

CHANGES IN GROUNDWATER SULPHATE CONTENT IN ESTONIAN OIL SHALE MINING AREA

K. ERG*

Department of Mining,
Tallinn University of Technology
5, Ehitajate Rd., 19086 Tallinn, Estonia

Dewatering of oil shale mines lowered groundwater level in the Keila-Kukruse aquifer and caused an increase in sulphate content about 50 times due to intensive oxidation of pyrite of natural origin. As sulphate ion is mobile, it may be used as an indicator to investigate changes in sulphate content of post-mining groundwater of the Keila-Kukruse and Lasnamäe-Kunda aquifers of the Ordovician system in the area of closed and working mines.

Investigation of sulphate ion distribution is one possible way to know how groundwater moves in lateral direction, both during flooding of underground mines and after reaching steady-state conditions.

Introduction

In many parts of the world more and more coal, ore and other mineral fields are closed [1–3]. Europe was once the most important mining region in the world, and nearly every European country has closed mining sites [4]. For the last decade, substantial efforts have been put into reclamation of closed underground mine sites over the world [4, 5]. Closed underground mines involve risks for the environment. Some of these risks are linked to the shut-down of the operations of mine water pumping leading to a rise in water level and the pollution of groundwater by sulphates and other chemical elements. After closing of the mines these risks may exist during a short, long or very long period of time depending on the quantity and flow of water involved and the volume of the mine workings concerned [6].

Oil shale production started in 1916, and until today different numbers of mines have been operating in Estonia. By 2005, eight mines have been closed in the central part of the oil shale deposit, two mines (*Estonia* and *Viru*) are in operation, and new reserves are continuously being identified and developed. Oil shale production in the Estonia deposit has directly

* E-mail erg@staff.ttu.ee

spoiled overlying geological layers over 220 km² [7]. Shut-down of pumping operations at first induced rise in water level, and sulphate content of water sharply increased. This was caused by oxidation of pyrite. It is one of the environmental problems associated with underground mining. As sulphate ion is mobile, it is possible to follow changes in sulphate content of groundwater of the Keila-Kukruse and Lasnamäe-Kunda aquifers in the post-mining time and to understand how waters move in lateral direction in both aquifers of the Ordovician system.

Description of Study Area

The Estonia oil shale deposit is located in north-eastern Estonia. East-westward length of the wedge-shaped deposit is about 130–140 km. From south to the northern erosional boundary, the extent of the deposit increases eastward; reaching about 40–45 km. The closed and two working oil shale mines are located in the central part of the oil shale deposit (Fig. 1).

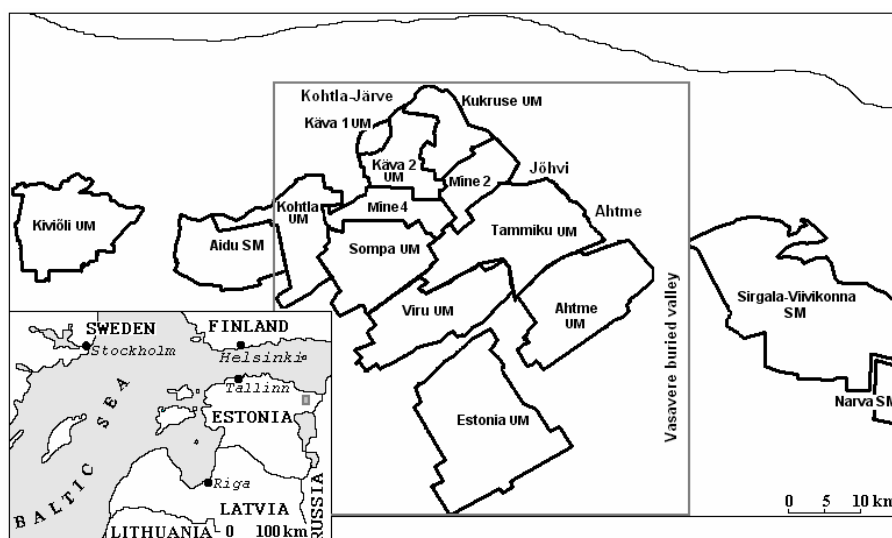


Fig. 1. Location of surface and underground mines in the research area in the Estonia oil shale deposit. UM – underground mine, SM – surface mine

From the hydrogeologic standpoint, the oil shale deposit is divided into two principal hydrostratigraphic units associated with a northeast- and east-driven flow with recharge zones at the highest outcrops and discharge zones in the rivers. Oil shale mining has affected the groundwater regime and chemistry of the system of Quaternary and Ordovician aquifers. Hydraulic properties of the aquifers are summarized in Table 1.

Table 1. The Hydraulic Properties of Aquifers in the Estonia Oil Shale Deposit [8]

Age	Quaternary	Ordovician		
Aquifer system	Q	Nabala-Rakvere, O ₂ nb-rk	Keila-Kukruse, O ₂ kl-kk	Lasnamäe-Kunda, O ₂ ls-kn
Rock type	Sand, till, peat	Limestone, marl, dolostone		
Depth, m	0	2–20	0.5–50	0.5–100
Thickness, m	0–77	0–50	0–44	17–24
Water table (piezometric), m below surface	+0.3–16	+0.1–13.2	0.2–28.2	0.6–15.6
Specific capacity, l/sec/m drawdown	0.001–54	0.025–11.0	0.007–8.3	0.001–2.1
Hydraulic conductivity, m/day	0.02–175	0.40–185	0.04–170	0–48
Transmissivity, m ² /day	0.1–1980	4–2546	0.03–2308	0.01–187

Quaternary deposits comprise a thin layer of peat, sand and till and unconsolidated glaciophysical sediments that constitute porous aquifers with mainly unconfined groundwater [9], influenced directly by meteorological conditions. Surface water percolates directly into the Quaternary cover, and most of the groundwater flows through the cover as the groundwater discharges into springs, streams, rivers and wetlands. The main, Quaternary aquifer in the oil shale deposit is the Vasavere buried valley, which is up to 70 m deep and filled with sand and gravel with very high hydraulic conductivity and storage capacity.

Ordovician aquifer system (O) consisting of lime- and dolostones with clayey interlayer lenses are found below the shallow cover of the Quaternary sediments in oil shale deposit. The limestone may be divided into a near-surface karst aquifer, cutting across the stratigraphic units, and several deep fracture aquifers, corresponding to the stratigraphic units [10, 11]. The upper part of the Ordovician system is heavily karstified and fissured to a depth of 30–40 m [8], with deep vertical fissures and karst cavities up to half a metre in diameter, forming open channels or filled by sand and soil from the surface.

The karstified zone is an excellent aquifer, with high storage capacity and hydraulic conductivity in the karst fractures and cavities. The karst aquifers are closely coupled with the overlying Quaternary aquifers; the karst aquifers are inseparable from the Quaternary aquifers at locations where sand and soil from the surface have been washed or fallen into the fractures and cavities of the karst aquifer. The deeper layers, where water storage and flow is limited to fractures, are generally poor aquifers; at some locations they may even be weak aquitards.

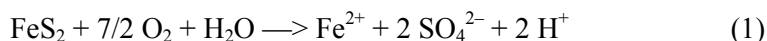
The individual limestone aquifers are separated from each other, both vertically and horizontally, by aquitards of clayey shale. One of these aquitards is the kukersite oil shale. According to the data from 235 pumping

tests [12] carried out during oil-shale exploration, the lateral near surface hydraulic conductivity of the Ordovician limestone is in the range of 5–300 m/d, whereas it is only 0.1 m/d [12] at a depth of 80–100 m. Vertical conductivity of the clayey layers separating the water-bearing zones is 10^{-5} – 10^{-2} m/d [13]. Therefore, these clayey layers serve as aquitards, dividing the limestone into many local aquifers of different vertical and horizontal extent. The water flow in the karst fractures and in the cavities is relatively fast, resulting in good water yields; thus, the specific capacity of wells tapping the upper portion of the aquifer system is in the range of 0.001–11 l/sec/m drawdown, with an average of 0.5–3 l/sec/m [9] drawdown.

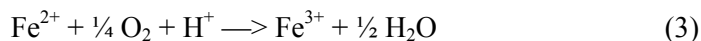
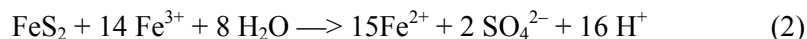
The local flow system mainly comprises the unconfined (or locally confined) shallow groundwater, moving from its recharge area toward the nearest ditches and rivers, and discharging directly into the sea. The total net infiltration first enters the Quaternary cover; part of the groundwater flows downward into the underlying bedrock, part discharges through springs or flows directly into the sea. In order to dewater the oil-shale mining areas, 159–226 millions m³ of groundwater depending on the amount of precipitation were extracted in northeastern Estonia in the 1980s.

The hydrogeology of underground working (*Viru* and *Estonia* mine) and closed mines was studied by Savitski and Boldõreva [14] and most recently by the researchers of the Department of Mining. Many other scientists have summarized localized environmental impacts from mining. The data obtained are generally well documented; however, the sulphate migration to the underlying aquifer and in the water of different mines remains relatively unclear.

Mine water in a closed mine is affected by sulphide oxidation. During mining processes pyrite (FeS₂) has been extensively mixed with air oxygen. It acts directly in oxidizing sulphide and iron (II) as shown by the reaction [15, 16]



or indirectly generating Fe(III) which then oxidizes pyrite. The reaction equations are as follows



Dissolution of pyrite leads to high concentrations of sulphates. The sulphide oxidation rate increases during the warm months, in other time the sulphide oxidation rate is low [15].

Material and Methods

The official data on the chemical content of water and water regime for the period 1991–2004 were received from the Estonian Oil Shale Company and from the Geological Survey of Estonia for the period 1970–2004. Groundwater samples were taken from permanently installed groundwater monitoring wells. The author carried out hydrogeological studies in the area of closed mines and lakes of Vasavere buried valley in 1982–1993 and 2004. During the last investigations water samples were taken from tectonic faults, karst fissures, and mine dewatering system in *Viru* and *Estonia* underground mines. Water samples taken from almost 50 sampling sites (Fig. 2) were analyzed on the content of more than twenty constituents.

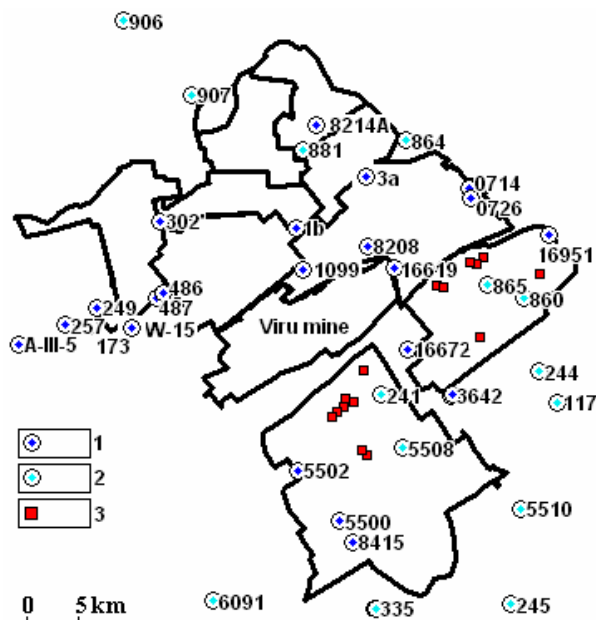


Fig. 2. Location of observation wells in the underground mine area:
 1 – water level and chemistry observation well in the Keila-Kukuruse and Lasnamäe-Kunda aquifers, Geological Survey of Estonia;
 2 – water level observation well in the Keila-Kukuruse and Lasnamäe-Kunda aquifers, Estonian Oil Shale Company;
 3 – observed groundwater in karst and tectonic faults

To investigate water movement different hydrogeological models have been compiled by various institutions and authors. In the Geological Survey of Estonia, Vallner [17] constructed a hydrogeological model for the Estonian territory and adjacent areas, covering a total of 88 032 km². The thirteen model layers include all main aquifers and aquitards from the ground surface down to the impermeable part of the crystalline basement.

Three-dimensional distribution of groundwater heads, flow directions, velocities, and rates as well as transport characteristics can be simulated by this model. Groundwater flow of the oil shale mining area was modelled by Savitski and Boldõreva [14].

A hydrodynamic model predicts the surface elevations and current velocity field across the model grid. It provides the flow data that can be used to run other models, such as water quality. A water quality model simulates the sulphate distribution that takes place within the modelled underground water basin. Different types of models to study sulphate transport processes were used. In the central part of the oil shale mining area sulphate distribution in groundwater of the Keila–Kukruse and Lasnamäe–Kunda aquifers was modelled.

We used software MapInfo Professional together with spatial analysis modelling package Vertical Mapper. Comparing intermediate results, calibrating can be achieved by different interpolation methods [18], including triangulations with smoothing, inverse distance weighting, rectangular (bilinear), kriging, custom point estimation and natural neighbour. The latter is the most suitable interpolation method for modelling sulphate distribution. This area stealing method creates natural neighbourhood regions for each data point and each grid cell. Cell values are derived using a point-weighting system based on the area of overlap of the grid cell natural neighbourhood region and the regions of surrounding data points. Therefore, it is important that the tool used is demonstrated to be suitable for the problem to be solved.

Results and Discussion

When the underground mines are closed, the pumping of water stops and the old shafts and tunnels fill up with water. Hydraulic conductivity is mainly determined by the degree of fracturing, by local karst and by human impact. As a rule, the hydraulic conductivity of host rock is at its highest near the surface and lowers gradually downwards. This causes the highest influx of shallow groundwater into shallow (40 m) underground mines. Long-term observation results [15, 19] show that the inflow to the mines varies with the seasons. Of the total water amount 20% falls on the winter months, 29% on the spring months, 27% to the summer months and 24% to the autumn months. Rainfall is characterized by an inter-annual irregularity. The above data show that the inflow is irregular in mines. This is characterized by the irregularity coefficient, which is received when the maximal inflow is divided by minimal inflow ($K = Q_{\max}/Q_{\min}$); the daily coefficient of irregularity is between 2 and 30 in the oil shale deposit area (Fig. 3).

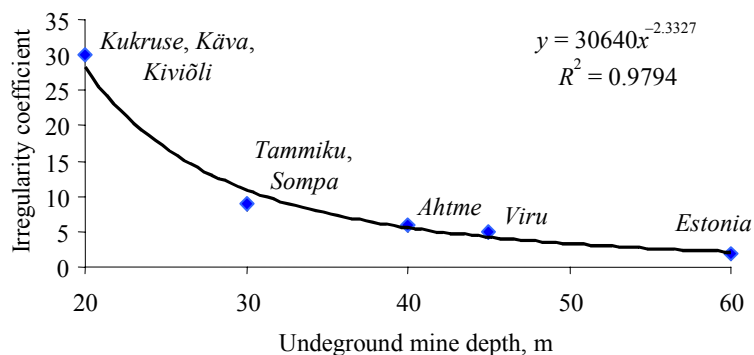


Fig. 3. Dependence of irregularity coefficient on mine depth

The impact of weather on the mine decreases with the depth of the mine. Figure 3 shows 2.3% decrease in the coefficient of inflow irregularity while mine becomes deeper by 1%.

In the area of underground oil shale mines the water level was studied in about fifty wells. Figure 4 shows only the wells located in the closed *Tammiku*, *Ahtme*, *Sompa* and *Kohtla* mines.

There may be a number of disconnected pools at the early stage of flooding in underground oil shale mines. Before flooding water sub-pools may exist at various locations and elevations within the mine. The abundance of sub-pools is greatest at the back of the mine where recharge and leakage collect. These sub-pools tend to coalesce and form a main pool, which will rise from the back of the mine in an up-dip direction. As flooding progresses, the sub-pools join into a single main pool with big water volume (Table 2). However, the main pool may stabilize at a lower elevation, if water-control measures are implemented or the mine spills into an adjacent mine.

Flooding situation is a transient scenario, while the flooded case is a steady-state one. In transient groundwater flow systems, hydraulic head is continuously changing with time, with minor seasonal or annual fluctuations. In 2003, the volume of water in the pools of the closed underground mines was about 160 million m³ [14]; in August 2003 it amounted to 170 million m³ (author's estimations) (see Table 2).

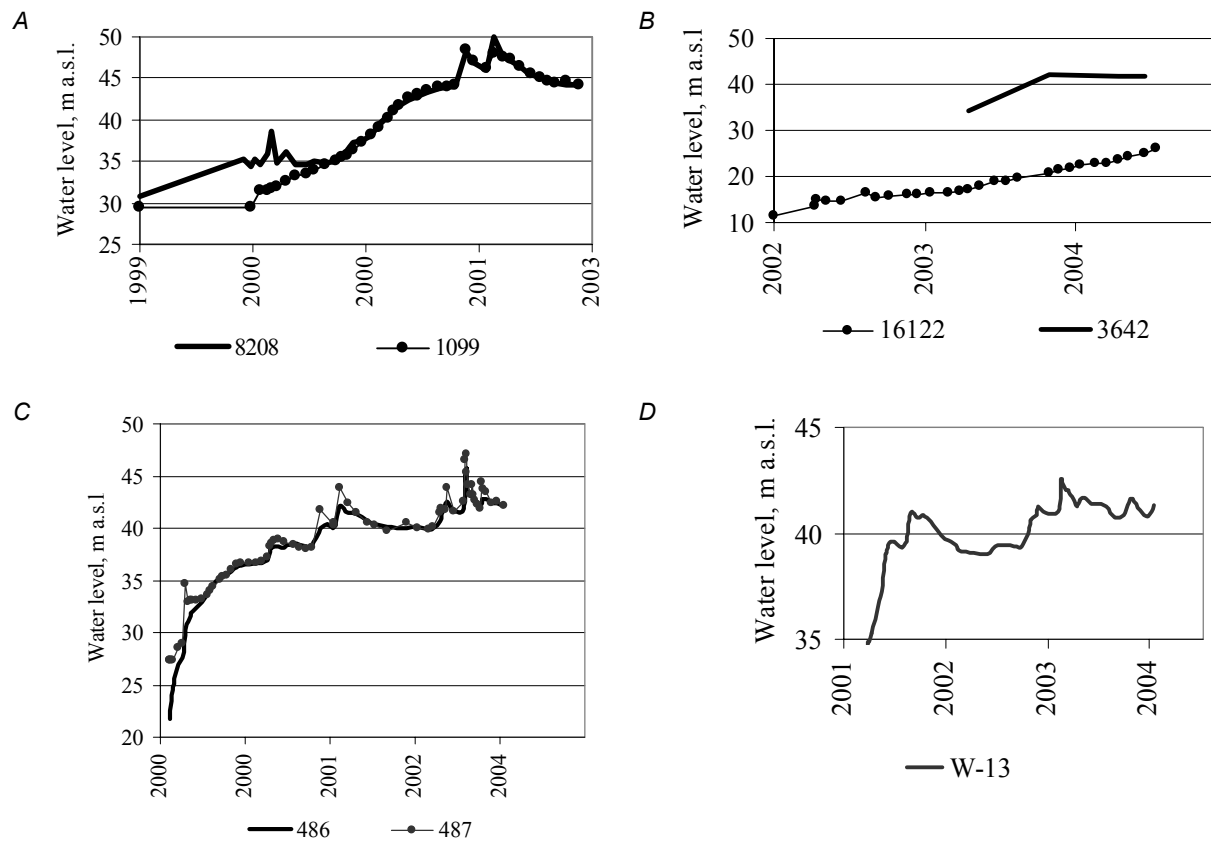


Fig. 4 Flooding of underground mines:
 A – Tammiku mine;
 B – Ahtme mine;
 C – Sompa mine;
 D – Kohtla mine

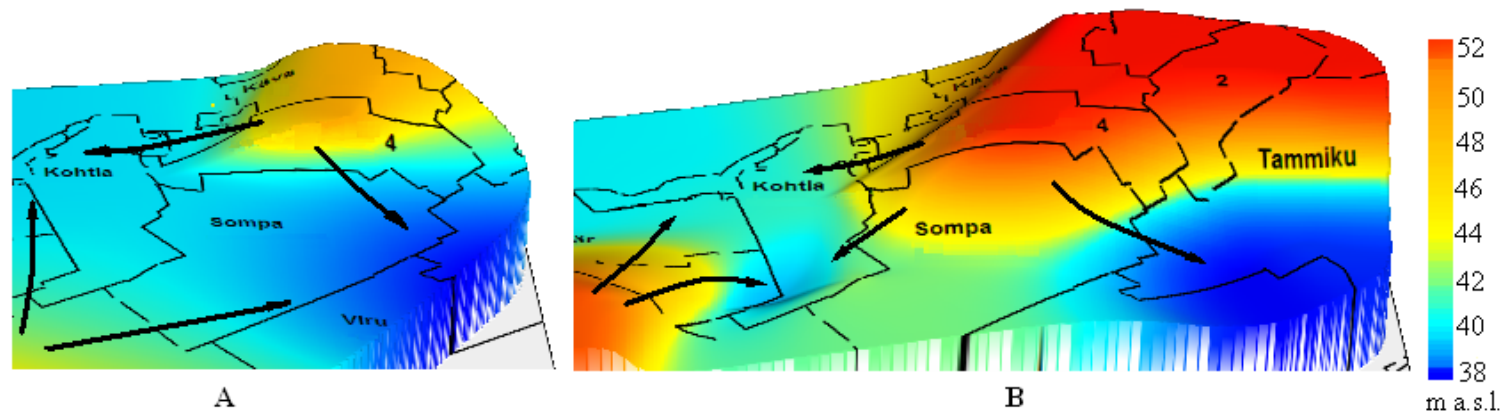


Fig. 5. Water level in the Keila-Kukuruse groundwater aquifer in 1990 (A) and in 2003 (B)

Table 2. Approximate Water Volume in Closed Underground Oil Shale Mines

Underground mine	Work started	Closed (pumping stopped) [14]	Water table in 2003 [14]		Mined out area, km ² [14]	Water volume (approximately), 10 ⁶ m ³	
			Obs. well No.	m, a.s.l.		[14] in 2003	Author's results, 2004
<i>Kukruse</i>	1916	1967	8214A	52	13	3.6	5
<i>Käva</i>	1924	1973	2	51.5	18	9	10
<i>Kohtla</i>	1937	28.06.2001	W-15	41	17	10	13
<i>Ahtme</i>	1948	1.04.2002	16122	25	35	60	63
<i>Sompa</i>	1949	2.12.2000	487	43	27	23	23
Mine No. 2	1949	1974	3a	51.41	13	7	7
<i>Tammiku</i>	1951	28.12.1999	714	47.95	40	~40	42
			8208	50.04			
			1099	47.92			
Mine No. 4	1953	1975	302	41.18	13	1.4	2.0
			1b	40.26			

The elevation of the water table in 1990 was about 42–50 m above the sea level (Fig. 5A) in *Käva* and *Kukruse* mines, Mine No. 2 and Mine No. 4.

In some cases, water levels in two or more adjacent mines will fluctuate in conformity with the seasonal or man-induced stresses. Hydrologic investigations indicate that the elevation of the water table has fluctuated over time, especially in Mine No. 2. The maximum elevation was about 51 m a.s.l., but seasonally it fluctuated between 50–56 m a.s.l., primarily as a result of variation in climate and increased precipitation.

If the inflow rate is all the time greater than the outflow rate, the water storage and hydraulic head in the saturated portion of the mine will increase. If outflows are greater than inflows, the hydraulic head will decline. During the rainy August of 2003, the water table rose 4 m in *Sompa* underground mine, 2 m in *Kohtla*, 2.1 m in *Kukruse*, 1.8 m in Mine No. 4 and 0.5 m in *Ahtme* mine. Filling of closed mines with water and restoration of Keila-Kukruse underground water level in 2003 are presented in Fig. 5B.

Estonia and *Viru* underground mines have advanced from shallow to deep cover and lie below regional drainage elevations. As mining progresses, groundwater can infiltrate into the mine. Therefore, the mine is progressively dewatered to allow mining to continue. As deeper mines are commonly separated from shallower up dip mines by thick barriers of unmined oil shale, the shallow closed mines may be flooded while deeper mining continues. One of several regulatory issues regarding the closure of such mines is long-term discharge of water after the mines have fully flooded. Following mine closure, pumping from the active mine is terminated and mine voids are allowed to re-saturate. Flooding of closed mines will generally continue until groundwater achieves a new equilibrium, either by surface discharge of mine water or by controlled pumping and

treatment. Eight of the underground mines located in the central part of the oil shale deposit were flooded in the beginning of 2004 by groundwater, which caused flooding in the northern part of the town of Jõhvi.

During mining the drowning of water level and increasing aeration zone cause intensive oxidation of pyrite, which is the biggest problem of groundwater pollution associated with underground mining. After mine closure water level rises, and pyrite oxidation slows down. The most noticeable change will take place in sulphate content. Evidently, the rise in the amount of sulphate anions (Fig. 6) in water has been caused by oxidation of pyrite in well-aerated water, which percolates down through the overburden. In the water, which fills underground mines, the content of this element is high (Fig. 6 – *Ahtme* mine), though lowering, and still stays 10 times higher than its natural background.

This is naturally accompanied by intensive removal of the sulphates recharging Ordovician carbonate rocks. Significant enrichment of water with sulphates takes place in the carbonate rocks in the aeration zone. There is an 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. In many cases, water is of low pH and contains elevated levels of sulphate ions.

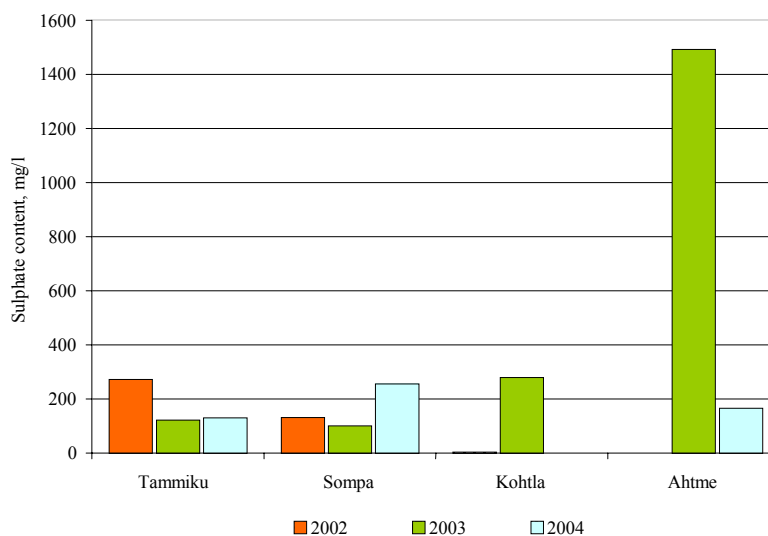


Fig. 6. Sulphate content of groundwater in underground mines: *Tammiku* observation well No. 0714; *Sompa* – 486; *Kohtla* – 0705; *Ahtme* – 16122

In recent years, in the area of oil shale underground mines, chemical composition of the groundwater has been stable. The content of SO_4 in groundwater in spring was 2 times higher than during the remaining seasons of the year (Fig. 7). It can be caused by dissolution of pyrites in the oxygen-abundant water in spring.

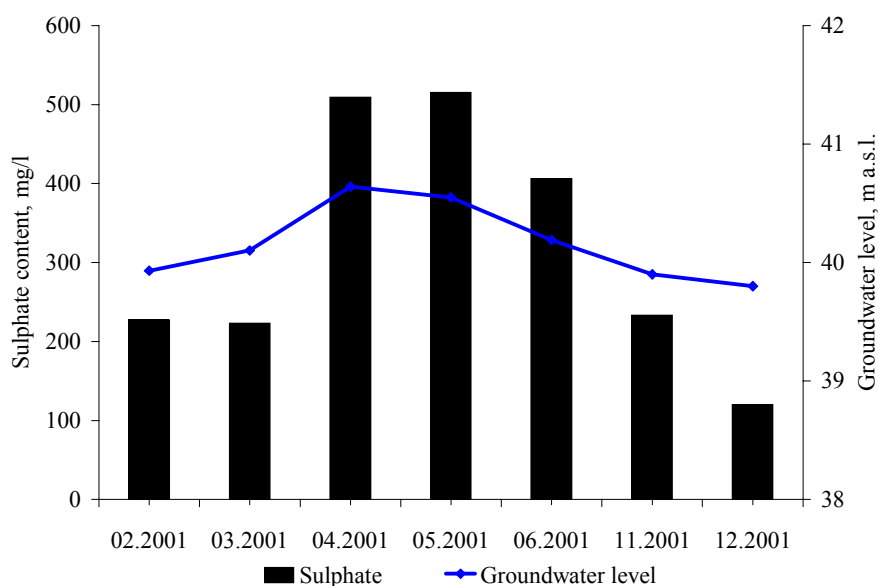


Fig. 7. Sulphate content of the water of the underground oil shale Mine No. 4 in 2001

Mine No. 4 closed in 1975 was filled with water in 1990. Mainly precipitation, groundwater flow from each side and rise in water level caused fluctuations in the sulphate content of Mine No. 4 (see Fig. 7).

Sulphate content of the water filling a mine is high; in the closed mines it is low. Water washes already oxidized pyrite products out of the limestone, and the sulphate content of groundwater will increase. Sulphate may be distributed in lateral direction many times more intensively than in transversal direction. This may be explained by the permeability of groundwater aquifer or aquifer system. Sulphate distribution in the water of underground mine in 2003 is shown in Fig. 8A.

In 2003, in the earliest closed underground mines (*Kukruse*, Mine No. 2) sulphate content was high in the Lasnamäe-Kunda aquifer. In the western part of *Tammiku* mine the Lasnamäe-Kunda aquifer was very rich in sulphate (Fig. 8B). This is promoted by karst and technogenic faults. In the southern part of *Kohtla* mine and in the northern part of *Sompa* mine sulphate content of the Lasnamäe-Kunda aquifer was between 100–200 mg/l (see Fig. 8). The distribution of sulphate in the Lasnamäe-Kunda aquifer may be due to the circumstance that the permeability of carbonated rock in a lateral direction can be up to 100 times higher than in transversal direction.

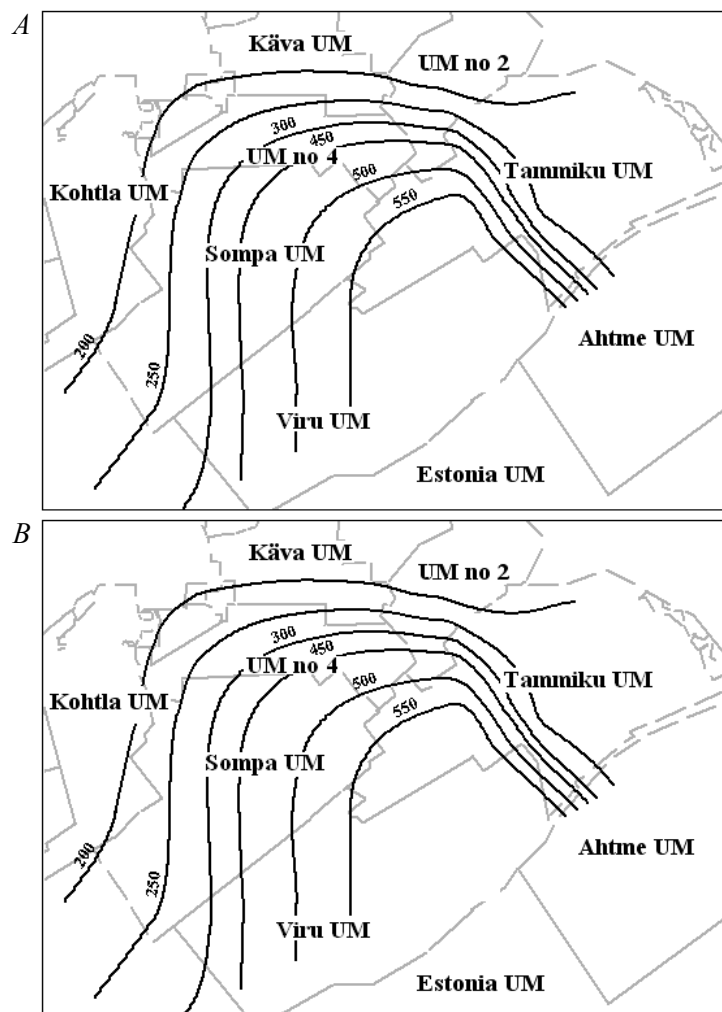


Fig. 8. Sulphate content in the Ordovician Keila-Kukruse aquifer (A) and Lasnamäe-Kunda aquifer (B) in the area of underground mines

Conclusions

In closed mine workings there form underground water pools with elevated sulphate content, that in any individual case depend on mining methods and underground mining operations.

The main results of this study may be summarized as follows:

1. The hydrogeological regime in oil shale mines is controlled by the thickness of the aeration zone, tectonical faults and fractures in the geological section, alteration of hydraulic gradients causing changes in flow direction and rate.

2. Closing and flooding of underground mines has changed the conditions of groundwater formation in the Lasnamäe-Kunda aquifer and its sulphate content.
3. Due to technogenic impact, the water of closed mines is connected with the Lasnamäe-Kunda aquifer.

Acknowledgements

I wish to express my sincerest gratitudes to Mr. Kalmer Sokman from Estonian Oil Shale Company who gave me useful advice in mining impact to environment and with whom I have been lucky to work for a long time. I would like to thank prof. Jaan-Mati Punning for his constructive criticism and help in preparing this paper.

The author is grateful to Estonian Scientific Foundation for financial support of this investigation (Grant No. 5913).

REFERENCES

1. *Younger, P. L.* Coalfield closure and the water environment in Europe // *Mining Technology*. 2002. Vol. 111, No. 3. P. 201–209.
2. *Rogoz, M., Posylek, E.* Hydrogeological problems connected with closure of mines on the Upper Silesian Coal Basin // *Proceedings, 7th International Mine Water Association Congress*. 2000. P. 319–330.
3. *Barnes, T. M.* Treatment of the gravity minewater discharge at Deerplay Mine, Burnley, UK // *Proceedings, 7th International Mine Water Association Congress*. 2000. P. 344–351.
4. *Wolkersdorfer, C., Bowell, R.* Contemporary Reviews of Mine Water Studies in Europe, Part 1 // *Mine Water and the Environment*. 2004. Vol 23, No. 4. P. 162–182.
5. *Wolkersdorfer, C., Bowell, R.* Contemporary Reviews of Mine Water Studies in Europe, Part 2 // *Mine Water and the Environment*. 2005. Vol 24, No. 1. P. 2–37.
6. *Banks, D., Younger, P. L., Arnesen, R.-T., Iversen, E. R., Banks, S. D.* 1997. Mine-water chemistry: the good, the bad and the ugly // *Environmental Geology*. 1997. Vol. 32, No. 3. P. 157–174.
7. *Reinsalu, E., Toomik, A., Valgma, I.* Mined Land. Tallinn: Soha OÜ, 2002, 97 [in Estonian].
8. *Perens, R., Andresmaa, E., Antonov, V., Roll, G., Sults, Ü.* Groundwater management in the northern Peipsi-Narva river basin. Background report, seminar "Support to Estonian-Russian Joint Peipsi-Narva Transboundary Water Commission through Capacity Building and Development of Recommendations" (CEE 008, supported by the Swedish Environmental Protection Agency). Tartu, Estonia, 2001, April 18–19.

9. *Perens, R., Vallner, L.* Water-bearing formation // Geology and Mineral Resources of Estonia. Raukas, A., Teedumäe, A. (eds.), Tallinn, Estonian Academy Publishers. 1997. P. 137–145.
10. *Vallner, L.* Groundwater. Estonian Environment. Past, Present and Future // Raukas, A. (ed.), Ministry of Environment, Tallinn, 1996. P. 60–71.
11. *Vallner, L., Savitskaja, L.* Groundwater extraction and safe yield // Geology and Mineral Resources of Estonia. Raukas, A., Teedumäe, A. (eds.), Tallinn, Estonian Academy Publishers, 1997. P. 159–162.
12. *Riet, K.* Generalized hydraulic conductivities of carbonate series of Ordovician strata of Estonian oil shale deposit // Geological notes, 3, Rõõmusoks, A., Kivila, H. (eds.) Tallinn, 1976. P. 88–92 [in Estonian].
13. *Perens, R., Savitski, L.* Hydrogeological conditions and problems on the left bank catchment area of the Narva River //: The Narva River and Reservoir (articles of hydrology, ecological conditions and water management). 2000. P. 24–35 [in Estonian and Russian].
14. *Savitski, L., Boldõreva, N.* Observation Area of Ida-Viru. Groundwater State in 1999–20039. – Tallinn, 2005. P. 43–50 [in Estonian].
15. *Karise, V., Pill, A., Johannes, E., Erg, K.* Water chemical content forming in Estonian oil shale mining area : Manuscript in Institute of Geology. Tallinn, 1987. P. 286.
16. *Singer P. C., Stumm, W.* Acidic mine drainage: the rate – determining step // Science. 1970. Vol. 167. P. 1121–1123.
17. *Vallner, L.* Hydrogeological model of Estonia and its applications // Proceedings of the Estonian Academy of Sciences. Geology. 2003. Vol. 52, No. 3. P. 179–192.
18. *Badman, T. D., Gravelle, M. A., Davis, G. M.* 2000. Seabed imaging using a computer mapping package: an example from Dorset // The Quarterly Journal of Engineering Geology and Hydrogeology, 33, 2, P. 171–175.
19. *Savitski, L.* The hydrogeological conditions. // Estonian Oil Shale / P. Vingissaar (ed.). Tallinn : Estonian Academy Publishers, 2000. P. 93–104.

Presented by E. Reinsalu

Received January 21, 2005