SULPHATE BALANCE OF LAKES AND SHALLOW GROUNDWATER IN THE VASAVERE BURIED VALLEY, NORTHEAST ESTONIA^{*}

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Groundwater is an important component of many water resource systems supplying water for domestic use, industry, and agriculture. Management of a groundwater system, an aquifer, means making such decisions as annual withdrawal of the total quantity of water, location of wells for pumping and for artificial recharge and their rates, and control conditions at aquifer boundaries. The decisions related to groundwater quality are of no less importance. In fact, the quantity and quality problems cannot be separated. In many parts of the country, with the increased withdrawal of groundwater, the quality of groundwater has been continuously deteriorating. In recent years the attention has been focused on groundwater contamination by mine water. A thorough understanding of the system, and the processes that take place in it are needed for modeling procedure. It is important to identify those parts of the system's behavior that are relevant to the considered problem, while other parts may be neglected. On the basis of this understanding, summarized as a conceptual model of the given problem, a numerical model was constructed. Using the conceptual model the groundwater flow model of the Quaternary aquifer, in the eastern part of oil shale deposit area, in the Vasavere buried valley, was constructed. The infiltration of contaminants grows, and the Quaternary aquifer is polluted by sulphate in a large area of the Vasavere buried valley.

Introduction

The handling of groundwater needs problem-oriented monitoring in oil shale mining and processing area of Estonia. The analysis of the extensive field data of good quality collected over a relatively long period of time leads to basic understanding of the mechanics of groundwater movement and the influence of pumping water to the environment. Groundwater is the principal source of water supply for domestic, industrial and agricultural needs in

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As water quality is inextricably linked to water quantity, it is important to understand the significance of developing modeling techniques that can accommodate both features. Transport modeling tries to predict the behavior of contaminants in groundwater and thus gives a possibility to evaluate the severity of contamination. Therefore, transport modeling is important to judge existing subsurface pollution. Two points are of main interest in transport modeling – groundwater flow and geochemical behavior of contaminants.

The determination of groundwater volumes and flow rates in the Vasavere buried valley requires a thorough knowledge of its geology. The character and arrangement of soil are important and variable factors within a groundwater reservoir. The Vasavere buried valley intersects the Ordovician limestone and represents a natural groundwater collector filled with sands.

In this paper the influence of sulphate of groundwater on a sandy sediment-groundwater system is calculated.

Description of Study Area

The Vasavere buried valley (Fig. 1), situated in the eastern part of the Estonia oil shale deposit area, is of north-south orientation and surrounded from all sides by oil shale underground and surface mines. The area is about 30 km² with 39 lakes that are more or less influenced by mine water, water consumption and industrial activities. The Vasavere buried valley groundwater quality is affected by the drainage of the *Sirgala* surface oil shale mine, peat cutting in Oru, oil shale extraction in the *Ahtme* underground mine, sand production in the *Pannjärve* pit, and water consumption in the Vasavere water intake.



Fig. 1. Location map of Estonia oil shale deposit and the Vasavere buried valley: 1 - oil shale underground and surface mines, 2 - claims and names of underground and surface mines, 3 - buried valley

The state of the lake ecosystems is, first of all, determined by the groundwater regime. Some of the effects are as follows:

- 1. Introduction of airborne pollutants into groundwater
- 2. Increase in the concentration of sulphate in shallow groundwater
- 3. Increase in the concentration of sulphate in lakes

The Quaternary aquifer in the valley often represents productive resources for groundwater use and supply. The Vasavere linear water intake is operating since 1972 and uses the groundwater of limno-glacifluvial aquifer. The groundwater of the Quaternary aquifer system is of the HCO₃–Ca–Mgtype with TDS up to 0.5 g/l [1]. The groundwater consumption from the Vasavere intake exerts an obvious influence on the lakes Martiska, Kuradijärv and Ahnejärv, where the water level has dropped.

Besides, there are exploitable high-quality sand deposits in *Pannjärve* sand pit, which is situated close to the water intake. In winter the wells in the pit do not function. As a result the water level rises, and the gradient of its outflow towards the Vasavere water intake increases. The natural water level lowering in some lakes is caused by drainage in peat cutting field and *Sirgala* surface mine, which are situated in the northeastern part of the area.

The *Ahtme* underground oil shale mine is situated in the northwestern part. It reached this area during the 1950s. By now the mine is closed.

The excavation of oil shale upsets water dynamical and chemical regime in the northern and eastern parts of the Vasavere buried valley. The groundwater of the aquifer begins to move concentrically towards excavation cavities. The comparatively big and uneven influx of water into the *Sirgala* surface mine and the *Ahtme* underground mine is caused mainly by irregular distribution of the industrial oil shale layers in the carbonate rocks of the split and eroded Ordovician deposit.

Data and Methods

For the purpose of modeling, a groundwater aquifer may be defined as a region of porous media in which void spaces are totally occupied by water. The movement of water within a given groundwater region is controlled by the flows crossing its boundaries, such as recharge or discharge across the water table, and by hydraulic relationships within the region itself. More specifically, the flow through saturated porous media is determined by the relationship defined as Darcy's law. A water quality model is mathematical statement or set of statements that equate water quality at a point of interest to causative factors. In general, water quality models are designed

- 1. to accept as input, constituent concentration *versus* time at points of entry to the system
- 2. to simulate the mixing and reaction kinetics of the system
- 3. to synthesise a time-distributed output at the system outlet

To investigate the Vasavere buried valley the official data from different enterprises and institutions were used. In the present paper official data from Estonian Geological Survey, *Eesti Põlevkivi (Estonian Oil Shale)* Ltd. and Estonian Meteorological and Hydrological Institute were used.

Human impact on the lakes and shallow groundwater in the Vasavere buried valley was rather small up to the middle of the last century, because the area was sparsely populated and the land use was rather modest. A sharp increase in the anthropogenic impact after World War II was caused by gradual expansion of mining and related industries, especially after the construction of several powerful oil-shale-based power plants put into operation in the early 1950s of the last century. The data available on the state of the lakes before World War II is quite limited. It may be assumed that up to the 1950s natural conditions prevailed. The earliest published data characterizing the end of the 1930s support this assumption. Comparing these data with the present ones, it is possible to evaluate the extent of man-made changes in the lakes and shallow groundwater.

Local hydrogeological conditions depend mainly on the hydraulic conductivity which is mainly determined by human impact. This causes the highest influx of shallow groundwater into surface mines and water intake. Precipitation, which is considered as a main source for groundwater replenishment, swiftly passes through the thin cover of limno-glacifluvial deposits and infiltrates into the relatively highly fractured Ordovician carbonate rocks. Seasonal factors, changing flow and various forms of recharge may all produce water level fluctuations of about 1 m.

Conceptual Model

Characterization of local conditions at a specific site provides the basic background information for the selection and design of the most appropriate system for restoration. This implies previous characterization and interpretation of flow and transport in the aquifer. The following factors must be characterized:

- 1. The source of sulphate
- 2. Aquifer hydrogeology and hydrochemistry (through aquifer monitoring)
- 3. Sulphate present

The information gathered from groundwater monitoring, sampling and study of sulphate behavior give an understanding of the aquifer hydrogeology, groundwater flow path, as well as sulphate behavior, concentration and distribution.

The aquifer to be modeled is the uppermost one, i.e. the aquifer in the Quaternary sand. It is a water-table aquifer, since it does not have any confining bed on the top. The water enters the aquifer by precipitation and moves from it to the oil shale underground and surface mines, lakes and the Vasavere water intake.

At modeling the following assumptions were made:

- 1. A porous medium continuum approach based on Darcy's law is applicable
- 2. The aquifer is considered being isotropic
- 3. Vertical flow components are ignored, so reducing the dimensions of the flow model from three to two (2D horizontal)
- 4. The groundwater divide is assumed to coincide with the surface water divide



The area to be modeled is restricted by the groundwater divide from the north-south side, and by the west-eastern side. The length of the area is 8.250 km, the width is 5.250 km, and the total area is 43.3125 km². The modeled domain is discretised using a 15×33 uniformly spaced finite difference grid of spacing 350 m as shown in Fig. 2. Specified head boundaries are located along row 33 and along column 1. Only a single aquifer is modeled; therefore only one layer is used. The aquifer is treated as confined be-

cause it is relatively thick and does not experience large changes in saturated thickness. Intergranual permeabilities average 2500–2700 m³ d⁻¹ m⁻² in its central part and drop to 10–50 m³ d⁻¹ m⁻² at the border of the area [2].

In all 18 groundwater wells exist within the research area, including monitoring wells and well field of water intake. The boundary condition for the groundwater divide is by definition a no-flux boundary. A much more difficult problem is to choose a proper boundary condition for the underground and surface mines, through which groundwater is discharged from the aquifer. One possibility is to use a prescribed head boundary condition. The problem is that any external water body does not control the heads along the boundary. The other possibility is to extract the water from the aquifer by applying a prescribed flux condition. Again, we do not know the magnitude of the flux. In this study the flux-out of the aquifer in a boundary block was calculated by multiplying the product of hydraulic conductivity, distance of the water level from the aquifer bottom and width of the block by the gradient of the water table at the boundary. The gradient of the water table was assumed to be constant along the whole boundary.

The initial state for dynamic calculations was created by calibrating a steady-state model to the observed water levels in the beginning of the calculation period. In generation of the initial condition, prescribed head boundary condition along the surface and underground mines was used. The head estimations near wells were based on the water-level measurements, and the head estimation at the top of the cape was obtained by visual observation.

The Quaternary aquifer is an unconfined water-bearing stratum fed by precipitation. A part of the rainfall is evaporated, a part is transported as surface runoff to the Vasavere River, channels and lakes. The remaining part of precipitation enters the aquifer. There is a meteorological station at the town of Jõhvi, and therefore rainfall measurements of good quality are available. The delay before rainwater enters the aquifer was modeled as water being released from a linear storage fed by rainfall. Evapotranspiration was taken into account by subtracting the evaporated water from storage. Outflow from the storage is the upper limit for the recharge value used in the model. The effect of surface runoff is included by multiplying the maximum amount of recharge by an appropriate coefficient (smaller than one).

Water level measurements in the wells were used in the calibration process. All wells are screened in the same aquifer. Groundwater level elevations in the wells are not very different and such differences can be explained with this kind of model. The location of the observation wells is shown in Fig. 2.

The groundwater model was applied to calculate the groundwater table in the model area in steady state. Comparing the computed groundwater level elevation values with the measured ones one can see that the computed values are too low. The value used as the maximum amount of recharge is already so high that its increasing is not realistic. The porosity and permeability values of a Quaternary aquifer depend to a large extent upon the degree of the cementation in the sand aquifer. Consequently, these values are generally expected to be much higher for the central part of the Vasavere buried valley than those for slopes. The results indicate that the values of calibrated hydraulic conductivities are too high (Fig. 3).

The aquifer is considered to be isotropic. This is a simplifying assumption, based on not knowing the nature of the prevailing anisotropy, and lack of data, rather than on a physical reality. If there is no natural barrier blocking the groundwater flow, the location of the groundwater divide can only be determined by simulating the groundwater flow on the area. In this case the location of the divide also depends on the recharge, i.e. the precipitation pattern and water pumped out of mines.

The mass balance is a very useful aspect of a model and can be used to check the conceptualization of an aquifer system, thus verifying the numerical accuracy of the solution, and to assess flow rates in discrete portions of the aquifer.

The groundwater level measurements and the chemical analysis results also include observation error. In this study, however, the observation error (probably ≤ 20 cm) is very small in comparison to all other sources.



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-19 -21 -23 -23 -25 -27 -29 -29 -31

-33

Fig. 3. Hydraulic conductivity of Quaternary sediments

Results and Discussion

A preliminary study of the hydrochemical properties of the shallow groundwater and sulphate balance from different parts of the area indicates the impact of different kinds of land use on the groundwater. The content of sulphate in the groundwater indicates directly the influence of the mining waters. Oxidation of the pyrite present in Ordovician deposits serves as a source of this compound, and concentrations approximately 500 mg l⁻¹ [3] have been found in mining waters. The data have been obtained by regular long series of analysis of water samples taken from observation wells located beyond the mining area and containing water flowing out of the undamaged bed, and of water pumped out from underground and surface mines. The processes that determine the chemical composition of the investigated water samples can be subdivided as follows:

- 1. Chemical composition and balance of the natural tectonically undisturbed aquifers
- 2. Chemical processes taking place at the sites of tectonic damage and erosion, and alteration of hydraulic parameters
- 3. Infiltration of groundwater through recharging layers
- 4. Changes in the natural aquifer due to mining



Fig. 4. Development of groundwater depressions during the last ten years

As a result of the joint influence of these processes, the natural hydrocarbonate water, characteristic of the Quaternary aquifer in this region, has changed into a hydrocarbonate-sulphate one. A very significant role in the formation of the chemical composition of water is played by depressions (Fig. 4) that have developed during the exploitation of underground and surface mines and water consumption. Their impact is two-fold: infiltration and water exchange increase significantly, and with the change of aeration conditions a geochemical environment with a new physical-chemical properties are formed. This change in geochemical conditions – because of the increased oxygen content – is evidently one of the reasons why the concentration of sulphate ions in the mine water has increased sharply posing a serious threat to water basins by which mine waters are removed, and is also a serious impediment to using the mine waters for technological purposes.

Oil shale mining brings about changes in the groundwater regime and chemical composition. As a result of extensive draining of mining shafts and water consumption, the groundwater table has noticeably lowered in the area of the Vasavere buried valley, and sulphate content in lakes and groundwater will be especially high in this area. The most noticeable change is a sharp increase in sulphate anions, a natural background for sulphate being between $2-10 \text{ mg l}^{-1}$. Evidently, the rise in sulphate anion content of such waters has been caused by oxidation of pyrite in well-aerated water, which percolates down through the overburden.



Fig. 5. Sulphate content in Quaternary sediments in 1947

It must be noted that this sharp rise in the sulphate ion concentration that exceeds concentrations characteristic of loading and reposing water zones by up to 50 times is probably caused by several processes: the sulphates partly penetrate with precipitation where the concentration of sulphate attains $30-40 \text{ mg l}^{-1}$, and with pumped-out mine water partially flowing back into the depression. This is naturally accompanied by intensive removal of the sulphates from the Quaternary sediments and recharging Ordovician carbonate rocks. Significant enrichment of water with the sulphates takes place due to oxidation of finely dispersed pyrite found in the carbonate rocks in the aeration zone. Rising concentration of the sulphate ions is accompanied by increasing concentration of calcium.

In 1947, when the lakes studied were mostly in natural conditions, the sulphate content was in the range of $1.0-6.7 \text{ mg l}^{-1}$ in the lakes and, presumably, also in the groundwater (Fig. 5).



Fig. 6. Sulphate content in directly and indirectly influenced lakes

Variations in this value seem to have been caused mainly by natural factors. At that time the land use was rather modest and the area was sparsely populated.

In recent years the content of sulphate has increased both in the closed lakes and in those influenced by mining waters (Fig. 6). This rise has been especially high in the lakes affected directly (discharge) or indirectly (infiltration) by mining waters, having the sulphate values in the range of 160–259 mg l⁻¹. When, for example, in 1946 the sulphate content in Lakes Nõmmjärv and Konsu was 5.8 and 1.0 mg l⁻¹, then in 2000 it was 259 and 184 mg l⁻¹, respectively; in the shallow groundwater the content of sulphate increased more than 50 times during 1970–2000.

Hydrotechnogenic influxes generated by human activities have seriously deformed the hydrochemical conditions of the surfacewater and groundwater. At strong human impact (mining activities and water consumption), the sulphate content of groundwater has risen about 50 times (Fig. 7).

We do hope that self-regulation of natural biogeochemical processes may be rehabilitated in these lakes and shallow groundwater during 20–25 years. Water resource and sulphate content development have often been based on the predominant use of either surfacewater or groundwater; it must be emphasized that these two components of the total water resource are interdependent. Changes in one component can have far-reaching effects on the other.



Fig. 7. Sulphate content in Quaternary aquifer in 2000

Conclusions

Oil shale mining has a serious impact on the environment also due to the pollution of surfacewater and groundwater by contaminated mine drainage waters, lowering groundwater level, and changing soil properties. There is an increasing evidence that portions of the water infiltrating through the soil surface may rapidly move through the aeration zone along preferred flow paths such as macrospores and fractures. This rapid, concentrated flow may also have significant impact on the transport of pollutants to the groundwater

body. Decline in mining activities and introduction of new technologies together with economic measures has improved the situation but much should be done during coming years.

In recent years, in the area of oil shale mines, chemical composition of groundwater was unstable: the SO_4 content of groundwater was in spring 4–6 times higher than in summer. It can be caused by dissolution of pyrites in oxygen-abundant water in spring, but the influence of agricultural pollution cannot be excluded either.

Due to the reasons explained above, the author believes that in spite of relatively good fit between the model outcome and observations, application of the model to the actual problems may not give sufficiently reliable results. To get a reliable picture of the groundwater system studied in this work, e.g. in order to predict contaminant transport in different scenarios, much work is still needed. And not only hydrogeology of the area, but more research on hydrochemistry would also be necessary.

Numerous models have already been developed to simulate the abovementioned hydrochemical processes. Balancing the input and output of sulphate in the Vasavere valley enables to provide a provisional hypothesis about internal hydrochemical processes in the Quaternary aquifer. A chemical model can describe possible paths of reaction. A description of the influence of different kinds of land use on sulphate content of groundwater can be achieved by this method. In order to estimate the kinetics of reactions involved, it is necessary to obtain detailed information about groundwater flow in the aquifer.

The correlation between the natural (meteorological and hydrological) and technogenic (mining-technological, hydrogeological, hydrochemical) factors caused by the oil shale mining in the Vasavere valley during 1970–2000 has been studied. Based on different factors, a conceptual balance-scheme of water circulation for the Vasavere valley has been worked out. The scheme shows that under the influence of anthropogenic flows of water, a new, anthropogenic geochemical matter cycling from the geological environment into the lakes and groundwater has been formed in the valley. Oil shale production and water consumption seriously influence the hydrological, hydrogeological and hydrochemical regime and conditions of the Vasavere lakes and Quaternary aquifer.

Due to the combined effect of natural and anthropogenic factors, a persistent tendency for transformation of physical-chemical composition of the waters has been developed (heightened content of sulphate). This tendency will not disappear after finishing the active production processes and closing the mine in the years to come.

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