LATVIJAS UNIVERSITĀTE
ANNO 1919
UNIVERSITY OF LATVIA

The 70th Scientific Conference of the University of Latvia

Session of Geology

Section

„Groundwater in Sedimentary Basins”

ABSTRACT BOOK

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Faculty of Geography and Earth Sciences,
Alberta street 10, room 313, Riga

January 30, 2012

Riga, 2012

The session is organised in the scope of the European Social Fund project *"Establishment of interdisciplinary scientist group and modelling system for groundwater research"* (Project contract Nr. 2009/0212/1DP/1.1.1.2.0/09/APIA/VIAA/060). Project is implemented by University of Latvia, Faculty of Geography and Earth Sciences and Faculty of Physics and Mathematics in collaboration with Latvia University of Agriculture, Faculty of Rural Engineering, Department of Environmental Engineering and Water Management.
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ISBN 978-9984-45-443-6

CONTENTS

THE STATE OF ART AND NEW TRENDS IN THE APPLICATION OF ISOTOPE-GEOCHEMISTRY FOR GROUNDWATER RESEARCH

Rein VAIKMÄE

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Environmental isotopes are now almost routinely applied in studies of groundwater resources.Tritium, radiocarbon and stable isotope ratios of D/H and $^{18}O/^{16}O$ have been most widely used, and increasing use is being made of inert gases.

In the past decade, aquifers have increasingly become palaeoclimatic archives in their own right alongside ice cores, sediments and other proxy records. The main tool for this task has been the noble gas palaeo-thermometer in combination with quantitative groundwater dating using radionuclides. Noble gas radionuclides play a unique role as tracers in environmental studies due to their chemical inertness and low concentration, making them ideal tracers. The same properties on the other hand make them difficult to measure on natural concentration levels. Therefore, for decades, low level counting (LLC) was the only method for detecting radioisotopes of argon and krypton at an atmospheric level. In recent times and with the increase in interest and potential applications, analytical efforts with novel detection methods have been intensified. In this talk, noble gas groundwater dating techniques over time scales from decades to millions of years are also discussed in relation to noble gas palaeo records at different locations in Europe.

THE SPECIFICS OF DETERMINING HYDROGEOLOGICAL PARAMETERS FOR TWO-PHASE LIQUID FLOWS IN POROUS MEDIA

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The two-phase fluid vertical distribution in the groundwater aquifer concept has been repeatedly changed and hydro geological calculation methodology has changed as well. Yet, in the 1950's-1960's, it was based on a standard (API-Publication, 2004), but erroneous impression, that the free-phase liquid layer in a ground-water aquifer forms a lens that is strictly separated from the water and floats above it. It was considered that in this lens, 100% from the pore volume is filled with free-phase liquid.

As of the 1970's (Lefebvre, 2006), subsequent studies (Brook and Corey, 1964; Genuchten, 1980) showed that the capillary force influence on free-phase liquid layer deposits range in the groundwater aquifer creates a complex multiphase system, in which free-phase liquid saturation of soil pores changes from its maximum percentage content in the top layer to the minimum at the bottom layer (Fig. 1). Field studies have shown (API-Publication, 2004) that the multiphase concept is applicable to coarse granular, as well as, fine sand. However, the sand grain size in this concept has fundamental importance because depending on whether the sand is coarse or fine, a difference can be seen there between in-the-bore-hole-observed (apparent) and in the groundwater aquifer's saturated part's actually existing (real) free-phase liquid layer thickness (Abdul *et al.*, 1989).

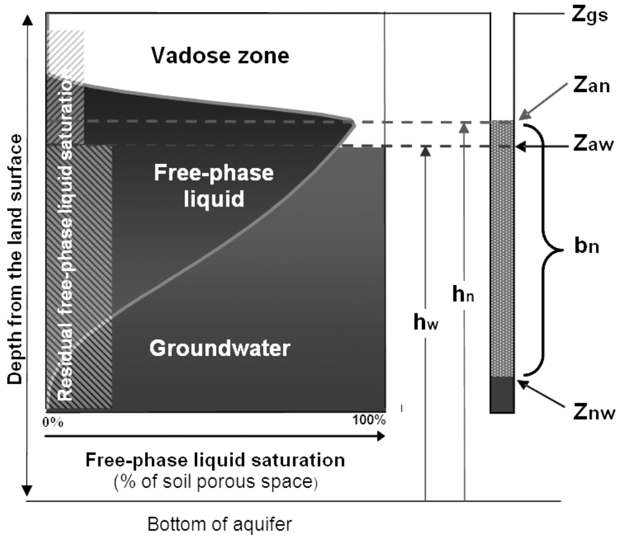

Fig. 1. Physical model of two-phase liquid vertical distribution in the groundwater aquifer. LEGEND. z_{gs} – reference level (top of surface), z_{an} – air and free-phase liquid interface, z_{aw} – hypothetical (pizometric) groundwater level, b_n – observed free-phase liquid layer thickness in the monitoring well, z_{nw} – free-phase liquid and water interface, h_w – water rising altitude (pressure) in the groundwater aquifer, h_n – free-phase liquid rising altitude (pressure) above its and groundwater interface.

The main factors resulting in the difference between in-the-borehole-observed (apparent) and the groundwater aquifer saturated part of the actual (real) free-phase liquid layer thickness, are the capillary forces in the horizon, the free-phase liquid density and the degree of commitment with the mineral soil (Blake and Hall, 1984). The finer the soil and the higher the capillary forces act, the greater is the difference between the in-the-borehole-observed and the real free-phase liquid layer thickness in the groundwater aquifer's saturated part. Given the fact that borehole capillary forces practically don't exist, the water level will always be located below the groundwater capillary elevation zone. As a result, free-phase liquid from the groundwater capillary elevation impact zone, where water pressure from below acts on it through the pores, will move to the borehole, where capillary water pressure of this type does not exist, and fill in this space in the bore-hole, starting from the lowest level corresponding to the groundwater capillary zone elevation, beginning in the horizon.

However, the described approach is valid for limited free-phase liquid layer thickness values. It was found that the free-phase liquid layer in the groundwater aquifer increases and reaches a certain thickness (Lefebvre, 2006). This layer mass pressure begins to overwhelm the underlying water capillary elevation and as a result, the difference between the in-the-borehole-observed horizon and the free-phase liquid layer thickness declines (Testa and Paczkowsk, 1989). And vice versa – a decrease in the free-phase liquid layer in the groundwater aquifer increases the part of free-phase liquid that is mechanically linked to the soil, resulting in this liquid losing its mobility that interferes with its accumulation in the borehole. In this case, the in-the-borehole-observed free-phase liquid layer thickness is less than what it should be in accordance with the study design.

The fact that all these processes result in significant changes in the rock filtration properties themselves, which even further reduces the free-phase liquid mobility in soil and with it the likelihood that such a layer will form at all (Burdine, 1953), should also be mentioned as an important additional factor in the mobile fluid volume loss by adsorption.

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TOWARDS "REALISTIC" FAULT ZONES IN A 3D STRUCTURE MODEL OF THE THURINGIAN BASIN, GERMANY

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3D computer models of geological architecture are presently evolving into a standard tool for visualization and analysis. Such models typically comprise the bounding surfaces of stratigraphic layers or stratigraphic volumes, and faults. Faults are ubiquitous in the Earth´s crust; they are shear fractures across which the stratigraphic layers are offset. Faults thus affect the continuity of aquifers and can themselves act as fluid conduits or barriers. This is one reason why a "realistic" representation of faults in 3D models is desirable. Still, many existing models treat faults in a simplistic fashion, e.g. as vertical downward projections of fault traces observed at the surface. Besides being geologically and mechanically unreasonable, this also causes technical difficulties in the modelling workflow. Boreholes located close to a fault at the surface can cross dipping fractures at depth, resulting in stratigraphic control points being allocated to the wrong block. Most natural faults are inclined and may change dips according to rock type or flatten into mechanically weak layers. Also, faults tend to split up into several branches, forming fault zones. Obtaining a more accurate representation of faults and fault zones is therefore challenging.

Here we present work-in-progress from the Thuringian Basin where we attempt to integrate complex fault zones in both a 3D architecture model and a numerical flow model. The Thuringian Basin is a doubly-plunging, NW-trending syncline some 150 km long and 75 km wide. Its mostly Triassic strata includes sandstone, limestone, shale and evaporite. The Thuringian Basin became separated from the much larger North German Basin only in the Late Cretaceous time, when contractional tectonics created its synclinal geometry. The syncline is dissected by several longitudinal fault zones. For some of these, a history of early normal faulting followed by reverse reactivation has been demonstrated. Wholesale uplift in the latest Cretaceous and early Paleogene time led to exhumation of the Thuringian Basin. Deposition is presently restricted to its north-eastern corner. The uplifted Thuringian Basin grants access to strata deeply buried in the North German Basin, Germany's most important hydrocarbon province. It can thus be viewed as a very large outcrop analogue of a geologic situation that has been intensely investigated by seismics and drilling in the North German Basin. The TB is geologically mapped at the 1 : 25.000 scale. Subsurface data is much scarcer.

There are a number of boreholes penetrating the basement and many more that have reached the Zechstein. Localized information is available from a few deep salt mines. Modern seismic data is limited to a few 2D lines, including two, newly acquired by the INFLUINS project. Away from the fault zones, extrapolation to depth is facilitated by the relatively constant thickness of most stratigraphic units.

In the fault zones, there is never enough data to fully constrain the geometries, so we need to make educated guesses as to how the faults continue to depth. We use balancing of serial, parallel cross-sections as a method of constraining subsurface extrapolations. The fundamental assumption is that rock volume does not change during deformation. Under plane strain conditions, i.e. with all particles moving in the cross-section plane, this translates into constant cross-section areas before and after deformation. The structure sections are checked for consistency by restoring them to an undeformed state with original layer thicknesses. If this is possible without producing any gaps or overlaps between strata, the interpretation is considered valid (but not unique) for a single cross-section. Such valid solutions are found in a computer-aided yet intuitive, trial-and-error procedure using Midland Valley´s 2DMove program.

Additional constraints are provided by comparison of adjacent cross-sections. Structures should change continuously from one section to another unless there are obvious cross-faults. Also, from the deformed and restored cross-sections, we can measure the length change (strain) incurred during deformation. The strain should be compatible among the cross-sections: If at all, it should vary smoothly and systematically along a given fault zone.

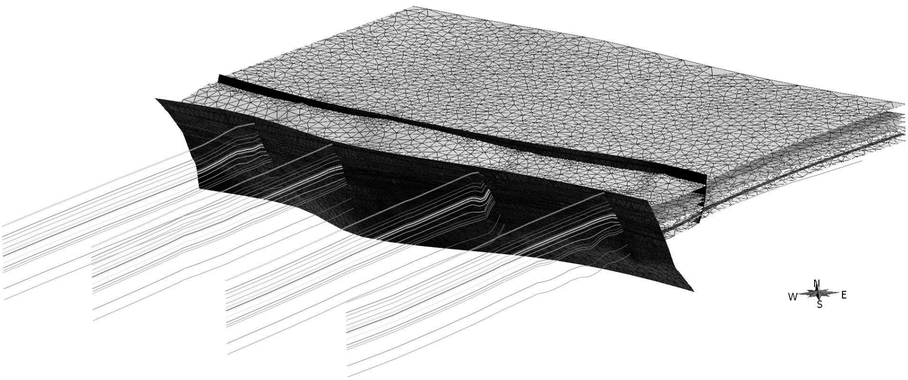

Fig. 1. High resolution model of a graben structure in the north-western Thuringian Basin, showing serial cross-sections and triangulated surfaces representing stratigraphic boundaries and faults. Notice short-wavelength folding close to the fault visible in some cross-sections.

The stratigraphic contacts and faults in the resulting grid of parallel balanced sections are then interpolated into a model containing stratigraphic boundaries and faults as triangulated surfaces in gOcad (Fig. 1). The interpolation is also controlled by borehole data located off the sections and the traces of stratigraphic boundaries at the surface. We have written customized scripts to largely automatize this step, with particular attention to a seamless fit between stratigraphic boundary surfaces and fault planes which share the same nodes along their contacts. Additional attention was paid to the creation of a uniform triangulated grid with maximized angles. This ensures that uniform triangulated volumes can be created from the model for further use in numerical flow modelling. A 3D balancing of the structure model is also planned, to check and increase the accuracy.

An, as yet, unsolved problem is the implementation of the fault zones and their hydraulic properties in a large-scale model of the entire basin. Short-wavelength folds and subsidiary faults control which aquifers and seals are juxtaposed across the fault zones. It is impossible to include these structures in the regional model, but neglecting them would result in incorrect assessments of hydraulic links or barriers. We presently plan to test and calibrate the hydraulic properties of the fault zones in smaller, high-resolution models and then to implement geometrically simple "equivalent" fault zones with appropriate, variable transmissivities between specific aquifers.

LIMITS AND PRESUPPOSITIONS ON CREATING AND USE OF THE REGIONAL HYDROGEOLOGICAL MODEL OF LATVIA

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The main limits regarding the regional hydrogeological model (HM) of Latvia (see Fig. 1) are, as follows:

- The HM will be used for the management of drinking groundwater resources of Latvia;
- The HM is created by the Environment Modelling Centre team of the Riga Technical University (RTU); the project is co-financed by the European Fund of Regional Development;
- The duration of the project is 24 months; the HM must be established in 2013;
- The geological and hydrogeological information required for establishing the HM, is provided by the Latvian Environment, Geology and Meteorology Centre (LEGMC);

- The principal parameters of the HM must be agreed between RTU and LEGMC;
- Data carried by the HM must be publicly available as a part of Latvia's environment information system; the system is supported by LEGMC;
- During the five years (till 2017), RTU and LEGMC cannot use the HM commercially.

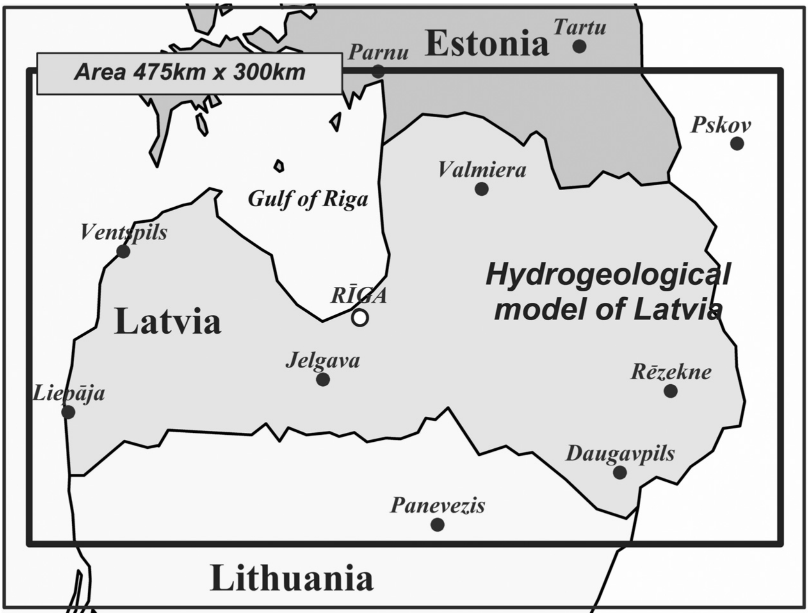

Fig. 1. Location of Latvia's HM.

The HM of Latvia will generalize geological and hydrogeological information accumulated by LEGMC. The HM will also serve as the base for creating more detailed local HM's.

It is not possible to incorporate all the data that can be provided by LEGMC into regional HM's. A reasonable reduction in HM complexity can be achieved by implementing the following presuppositions:

- The complexity and dimensions of the HM must not exceed the feasibility of a modern personal computer used to run the HM; The HM simulates the steady state average regimes of the groundwater flow; the HM area size is 475 km × 300 km; the HM volume is approximated by the finite difference method; its plane approximation step is 500 meters; the spatial HM grid contains 25 planes; therefore, the grid consists of $951 \times 601 \times 25 = 14.86 \times 10^6$ nodes; the HM volume represents the active groundwater zone that is bedded by the regional Narva aquitard;

- To ensure compatibility with the models and software tools of other countries, the "Groundwater Vistas" (GV) commercial program is used for the running of the HM; the program is being regularly updated (GV-6 version is available); it contains the MODFLOW, MODPATH and MT3D software tools applied for groundwater modelling worldwide;
- At the present, the HM consists of its active and passive parts; the active part includes the land territory of Latvia and the Gulf of Riga; the passive part represents the border areas of neighbouring countries. However, the HM is open for trans-boundary modelling projects; a neighbouring country would then provide data for activating the HM area involved;
- Although buried valleys may be of considerable importance, they are not accounted for by the current HM version; it is difficult to create them geometrically as the filling material of valleys may be unknown;
- In the HM, only the Narva aquitard is continuous; the other geological layers are discontinuous, because they include areas with a zero thickness; for the model, these areas have a thickness of 0.02 meters and their permeability is 1.0 m/day;
- Three elevation surfaces of the HM are especially important:
  - the hydrogeological relief *relh* that represents the ground surface where the hydrographical network is incorporated;
  - the geological relief *relg* that gives land surface elevations;
  - the sub-Quaternary surface *subQ* that covers the system of basic geological layers.

The difference $m_w = relh\text{-}relg$ is the thickness of surface water bodies. (in the HM, $m_w > 0$ for the sea area and for the Daugava river with its three lakes with hydroelectric power stations); for other water bodies (lakes, rivers), $m_w = 0$.

The difference $\Delta = relg\text{-}subQ$ is used for obtaining the Quarternary system thickness m_Q: $m_Q = \Delta$ if $\Delta > 1.0$; $m_Q = 1.0$ if $\Delta \leq 1.0$ and $relg = subQ + 1$; by correcting *relg*, along the river valleys where $\Delta < 0$, the *subQ* surface remains unchanged (no deep valleys are cut into it); otherwise, the grid nodes will be lost where river long lines elevations must be connected (option <u>River</u> of GV):

- The *relh* map serves as the piezometric boundary condition, on the HM top; due to this condition, the HM automatically creates a feasible infiltration flow distribution;
- No real thicknesses of bogs, of the aeration zone and of the unconfined Quaternary aquifer are used during the HM calibration; the aeration zone of the thickness of 0.02 meters acts as a formal aquitard that controls the intensity of the infiltration flow; the bogs are located within this formal layer; if necessary, the real thicknesses of the abovementioned layers can be restored;
- In HM, real thicknesses are used for layers (bogs, the aeration zone and the quaternary unconfined layer are exceptions); to account for admixtures that exist in the layers, the maps of their permeability are corrected;

- For aquifers, along the borderline of the HM active part, piezometric boundary conditions (heads) are applied; an impervious border surface cannot be used, because the cross border groundwater flow is notable everywhere;
- As the piezometric boundary condition, on the HM bottom, the Pernava aquifer map of its head distribution is used.

Most of the abovedescribed measures can be used, if complex hydrogeological models have to be created.

STUDIES AND PROJECTIONS OF HYDRAULIC CONDUCTIVITY OF DEVONIAN AND CAMBRIAN CLASTIC SEDIMENTS

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Aquifer fluid conductivity properties describe the ability of sediments to transmit groundwater, and consequently govern the groundwater flow. Hydraulic conductivity mostly depends on the different physical properties of the sediments and their liquid filtering properties. Studies and knowledge of hydraulic conductivity (K), transmissivity, storativity and aquifer properties for the particular aquifer are very important for the hydrogeological problem solving process.

This study presents the results of the comparative study between hydraulic conductivity, grain size distribution and sediment lithology of the lower Devonian Emsian stage, the middle Devonian Eifelian and Givetian stage, the upper Devonian Frasnian stage, and Cambrian clastic sediments in the central part of the Baltic Basin. The aim of this study was to find characteristic hydraulic conductivity values for each aquifer based on aquifer grain size distribution and lithology on the one hand and pumping test results on the other.

For the calculation of the hydraulic conductivity, one has to take into account not only grain size distribution, but effective porosity, temperature and kinematic viscosity of the fluid as well, which are lacking in this study.

Pumping test results provide a range of at least two orders of hydraulic conductivity values for each aquifer. To characterize the typical values for each aquifer and further subdivide each aquifer into regions of different hydraulic conductivities, the pumping test results were correlated with grain size distribution. A fraction of fine particles, with a size less than 0.05 mm, was chosen as a limiting factor for the hydraulic conductivity in the sandstones. The correlation of hydraulic conductivity and grain size distribution was carried out by comparing the <0.05 mm fraction and the respective hydraulic conductivity values in the wells. The results suggest that grain size distribution in general does not correlate with conductivity obtained from the pumping tests. In general, in comparing the hydraulic conductivity values obtained from pumping tests with calculated values from grain size distribution, the calculated values in some cases differ from those obtained for some units (1 – 3 m/dnn), but in some cases more than two times. This is connected with the uncertainty of existent data and imperfections in calculation methods. A correlation with the lithology of the aquifer (as described in boreholes) shows better results and allows the subdivision of the aquifer into two clusters of typical K values.

A correlation of the grain size and hydraulic conductivity provided a range of the average hydraulic conductivity values for each aquifer. For example in D_2ar and D_2br aquifers K values varied from 1 – 7 m/day, in D_3gj 1 – 8 m/day and in D_3am aquifer 1 – 5 m/day.

This study is supported by the European Social Fund project
No. 2009/0212/1DP/1.1.1.2.0/09/APIA/VIAA/060.

SENSITIVITY OF HYDROGEOLOGICAL MODEL TO THE SURFACE ROUGHNESS AND SPATIAL VARIABILITY OF HYDRAULIC CONDUCTIVITY

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The calibration of the hydrogeological model fits the calculation result to the observation data (in our case water pressure in a borehole) by changing the coefficients of effective conductivity. Thus, calibration coefficients include all the uncertainties of the forcing data, layers thicknesses and variability of the conductivity of the material. Such an approach does not allow for the making of spatially distributed models without calibration.

The calibration procedure requires a lot of computational power. The calibration of the property of each model grid cell is still an unmanageable process and does not lead to a unique solution. In our case the accuracy of calibration is strongly limited by the quantity of available geometric and observation data.

In the present work, the spatial variability of the roughness of surfaces and conductivity was estimated. Surface shape and hydraulic conductivity of the layer were perturbed and the sensitivity of the model to the surface roughness and spatial variability of hydraulic conductivity was calculated. Additional attention was paid to the estimation of the impact of subgrid scales of the topological surface to the calculated hydraulic head distribution.

This study is supported by the European Social Fund Project
No. 2009/0212/1DP/1.1.1.2.0/09/APIA/VIAA/060.

TRANSIENT MODELLING OF GROUNDWATER DYNAMICS IN THE BALTIC ARTESIAN BASIN

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The transient solver of groundwater flow for the model of the Baltic Artesian Basin (BAB) was developed using the finite volume OpenFOAM libraries. Solver potentialFoam was employed as a basis for the development, adding the transient and water abstraction terms, as well as the management of the unconfined zone. The geometry of the model consists of a subsequent combination of aquifers and aquitards with enormous differences of conductivities by 9 orders and thicknesses by 2 orders between them. Due to the large area of the BAB and the complex structure of sediments only one element per layer is tolerable. Therefore, high accuracy schemes with the ability to handle large pressure gradients should be used. Investigations show that Monotone Upstream-centred Schemes for Conservation Laws (MUSCL) are best suited for such typical hydrogeological structures.

Mesh structure, conductivity and boundary conditions are prepared in the pre-processing routines of HiFiGeo software and exported to the OpenFOAM file formats.

A method for the consideration of the unconfined zone has been developed. The storativity and conductivity of the unconfined volume elements are reduced thus excluding the unconfined zone from the flow field.

The influence of storativity and conductivity on the transient solutions has been demonstrated for typical ranges of material properties in the BAB.

Distinct time dependence of the groundwater abstraction is typical for the BAB over the last 50 years with a maximum in the 1980's. The resulting transient behaviour of the groundwater flow, the piezometric head and the development and disappearing of the cones of depression has been calculated.

This study is supported by the European Social Fund Project
No. 2009/0212/1DP/1.1.1.2.0/09/APIA/VIAA/060.

FORMATION OF GROUNDWATER IN SEDIMENTARY BASINS: TRADITIONAL AND ALTERNATIVE MODELS

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According to the accepted "classical" model of the Baltic Artesian Basin groundwater dynamics, currently acknowledged and widely used by researchers, groundwater recharge is the most intensive in the heights, and groundwater runoff moving towards the periphery of the artesian basin, *i.e.* to the central part of the Baltic Syneclise in the area of the Baltic Sea, where its submarine discharge takes place (Juodkazis, 1979; Региональная..., 1989; Mokrik, 2003; and others). The greatest thickness of the fresh groundwater layer in the Eastern Baltic region reaches 500 meters, and this has been determined in the northwestern part of Lithuania (Mokrik, 2003). However, an alternative model of fresh groundwater formation – meltwater injections into aquifers – is also possible. It has been established that the oxygen isotopic composition of fresh groundwater of the Cambrian-Vendian aquifer in the Estonian Monocline is abnormally light and that $\delta^{18}O$ values reach -20‰ – -22‰, *i.e.* the groundwater is of glacigenous origin (Vaikmäe, 1999; Mokrik, 2003). Referring to these data, the researchers explain the formation of fresh water resources by meltwater injections into the aquifers during the degradation of the ice sheet and deglaciation of the area (Vaikmäe *et al.,* 2001; Mokrik, 2003; Zuzevičius, 2010; and others).

According to other researchers, ways for fresh groundwater to form could be explained by the mechanism of meltwater with high hydrostatic head circulation under the continental ice sheet during the transgressive phase of glaciation (Boulton *et al.,* 1995, 1996; and others). A great amount of water was infiltrated into aquifers of the subglacial substratum in this way. We think that the model of infiltration mentioned has been proved by the results of groundwater (dissolved carbonates) radiocarbon dating. A few hundred groundwater dating results from different aquifers in Lithuania and the surrounding regions show that the entire amount of fresh groundwater was formed only commencing from the second half of the Last Glacial (Middle and Late Weichselian), *i.e.* there is practically no water older than 34-35 kyrs (Bitinas, 1999, 2011). It is important to note that the results of groundwater radiocarbon dating are not accurate (they are usually "aged"), and that is why it is necessary to make corresponding corrections in their interpretation (Mokrik and Mažeika, 2006; Mokrik *et al.,* 2008). The distribution of groundwater dating results of the Upper Permian aquifer in the Western Lithuania is presented (Fig.) as an example. According to the traditional ("classical") model,

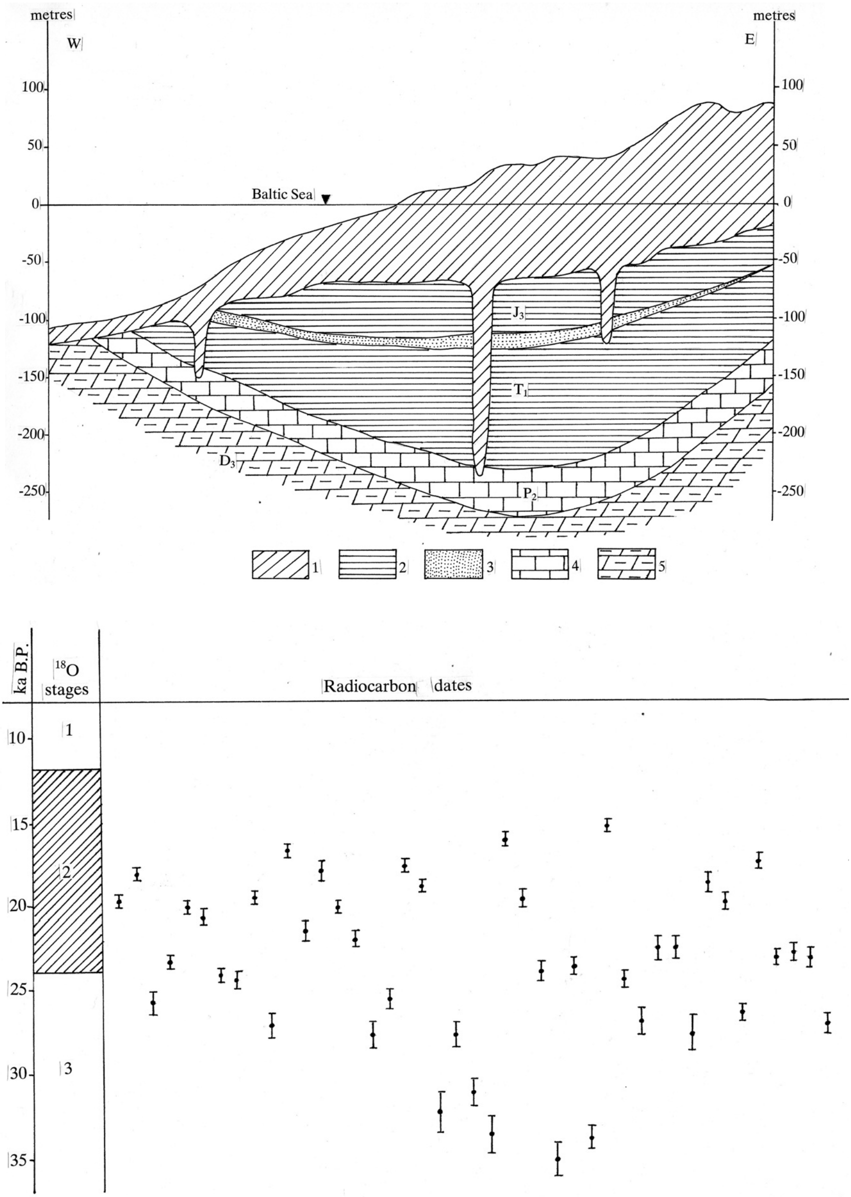

Fig. Geological occurrence of the Upper Permian aquifer in Western Lithuania and the radiocarbon age of groundwater (dissolved carbonates) of this layer.

1 – Quaternary deposits; 2 – clay; 3 – sand; 4 – fractured limestone; 5 – clayey marl and limestone. Age of deposits: D_3 – Upper Devonian; P_2 – Upper Permian; T_1 – Lower Triassic; J_3 – Upper Jurassic

the formation of fresh water in the Upper Permian aquifer took place more or less regularly during the Middle and Late Weichselian, despite the fact that the area before glaciation had been frozen over, so any water infiltration was hardly possible. Another model of fresh groundwater formation – meltwater injections with high hydrostatic head – gives a better explanation of fresh groundwater distribution and the reason why freshwater injections reach such depths.

A different interpretation of groundwater dating results, as well as a new perception of continental glacier dynamics and the mechanism of its meltwater circulation, enable one to change the attitude not only of the almost dogmatic approach to glacial geology, but also to modify the already settled viewpoints on groundwater dynamics in aquifers during ice ages and the formation of fresh groundwater resources. Thus, we have to answer a number of new questions: supposing that part of the fresh groundwater resources in the Eastern Baltic region have been formed due to meltwater injections, what are their real exploitation resources? Are they possibly much smaller than we imagine? Can most of them be reasonably considered as the renewing ones?

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WATER FILLED UNDERGROUND OIL SHALE MINES AS A HEAT SOURCE

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Underground oil shale has been mined for 90 years in the middle-north part of the Baltic oil shale deposit, in the Estonian deposit. After the closure of the mine, the mine works filled with water. Underground oil shale mining creates underground water pools called technogenic water bodies (Figure 1). The Estonian oil shale deposit is comprised of ten closed mines that are fully or partly filled with water. Eight mines in the central part of the deposit: Ahtme, Kohtla, Kukruse, Käva, Sompa, Tammiku, mine No. 2 and mine No. 4 form one water body. Ubja mine and Kiviõli mines are located in the western part of the deposit, away from the other mines.

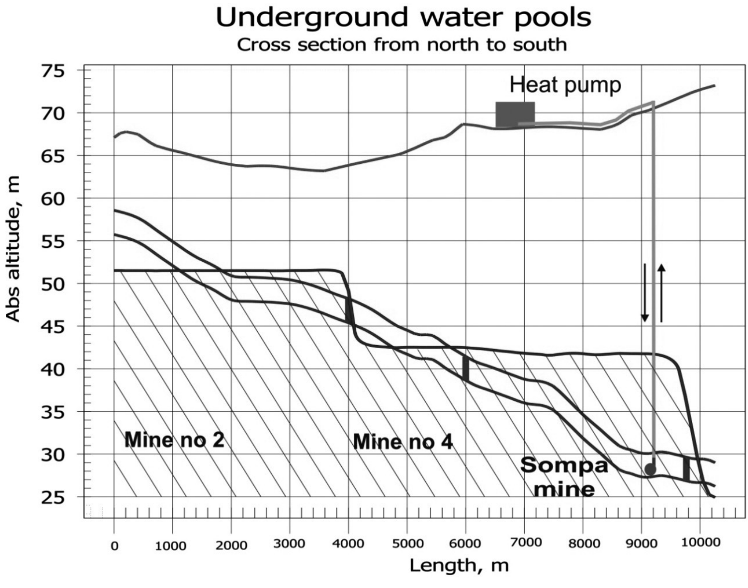

Fig. 1. North-South cross section of underground mining area and heat pump installation example.

The main aim of this paper is to analyze the feasibility of using mine water as a heat source for heat pumps and to find suitable places to set up such systems. It would be useful to use this mine water as a heat source for heat pumps to produce heat.

Technologies have to be created and evaluated for defining hydro geological parameters to define underground space, properties and classification of used mines. Classification helps to define the space that is available for water in abandoned mines. A 3D model was built with geometrical data from mine plans, mine closing acts and borehole data and from land survey data. The main tools chosen for spatial modelling were spreadsheets and MS Access databases for systemising and querying data, MapInfo for georeferencing, Vertical Mapper for interpolating and grid calculations and Modflow for the pumping simulation. With the help of interpolated grids, the surface elevations, layer thicknesses and required properties were calculated.

The best possible technical solution for using mine water in heat pumps is:

1) pumping the water through the drill hole onto the ground surface
2) water goes to the heat exchanger unit
3) mine water temperature will be lowered in the heat pump heat exchanger by about 1...4 degrees,
4) after that, the mine water is directed back to the mine.

Table 1. COP values for Ahtme 10MW heat pump complex

ΔT	Heat producti on kW	Water needed m3	Pumps needed	Electricity for pumps,kW	Heat pumps needed	Electricity for heat pumps, kW	Summary of elecricity,kW	COP
1	10000	8592	8	1 320	7	2 562	3 882	2.58
2	10000	4296	4	660	7	2 562	3 222	3.10
3	10000	2864	3	495	7	2 562	3 057	3.27
4	10000	2148	2	330	7	2 562	2 892	3.46

Table 2. Kiikla 500kW heat pump

Pilot project in Kiikla 500kW heat production unit									
Date	Heat producti on, MWh	Elecricity for heat pump, kWh	Electricit y for pumps, kWh	Summary of elecricity , kWh	Pumped amount of water, m3	Water from mine, °C	Water to mine, °C	ΔT	COP
20.04.	3.559	1415	341	1756	1488	6.4	5.7	0.7	2.03
21.04.	2.104	655	268	923	1534	6.1	4.9	1.2	2.28
25.04.	8.381	2749	1245	3994	6852	6.4	5.1	1.3	2.10
29.04.	3.321	1131	632	1763	3151	5.9	4.6	1.3	1.88
02.05.	6.97	2421	893	3314	4671	6.2	5.2	1	2.10
04.05.	4.954	1743	558	2301	2856	6.1	5.2	0.9	2.15
06.05.	4.82	1649	599	2248	2978	6.6	5.2	1.4	2.14
18.10.	45.684	15600	3468	19068	24673	0	0	0	2.40
27.10.	29.015	9671	1800	11471	10498	0	0	0	2.53
31.10.	11.828	4299	842	5141	4753	0	0	0	2.30
08.11.	20.068	6081	1538	7619	8688	0	0	0	2.63

If we use underground water pool water, then the recommended temperature reduction must be more then one degree. It depends on how large a heat exchanger is economical. When the temperature is lowered less, we have to use large volumes of mine water.

The best location for the heat pump complex is near Ahtme Power Plant. The heat pump complex in Ahtme will need water pumps and heat pumps. By building this unit at Ahtme, the water requirement and the COP values are as shown in Table 1.

Mine water usage for a heat pump complex is unique in the world. The first pilot pump in Estonia was opened in 2011 at Kiikla settlement in Estonia. At Kiikla settlement the installed heat pump is like a pilot unit for using mine water as a heat source and the COP values and other parameters are shown in Table 2.

RECONSTRUCTING THE GROUNDWATER FLOW IN THE BALTIC BASIN DURING THE LAST GLACIATION

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Groundwater flow beneath the ice sheets has caught the imagination of scientists relatively recently. The groundwater in the Baltic basin can be subdivided into three identifiable groups according to its chemical and isotopic composition: water of the Quaternary age of either the warm (1) or cold (2) stages, or Pre-Quaternary brine (3). The first two are readily identified by the stable isotope values and chemical composition (Radla, 2010), while the last is characterised by high concentrations of dissolved salts and is found in the deeper part of the basin.

The aim of this study is to test the assumption that the groundwater body of glacial origin found in the Cambrian–Vendian aquifer in Northern Estonia originated as a result of sub-glacial meltwater infiltration during the reoccurring glaciations. In the last decades it has been proved that most large ice sheets tend to reside on warm beds even in harsh climatic conditions and that sub-glacial melting due to geothermal heat flow and deformation heat from the ice flow is taking place.

A steady state regional groundwater flow model of the Baltic basin was used to simulate the groundwater flow during the last glaciations with model geometry adjusted to reflect the sub-glacial topography. Ice thickness model data (Argus and Peltier, 2010) was set as a constant head boundary condition on the topographical surface. In total, 19 calculation scenarios from 28 ka BP to 10 ka BP were chosen. The meltwater pressure at the ice sole was assumed to be equal to the ice pressure.

The modelling results suggest two main recharge areas of the Cm-V aquifer system, and a reversed groundwater flow that persisted for at least 14 thousand years. Modelling suggests that the groundwater flow velocities in the Cm-V beneath the ice sheet exceeded the present velocities by a factor of 10 on average. The volume recharged during the reversed groundwater flow amounts to ~ $2*10^{12}$ m^3. Assuming mean porosity of the Cm-V to be around 25% (Brangulis, 1995) this volume corresponds to approximately 200 km of the intrusion length, which is an obvious overestimation.

It is likely that the sub-glacial groundwater recharge operated on shorter time scales than modelled, perhaps due to the slow degradation of the permafrost which developed in front of the advancing ice margin, or pore water pressure at the ice sole less than that of the ice weight due to meltwater evacuation by the englacial drainage system. However, the most difficulty in quantifying the parameter is the development of a rather thin but continuous till layer that could effectively hamper the sub-glacial meltwater infiltration.

This study is financed by the European Social Fund Project No. 2009/0212/1DP/1.1.1.2.0/09/APIA/VIAA/060.

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THE UNCERTAINTY OF THE FUTURE ANNUAL LONG-TERM GROUNDWATER TABLE FLUCTUATION REGIME IN LATVIA

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Varying annual regimes of shallow groundwater levels affect the overall hydrological system significantly and in differing ways, as do related causes such as agricultural and forestry production. These regimes can be constructed and compiled if groundwater level monitoring is used and the groundwater levels are known. The primary objective of ground water regime monitoring is to record information on ground water levels in space and time. Measurements of water levels in wells provide the most fundamental indicator of the status of this resource and are critical to a meaningful evaluation of the effects it causes. Modelling groundwater levels using future daily climate data allows the prediction of future groundwater table fluctuations. The ability to adapt to changes depends on knowing the possible alterations of the groundwater level regime. Such knowledge could form the basis for different and flexible approaches to sustainable development in the future. The classical Latvian long-term groundwater level fluctuation regime can be described as an M-shaped function which represents two groundwater level maximums (in spring and late autumn) and two minimums (in winter and late summer). The aim of this paper is to model the long-term annual regime of relatively shallow groundwater levels using 14 climate scenario groupings and in addition, to analyze them according to the dominance of continentality in Latvia (Draveniece, 2007). Using relative groundwater levels allows one to compare wells with different absolute amplitude and average levels, as well as removing inter-annual trends. Such a method has been successfully used in Poland (Chelmicki, 1993). The mathematical model METUL was chosen as the best known and most appropriate model for Latvian climatic conditions for modelling future daily groundwater levels, using daily temperature, precipitation and humidity. To characterize how the variability of different climate scenarios affects the annual regime of shallow groundwater levels, statistical methods focusing on percentile analyses were applied. Results from one freely chosen model were used to analyze the differences and similarities between the single climate scenario model and the multiple climate scenarios model ensemble. Results show definite annual long-term groundwater regime changes in the future period (2070-2100) compared to the reference period (1961-1990) over all of Latvia. The future Latvian long-term groundwater level fluctuation regime can be described as an A-shaped function with one maximum and one minimum. Spatiotemporal differences are similar in both periods with a gradual transition adjusted for continentality, being most apparent in the spring months.

This study is financed by the European Social Fund Project
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EVOLUTION OF GROUNDWATER COMPOSITION IN THE DEPRESSION CONE OF THE RIGA REGION

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Groundwater is one of the major sources of drinking water, but its effective usage is very important, otherwise problems related to water quality and the quantity of the resource can arise. An example of ineffective groundwater usage has been observable from 1950 to 1990 in Riga, where intensive groundwater extraction from the Arukilas-Amatas multi-aquifer system has caused a sharp and significant lowering of piezometric surfaces.

Riga is the capital of Latvia, a city where the main water supply is both centralized and decentralized, mostly from groundwater sources, that is, from the Arukilas-Amatas multi-aquifer system, which consists of sandstones and siltstone. These rocks belong to the middle and upper Devonian and have good properties for the extraction of groundwater: they have high permeability and are widely spread. This system covers the upper Devonian Plavinas formation which consists of dolomite, but above it lies the Salaspils formation which consists of marl and gypsum, and is located in the southern and western part of Riga. Below this system lies the middle Devonian Narvas aquitard which consists of marl and clay.

Initially, the lowering of the water table was quick and the maximum decline of the piezometric surface was observed in 1972, when it was about 16 m lower than the average. Regeneration of the water table began at the end of the 1980's, when groundwater usage decreased. Nowadays, the piezometric surface is being renewed, and fluctuations are insignificant and described as natural. The area where the natural groundwater regime has changed, induced by the anthropogenic effect, is called "Large Riga".

In the study, long–term monitoring data has been used to track groundwater chemical changes and evolution in the Arukila-Amatas multi-aquifer system. Data on piezometric surfaces and major ions from 45 monitoring wells have been analysed. Before the data was analyzed, statistical analysis was done to exclude values which may be incorrect. Based on a map that shows the piezometric surface difference between two periods: 1949-1951 (which describes the natural situation), and 1970-1972 (where the minimal groundwater level in the Gauja aquifer was observed), the territory was divided into three zones – the central, middle and periphery, which differ from each other by the volume of decline in the piezometric surfaces. The map was created using Surfer 9 software. Changes in groundwater flow in the "Large Riga" area were studied using a hydrogeological model. Using this as a basis, the speed with which changes in water chemical composition in aquifers shows up and how these trends change in time were studied.

It was discovered that the sources of water with high SO_4^{2-} which worsen the quality of water in deeper aquifers, come from the Salaspils aquifer, because the first signs were observed in the Plavinas aquifer, which lies below the Salaspils formation. The same changes in water composition in deeper aquifers can be observed with a time lag.

Significant changes in water composition were observed in the central part, where the greatest lowering in the piezometric surface is found, and was sufficient to cause stronger downward flow from upper aquifers, which induced the mixing of water from different aquifers in this territory. As a result, there are great changes in water composition in this zone. In addition, the first signs of changes in water composition show up very quickly, but the return to a natural situation is relatively slow.

The fact was also observed, that when the piezometric surface rose up at the end of the 1980's, the mixing from different aquifers declined. This can be clearly observed in the upper Devonian Plavinas aquifer in the central part, where there is an increasing concentration of the HCO_3^- ion in the latest samples. These are the first signs that the situation in this multi-aquifer system is beginning to return to its natural condition.

Despite the fact that Riga lies near the sea, the lowering of the water table in the Arukilas-Amatas multi-aquifer system hasn't induced an intensive intrusion of sea water. This process has only been observed in some areas, where the intrusion has occurred through the bed of the River Daugava, where the Plavinas aquifer dolomites are situated.

This study is supported by the European Social Fund project
No. 2009/0212/1DP/1.1.1.2.0/09/APIA/VIAA/060.

APPLICATION OF STABLE ISOTOPE CONTENT IN GROUNDWATER TO VALIDATE THE RESULTS OF THE HYDROGEOLOGICAL MODEL OF THE BALTIC ARTESIAN BASIN

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Groundwater levels and their fluctuations are most commonly used to calibrate the developed numerical groundwater models. However, environmental water isotopes and groundwater chemistry, especially, trace elements and conservative ions such as chlorine can also be used in this validation process. Isotope and geochemical data are mainly used to calibrate transient flow or mixing models backwards, for instance if paleoreconstructions are performed. Isotope or chemical tracers are used in a small scale surface or subsurface hydrological models. They can also be used in large scale models if a large set of data is obtained and different isotope ratios occur, thus different end members can be distinguished and mixing lines can be detected.

Theoretically, in a case of relatively stable groundwater and a constant climate regime, stable water isotope ratios as well as chlorine concentrations shouldn't change much along the flow paths to the groundwater discharge areas. If recharge occurred in the same conditions as observed nowadays, then oxygen and hydrogen stable isotope composition should be more or less around the mean values in precipitation. Again, the TDS values should increase from recharge to discharge due to more or less intensive groundwater/rock interaction. Hydraulic connection thus mixing between deeper and shallower aquifers as well as in the case of interaction between aquifer and surface water bodies can change the chemical as well as the isotopic values, although if such interaction is significant in well built numerical models, it should be apparent.

In this study we tried to apply isotopes and major ion chemistry to verify the response of the developed and calibrated Baltic Artesian Basin numerical model. The developed large scale steady state model has a total area of approximately 480,000 km^3.

The stable isotope ratios from more than 200 samples covered a large range and no consequences could be observed when the chemical or isotopic data was treated alone. Due to this, smaller places for model verification were chosen and in those areas, where hydrogeological conditions were previously investigated with high accuracy. One of the chosen sites was the Upper Devonian Gauja aquifer in the Riga district. The Quaternary and Middle Devonian Pernava aquifers were other study areas analyzed in almost all of the territory of the Baltic Artesian Basin where new data was collected or old data was available.

Modelled and observed piezometric levels in most aquifers above the regional aquitard which consists of Middle Devonian Narva clayey sediments are very close and the main flow paths are almost equal. Despite a fine matching level above the Narva aquitard, below it, neither levels nor flow paths were modelled to a satisfactory level. In some cases piezometric level mismatch can increase up to 80 m. In these aquifers where modelled piezometric levels differ, such a great distribution of isotopic and chemical values can't be explained with the developed model.

The great mismatch in the model can be explained in two controversial ways. One is that the problem is in the model, which can be explained by an inaccurate model structure or the incorrectly defined permeability properties of materials. Another problem could be the remains of the Quaternary evolution, which with the rapid climate changes and hydrogeological conditions during the Quaternary period may still have an impact on subsurface hydrology nowadays.

If we assume that the model was built properly and that the structure calculates piezometric levels very well in aquifers with faster water exchange, it is more likely that the problem occurred due to not taking into account the evolution of groundwater hydrology during the Quaternary period, especially the Late Pleistocene and the Holocene.

The present work has been funded by the European Social Fund Project "Establishment of interdisciplinary scientist group and modelling system for groundwater research" (Project Nr. 2009/0212/1DP/1.1.1.2.0/09/APIA/VIAA/060).

THE ANALYSIS OF GROUNDWATER QUALITY PROBLEMS IN BALTIC SEA REGION COUNTRIES

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Groundwater resource quality and availability is a world wide problem and research field. Groundwater is the largest reservoir of freshwater in countries in the the Baltic Sea region. One of the current problems in the region is agricultural activity. The use of nitrogen fertilizers in agriculture is one of the main factors contributing to the contamination of surface water and groundwater, from various types of nitrogen.

The aim of the study presented is to analyze the current agricultural impact on groundwater quality in the Baltic Sea region and to give guidance and recommendations for better international cooperation based on EU directives. The main task is to compare the current situation of groundwater quality in three groups of Baltic Sea region countries and to define development scenarios.

The study presents general information about groundwater quality in the following Baltic Sea region countries: Denmark, Sweden, Finland, Estonia, Latvia, Lithuania, Poland and Germany. The countries from the region are divided into three groups based on similar geographical, agro climatic and agro historical conditions and applied methods. The first group is the Baltic States – Estonia, Latvia and Lithuania, the second is the Northern European countries – Finland and Sweden, and the third group is Denmark, Germany and Poland.

The major contamination in the Baltic Sea region is from agricultural chemicals, for example, fertilizers (nitrate, phosphorus) and pesticides. The role of diffuse source pollution in agriculture is a common problem in the region and is solved in a different way in each country. All of the countries in the region, except Russia, are subject to European Union (EU) groundwater directive restrictions. European Environmental Agency (EEA) EUROWATERNET data bases are available for a better understanding of the quality of groundwater and its contamination in the region.

In the research, the present situation is analyzed and advice and conclusions have been provided for future groundwater quality scenarios for the Baltic Sea region. A monitoring system has been established in all Baltic region countries in accordance with EU directives to determine and forecast groundwater quality.

This study is supported by the European Social Fund project “Establishment of interdisciplinary scientists group and modelling system for groundwater research”. No. 2009/0212/1DP/1.1.1.2.0/09/APIA/VIAA/060.

THE VISUALISATION OF GROUNDWATER CHEMICAL COMPOSITION USING THE RGB SCALE. AN EXAMPLE FROM THE D12 AQUIFER, LATVIA

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Groundwater composition is traditionally visualised using the Piper diagram where the relative concentration of all major inorganic components are plotted in ternary diagrams. This is a convenient approach to identify different water types or, for example, to study the evolution of water composition along the flow lines. However, it is not easy to examine more complex spatial variations of water composition using ternary diagrams.

The ternary diagrams where the relative concentration of anions (or cations) is plotted are remarkably similar to the RGB colour scale which is usually used for colour coding in colour photography. In digital images, the RGB colour scale is composed of three bands – red, green and blue (RGB) – where simple numbers indicate the relative intensity of each of the bands. The colour values in the RGB scale can be substituted by the relative concentrations of the three desired chemical components, thus allowing the visualisation of three parameters with a single colour code. The conversion of relative concentrations of the dominant anions to RGB colour is straightforward: calculate the proportion of each anion (e.g. in %) and convert theses values to the respective colour intensities in the RGB scale. Thus, the spatial variation of concentrations of the three chemical components can be visualised with a colour code on the map or a cross section.

But there is more: the absolute concentration values can be converted to the RGB colour values, thus representing the absolute and not only the relative concentrations of given chemical compounds. To do this, arbitrary minimum and maximum concentrations of all desired compounds need to be defined. The minimum value in most cases obviously will be 0, but the maximum value possibly needs to be specified for each compound and for each case individually. For example, the HCO_3^- concentration rarely exceeds 8 mmol/l in groundwater, while the Cl^- can exceed 2,500 mmol/l. Actually the problem of defining the conversion criterions is a secondary one. The primary problem is how to visualise the scale for colour coding where the summary colour intensity (the sum of all three colour band values) is the fourth parameter or dimension, besides the relative values of three colour bands.

The most significant disadvantage of the presented colour coding scheme is that the ability to distinguish colour is not the same for everyone. The ability of a person to discriminate between similar colours depends on the person's physical abilities and even from the person's cultural background.

In the territory of Latvia, the lower-middle Devonian aquifer system, confined by the Narva formation's regional aquiclude, hosts fresh, HCO_3^- dominated water in the NE of the territory and Cl^- and SO_4^{2-} dominated water with downward increasing Cl^- concentration in the rest of the territory. Cl dominated brine intrusions from below, freshwater infiltration from above and dissolution of gypsum that is often found as an accessory mineral are dominant factors controlling groundwater composition in the horizon. Thus, the lower and middle Devonian sub-Narva water horizon complex is a good example for using the RGB scale to visualise the water composition and in identifying the distribution of different water types.

This study is supported by the European Social Fund project
No. 2009/0212/1DP/1.1.1.2.0/09/APIA/VIAA/060.

CREDIBILITY CRITERIA OF THE RESULTS OF UNDERGROUND WATER ANALYSIS

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The wells data base compiled by the LVGMC includes the results of several thousand groundwater sample analyses made as early as the 1960's. As the data has been accumulated during a period of more than 50 years, which includes progress in analytical technologies and turmoil in the political system, the credibility of data needs to be evaluated before it can be used for scientific research.

Infiltration of precipitation water is the force moving groundwater flow, while the shape of the flow is determined by geological structures and earth surface elevation. As none of these factors has changed significantly during the last 10,000 years since the end of the ice age (Mokrik and Mazeika, 2002), it can be concluded that the configuration of groundwater flow has not substantially changed and the dynamic equilibrium of water composition has been maintained. The steady groundwater flow means that any systematic changes in composition will happen gradually over a long period of time, determined by gradual disarrangement of flow patterns smoothed out by flow dispersion and diffusion (Ingebritsen *et al.*, 2008). The measured variations in groundwater composition will have the character of an unintentional fluctuation which could even be induced by perturbations created during water sample collection. An exception is regions with intensive water extraction such as the surroundings of Riga and Jelgava, where during the 1960's, 1970's and 1980's huge depressions in the groundwater level developed (Levina and Levins, 2005). But, even here gradual rather than abrupt groundwater composition changes will be taking place.

In order to identify analytical mistakes, we suggest the use of total validation criterion (TVC) which is calculated by dividing the concentration of Ca^{2+} ion (mgekv/l) with the sum of the concentration of anions (mgekv/l). The TVC is calculated from the only chemical components that were determined directly, even in the early days of systematic groundwater composition exploration, that is HCO_3^-, SO_4^{2-}, Cl^- and Ca^{2+}. The rest of the major components – Na^+, K^+, Mg^{2+} – according to GOST standards, were calculated from the ion balance (Na^+ and K^+) or from indirect measurements (Mg^{2+}).

The TVC can be used to validate long term series of groundwater monitoring data in observation points where short term variations caused by climatic and meteorological or human factors can be excluded. Divergences from the general trend at each monitoring spot should be considered bad measurements and dismissed (Fig. 1.).

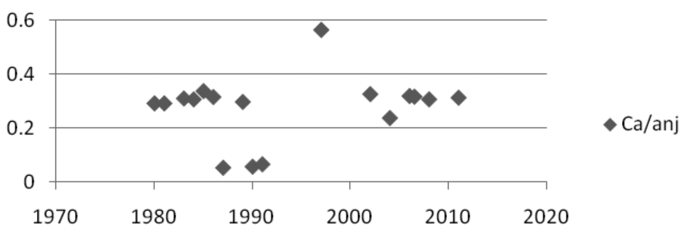

Fig. 1. TVC value of monitoring well No. 1508 at Baldone. There is no feasible geological explanation for the four divergences from the general trend and these analyses should be regarded as wrong.

In order to identify sample spoiling during storage due to freezing or aeration which can result in calcium carbonate precipitation, the results need to be checked for consistency of concentration of Ca^{2+} and HCO_3^- ion concentrations in (mgekv/l). If the changes are severe and correlate for both ions, then the results should be considered as a bad mistake. It's possible that TVC won't spot this deviation (Fig. 2. and 3.).

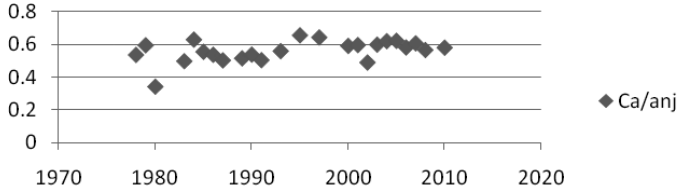

Fig. 2. TVC value of monitoring well No. 1492 at Inčukalns. Only one bad measurement can be identified using TVC.

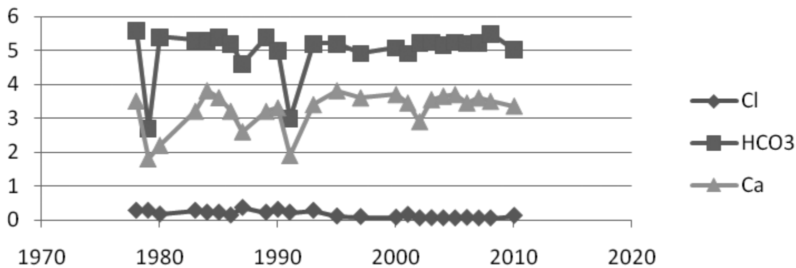

Fig. 3. Cl^-, HCO_3^- and Ca^{2+} concentration (mg/L) at monitoring well No. 1492 at Inčukalns. Two coherent drops in the Ca2+ and HCO3- concentrations suggest that CaCO3 precipitated from the sample during the storage possibly due to sample freezing or loss of CO_2 gas.

We speculate that the proportion of incorrect analyses in the data base are similar for the monitoring wells where long term series of observations are available and other well types where only one or a few groundwater samples have been collected and analysed. If this is so, the overall quality of the groundwater composition data base can be quantified and considered when performing statistical or individual analyses of water composition data.

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TRACE ELEMENTS IN GROUNDWATER IN LATVIA: EXISTING DATA AND FIRST NEW RESULTS

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The territory of Latvia is a part of the Baltic Artesian (Sedimentary) Basin which can be subdivided into three major water exchange zones: freshwater (active water exchange), saline (delayed exchange), and brine (stagnant) hydrogeological zones, in considering the water chemistry and intensity of water connection between aquifers. The occurrence of trace elements in groundwater can be due to natural sources such as dilution of water bearing rocks, surface impact or anthropogenic influence.

An extensive set of data about groundwater in Latvia is available from the beginning of the 1960's and contains mainly information about groundwater levels, major ion chemistry and physical parameters. It is impossible to test the quality of the old data on trace element concentration from geological mapping and hydrogeological exploration during Soviet times. The more recent studies contain mainly data about the Quaternary sedimentary aquifer and are limited to the active water exchange zone. A lack of available trace element data in the deeper strata led to the implementation of this study.

The aim of this study is to determine the distribution and sources of trace elements in groundwater in Latvia and compare the results with the major ion chemistry data and WHO and EU potable water standards. At the end of this study there will be new data about approximately two hundred groundwater samples. Groundwater samples from monitoring and supply wells, as well as boreholes and springs, were analyzed by total x-ray fluorescence (TXRF) and atomic absorption spectroscopy (AAS) techniques to determine the concentration of trace and some major elements. The contents of cations and anions, *pH*, electrical conductivity (EC), redox potential (ORP), TDS and dissolved oxygen were analysed to assess the quality of groundwater.

Generally, the concentration of trace elements in uncontaminated shallow groundwater samples is bellow WHO and EU potable water standards. Previous studies suggest that uranium, arsenic, cobalt, and copper in groundwater can often be derived from agricultural fertilizers and due to the high flux of nitrate present in infiltrating water, some metals can be released from deposits (Gosk *et al.*, 2006). The influence of the lithology of aquifer deposits on concentrations of trace elements is statistically significant only in cases where aquifer deposits are rich in organic matter or contain well-soluble minerals. Some exceeding trace element concentrations are associated with gypsum dissolution in shallow groundwater samples. Studies show that the concentration of barium, iron, lithium and strontium increases with increasing residence time and the confinement degree of an aquifer (Levins and Gosk, 2007).

Due to incomplete studies, it is essential to determine the trace element baseline values in Latvian confined aquifers, to avoid migration of pollutants to lower aquifers.

This study is supported by the European Social Fund Project
No. 2009/0212/1DP/1.1.1.2.0/09/APIA/VIAA/060.

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RECESSION CURVE ANALYSIS APPROACH FOR GROUNDWATER

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Recession curve analysis is a powerful and effective analysis technique in many research areas related to hydrogeology, where observations have to be made, such as water filtration and absorption of moisture, irrigation and drainage, planning of hydroelectric power production and chemical leaching (elution of chemical substances), as well as in other areas.

Upward areas were cut out from the initial curve within the process of decreased recession curve analysis, leaving only the drops of the curve. Consequently, the curve is transformed more closely to the groundwater flow, in an attempt to remove the impact of rain or drought periods from the curve. Respectively, the drop-down curve is part of the data, collected with a hydrograph, where data with the discharge dominates, without considering the impact of precipitation.

There are manually prepared hydrographs for the analysis of recession curves for observation wells (MG2, BG2 and AG1) in agricultural monitoring sites in Latvia. Data of declining periods, split by month, was extracted within this study from the available monitored data of groundwater levels. The drop-down curves were manually (by changing the date) moved together to find the best match, thereby obtaining monthly drop-down curves, representing each month separately. Monthly curves were combined and manually joined to obtain characteristic drop-down curves for the year for each well. The mathematical model of data equalization was used for displaying data, finding the corresponding or closest logarithmic function of the recession for the graph. Using recession curve analysis theory, a readymade tool – "A Visual Basic Spreadsheet Macro for Recession Curve Analysis" (Posavec *et al.*, 2006) was also used to prepare similar curves with superior accuracy in the selection of data and matching of logarithmic functions, than the functions which were developed by the manual processing of data. The recession curves obtained were similar but not identical.

With a full knowledge of the fluctuations of ground water levels, it is possible to indirectly (without taking soil samples) determine the filtration coefficient: a more rapid decline in the recession curve corresponds to better filtration conditions. This research could be very useful in construction planning, road constructions, agriculture etc.

The authors gratefully acknowledge the funding from the ESF Project "Establishment of interdisciplinary scientist group and modelling system for groundwater research" (Agreement No. 2009/0212/1DP/1.1.1.2.0/09/APIA/VIAA/060).

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INFLUENCE OF WATER ABSTRACTION ON GROUNDWATER FLOW IN THE BAB

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There have been 3 different scenarios of groundwater abstraction trends in the region of the Baltic Artesian Basin starting from 1950 until the present day:

1. Natural scenario of groundwater abstraction – minimal water extraction, almost unnoticeable compared to natural processes of water circulation;
2. Wasteful scenario of groundwater abstraction – significant water usage leading to reveal influence to the groundwater resources;
3. Medium scenario of groundwater abstraction – reasonable water extraction, minimal influence to groundwater resources, maintaining natural groundwater resource restoration.

The groundwater extraction trend in all the Baltic countries is similar – starting from 1950 until about 1965, the representative trend was minimal water extraction (natural scenario of groundwater extraction); from 1965 until 1990, the characteristic trend was an increasing amount of groundwater extraction, which led to the wasteful scenario of groundwater extraction at the end of the given time period; after 1990, a rapid decrease in the amount of extracted groundwater was observed, referring to the medium scenario of groundwater extraction which exists presently.

The groundwater extraction sources have a non-homogeneous distribution throughout the Baltic Artesian Basin, where most of the wells are concentrated around the biggest cities and industrial regions (in the territory of Latvia this means the largest cities: Riga, Liepaja, Daugavpils, Jurmala etc.). The spatial localization of groundwater wells leads to vulnerability in local groundwater resources, as well as a limitation on groundwater resource usage.

Stationary calculations using the BAB version 1 hydrogeological model shows that increased groundwater extraction causes areas of depression. In Latvia, the areas of depression are observed in aquifers D3 gj, D3 am, and D2 br near Riga, as well as in the Cambrian aquifer in Estonia near Tallinn and are cases of the medium and wasteful scenario of groundwater abstraction.

The preferred future mode of groundwater extraction should maintain the balance between the extraction and restoration of groundwater resources. In cases where an increase in the volume of groundwater extraction is necessary, one must consider the influence on the potentially available groundwater resources.

This study is supported by the European Social Fund Project
No. 2009/0212/1DP/1.1.1.2.0/09/APIA/VIAA/060.

USEFUL STATISTICS FOR DESCRIBING HYDRAULIC CONDUCTIVITY OF THE QUATERNARY STRATA FROM THE LATVIAN BOREHOLE LOG DATA

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Borehole log data in the territory of Latvia is highly variable in quality, spatial distribution and accuracy of georeferencing, making direct comparisons between different boreholes difficult. (It is, however, useful to compile some global statistics describing the broader geological characteristics present in the Quaternary lithology.

The regional groundwater modelling system for the Baltic Artesian Basin – MOSYS V1 required hydraulic conductivities for the Quaternary strata. To derive this, we started by looking at the ways to compare the quality of the borehole log data available. To make the log structures comparable in a universal form, useful for finding structural patterns, a range normalization for all layer depth entries in all boreholes was performed. This range-normalized borehole log data is used to generate 1000 bin histograms for boreholes that have at least 2 layers in the Quaternary part of the log data. A 1000 bin histogram was selected because 1 unit of normalized depth represents, on average, ~5 cm of the log depth in the Quaternary sequence, thus providing an adequate precision for this analysis as well as allowing for an amount of "noise" in the log data precision.

For generalization of the lithology structure in the Quaternary sediments, a simplified 4 layer model was used in MOSYS V1. This was based on the results of generalized aquifer-aquiclude transition statistics of the borehole logs as inferred from the lithology. There are two such transitions for 88% of boreholes in Latvia, suggesting that a simplified yet representative version of the Quaternary sequence can be made out of 4 layers – 2 aquitards and 2 aquifers respectively.

To further improve this model, another set of layer transition counts were performed. Using the lithology classifier for aggregation of lithological codes into more general groups of sedimentary rock types, a global count of all transitions between rock type classes was performed using the SQL database language. The most common layer transitions are sand-loam, loam-sand and sand-silt, siltstone.

These results have been generated for use in different experiments during our work on the development of a regional groundwater model. The normalized layer transition histograms show that some transitions are particularly numerous, such as the 0.500, 0.333 and 0.250 normalized depth transitions in the Quaternary sediments, pointing perhaps towards a requirement for borehole log quality control. The aquifer-aquiclude counts allow for a coarse generalization of hydraulic conductivity in a simplified 4 layer model of the Quaternary strata. The layer transition counts for aggregate lithology types served as a reference for implementing a sequence based interpolation of inter-cluster boundary transition gradients for spatial clusters of similar lithological structure in the Quaternary part of the borehole logs. This information has been gradually complied during the PUMa project. It may, however, be possible for other studies to benefit directly from these results.

This study is supported by the European Social Fund Project
No. 2009/0212/1DP/1.1.1.2.0/09/APIA/VIAA/060.

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ENVIRONMENTAL SITUATION IN THE AREA AROUND INČUKALNS PONDS AND THREATS TO GROUNDWATER

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The Southern and Northern acid tar ponds at Inčukalns are historical contaminated sites located 30-35 km from Riga. During the period from 1950-1980, acid tar was generated as waste from the production of medical and perfumery oil. With a disregard for environmental protection measures, acid tar and other chemical waste was dumped in sandy pits in this forested area. The dump site was closed in 1986.

The geological conditions in Latvia are such that the surficial sediment layers often have good filtration characteristics, meaning that potential contamination can migrate to the deeper horizons of groundwater (artesian waters). This means that surface waters as well as soils must be protected from different kinds of pollution. Studies and comparisons in the Inčukalns area are slightly complicated because of the many factors influencing groundwater. Therefore, monitoring has been done here more or less regularly for at least a decade. Groundwater analysis has been conducted mostly using the methodological approaches used in geology, not surface water analysis. The results from monitoring show that the area around both acid tar ponds – the Northern and Southern are contaminated at both levels – the upper groundwater and the deeper Gauja horizon. The complexity arises because of the differentiation of the contamination layers. In addition, the parameters for the upper groundwater and the deeper Gauja horizon are not completely understood. The contamination plumes are migrating in the horizontal as well as the vertical plane, and thus the polluting substances from the Northern and Southern ponds are getting closer to the Gauja River and also threating the groundwater resources of the Riga City. Considerable contamination in the permeable and percolation sandy layers has reached groundwater and artesian waters at a depth of 70-90 m by way of infiltration.

In 2010, a remediation project for the Inčukalns acid tar ponds began, the main task being to prevent contamination of the territories next to the acid tar ponds. Project implementation tasks will include treatment with lime and the replacement, excavation and disposal to landfill (with or without treatment) and un-engineered capping, as well as the pumping out of contaminated water, then its treatment and injection back into the subterranean level in order to stop the mobility of the contaminated plume. The sandy layers, in which the acid tar is located, are permeable and have good filtration properties. This means that there is a potential hazard to be dealt with in the next 25 years, if the movement of the contamination plume in the groundwater is not stopped before the Gauja River. The main idea is to stop the plume by pumping, then treating and injecting the groundwater back into the subterranean. Environmental impact and monitoring should be undertaken using previous knowledge from other case studies. Air emissions, as well as the behaviour of acid tar and its ingredients in soil and groundwater during excavation and neutralization works, must be strongly taken in account. Legislation and funding should be considered in the planning of any remediation activities. Emissions and residuals during the works must also be strongly controlled.

The opportunity to collaborate within the EU remediation framework of historically contaminated territories provides an opportunity to carry out environmental research and follow-up with remediation projects in problematic areas. The remediation in Inčukalns will demand a multidisciplinary approach in order to have a successful result. The acid tar in the ponds and in the soil has complicated chemical properties and therefore, similar case studies on acid tar lagoon remediation should be used in the planning of such activities in the Republic of Latvia.

THE DEVELOPMENT TRENDS OF MŪRU-ŽAGARES AND JONIŠĶI-AKMENES GROUNDWATER HORIZON SURFACE DEPRESSION AND SEA WATER INTRUSION IMPACT IN LIEPĀJA CITY

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In ancient times, Quaternary groundwater was used as the source of water supply in Liepāja City. It was only in the middle of the 19th century, that water supply wells were first drilled deeper – into bedrock. The Žagare horizon of the Upper Devonian ($D_{3\ žg}$) was mostly exploited and a more extensive use of it commenced in the 1930's and 1940's. At this time Liepāja's industrial development was increasing, the volume of exploited water resources was growing and the first reports were appearing on the development of sea water intrusion, which was percolating the horizons of artesian groundwater. Laboratory tests showed that water quality diminished due to this and no longer met the standard for drinking water.

Hydrogeological mapping on a scale of 1:100 000 was done for Liepāja City and surroundings in 1947, and the authors insisted that only the Naujoji Akmene-Middle Ketleri and Mūru-Žagare artesian horizons of the Upper Devonian could be used as a useful source of drinking water. It was recommended that a system of drinking water extraction wells be developed to the east of the Liepāja Lake between Grobiņa and

Otaņķi. The first two experimental research-exploitation wells were drilled in this area in 1953, with the depths of these wells being 102.8 m and 117.0 m respectively. Research was carried out on water supply needs in the artesian horizons of Lower Carboniferous and Upper Devonian (Žagare Formation). Further analysis showed that the capacity of these horizons was sufficient for use as the water supply for Liepāja City. Two additional drinking water wells were drilled in 1959 in Otaņķi, with their construction being the same as for the previous two. Water quality results in 1960 already showed that the content of dry matter and chlorides in the water horizons of the Lower Carboniferous and Upper Devonian Žagare Formation in the centre of Liepāja City were 2-3 g/l and 1.6 g/l respectively.

In the period from 1960-1970, the size of the groundwater horizon surface depression in the centre of Liepāja City grew because of the increasing intensity of water extraction. Thus, during that time, this surface depression had already started to fulfil a particular protective function from the intensive sea water intrusion into the Otaņķi water supply prospect. In order to improve the situation in the water supply to Liepāja City, several research-exploitation wells were drilled in the Aistere prospect. During the 1983-1985 research in Liepāja City, it was observed that the piezometric surfaces of the Upper Devonian Mūru-Žagares and Jonišķi-Kursas groundwater horizons were almost the same (around -7 m). Both of these horizons are separated by up to 20 m thick sedimentary rocks of the Akmene Formation with low filtration properties.

The intensive and long-term exploitation of the Upper Devonian Mūru-Žagare groundwater horizon in Liepāja City and surroundings has caused, and further developed, a complicated hydrodynamic and hydrochemical situation: the intrusion of sea water (enriched with chlorides) and the shifting of the lower situated Eleja-Pļaviņas water horizons (with sulphates). As a result of the mentioned obstacles, the concentration of chlorides in the groundwater of Mūru-Žagare horizon had already risen, in 1944, from 10-20 mg/l to 245 mg/l, but the concentration of sulphates averagely from 100 mg/l to 200 mg/l. The piezometric surface of this horizon in 1944 was 2-3 m, while ten years later it was 3-4 m below sea level, but the concentration of chlorides in wells rose up to 600 mg/l. From 1976 it was observed that the eastern part of the sea water (and chloride) intrusion zone had started to move in the direction of the groundwater prospect Otaņķi, with the main reason for this process being the intensive exploitation of wells in this prospect.

The lowest levels of the piezometric surface in Liepāja City were observed from 1985-1990 (-14 m), when the exploitation of the Mūru-Žagare horizon was the most intense. If we compare the development trends of the surface depression in Liepāja City territory and Otaņķi groundwater prospect, according data from last year from "Liepājas ūdens" Ltd., static groundwater level in Otaņķi prospect for the Mūru-Žagares horizon surface is systematically lowering and in 2008 reached -9.0 to -16.5 m. At the beginning of the exploitation of the Otaņķi prospect from 1961-1976, the piezometric surface of the Mūru-Žagares horizon was fixed at a level of -5.0 to -5.5 m.

The lowering of the piezometric surface of the Mūru-Žagares groundwater horizon is the reason for the increase in the concentration of chlorides in water. The maximum concentration (up to 2000 mg/l) can be observed in wells in Liepāja City, which is the centre of the depression, but the results of groundwater chemical analysis from observation wells in Liepāja City (DB 2642, Zemnieku St. 28) show a chloride concentration of up to 1200 mg/l.

The results from the "Liepājas ūdens" Ltd. central testing laboratory for the period 2007-2011 from Otaņķi groundwater prospect are showing chloride concentrations, which do not exceed water quality norms (250 mg/l). A problematic issue is that, according to some evaluations, sea water intrusion is shifting frontally with a speed of 50 m/yr. A possible solution to this problem could be more intensive pumping in the Liepāja City area in order to generate an artificially bigger surface depression for the groundwater surface in order to stop migration of the sea water intrusion further inland, for example, to Otaņķi prospect.

ADDITIONAL DATA ON THE CFC CONCENTRATION AND CORRESPONDING GROUND WATER AGE IN THE FRESH GROUNDWATER OF LATVIA

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One of the main issues in groundwater studies is groundwater age, also known as residence time. This is important for investigating the groundwater filtration rate and to solve various issues such as groundwater use, management and protection.

The concentration of tritium will be determined in about 60 samples, but the CFC concentration has now been analyzed in 39 samples – 19 samples in year 2010 and 20 new samples in 2011 (Fig. 1.). Previous studies have shown that the Latvian CFC method is appropriate for aquifers to an average depth of 30-50 m (Gosk *et al.*, 2006). Therefore, new samples for the year 2011 were taken from an average depth of 37 m, although the depth varies from 6 m to 128 m and one sample was taken from surface water (Baltezers basin). CFCs concentrations were analyzed in the laboratory at GEUS after Busenberg and Plummer (Busenberg and Plummer, 1992) described a method using gas chromatography equipped with an EDC detector. Interpretation of the results was carried out by laboratory expert, Troels Laier. Many samples were taken from one place at different depths to obtain a better view of residence time distribution and these new samples from 20 wells were located in 8 places (Fig. 1.).

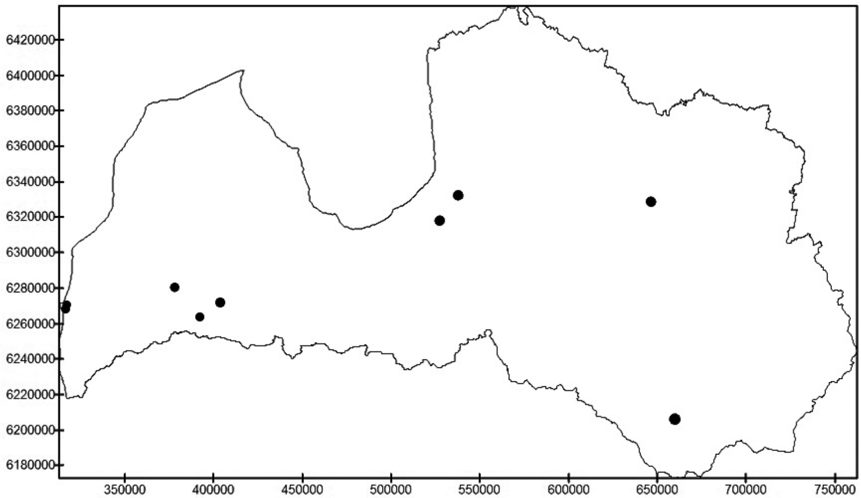

Fig. 1. Sites of new samples for CFC analysis.

New samples were taken both from the unconfined aquifer and the first confined aquifer. The sampling interval for CFC analysis varies from 6-14 m up to 108-128 m. It was found that most of the groundwater determined from both – the confined and the unconfined aquifer – had a residence time of 35-60 years (Fig. 2.).

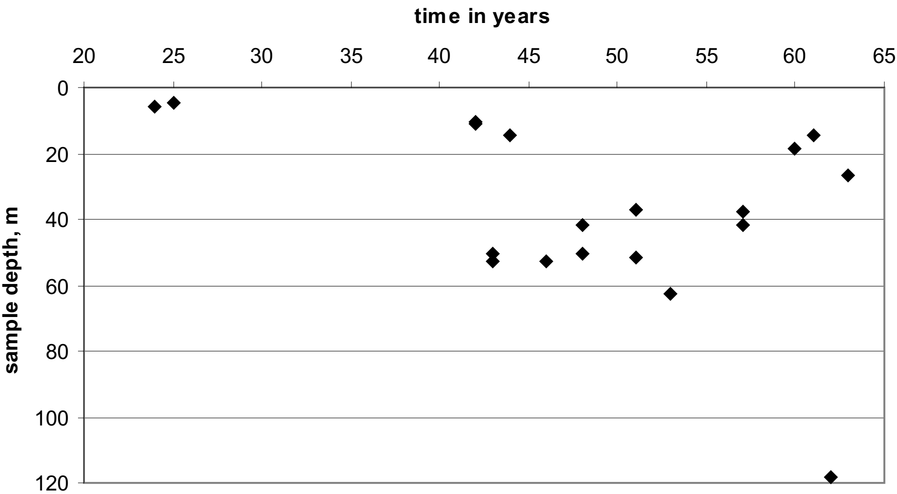

Fig. 2. Water residence time at different depths.

An interesting situation was found at the Ēvarži wells, where water residence time at a depth of 24 – 29 m was ~63 years but at a depth of 48 – 53 m, the water age was 43 years (the distance between these 2 wells is only 10 meters). In such a situation, the major role in water age distribution is from water horizontal flow.

This study is supported by the European Social Fund project
No. 2009/0212/1DP/1.1.1.2.0/09/APIA/VIAA/060.

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GROUNDWATER ABSTRACTION DYNAMICS IN THE BALTIC ARTESIAN BASIN

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The Baltic Artesian Basin consists of a number of multi-aquifer systems that contain fresh groundwater. The fresh groundwater is the main drinking water source for the centralised and decentralised water supply in Estonia, Latvia and Lithuania. The fresh groundwater resources in the Baltic Artesian Basin are pretty large, roughly about 4.1 million m^3, which is 10-15 times more than is abstracted for drinking water supply needs (Jodkazis, 1980). However, the groundwater abstraction is distributed unevenly spatially and in time. In order to model groundwater flow in the transient conditions, one has to know the changes in groundwater abstraction in time and space.

The main task of the study was to provide knowledge on groundwater abstraction dynamics in order to develop different scenarios for the model calculations. The data on groundwater abstraction was collected from the geological surveys of Estonia and Lithuania and the Latvian Environment, Geology and Meteorology Centre.

Groundwater has already been used in water supply for several centuries, taking water from springs or from shallow dug wells. The first deeper wells drilled for groundwater abstraction were already installed at the end of the 19th century, but these were just a few, and did not cause any impact on groundwater resources. With the development of drilling techniques, the number of wells and abstracted water amount increased continuously. The most wells were drilled from 1955-1970's. For example, during this period 10-30 wells were installed in Riga every year, and the amount of abstracted groundwater increased to 5,000-6,000 m³/day per year. The development of a centralised water supply was typical for this period, and a lot of well fields for public water supply were installed. The maximum water abstraction was reached at the end of the 1970's – beginning of the 1980's after which more or less stable water abstraction continued until the beginning of the 1990's. With the collapse of industry, water abstraction dropped sharply and the volume of abstracted water decreased about two times. Today, water abstraction has increased compared to the 1990's, but below the overexploitation during the 1970-1980's.

It is typical that the amount of water abstracted for the centralised and decentralised water supply is rather similar, a little more for the centralised water supply, but the number of wells is 5–10 times more in the decentralised water supply. For the model calculations, this means that it is most important to take into account the centralised water supply, where a large amount of water abstraction is concentrated in one spot (cell), but the individual locations of decentralised water supply wells could be omitted for the regional studies, because the yield of each well is very low (100-200 m³/day in average). The only exception is large industrial enterprises having local semi-centralised water supply systems.

For model calculations, it is assumed that the natural conditions could be assumed for the time period before year 1950, which is followed by the disturbed groundwater regime due to the overexploitation of the groundwater resources until 1992, and the change back to semi-natural conditions after the year 2000.

This study is supported by the European Social Fund Project
No. 2009/0212/1DP/1.1.1.2.0/09/APIA/VIAA/060

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RECONSTRUCTING THE CALEDONIAN STRUCTURAL COMPLEX DEFORMATION THROUGH THICKNESS ANALYSIS

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In this study we attempt to interpret the deformation that occurred in the Early Paleozoic time, known as the Caledonian tectonic event, which compared to other tectonic events had the most significant impact. Theoretical material was collected and analyzed from previous studies during this research, and a considerable amount of published material was gathered about tectonic structures within the territory of Latvia. Based on this data, a rough resolution 3D geological tectonic block model was developed. Although geophysical research has previously been carried out by Geological Survey, the studies offer insight of structures in general, but do not determine their kinematics or possible evolution, except for some local scale structures (Brangulis and Kaņevs, 2002).

To reach the goal, the *MOSYS* modelling system which was developed within the PUMA project was used for the geological structure modelling (Seņņikovs *et al.*, 2011). An algorithmic genetic approach was applied to interpolate the data of well logs as strata volume and to sequentially reconstruct the post-deformation situation. This approach allows for the modification of the model construction at any step and all processes are fully documented and are repeatable. The geometrical model consists of 33 tectonic blocks bordered by the faults, which were distributed by interpreting the displacement volume of the blocks along the faults, providing an opportunity to characterize the common tectonic evolution.

Thickness analysis was performed to determine the tectonic events which led to defined uplift or sinking events, by comparing the volume in each tectonic block and the eroded amount of strata. This method showed that the smallest strata thickness change was in the Ordovician and Llandovery, Wenlock epochs of the Silurian but the largest changes of strata thickness and erosion occurred at the end of the Silurian and in the Early Devonian. Both thickness changes and erosion show that processes occurred very rapidly and that there was a major compression event at the Caledonian orogeny and should be linked to *Scandian* orogeny.

The methodology applied allowed us to reach good model strata surfaces and compatibility of well logs which was no larger than 3 meters and indicated that the model is reliable for strata thickness analyses. The constructed model is respective for the studied region, which is confirmed by the analysis that strata thickness is well maintained and strata thickness does not vary radically in each tectonic block. Although research data is sparse, the dip angles of fault structures are unknown and the fault planes in the model are vertical, it still indicates major characteristics of deformation and strata thickness changes which, as a preliminary study in tectonic evolution, is enough to draw the first major conclusions.

This Project was supported by ESF project "Establishment of interdisciplinary scientist group and modelling system for Groundwater research"
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BAB V1 GEOMETRICAL MODEL: INTEGRATING HETEROGENEOUS AND UNEVEN DENSITY DATA INTO A 3D GEOLOGICAL MODEL

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Although specialized tools allow one to model complex geological bodies in 3D using geological maps, survey records and borehole data, the building of a viable geological model is still a challenge. One of the main difficulties in 3D reconstructions lies in the heterogeneity of the data and its interpretation, where there is a need for accuracy, representation at the scale of interest and reliability.

A 3D regional geological model was created for the Baltic Artesian Basin (BB) – for modelling the groundwater flow. A large volume of geological data describing the geological structure of the BB was available; however, the data coverage is very uneven.

In previous studies a number of problems have been solved associated with the collection, harmonization and post-production of cartographic data in different resolutions and various formats, which includes the control of various data input, generalization and topological issues (Dēliņa *et al.*, 2011). Mathematical algorithms have also been created that consider the priority, importance and plausibility of each data source in integrating topography and lithology data as well as borehole data (Seņņikovs *et al.*, 2011). However, there is a need to use low resolution data and interpretations from information in the literature for certain areas, making geological generalizations and interpretations that are based on knowledge about the geological evolution of the territory.

Geological reconstruction is subordinated to geological preconditions, where structure generation is based on an assumption that post-depositional deformation produces no significant change in the sedimentary strata volume – strata thickness and its length in a cross sectional plane remains unchanged, except as a result of erosion (Dahlstrom, 1969). In the case of the BB, tectonic deformation occurred in sequential cycles where subsequent tectonic stage strata was separated by regional unconformities (Brangulis and Kanevs, 2002), providing an opportunity for an algorithmic approach in the reconstruction of these conditions for the whole BB territory.

The methodology for the model reconstruction includes several steps, including 3D reconstruction of the structural surfaces with known tectonic structures, determination and reconstruction of the unconformities, which together with structural surfaces form an ensemble of base surfaces which are further used to reconstruct sedimentary layer distribution and thickness variations.

The 3D reconstruction of the base surfaces: known tectonic structures and amount of slip along the faults and unconformities, are reconstructed using all available data after considering the priority of each data source. For areas without data, surfaces are reconstructed using extrapolation of thickness data between the base surfaces.

All depositional layers in the territory of Latvia and Estonia are reconstructed using initial thickness data from the boreholes, adding additional surface data from cartographic and published cross sections, while the thickness between the current and underlying surface is extrapolated to the territories for which there is no data.

The topography of each model layer was obtained by sequentially summing thickness to the initial base surfaces. Thereby, each layer reflects the topography and amount of slip along the fault of the underlying layer. An overlying tectonic cycle sequence is implemented into the model structure by using an unconformity surface as an initial reference surface.

Applied techniques made it possible to reliably reconstruct the 3D geological structure of the BB and allowed the prediction of the surface geometry of the layers in areas with sparse data. Modelling results allows the quantifying of areas in the model where additional data is necessary for geological reconstruction. The approach used has good potential for the development of regional geological models of sedimentary basins and is valid for the spatial interpretation of geological structures from heterogeneous and sparse data, subordinating this process to the prerequisites of geological evolution.

The present work has been funded by the European Social Fund Project "Establishment of interdisciplinary scientist group and modelling system for groundwater research" (Project Nr. 2009/0212/1DP/1.1.1.2.0/09/APIA/VIAA/060).

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THE SENSIBILITY ANALYSIS OF CL^- AND SO_4^{2-} TITRATION IN GROUNDWATER SAMPLES

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The dominant cations in groundwater are K^+, Na^+, Ca^{2+}, Mg^{2+} and the anions are HCO_3^-, Cl^- and SO_4^{2-}. The method for determining the concentration of anions used in the "Establishment of interdisciplinary scientist group and modelling system for groundwater research" Project is described here. The concentration of ions in groundwater can vary across several orders of magnitude, e.g. in the case of Cl^- from several mg/L to more than 100 g/L. The measurement of low concentrations of SO_4^{2-} and Cl^- can be particularly difficult.

The turbidimetric method is used to measure the concentration of the sulphate ion. By adding barium chloride, barium sulphate is obtained. $BaSO_4$ solubility is about 1 mg/L and it defines the determination limit of the method. If SO_4^{2-} concentration is lower than 5 mg/L, it cannot be measured, because in it the linear dependence of light absorption to SO_4^{2-} concentration is breaking down.

Cl^- concentration is measured using the argentometric method. Potassium chromate is used to indicate the end point of the silver nitrate titration of chloride (Eaton *et al.*, 2005). In measuring chloride with a low concentration, the solution colour changes later. The accuracy of both methods in a low concentration range was tested and procedures were developed to improve it.

Using test solutions it has been found that in the solutions with chloride concentration less than about 40 mg/L, the measured results are higher than the theoretical concentration known for the test solutions (Fig. 1.).

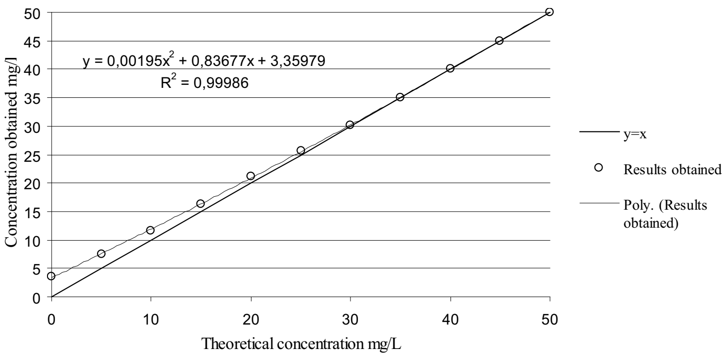

Fig. 1. Comparison of measured and theoretical Cl^- concentration.

Using mathematical regression the best fit curve can be found which is described by the equation:

$$\gamma_{Obt.} = 0,00195\gamma_{Cl} + 0,83677\gamma_{Cl} + 3,35979 \qquad (1)$$

Solving equation (1) the Cl^- concentration corrected value can be found by the equation:

$$\gamma_{Cl.} = \frac{-0,83677 + \sqrt{0,67397 + 0,0078\gamma_{\cdot Obt.}}}{0,0039} mg/L, \qquad (2)$$

where, $\gamma_{Obt.}$ – measured result and γ_{Cl} – corrected value, which should represent the reality more closely. Equation (2) can be used to correct the measurement accuracy if the Cl^- concentration is less than 35 mg/L.

Sulphates cannot be measured if the concentration is less than 5 mg/L, as there is no linear dependence of light absorption by the precipitated $BaSO_4$. It is possible to add Na_2SO_4 to a shifting sulphate concentration in the range of linear absorption. In knowing exactly the amount of extra SO_4^{2-} ions introduced in the solution, it is possible to calculate the initial SO_4^{2-} concentration. This approach can be used if the SO_4^{2-} concentration is lower than 10 mg/L. The offset of the results is demonstrated in Fig. 2.

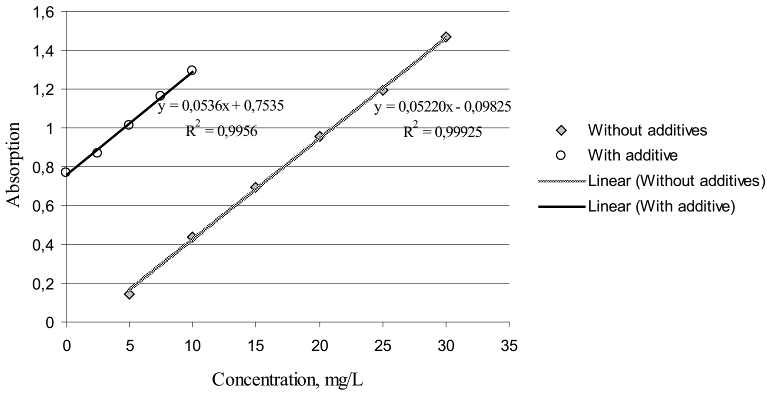

Fig. 2. Calibration curves for SO_4^{2-} measurement with and without additives or extra added Na_2SO_4.

This study is supported by the European Social Fund project No.2009/0212/1DP/1.1.1.2.0/09/APIA/VIAA/060.

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The session is organised in the scope of the European Social Fund project „Establishment of interdisciplinary scientist group and modelling system for groundwater research" (Project contract Nr. 2009/0212/1DP/1.1.1.2.0/09/APIA/VIAA/060). Project is implemented by University of Latvia, Faculty of Geography and Earth sciences and Faculty of Physics and Mathematics in collaboration with Latvia University of Agriculture, Faculty of Rural Engineering, Department of Environmental Engineering and Water Management.
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ISBN 978-9984-45-443-6
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