modelling results were gained.

Dewatering of the Tammiku-Kose opencast pit. Hydrogeological modelling (Lind H 2005). The area of the possible Tammiku-Kose opencast pit is situated southeast from Jõhvi being adjacent to the eastern border of the Tammiku mine. The shape of this new pit designed is oblong in the plane: its length reaches about 3 km in southeast direction but the width ranges from 100 to 450 m. The main goal of the project was to elaborate the methodology of investigation of the groundwater flow situation by means of the modelling code Visual MODFLOW v. 4.0. Thereat the area of the Tammiku-Kose opencast pit was considered as a pilot site.

The Tammiku-Kose opencast pit planned and the eastern part of the Tammiku mine closed in 2002 were included in a quadratic modelling area with a side size of 2.7 km (Fig. 1.2.1). The spacing of the quadratic computational grid was 50 m. The model consisted three layers from top to bottom as follows: the upper part of the Idavere-Kukruse aquifer, the oil shale production seam in the Kukruse Stage, and the semi-confining upper portion of the Uhaku Stage. The value of lateral hydraulic conductivity omitted to all model layers was 10 m/d. This was a too rough generalization whereas the real value of lateral hydraulic conductivity of layers modelled varied between 0.01–200 m/d. The boundary conditions used for modelling were not clarified in the project report. The net infiltration and the hydraulic impact of the river network were not accounted at modelling. Two pumping wells with a discharge rate reaching 10,000 m^3/day in all were designed for dewatering of the new pit. According to the simulation, the two planned pumping wells were not sufficient to dewater the pit. Thus, the task posed by the project was not fulfilled. Unfortunately, the performer of the project was not able to use all flexible potentialities of the Visual MODFLOW.

Estimation of the safe yield of the Kurtna-Vasavere groundwater intake until 2035 (Savitski L, Savva V 2005a). Flow modelling was used to estimate the safe yield of the Vasavere groundwater intake. The Vasavere buried valley with the KLR and surrounding mines and pits were enfolded into a quadratic modelling area (Fig. 1.2.1). The sizes of this site were about 12 km × 12 km. The model comprised five layers including the Quaternarian aquifer system (boggy, glaciolacustrine, glacifluvial, and glacial deposits), the Ordovician Keila-Kukruse aquifer, and the Uhaku aquitard. Boundary conditions of the model were not elucidated. The mode of hydraulic connection between model layers and the river network of the KLR was not explained.

The groundwater table was simulated for the study area at pumping rates corresponding to safe yields designed before. The lowering of the water table of lakes was not calculated. It was concluded in compliance with the groundwater table simulated that the safe yield of the Vasavere groundwater intake could be 8,000 m^3/day without unfavourable impact on the lakes until 2035. Moreover, it was foreseen an additional groundwater intake Vasavere-2 northward from the existing one. The safe yield of the new intake proposed was 4,000 m^3/day from 2010 until 2035. At calculation of safe yields was assumed that the pits bordering the KLR from the east would be closed in 2010.

Re-evaluating the safe yield of the Cambrian-Vendian aquifer system and the changes in water chemistry in the area of Kohtla-Järve until 2035 (Savitski L, Savva V 2005b). The need for the new evaluations was initiated by the decreasing demand for potable water supply in the area of Kohtla-Järve (Fig. 1.2.1). For that purpose, the hydrogeological model of Northeast Estonia was used (Vallner L 1996b).

The needed discharge rates were assigned to groundwater intakes of the model and corresponding distribution of hydraulic head was simulated. It was established that a safe yield reaching 13,180 m^3 per day all together would lower the hydraulic head in the Cambrian-Vendian aquifer system up to 21–24 m b. s. l. during the period from 2013 until 2035. The drawdown predicted was considered as an acceptable one despite concurring saline seawater intrusion into the Cambrian-Vendian aquifer system. This encroachment amounts to 7,300 m^3/day making 55% of the safe yield estimated. The effects of saline water intrusion on the groundwater quality were not analysed in the project report.

Life cycle analysis of the Estonian oil shale industry (Gavrilova O *et al.* 2005). The main topic of this project was the efficiency of the Estonian oil shale industry but the problems of sustainable consumption of groundwater resources in Lääne-Viru County and Ida-Viru County were profoundly studied by means of the RHME. It was done because of requirements of the Water Framework Directive (WFD) of the European Parliament and of the European Union (Directive 2000/60/EC…2000). According to the WFD, the Member States of the EU must protect, enhance and restore all groundwater bodies (GWB), ensure a balance between abstraction and recharge of groundwater, with the aim of achieving a good quantitative status and chemical status of GWB.

Pursuant to WFD definitions, the groundwater quantitative status is good when the level of groundwater in a GWB 'is such that the available groundwater resource (AGR) is not exceeded by the long-term annual average rate of abstraction'. Alterations to flow direction resulting in level changes may ‘occur in a spatially limited area, but such reversals must not cause saltwater or other intrusions’. Groundwater chemical status is good when it does not exhibit the effects of saline or other intrusions and the concentrations of pollutants does not exceed the EU quality standards. If these quantitative and qualitative requirements have not been satisfied then the groundwater status of a GWB is poor (not good) and special measures must be carried out to attain its good status by this EU Member State where the GWB situates.

In compliance with main standpoints of the WFD, it is necessary to estimate the available groundwater resources of groundwater bodies and to compare them with groundwater abstraction. If the abstraction from a GWB exceeds the AGR calculated then the quantitative state of this GWB should be assessed as a bad one.

The groundwater bodies of both, Lääne-Viru County and Ida-Viru County, were bordered before (Fig. 1.2.5) as water management monads (Põhjaveekogumite veeklassid…2003; Viru-Peipsi…2004). In frames of the project (Gavrilova O *et al.* 2005) every GWB was distinguished from the other water bearing layers as a special water budget zone of the RHME. All budget zones delineated formed a united hierarch water budget system. Then all pumping wells were deactivated in the model and a detailed water budget of each GWB was simulated for the predevelopment (natural) conditions by means of the RHME. The available resource was estimated as a natural inflow into the GWB under consideration less the outflow needed for the formation of the available resource of the underlying GWB (Table 1.2.1).

Accordingly, to water budget simulation, the abstraction from the Quaternarian Vasavere GWB, Silurian-Ordovician GWB, and Ordovician-Cambrian GWB was less than their AGR in 2003. The safe yields of these groundwater bodies certified by the Estonian Ministry of the Environment were acceptable, except the official safe yield planned for the Vasavere GWB. A pumping rate reaching 12,000 m^3/day (Savitski L, Savva V 2005a) would likely cause an unfavourable lowering of the water table in the Kurtna Lakes. The amount of water pumped out from mines and opencast pits exceeded the AGR of the Ordovician carbonate bedrock in the Ida-Viru oil shale basin more than five times. In 2003 the abstraction from the Voronka GWB and from the Gdov GWB was two-three-fold more than their AGR. The safe yield certified for Cambrian-Vendian groundwater bodies exceeded the AGR by five-seven times!

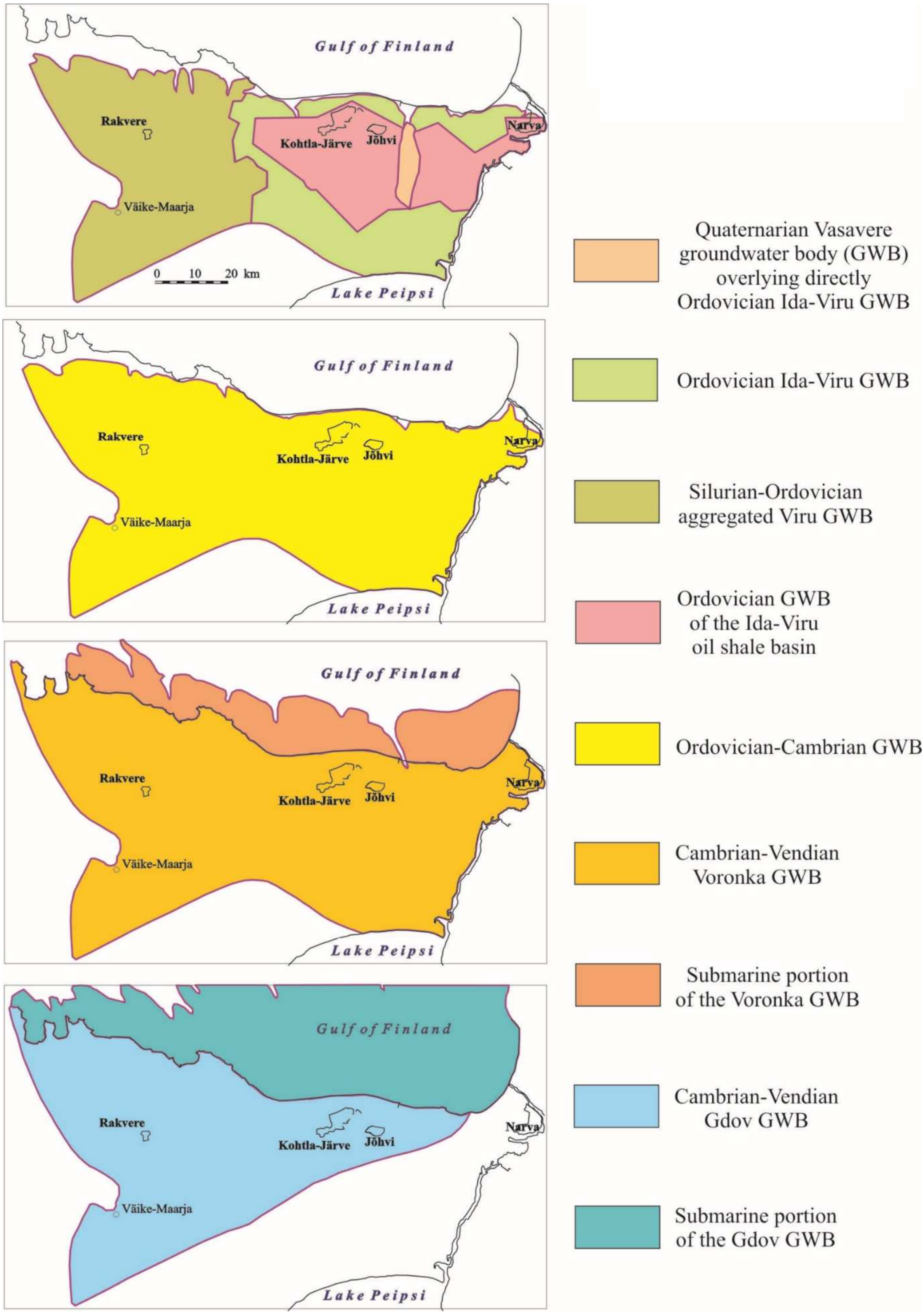

Fig. 1.2.5. Groundwater bodies of West-Viru County and East-Viru County (Põhjaveekogumite veeklassid…2003; Viru alamvesikonna…2006; Viru-Peipsi…2004).

Human impact on water resources in Virumaa (Vallner L 2008b). This study upgraded the former investigations presented in the above report (Gavrilova O *et al*. 2005). The additional data on groundwater abstraction rates and proved reserves (Perens R, Savva V 2007b, 2006) were taken into account. Apart from the abstraction problems the sea water encroachment was investigated by modelling. It was demonstrated that an inverse flow from seaside toward the coast lasted in the Cambrian-Vendian aquifer system during 60 years because of too intensive pumping (Fig. 1.2.6).

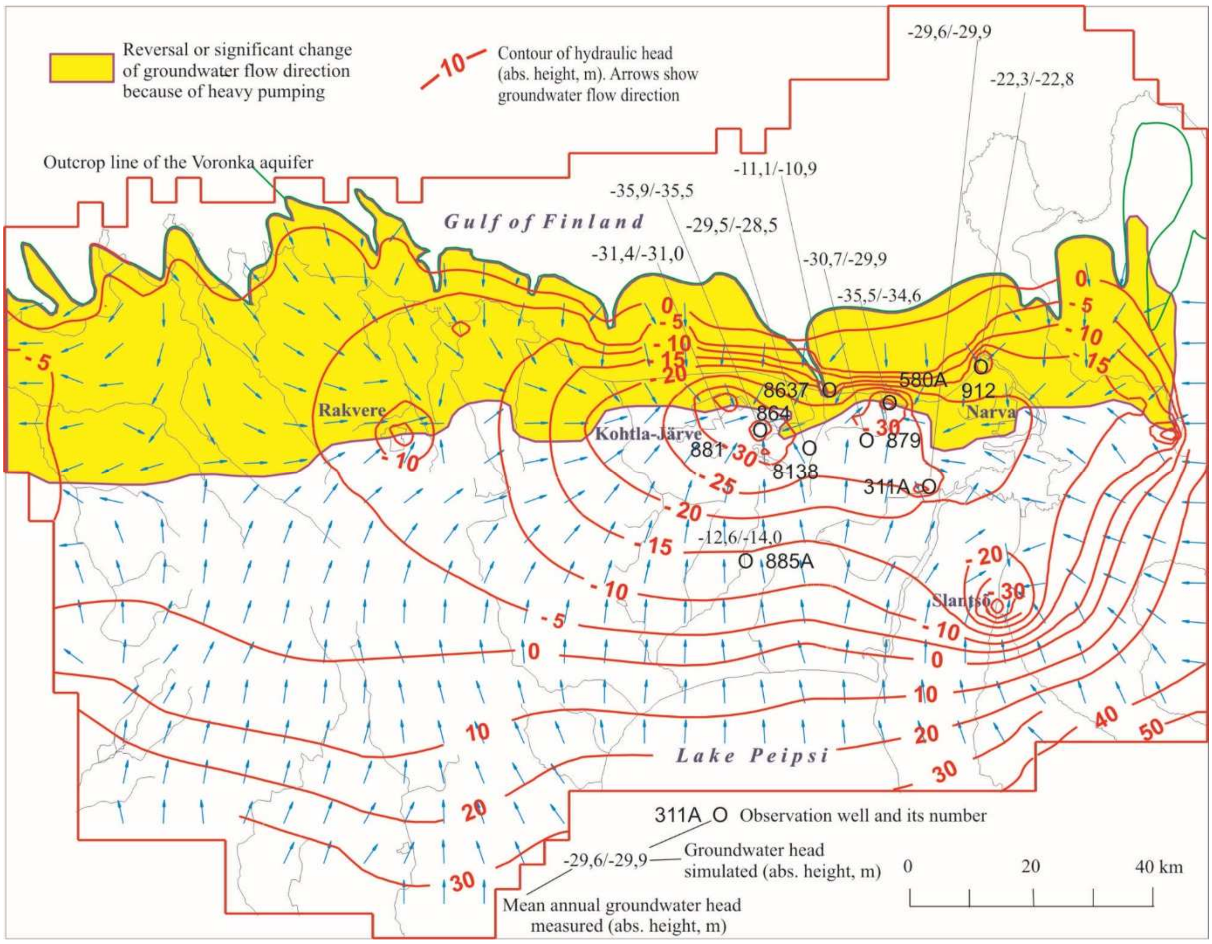

Fig. 1.2.6. Groundwater head and flow direction in the Voronka aquifer in 1991.

The available resources, the certified safe yields, the actual abstraction, and their relations were juxtaposed for every GWB studied in Tables 1.2.2 and 1.2.3. It rendered feasible to assess the general status of groundwater bodies (Table 1.2.3). The groundwater status was assessed on the basis of the both, actual and planned groundwater abstraction. It allowed evaluating the water management projects resting on officially certified groundwater safe yields.

Following the prescriptions of the WFD (Directive 2006/118/EC…2006; Directive 2000/60/EC…2000) the general status of a groundwater body was assessed by the poorer of the quantitative and chemical state of groundwater.

Table 1.2.1. Predevelopment water budget of groundwater bodies and their available resource, m^3/day* (Gavrilova O *et al*. 2005; Vallner L 2008b)

Groundwater body	Inflow				Outflow					Available groundwater resource
	From above	Lateral	From below	Total in	Up	Lateral	Down	Into channel network	Total out	
Quaternary Vasavere	21,600 (infiltration)	13,300	0	34,900	0	8,000	300	26,600	34,900	8,000
Ordovician of the Ida-Viru oil shale basin	239,400 (infiltration) + 300	79,700	300	319,700	0	116,000	1,400	202,300	319,700	104,000
Ordovician, Ida-Viru area	288,400 (infiltration)	225,200	2,800	516,400	0	208,500	8,300	299,600	516,400	208,000
Silurian-Ordovician, Viru area	763,300 (infiltration)	98,800	600	862,700	0	166,500	16,500	679,700	826,700	167,000
Ordovician-Cambrian	26,200	1,000	0	27,200	4,200	3,200	5,000	14,800 (spring discharge on Klint)	27,200	20,000
Cambrian-Vendian Voronka, terrestrial part	3,500	2,200	500	6,200	<100[1]	3,100	3,100	0	6,200	3,000
Cambrian-Vendian Gdov	3,000	4,100	0	7,100	600	6,500	0	0	7,100	5,000

*Flow rate <100 m^3/day has not been accounted at summation.

The status of some groundwater bodies was assessed as critical in the Table 1.2.3. It means that the status of these groundwater bodies may easily turn to a bad one in the future. An effective monitoring is needful to detect the unfavourable changes in time.

In the framework of the study (Vallner L 2008b), it was shown that the groundwater abstraction from the Quaternarian Vasavere GWB was less than the available resource and, consequently, acceptable in 2006. The chemical status of the GWB was critical because of possible intrusion of water enriched by excessive amounts of sulphates from the Ahtme underground mine. Thus, the general status of this GWB was assessed as critical (Table 1.2.3). The safe yield calculated ranging 12,000 m^3/day exceeded the maximum pumping rate of the Vasavere groundwater intake recorded in 1995–1999 by 1.2 times. Unfavourable lowering of the water tables was greatest during this period of intensive abstraction. Therefore, by using up the whole safe yield, the quantitative status, as well as the general status of the Quaternarian Vasavere GWB, will be bad.

The chemical status and the quantitative status of the Ordovician GWB in the Ida-Viru oil shale basin were undoubtedly bad. Consequently, its general status was also bad. Moreover, this GWB engenders steadily surrounding groundwater bodies and especially the underlying Ordovician-Cambrian GWB by spreading variable contaminants.

The Ordovician GWB in the Ida-Viru area and the Silurian-Ordovician GWB of the Viru area were in good chemical status and practically not suffering from industrial pollution sources. The groundwater abstraction was significantly less than its available resource. The general status of these groundwater bodies was good.

In 2006 the groundwater abstraction from the Ordovician-Cambrian GWB did not exceed its AGR and, therefore, the quantitative status of this GWB was good. However, the encroachment of contaminated water from semi-coke and ash landfills through the Lower Ordovician aquitards into the Ordovician-Cambrian GWB in Kohtla-Järve and Ahtme was detected. There is a potential risk that the contamination coming from overlying layers may spread in the Ordovician-Cambrian GWB. Therefore, the chemical status of the GWB was assessed as critical. For that reason, the general status of the Ordovician-Cambrium GWB was also assessed as critical.

The terrestrial part of the Cambrian-Vendian Voronka GWB is not vulnerable to polluting leakage from overlaying strata. Therefore, its chemical status is still good. However, the abstraction from the GWB exceeded the available resource 3 times in 2006 and pumping allowed (safe yield) was 10.5 times greater than the available resource. The natural seaward flow was replaced by a backward flow lasting more than 60 years because of excessive abstraction. This situation will continue until 2035 and there is a risk of saline sea water intrusion into the coastal intakes tapping the Voronka aquifer. On that account, the quantitative status and, respectively, also the general status of the Voronka GWB was bad in 2006.

In 2006 the abstraction from the Cambrian-Vendian Gdov GWBs was two-fold more than the available resource. The certified safe yield until 2012 exceeded the available resource 6.3 times. Despite a later reassessment and decreasing of the safe yield (Savitski L, Savva V 2005b) its value certified for the period 2013-2035 still will exceed the available resource by 5 times. Due to heavy pumping exists a long-term anthropogenic groundwater flow from seaside to the coast as well an upconing of brines from the underlying crystalline basement into the Gdov aquifer. It makes the chemical status of this GWB critical. Because of the bad quantitative status, the general status of the Gdov GWB was assessed as bad in 2006.

The critical or bad status of groundwater bodies estimated demonstrates that the lawful rules and methods used for determination of groundwater resources (Keskkonna-alaste...1998; Põhjaveekogumite veeklassid...2004) need a profound audit.

Table 1.2.2. Water management characteristics of groundwater bodies, m^3/day (Vallner L 2008b)

Groundwater body	Available resource (*D*)	Abstraction in 2006 (*B*)	Certified safe yield (*Q*)	Ratio *B/D*	Ratio *Q/D*
Quaternarian Vasavere	8,000	4,284	8,000	0.54	1.00
Ordovician Ida-Viru	208,000	No data (<<208,000)	Not determined	Not determinable	Not determinable
Ordovician of the Ida-Viru Oil shale basin	104,000	463,714 (dewatering of mines and pits in Ida-Viru County)	Not determined	4.46	Not determinable
Silurian-Ordovician aggregated Viru	167,000	4,915	5,730	0.03	0.03
Ordovician-Cambrian	20,000	3,490	10,760	0.18	0.54
Cambrian-Vendian Voronka (terrestrial part)	3,000	9,128	31,443 (until 2012)	3.04	10.48
Cambrian-Vendian Gdov (terrestrial part)	5,000	10,231	31,487 (until 2012)	2.05	6.30

Table 1.2.3. Status of groundwater bodies accordingly to requirements of the WFD in North-East Estonia (Directive 2006/118/EC ...2006; Directive 2000/60/EC...2000; Vallner L 2008b)

Groundwater body	Chemical status	Quantitative status accordingly to pumping rate in 2006	Quantitative status at the consumption of whole certified safe yield	General status in 2006	General status at the consumption of whole certified safe yield
Quaternarian Vasavere	Critical	Good	Good	Critical	Critical
Ordovician Ida-Viru	Good	Good	Not determinable	Good	Not determinable
Ordovician of the Ida-Viru Oil shale basin	Bad	Bad	Not determinable	Bad	Not determinable
Silurian-Ordovician aggregated Viru	Good	Good	Not determinable	Good	Not determinable
Ordovician-Cambrian	Bad	Good	Good	Bad	Critical
Cambrian-Vendian Voronka (terrestrial part)	Good	Bad	Bad	Bad	Bad
Cambrian-Vendian Gdov (terrestrial part)	Critical	Bad	Bad	Bad	Bad

Groundwater monitoring in the Ojamaa mining district (Perens R, Savva V, Boldõreva N 2006). It was declared that a groundwater model of the Ojamaa mining district and the Muraka Moor was completed by means of the code GMS v. 3.1 (Fig. 1.2.1). Any more information about the model characteristics was not given. A map of the groundwater table of the Keila-Kukruse aquifer simulated for 2005 was presented.

Groundwater flow model of oil shale mining area (Lind H 2010). It was alleged that a groundwater flow model was completed covering an area of 1,650 km^2 where Aidu, Vanaküla, Kohtla, Sompa, Tammiku, Viru, Estonia, and Ahtme oil shale mines were situated. The borders of the modelling area were not shown in the paper. The most of the paper content was devoted to the retelling of well-known fundamentals of groundwater modelling. The only result presented was the water flow calculated from Ahtme mine into the Estonia mine ranging from 27,000 m^3/day to 42,800 m^3/day. The interval of dates belonging to flow rates calculated was not specified.

A life cycle environmental impact assessment of oil shale produced and consumed in Estonia. (Gavrilova O, Vilu R, Vallner L 2010). The data of the AGR determined with groundwater modelling for Ida-Viru County, the real groundwater abstraction, and their conformity to requirements of the WFD (Gavrilova O *et al.* 2005, Vallner L 2008b) were discussed in this paper.

2. MODELLING PLATFORM

To elaborate an effective monitoring system supporting the optimum groundwater utilization and protection in Ida-Viru County it is needful the development a functionally coupled system of groundwater models. An acceptable groundwater regime should be determined by means of these models.

Such groundwater regime must satisfy requirements of the WFD (Directive 2006/118/EC…2006; Directive 2000/60/EC…2000). Accordingly, to main demands of these documents, the pumping rate must not exceed the AGR and harmful intrusions have not been allowed into a GWB. Concentrations of pollutants must not outnumber the EU quality standards.

Some circles of the water management staff of Estonia contend that the restrictions of the WFD are too rigid for a working groundwater consumption and protection. Some decision makers are thinking that it is not important if at a needful pumping rate the front of a saline water intrusion would reach a groundwater intake, say, after a hundred years. Then the water politics will be completely dissimilar and we should not care about it at present. Though this standpoint is not passable from the WFD position, nevertheless it could be discussed in some extent. The rational deviations from the WFD requirements should be an object of a social agreement founded on a profound economical and hydrogeological analysis. The arguments and criterions for such analysis should be obtained by a comprehensive and trustworthy groundwater modelling.

The heights of the groundwater table and hydraulic heads characterising the sustainable and acceptable groundwater regime must be simulated by the system of groundwater models developed. The same way the threshold values of the groundwater chemistry (Marandi, A 2007; Marandi A, Karro E 2007) must be established for both regimes mentioned. These checking criterions should be juxtaposed with the data of groundwater monitoring network to assess the groundwater status.

Some of the existing monitoring points may not satisfactorily represent the current status of groundwater. Therefore, the most suitable localities of observations points, monitoring objects, and frequency of measuring or sampling should be determined by the sensibility analyses of the model system.

The system of groundwater models must be regularly calibrated against the variable sets of monitoring data. It enables to achieve the best trustworthiness of models and consolidate an effective feedback between monitoring and modelling.

The modelling system should consist of a regional basic model enfolding the whole water-bearing formation of North-East Estonia and its outcrop on the bottom of the Gulf of Finland, and of some local models representing landfills, oil shale mines, groundwater intakes, landscape reserves etc. All models must form a united water budget and transport system. Thereat, the transport modelling of liquids of different densities must be carried out to investigate the development of saline water intrusions in submarine portions of the Cambrian-Vendian aquifer system. The modelling system must work in both, steady state and transient state.

The hydrogeological model of Estonia (Vallner L 2003, 2002) is the most suitable base to develop a coupled groundwater modelling system for Ida-Viru County. All hydrogeological and hydrological data of the model of North-East Estonia (Vallner L 1996b) used at seven projects completed have been incorporated into the regional model of Estonia (RHME). This model is well calibrated and verified by different sets of data at performing of variable tasks (Bruin de E *et al*. 2006; Gavrilova O *et al*. 2008, 2010; Marandi A, Vallner L 2010; Perens R *et a*l. 2006; Vallner L 2008, 2011). It includes all main aquifers and aquitards from the ground surface as low as the waterproof portion of the crystalline basement.

For the present, the RHME has been updated by including additional local aquifers and aquitards into the Silurian-Ordovician aquifer system. They are Vormsi aquitard, Nabala-Rakvere aquifer, Oandu aquitard, Keila-Kukruse aquifer, Uhaku aquitard, and Lasnamäe-Kunda aquifer. Further splitting of model layers representing the Quaternarian aquifer system and Keila-Kukruse aquifer is possible. The sophisticated problems of the groundwater transient transport can be solved by means of the RHME coupled with the SEAWAT engine (Marandi A, Vallner L 2010). It enables a detailed investigation of the man-made hydrogeological situation in the Ida-Viru County.

The RHME has been converted to the code Visual MODFLOW v. 4.5 (2010.1). This is the most popular, complete, and user-friendly modelling environment suitable for a profound research as well for training and demonstration purposes. This software is recognized, accepted, and used by more than 10,000 groundwater professionals in over 90 different countries around the world.

Thus, the RHME could be the regional basic groundwater model of the Ida-Viru County. Provisionally it appears to be a too big one (enfolding the whole Estonia) but this is not important accounting the capability of modern computers. On the contrary, it enables to estimate the mutual hydrogeological influence between the Ida-Viru County and adjacent regions of Estonia and Russian Federation covering a wider area than other models (Perens R, Savva V. 2007a). This creates a productive platform for the further development of an integrated monitoring system supporting sustainable groundwater consumption and protection in whole Estonia.

The local models should be developed based on the RHME. The main hydrogeological parameters can be incorporated into local models directly from the data files of the RHME. Thereat, the additional data, derived from field experiments and complementary monitoring can be used for the adjustment of local models. The boundary conditions and the initial condition of local models should be determined in accordance with simulation results obtained by running of the RHME. It will enhance the trustworthiness of simulation results get by local model.

3. DEVELOPMENT OF HYDROGEOLOGICAL MODELS OF LANDFILLS

3.1. General goal

The groundwater models of Kohtla-Järve, Kiviõli, and Narva landfills completed formerly by L. Vallner (Sørlie J-E *et al.* 2004) were significantly developed in course of the current research to improve their predictive power. For that purpose, the input parameters of models were corrected and adjustable simulation series were carried out. It allowed to enhance the trustworthiness of models and profoundly to analyse the speeds and rates of groundwater flow depending on values and exactness of hydrogeological input characteristics. A special approach was elaborated to overcome the imperfection or lack of groundwater transport parameters. It enabled to specify the development of contaminant plumes both in a horizontal and vertical direction depending on natural biodegradation and attenuation of contaminants in course of time.

In this connection, a short review is given below considering contemporary principal standpoints of groundwater contamination modelling at first. It is necessary for a better understanding of principles, terms, and notifications performed further. After it, the layers modelled are characterised and a critical review of input data used for completing the landfill models is represented. Next, the landfill models completed are described and the results of simulations are analysed.

3.2. Modelling of groundwater contamination

For the assessment of environmental risks induced by groundwater contamination, it is necessary to predict the pathways of pollutants and their spreading time in water-bearing layers. The hydrogeological digital modelling is a most efficient and authoritative tool for managing such task. This method lies in solving a system of fundamental differential equations describing the groundwater flow and transport in a porous or fractured media.

The movement and modification of variable groundwater ingredients including potential contaminants are known as mass or solute transport. Solutes transported in groundwater are subject to processes of attenuation (Fetter CW 1993).

Diffusion will cause solutes to move in the direction of the concentration gradient – from areas of higher concentration to lower concentration. This process is known as molecular diffusion. It is especially important to point out that diffusion will occur as long as a concentration gradient exists, even if the fluid is not moving. Diffusion may be the major factor in mass transport in geologic materials of very low permeability.

The process of advection (also known as convection) also transports solutes. It occurs as the flowing groundwater carries the dissolved solutes with it. At the scale of a few pore diameters, groundwater will move parallel to the flow path. The flow rates are different because of differences in pore sizes. This causes the solute plume (a domain where the concentration of solute differs from the concentration of surrounding natural groundwater) to spread along the direction of the flow path. This is the longitudinal dispersion. The solute plume will also spread laterally as flow paths diverge around mineral grains, a process known as transverse dispersion. The sum of longitudinal dispersion and transverse dispersion is considered as an expression of hydrodynamic dispersion.

Besides advection-dispersion process, a number of solutes are removed from solution by sorption, chemical reaction, and biological and radioactive decay. Sorption of inorganic solutes occurs primarily on mineral surfaces and is a function of the surface area available for sorption. Organic and radioactive components of a solute can decay. As a result, some solutes will move much more slowly through the layers than the groundwater transporting them. Solutes can also attenuate because of sorption. These effects are known as retardation and as attenuation of solutes. Special mathematical functions describing the disappearance of a solute due to sorption and biological or radioactive decay have been elaborated.

The partial differential equation describing the fate and transport of contaminants of species of k in 3D, transient groundwater flow system is (Zheng C. 1999; Zheng C, Wang P 2003, 1998; Yu C, Zheng C 2010):

$$\partial(\theta C^k)/\partial t = (\partial/\partial x_i)\,(\theta D_{ij}\partial C^k/\partial x_i) - (\partial/\partial x_i)\,(\theta\partial v_i\, C^k) + q_s\, C^k{}_s + \Sigma R_n \qquad (3.2.1)$$

where

C^k is the dissolved concentration of species k, [ML^{-3}];

θ is the porosity of the subsurface medium, (dimensionless);

t is time, [T];

x_i is the distance along the respective Cartesian co-ordinate axis, [L];

D_{ij} is the hydrodynamic dispersion coefficient tensor, [L^2T^{-1}];

v_i is the seepage or linear pore water velocity, [LT^{-1}]; it is related to the specific discharge or Darcy flux through the relationship $v = q_s/\theta$;

q_s is the volumetric flow rate per unit volume of aquifer representing fluid sources (positive) or sinks (negative) [T^{-1}];

$C^k{}_s$ is the concentration of the source or sink flux for species k [ML^{-3}]:

ΣR_n is the chemical reaction term [$ML^{-3}T$].

A solution of equation (3.2.1) is possible at the known 3D distribution of dissolved concentration C^k for the moment $t = 0$ in the groundwater domain (the initial condition), the boundary conditions of solute concentration must be established beforehand, too.

The transport equation (3.2.1) is related to the flow equation (1.1.1) through the Darcy's Law:

$$v_i = q_i / \theta = - (K_i / \theta)(\partial h/\partial x_i) \qquad (3.2.2)$$

where

K_i is a principal component of hydraulic conductivity tensor, [LT^{-1}]:

h is hydraulic head [L].

It means that to solve the equation (3.2.1) for a certain hydrogeological environment the equation (1.1.1) must be solved for the same environment beforehand.

There are three types of boundary conditions for mass transport (Fetter CW 1993). The boundary condition of the first type is a fixed concentration. The boundary condition of the second type is a fixed concentration gradient. A variable flux boundary constitutes the boundary condition of the third type.

Variable codes of hydrogeological modelling enable the numerical solution of fundamental equations (1.1.1) and (3.2.1). Most of these codes contain a coupled solution of both, flow and transport equation (Visual MODFLOW…2006, 2010, etc.)

The main problem of hydrogeological modelling is the trustworthiness of available parameters characterizing flow and transport properties of water-bearing layers. The parameters can be determined by laboratory or field tests.

The laboratory tests are mostly carried out by means of columns packed with the porous media under investigation. In general, the average linear velocity of the fluid as well diffusion and dispersivity characteristics of the porous media in the column can be found from the quantity of fluid discharging per unit time divided by the product of the cross-sectional area and porosity. However, only a very small portion of a sandy or loamy water-bearing layer can be investigated by a laboratory test. Especially difficult is to test a hard fractured rock (limestone, sandstone, granite). Therefore, results of laboratory tests are often not representative enough.

Dispersivity can be determined by two means in the field. If there is a contaminated aquifer, the plume of known contamination can be mapped in accordance with data of observation wells and the transport equation solved with dispersivity as unknown. It is possible to modify initial guesses about the values of longitudinal and transverse hydrodynamic dispersivity during model runs until the computer model will yield a reasonable reproduction of the observed contaminant plume. One of the most serious difficulties of this approach is that the concentration and volume of the contaminant source are often not satisfactorily determined by means of observation wells and groundwater sampling.

A much more common approach is the use of a tracer injected into the water-bearing layer via a well (Fetter C 1993). There are numerous variations of this method. Natural gradient tests use the injection of a tracer into an aquifer, and require the measurement of the contaminant plume that has developed under the existing gradient of the hydraulic pressure. The plume is measured by means of small amounts of water pumped out from down-gradient observation wells and multilevel piezometers. A single well tracer test involves the injection of water containing a conservative tracer into an aquifer through an injection well and then the subsequent pumping of that well to recover the injected fluid. Formulas have been derived out to calculate the dispersivity parameters based on injection and pumping rates measured at the test.

Pumping tests, tracer observations, resistivity measures, groundwater samplings, etc. perform data indeed for the point where they were carried out but an extrapolation of their values to a further distance, say, until one kilometre, is quite questionable. Therefore, to get reliable data on groundwater contamination parameters several testing sites should be placed in a study area.

3.3. Hydrogeological units modelled

Due to the similarity of the hydrogeological conditions in Northeast Estonia, the models of all landfills enfold the same hydrogeological units. They are from top to bottom as follows:

The dump deposits (dQ_{IV}) are the ash and semi-coke of oil shale. They have been heaped as landfills which relative height reaches 100—160 m in Kiviõli and Kohtla-Järve. The fresh semi-coke dumped is a granulose gritty substance with consistence similar to moist sandy loam in field conditions. It consists predominantly of calcite, illite, ettringite, quartz, K-feldspar, and dolomite (Mõtlep R *et al*. 2007).

The layer of the fresh semi-coke having a thickness of 0.5 m is underlain by a formation of conglomeratic semi-coke cemented with a significant amount of detritus. The lower surface of conglomeratic semi-coke is between 68–70 m a. s. l. in Kohtla-Järve (Metsur M 2005). This formation is mostly unconfined with lenses of perched water and predominantly unsaturated. Its porosity is 0.63–0.71 and, therefore, the bulk density is 0.9–1.3 g/cm^3, and dry bulk density is 0.7–0.9 g/cm^3. The mass density is about 2.3 GS (Tööstusjäätmete ja…2007, 2006; Viru Keemia…2004). Beneath the conglomeratic formation as low as the natural ground surface lies the layer of cemented schistose semi-coke saturated with leachate. Its porosity is 0.62–0.77, the bulk density is 1.1–1.4 g/cm^3, dry bulk density is 0.5–0.8 g/cm^3, and the mass density is 2.0–2.4 GS (Metsur M 2005; Tööstusjäätmete ja…2007, 2006; Viru Keemia…2004). The hydraulic conductivity of semi-coke layers ranges from 0.01 to 0.028 m/day accordingly to five field tests described in the next paragraph 3.4.

The portion of kerogen left in semi-coke after retorting reaches up to 23% in places (Metsur M 2005; Tööstusjäätmete ja…2007, 2006; Viru Keemia…2004). The kerogen buried can ignite spontaneously and cause long-term landfill burnings. A mechanical mixture of heavy fractions of oil, fine particles of coke and water of variable composition called the fuss, forms up to 1.6% of the mass of semi-coke in landfills (Sørlie J-E *et a*l. 2004). A pond of fuss covering an area approximately of 3,500 m^2 was situated in the central part of the Kohtla-Järve landfill.

At the thermal power plants, the oil shale organic matter is completely burned. The main component of ash is calcium oxide (lime), furthermore considerable are portlandite, magnesium oxide, anhydrite, calcite, quartz, and clay minerals (Bityukova L, Mõtlep R, Kirsimäe K 2010). The lower portion of ash and semi-coke storages with the thickness up to 10–20 m in Kiviõli and Kohtla-Järve and the ash plateaus at Narva is saturated by leachate containing contaminants. The hydraulic conductivity of ash ranges from 0.01–0.06 m/d in compliance with three slug tests performed (Table 3.4.1).

Quaternary deposits (Q) consist predominantly of glacial till covered by glaciolacustrine sand and sandy loam or varved clay (Perens R, Vallner L. 1997). Their thickness ranges from 0.5 to 30 m. In boggy areas, the uppermost portion of Quaternary deposits is represented by a peat layer, which thickness usually does not exceed 3 m. Water table conditions prevail in Quaternary deposits. The lateral conductivity of Quaternary deposits is mostly 0.1–3 m/day, their vertical conductivity changes in an interval of 10^{-2}–10^{-1} m/day. The transversal conductivity of varved clay may be less than 10^{-4} m/day. The depth of the water table from the ground surface around landfills varies mostly from 1 m to 3 m in the period of the summer low flow. The rate of net infiltration into the natural Quaternary cover is predominantly 60–80 mm/year (Vallner 1996b).

The Silurian-Ordovician aquifer system (S-O) lies directly beneath the Quaternary cover and is represented by diverse Middle and Lower Ordovician limestones with clayey interbeds forming local aquitards in places. This part of the geological section having a total thickness up to 50 m comprises of seams from the Kukruse Stage to the Kunda Stage (Perens R, Vallner L 1997; Raukas A, Teedumäe A 1997). The limestones are fissured and heavily karstified in spots. Therefore, they can easily become polluted. The lateral conductivity of the carbonate bedrock changes usually from 2 to 20 m/day and the storage coefficient is between 10^{-6}–10^{-3} 1/day depending on the degree of fissuration and karstification. Effective porosity of the fissured carbonate not containing significant karst conduits ranges mostly from 0.02 to 0.03. The transversal conductivity of the local aquitards is 10^{-5} m/day or even less. In the places where the thickness of the Quaternary cover does not exceed a couple of metres, the uppermost portion of the carbonate bedrock belongs to the unsaturated zone. The saturated zone of the carbonate bedrock lying on a local or regional aquitard is in water table conditions. The average specific yield of the unconfined portion of the bedrock is from 0.02 to 0.05. Under an aquitard, the bedrock layers are confined. The water of the S-O aquifer is mostly polluted and not suitable for drinking in study areas (Viru alamvesikonna... 2006). The Silurian-Ordovician aquifer system is recharged from overlying Quaternary cover including the downward flux from the landfills and with lateral flows coming from local watersheds. The Silurian-Ordovician aquifer system is not suitable for a public water supply in the oil shale mining area (Gavrilova O, Vilu R, Vallner L 2010; Viru alamvesikonna... 2006).

The Silurian-Ordovician regional aquitard ($S–O_{aquitard}$) enfolds the Lower Ordovician layers from the Volhovi Stage to the Varangu Stage (Perens, Vallner 1997; Raukas, Teedumäe 1997). They consist of limestones, marls, siltstones, clays, and argillites with a total thickness of 2 to 10 m. The transversal conductivity of the aquitard is 10^{-7}–10^{-5} m/day. This aquitard prevents spreading of polluted water from S-O aquifer downwards to a certain extent.

The Ordovician-Cambrian aquifer system (O-Є) is represented by fine-grained sandstone and siltstone with a total thickness of 20 m. This confined aquifer system is a significant source for public water supply in Northeast Estonia. Its lateral conductivity is mostly 2–4 m/day. The well yields are predominantly 400–600 m^3/day per 10–15 m of drawdown. The storage coefficient is from 2.5×10^{-5} up to 6×10^{-3} 1/day; the specific capacity of a drained aquifer is 0.12–0.14 m^2/day. The O-Є aquifer system is recharged with downward flux coming from S-O aquifer system and penetrating the Lower Ordovician aquitard. Due to an intensive water extraction, the regional head depressions have been formed with centres southward from Kohtla-Järve and in Slancy (Russian Federation). Water moves toward the local centres of the head depression or in direction North-Estonian Klint draining the O-Є aquifer system.

The Lükati-Lontova regional aquitard ($Є_1$Lk-$Є_1$Ln) with the thickness of 70 m consists of siltstones and clays which transversal conductivity is 10^{-6}–10^{-9} m/day. The intensity of downward flows penetrating this aquitard is very small. Therefore, the lower impermeable boundary of landfill models has been combined with the upper surface of the Lükati-Lontova regional aquitard. Under the Lükati-Lontova aquitard lays the principal source of the public groundwater supply of North-Estonia, the Cambrian-Vendian aquifer system on the crystalline basement (Perens R, Vallner L 1997). The Lükati-Lontova aquitard and the Cambrian-Vendian aquifer system are not incorporated into landfill models.

3.4. Input data

Hydraulic conductivity of layers modelled is estimated by former single-well pumping tests for few points only in the landfill study areas. Non the less, the conductivity of regional hydrogeological units described above was predominantly specified for landfill models on the basis of data and experience get with hydrogeological mapping (Érisalu É *et al*. 1965; Saadre T *et al*. 1987; Suuroja K *et al*. 2009a, 2009b, 2008; Tassa V 1967; etc.), investigation of groundwater intakes (Perens R *et al.* 2005, Perens R, Savva V. 2007b, 2006; Savitski L, Savva V. 2005a, 2005b, 2001a, 2000a, 2000b; Savitski L, Vallner L 1999a, 1999b; etc.), mining studies (Erg K 2005; Riet 1976; Savitski L, Savva V 2001b, 2001c; etc.), and completing of previous hydrogeological models (Vallner L 2003, 2002, 1996b). Real conductivity values of bedrock aquifers can alter by several times and lateral conductivity of Quaternary deposits ten-folds or even more in dependence on the locality of the point under consideration. Variations of the transversal conductivity of aquitards can merge in several magnitudes. However, it is hoped that the quite truthful value of conductivity for a certain study area can be estimated by a profound model calibration as it was elucidated above.

In each study area, 1–2 single-well slug tests were made (Sørlie J-E *et al.* 2004) to determine the conductivity of ash or semi-coke (Table 3.4.1). The bottom seam of landfills consisting of dump sediments was characterized by one of these tests for Kohtla-Järve and Narva landfills. Another test was carried out 10–20 m higher than the lower test point. Accordingly to these experiments conductivity of semi-coke was 0.02 m/d in Kohtla-Järve and conductivity of oil shale ash ranged between 0.01–0.06 m/d. The relative error of these data may reach 200% or even more because of filtrational heterogeneity of layers tested and of the methodological inexactness of single-well slug tests (Mills A 2010).

Table 3.4.1. The results of slug tests

Study area	Well	Altitude, m a.s.l.	The composition of the layer tested	Conductivity, m/d
Kiviõli	RA-KV-6SL	54	Till and limestone	0.18
Kohtla-Järve	RA-KJ-5SU	81	Semi-coke	0.08
Kohtla-Järve	RA-KJ-6SL	69	Oil shale ash	0.02
Narva	RA-N-2SU	55	Oil shale ash	0.06
Narva	RA-N-3SL	35	Oil shale ash	0.01

Four shallow perforations were dug into the flat portion of Kohtla-Järve semi-coke landfill (Metsur M 2005; Tööstusjäätmete ja…2007, 2006; Viru Keemia…2004). The borings, having a depth of 0.2 m and a diameter of 30 mm, were sunk into the bottom of perforations. The hydraulic conductivity of semi-coke was determined by a Swedish device GeoN Permeameter Pi301 (BAT) in these borings. The values of conductivity gained were 0.010, 0.020, 0.023, and 0.028 m/day. Unfortunately, the size of the measuring sphere of the permeameter used did obviously not exceed a couple of meters. Therefore, a generalization of some single conductivity values got by means of the permeameter for the whole semi-coke formation of a landfill was not grounded enough.

Groundwater heads and quality monitoring was started in the area of the Kohtla-Järve landfill in 1992 (Razgonjajev A 1997; Razgonjajev A, Timaškin R, Razgonjajeva L 1993). The groundwater head was measured once a month in 24 borings tapping the S-O and O-Є aquifer system until 1998 and in 2001. The water table in dump deposits of the landfill lower portion was registered in two borings in July 2003 (Sørlie J-E *et al.* 2004) and in six borings in August 2004 (Metsur M 2005). Standard chemical analyses (Cl^-, SO_4^{2-}, HCO_3^-, Ca^{2+}, Mg^{2+}, K^+, and Na^+ determined) of water sampled from all observation wells were made in 1992. Since 1996 until 2008, the content of aromatic hydrocarbons (PAH), BTX, phenols, also oils, and some harmful elements (As, Hg, Mb, and Zn) was analysed (Fenoolide seire…2007; Kirde-Eesti…2008). Up to 14 borings opening both the S-O and O-Є aquifer system were sampled mostly 2–3 times yearly. The content of 46 trace elements was determined in the water of the same sampling points in 2003 (Sørlie J-E *et al.* 2004). In 2010 in the area of the Kohtla-Järve landfill, the water of 11 borings was sampled three times for analyses of PAH, BTEX, and phenols by L. Bityukova (Bityukova L *et al.* 2011). All head and concentration observation wells used for model calibration have been shown in Fig. 3.5.1.1.

The water of 9 borings was sampled and its head measured In the Kiviõli study area once in 2003 (Fig. 3.5.2.1). The content of PAH, BTX, phenols, and 46 trace elements were investigated.

Groundwater monitoring of the ash plateau at the Balti PP (Narva) has been carried out since 1991 (Sahnovskij B 1993, 1995). Groundwater head was measured and water sampled in 42 borings yearly twice until 2008 (Fig. 3.5.3.1). The standard chemical analyses and analyses for petroleum products, PAH, BTEX, phenols, trace elements were performed (Kivit N 2008a, 2008b, 2007a, 2007b, 2006a, 2006b, 2005a, 2005b, 2004a, 2004b, 2003a, 2003b, 2002, 2001, 2000, 1999, 1998, 1997; Sørlie J-E *et al.* 2004).

The data of groundwater monitoring and episodic measuring mentioned were used for creating landfill models. The elevation of the groundwater table and the potentiometric head along layer boundaries of model areas was given based on former basin-wide modellings (Vallner L 2003, 2002, 1966b). The lower or median groundwater levels from monitoring series or episodic measuring performed in 1991–2008 were used for calibration of steady-state models. Unfortunately, no systematic monitoring of groundwater heads of the Ordovician-Cambrian aquifer system was carried out in Kiviõli. Therefore, the heads of this aquifer system measured at boring of production wells in 1965—1983 were used for model calibration.

<u>The groundwater recharge</u> was given as the net infiltration (total groundwater recharge minus evaporation from the zone of saturation or capillary fringe) onto the ground surface. There was no instrumental data (lysimeters or multi-level well tests, etc.) for its estimation in study areas. It is supposed that net infiltration into the landfills varies mostly from 40 to 100 mm/year. Furthermore, a significant amount of water is used for the ash transport. In a landfill, water moves downward and leaches semi-coke and ash stored. This flux penetrates the thin layer of natural Quaternary deposits beneath landfills and intrudes into bedrock.

For that reason, the net infiltration into a landfill was put into models as an assumed value keeping the maximum altitude of the groundwater table by 10–20 m higher in the central part of Kohtla-Järve and Kivõli landfills than on their margins. This difference reached 30 m in the landfill of the Balti PP at Narva. The rate of the net infiltration assumed was corrected by model calibration. The net infiltration outside of landfills was estimated based on former investigations carried out on the whole territory of Estonia (Vallner L 2003, 1997a, 1996b, 1980).

The average annual air temperature is 4.2°C in study areas. The coldest month is January with an average temperature of –6.5°C and the hottest – July with a temperature of 16.8°C. Average annual precipitation is about 730 mm/year. The average total surface runoff reaches 260 mm/year (Resursy poverhnostnyh…1972).

<u>The riverbed conductance</u> for assigning the river boundary conditions was estimated accordingly to the experience of former investigations and modellings of the groundwater runoff (Vallner L 2003, 2002, 1997a, 1996a, 1996b, 1980, 1976).

The parameters of the groundwater transport are needed for predicting the contaminant development based on the equation (3.2.1). No field or laboratory tests have been performed to determine the rates of the hydrodynamic dispersion coefficient tensor, and components of the chemical reaction term of this equation in Northeast Estonia up do date.

Thereat, a special attention serves the half-life time $t_{1/2}$ = 7.0 days of phenols used to calculate the maximum extent of the phenol plume permeating the carbonate bedrock beneath and around of landfills in Kohtla-Järve and Kiviõli by K. Rudolph-Lund (Sørlie J-E. *et al.* 2004). The half-life time of phenol and chlorophenols degradation is 7 days at an abundant sunlight in warm estuarine water (Hwang H, Hodson R 1986). However, this time could be significantly longer at a lower temperature in the underground. Anyhow, the half-life time of 2, 4, 6-Trichlorophenol is 917 days (Howard P *et al*. 1991). On the other hand, a wide spectrum of PAHs and BTEX originated from oil shale processing wastes occurs next to phenols in area of landfills (Sørlie J-E *et al.* 2004). The half-life times of some organic pollutants detected over there are as follows: Benzo(a)anthracene – 782 days, Benzo(k)fluoranthen – up to 4,280 days, very toxic Benzo(a)pyrene – up to 1,060 days, Chrysene – up to 2,000 days, and Indeno(123-cd)pyrene – 1,330 days (Howard P *et al*. 1991). Thus, proceeding from half-life times, some organic compounds are likely able to contaminate the groundwater during a longer time and to move deeper than it was supposed about phenols in the former report by K. Rudolf-Lund (Amiri F. 2005; Siedlecka E, Stepnowski P 2005; Sørlie J-E *et al.* 2004; Vessely M *et al.* 1997).

Modelling codes. Hydrogeological processes are very complicated in study areas because of great differences in the surface topography, filtrational heterogeneity of layers and spatial dissimilarity of pollution sources. The Visual MODFLOW v. 4.5 (2010.1) software including the MT3DMS code was used to create the coupled flow and transport models of landfill areas (Harbaugh AW 2005; Bear J, Cheng A-D 2010; Konikow, 2011; Visual MODFLOW…2010; Zheng C 1999; Zheng C, Bennet GD 2002; Zheng C, Wang PP 2003, 1998). This program package has been chosen since it is widely acknowledged around the world.

3.5. Groundwater models of landfills

3.5.1. Kohtla-Järve model

The model area is situated on the Viru plateau in northeast Estonia where semi-coke generated with the Kohtla-Järve oil factory and ash from an accompanying power plant stored during 73 last year's form a prolonged landfill (Fig. 3.5.1.1, 4.1.1). The Pulkovo 1942 co-ordinates of this area (m) are for the lower left corner 5510250, and 6584000; for the upper right corner 5515500, and 6589000. The relative height of the landfill was up to 120 m in 2010. The rectangular study area around the landfill is 5,250 m from west to east and 5,000 m from north to south covering 26.25 km^2. The flat Ahtme height with the maximum absolute elevation of 72 m serving as a local watershed on the Viru plateau borders the study site from the east. Absolute elevations of the ground surface decrease from 55–66 m on the eastern border of the site to 46–47 m on its northern and western borders.

Oil shale ash and semi-coke have been piled in the central part of the study area where the absolute top elevation of the landfill reaches 172 m. The total bulk of the landfill is about 6.6 × 10^7 m^3 covering the area of 2.14 km^2, the quantum of wastes stored should reach 8.3 × 10^7 t at present (Sørlie J-E *et al.* 2004). The landfill is drained with a surrounding network of ditches. The water collected is directed into Kohtla-Järve Regional Sewage Treatment Plant. In the high flow periods, a portion of the run-off from the landfill does not find room in the ditches surrounding immediately the landfill. This excessive contaminated water intrudes into farther ditches, which are branches of Purtse River situating westward from the area. Woodlands and farmlands occur mostly westward from the landfill. A shale oil factory and power plant are situated alongside the eastern flank of the landfill. Suburbs of the Kohtla-Järve town lie in the northeast part of the study area. The Käva 2 underground oil shale mine exhausted adjoins the study area from the southeast. This mine was closed and flooded in 1973.

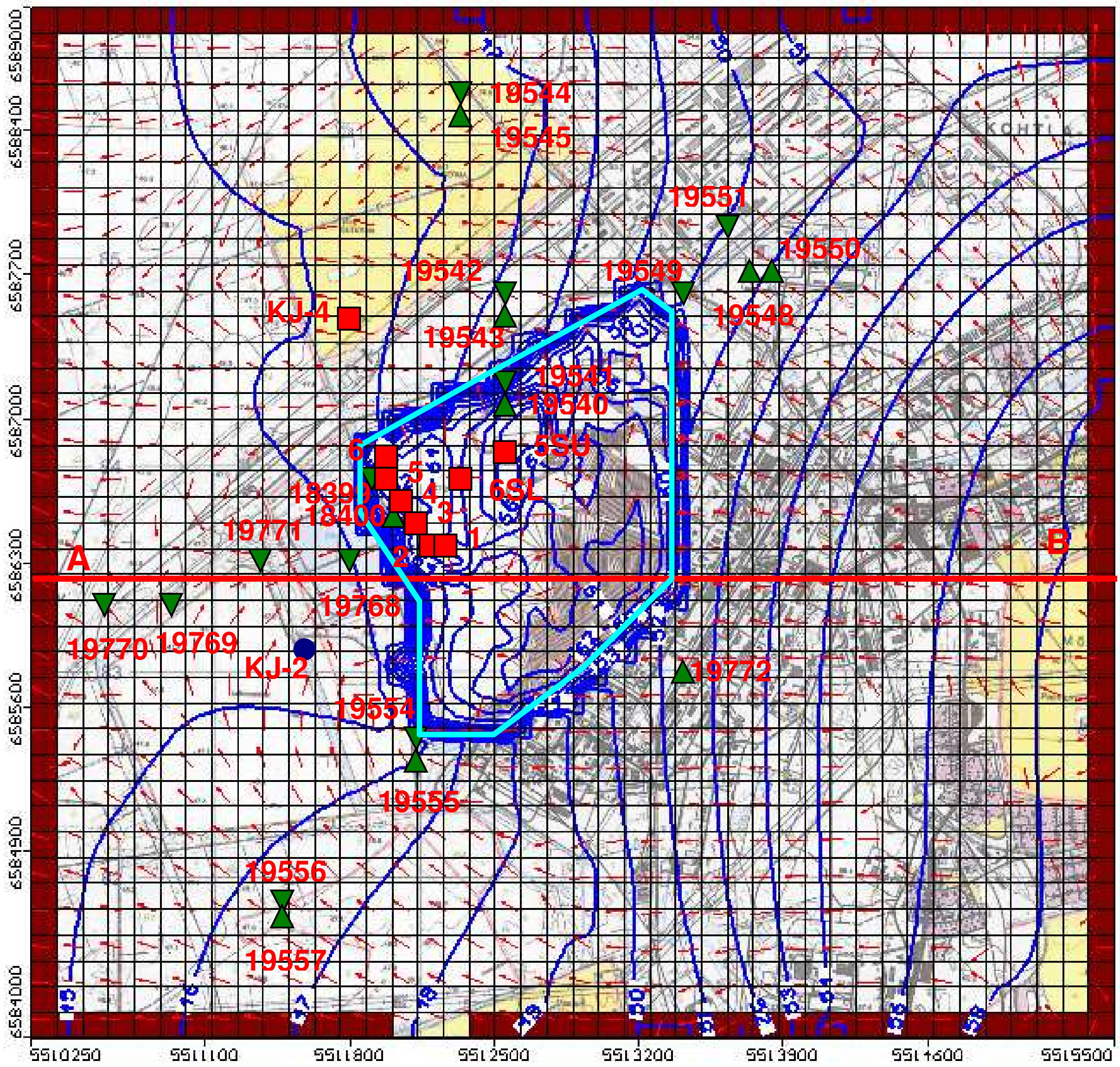

Fig. 3.5.1.1. Area of the Kohtla-Järve model. Model Pulkovo co-ordinates are in metres. Water table: blue contours – m a.s.l.; red arrows – the direction of the groundwater movement in the 1st model layer. Boundary conditions: dark red lines – constant head on the borders of the

model area; Observation wells and their numbers: red quadrates – model 1st layer, dark blue circle – 2nd layer, green triangle down – 3rd layer, green triangle up – 5th layer. Light blue contour closed – an area of the landfill. A–B – line of the hydrogeological cross-section. The spacing of the orthogonal virtual computational grid is 125 m × 125 m.

Model layers and boundary conditions. A coupled groundwater flow and transport model of the Kohtla-Järve study area was built. For that purpose, the study area was covered by a virtual orthogonal computational grid with the spacing of 125 m × 125 m forming 40 rows and 42 columns, and 5 model layers were generated to represent the cross section of hydrogeological units described above in paragraph 3.3 (Fig. 3.5.1.1, 3.5.1.2).

The 1st model layer enfolds the ash and semi-coke of the landfill and the upper seam of natural Quaternary deposits around the landfill. The thickness of the 1st model layer reaches up to 125 m beneath the top of the landfill. The lateral hydraulic conductivity given to the portion of the 1st layer represented by semi-coke and ash varies from 10^{-3} m/day to 10^{-4} m/day, the transversal conductivity is $9*10^{-5}$–10^{-4} m/day. Outside the landfill borders, the average lateral hydraulic conductivity of natural Quaternary deposits is 5 m/day and their transversal conductivity does not exceed 0.5 m/day. The 1st layer is unconfined.

The net infiltration given on the surface of the 1st layer of the flow model is 50–60 mm/year. A Cauchy boundary condition specifying the hydraulic relationship between groundwater head modelled and a surface water body (Drain Boundary Condition of the Visual MODFLOW) is assigned to the channel network draining the study area. At that, the drain conductance ranging mostly 100–200 m^2/day was estimated accordingly to the experience of former investigations and modellings of the groundwater runoff (Vallner L 2003, 2002, 1997a, 1996a, 1996b, 1994, 1980, 1976).

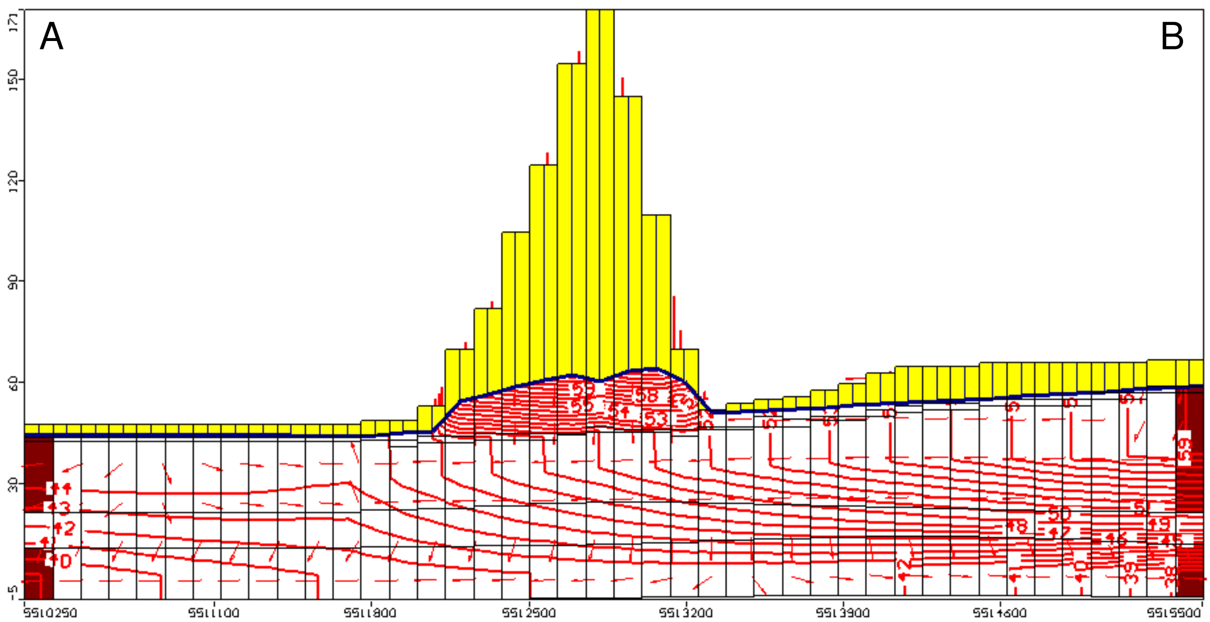

Fig. 3.5.1.2. Hydrogeological cross-section A–B of the Kohtla-Järve model. Vertical axis – absolute heights, m. Red isolines – absolute elevation of the groundwater head, m. Red arrows – the direction of the groundwater movement. Yellow – soils above groundwater table (vadose zone). Other graphical symbols – see the Fig. 3.5.1.1.

The 2nd model layer is a bed representing the natural Quaternary deposits beneath the landfill, and the lower seam of Quaternary deposits around the landfill. The thickness of the 2nd model layer ranges mostly from 2 m to 4 m. It was assumed that the portion of this layer directly beneath the landfill forms a relatively great hydraulic resistance to the downward flux coming from above. Such presumption is logical insofar as the originally unconsolidated deposits of this zone must be impregnated by oil products of leachate and they have strongly compressed by the weight of overlying landfill. Therefore, the lateral conductivity of this zone was equalled to 0.1 m/day and the transversal conductivity to 0.01 m/day. Outside the landfill, the lateral conductivity of the 2nd layer is 5.0–8.0 m/day, and the transversal conductivity is predominantly 0.5 m/day. The layer is considered as unconfined-confined.

The 3rd layer consists of diverse fissured and karsted limestones with clayey interbeds from the Uhaku Stage to the Kunda Stage, belonging to the S-O aquifer system. Its total thickness is 20–25 m. The lateral conductivity ranges mostly from 10 m/day to 15 m/day, the transversal conductivity varies between 1.0–1.5 m/day. The hydraulic type of the layer is unconfined-confined.

The S-O aquifer system is recharged from overlying Quaternary cover including the downward flux from the landfill and with lateral flow coming from Ahtme height. Groundwater moves mainly westward in the carbonate bedrock discharging partly to the channel network. Before heaping of the landfill, the intensity of this discharge was 90–260 $m^3/(day*km^2)$ (Vallner L 1996b). The annual amplitude of fluctuation of groundwater table or head is 1–3 m.

The 4th layer enfolds the S-O regional aquitard with a total thickness of 4–5 m. Groundwater moves predominantly in the vertical direction in this layer. Therefore, the boundary conditions are not specified along outer borders of the layer. The transversal conductivity corrected by model calibration is mostly 10^{-4} m/day.

The 5th layer represents the confined O-Є aquifer system which total thickness is 15–18 m. Its lateral mean hydraulic conductivity is about 3 m/day. The O-Є aquifer system is recharged with downward flux coming from S-O aquifer system and penetrating the Lower Ordovician aquitard. Therefore, the potentiometric surface of the O-Є aquifer system is convex. It decreases from an absolute elevation of 42–43 m in the central part of the study area to elevations of 37–40 m on its borders. The O-Є aquifer system is not used for a public water supply in the study area at present.

The Constant Head boundary conditions have been assigned to outer borders of the 1st, 2nd, 3rd, and 5th model layer. Under the O-Є aquifer system lies the Lükati-Lontova regional aquitard ($Є_1$Lk-$Є_1$Ln) consisting of siltstones and clays with the total thickness of 70 m which transversal conductivity is 10^{-9}–10^{-6} m/day. The downward flux penetrating this aquitard is irrelevant from the viewpoint of the present research.

Therefore, the bottom surface of the O-Є aquifer system is considered as a lower impermeable boundary of the Kohtla-Järve model.

Table 3.5.1.1. Sum of phenols in groundwater coupled observation borings in the Kohtla-Järve area, mg/L (Bityukova L *et al*. 2011; Kirde-Eesti...2008, Tööstusjäätmete ja...2007)

Sampling date	Number of the observation boring and the hydrogeological unit tapped								
	18399	18400	19540	19544	19545	19554	19555	19556	19557
	O	O-Є	O	O	O- Є	O	O- Є	O	O- Є
21.05.1996			2,231			0.094	0.038	<0.0001	<0.0001
3.07.1996			11,040	<0.0001	0.007	<0.0001	0.030	<0.0001	0.020
22.10.1996			9.110	<0.0001	<0.0001	0.095	0.101	<0.0001	<0.0001
24.11.1996	18,820	0.008	5.410	<0.0001	<0.0001			<0.0001	<0.0001
22.05.1997	62,800	0.040	4.060	0.110				<0.0001	<0.0001
5.08.1997	6,830	0.004	9,100	<0.0001				<0.0001	<0.0001
1.10.1997	17,180	0.017	5.520	<0.0001				<0.0001	<0.0001
7.12.1997	27,330	0.051	4,400						
21.04.1998	31,290	0.055		<0.0001	<0.0001			<0.0001	0.005
27.05.1998	31,230	0.108		<0.0001				<0.0001	<0.0001
28.07.1998	27,900	0.017		<0.0001				<0.0001	<0.0001
29.09.1998	18,290	0.011		<0.0001				<0.0001	<0.0001
4.06.1999	27,400	0.033		0.085	0.230			0.050	0.030
28.07.1999	27,720	0.141		0.063	<0.0001			<0.0001	0.015
22.09.1999	44,430	0.097		0.020	<0.0001			<0.0001	<0.0001
28.10.1999		0.054		0.005					
7.04.2000	28,900	0.130		0.006	0.012				
29.06.2000	25,950	0.111		<0.0001	<0.0001			<0.0001	<0.0001
22.08.2000	12,910	0.769		0.006	0.004			<0.0001	0.002
4.10.2000	19,270	0.005		0.024	<0.0001			<0.0001	<0.0001
15.05.2002	6,310	0.100		<0.0001	<0.0001			<0.0001	<0.0001
2.07.2002	16,625	0.003		<0.0001	<0.0001			<0.0001	<0.0001
29.08.2002	13,000	0.005		<0.0001	<0.0001			<0.0001	<0.0001
8.10.2002	16,560	0.003		<0.0001	<0.0001				<0.0001
14.05.2003	5,916	0.020		<0.0001	<0.0001			0.016	<0.0001
29.07.2003	12,441	0.032		0.007	<0.0001				
7.10.2003	6,603	0.014		<0.0001	<0.0001				
6.07.2004	6,639			0.003	0.003			0.020	
2.09.2004	7,127			0.048	13,150			2,631	0.018
12.10.2004	11,500			<0.0001	0.017			0.005	0.058
12.07.2005	15,514			0.025	<0.001			0.034	0
31.08.2005	10,160			0.006	0.002			0.033	0.029
19.10.2005	17,890			<0.0001	0.019			<0.0001	<0.0001
21.11.2005								<0.0001	<0.0001
5.06.2006	10,788								
13.06.2006				<0.0001	<0.0001			<0.0001	<0.0001
30.08.2006	17,230			0.004	<0.0001				0.020
17.10.2006	49,100			<0.0001				<0.0001	
13.10.2008	17,267			0.029	<0.0001			0.054	0.040
3.05.2010	11,217								
12.07.2010	14,069								
15.09.2010	4,000								

Table 3.5.1.2. Sum of phenols in groundwater in the Kohtla-Järve area, mg/L (Bityukova L *et al.* 2011; Kirde-Eesti...2008, Tööstusjäätmete ja...2007)

Sampling date	Number of the observation boring and the hydrogeological unit tapped									
	19542	19543	19548	19549	19550	19551	19768	19769	19771	19772
	O	O- Є	O	O- Є	O	O- Є	O	O	O	O- Є
21.05.1996	3,400						9,340	<0.0001		
3.07.1996	19,030	<0.0001								
24.11.1996	0.195	<0.0001								
22.05.1997	6,220	0.080							3,600	0.020
5.08.1997	1,960	<0.0001					15,980	<0.0001	4,070	<0.0001
1.10.1997	3,620	<0.0001					25,780	<0.0001	1,280	<0.0001
7.12.1997							56,570	<0.0001	4,260	
21.04.1998	0.063	<0.0001							4,880	<0.0001
27.05.1998	1,955	<0.0001							12,630	0.075
28.07.1998	2,030	<0.0001							9,430	<0.0001
29.09.1998	2,250	<0.0001							5,270	<0.0001
4.06.1999	4,536	0.003					38,280	0.055	2,139	0.008
28.07.1999	2,780	<0.0001					26.970	0.011	1,219	0.007
22.09.1999	17,430	0.0030					41,330	0.023	14,540	<0.0001
28.10.1999		0.077					16,919	0.024		
7.04.2000	0.228	0.009							4,180	<0.0001
29.06.2000	7,620	<0.0001							2,950	<0.0001
22.08.2000	2,757	0.003							0.769	<0.0001
4.10.2000		0.0011							1,340	0.011
3.05.2001	2,757									
15.05.2002	0.945	<0.0001							0.948	<0.0001
2.07.2002	5,200	<0.0001							1,112	<0.0001
29.08.2002	6,094	<0.0001							1,885	<0.0001
8.10.2002		<0.0001			<0.0001				2,516	<0.0001
14.05.2003	10,702	0.004							1,888	
29.07.2003	0.036	<0.0001							3,816	<0.0001
7.10.2003	1,983	<0.0001							1,581	0.007
6.07.2004	0.113	0.004							1,619	0.013
2.09.2004	0.008	0.006							7,640	0.028
12.10.2004	0.615	0.609							4,810	0.006
12.07.2005	0.637	0.018	0.034	0.003	<0.0001	<0.0001			7,628	0.014
31.08.2005	0.088	0.021	0.058		0.031	0			3,426	0.164
19.10.2005	0.175	<0.0001	0.012	0.002	<0.0001	0			3,049	<0.0001
5.06.2006			0.017		0.004				24,642	0.002
13.06.2006	0.008	<0.0001	0.005	0.003	0.005	0.003				
30.08.2006	0.161	<0.0001	0.003		<0.0001				9,550	0.003
17.10.2006	0.449	<0.0001	<0.0001		<0.0001				7,230	
27.11.2006	0.020		<0.0001		<0.0001					
25.07.2007	0.013	0.027	0.013	<0.0001	0.007	<0.0001				
4.10.2007	0.019	0.006	0.006	<0.0001	<0.0001	0.039				
13.11.2007	0.010		0.007							
21.08.2008	0.001	0.010	0.015	0.005						
13.10.2008	0.044	0.027	0.010							0.036
3.05.2010	0.005	0	<0.0001	0						0
8.07.2010	0	0	0	0						0
9.09.2010	0	0.031	0	0						0

<u>Modelling of the groundwater flow.</u> The hydraulic properties of the model layers and model boundary conditions were determined based on former numerous field tests and their generalizations (Perens R, Vallner L 1997; Razgonjajev A 1997; Razgonjajev A, Timaškin R, Razgonjajeva L1993; Riet K 1976; Vallner L 2002, 1997a, 1996b; etc.). The data incorporated into the flow model were proved an adjusted by calibration calculations. They were carried out using the trial-and error correction of hydraulic conductivity and net infiltration values for achieving the optimum match between simulated parameters and calibration targets. Simulated elevations of groundwater table and heads were checked against data of the observation wells until the correlation coefficient between computed, measured data reached 0.999, and the standard error of the estimate was 0.105 m (Fig. 3.5.1.3). All calibration points fell into the interval where 95% of them were statistically expected to occur (Visual MODFLOW…2010).

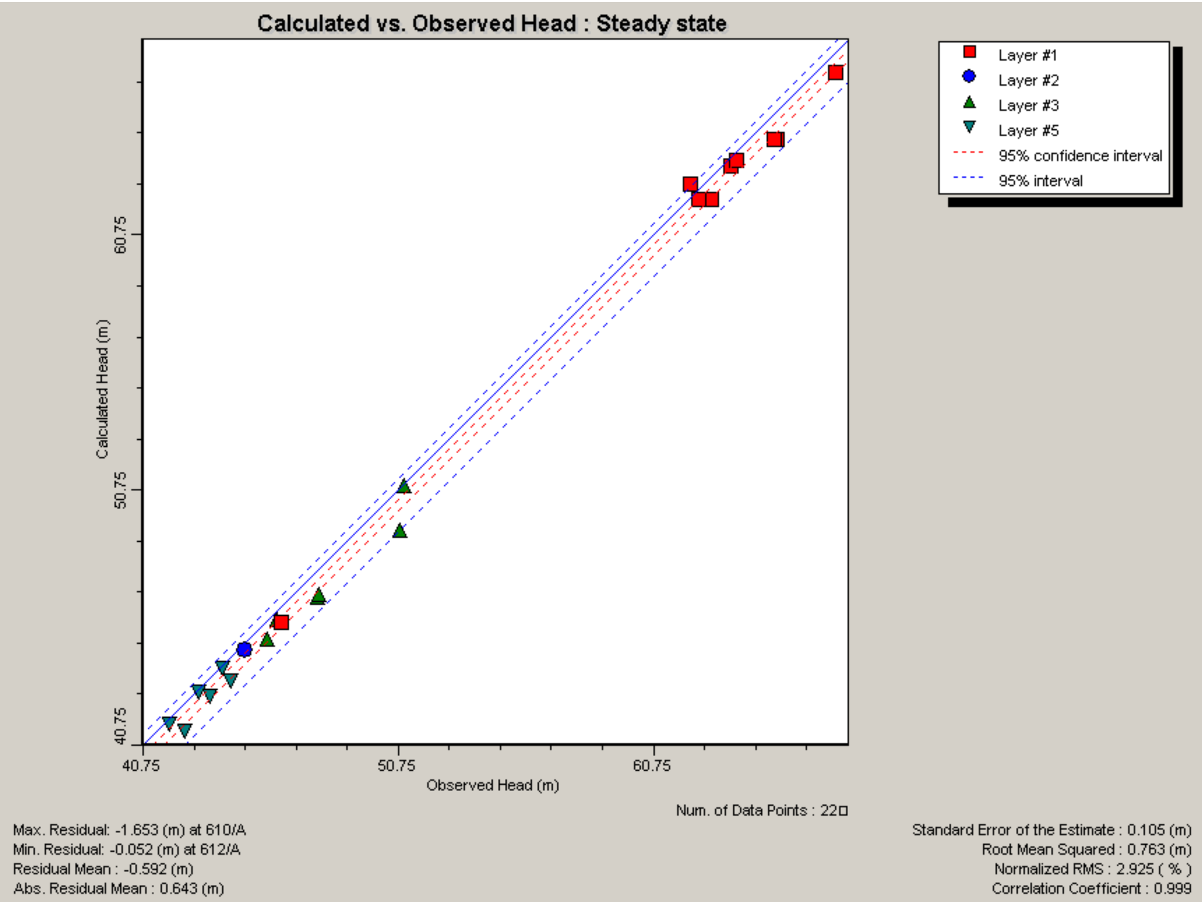

Fig. 3.5.1.3. Calibration graph of the Kohtla-Järve flow model.

The profoundly calibrated model gave a scientifically founded synopsis of contemporary groundwater flow characteristics in the study area. At that, the mass balance rates and 3D particle tracking pathlines were determined with a special calculation modules Zone Budget and MODPATH included into Visual MODFLOW Premium code (Harbaugh 1990; Pollock 1998).

The performed simulations show that the total net infiltration from precipitation is about 3,600 m^3/day or 50 mm/year or 1.6 L/(s × km^2) in the study area. The intensity of the direct net infiltration into the landfill is the same; its amount is 370 m^3/day. An unconfined aquifer has been formed in deposits of the landfill (Fig. 3.5.1.1, 3.5.1.2). The thickness of the unsaturated (vadose) zone of this aquifer varies from 1–2 m to 80–100 m depending on the topography of the landfill. In vadose zone, a preferential flow takes evidently place (Bowman R, Bouwer H, Rice R 1987; Bouwer H 1991; Molz F *et al.* 2006; Pontedeiro E *et al.* 2010). It is caused by the conglomeratic and slaty structure of landfill deposits coupled with an extremely great porosity of this formation. The water flows downward along the fingering of the vadose zone. (Scotter D 1978; Zheng C, Gorelick S 2003). The lower portion of the landfill has saturated because of comparatively great hydraulic resistance of deposits. The absolute elevation of the water table is mostly 62–66 m in the north-western part of the landfill and 68–70 m or even more beneath higher parts of the landfill. The thickness of the saturated zone of landfill deposits varies mostly between 10–15 m.

The downward flux into the zone beneath the landfill (ZBL) of the 2nd model layer is 360 m^3/day. Practically all of this water intrudes into ZBL of the 3rd layer. The total lateral flux from the landfill and from the ZBL of the 2nd layer into surrounding parts of the 1st and 2nd layer does not exceed 10 m^3/day. In the ZBL of the 3rd model layer, the downward flux of contaminated water mixes with the lateral uncontaminated flux coming from the east along the 3rd layer and amounting 1,700 m^3/day (Fig. 3.5.1.4). About 1,900 m^3/day from the blend formed flows westward out from the ZBL of the 3rd model layer and approximately 120 m^3/day intrudes downward through the S-O regional aquitard into the 5th layer. The discharge of groundwater in ditches directly surrounding the landfill is 300–400 m^3/day. Thus, the most of contaminated water forming in the landfill passes these ditches from underneath and intrudes into the surrounding parts of the 2nd and 3rd layer. The total groundwater run-off forming in 1st, 2nd, and 3rd layers and discharging in channel network of the study area is 3,700 m^3/day.

The 3rd model layer (the S-O aquifer system) is recharged by the lateral inflow reaching 7,400 m^3/day. This flux comes through the eastern border of the study area, partly from the oil shale mine abandoned. The downward flux from the overlying 2nd layer is in total 3,400 m^3/day. From this flux, about 350 m^3/day goes into the ZBL of the 3rd layer. Heads decrease from 52–59 m on the eastern border of the study area to 44–45 m on the western border due to the drainage impact of the channel network of the Purtse River (Fig. 3.5.1.4). Therefore, the water stored in the eastern portion of the 3rd layer moves laterally mainly westward partially penetrating the ZBL. The velocity of these lateral fluxes is up to 40–60 m/year.

Due to the drainage effect of the channel network, a portion of water rises up from the 3rd layer into the 2nd layer in western half of the study area. The amount of this flow is 5,000 m^3/day. The lateral outflow through the western border averages 1,800 m^3/day. The total downward flow from the 3rd layer into the 4th layer (the Silurian-Ordovician regional aquitard) reaches 4,000 m^3/day.

The lateral flow is insignificant in the 4th model layer because it's small hydraulic conductivity. However, due to the head difference between the 3rd and 5th layer ranging from 7 m to 21 m, an essential downward flux penetrates the 4th layer.

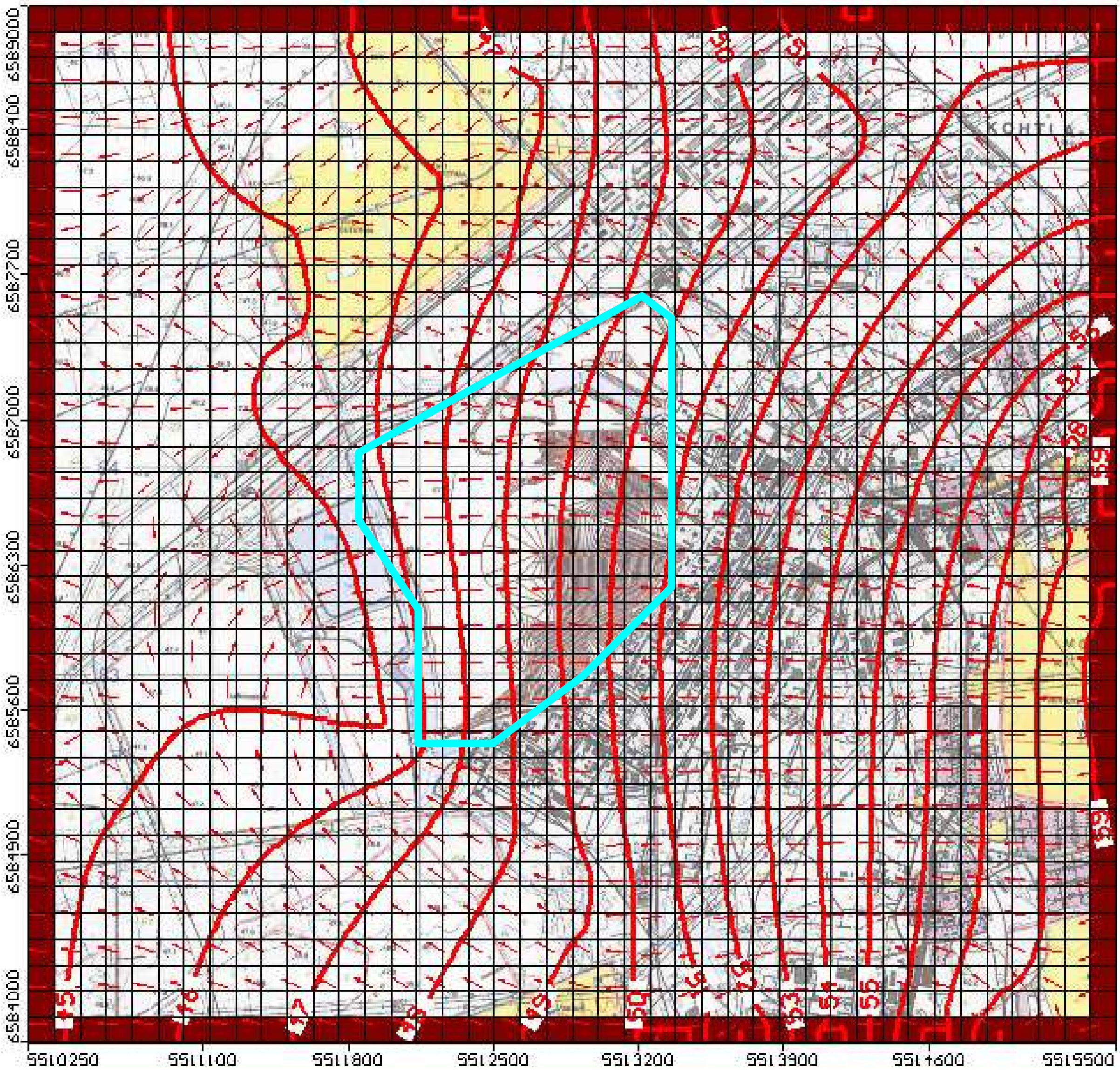

Fig. 3.5.1.4. Groundwater heads in the 3rd model layer (S-O aquifer system) of the Kohtla-Järve model – red isolines (absolute elevation, m). Other graphical symbols – see the Fig. 3.5.1.1, 3.5.1.2.

The 5th model layer (the Ordovician-Cambrian aquifer system) is recharged mainly by the downward flow reaching in total 4.000 m^3/day. It comes from the overlying regional aquitard. The downward flux into the ZBL of the 5th layer amounts 120 m^3/day. A zone of the increased hydraulic head reaching 42–44 m prolonged in SW-NE direction exists in the central portion of the 5th layer (Fig. 3.5.1.5). It is formed due to the vertical transmitting of the head from the top of the groundwater table and because of sporadic relatively major hydraulic conductivity in the 4th layer. Groundwater flows radially from the central zone of the increased head towards the outer borders of the 5th layer where the head is mostly 38 m. The lateral outflow through the borders is 4.000 m^3/day in total. The velocity of the groundwater flow is 2–4 m/year in the Ordovician-Cambrian aquifer system.

It is possible, that a significant amount of water infiltrated into the semi-coke was or will be consummated by forming the ettringite that comes into being because of hydration of semi-coke. This water should be disjoint from general groundwater circulation, but special experimental investigations are needful to ground this procedure.

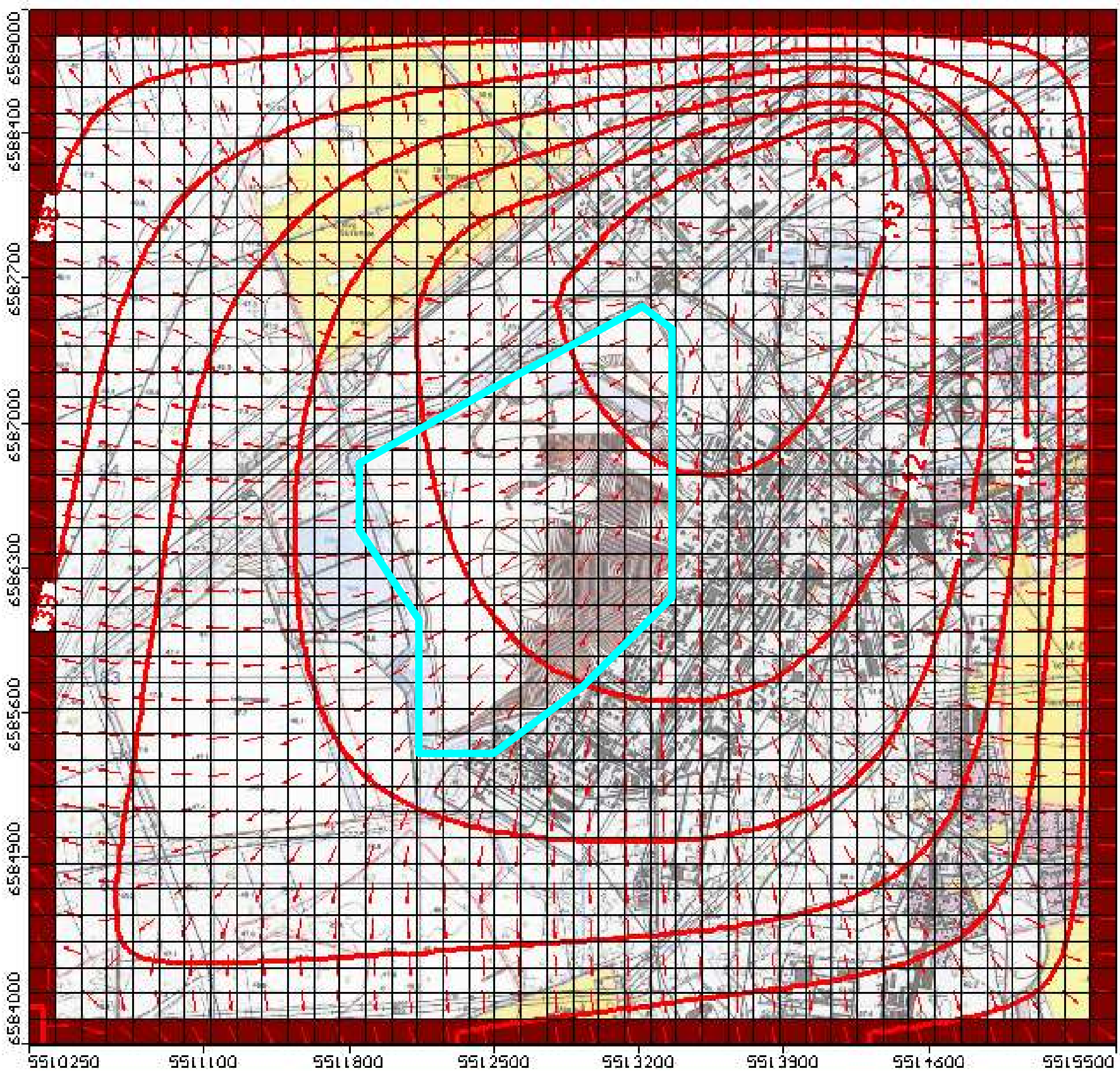

Fig. 3.5.1.5. Groundwater heads in the 5th model layer (O-Є aquifer system) of the Kohtla-Järve model – red isolines (absolute elevation, m). Other graphical symbols – see the Fig. 3.5.1.1, 3.5.1.2.

<u>The model of the conservative transport</u> was based on the flow model. Spatial distribution of the Total of Dissolved Solids (TDS) was studied with transport modelling assuming that the TDS could be considered as a conservative tracer no participating in chemical reactions. The concentration of other non-reactive groundwater ingredients might presumably be change proportionally to the TDS at the groundwater transport. The TDS value is not rationed with Estonian standards of drinking water (Joogivee kvaliteedi... 2001; Ohtlike ainete... 2010), but on the background of the initial TDS equalled to 500 mg/L, the TDS values exceeding this threshold must be considered as evident signs of the groundwater contamination.

Estimation of parameters for the transport model was very complicated. No special tests were carried out for experimental determination of porosity and hydrodynamic dispersion of the groundwater environment in the study area. The sole data source for construction of an adequate transport model was the water quality monitoring performed in 1996–2008 (Kirde-Eesti... 2008; Tööstusjäätmete ja... 2007). Therefore, it was decided to reproduce the observed contamination plum as a calibration target by modifying of input parameters at model runs (Schwede RL, Cirpka OA 2010). In such of way, *the model calibration was turned from an adjusting and checking operation to a principal method for establishing of model parameters.*

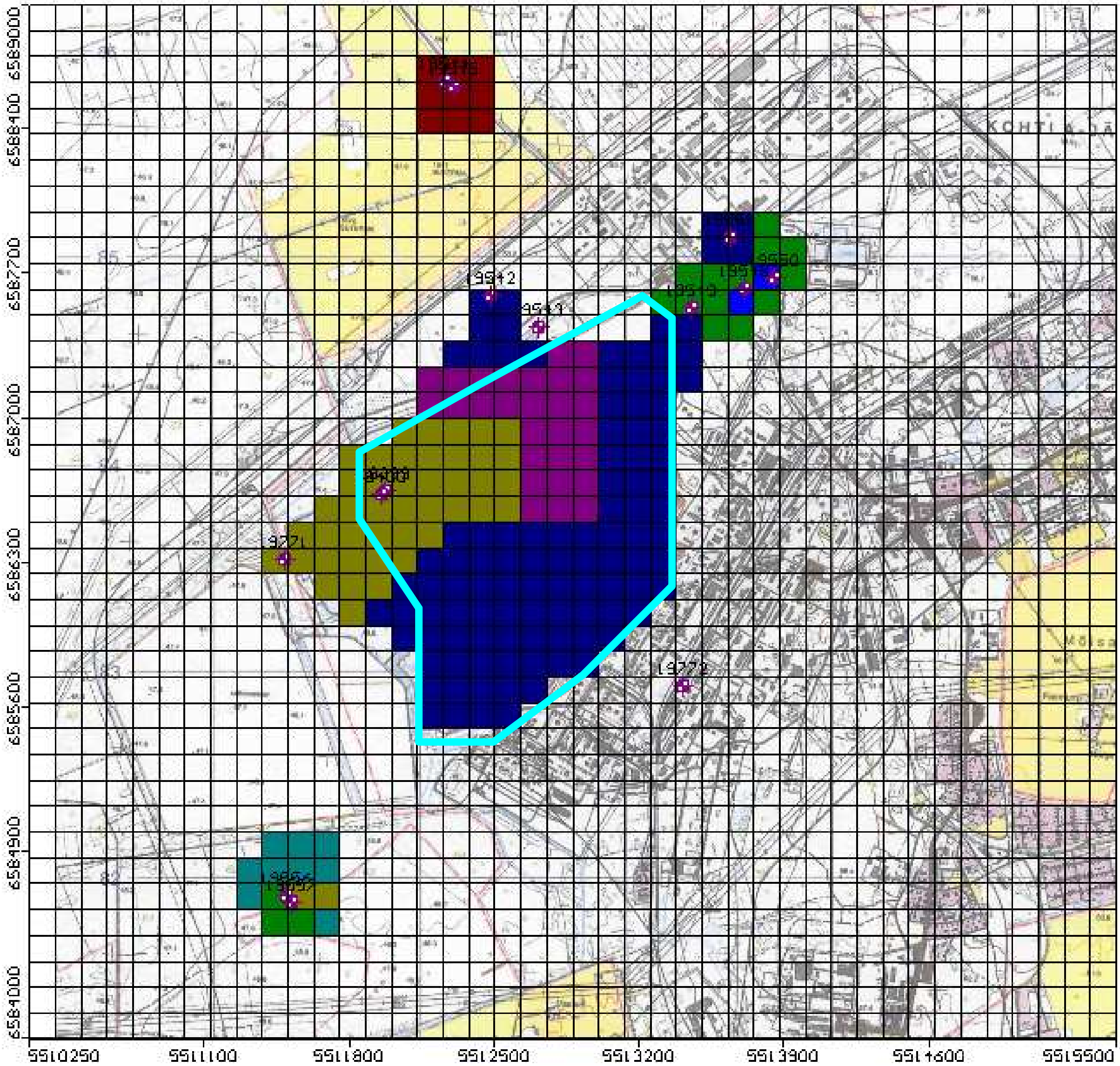

Fig. 3.5.1.6. Constant Concentration boundary condition given to the 1st layer (Quaternary deposits) of the Kohtla-Järve model. TDS value, mg/L: maroon – 700; greenish blue – 1,000; green – 1,300; light blue – 2,000; dark blue – 3,000; violet – 6,000; olive – 12,000. Other graphical symbols – see the Fig. 3.5.1.1, and 3.5.1.2.

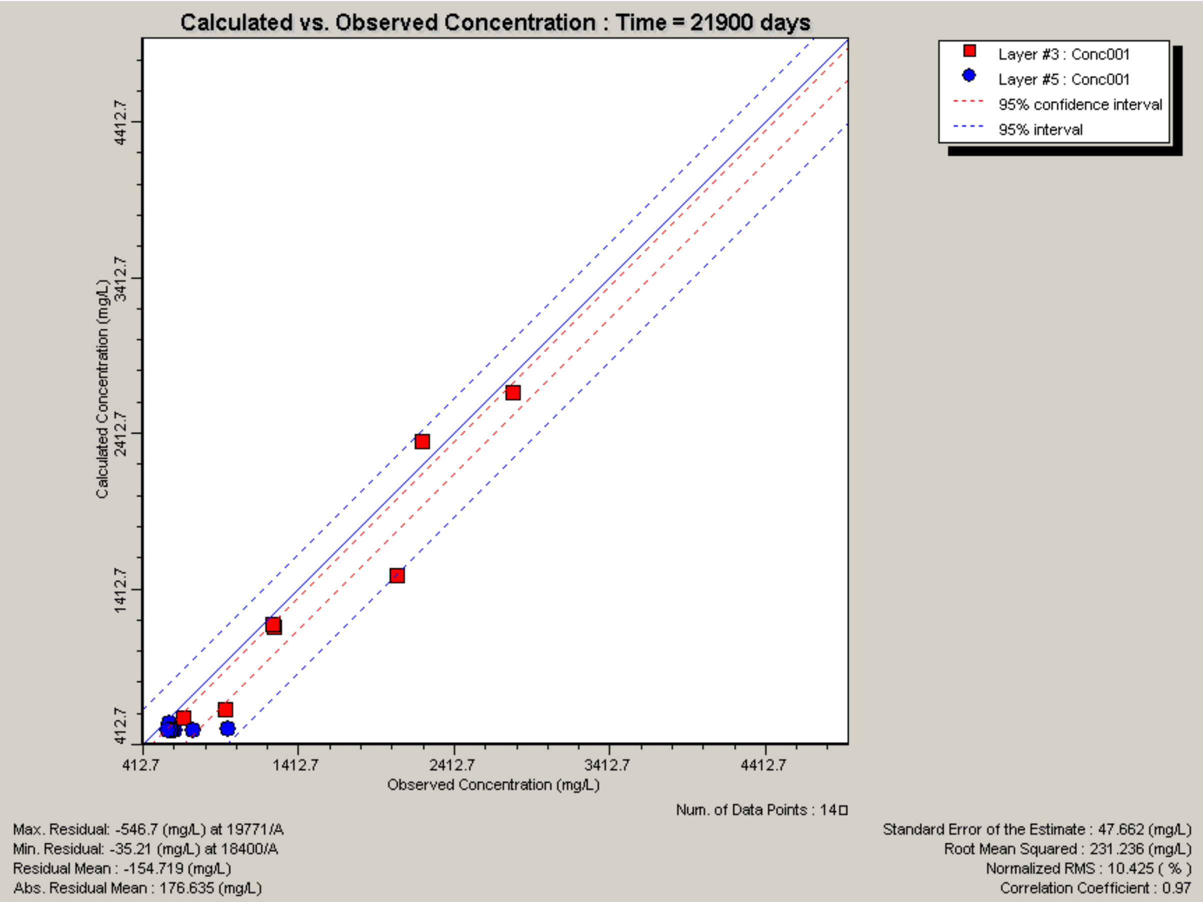

Fig. 3.5.1.7. Calibration graph of the Kohtla-Järve TDS transport model.

Following this conception, the initial TDS was set equal to 500 mg/L for all model layers. The value of longitudinal dispersivity was varied from 20 m to 200 m, the ratios of horizontal and vertical dispersivity to the longitudinal one were mostly 0.1, and the coefficient of diffusion given was 0.1 during testing runs of the model. Effective porosity accepted was 0.15–0.45 for the 1st and 2nd layer (Metsur M 2005), 0.03 for the 3rd and 4th layer, and 0.3 for the 5th layer. Such values of transport parameters answered generally to determinations of other researchers (Bredehoeft J *et al*. 1973; Egboka B *et al*. 1983). The MT3DMS engine (Zheng C, Wang P 2003) was used for transport calculations.

It was found out that the output of the transport model was most sensible to fixed TDS values given as Visual MODFLOW Constant Concentration boundary conditions to the 1st layer (Fig. 3.5.1.6). Actually, there was no experimental information about the distribution of TDS in the bottom of the landfill. It was established manually trying many variants until an optimum match between observed and calculated TDS was achieved (Fig. 3.5.1.7). Accordingly, to observation wells 19544, 19545, 19548, 19549, 19550, 19551, 19556, and 19557, the contamination occurs also as detached spots northward, north-eastward, and south-westward from the landfill. The TDS reaches there up to 1,500 mg/L in the 3rd model layer. Thus, the local surficial contamination sources not connected with the landfill must exist in these spots. It was also accounted for the setting of the Constant Concentration boundary condition.

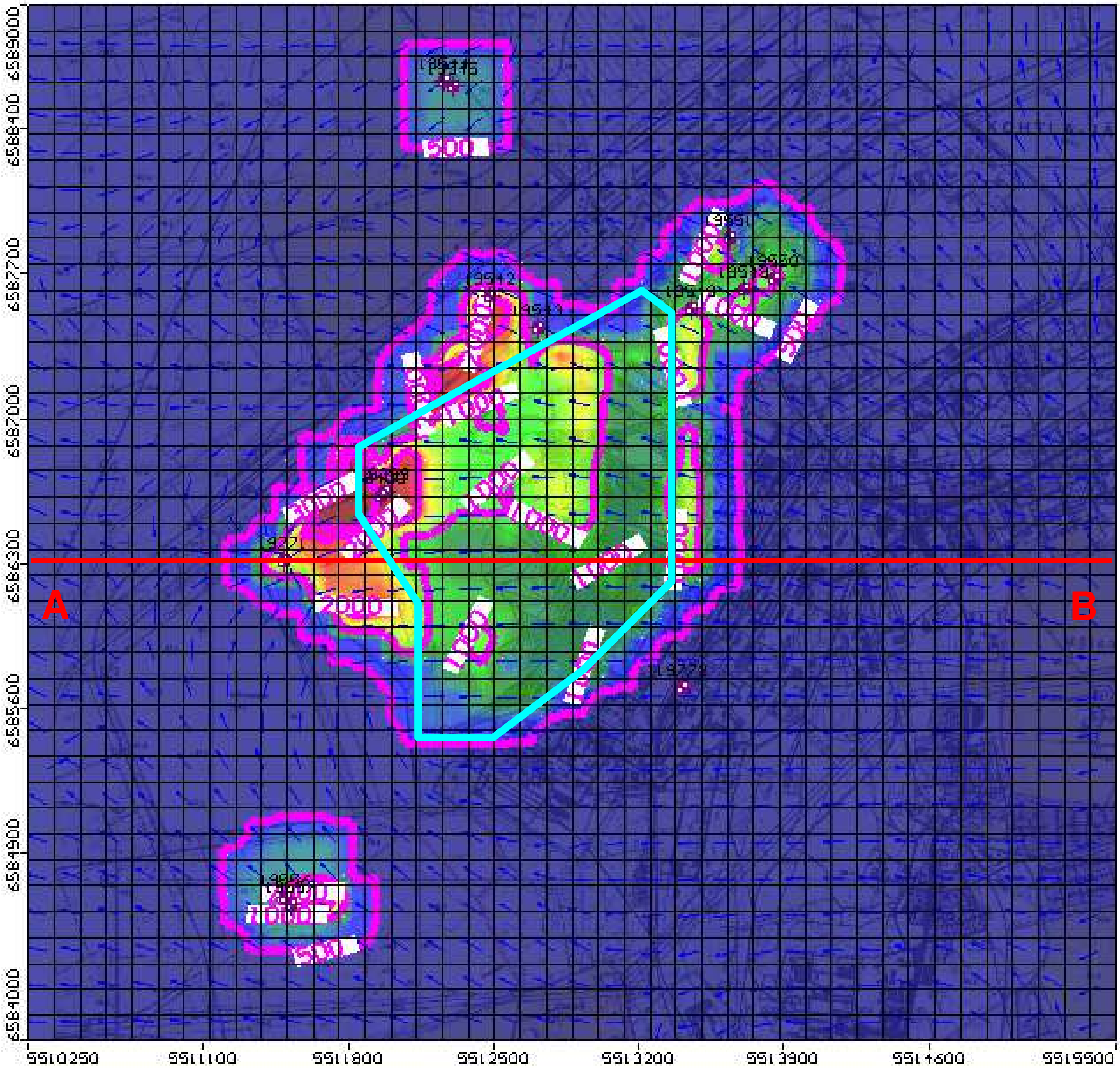

Fig. 3.5.1.8. TDS in the 3rd layer (S-O aquifer system) of the Kohtla-Järve model in 2010. TDS content, mg/L: dark blue – up to 500, light blue – 500–600, glaucous – 600—800, dark green – 800–1000, light green – 1000–1,500, yellow – 1,500–2.000, light brown – 2.000–3,000, brown – >2,000. Violet isolines – the value of TDS, mg/L. Small violet circles indicate concentration observation wells and their numbers. Blue arrows indicate directions of the groundwater movement. Other graphical symbols – see the Fig. 3.5.1.1, and 3.5.1.2.

The year 1938 was considered as an initial date of spreading of high TDS when a significant amount of retorting wastes was accumulated in borders of the existing landfill for the first time. The output time of transport calculations carried out by the MT3DMS engine was set in 2010. This conception should be acceptable since the main sources of groundwater contamination are liquids formed at leaching of semi-coke and oil shale ash by rain and thaw waters infiltrating into the landfill. The water used for washing of semi-coke transport bogies before 1995 should be added to these sources.

Modifying the input parameters was continued until the value of the correlation coefficient between observed and modelled TDS values reached 0.972 based on data of 21 observation wells (only 10 of them is seen in the Figure 3.5.1.7 because of overlapping). All calibration points fell into the 95% interval of statistical exception. The standard deviation of TDS values calculated was 48 mg/L or about 9.6% of the initial homogeneous TDS attributed to the model. The mass transport model completed was reputed to be adequate because of authentic calibration results.

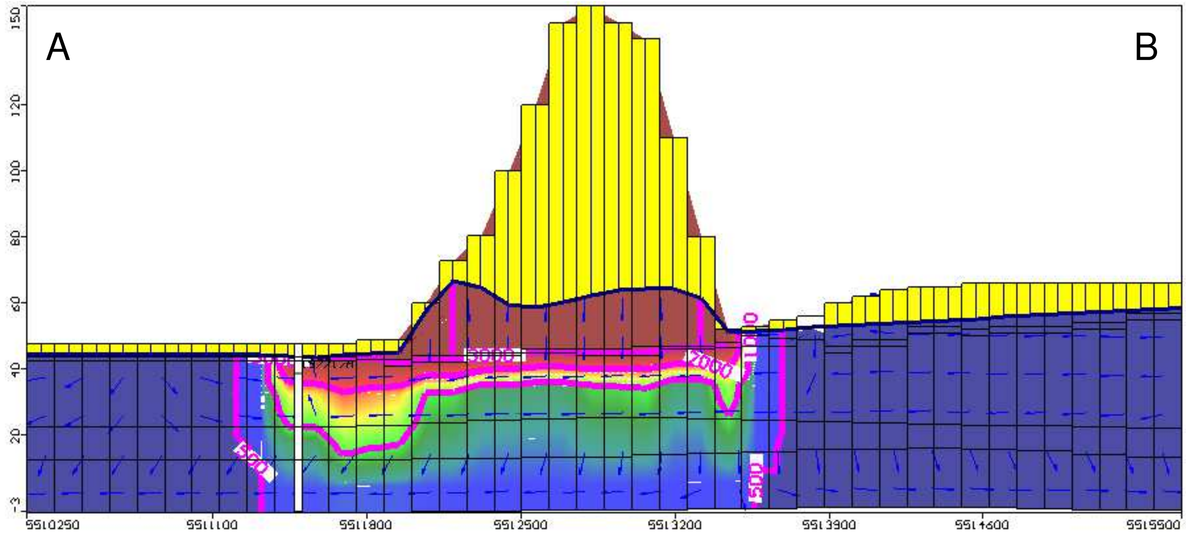

Fig. 3.5.1.9. TDS in the cross-section A–B of the Kohtla-Järve model in 2010. Graphical symbols – see the Fig. 3.5.1.1, 3.5.1.2, and 3.5.1.8.

Groundwater transport simulations carried out demonstrates that because of advection and hydrodynamic dispersion, the increased TDS intrudes from the 2nd model layer into the 3rd layer spreading both in a lateral and downward direction (Fig. 3.5.1.8, 3.5.1.9). The TDS value is predominantly 800–1,500 mg/L in the ZBL of the 3rd model layer in 2010. Westwards and northwards the TDS ranges from 1,500 mg/L to 3.0 mg/L or even more.

The lateral outline of the contamination plume has been spread eastward and south-eastward in the 3rd layer compared to areas of polluting sources determined as Constant Concentration boundary conditions in the 1st layer. This shift is almost equal along the perimeter of the plum and reaches up to 350 m. It is predominantly caused by longitudinal and horizontal components of the hydrodynamic dispersivity (Fetter C 1993; Matheron G, de Marsily G 1980; Neuman SP 2005; West M, Kueper B 2010; Woolfenden L, Ginn T 2009; Zheng C, Wang P 2003).

The TDS decreases downward from 3,000 mg/L to 700 mg/L in the eastern portion of the ZBL of the 3rd layer but in the western portion of the same ZBL – only to 2,000 mg/L (Fig. 3.5.1.9).

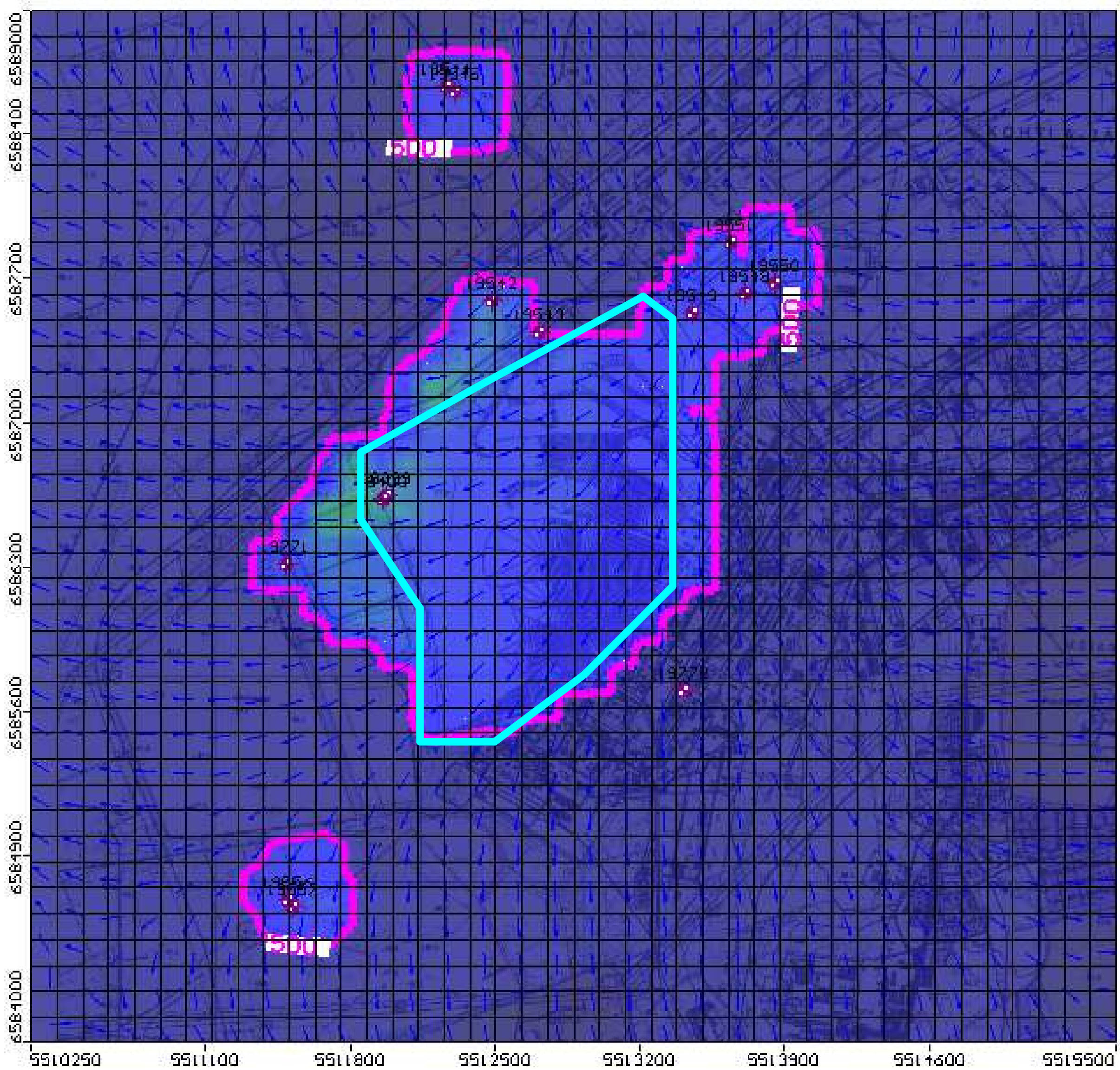

Fig. 3.5.1.10. TDS in the 5th model layer (O-Є aquifer system) of the Kohtla-Järve area in 2010. Graphical symbols – see the Fig. 3.5.1.1, 3.5.1.2, and 3.5.1.8.

The TDS decreases by 2–10 times at downward penetrating the 4th model layer. Therefore, TDS ranges only from 500 mg/L to 850 mg/L in the 5th model layer (Fig. 3.5.1.9, 3.5.1.10). Major TDS values are mostly situated in the north-eastern portion of the contamination plume. Lateral configurations of the pollution plume practically coincide in 3rd and 5th model layers. Groundwater transport modelling verifies that the pollution observed in the Ordovician-Cambrian aquifer system (Razgonjajev A 1997; Razgonjajev A, Timaškin R, Razgonjajeva L 1993; Savitski L, Savva V 2001) can be caused by downward migration of pollutants through the Lower Ordovician regional aquitard.

The output times of the MT3DMS engine were set to predict the transport situation in the study area in 2030, 2060, and 2110. The TDS was calculated for these dates.

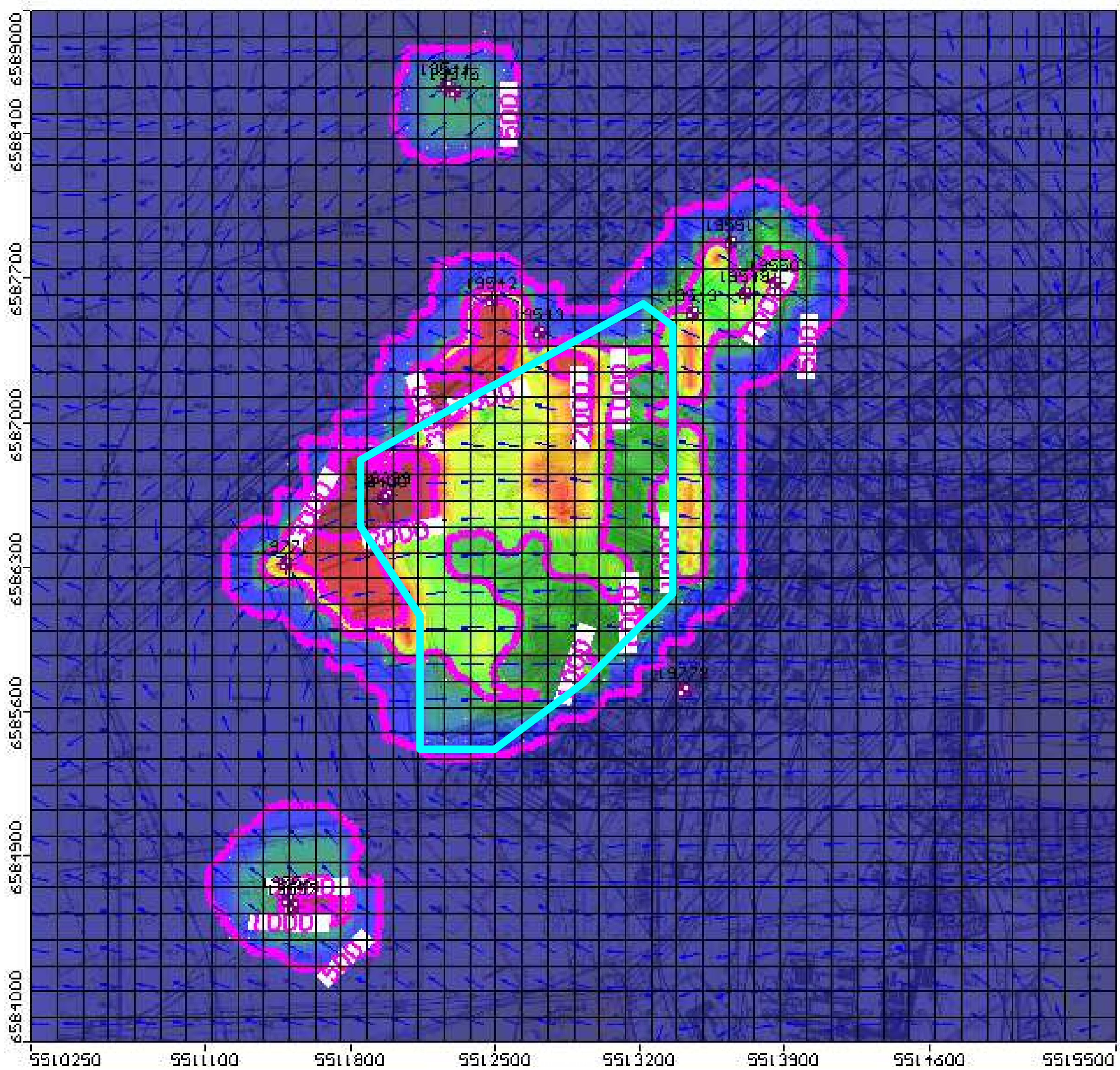

Fig. 3.5.1.11. TDS in the 3rd layer (S-O aquifer system) of the Kohtla-Järve model in 2060. Graphical symbols – see the Fig. 3.5.1.1, 3.5.1.2, and 3.5.1.8.

The contour of the TDS value equal to 1.000 mg/L will deepen by 1–2 m in the ZBL in 2030. The maximum TDS will reach 700 mg/L in the north-eastern portion of the pollution plume in the Ordovician-Cambrian aquifer system. The lateral area of pollution plume will practically not change.

In 2060, the lateral range of the contamination plum will also not significantly change but its downward intrusion will be considerable. The TDS will reach up to 8,000 mg/L in the Ordovician aquifer system and up to 900 mg/L in the Ordovician-Cambrian aquifer system (Fig. 3.5.1.11, 3.5.1.12, and 3.5.1.13).

In 2110, the contamination plume has been penetrated deeper, but its lateral extent has been not considerably expanded. The TDS will be up to 9,000 mg/L in the Ordovician aquifer system and up to 1,200 mg/L in the Ordovician-Cambrian aquifer system (Fig. 3.5.1.14, 3.5.1.15, and 3.5.1.16).

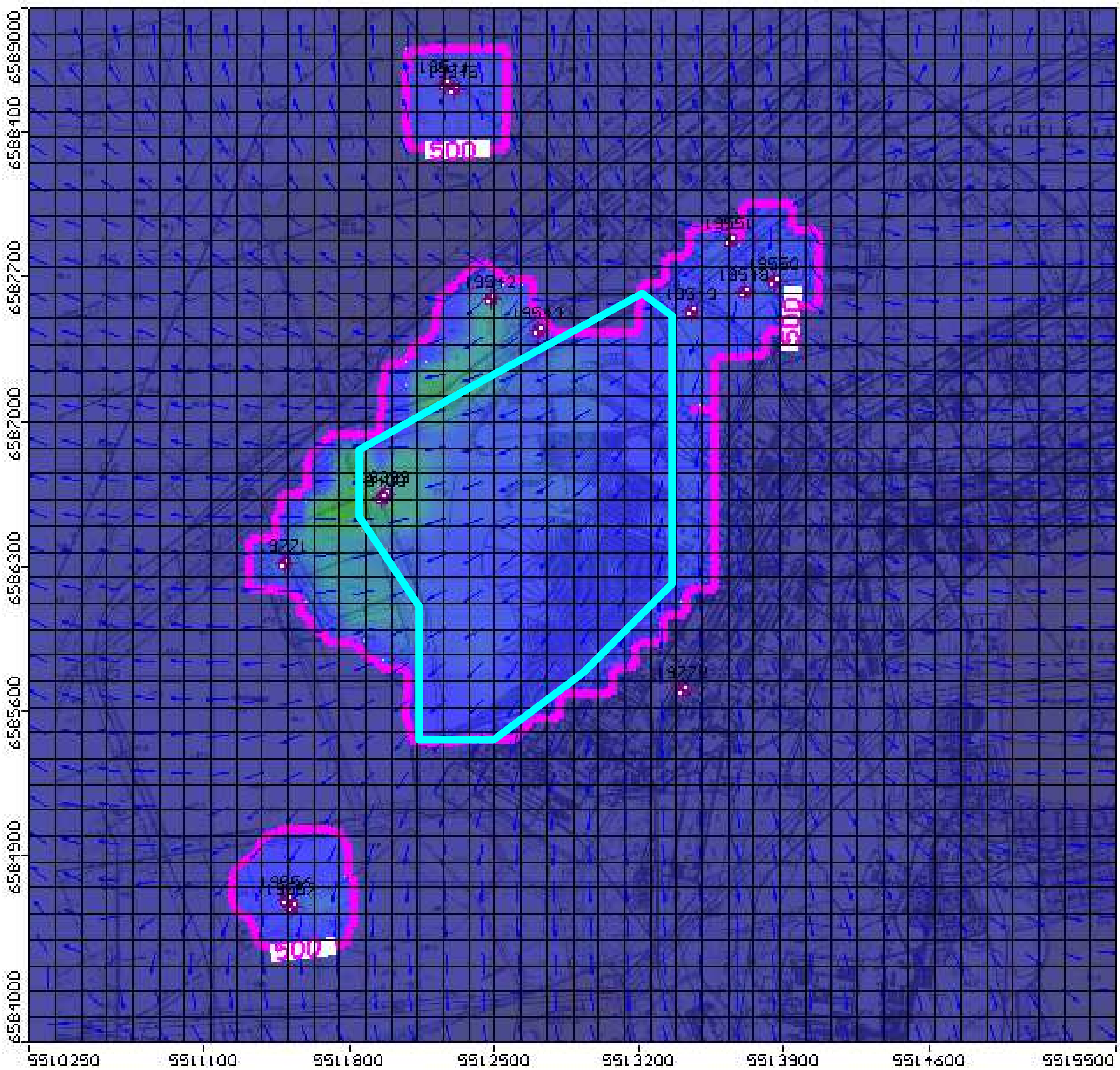

Fig. 3.5.1.12. TDS in the 5th layer (O-Є aquifer system) of the Kohtla-Järve model in 2060. Graphical symbols – see the Fig. 3.5.1.1, 3.5.1.2, and 3.5.1.8.

<u>The transport of phenols</u> occurring in the study area is subject to processes of sorption and biological degradation. It should be expressed by data of groundwater quality monitoring, but, unfortunately, they are too sparse for a detailed and completely convincing study of the phenol transport (Fig. 3.5.1.1, Table 3.5.1.1, 3.5.1.2).

The total of representative observation wells is 21, but only nine of these were more or less regularly sampled after every 3–4 months in between 1996–2010. The remainder of wells were sampled with the same frequency during 2–3 years. A number of gaps (blank cells in tables) occur ranging from a half of year to a year in observation series.

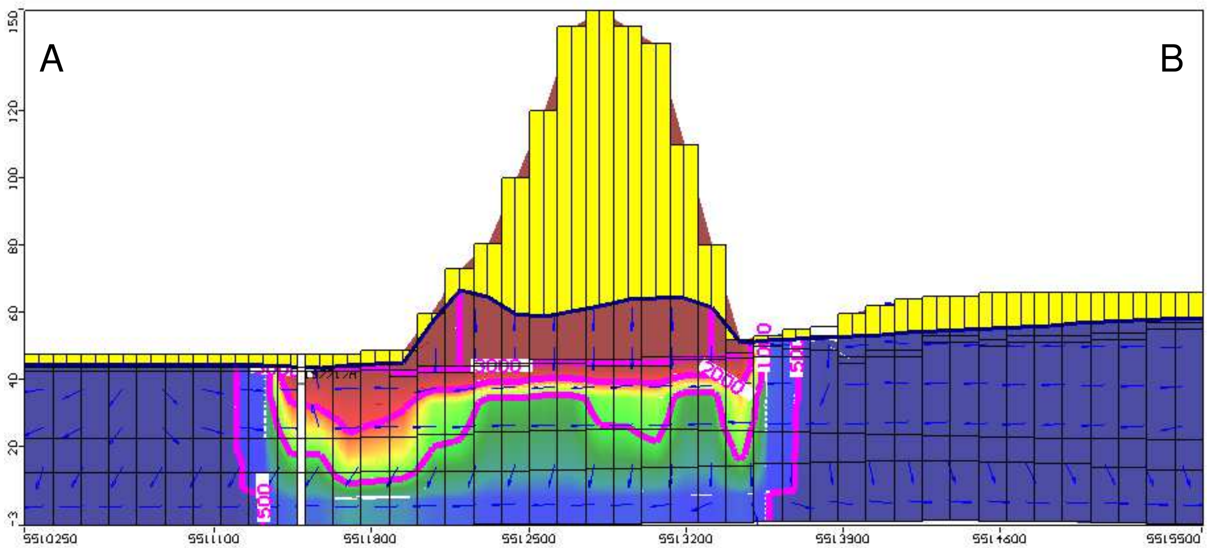

Fig. 3.5.1.13. TDS in the cross-section A–B of the Kohtla-Järve model in 2060. Graphical symbols – see the Fig. 3.5.1.1, 3.5.1.2, and 3.5.1.8.

Therefore, it is difficult to detect clear evidence of the transient degradation of phenols which half-life does not exceed a couple of weeks. Nevertheless, certain periods of a monotonic depletion of the phenol content lasting 6–14 months are still distinguishable in some observation series (blue numbers in Tables 3.5.1.1, and 3.5.1.2). These data refer to phenols having a comparative long half-life. On the other hand, the periods of monotonic increasing of the phenol content (red numbers in the Tables) have also been detected with monitoring. In some borings sudden sharp peaks of the phenols content alternate with the relatively long periods when phenols concentration is lower than precision their laboratory determination (LeFrancois M, Poeter E. 2009) or when phenols lack in water samples (cells containing the values <0.0001 or 0, respectively, in Tables 3.5.1.1, and 3.5.1.2).

The most of observation borings are coupled in the study area (Fig. 3.5.1.1). One of them is opening the Ordovician aquifer system and another taps the underlying Ordovician-Cambrian aquifer system in the same place (wells 18399/18400, 19540/19541, 19542/19543, 19544/19545, 19548/19549, 19550/19551, 19554/19555, and 19556/19557 in Tables 3.5.1.1, and 3.5.1.2). It allows preliminary to assess the distribution of chemicals in the vertical direction

However, observation wells tap the Ordovician carbonate bedrock or the Ordovician-Cambrian aquifer system as the whole without refining them into the thinner intervals. Therefore, the further details are unavailable about attenuation of the phenol content in the vertical direction. There are no sections of observation wells specially oriented along the main radial directions of the groundwater movement from the highest portions of the landfill toward the surrounding lowland. The distances between observation wells are too long to detect the rate of degradation of phenols at their lateral movement. All it aggravates the study of phenol distribution in a three-dimensional environment.

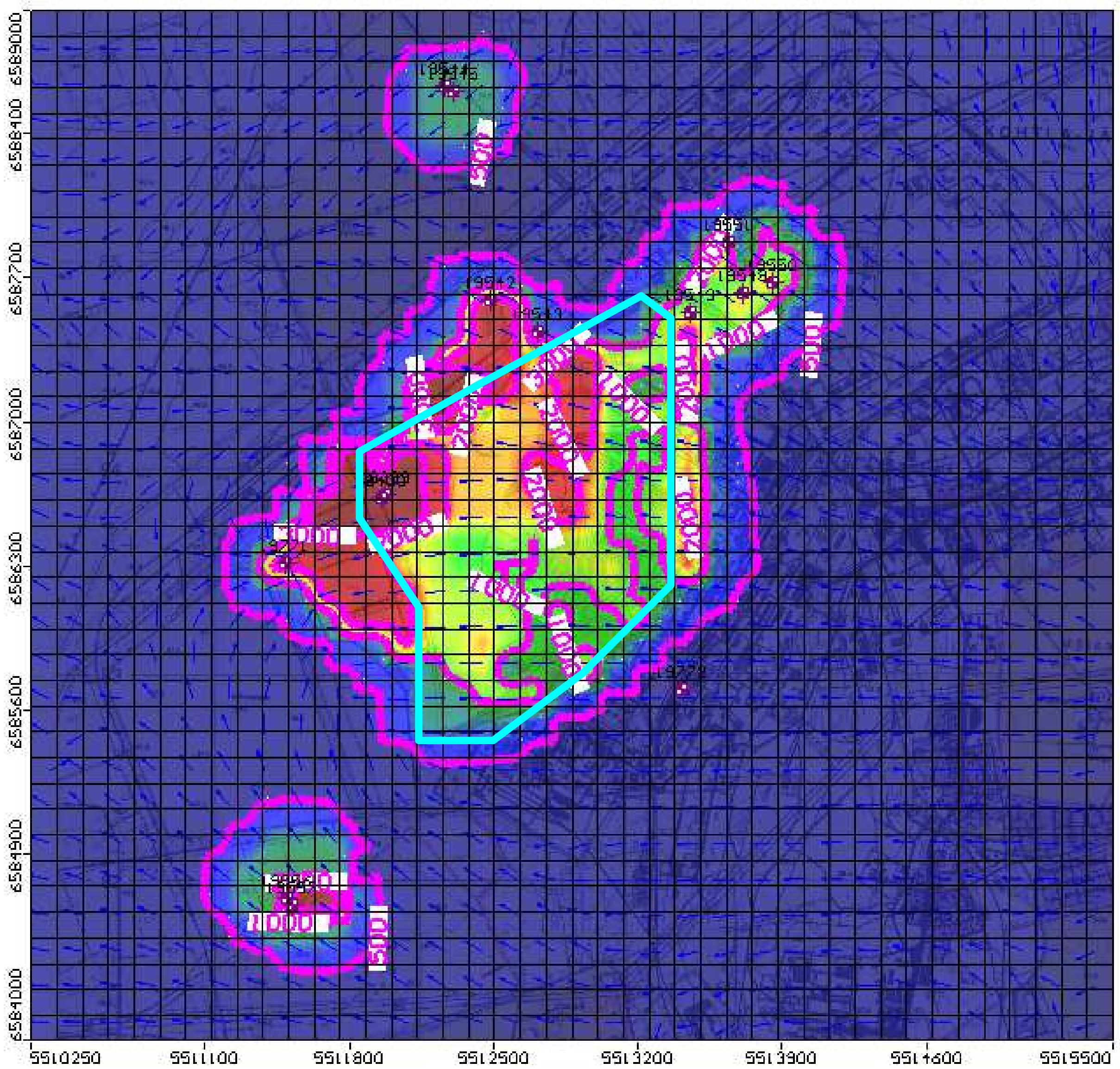

Fig. 3.5.1.14. TDS in the 3rd layer (S-O aquifer system) of the Kohtla-Järve model in 2210. Graphical symbols – see the Fig. 3.5.1.1, 3.5.1.2, and 3.5.1.8.

In spite of incomplete monitoring data, it was decided to construct a phenol transport model of the Kohtla-Järve landfill. For that purpose, the MT3DMS engine (Zheng C, Wang P 2003) incorporated into the Visual MODFLOW v. 4.5 was chosen for non-conservative transport calculations. It was assumed that equilibrium conditions existed between the solution-phase and solid-phase concentrations of phenols and that the sorption reaction was fast enough relative to groundwater velocity so it could be treated as instantaneous. Thus, the chemical reactions modelled were equilibrium-controlled linear sorption and first-order irreversible decay.

The conceptual model was based on a presumption that the initial concentration of phenols was zero in the study area. A leaching of phenols from the dump deposits into groundwater in the site of the existing landfill started in 1938. During the following 60–70 years the sum of phenols reached their values registered with monitoring performed from 1996 until 2010 (Fig. 3.5.1.1, Table 3.5.1.1, 3.5.1.2). The significant fluctuation of the phenols content complicated with peak concentrations detected with monitoring demonstrates that the real leaching intensity of phenols from dump deposits into groundwater was irregular both in the area and in the course of time.

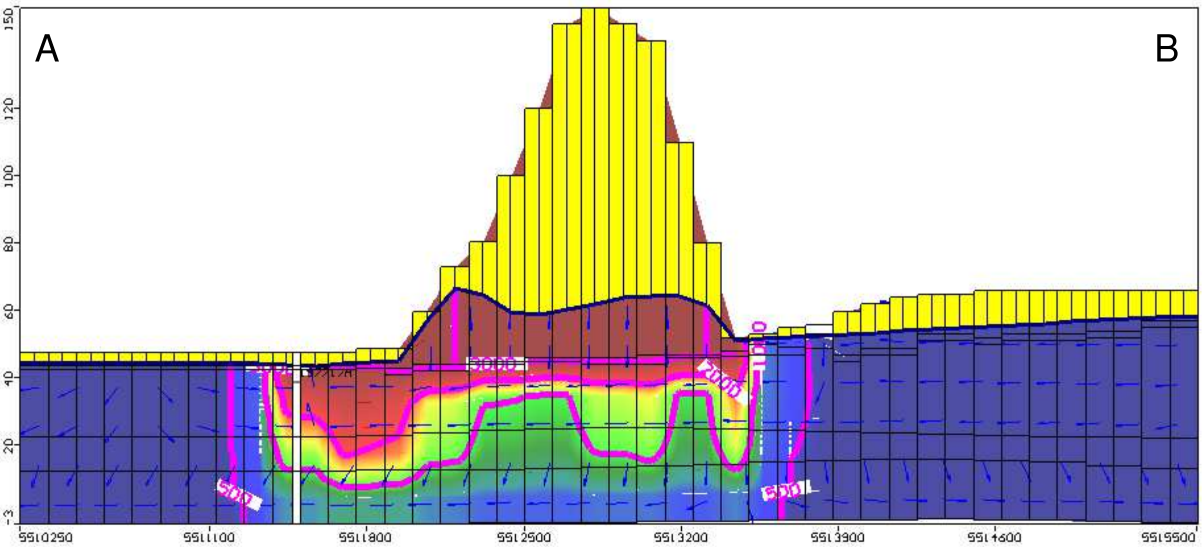

Fig. 3.5.1.15. TDS in the cross-section A–B of the Kohtla-Järve model in 2110. Graphical symbols – see the Fig. 3.5.1.1, 3.5.1.2, and 3.5.1.8.

The sharp or monotone increasing of phenols as well their episodic absence in monitoring wells can be explained by specialities of the preferential flow penetrating the vadose zone. During the snow smelting and intensive rains, a significant amount of water infiltrates into the landfill. More leachates of semi-coke are generated and they penetrate quite quickly the fingering of the vadose zone raising the content of phenols in the saturated zone. When a dry period arrives then the downward leaching flux decreases or dies away in some portions of the zone of the preferential flow. Alimentation of the saturated zone by chemicals from above decreases or stops provisionally and it reflects on their concentration. The interim disappearance of phenols from the water of the observation well can be explained by their washout from a portion of layer sampled or by biodegradation. These processes are also complicated by a permanent dumping and replacing of new amounts of semi-coke on the landfill.

Unfortunately, there were not enough monitoring data to reproduce the transient flux of phenols from the landfill into the underlying layers as an adequate transient boundary condition given to the 1st model layer. Therefore, it was presumed that the Constant Concentration boundary condition omitted to the uppermost model layer could be serving as a suitable supplementation of the transient boundary. Of course, this presumption was a rough approximation of the reality, but because of the lack of correct data in this manner was only possible to determine the three-dimensional distribution of phenols corresponding to observations. The concentration of phenols or their fluxes were not specified on the outer borders of the model

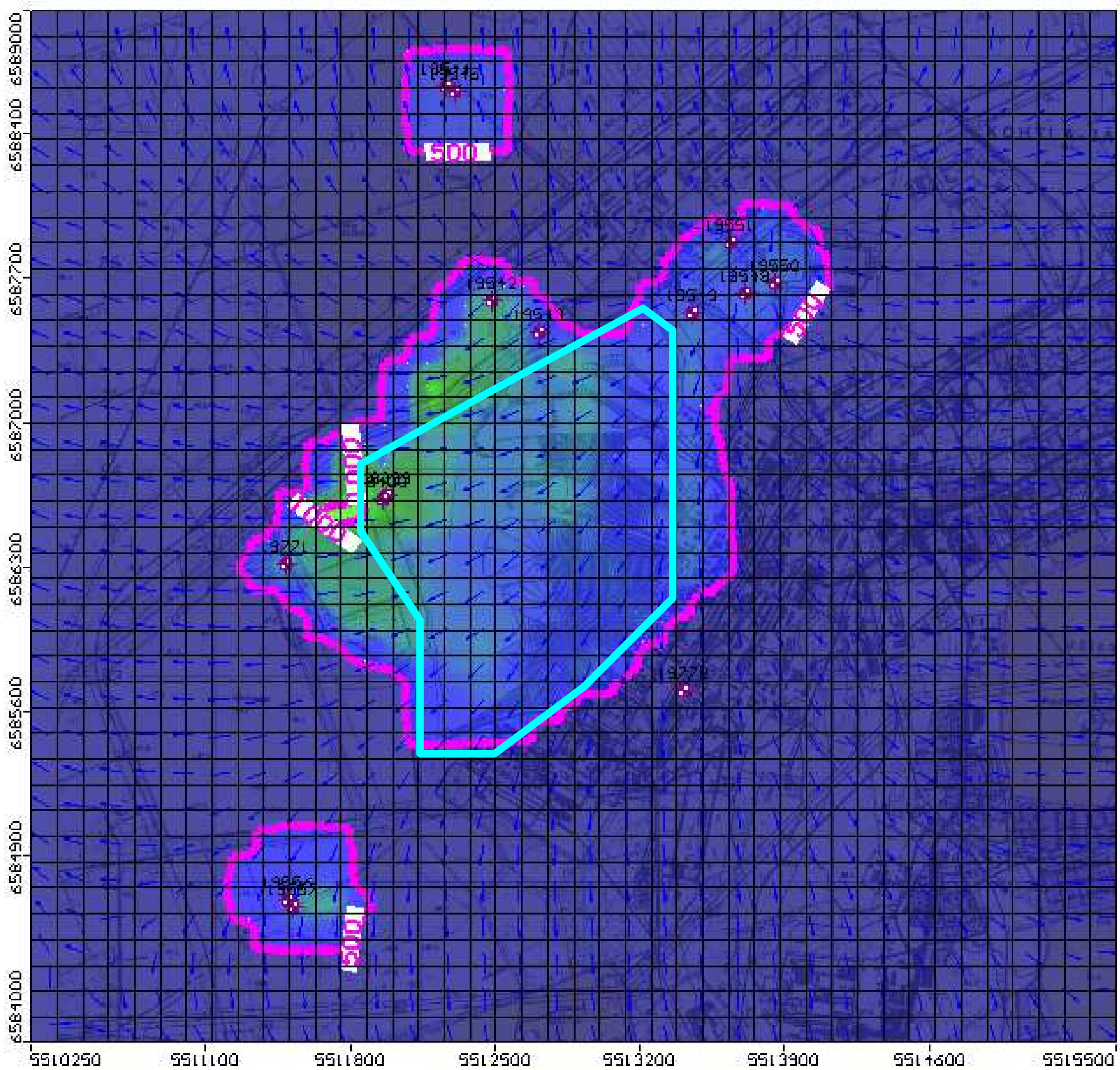

Fig. 3.5.1.16. TDS in the 5th model layer (O-Є aquifer system) of the Kohtla-Järve area in 2110. Graphical symbols – see the Fig. 3.5.1.1, 3.5.1.2, and 3.5.1.8.

Correspondingly to this conception, the Constant Concentration boundary condition was given to the 1st layer of the simulation model (Fig. 3.5.1.17). Initially, the values of the Constant Concentration were specified for cells of the 1st layer more or less conformably to the mean or median phenols concentrations of monitoring borings. Afterwards, the areal distribution of the Constant Concentration was corrected in the course of the model calibration set to 2010. The phenol content simulated was very sensible to the Constant Concentration boundary condition given. This condition established served as a source of phenol contamination of constant intensity during the whole simulation period ranging from 2010 until 2110.

The equation (3.2.1) is used to model the non-conservative transport of phenols, but for its numerical solution the values of the hydrodynamic dispersion coefficient tensor D_{ij} and the chemical reaction term ΣR_n should be determined beforehand (LeBlanc DR 2006; Voss CL, Provost AM 2002). The latter is written as follows (Bear J, Cheng AH-D 2010; Domenico PA, Schwartz FW 1998):

$$\Sigma R_n = - \rho_b (\partial \check{C}^k/\partial t) - \lambda_1 \theta C - \lambda_2 \rho_b \check{C}^k \qquad (3.2.3)$$

where

ρ_b	is the bulk density of the subsurface medium, $[ML^{-3}]$;
$\check{C}^k$	is the concentration of species k sorbed on the subsurface solids, $[MM^{-1}]$;
$\lambda_1 \theta C$	is the first-order reaction rate for the dissolved phase, $[T^{-1}]$;
θ	is the porosity of the subsurface medium, (dimensionless);
$\lambda_2 \rho_b \check{C}^k$	is the first-order reaction rate for the sorbed (solid) phase, $[T^{-1}]$.

The input of parameters *SP1, RC1*, and *RC2* is required by Visual MODFLOW code for executing of transport simulations. Their meaning is as follows (Zheng C. 1999; Zheng C, Wang PP 2003, 1998):

The distribution coefficient $SP1 = K_d$ $[L^3M^{-1}]$ belongs to the model of the equilibrium-controlled linear sorption chosen for the study of phenols migration. The function called the linear sorption isotherm

$$\check{C} = K_d C \qquad (3.2.4)$$

assumes that the sorbed concentration $\check{C}$ is directly proportional to the dissolved concentration C.

The reaction parameters $RC1 = K\_mobile = \lambda_1 \theta C$ and $RC2 = K\_sorbed = \lambda_2 \rho_b \check{C}^k$ are the first-order decay rates $[T^{-1}]$ both for the dissolved phase and for the sorbed (solid) phase. They express the biodegradation of phenols.

The first-order decay rate might be calculated by a formula

$$RC1 = (\ln 0.5)/(-t_{½}) \qquad (3.2.5)$$

where $t_{½}$ is the half-life time [T] of a groundwater ingredient subjected to biodegradation (LeBlanc DR 2006; Visual MODFLOW…2006). Experimental tests are needed for a correct determination of the half-life time of phenols in the real world conditions of Estonia (Studicky EA *et al*. 2010).

On the other hand, the formula (3.2.5) can obviously be rewritten for determination of the half-life time based on a *RC1* value given

$$t_{½} = (\ln 0.5)/RC1 \tag{3.2.6}$$

Then information about the trustworthy *RC1* value should be available.

Moreover, the longitudinal dispersivity α_L, the horizontal dispersivity α_H, the vertical dispersivity α_V, and the diffusion coefficient D^* must be incorporated into the model of the phenols transport.

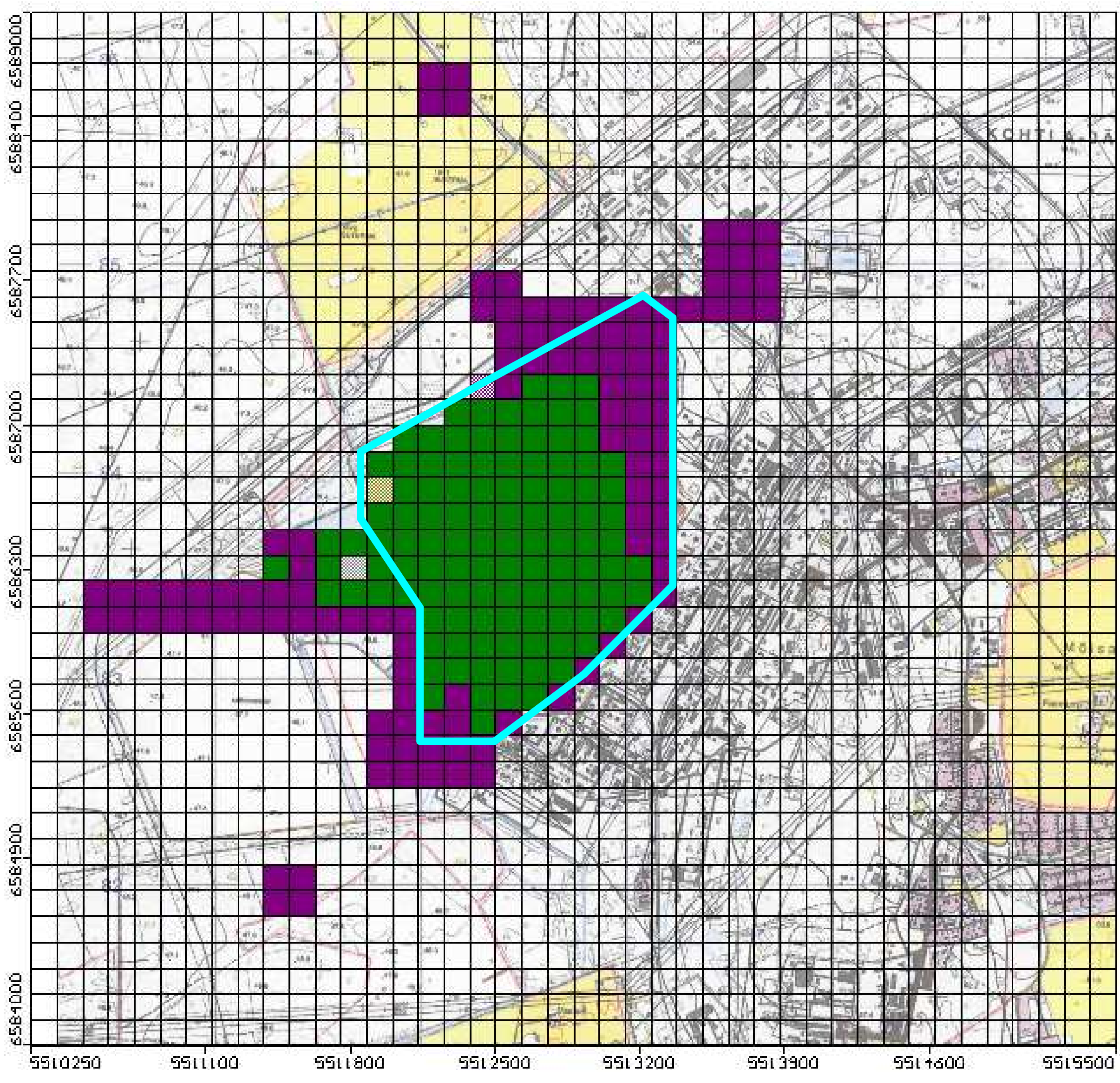

Fig.3.5.1.17. Constant Concentration boundary condition in the 1st layer (Quaternary deposits) of the Kohtla-Järve phenols transport model. Sum of phenols, mg/L: violet – 1.0, green – 20.0, light grey – 400.0, light olive – 500.0, light violet – 1000. Other graphical symbols – see the Fig. 3.5.1.1, and 3.5.1.2.

Since no field experiments were carried out to determine the parameters D^*, K_d, $SP1$, $RC1$, $RC2$, α_L, α_H, and α_V in the Kohtla-Järve study area, an inverse calibration method was used to compile a tried and true transport simulation model. As it was already described above at the modelling of the conservative transport of the TDS the supposed values of Constant Concentration boundary condition given to the 1st model layer (Fig. 3.5.1.17) and input parameters D^*, K_d, $SP1$, $RC1$, $RC2$, α_L, α_H, and α_V were modified. This procedure was continued until an optimum match between observed and calculated median values of the phenol sum was achieved with series of simulations.

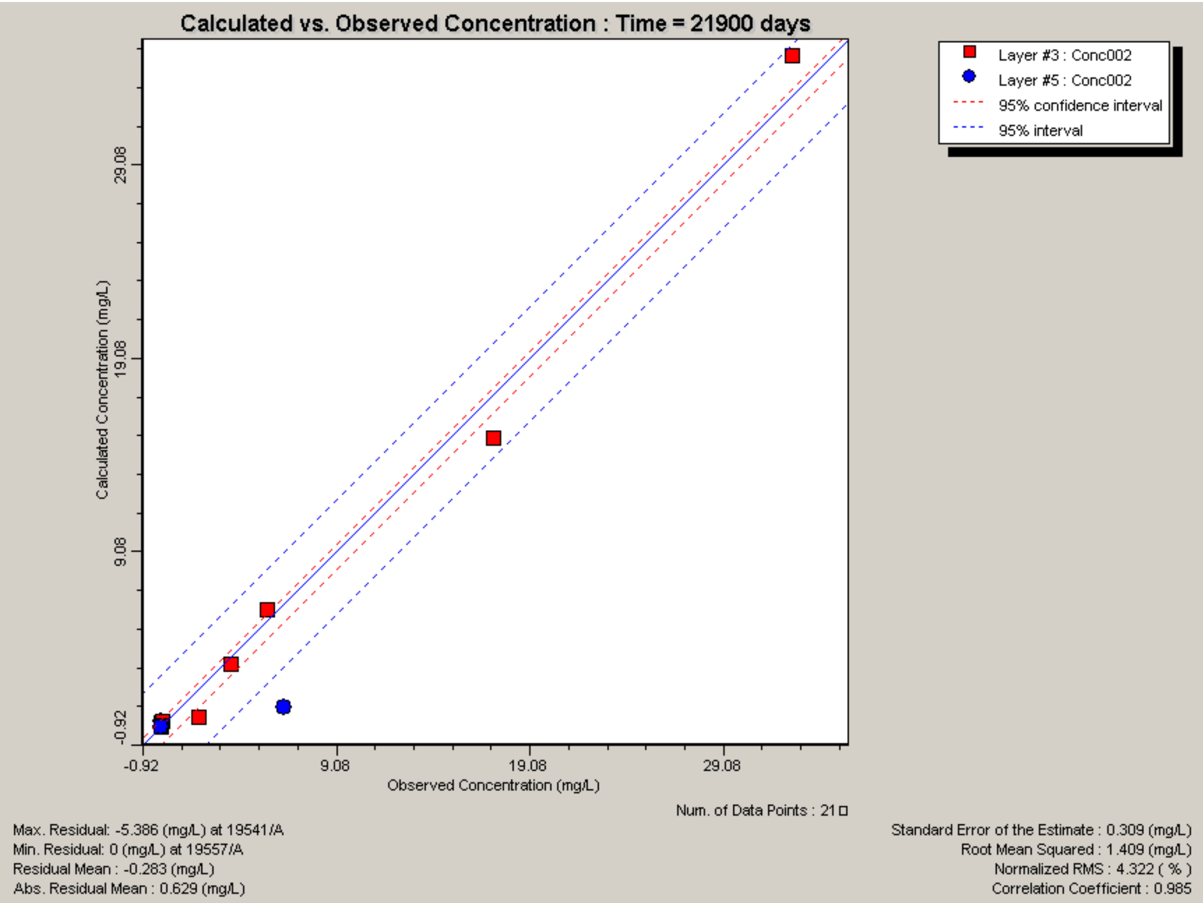

Fig. 3.5.1.18. Calibration graph of the Kohtla-Järve model of the phenols transport. Calibration was carried out based on data from 20 observation wells (Fig. 3.5.1.19), but because of graphical overlapping, only eight of them are seen on the plot.

Because of a significant variability of records, the median values of phenols content from monitoring series were mostly used as calibration targets. The test simulations were continued until the value of the correlation coefficient between the observed sum of phenols and modelled one reached 0.985 based on data from 21 monitoring wells (Fig. 3.5.1.18). Practically all calibration points fell into the 95% interval of statistical exception. The standard deviation of the sum of phenols simulated for a calibration point was 0.31 mg/L. The spatial distribution of phenols simulated for 2010 seemed to be realistic fitting the data monitoring. Such calculation result attesting a sufficient formal adequacy of the transport model of phenols completed was got at values: $D^* = 0.1$ m^2/day, SP1 $= K_d = 1.1 \cdot 10^{-6}$ L/mg, RC1 = 0.001 1/day, $RC2 = 0.0001$ 1/day, $\alpha_L = 50$ m, $\alpha_H = 5$ m, $\alpha_V = 0.5$ m, $\rho_b = 1{,}700$ kg/m^3 = $1.7 \cdot 10^6$ mg/L, and $\theta = 0.25$.

As much as the value *RC1* = 0.001 1/day was affirmed by model simulations it was substituted into equation (3.2.6) for determination of the half-life time of phenols:

$$t_{½} = 0.4262/0.001 = 426 \text{ days.}$$

It is about 60 times more than the half-life of phenols for the Kiviõli and Kohtla-Järve landfills determined by K. Rudolph-Lund (Sørlie J-E. *et al.* 2004).

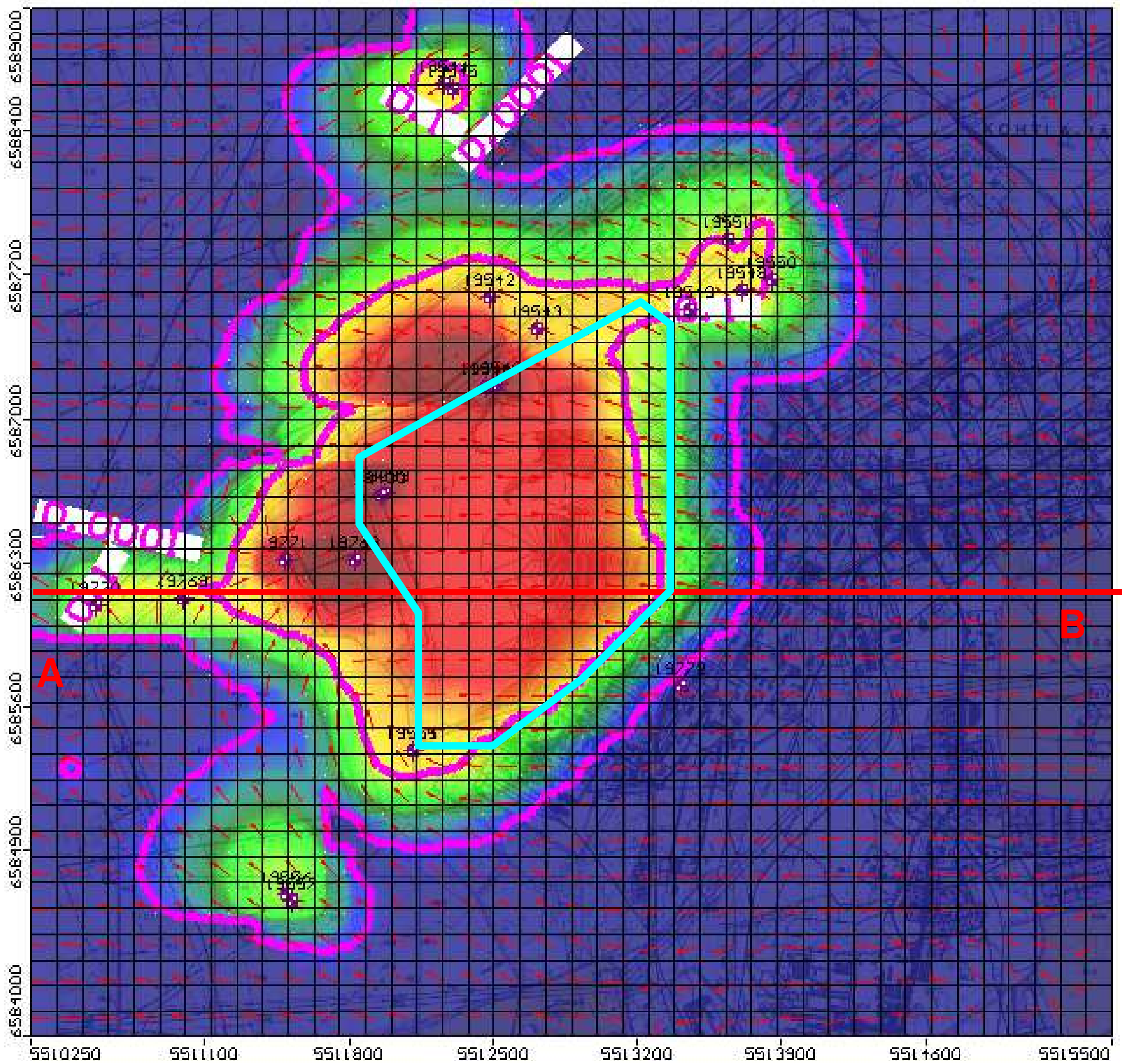

Fig. 3.5.1.19. Sum of phenols in the S-O aquifer system of the Kohtla-Järve model in 2010. Gradation of the sum of phenols, mg/L: dark blue – 0–0.0001, light blue – 0.0001–0.005, green – 0.005–0.01, light green – 0.01–0.1, yellow – 0.1–1.0, light brown – 1.0–5.0, brown – >5.0. Violet isolines – concentration of phenols, mg/L. Small violet circles indicate concentration observation wells and their numbers. Other graphical symbols – see the Fig. 3.5.1.1, and 3.5.1.2

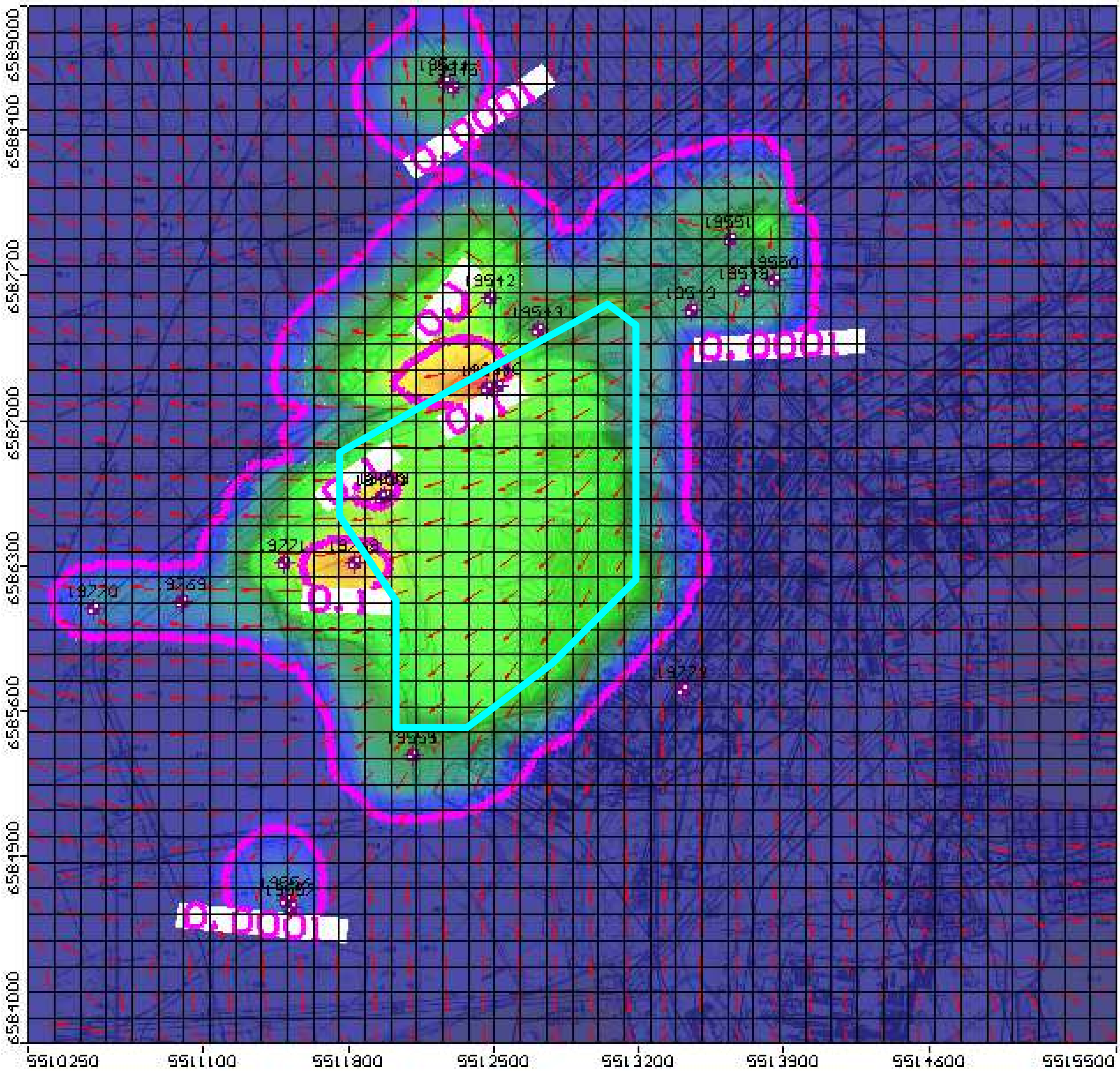

Fig. 3.5.1.20. Sum of phenols in the O-Є aquifer system of the Kohtla-Järve model in 2010. Graphical symbols – see the Fig. 3.5.1.1, 3.5.1.2, and 3.5.1.19.

The migration of contaminants (phenols) in the subsurface environment has generally delayed due to interactions between the water ingredients and the solid surfaces of the porous media. This phenomenon is described by the function (Bouwer H 1991)

$$R = V/V_c \tag{3.2.7}$$

where R is the retardation factor (dimensionless), V [LT^{-1}] is the velocity of groundwater, and V_c [LT^{-1}] is the velocity of the contaminant species.

If none of a particular species has retarded then R = 1 and the contaminant travels along with the water at the groundwater flow rate. When R is large, the contaminant can take many times to migrate offsite.

The retardation factor R is defined as (Bouwer H 1991; Zheng C. 1999; Zheng C, Wang P 2003, 1998):

$$R = 1 + (\rho_b/\theta)\ K_d. \tag{3.2.8}$$

Substitution of parameters $\rho_b = 1.7*10^6$ mg/L, $\theta = 0.25$ (O-Є aquifer system), and $K_d = 1.1*10^{-6}$ L/mg into the formula (3.2.8) gives the value of the retardation factor:

$$R = 1 + [(1.7*10^6)/(0.25)]*1.1*10^{-6} \approx 1 + 7.5 \approx 8.5.$$

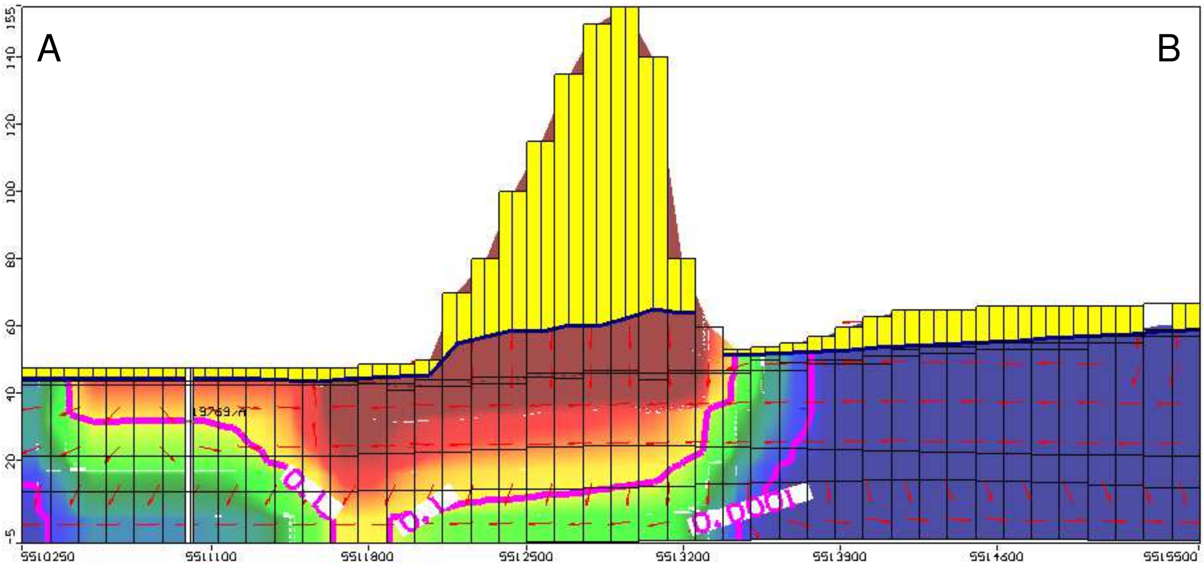

Fig. 3.5.1.21. Sum of phenols in the cross-section A—B of the Kohtla-Järve model in 2010. Graphical symbols – see the Fig. 3.5.1.1, 3.5.1.2, and 3.5.1.19.

Unfortunately, the content of phenols in drinking water is not standardized by regulations of the Estonian Ministry of the Social Affairs (Joogivee kvaliteedi… 2001). However, the precision of chemical analyses at the determination of phenols used in the present report is mostly 0.0001 mg/L (personal communication of L. Bityukova). This value should mark the lowest concentration indicating that groundwater is contaminated by phenols. Another hand, accordingly to a regulation of the Estonian Ministry of the Environment the groundwater containing phenols more than 0.1 mg/L must be attested as a contaminated one and measures should be taken to remediate its quality (Ohtlike ainete… 2010). Both these criterions are marked with isolines of 0.0001 mg/l and 0.1 m/L in maps and sections representing the spatial distribution of phenols simulated for the model areas. Thereat, the concentration of phenols is also differentiated by colour shading in figures. The output times of phenol transport simulations performed were set on 2010, 2030, 2060, and 2110.

Accordingly, to transport simulations, the area of groundwater contaminated by phenols is about 10.6 km^2 in the Ordovician aquifer system of the Kohtla-Järve model in 2010 (Fig. 3.5.1.19). It takes nearly 40% of the total area of the Kohtla-Järve site.

Phenols concentration ranges predominantly from 0.1 mg/L to 5 mg/L in the ZBL, but increases to 10–34 mg/L beneath sedimentation basins and their connecting canals surrounding the western side of the landfill. The contamination plum is stretched by 300–400 m eastward from the landfill upstream to the prevailing groundwater flow because of dispersion. In return, the contamination plum extends to the distance of 1000–1300 m downstream of the groundwater flow westward and northward from the landfill and sedimentation basins. In the same directions, the phenols contamination should even cross the borders of the study area.

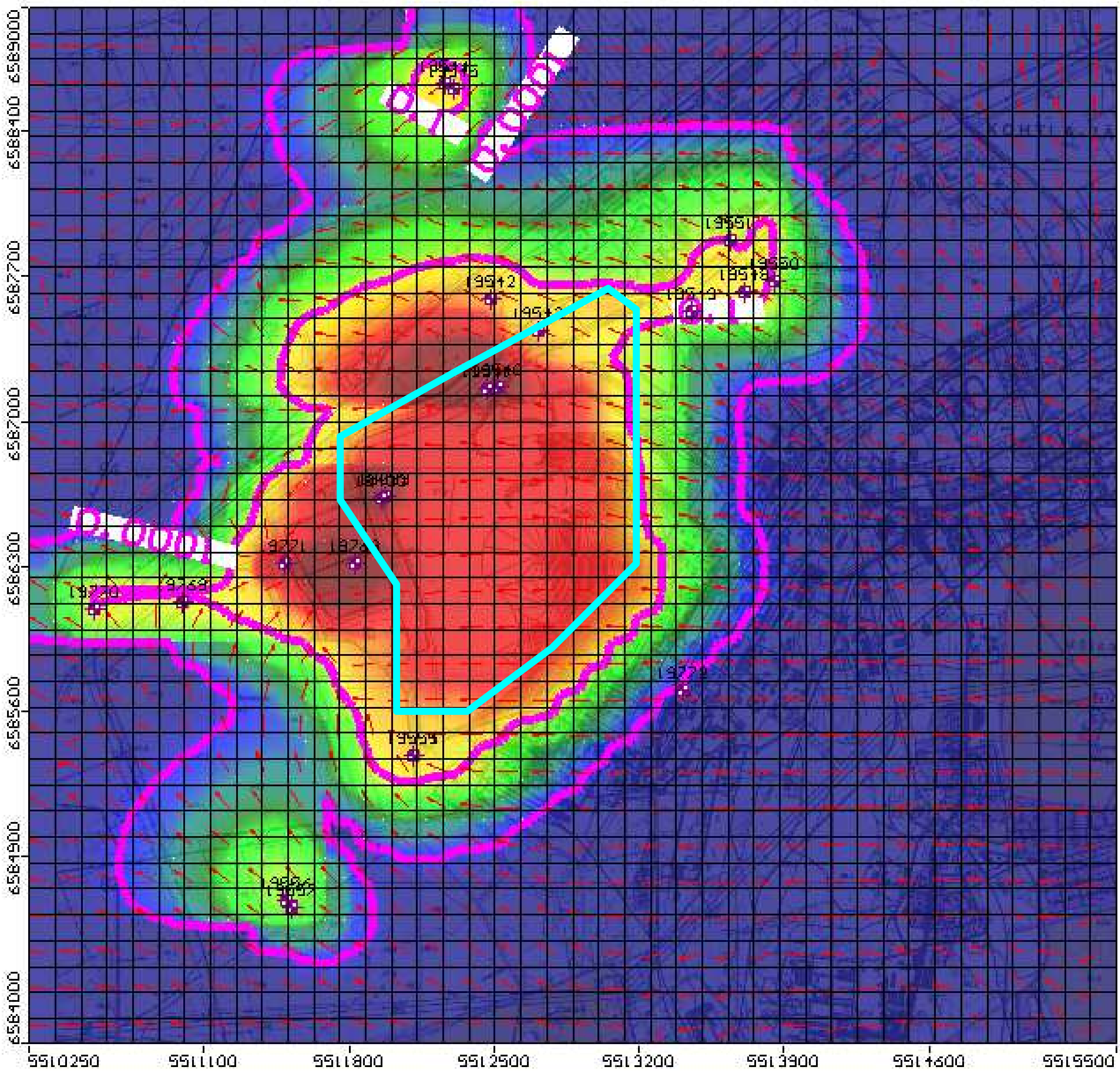

Fig. 3.5.1.22. Sum of phenols in the O aquifer system of the Kohtla-Järve model in 2110. Graphical symbols – see the Fig. 3.5.1.1, 3.5.1.2, and 3.5.1.19.

The downward groundwater flux contaminated by phenols has been penetrated the regional Silurian-Ordovician regional aquitard and is spreading in the Ordovician-

Cambrian aquifer system in 2010 (Fig. 3.5.1.20, 3.5.1.21). The present concentration of phenols is chiefly 0.1 mg/L in the ZBL of the O-Є aquifer system but rises up to 1–3 mg/L under the western sedimentation basins. The area of the contamination plum is 7.5 km^2 in the Ordovician-Cambrian aquifer system in 2010. It takes some 29% of the total of the Kohtla-Järve study area. In comparison with Ordovician aquifer system, the lateral extent of the phenols contamination plum is narrower by 100–200 m in the Ordovician-Cambrian aquifer system.

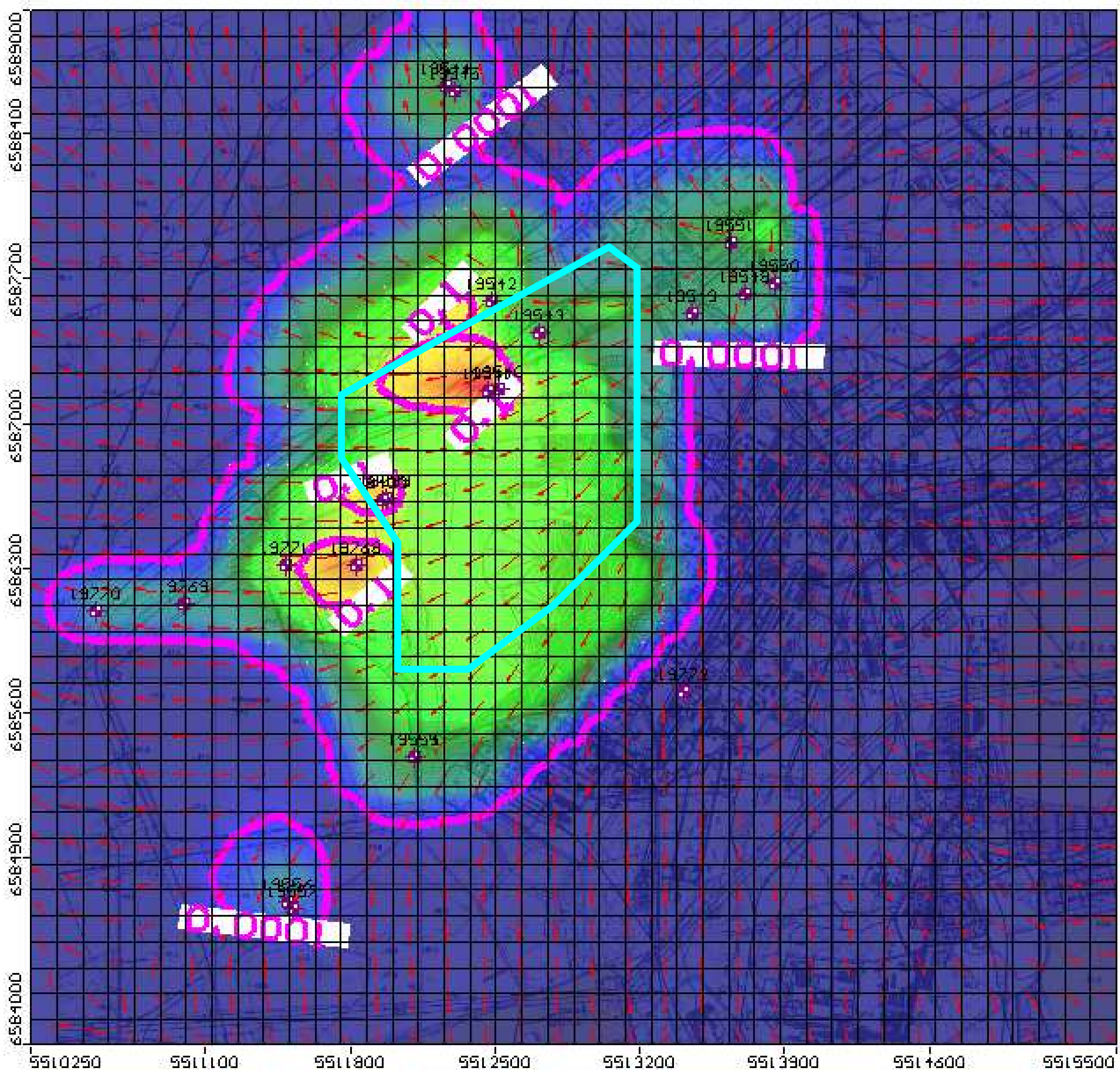

Fig. 3.5.1.23. Sum of phenols in the O-Є aquifer system of the Kohtla-Järve model in 2110. Graphical symbols – see the Fig. 3.5.1.1, 3.5.1.2 and 3.5.1.19.

The simulations carried out demonstrate that the lateral phenols contamination will remarkably not expand in the Ordovician aquifer system during next 100 years (Fig.

3.5.1.22). If the intensity of the source of phenol contamination will not change in course of next 100 years then the area of phenols contamination will also essentially not expand laterally in the Ordovician-Cambrian aquifer system (Fig. 3.5.1.23), but the concentration of phenols simulated will increase in all layers. It will reach 57 mg/L in Ordovician carbonate bedrock beneath the northern border of the landfill and 10 mg/L under the western sedimentation basin in the O-Є aquifer system.

The transport simulations of both the TDS and phenols prove that the Silurian-Ordovician regional aquitard does not prevent the downward intrusion of contaminated groundwater from the overlying carbonate bedrock into the Ordovician-Cambrian aquifer system in the Kohtla-Järve study area. Therefore, the contaminants can intrude into the Ordovician-Cambrian aquifer system everywhere in the East-Viru County where the landfills of polluting residuals of the oil shale industry occur.

Adequacy of simulations performed. An optimum suitability of existing groundwater monitoring data and probable values of transport parameters was sought by simulations carried out. Since the transport parameters (the half-life of phenols $t_{½}$, the distribution coefficient K_d, the diffusion coefficient D^*, the coefficient of the hydrodynamic dispersion D, coefficients of longitudinal, horizontal, and vertical dispersion α_L, α_H, and α_V, respectively) were not experimentally determined for the model area, their values were established on ground of scientific literature. At that, certain deviations of transport parameters from their real values were inevitable. Because of these deviations and due to inexactness of monitoring data the modelling results cannot be completely adequate to the real world.

The position of the outer contour of the phenols contamination plum equalled to their concentration of 0.0001 mg/L has probably an error of ±100 m upstream to the groundwater flow and up to ±500 m downstream. The same error in the vertical direction can reach ±20 m. The error of phenols concentration simulated can be taken equal to their standard deviation of the estimate at model calibration that is 0.13 mg/L (Fig. 3.5.1.18).

Thus, results of simulations are not unique from a rigid formal standpoint. However, they try to offer a possible best fit between theoretical expectations and real opportunities. Despite the lack of sound and sufficient source data, the conclusions drawn are logical and they determine quantitatively the main characteristics of phenomena's studied.

3.5.2. Kiviõli model

Model area. The Pulkovo 1942 co-ordinates of the rectangular Kiviõli model area are: lower left corner 5494500; 6581000, and upper right corner 5499500; 6584500 (Fig. 3.5.2.1). The area measures 5000 m from west to east and 3500 m from north to south. Before the start of industrial operations, the topography of the area was flat with absolute elevations between 45–55 m. The solid residuals of the oil shale enrichment and processing (semi-coke) were piled in the central part of the model area where three detached landfills occur. The absolute elevation of two main semi-coke landfills reaches 120–130 m. Between main landfills is situated a lower one containing the waste rocks with a top elevation about 65 m. The area of landfills is surrounded halfway from the northwest, north, and northeast by the Koljala Creek and its branches. The local drainage basis for the channel network and upper aquifers is Purtse River remaining from 1 to 3 km eastward from the model area. Oil-shale mines and opencast pits abandoned and drained by ditches occur in the south-eastern part of the area. They extend up to 0.3–0.8 km northward from the Tapa-Narva railway line (Fig. 3.5.2.1). The study area was covered by a virtual orthogonal modelling grid including 28 rows and 40 columns with a spacing of 125 m.

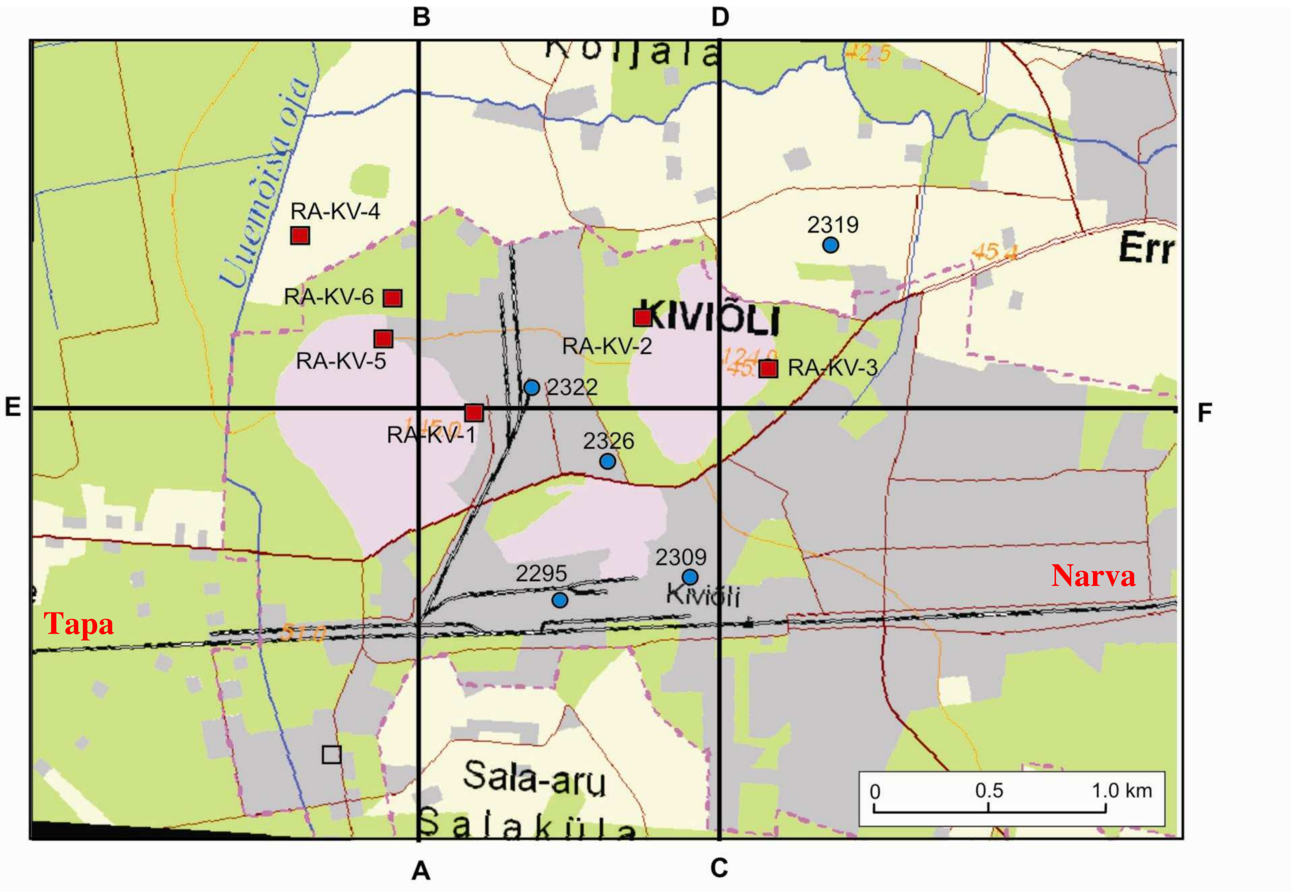

Fig. 3.5.2.1. Kiviõli model area. Localities: light green – woodland, beige – field or clearing, grey – industrial or dwelling area, pink – landfills. Hydrogeological units tapped by observation borings: red quadrates – Quaternarian deposits and Ordovician carbonate bedrock, blue circles – the O-Є aquifer system. Hydrogeological cross-sections: A–B, C–D, and E–F. Tapa-Narva – railway line.

<u>Model layers and boundary conditions.</u> The unconfined 1st layer enfolds the semi-coke and oil shale ash inside borders of landfills. The vertical hydraulic conductivity assigned to dump deposits is 0.005–0.05 m/day. The natural Quaternary deposits belonging to the layer are represented predominantly by till outside the landfills. The average conductivity given to the natural Quaternary deposits ranges from 0.5 m/day to 2.5 m/day. The southern third of the 1st layer is dried up due to the impact of channel network and mine drainage.

The Constant Head boundary conditions were assigned along the northern and partially western and eastern borders of the 1st layer. Boundary conditions were not specified for the remaining portion of layer borders. In the last case was supposed that the groundwater stage depends predominantly on the mine drainage impact and it can be estimated by modelling.

The net infiltration into the 1st layer of the flow model equalled to 60 mm/year and the riverbed conductance reaching 500–1000 m^2/day were estimated accordingly to former investigations (Vallner L 2003, 2002, 1996b).

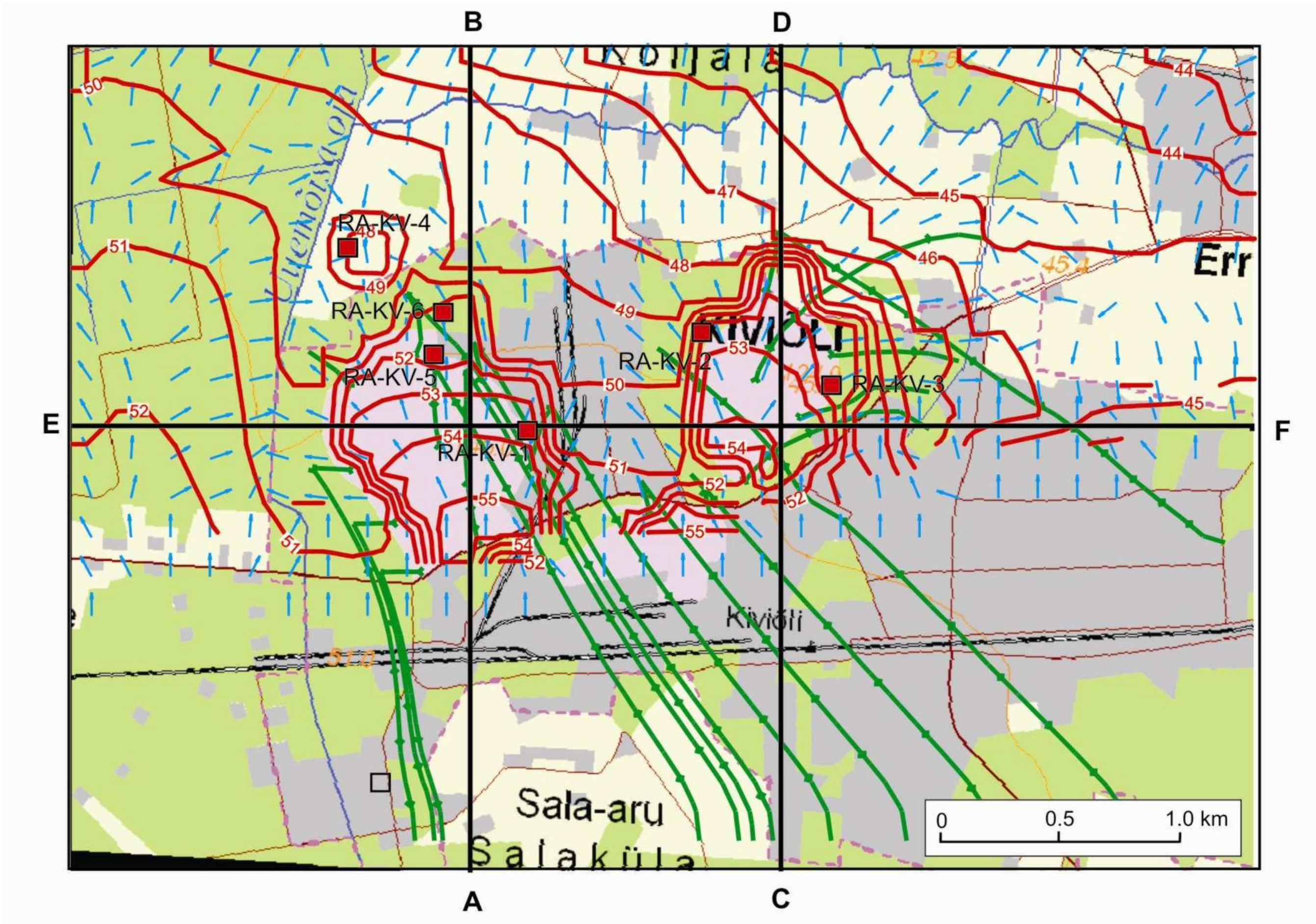

Fig. 3.5.2.2. Groundwater table in the topmost model layer in the Kiviõli area: red isolines – water table, m a.s.l. Blue arrows – the direction of the groundwater movement in saturated portions of 1st, 2nd, and 3rd model layer. Groundwater flow pathlines in the 1st, 2nd, and 3rd layer: green; the distance between green arrowheads is equal to 10 years of the groundwater movement.

The 2nd model layer with the thickness of 3–4 m serves as an arbitrary tool. It is needful for creating a semi-pervious screen beneath the 1st layer. Without such of the screen, the water table elevation modelled would be significantly lower than measured one in semi-coke landfills. To the 2nd layer belong the lowermost seams of semi-coke compressed by the weight of overlying layers, the lower portion of natural Quaternary deposits and uppermost seams of the Ordovician carbonate bedrock. The vertical conductivity of the 2nd layer is 10^{-6}–10^{-5} m/day in the area of semi-coke landfills and their vicinity. In the remained area, the conductivity of this layer changes from 0.005 to 0.1 m/day**.** The 2nd layer is considered as confined-unconfined at modelling. The southern third of the 2nd layer is also dried up due to the impact of channel network and mine drainage. The boundary conditions of the 2nd layer are the same as in the 1st layer.

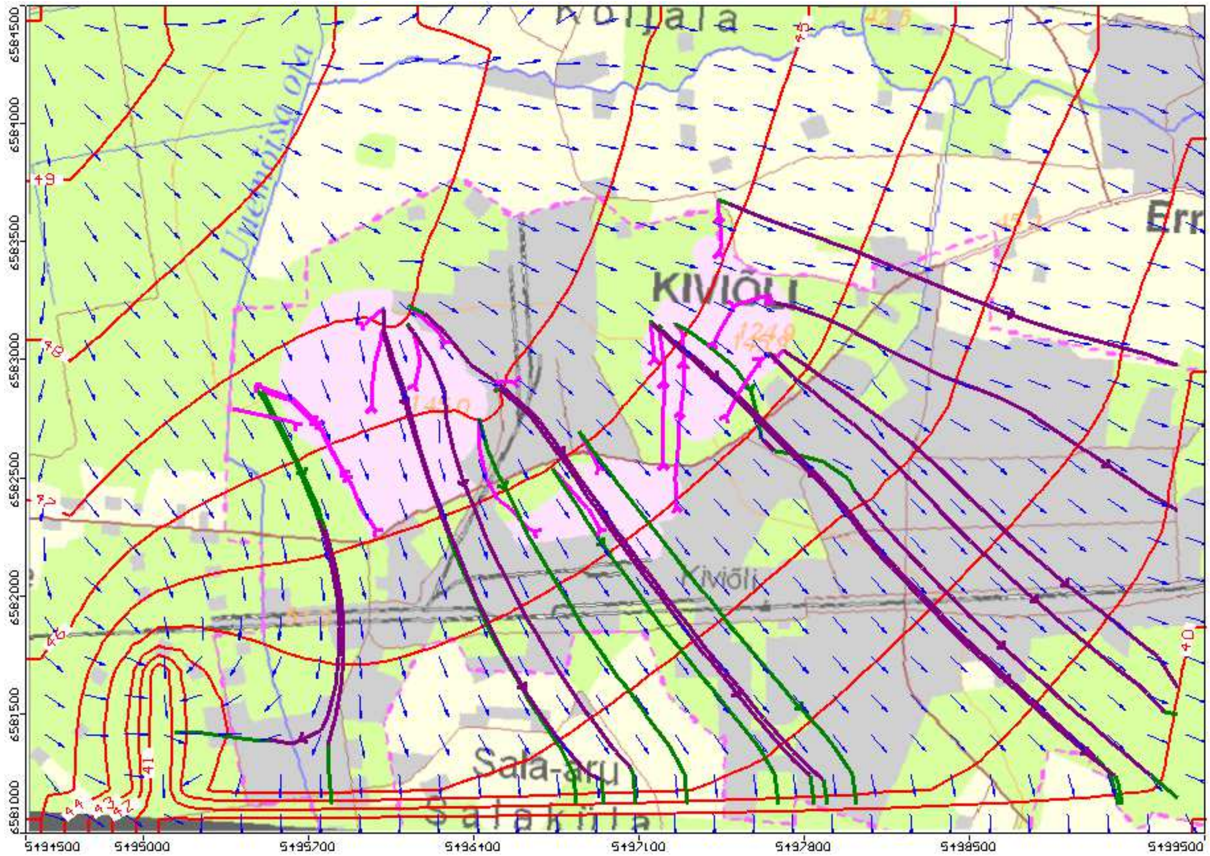

Fig. 3.5.2.3. Groundwater head in the 3rd and 4th model layers (O aquifer system) in the Kiviõli area: red isolines m a. s. l. Groundwater flow directions: blue arrows. Flow pathlines in the 3rd model layer – green, pathlines above the 3rd layer - light violet, below the 3rd layer – dark violet. The distance between arrowheads on pathlines is equal to 10 years of the groundwater movement. The distance between outer ticks is 700 m on the X-axis and 500 m on the Y-axis of the site map.

The 3rd layer comprising the oil-shale commercial bed enfolds the Kukruse Stage and upper seams of the Uhaku Stage which total thickness ranges from 12–15 m to 2–3 m. In the southern third of the study area, the oil-shale commercial bed has been excavated. The former opencast pits and underground mines have been abandoned and partially or completely filled with water at present. An underground cracking of oil shale was carried out in the southwest portion of the 3rd layer in the Soviet era. This experiment failed and the bedrock layers were heavily contaminated by oils and phenols. To prevent the underground spreading of this pollution the water table is kept at the absolute elevation of 41 m by a ditch system situated in the southwest corner of the 3rd layer. This situation as well the general distribution of groundwater head on the carbonate bedrock was taken into account at assigning the boundary conditions to the 3rd layer. Because of mine draining the layer is considered as unconfined-confined at modelling. The conductivity of the layer ranges from 0.1 to 8 m/day.

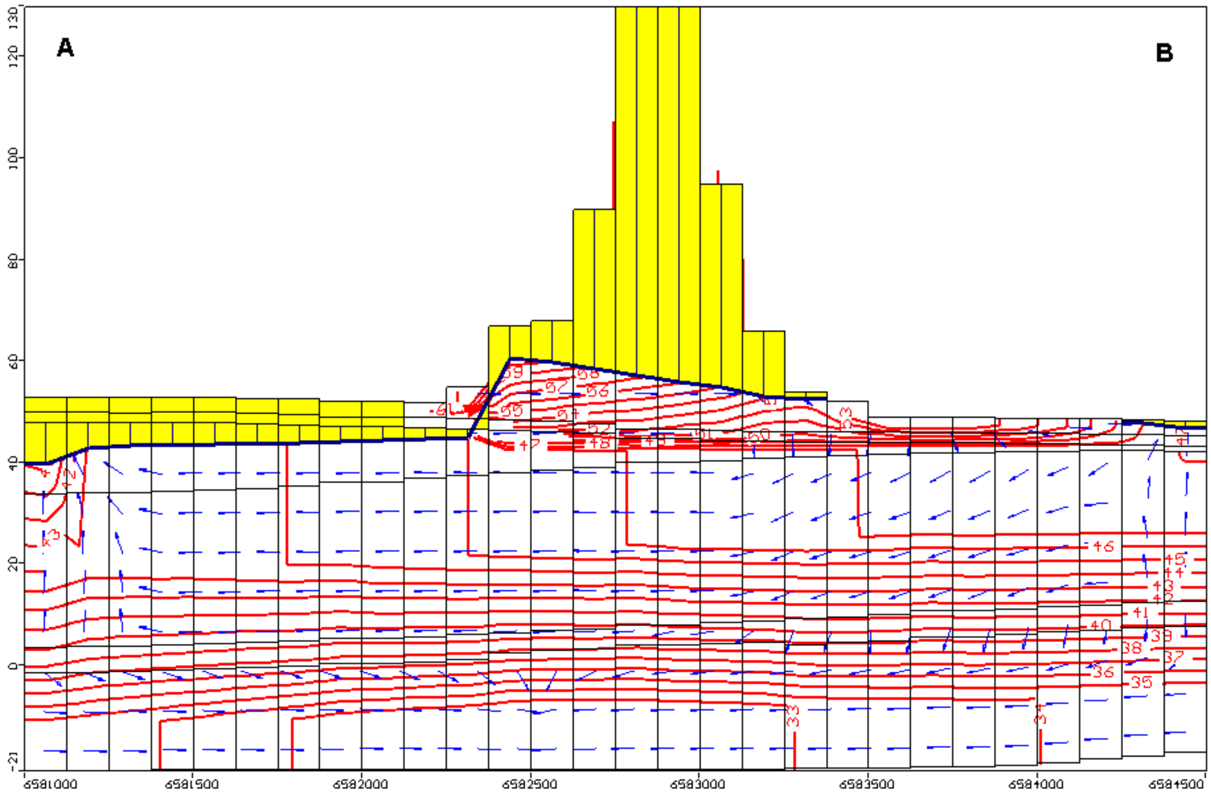

Fig. 3.5.2.4. Hydrogeological cross-section A–B in the Kiviõli area. Groundwater table – dark blue contour, vadose zone – yellow. Groundwater head – red isolines, m a.s.l. Groundwater flow directions – blue arrows. The distance between outer ticks on the X-axis is 700 m, absolute elevation on the Y-axis, m.

The 4th layer having the mean thickness of 30 m represents the lower portion of the confined Silurian-Ordovician aquifer system from the Lasnamäe Stage until the Kunda Stage (Perens R, Vallner L 1997; Raukas A, Teedumäe A 1997). Its lateral conductivity is 4 m/day and transversal conductivity is 0.4 m/day in average.

The 5th layer conforms to the Silurian-Ordovician regional aquitard which lateral conductivity $K_{x,\,y}$ is $2*10^{-4}$ m/day and vertical conductivity K_z is $5*10^{-5}$ m/day. Its total thickness ranges from 2 m to 10 m.

The 6th layer enfolds the Ordovician-Cambrian aquifer system represented by fine-grained sandstone and siltstone with a total thickness of 20 m. Its lateral conductivity is 2–4 m/day. This aquifer system provides water mostly for the oil shale cracking enterprise in Kiviõli. The abstraction from the Ordovician-Cambrian aquifer system was 367 m^3/day and from this 356 m^3/day was pumped out on the territory of the oil shale processing plant for technological purposes in 2007 (Perens R, Savva V 2006, 2008). The total amount of water allowed to abstract from the 6th model layer by Ministry of the Environment of Estonia is 1000 m^3/day in Kiviõli until 2020 (Perens R, Savva V 2008). Based on this regulation the pumping rate for the 6th model layer given for groundwater flow and transport modelling is 1000 m^3/day.

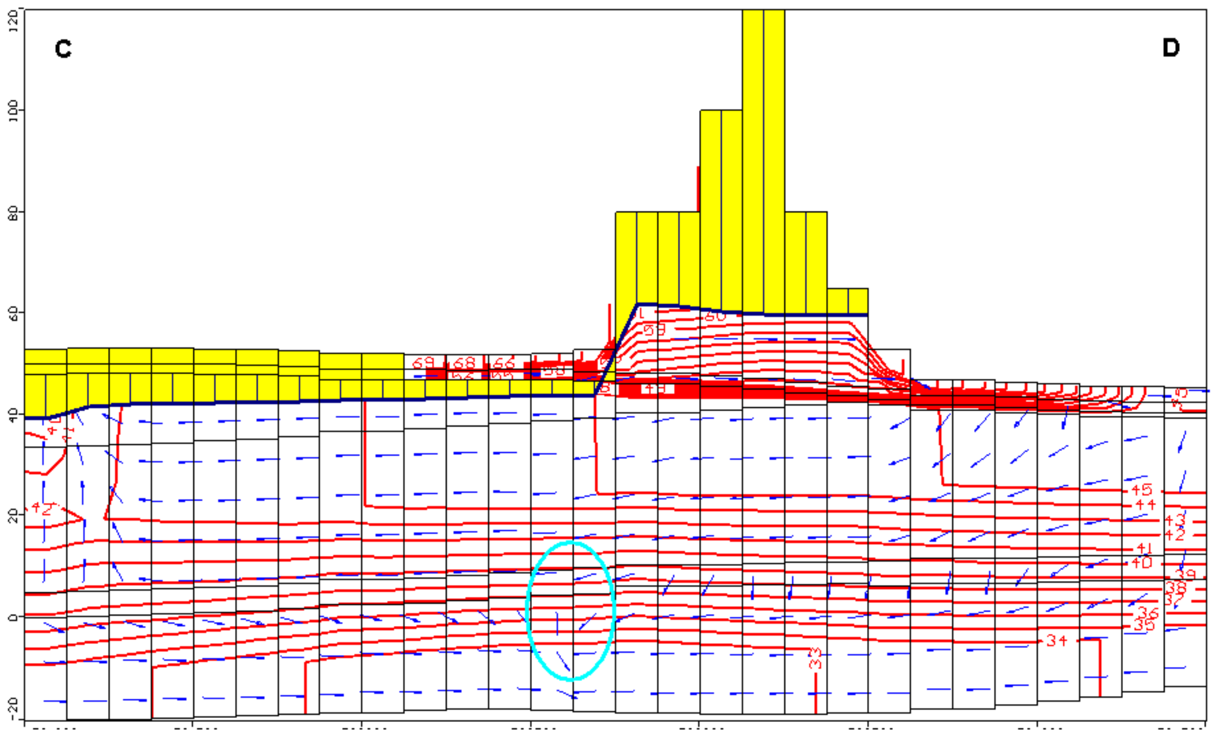

Fig. 3.5.2.5. Hydrogeological cross-section C–D in the Kiviõli area. Groundwater table – dark blue contour, vadose zone – yellow. Groundwater head – red isolines, m a.s.l. Groundwater flow directions – blue arrows. Intensive downward flux through the aquitard – the light blue oval. The distance between outer ticks on the X-axis is 700 m, absolute elevation on the Y-axis, m.

<u>Groundwater flow.</u> The total net infiltration into the topsoil given at the modelling is 2,800 m^3/day in the study area. About 460 m^3/day from it seep into the landfills. For that reason, the simulated topmost elevations of the groundwater table reach 55 m a.s.l. in southern portions of all three semi-coke heaps (Fig. 3.5.2.2). Thus, the thickness of the vadose (unsaturated) zone determined by modelling is 70–80 m in the central part of both larger landfills. In the southern portion of the small landfill, the thickness of the vadose zone does not exceed 10 m. In the 1st model layer, the water moves radially from landfills towards the surrounding ditches and mines abandoned. However, only 1,300 m^3/day of this water intrudes into the river network directly.

Groundwater particle tracking performed by means of the MODPATH code shows that most of the fluxes formed in semi-coke landfills move to west, north, or northeast at first. Their velocity is up to 25 m/year. A portion of the water flows out from the eastern big landfill toward the right-side branch of the Koljala Creek. The flow velocity is up to 50 m/year in this case. After moving by 250–750 m the lower branches of radial flows bend downward and intrude into the 3rd model layer (Fig. 3.5.2.3, 3.5.2.4, 3.5.2.5, 3.5.2.6). It is caused by a significant draining influence of 3rd layer. The latter is in one's turn intensively drained by the goaf of the exhausted mine. It forces the water move mostly from northwest to southeast in the 3rd layer. Because of a difference of heads, the water flows from overlying beds into the 3rd layer.

Due to the general trend of the water movement in the 3rd layer, the downward flows coming from overlying layers change their course sharply and start to move in a southeast direction toward the oil shale mine abandoned.

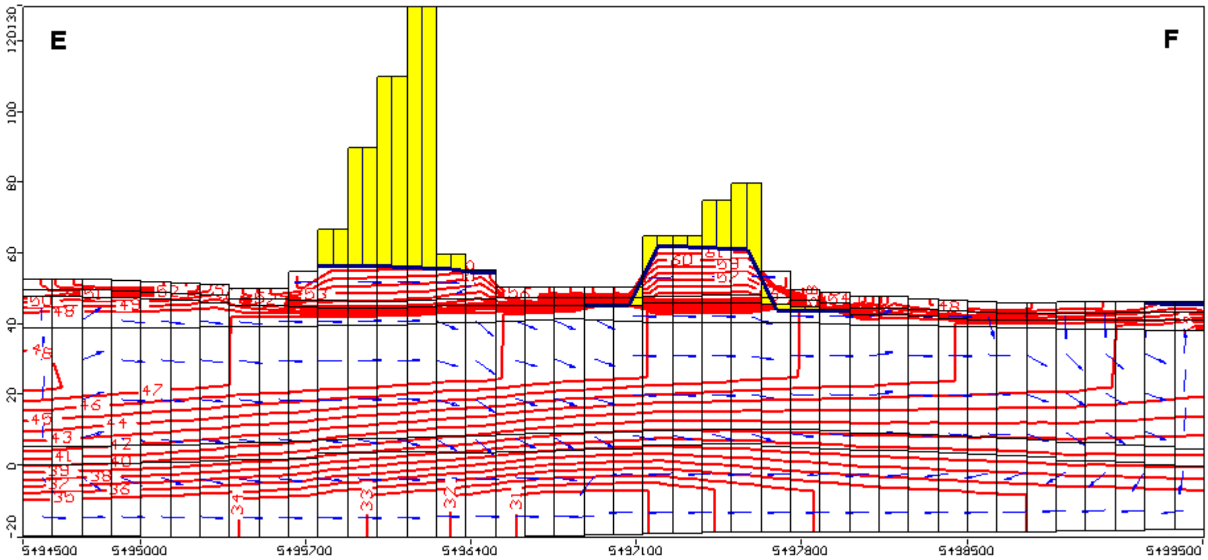

Fig. 3.5.2.6. Hydrogeological cross-section E–F in the Kiviõli area. Groundwater table: dark blue contour, vadose zone – yellow. Groundwater head – red isolines, m a. s. l. Groundwater flow directions – blue arrows. The distance between outer ticks on the X-axis is 700 m, absolute elevations on the Y-axis, m.

From the all water formed by infiltration, about 1,200 m^3/day flow downward through the 2nd model layer into the 3rd layer. This water joins with the general flow of the 3rd layer toward the mine abandoned at the south-eastern part of the site. The velocity of fluxes moving in southeast direction may achieve 150 m/year in the 3rd layer. The total inflow into the 3rd layer is 8,700 m^3/day. From it discharges into goafs of mines closed about 8,000 m^3/day. Due to an intensive draining mostly by abandoned mines, the 3rd layer is in confined-unconfined conditions. Its southern portion is partially dried up. Respectively, overlying 1st layer and 2nd layer have completely been dried up in the southern area of the study site.

The 4th model layer is recharged mainly through its lateral boundaries. The lateral inflow coming mostly from west and south-west is about 6,900 m^3/day. Furthermore, about 900 m^3/day of water intrudes from the 3rd layer into the underlying 4th layer. A relatively high conductivity of the 4th layer essentially favours the transport of pollutants.

The flow patterns in the 4th layer are altogether similar to them in the 3rd layer. The velocity of south-eastward fluxes reaches also up to 150 m/year.

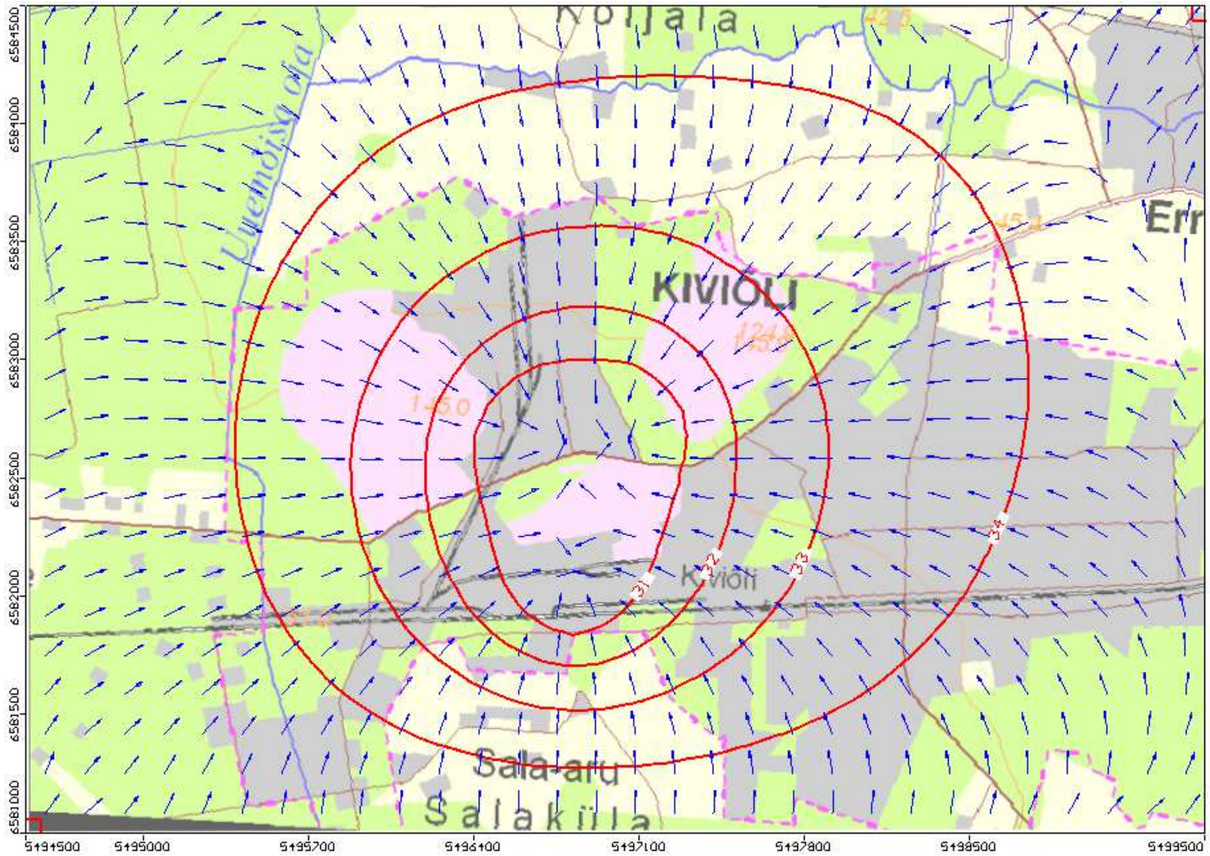

Fig. 3.5.2.7. Groundwater head in the 6th model layer in Kiviõli: red isolines m a.s.l. Groundwater flow directions - blue arrows. The distance between outer ticks is 700 m on the X-axis and 500 m on the Y-axis of the site map.

The total outflow from the 4th layer is about 6,800 m^3/day. Thereat, 240 m^3/day of water flows downward through the 5th layer (Silurian-Ordovician regional aquitard) into the 6th layer (Ordovician-Silurian aquifer system). Simulations performed clearly indicate a potentiality of such downward flux at a difference of heads between the 4th layer and 6th layer reaching some meters only (Fig. 3.5.2.5). The velocity of the downward flux is about 0.5 m/year. It means that the water contaminated may penetrate the Silurian-Ordovician regional aquitard in the transversal direction in course of 30–50 years if the value of the head gradient is close to 1 (taking into account a possible retardation). If the head gradient is major (at the more intensive pumping from the Ordovician-Cambrian aquifer system) then the velocity of the downward flux increases and the penetration time decreases proportionally.

The inflow into the 6th model layer through its lateral boundaries is less than 50 m^3/day at the pumping rate reaching 1,000 m^3/day. The abstraction given induces a concentric drawdown reaching up to 5–6 m in the 6th layer (Fig. 3.5.2.7). The lateral velocity of fluxes moving toward the centre of head depression is 5–20 m/year.

The calibration plot completed on the basis of simulation results is fully acceptable (Fig. 4.5.2.8). The total coefficient of correlation accounting observations of both, 1st and 6th layer, ranges 0.994 at the standard error of estimation equal to 0.41 m. The most of calibration points fall into the 95% confidence interval, all they are in the 95% interval.

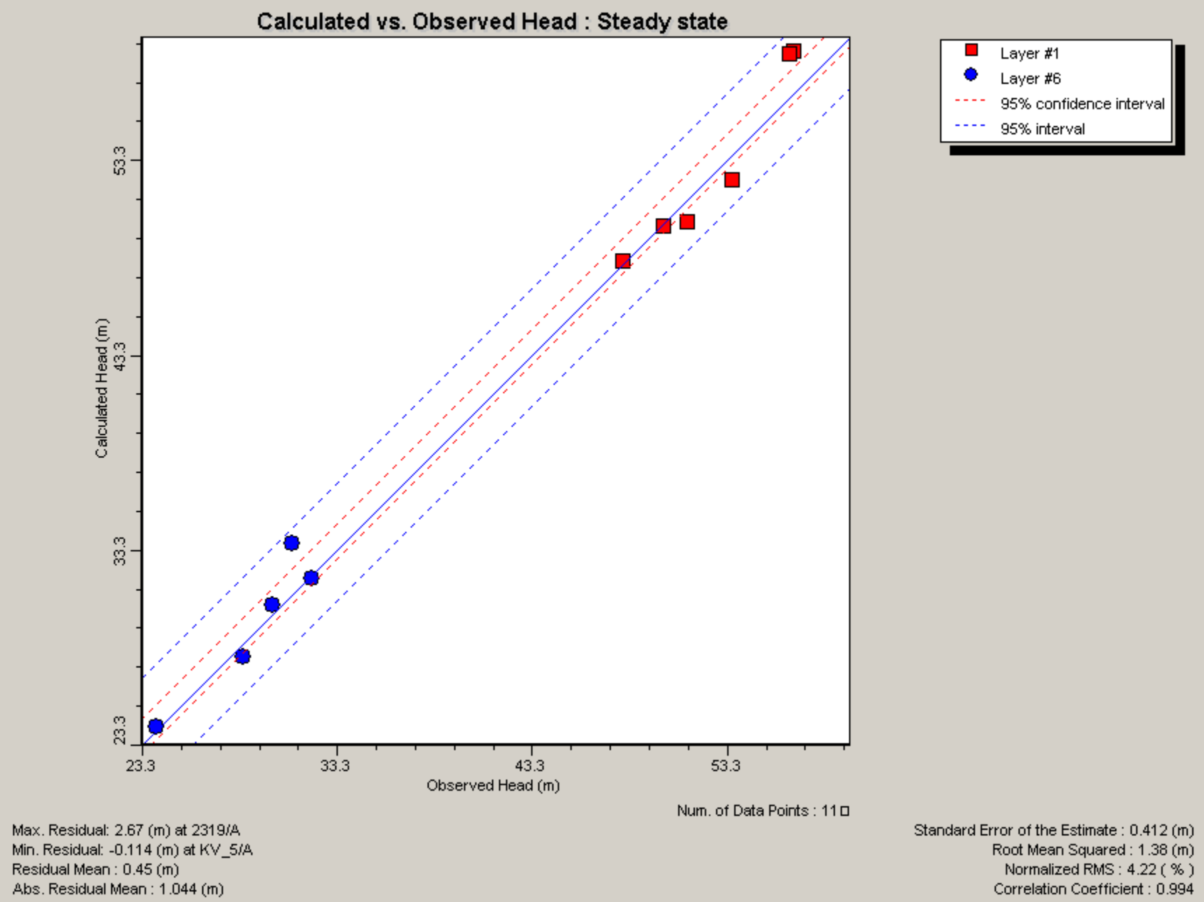

Fig. 3.5.2.8. Calibration graph of the Kiviõli flow model. Head in the 1st model layer - red quadrates, in the 6th layer – blue circles

<u>The transport of phenols</u> was again modelled by means of the engine MT3DMS. The boundary conditions of the transport model have established accordingly to existing sparse data. It was recorded in 2003 that in research borings RA-KV-1 and RA-KV-2 tapping the 3rd model layer the sum of phenols was 4.763 mg/L and 0.020 mg/L, respectively (Sørlie J-E. *et al.* 2004). The content of phenols was less than 0.0005 mg/L in borings RA-KV-3, RA-KV-4, RA-KV-5, RA-KV-6, and in borings opening the Ordovician-Cambrian aquifer system (Fig. 3.4.2.1). In a report of the AS Maves is noticed that the content of 5-Metylresorcin reached 2,190 mg/L in the boring PA-15, but the locality of this sampling point was not specified (Kiviõli Keemiatööstuse… 2007; Tööstusjäätmete ja… 2007). No additional data have at our disposal about the occurrence of phenols in groundwater in the Kiviõli area at present (Fenoolide seire...2007). Therefore, the data of wells RA-KV-1 and RA-KV-2 mentioned were posited to the model of phenols transport. The Constant Concentration boundary condition with the phenols content of 0.020 mg/L was

assigned to the 3rd model layer at the vicinity of the boring RA-KV-1 in the area of 36 ha and the phenols content of 2.0 mg/L was omitted to the locality of the boring RA-KV-2 in the area of 1.6 ha. The zero initial concentration of phenols was given to the remaining portion of the model. The values of other transport parameters (*D*, K_d, SP1, RC1, RC2,* α_L, α_H, and α_V) incorporated into the Kiviõli model were identical with the same parameters of the Kohtla-Järve model.

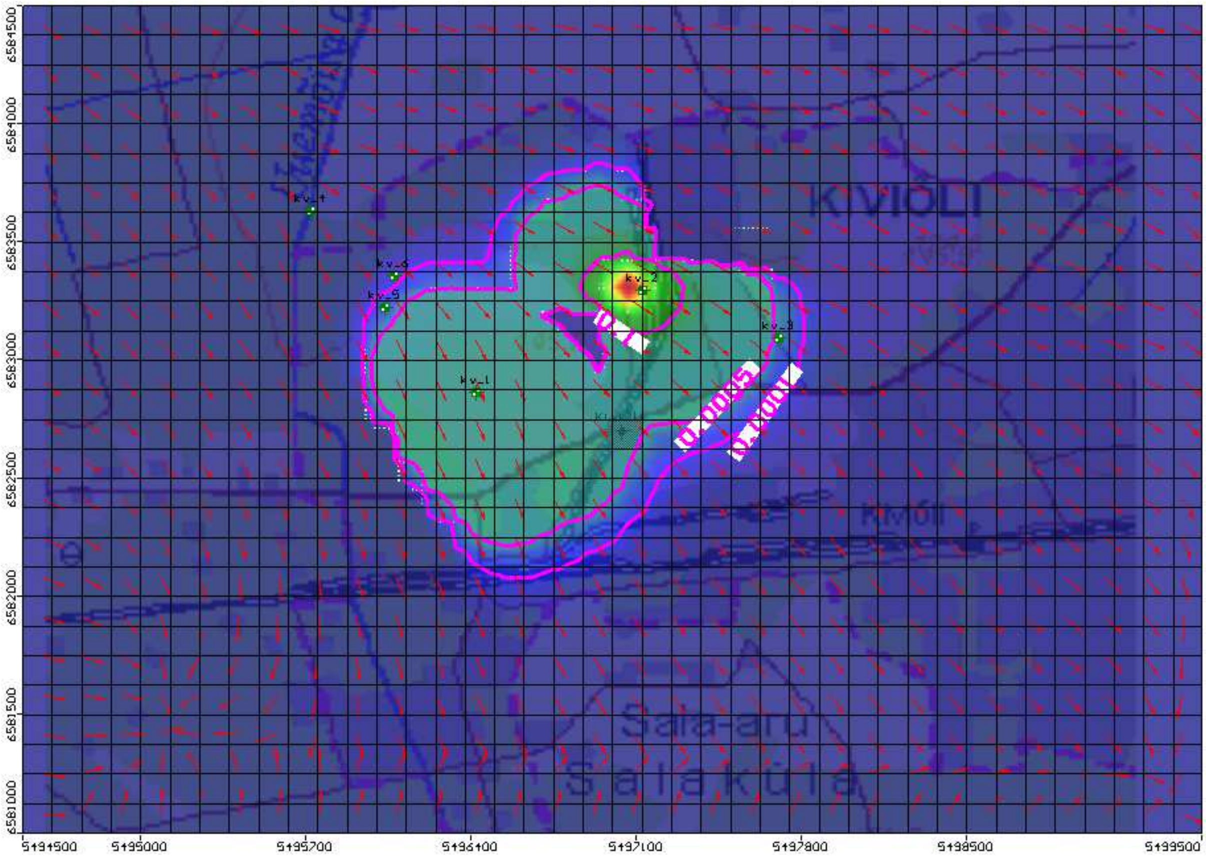

Fig. 3.5.2.9. Sum of phenols in the 4th model layer (S-O aquifer system) in the Kiviõli area in 2060. Concentration of phenols, mg/L: blue – 0.0–0.001, light blue – 0.001–0.005, green – 0.005–0.1, light green and brown – 0.1–2.0. Groundwater flow directions – red arrows.

Accordingly to simulations performed, the sum of phenols will range predominantly from 0.0001 mg/L to 0.1 mg/L in the confined portion of the O-S aquifer system beneath landfills in 2060 (Fig. 3.5.2.9). Only in close vicinity of the boring RA-KV-2, the content of phenols will reach 2.0 mg/L. It is very liable that the phenol contamination occurs also beneath the eastern landfill, but to simulate it the measured data lack. The phenols contamination up to 0.1 mg/L will intrude into the underlying O-Є aquifer system in 2060. If the intensity of the source of phenol contamination and pumping rate given to Ordovician-Cambrian aquifer system not alter hereafter then the content of phenols will increase up to 0.1 mg/l in this aquifer system in 2060 (Fig. 3.5.2.10) .

The adequacy of the Kiviõli model depends on the same circumstances analysed above at the Kohtla-Järve model. Unfortunately, because of the lack of a regular groundwater monitoring the source data are much poorer for the Kiviõli area. Therefore, the errors of simulation results are likely greater. In spite of exactness problems, the main conclusions drawn should be right in principle.

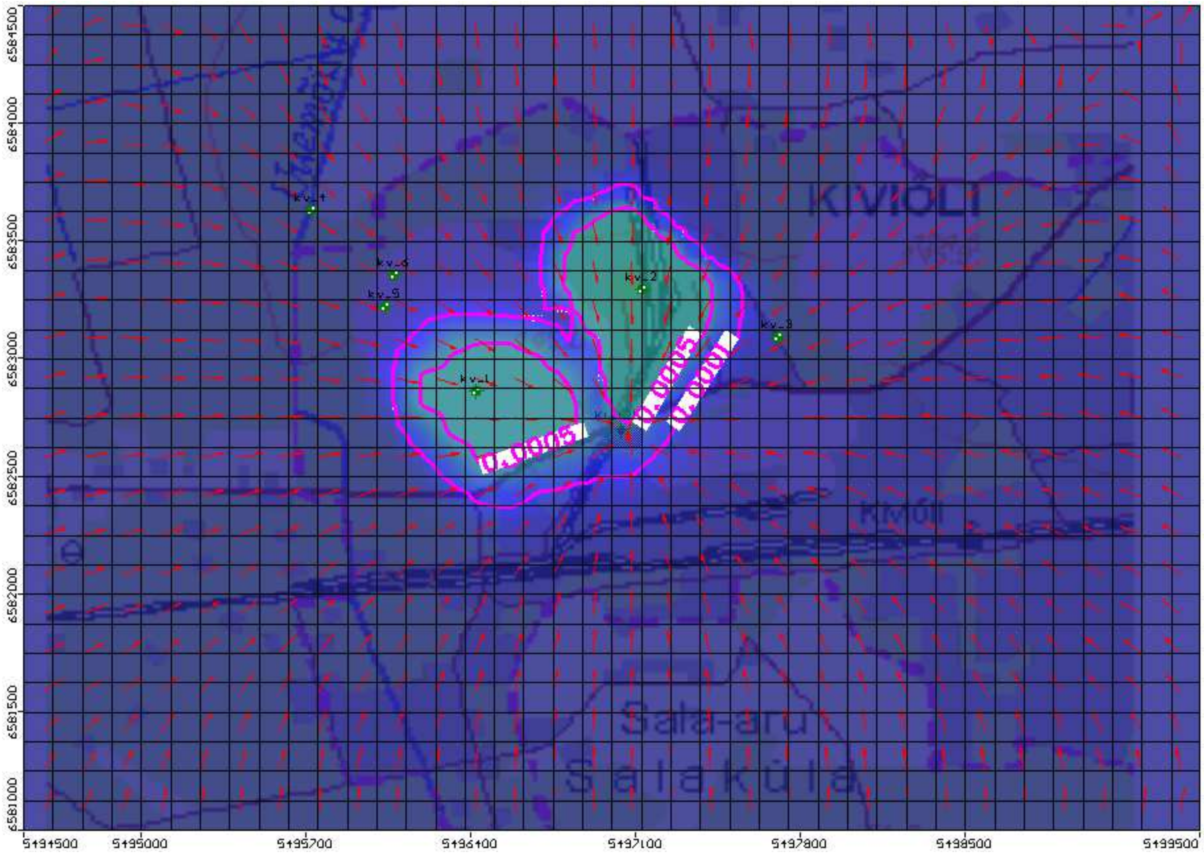

Fig. 3.5.2.10. Sum of phenols in the 6th model layer (O-Є aquifer system) in the Kiviõli area in 2060. Concentration of phenols, mg/L: blue – 0.0–0.001, light blue – 0.001–0.005, green – 0.005–0.1. Groundwater flow directions – red arrows.

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3.5.3. Narva model

Model area. The quadratic study site is situated southward of Narva on the left bank of the Narva Reservoir (Fig. 3.5.3.1). The Pulkovo 1942 co-ordinates of the model area are: the lower left corner – 5558000, 6578000; upper right corner – 5565000, 6585000. The area covers 49.0 km^2, the side length of the site is 7,000 m. The natural ground surface was flat with absolute elevations between 25 and 32 m. The main drainage base for surface water and shallow groundwater is Narva Reservoir bounding the study area from the southeast. In the central part of the model, area two rectangular plateaus are formed by oil shale ash of the Balti Power Plant stored. Sizes of plateaus are about 1.7 km × 2.2 km and the absolute elevations of their top surface reach from 30 m up to 70 m.

On the top surface of plateaus, large ponds have occurred. They were formed by water used for transporting ash. The ash plateaus are drained by surrounding channels. Two sedimentation basins with sizes of 0.6 km × 1.7 km bound the ash plateaus from the south (they have been marked as 'Roheline järv' on the site map).

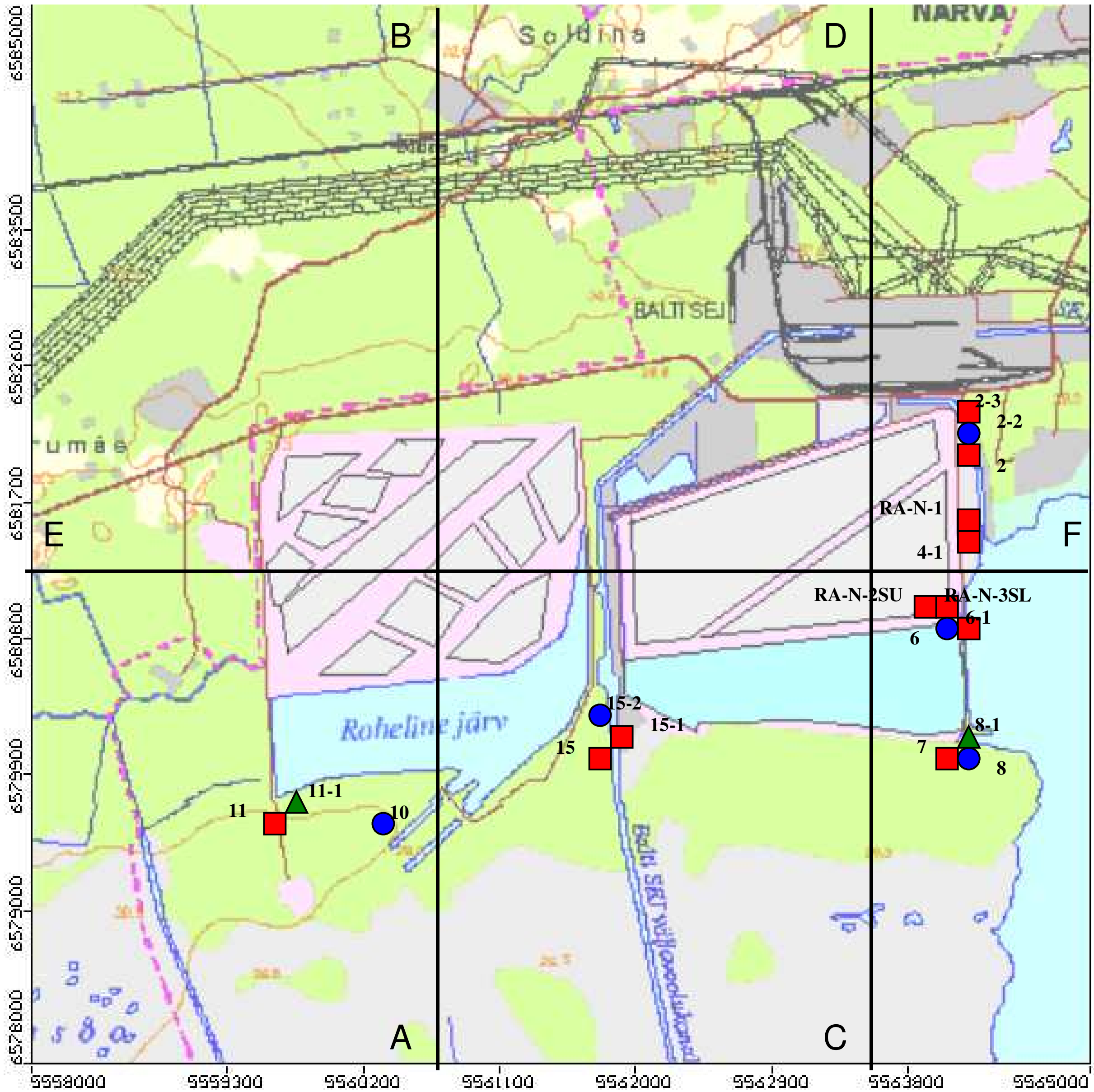

Fig.3.5.3.1. Modelling area with ash plateaus (surrounded by pink stripes) and head observation wells (red quadrates – 1st layer, blue circles – 2nd layer, green triangles – 4th layer). Hydrogeological cross-sections: A–B, C–D, E–F.

Dumping of ash on the eastern plateau has been ceased. This plateau is used for the arrangement of wind generators at present. The eastern plateau is still used for ash dumping by hydro transport whereat the water used is recycled and treated to decrease its high alkalinity. When the amount of water in the ponds and channels is too high, it is discharged after a purification treatment into the Narva Reservoir.

Model layers and boundary conditions. The 1st layer enfolds Quaternary deposits (Q), which consists predominantly of peat, glaciolacustrine sand, sandy loam (varved clay), and underlying till (Perens R, Vallner L 1997). The total thickness of these seams is 10–15 m (Fig. 3.5.3.2, 3.5.3.3). The prevailing conductivity value of sandy deposits does not exceed 1.5 m/day. However, the transversal conductivity of clayey Quaternary seams is mostly less than 0.01 m/day and, therefore, they form a semi-pervious confining bed for the underlying 2nd model layer. Inside the borders of ash plateaus, the 1st model layer comprises oil shale ash and the underlying Quaternary deposits that total thickness reaches up to 30 m on the western plateau and up to 50 m on the eastern one. The conductivity of this formation changes from 0.01 to 0.05 m/day. Water table conditions prevail in Quaternary deposits. The lower portion of ash storages and underlying Quaternary deposits is saturated by leachates of ash.

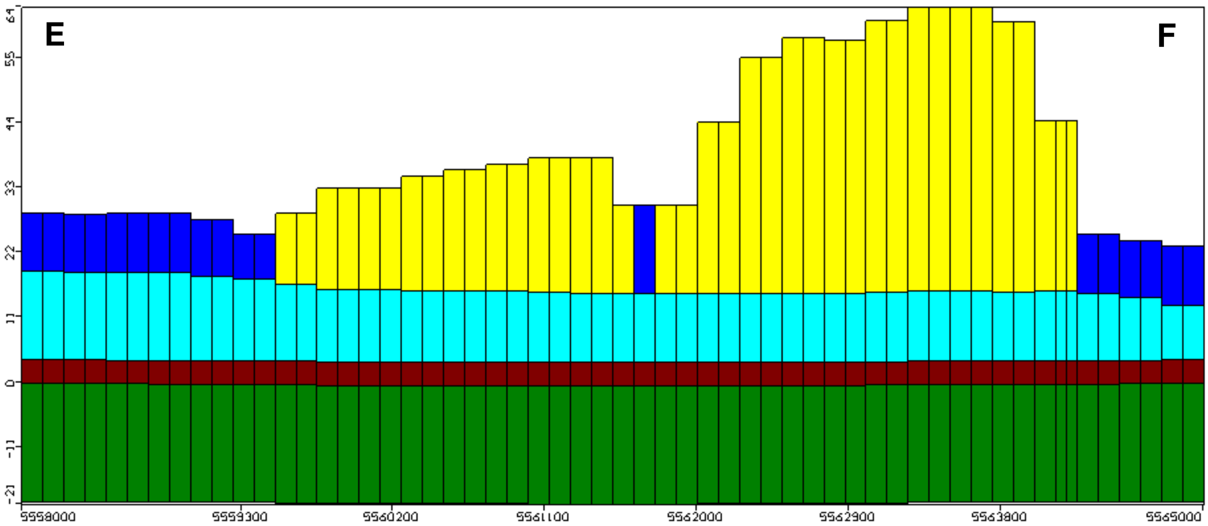

Fig. 3.5.3.2. Hydrogeological section E–F. Model 1st layer: blue – natural Quaternary deposits, yellow – oil shale ash with underlying Quaternary deposits; 2nd layer: light blue – Lasnamäe-Kunda aquifer; 3rd layer: maroon – Ordovician regional aquitard; 4th layer: green – Ordovician-Cambrian aquifer system. The distance between outer ticks is 900 m on the X-axis, absolute elevations on the Y-axis, m.

The River boundary condition has been assigned to the southern portion of layer border conforming to Narva Reservoir. The Constant Head conditions have been given to the remainder border portion of the 1st layer. In the areas of ash plateaus, the Constant Head boundary conditions have been assigned to the bonds mentioned above. The net infiltration given to the 1st unconfined model layer ranges mostly from 40 mm/year to 60 mm/year.

The 2nd model layer includes the semi-confined Lasnamäe-Kunda aquifer (O_2Ls-O_2Kn) represented by diverse Middle Ordovician limestones with clayey interbeds. Their total thickness ranges from 10 to 20 m. The limestones are fissured and heavily karstified. Therefore, they can easily become polluted. The lateral conductivity of this carbonate aquifer varies mostly from 3 to 10 m/day and the storage coefficient is between 10^{-6}–10^{-3} 1/day depending on the degree of fissuration and karstification. The transversal conductivity is 0.1 m/day or less. Carbonate bedrock layers are confined under the clayey Quaternarian deposits forming an overlying local aquitard. The water of the Lasnamäe-Kunda aquifer is often contaminated and not suitable for drinking in the study area (Viru alamvesikonna... 2006). The Constant Head boundary conditions have been established along borders of the 2nd model layer.

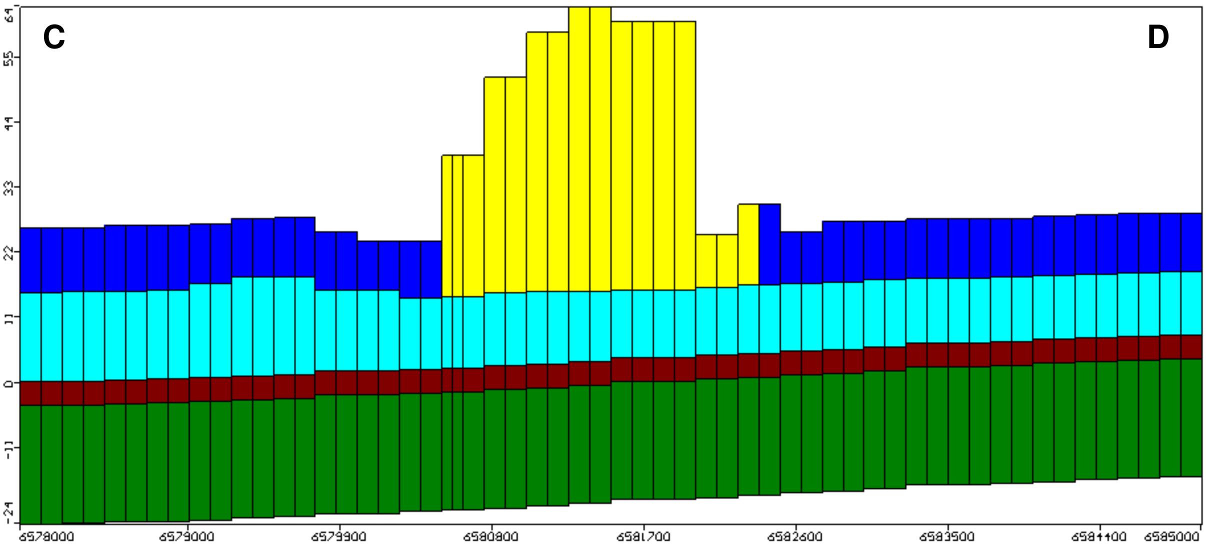

Fig. 3.5.3.3. Hydrogeological cross-section C–D. The legend in the Fig. 3.5.3.2.

The 3rd model layer enfolds the Silurian-Ordovician regional aquitard ($S\text{-}O_{aquitard}$) consisting of limestones, marls, siltstones, clays, and argillites with a total thickness of 2–4 m. Their transversal conductivity is 10^{-7}–10^{-5} m/day. This aquitard prevents spreading of polluted water from the $O_2Ls\text{-}O_2Kn$ aquifer downwards to some extents. The boundary conditions are not specified along outer borders of this layer.

The 4th model layer encompasses the confined Ordovician-Cambrian aquifer system (O-Є) represented by fine-grained sandstones and siltstones with a total thickness about 20 m. Their lateral conductivity changes mostly from 3 to 6 m/day. The well yields are predominantly 400—600 m^3/day per 10 to 15 m of drawdown. The storage coefficient is from 2.5×10^{-5} up to 6×10^{-3} 1/m. This aquifer system is a significant source for public water supply in Northeast Estonia. Due to the intensive water abstraction, regional head depressions have been formed with centres southward from Kohtla-Järve and in Slancy (Russian Federation). In the study area, water moves in southeast direction – toward Slancy. The Constant Head boundary conditions are omitted to the outer borders of the 4th layer.

Groundwater flow. A detailed groundwater budget of the study area was completed to obtain a clear quantitative conception about the structure of the groundwater flow. Therefore, the study area was divided into a number of budget zones. The inflow and outflow components were simulated for every budget zone.

The 1st model layer was divided as follows (Fig. 4.5.3.4): 2nd water budget zone (a zone number 1 was not specified) – eastern ash plateau; 3rd zone – western ash plateau; 4th zone – the relatively narrow area immediately surrounding ash plateaus; 5th zone – the area around the 4th zone.

To other model layers following budget zones were omitted (Fig. 4.5.3.5): 6th zone – 2nd layer; 7th zone – 3rd layer; 8th zone – 4th layer.

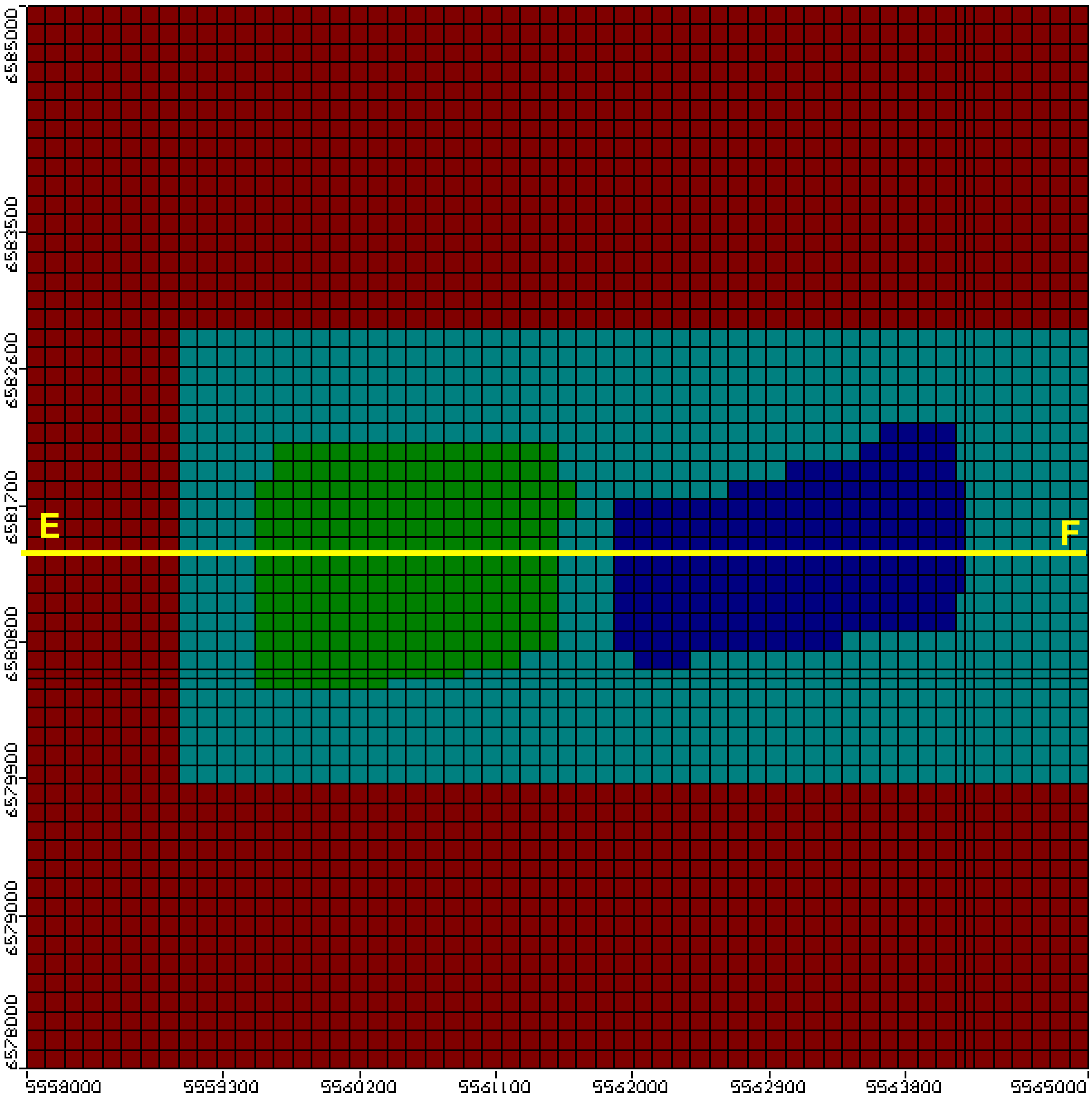

Fig. 3.5.3.4. Water budget zones of the 1st model layer. Zones: blue – 2nd zone, green – 3rd zone, greenish blue – 4th zone, maroon – 5th zone. E–F – hydrogeological cross-section.

The total outflow from the model domain simulated is equal to the inflow. The main outflow component, reaching 31,400 m^3/day, discharges to river network and directly to Narva reservoir. The total lateral outflow is about 2,000 m^3/day.

In ash storages, the water is formed mainly by the inflow from sedimentation bonds whereto the suspension of water and ash is pumped (Fig. 3.5.3.1). An additional recharge takes place at the cost of direct infiltration from precipitation reaching 800 m^3/day. Therefore, the topmost elevation of the groundwater table simulated reaches 60 m a.s.l. in the northeast portion of the eastern plateau (Fig. 3.5.3.6). Elevation of the groundwater table changes from 60 m to 26 m in the eastern plateau and from 30 m to 26 m in the western plateau. Thus, the thickness of the vadose (unsaturated) zone determined by modelling does mostly not exceed 10 m in ash landfills.

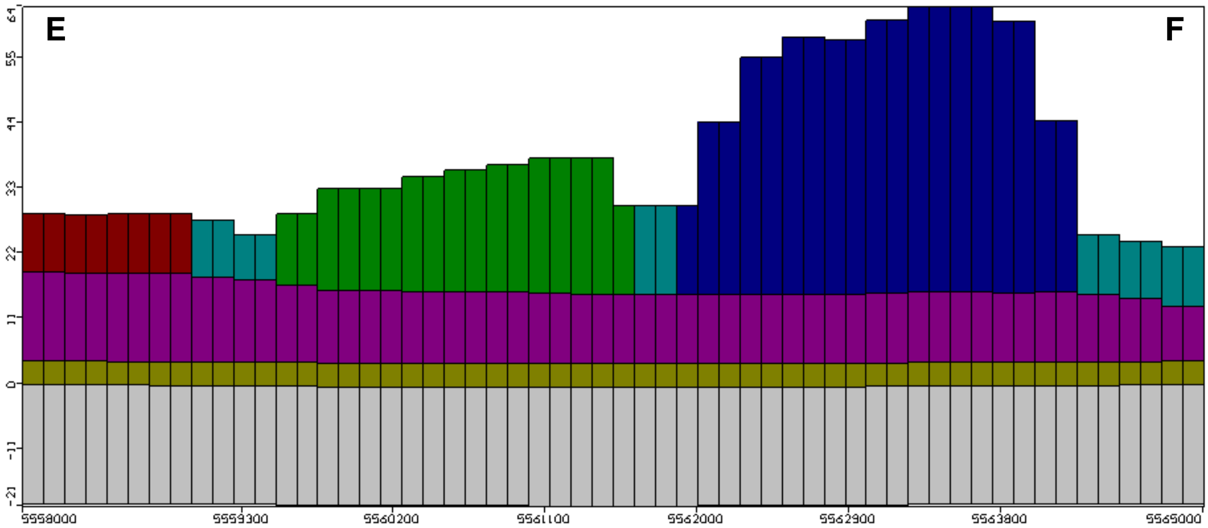

Fig. 3.5.3.5. Water budget zones of in the cross-section E–F. Zones: blue – 2nd zone, green – 3rd zone, greenish blue – 4th zone, maroon – 5th zone, violet – 6th zone (2nd model layer), olive – 7th zone (3rd layer), grey –8th zone (4th layer).

It is important to point out that the water forming in landfills is leaching out from the oil shale ash. The formed leachate contains various substances, which may be harmful to the environment. For instance, the ecotoxicological tests have been showed that some samples of laboratory ash leachates were very toxic. The pH of these leachates was measured to 12.9 (Sørlie J-E *et al.* 2004).

A portion of water stored in ash flows radially into the budget zone 4 surrounding the plateaus. This outflow from the plateaus amounts up to 2,400 m^3/day. Another portion of the outflow from the plateaus moving downward intrudes directly into the underlying carbonate bedrock (2nd model layer represented by 6th budget zone). This outflow component is about 7,200 m^3/day. Groundwater particle tracking shows that the velocity of the downward flow changes from 0.1 to 1 m/year and the velocity of the lateral outflow ranges from 2–5 m/year to 25–40 m/year in ash (Fig. 3.5.3.7, 3.5.3.8, 3.5.3.9).

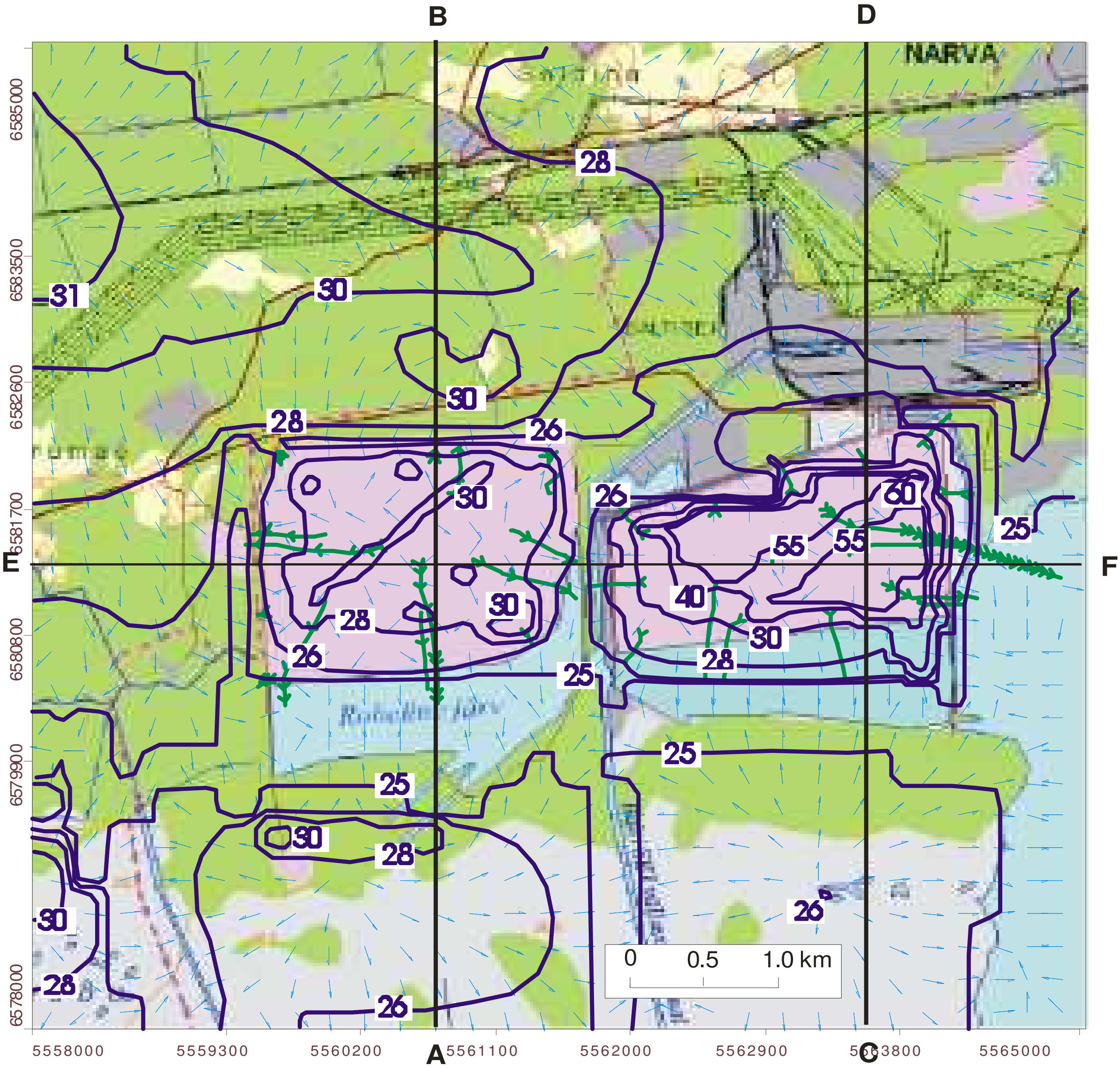

Fig. 3.5.3.6. Contours of the groundwater table in the 1st layer of the Narva model, m a.s.l – dark blue isolines. Groundwater flow directions – blue arrows. Flow pathlines – green. The distance between markers (arrowheads on pathlines) is equal to 10 years of the groundwater movement.

The budget zone 4 of the 1st model layer surrounding the landfills is recharged by the direct outflow from the ash plateaus which reaches 2,400 m^3/day, but its main inflow amounting 12,700 m^3/day comes from the underlying 2nd model layer. This flow component has formed in a central portion of ash storages and after a prior downward seeping into the 2nd layer, it bends upward intruding into the overlying 4th budget zone (Fig. 3.5.3.7, 3.5.3.8, 3.5.3.9). The chief outflow from the 4th budget zone, reaching 16,200 m^3/day, discharges in Roheline järv and Narva Reservoir. This is the principal groundwater flow carrying the leachates out from ash storages into surface water bodies.

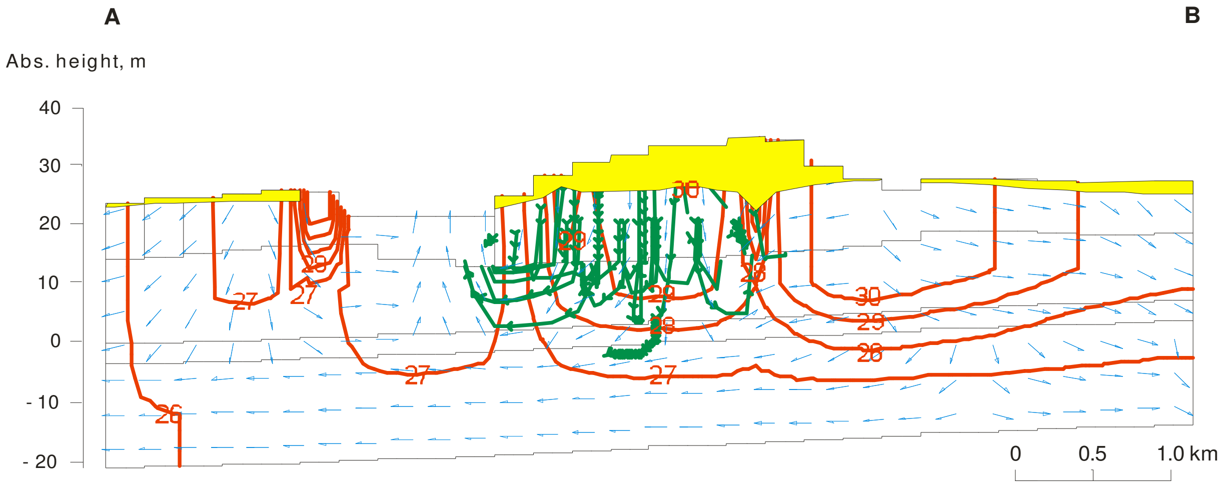

Fig. 3.5.3.7. Hydrogeological cross-section A–B. Vadose zone – yellow. Groundwater head, m a.s.l. – red isolines, groundwater flow directions – blue arrows. Flow pathlines – green. The distance between markers (arrowheads on pathlines) is equal to 10 years of the groundwater movement.

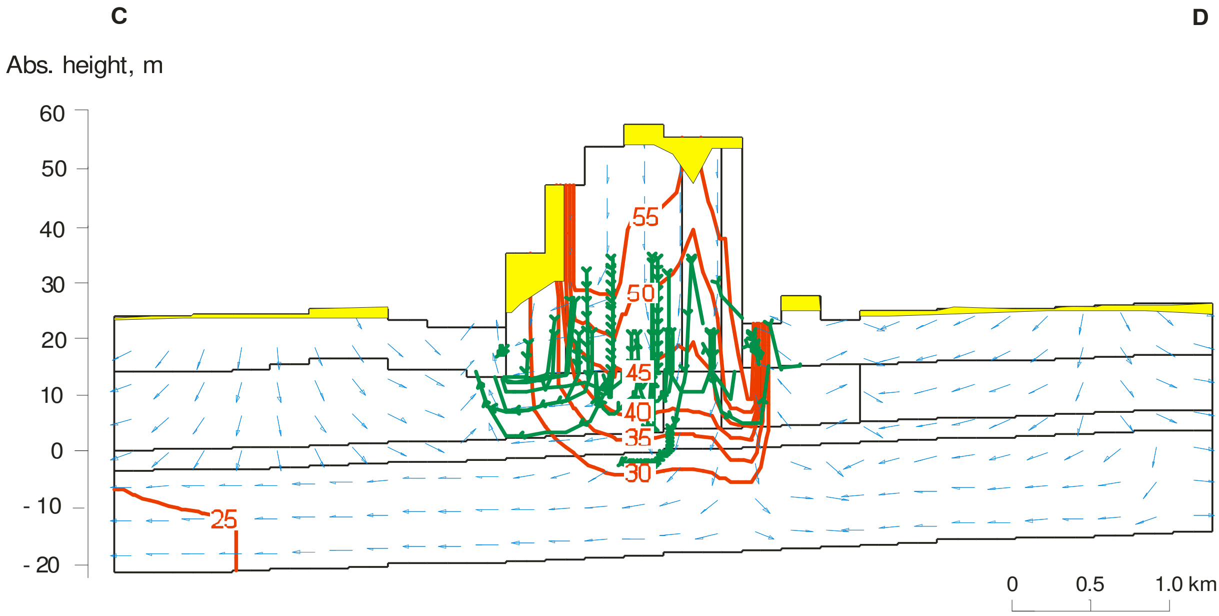

Fig. 3.5.3.8. Hydrogeological cross-section C–D. The legend in the Fig. 3.5.3.7.

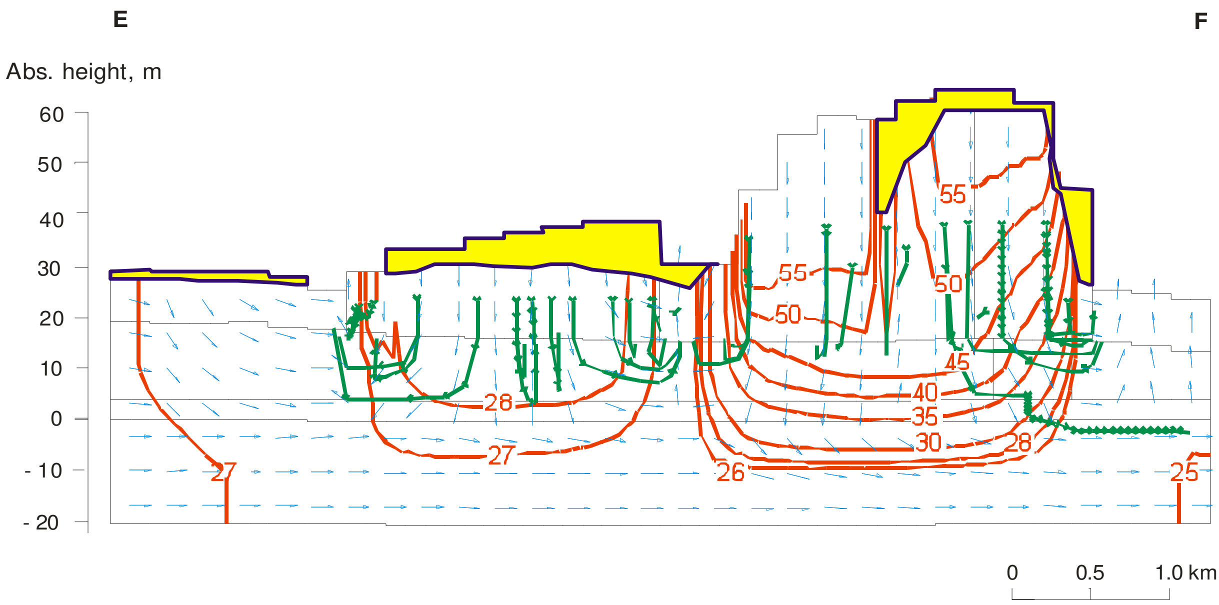

Fig. 3.5.3.9. Hydrogeological cross-section E–F. The legend in the Fig. 3.5.3.7.

The remainder portion of the 1st model layer which belongs to 5th water budget zone is recharged by direct infiltration equal to 3,500 m^3/day, but the main inflow amounting 14,200 m^3/day comes from the underlying 2nd model layer. Such comparatively significant rising flow is caused by the locality of the study area on the shore of Narva Reservoir having a mighty drainage capability. Here the regional deep groundwater flows bend upward and intrude in large surface water bodies. The total outflow from budget zone 5 into the hydrological network is about 15,200 m^3/day. In the northwest part of the modelling site, a downward flux exists from 1st layer into the 2nd layer reaching approximately 3,700 m^3/day.

The 2nd model layer representing the Lasnamäe-Kunda aquifer of the Ordovician carbonate bedrock is recharged by the downward flow coming from the overlying 1st model layer and amounting 8,500 m^3/day (Fig. 4.5.3.10). About 57% of this inflow originates from the eastern ash plateau. The lateral inflow into the 2nd layer, mainly through its western borders, is circa 16,300 m^3/day. The total inflow into the 2nd model layer reaches 28,500 m^3/day.

Due to the intensive downward inflow from the 1st model layer, the groundwater head has significantly increased beneath the eastern landfill in the model 2nd layer. Over there, heads range from 28 to 60 m a. s. l. Under the western ash plateau, the heads are between 26–30 m a. s. l. In the 2nd layer beneath the ash plateaus, groundwater moves laterally in radial directions at velocities mostly from 4 to 40 m/year. As it was pointed above, this flow is mostly bending upward to intrude into the overlying 4th budget zone around of ash landfills.

The main outflow from the 2nd model layer reaches 12,700 m^3/day and tends upward into the 1st model layer in the area of sedimentation ponds (Roheline järv) and Narva Reservoir. The downward flow into the 3rd model layer (the Silurian-Ordovician regional aquitard) is relatively small – approximately 200 m^3/day. The total outflow from the 2nd model layer, including the flow through its lateral borders, is about 28,500 m^3/day.

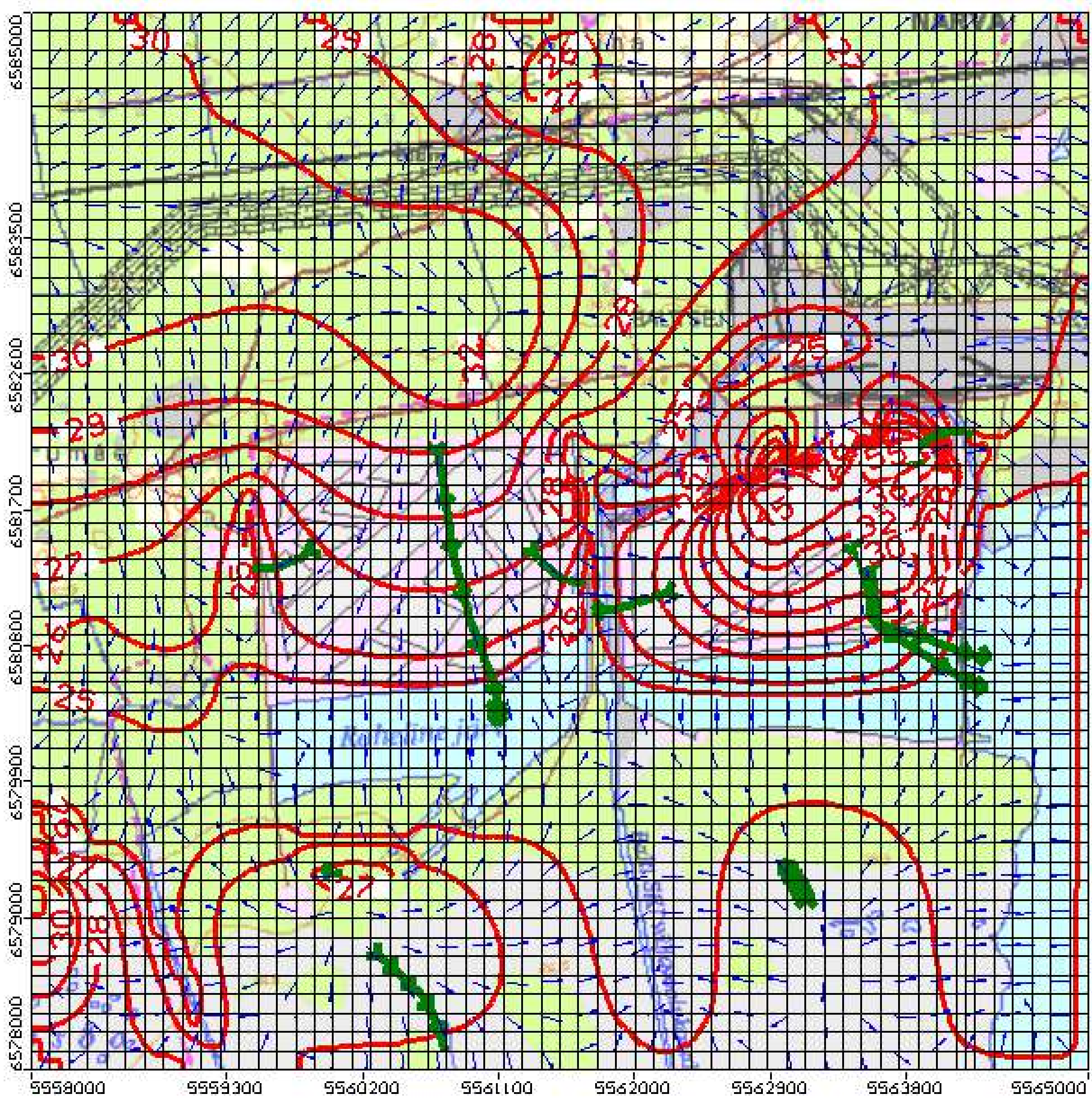

Fig 3.5.3.10. Contours of the groundwater head in the 2nd model layer, m a.s.l – red isolines. The direction of the groundwater flow - blue arrows. Flow pathlines – green. The distance between markers (arrowheads on pathlines) is equal to 10 years of the groundwater movement.

The duration of moving of groundwater particles from sedimentation ponds on the surface of ash plateaus to the draining network surrounding the plateaus range from 10 to 200 years. The shorter moving times occur when particles penetrate the upper ash seems on margins of landfills. If particles must penetrate the whole thickness of ash, underlying the landfill Quaternary deposits, and carbonate bedrock of the 2nd model layer, then their moving time will be remarkable longer. It is necessary to point out that the real velocity of chemicals is significantly lower than the velocity of water because of retardation.

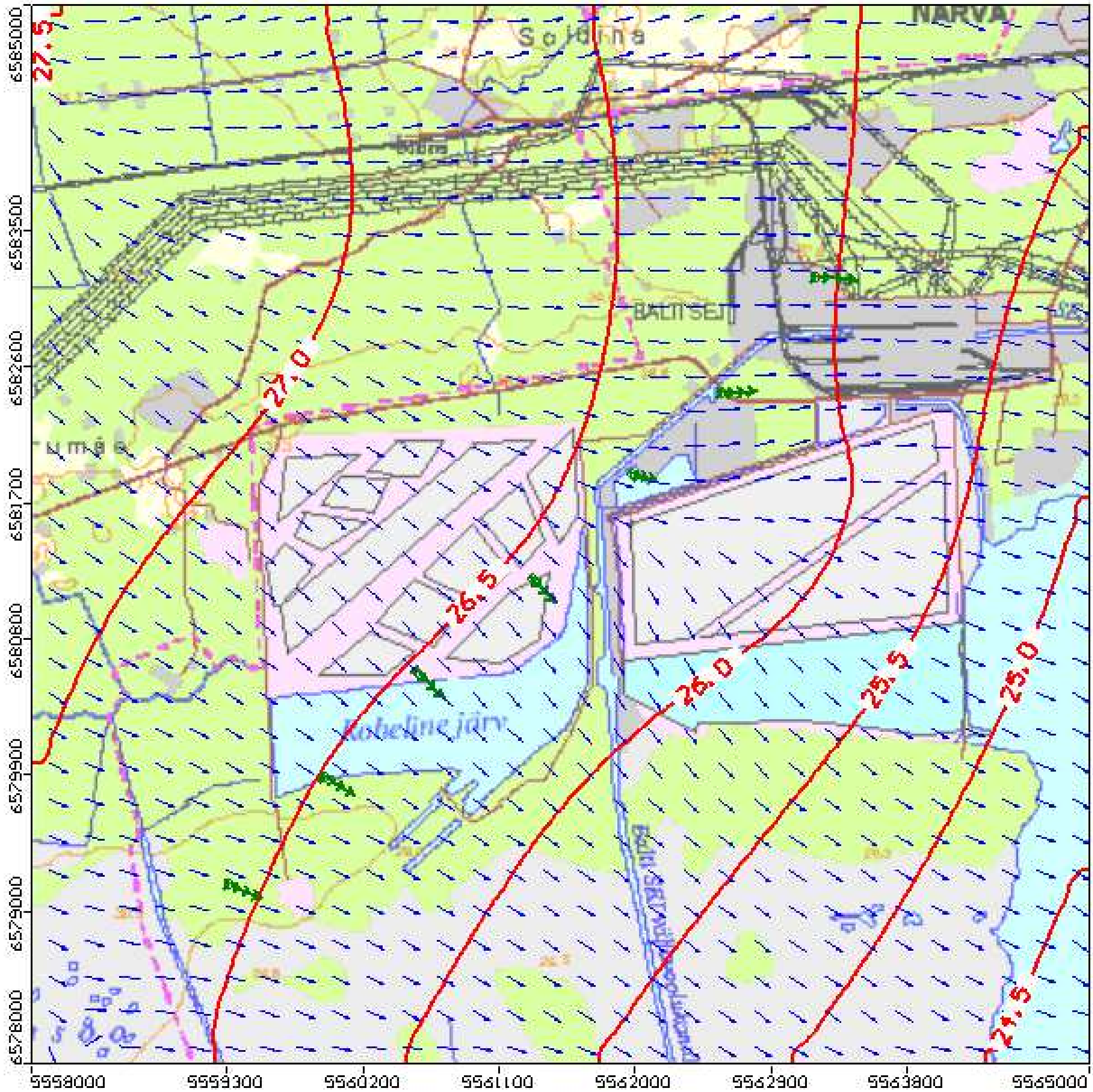

Fig. 3.5.3.11. Contours of the groundwater head in the 4th model layer, m a.s.l. – red isolines. The direction of the groundwater flow - blue arrows. Flow pathlines – green. The distance between markers (arrowheads on pathlines) is equal to 10 years of the groundwater movement.

In the 3rd model layer enfolding the Lower Ordovician regional aquitard, a downward flow prevails. It comes from the overlying Lasnamäe-Kunda aquifer and goes into the underlying 4th model layer (the Ordovician-Cambrian aquifer system). Due to the very low transversal conductivity of the regional aquitard, the downward flow is approximately 200 m^3/day, only. The time needed water particles to penetrate the regional aquitard in the transversal direction ranges from 70 to 100 years (Fig. 3.5.3.7, 3.5.3.8, 3.5.3.9). The flow velocity is 0.04–0.06 m/year. Consequently, leachates of ash plateaus have not reached the Ordovician-Cambrian aquifer system yet.

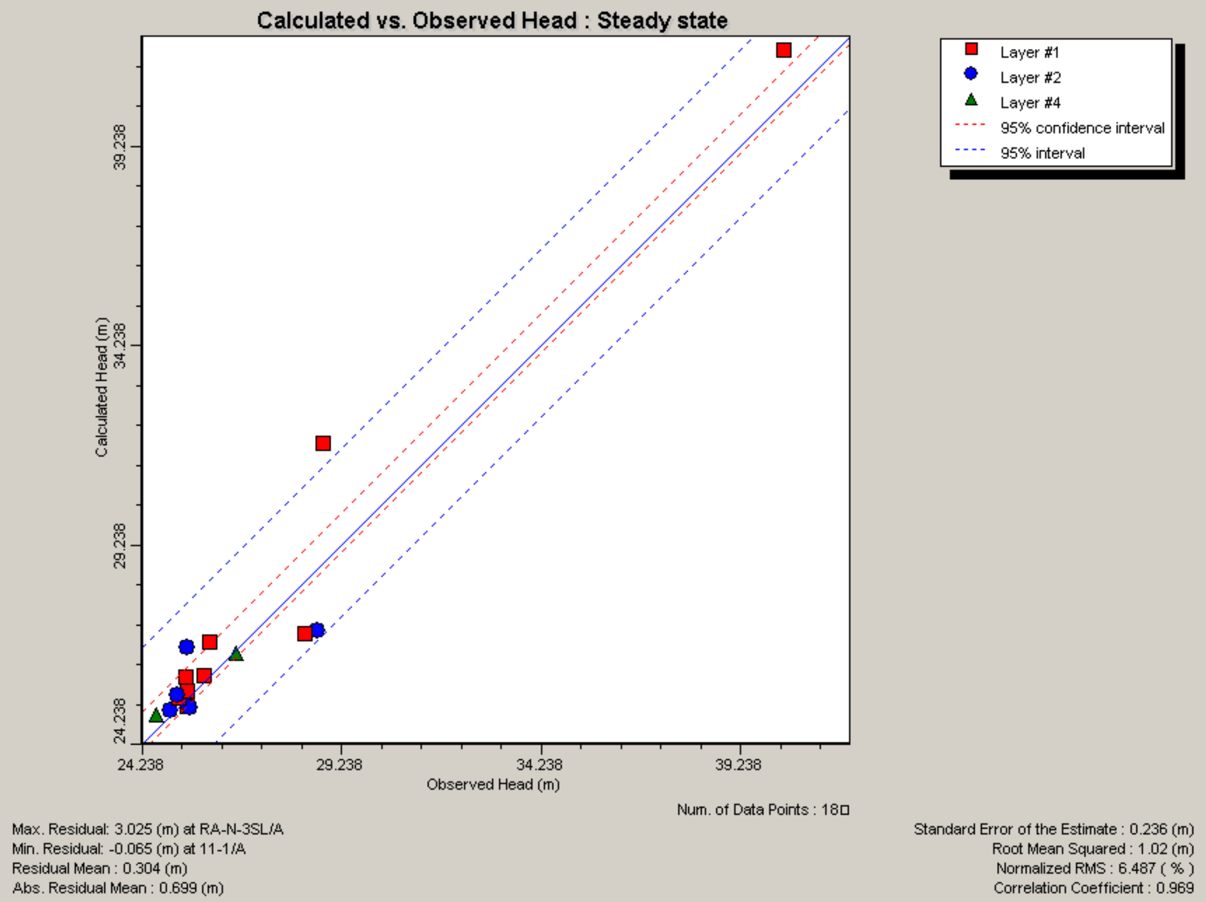

Fig. 3.5.3.12. Calibration graph of the Narva flow model. Head observation points: red quadrates – 1st layer, blue circles – 2nd layer, green triangles – 4th layer

The downward flow from above amounting 200 m^3/day is the main recharge source of the Ordovician-Cambrian aquifer system. The lateral inflow through its western border does not exceed 100 m^3/day. In the Ordovician-Cambrian aquifer system, the water moves mainly in southeast direction – towards the mighty intake in Slancy, Russian Federation (Fig. 3.5.3.11). The lateral outflow through the eastern and southern borders of the 4th model layer is about 300 m^3/day. The heads decrease from 27.5 m a. s. l. in the northwest corner of the study area to 24.5 m in the southeast corner.

In the area of ash plateaus, the groundwater levels were episodically recorded at different times from 1994 until 2006. The arithmetical mean of levels was used for model calibration. The calibration plot completed on the basis of simulation results is quite acceptable (Fig. 3.5.3.12). The total coefficient of correlation accounting the data of observation wells tapping 1st, 2nd, and 4th layer, reaches 0.964 at the standard error of estimation equal to 0.24 m. In general, the most of calibration points are in the 95% interval.

The groundwater samples taken from the observation wells surrounding the ash landfills were not contaminated and the outflow from ash storages was not expected to cause any environmental adverse effects in 2003 (Sørlie JE *et al.* 2004).

Furthermore, the mean annual discharge of groundwater from the model layers in the river network of the Narva Reservoir simulated is approximately 32,000 m^3/day or 0.37 m^3/s. This quantity of water takes only 0.1% of the mean annual runoff of the Narva River amounting 394 m^3/s at Narva (Resursy poverhnostnyh…1972). Therefore, the groundwater outflow from ash landfills due to its relatively inconsiderable quantity and acceptable chemical quality is practically not harmful to the river network of Narva Reservoir at present.

4. SUBSEQUENT INVESTIGATION OF LANDFILLS

4.1. Efficiency assessment of landfill remediation

An additional continuous investigation of the Kohtla-Järve landfill and Kiviõli landfill as pollution sources is relevant in connection with their remediation projects.

Accordingly to a convention between European Commission and Estonian Republic the landfills of East-Viru County must meet the requirements of the EU Landfill Directive at the latest in 2013 (Töötusjäätmete ja… 2007). To fulfil this task (2003/EE/16/P/PA/012) the projects of partial closing up the Kohtla-Järve landfill and the Kiviõli landfill were completed by enterprises AS Maves, EL Konsult OÜ, IPT Projektijuhtimine OÜ, and Ramboll Finland OY (Closing down… 2006; Kiviõli Keemiatööstuse… 2007; Töötusjäätmete ja… 2007).

The eastern portion of the Kohtla-Järve semi-coke landfill (Fig. 4.1.1) and the south-eastern portion of the western Kiviõli landfill (Fig. 4.1.2) have been determined to close up. The dumping of semi-coke will be ceased and existing great asperities of the ground surface will be smoothed in those areas. To prevent the leaching of harmful substances from semi-coke the smoothed surface of the semi-coke dumped will be covered with a screening layer. The latter must consist of fresh semi-coke or clay or loam with a total thickness of 0.5 m. The vertical hydraulic conductivity of this semi-confining layer should be decreased to 10^{-8} m/s ($\approx 10^{-3}$ m/day) by means of mechanical compression (Töötusjäätmete ja… 2007).

A more or less waterproof vertical barrier coupled with a drainage system shall be arranged into the loose deposits around the Kohtla-Järve landfill. This barrier and drainage system with the total length of about 7 km should catch the surface run-off and upper underground fluxes of contaminated water from the landfill. The water collected will be made harmless by a special treating. Dumping of fresh semi-coke is continued in the western portion of the Kohtla-Järve landfill and in the north-western portion of the Kivõli landfill.

The full cost of remediation of the Kohtla-Järve landfill and Kiviõli landfill including the costs of project designing, construction, supervisory etc. amounts up to 106 million of Euros (Kiviõli Keemiatööstuse… 2007; Tööstusjäätmete ja… 2007). The fulfilment of the remediation project was already started in 2011.

Fig. 4.1.1. Orthophoto of the Kohtla-Järve landfill (the area of the ash landfill is light grey). The portion of the landfill determined for closing up (92 ha) has been surrounded by a red contour in the figure (Tööstusjäätmete ja…2007).

In spite of the extremely expensive remediation project described its scientific argumentation is poor. A correct hydrogeological analysis of forming and spatial transport of contaminated groundwater in areas of landfills lacks in project documentation (Metsur M 2005; Tööstusjäätmete ja… 2007). It is not sufficiently proved whether the covering layer designed can significantly impede infiltration of rain and thaw water into the landfill in real-world conditions. The development of outer contours and inner concentrations of the contamination plum under the influence of measures designed have not been determined in the space-time. Therefore, it is not possible quantitatively to appraise the efficiency of the remediation planned. The project performers summarize the expected impact of project measures on the water environment by three sentences only as follows: "In result of fulfilling the project the quality of surface water and groundwater will improve. Primarily the quality of surface water will improve. A longer time will take improving the groundwater state" (Tööstusjäätmete ja…2007, p. 64).

The risk of mechanical smoothing of landfill asperities by mighty earth-moving machines has not been analyzed and assessed adequately. Earthworks may open the access of air oxygen to kerogen buried causing prolonged landfill burnings. Variable hazardous gazes will be emitted from burning places where the temperature may reach up to 1000°C. The management of landfill burnings is very bothersome and expensive (Adeolu O, Otitoloju A 2012; Landfill fires…2002; Landfill guidelines…1997).

Fig. 4.1.2. Orthophoto of the Kivõli western landfill. The portion of the landfill determined for closing up (17 ha) has been surrounded by a red contour (Tööstusjäätmete ja…2007).

From the viewpoint of taxpayers and state budget management, it is essential to estimate the environmental and financial virtue of the remediation project of landfills. For that purpose, the measuring criterions should be established at first. It means that the certain observation points and water characteristics should be determined by an objective monitoring of water quality. The remediation targets for these points are necessary to set up. Then a comparison of values of remediation targets with characteristics of the current or predicted water state will measure the real efficiency of remediation works planned or performed.

For instance, let us suppose that the content of a harmful chemical in water of a monitoring point is 10 mg/L at present. The target of remediation would be to decrease this concentration to a tolerable value of 1 mg/L for a certain date, say, for 2030. It should be possible correctly to predict: is achieving this target feasible for the date fixed. On the other hand, if fulfilling of this task will cost, say, 9 million of Euro, then the decreasing of the concentration of the contaminant considered by 1 mg/L will cost one million of Euros. Hence, a question can arise – is this remediation action economically rational or is it a misuse of funds? To date, such as analyses of remediation projects of Kohtla-Järve landfill and Kiviõli landfill lack.

The current state of groundwater can be assessed based on data of the existing monitoring network indeed. If monitoring data are not sufficient, then methods of sampling or analysis can be improved, or additional monitoring points set up. However, the real adequacy of the monitoring network can be evaluated only by a profound groundwater flow and transport modelling. It makes feasible to check: does the disposition of monitoring points give a dependable imagination about the spatial development of the contaminant plum. Without such of modelling, it is not possible to establish the remediation targets and correctly predict the changing of the content of contaminants in water. In return, the tempolabile concentration of contaminants depending on remediation measures can be simulated by a tried and true model. It allows to compare the real and expected (calculated) contaminant concentration after a reasonable time and to control the efficiency of the remediation process.

The Kohtla-Järve and Kiviõli models described in the previous chapter can be used for solving remediation problems. For that purpose, the 1st layer of these models must be refined to thinner seams reflecting the filtrational heterogeneity of semi-coke. The hydraulic conductivity designed for the capping cover of landfills should be omitted to uppermost additional seams. Cauchy boundary conditions and the Wall boundary condition of the Visual MODFLOW code reflecting the impact of the planned drain system must be set up around landfills modelled. An imaginary optimum placing of monitoring points should be designed. It is necessary to modify input parameters to establish the sensibility of models. The need for additional monitoring points and for specification of model input parameters should be based on the model sensibility analysis.

The data adjusted should be incorporated into models after performing of supplementary experiments and monitoring. Then the remediation targets for variable times assigned can be determined by modelling. It allows a posterior check of the efficiency of landfills remediation projects started already and plan their corrections.

4.2. Groundwater monitoring and determination of transport parameters

The landfill models are necessary not only for the Kohtla-Järve and Kiviõli, but they can also be useful for managing the landfill sites in Kukruse, Sinivoore and other dumping places of residuals of the oil shale industry. Despite an apparent stabilization of the transport of harmful substances in the groundwater environment of landfills, however, it is necessary to be on the alert. It refers especially to the spreading of contaminants in the O-Є aquifer system and eventual intrusions into the Є-V aquifer system in Kohtla-Järve. Therefore, an elaboration of the existing groundwater monitoring network and the establishment of some new observation systems are desirable in the East-Viru County.

In this connection, an integrated computerized database containing all information about monitoring of landfill areas should be completed at first. Constructions of observation wells, records of groundwater heads, results of water analyses, co-ordinates, and absolute elevations of monitoring points should be revised and concentrated. Elevations must be certainly determined by geodetic levelling or airborne LIDAR system.

Unfortunately, the existing groundwater monitoring system in the landfill areas does mostly not enable a profound study of the contaminant transport (Crane PE, Silliman SE 2009). The placement of monitoring points and objects is not rational. Sampling intervals are often too long or random and an assortment of water characteristics measured or chemically analysed insufficiently considered.

There are no monitoring borings in highest parts of landfills currently. It aggravates a correct completion of landfill models because of lack of data about extreme elevations of the groundwater table. For that reason, it is necessary to install supplementary observation borings in parts of landfills where the highest hydraulic pressures occur. The observation borings should be placed on the hydrogeological sections oriented along the main directions of groundwater flow from landfills toward the surrounding lowland.

The detached groups of observation borings should be assembled. The assembled observation wells should tap separately the upper, middle, and the lower portion of main hydrogeological units in monitoring spots. It renders possible to study the changing of water characteristics or displacement of chemicals in the vertical direction with a sufficient validity. It is especially pertinent for investigation of the preferential flow in the vadose zone of landfills.

The distances between groups of observation wells assembled on the hydrogeological cross-sections must not exceed 100 m. The outermost monitoring points on cross-sections should reach the most far outer contours of the contaminant plum predicted in both horizontal direction and vertical direction. It will give a possibility to check achieving the remediation targets established.

The frequency of the groundwater head measurement in borings and their sampling should not exceed 1–2 months. A longer interval between observations will hinder the study of biodegradation of contaminants. The observation series should last a couple years without interruptions. Shorter measuring and sampling intervals can be required for investigation of transport processes occurring in the vadose zone of landfills (Williams J. *et al*. 1998). Furthermore, it is recommendable to install a lysimeter or a couple of lysimeters into the semi-coke of landfills to a correct determination of the total precipitation and net infiltration.

The disposition of observation borings, vertical intervals of water sampling, and frequency of sampling recommended making feasible to calculate the values of the hydrodynamic dispersivity, longitudinal dispersivity, and vertical dispersivity on the ground of monitoring data.

The assortment of water ingredients analysed must surely include the ions: Cl^-, SO_4^{-2}, HCO_3^-, Ca^{+2}, Mg^{+2}, Na^+, and K^+. It gives a complete imagination about the chemical composition of water whereby the content of Cl^- and SO_4^{2-} has been limited by Estonian drinking water standard. It is necessary to fix the TDS, pH, and electric conductivity of water. In addition to components mentioned the sums of BTEX, PAH, oils, and phenols must be determined (Licha T, Herfort M, Sauter M 2001). The content of other chemicals (As, Fe, Se etc.), occurring in areas of landfills, which concentration is normalized by variable standards (Joogivee kvaliteedi… 2001; Joogivee tootmiseks… 2003; Ohtlike ainete… 2010; Pinnavees ohtlike… 2010), must also be analysed.

The groundwater transport parameters (diffusivity, dispersivity, sorption coefficients, the half-life of organic contaminants etc.) should also be determined by means of field experiments carried out for main species of water-bearing layers (sand, gravel, moraine, siltstone, sandstone, limestone). For that purpose, a group of several borings should be sunk in these layers (Bear J, Cheng A 2010; Domenico P, Schwartz F 1989; Fetter C 1994, 1993; Hagerman J *et al*., 1989). An eventual contaminant should be injected into one of the borings and the emerging of this contaminant and alteration its concentration should be recorded in other borings. Data collected in this way are useful as for the study of landfills as well for investigation other contamination problems in East-Viru County.

It is purposeful to run repeatedly the landfill models in accordance with getting new observation data. This way an operative feedback between the performance of models and monitoring network will be most efficiency. The transient flow and transport models should be also developed. It enables to account the impact of time-dependent boundary conditions on the modelling results and enhance their reliability.

Some model uncertainties may be become evident due to an unavoidable mismatch between conceptual and measured data. It appears as a no uniqueness of model predictions. A number of approaches exist providing different model averaging techniques to overcome this problem (Dausman A *et al.* 2010; Singh A, Mishra S, Ruskauff G 2010; Ye M *et al*. 2010).

CONCLUSIONS

Based on results of the present study it is possible to draw the next principal conclusions.

- More than 25 groundwater modelling projects considering variable hydrogeological problems of East-Viru County has been completed by many investigators from 1972 until 2010. Unfortunately, most models performed have become out of data and cannot produce useful ideas.

- A universal transient groundwater flow model coupled with a transport model is needful for the development of the water management in East-Viru County. This model should cover all territory of North-East Estonia with its coastal sea, the northern portion of Lake Peipsi, border districts of Russian Federation and include all main aquifers and aquitards from the ground surface to as low as the impermeable part of the crystalline basement. The regional hydrogeological model of Estonia (RHME) completed by L. Vallner fits these requirements.

- The RHME can be refined for a particular study of detached local problems (contamination impact of landfills and mines, designing, and protection of groundwater intakes etc.). It is recommendable to couple the local models with the regional model to explain their mutual effect.

- An efficient groundwater monitoring system can be developed based on demands of the RHME. On the other hand, the RHME can be corrected and improved based on data of groundwater monitoring.

- The RHME is a suitable tool to investigate the interaction of groundwater bodies, coastal sea, Lake Peipsi, surface water bodies, mines, landfills, and groundwater intakes in East-Viru County considering the impact of border districts of Russian Federation and other parts of Estonia. The groundwater heads, flow directions, velocities, and rates, as well concentrations of groundwater ingredients can be simulated for every point of water-bearing formation for every time moment.

- The detailed groundwater flow and transport models coupled with the RHME have been completed for the area of the Kohtla-Järve ash and semi-coke landfill. It was proved by a profound calibration of models that the transversal (vertical) hydraulic conductivity of landfill deposits averages predominantly 10^{-4} m/day or $1.2*10^{-9}$ m/s.

- Correspondingly to model calculations, the Total of Dissolved Solids (TDS) will increase up to 18 times in the Ordovician carbonate bedrock and up to 2.2 times in the Ordovician-Cambrian sandstone aquifer system beneath the Kohtla-Järve landfill until 2110. The area of increased TDS will cover 6.2 km^2 in carbonate bedrock and 5.5 km^2 in sandstones.

- Distribution of phenol contamination in water-bearing layers of the Kohtla-Järve area has been determined by transport modelling. The concentration of phenols reaches 36 mg/L beneath the landfill at present. The outer contour of the plume of phenol contamination exceeds the area of the landfill by 300 m upstream to the groundwater flow and 2,000 m downstream. The area of the contamination plum is 10.6 km^2 in the Ordovician bedrock and 7.5 km^2 in the Ordovician-Cambrian aquifer system. During next 50–100 years phenol contamination will spatially expand to a certain extent, but its concentration will significantly increase – up to 57 mg/L in Ordovician layers and up to 10 mg/L in the Ordovician-Cambrian aquifer system.

- The groundwater flow and transport models performed for the area of Kiviõli semi-coke landfills and connected with the RHME prove the existence of a moderate phenol contamination in the Ordovician carbonate bedrock beneath landfills. This contamination may intrude into the underlying Ordovician-Cambrian aquifer system in course of following 50–100 years.

- The Silurian-Ordovician regional aquitard does not prevent effectively the intrusion of contaminants from Ordovician carbonate bedrock into the underlying Ordovician-Cambrian aquifer system in East-Viru County.

- The detailed groundwater flow and budget model completed for the area of ash landfills of the Balti Power Station at Narva shows that the groundwater outflow from these landfills, due to its relatively inconsiderable quantity and acceptable chemical quality, is practically not harmful to the river network of Narva Reservoir and for public groundwater intakes at present.

- The remediation projects of the environmental state of Kohtla-Järve and Kiviõli landfills have been completed by enterprises AS Maves, EL Konsult OÜ, IPT Projektijuhtimine OÜ, and Ramboll Finland OY. The basic idea of these projects is to cover the landfills by a coating bed, which thickness is about 0.5 m and vertical hydraulic conductivity 10^{-8} m/s, to prevent infiltration of rain and thaw water. A more or less waterproof vertical barrier with the total length of about 7 km is planned to arrange into the loose deposits around the Kohtla-Järve landfill for catching fluxes of contaminated groundwater.

- The scientific argumentation of remediation projects mentioned is poor. A correct hydrogeological analysis of the spatial transport of contaminated groundwater lacks. Therefore, it is not possible quantitatively to appraise the environmental and economic efficiency of the remediation planned on the ground of project documentation.

- The coating liner designed will not substantially to impede formation and migration of contaminated groundwater since the vertical hydraulic conductivity of the liner is tenfold less than this one of existing landfill deposits. Therefore, the construction of the liner designed is useless.

- Contaminated groundwater formed in the landfill passes the vertical barrier from underneath and spreads in the bedrock.

- Smoothing the asperities of the Kohtla-Järve landfill may cause long-term burnings of this landfill in conjunction with emissions of hazardous gazes into the atmosphere.

- The groundwater flow and transport models completed and approbated in frames of the current project can be used for the study of remediation problems of the environmental state of Kohtla-Järve and Kiviõli landfills. It makes feasible to check the efficiency of remediation projects of landfills started already and plan their corrections if it will be needful. For that purpose, these models should be developed and provided with supplementary monitoring and experimental data.

- A development of the groundwater monitoring network is desirable for the efficiency assessment of landfill remediation and management projects in the East-Viru County. It is recommendable to install the supplementary observation wells tapping different vertical intervals of water-bearing layers and to sample the wells after

1–2 months during a couple of years. The groundwater transport parameters (diffusivity, dispersivity, sorption coefficients, the half-life of organic contaminants etc.) should be determined by means of field experiments carried out for main species of water-bearing layers.

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