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Environmental problems in the Estonian oil shale industry

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In Estonia technologies for oil shale mining and consuming have been continuously developed for more than 80 years. By March 2006 one billion tonnes of oil shale had been produced in Estonia. Since the 1960s, Estonia has been the largest oil shale producer and consumer in the world. In the 1980s about two-thirds of the world's oil shale output came from Estonia. Serious problems in the mining complex are connected with large losses of oil shale at mining and enrichment (more than 30%) and voluminous dewatering (25 m^3 per tonne of oil shale). After mining and beneficiation, much limestone remains unused and is deposited in waste dumps. Oil shale waste and waste heaps may be considered a rather innocent production residue; however, from time to time they are subject to self-ignition. Following combustion of enriched oil shale, ash remains which also has to be deposited. The most toxic waste (semicoke) comes from the oil-shale chemical industry. Power stations using oil shale emit large amounts of carbon dioxide and other gases; the groundwater regime, and often also the water quality, are altered in mined-out areas. Environmental effects and the resulting immediate hazards were greatest in the 1980s, now the situation is improving.

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Broader context

Reserves of oil shale are immense, exceeding the resources of all other solid fuels combined. Since the 1960s, Estonia has been the largest oil shale producer and consumer in the world and this has caused serious environmental problems (voluminous dewatering in underground mines, toxic waste

heaps, atmospheric emissions of sulfur, nitrogen and carbon dioxide, land subsidence). The negative aspects of oil shale energy could be reduced by a transition from pulverized combustion technology to circulating-fluidized-bed combustion technology. In shale oil processing vertical retorts will be changed with solid heat carrier devices. New technologies should have higher thermal efficiencies, larger oil yields and produce much less atmospheric emissions and wastes. In the coming years all environmental parameters in the Estonian oil shale industry should be in harmony with the requirements of EU directives.

1. Introduction

Oil shale deposits range from early Palaeozoic to Cenozoic in age. The reserves of oil shale in the world are immense, some 10^{13} tonnes,[1] exceeding the resources of other solid fuels (coal, lignite, brown coal) combined. Over 600 deposits are known located in more than 30 countries on all continents.[2] The largest resources are known in the USA (72%), Brazil (5.4%), Jordan (4.2%), Morocco (3.5%) and Australia (2.1%). World oil shale science and technology have a long history, however with considerable ups (around the World War II and after the Arab oil embargo) and downs. Depletion of oil reserves and emphasise on energy security can be expected to give an impetus to oil shale research in the near future. Hopefully Estonian achievements can demonstrate to other oil shale mining countries that shale oil production will be increasingly profitable and that power generation on the basis of oil shales is efficient.

2. Composition of oil shale

World oil shales vary widely in their content and composition of organic compounds – kerogen. Presently there is no unanimously accepted lower limit of sapropelic organic matter percentage for oil shales. The lower limit varies from 5 to 10% and the upper limit from 60 to 70%. It is difficult to find two analogous oil shales, mainly because of their extremely variable deposition conditions as well as post-sedimentation alterations. Besides, the composition and quality of oil shale vary also within the limits of one deposit. Depending on the character of the deposit, there could be considerable variations among results obtained by analysis of different samples taken from the same deposit. In order to take the best advantage of up-to-date analytical techniques and corresponding expertise, currently scattered at different laboratories worldwide, an oil shale sample bank should be developed.[3] This would make it easier to benefit from the progress made in engineering disciplines (fluid dynamics, thermodynamics, heat and mass transfer), chemistry and geochemistry, geology and mining of oil shales.

The Estonian kukersite oil shale seams contain organic matter, carbonate and clastic materials in various proportions. The carbonate material consists mainly of pore-filling micritic carbonate mud together with a variable content of fine to coarse skeletal debris. The carbonate content (mainly calcite) ranges in different kukersite seams from 20 to 70% and the organic matter content from 10–15% to 50–60%. The clastic matrix is composed mainly of silt-size quartz and illite, the minor clastic minerals are feldspars and chlorite, pyrite is a rather common authigenic mineral in kukersite oil shale.[4] The organic matter of Estonian oil shale (kerogen) represents a mixture of high-molecular polyfunctional organic compounds, the real structure of which is still a subject of studies.[1]

In fact Estonia has two different oil shales: kukersite and graptolitic argillite, also known as *Dictyonema* argillite or *Dictyonema* shale. The latter is a dark, blackish- or greyish-brown fine grained radioactive claystone with a low organic matter content (15 to 20%) and calorific value (4.2–6.7 MJ kg^{-1}). Argillite is at its thickest (more than 4 m) in western Estonia and its resources are estimated at 60 billion tonnes.[5] It contains several rare elements, among them molybdenum (up to 600 g t^{-1}), vanadium (up to 1200 g t^{-1}) and uranium (up to 300–400 g t^{-1}). From 1949 to 1952 graptolitic argillite was mined at Sillamäe in NE Estonia for the production of uranium. Of some 250 000 tonnes of ore unearthed, more than 60 tonnes of uranium compounds were produced. Later the Sillamäe plant switched to processing imported raw material.[6] Graptolitic argillite is undoubtedly a potential complex mineral resource.

3. The Baltic Oil Shale Basin and its resources

The Baltic Oil Shale Basin is situated mostly in NE Estonia and extends eastwards into Russia, covering a total area over 50 000 km^2. The basin is administratively divided into the Estonia and Leningrad fields, comprising two currently mineable (Estonia and Leningrad) deposits and the prospective Tapa deposit. To distinguish Estonian oil shale from the other kinds of oil shale in the world it is called kukersite (from the word Kuckers, the German name for Kukruse manor, where the rock was first described). The main kukersite sequence belongs to the Kukruse Regional Stage of the Upper Ordovician[7] and contains up to 50 individual kukersite beds, which have been traced over 250 km in an east–west direction. Unlike most other oil shales, kukersite beds contain a rich and diverse marine faunal assemblage indicating the presence of well-oxidized bottom waters at the time when kukersite organic matter was accumulated.

The Estonia deposit (Fig. 1) is the largest explored and commercially exploited oil shale deposit in the world; its total resources exceed 7 billion tonnes of oil shale. The resources of the prospective Tapa deposit are of the order of 2.6 billion tonnes. The mineable seam in the Estonia deposit consists of seven kukersite layers and four to six limestone interlayers at a depth of 0–100 m with a thickness of 1.4–3.2 m in an area of 2884 km^2. The energy rating of the bed is 15–45 GJ m^{-2}. The criteria of oil shale reserves are: energy rating, calorific value of the layers, thickness and depth of the seam, location, available mining technology, world price and transportation costs of the alternative fuel, oil shale mining and transporting cost, nature protection areas and other limiting factors for mining.[8] According to the calculations of Estonian Natural Resources, the oil shale reserve is some 5 billion tonnes (economic reserve ~1.5 billion and subeconomic 3.5 billion tonnes). At present oil shale is used for electricity generation in power plants, shale oil production and in small amounts for cement production.

Fig. 1 Location map of Estonia and Tapa oil shale deposits and Narva power plants.

In Estonia technologies for oil shale mining and exploitation have been continuously developed for more than 80 years. Since the 1960s, Estonia has been the largest oil-shale producer and consumer in the world. Until 1960, the main oil shale consumers were the Kohtla-Järve and Kiviõli shale oil plants and the railway. Fine oil shale was used as a fuel at local power stations. Later large power stations using oil shale were installed in Narva—the Balti Thermal Power Station in 1966 and the Eesti Thermal

Power Station in 1973. This altered the structure of oil shale consumption: now ~80% of mined oil shale was used for producing energy. Oil shale production reached its peak in 1980, when 31.3 million tonnes was mined. Now oil shale production has stabilized at a level of some 15 million tonnes per year. However, crude oil prices in the world market strongly influence the Estonian oil shale industry. Therefore, the mining capacity needs regulation because the production of oil shale and its use as a raw material in the oil and chemical industry and power engineering could cause serious environmental problems.

4. Improvement of mining technology and utilization

Currently, kukersite oil shale in Estonia is mined in six underground mines and in three open-cast pits. Until the 1990s, in underground mining longwall mining techniques were widely used; where the bed is mined with a coal cutting shearer-loader and the roof is temporarily supported by hydraulic jacks. This method was productive but capital-intensive and it caused noticeable changes in land surface. All reasons which led to the abandoning of longwall mining in the 1990s to be replaced by pillar mining.[9] In pillar mining the roof and mined out land are supported with pillars of unextracted oil shale (averaging 25% of the reserve). As a result the ground does not subside but losses of oil shale are rather large (Fig. 2).

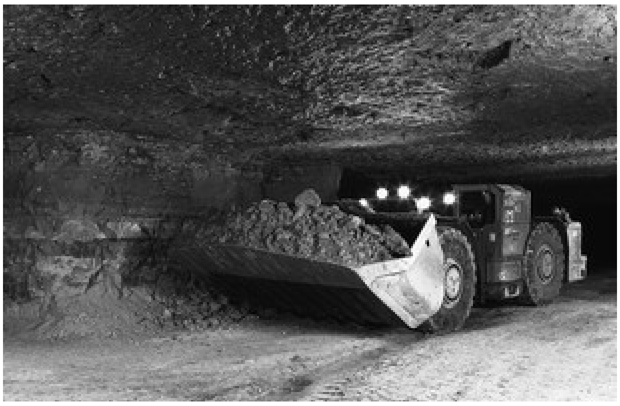

Fig. 2 In pillar mining, the roof and the mined out land are supported with pillars of unextracted rock, with significant losses of oil shale, some 25%. Photo by H. Bauert.

In surface mining technology developed after World War II large bucket (10–35 m^3) excavators, mainly draglines, started to be used. Both the overburden and the bed were first broken up by blasting. Stripping was done with smaller excavators in open casts with a thin overburden using front end loaders and hydraulic excavators. The overburden was transported with frontend loaders and trucks. In 2006 highly selective extraction was started, using a milling cutter surface miner from the German Company Wirtgen. The surface miner breaks, crushes and loads material in one operation. The size of the particles of the extracted rock depends on milling depth and operating speed.[10]

Most oil shale mined in Estonia today is used as a feedstock for the production of energy. Eesti Energia AS (Estonian Energy) with its 500 000 customers and more than 8000 employees is the leading Baltic energy utility as well as one of the biggest companies in Estonia; it generates 95% of the electric power produced in the country. Eesti Energia owns the largest oil-shale fuelled power plants in the world, the Balti and Eesti Power Plants (Fig. 1) burn about 12 million tonnes of oil shale annually. Since 2004, a new circulating fluidized bed (CFB) boiler technology has been used for electric power generation. The new technology has met expectations in all respects. The performance and efficiency of boilers have risen and the percentage of harmful industrial contaminants emitted has fallen substantially and is now below Estonian and EU permissible limits. About 1.4 kg of oil shale is burnt in older (pulverized combustion) Narva Power Plants boilers to generate 1 kW of electric power; the new technology has decreased this figure to 1.17 kg resulting in substantial fuel savings.[2]

The total oil resource in the Estonian deposit amounts to 985.7 million tonnes.[11] Presently shale oil is mainly used for producing fuel oil, and small amounts go to the production of calcined petroleum coke and road asphalt. Also phenols, resins, glues, impregnators, tanning agents, mastic, and other products are produced. The composition of crude shale oil is distinguished by a high content of oxygen compounds in addition to aliphatic and aromatic hydrocarbons.[1] During the last decades, two methods were used for shale oil processing. The Kiviter process (vertical retorts with internal heating, some 1000 t oil shale per day) with the use of enriched oil shale ensures 15–17% oil yield. Unfortunately, large amounts of organic matter get lost with harmful semicoke, which accumulates in large waste piles (Fig. 3).

Fig. 3 Kiviõli semicoke pile in May 2001. Photo by A. Käärd.

The Galoter or TSK-140 process with solid heat carrier in which poorer fine oil shale is used has lower environmental impact and is recommended in the national development plan. Narva Power Plants plan to install two new TSK-140 devices thus bringing the daily throughput of oil shale to 12 000 tonnes. The Viru Keemia Grupp in Kohtla-Järve started to do the same and this brings the total number of solid heat transfer retorts to six with a total estimated throughput of some 6 Mt of oil shale per year in 2012. All this together with the vertical retorts would supply more than 7 million barrels of high quality shale oil per year.

5. Environmental problems

Before the collapse of the Soviet Union Estonia was one of the biggest uranium producers in Europe. During 1948–1990, more than 4 million tonnes of uranium ore was processed and over 100 000 tonnes of uranium were produced at the Sillamäe Plant in NE Estonia. At first *Dictyonema* argillite was used as a raw material. In 1952 this was replaced by a richer ore imported from Central Europe, mainly from Hungary and Czechoslovakia. In parallel, the plant started processing loparite, a radioactive mineral mined in the Kola Peninsula, in order to separate rare earth metals such as niobium, tantalum, *etc.* Loparite also contains uranium (about 0.03%) and thorium (0.6%). A total of some 140 000 tonnes of loparite was processed at the plant.[6] The wastes from processing were conveyed to the first marine terrace of Cape Päite near the plant and stored on the surface until 1959, when a waste tailings pile was established. The pile has been reshaped a couple of times and in 1969–70 it was expanded to its present size with an overall area of some 350 000 m^2 and an elevation of about 25 m above sea level. It contains *ca.* 12.4 million tonnes of various wastes, including 6.3 million tonnes of waste from processed uranium ore and 6.1 million tonnes of oil-shale ash mixed with waste from loparite processing.[12] The estimated amount of naturally occurring radionuclides in the depository includes some 1830 tonnes of uranium, 800 tonnes of thorium and 7.8 kg (3×10^{14} Bq) of radium. Previously stored under an open sky at the waterfront of the Gulf of Finland, help and funding from Nordic countries and the European Commission enabled this extremely hazardous pollution source to be made safe in 2008.

The thermal combustion of oil shale in power plants leads to increasing amounts of heavy metals, included radioactive ones, in the ash deposited on ash fields or emitted into the atmosphere. So the concentration of radioactive isotopes in the oil shale dumps, which cover more than 20 km^2 in NE

Estonia, is many times higher than the background values in soils, reaching 5.5 g T^{-1}. Annually about 50 tonnes of uranium is transported with ash to the ash fields. In 1970–1990 more than 200 000 tonnes of fly-ash was emitted into the atmosphere. The concentration of uranium in the fly-ash passing precipitators even higher, 10% more than in the ash transported to the ash fields. This means that yearly some 1.2 tonnes of uranium is emitted. Due to dry and wet deposition, some 90% of the pollutants remain within 30 km from the pollution source and some 1.1 tonnes of uranium is deposited annually in the vicinity of oil-shale fired power plants. Data from the 1990s[13] demonstrate that in this area the estimated radiation doses were the highest (20–30 μR h^{-1}).

Where the bedrock consists of argillite and is covered only by a thin Quaternary cover, a high radon risk exists. The average and maximum radon concentrations reached 60 and 630 Bq m^{-3} for Tallinn, 160 and 1400 Bq m^{-3} for Kunda and 304 and 880 Bq m^{-3} for Toila.[14] The highest measured indoor radon level was 6700 Bq m^{-3}.[15]

5.1. Impact on hydrology and groundwater

Oil shale mining is accompanied by a lowering of the water level and discharge of mine water into bodies of surface water. The total amount of drained water is formed from precipitation water, surface water, subsurface water (free surface groundwater) and pressurized groundwater, and mine water is actually part of the natural cycle of water; the components of the balance of water pumped out from open-casts and underground mines differ significantly.

To calculate the balance of mine water inflow, results of monitoring of the used mine water inflow and borehole water level as well as hydrogeological modelling were used. In the case of Narva open-cast, which consists of Viivikonna, Sirgala and Narva open-cast fields, analytical solutions based on hydrodynamic equations were used to estimate the groundwater component. The area of mines flooded is presently over 220 km^2, the volume of water in these mines is more than 170 mln m^3. Calculations of the components of the inflow balance of open-cast mine fields showed that pressurized groundwater makes up 10% of the total inflow, the proportion of free surface groundwater in different open-cast mine fields accounts for 6–34% and precipitation water makes up 66–84% in the depleted area. In closed mines the inflow of groundwater makes up 60% of the total water pumped out.[16] Mining activities have a direct influence on groundwater quality due to the use of machinery, blasting, fuel and oil residues, *etc.* The kinetics of the oxidation reaction changes significantly as a result of grinding rocks and access of air oxygen due to the lowering of the water level. Due to oxidation of pyrite which occurs in Ordovician rocks the water pumped out from mines contains large amounts of sulfates; at concentrations up to 500 mg l^{-1}. Sulfates are easy to analyse and therefore are good indicators to describe the movement of mine waters. Sulfates are a potential threat to the environment as in strongly anaerobic environments toxic hydrogen sulfide may be released.

Oil shale mining and processing have a complicated impact on surrounding landscapes, on their ecological state, matter cycling, development *etc.* Detailed studies were conducted in the Kurtna Landscape Reserve.[17] The Kurtna (Illuka) Kame field formed in a marginal area of the last continental glaciation during the Pandivere stage about 12 200–12 300 radiocarbon years ago. The Kame field has formed above a deep (up to 105 m in depth) buried Vasavere ancient valley, filled with Quaternary deposits. It is a typical marginal formation of asymmetric cross-section with a relatively high, straight and abrupt ice contact slope to west and gradually lowering and undefined slope to the east. The relief is especially varied due to numerous glaciokarstic hollows, in which most of the lakes were formed during the Preboreal chronozone. Thirty-nine lakes of different shape, size, drainage catchment, hydrological regime and trophic level exist in the 30 km^2 area. Two main factors caused by the oil shale industry have affected the state of these lakes, as well as the surrounding mires. Since the 1950s water levels have started to fall in 24 of the lakes because of drainage into oil shale open-casts, peat cutting in the north and east of the Vasavere valley, sand production in the Pannjärve opencast and water consumption in the Vasavere intake structure.[18] As a result, at the start of the 1990s the water level in the centre of the Kame field had fallen by 3–4 m (Fig. 4).

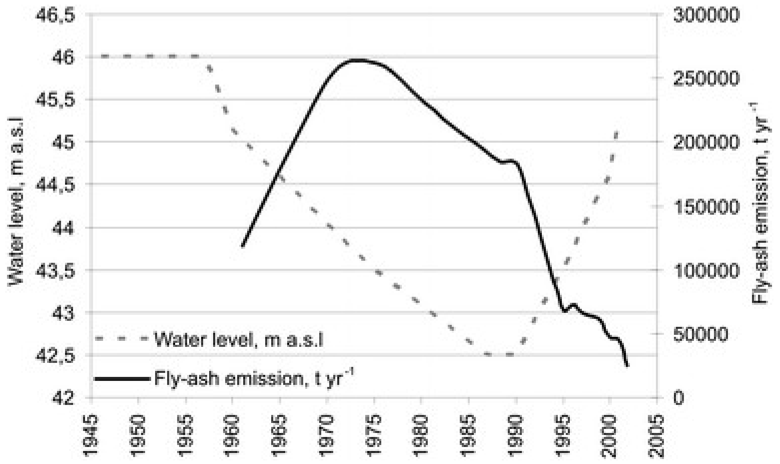

Fig. 4 In large areas of NE Estonia, landscapes have been affected by oil shale mining which has lowered the groundwater level by up to 3–4 metres. Power plants emit millions of tonnes of fly ash into the atmosphere.

5.2. Impact on the atmosphere

Combustion of oil shale in thermal power plants is another important factor that has affected ecosystems in NE Estonia during the second half of the last century. The first oil-shale fired power plant in NE Estonia was erected in 1937. Following this the Kohtla-Järve and Ahtme power plants were built in 1949 and 1951 correspondingly. The latter is located only 10 km from Lake Nômmejärv. Erected in the late 1960s and early 1970s the Balti and Eesti thermal power plants were much more powerful than the older plants. As a result of this increased power generation some 200 000 tonnes of fly-ash is emitted into the atmosphere annually.[19] Fly-ash deposition has resulted in an increase in the accumulation rates of several microelements in lake sediments and a decrease in their organic matter content. Therefore upper layers of the sediment contain excellent markers, which provide an exact time scale for studying the palaeobotanical and geochemical records (Fig. 5).

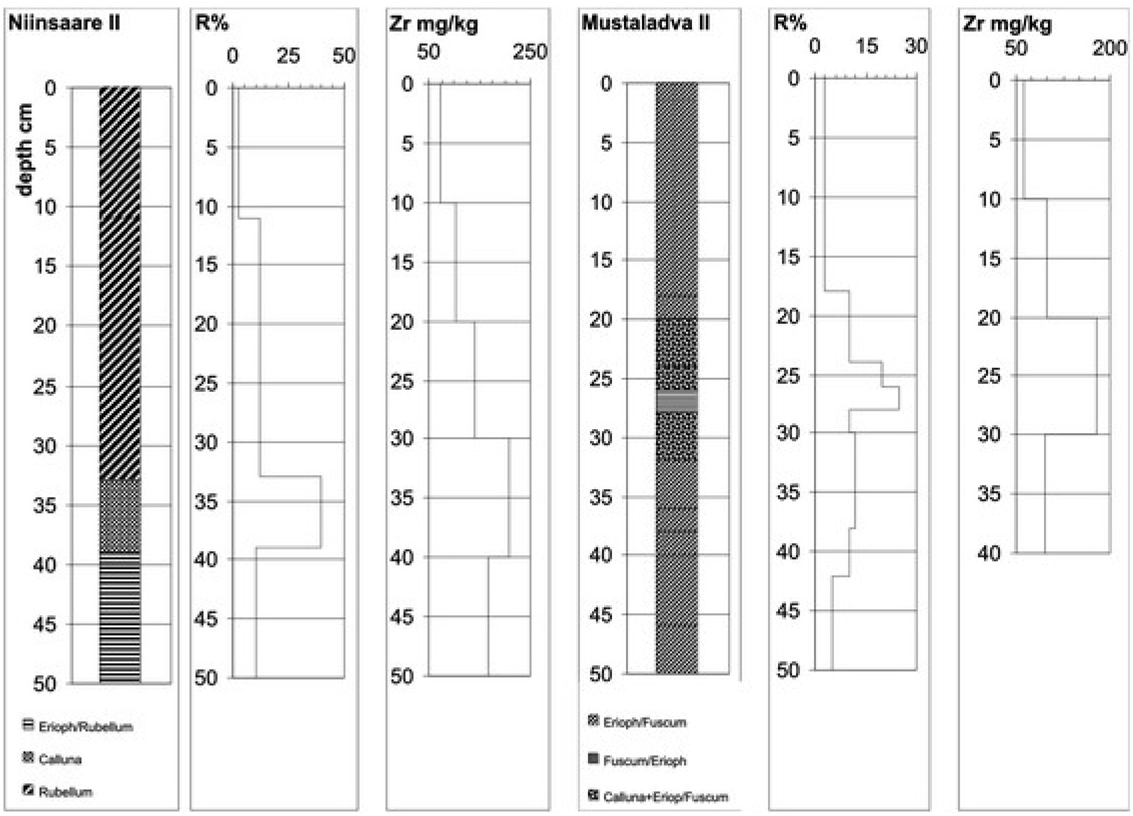

Fig. 5 Impact of atmospheric pollution on sphagnum cover of ombrotrophic peatland in NE Estonia. Stratigraphy, degree of decomposition (R%) and Zr content (mg kg^{-1}) in the topmost section of *Sphagnum* hummocks in the Niinsaare and Mustaladva bogs near the power plants.

Alkaline oil shale fly-ash is characterised by high concentrations of several heavy metals, carbonates, alkaline oxides, and harmful organic substances. The rapid increase and peaking of mineral fly ash particles and concentrations of various heavy metals (characteristic of oil shale ash) in the peat layers accumulated around 1976 is clear evidence of the atmospheric impact of oil-shale fired power plants. These maximums correspond also to the maximum content of ash in peat. The changes in the decomposition degree are in good temporal correlation in the sites situated closest to the emission sources; with increasing distance and decreasing number of emitted spheroidal fly ash particles in peat, the decomposition degree decreases. The degree of *Sphagnum* peat decomposition shows good correlation with the ash content but the restoration time of the *Sphagnum* cover depends directly on the amount of accumulated fly-ash.[20]

Energy-related activities are the most significant contributors to Estonian greenhouse gas emissions. Estonia satisfies most of its electrical energy demand by using fossil fuels, especially oil shale, its share of total fuel consumption is approximately 68%.[20] Due to the high carbonate content in oil shale, the content of alkaline oxides, especially CaO, in combustion products is very high. The carbonate fraction contains 90.5% $CaCO_3$; 9,2% $CaMg(CO_3)_2$; 0,3% $FeCO_3$.

It is rather complicated to calculate the proportion of free CaO that will react with atmospheric CO_2. The limiting conditions are the diffusion coefficient of CO_2 in water, temperature and also the fact that after a certain time the CaO-rich ash will be buried by new layers of ash. The pH values of precipitation in NE Estonia are high throughout the year, the mean values of summer months being 5.9–7.5. The pH value of the fly-ash precipitated on electric filters is in the range 12.3–12.6.[17] The content of free CaO in fine-grained ash from cyclones and electrical precipitators is in general some 40.0%. This percentage decreases with the decrease in the disintegration level. Therefore we can presume that the content of CaO in fly-ash is *ca.* 30% and its total annual emission into the atmosphere is some 50 thousand tonnes.

6. Summary and outlook

There is no other country in the world that can supply almost 80% of its electric power needs from oil shale. A reorganisation of the economy after Estonia regained its independence in 1991 has led to a continuous decline in the production of oil shale and the use of more environmentally friendly technologies. As a consequence there has been a restoration of lake environments in the surrounding areas towards a more natural state. Water levels in lakes in Kurtna Kame field have risen by up to 3 m which approaches the pre-disturbance level, and in 2002 fly-ash emissions from power plants had decreased more than 10-fold compared with maximum rates discharged during the late 1970s. Sustainable exploitation of oil shale in Estonia must be based on reforms of instruments and institutions, which should include technical, administrative, economic and environmental measures. New technologies should have higher thermal efficiencies and produce much less atmospheric emissions and wastes.

Research into sustainable mining is needed to preserve the natural environment. As large-scale tests are complicated, these studies should be performed by computer modelling. Oil shale mining causes a large number of technical, economic, geological, ecological and legal problems and therefore risk management is necessary. Risk management methods are focused on health and safety problems. In 2008 the Government of the Republic of Estonia accepted a State Development Plan of Oil Shale utilization up to 2015. In the compilation of the plan all counties, towns and rural municipalities of NE Estonia, nine large enterprises, 14 research institutions and 16 non-governmental organizations participated. Analyses showed that the negative aspects of oil shale energy could be significantly reduced by a transition from pulverized combustion technology to circulating-fluidized-bed combustion technology. All environmental parameters should then be in harmony with the requirements of EU directives. After the installation of two 215 MW units with new boilers in Narva Plants, emissions of carbon dioxide, nitrogen oxides and sulfur dioxides has fallen substantially. Other important tasks are to reduce the negative environmental impacts from the hydraulic transport of ash to the ash fields and subsequent release of highly alkaline ash field water into the surrounding environment.

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