SULPHATE BALANCE OF LAKES AND SHALLOW GROUNDWATER IN THE VASAVERE BURIED VALLEY, NORTHEAST ESTONIA

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Groundwater constitutes an important component of many water resource systems, supplying water for domestic use, for industry, and for agriculture. Management of a groundwater system, an aquifer, means making such decisions as to the total quantity of water to be withdrawn annually, the location of wells for pumping and for artificial recharge and their rates, and control conditions at aquifer boundaries. Not less important are decisions related to groundwater quality. In fact, the quantity and quality problems cannot be separated. In many parts of 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 most important part of modelling procedure is a thorough understanding of the system, and the processes that take place in it. It is important to identify those parts of the system’s behaviour that are relevant to the considered problem, while other parts may be neglected. On the basis of this understanding, summarised as a conceptual model of the given problem, a numerical model is constructed. A groundwater flow model of the Quaternary aquifer, in eastern part of oil shale deposit area, in Vasavere buried valley, was constructed by using the conceptual model. The infiltration of contaminants grows and the Quaternary
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 of water to environment. Groundwater is the principal source of water supply for domestic, industrial and agricultural needs in northeast Estonia.

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

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

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

Description of Study Area

The Vasavere buried valley (Fig. 1), which is situated in the eastern part of the Estonian 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, and contains 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 surface oil shale mine in Sirgala, 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. The state of the lake ecosystems is, first of all, determined by the groundwater regime. Some of 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 Quaternary aquifer system is of HCO₃-Ca-Mg-type with TDS up to 0.5 g/l [1]. The groundwater consumption from the Vasavere intake exerts an obvious influence on the Lakes Martiska, Kuradi- and Ahnejärv, where the water level has dropped.

Besides, there are exploitable high-quality sand deposits in Pannjärve sand pit, which situated close to 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 caused by drainage in peat
cutting field and Sirgala surface mine, which are situated in the north-eastern part of the area.

The Ahtme underground oil shale mine is situated in the north-western part and reached to this area during 1950s. By now the underground mine is closed.

The excavation of oil shale upsets the 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 the excavation cavities. The comparatively big and uneven influx of water into the Sirgala surface mine and the Ahtme underground mine is caused mainly by the 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 modelling, 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 one 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 to

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

To investigation of Vasavere buried valley the official data from different enterprises and institutions were used. In the present paper used official data from Estonian Geological Survey, Eesti Põlevkivi Ltd. and Estonian Meteorological and Hydrological Institute.

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 that started to operate in the early fifties 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, characterising 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 fluctuations in the water level of about 1 m.

**Conceptual model**

The characterisation 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 the previous characterisation and interpretation of flow and transport in the aquifer. The following factors must be characterised:

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 behaviour give an understanding of the aquifer hydrogeology, groundwater flow path, as well as sulphate behaviour, concentration and distribution.

The aquifer to be modelled is uppermost aquifer, i.e. the aquifer in the Quaternary sand. The aquifer is a water table aquifer, since it does not have any confining bed on the top of it. The water enters the aquifer by precipitation and it discharges from the aquifer to the oil
shale underground and surface mines, lakes and Vasavere water intake. Furthermore, the following assumptions are made:

1. a continuum approach of porous medium is based Darcy’s law is applicable.
2. the aquifer considered being isotropic.
3. vertical flow components are ignored, which reduces 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 modelled 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 kilometres, the width is 5.250 kilometres, and the total area is 43.3125 square kilometres. The modelled domain is discretised using a fifteen by thirty three uniformly spaced finite difference grid of spacing 350 m as shown in Figure 2. Specified head boundaries are located along row 33 and along

Fig. 2. Conceptual model of Vasavere buried valley.
1 – well of Quaternary aquifer, 2 – boundary of buried valley, 3 – peat cutting area, 4 – sand pit, 5 – water intake, 6 – boundary of surface and underground mines, 7 – lakes.
Only a single aquifer is modelled; therefore only one layer is used. The aquifer is treated as confined because it is relatively thick and does not experience large changes in saturated thickness. Intergranular permeabilities average 2500-2700 m$^3$d$^{-1}$m$^{-2}$ in its central part and drop to 10-50 m$^3$d$^{-1}$m$^{-2}$ at the border of the area [2].

A total of 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 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 calculated the flux out of the aquifer in a boundary block by multiplying the product of hydraulic conductivity, distance of the water level from the aquifer bottom and width of the block by gradient of the water table at the boundary. The gradient of the water table is assumed to be constant the whole boundary.

The initial state for dynamic calculations is 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 is used along the surface and underground mines. The head estimations near wells are based on 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 and 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 Jõhvi and therefore rainfall measurements of good quality are available. The delay before rainwater enters the aquifer is modelled as water being released from a linear storage, which is fed by rainfall. Evapotranspiration is 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
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, that such differences can be explained with this kind of model. The location of the observation wells are shown in Figure 2.

The groundwater model is applied to calculate the groundwater table in the model area in steady state. Comparing the computed groundwater level elevation values with the measured values, the computed values are too low. The value used, as the maximum amount of recharge is already so high that increasing it is not realistic opinion. 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 Vasavere buried valley than those for slopes. The results indicate that the values of calibrated hydraulic conductivities are too high (Fig 3).

Fig. 3. Quaternary sediments hydraulic conductivity

The aquifer is considered to be isotropic. This is simplifying assumption, which is more
based on not knowing the nature of the prevailing anisotropy, and lack of data, than a physical reality. If there is no natural barrier, which blocks 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 as a check on the conceptualisation of an aquifer system, as a check on 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 also contain observation error. In this study, however, the observation error (probably in magnitude <20 cm) is very small in comparison to all other sources.

**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 contained 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, of water pumped out from underground and surface mines. The processes that determine the chemical composition of water in the investigated 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. man-made changes in the natural aquifer, due to mining.

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 the sulphate ions has increased sharply in the mine water
and poses a serious threat to water basins, by which mine waters are removed, and is also a
serious impediment to using the waters of mine for technological purposes.

Fig. 4. Development of groundwater depressions during last 10 years.
The 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 Vasavere buried valley and especially high in this area will be sulphate content in lakes and groundwater. The most noticeable change is a sharp increase of sulphate anions, a natural background for sulphate being between 2-10 mg l$^{-1}$. Evidently, the rise of sulphate anions in such waters has been caused by oxidation of pyrite in well-aerated water, which percolates down through the overburden. 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 are partly penetrating with precipitation where the concentration of sulphate attains 30-40 mg l$^{-1}$, and with pumped out mine water partially flowing back in to 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 mg l$^{-1}$ in the lakes and, presumably, also in the groundwater (Fig. 5).
Variations of this value seem to have been caused mainly by natural factors. This time the land use was rather modest and 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 were 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.

Fig. 6. Sulphate content in directly and indirectly influenced lakes
Hydrotechnogenic influxes generated by human activities have seriously deformed the hydrochemical conditions of the surface and groundwater. Accordingly strong human impact (mining activities and water consumption) the sulphate content in groundwater rises about 50 times (Fig. 7).

Fig. 7. Sulphate content in Quaternary aquifer in 2000.
1 – sulphate content <100 mg/l, 2 – 100-150 mg/l, 3 - >150 mg/l.

Have a 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 surface or groundwater; it must be emphasised that these two components of the total water resource are interdependent. Changes in one component can have far-reaching effects on the other.

Conclusions

The oil shale mining has a serious impact on the environment also due to the pollution of surface and groundwater by polluted mine drainage waters, lowering of groundwater level, changing of soil properties. There is increasing evidence that portions of the water infiltrating through the soil surface may move rapidly through the aeration zone along
preferred flow paths such as macrospores and fractures. This rapid, concentrated flow may also have significant implications for the transport of pollutants to the groundwater body. Decline in mining activities and the 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, the chemical composition of groundwater was unstable: the content of SO$_4^{2-}$ in 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, also cannot exclude the influence of agricultural pollution.

Due to reasons explained above, the author believes that in spite of relatively good fit between the model outcome and the observations, application of the model to real world problems may not give sufficiently reliable results. To get 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 above-mentioned 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 in 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. Scheme shows that under the influence of anthropogenic flows of water the new, anthropogenic geochemical matter cycling from 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 developed (heightened content of sulphate). This tendency will not disappear after finishing the active production processes and closing the mine in first years.

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References:

