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DICTYONEMA SHALE AND URANIUM PROCESSING AT SILLAMÄE*

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The present paper deals with the foundation and operation of the Sillamäe Metallurgical Plant in 1946-1989. It presents data on uranium production from the local Dictyonema shale as well as from the imported ore. Data on the processing of enriched uranium are also given. The article describes the work of the Narva pilot plant and construction of the waste depository in Sillamäe, which is an environmental hazard. The pre-investment planning of the initial remedial measures is presented.

The Estonian town of Sillamäe is situated on the southern coast of the Gulf of Finland at the mouth of River Sõtke, 172 km east of Tallinn and 25 km from the Russian border. Processing of uraniferous black Dictyonema shale in Sillamäe was started after the World War II. The earlier Swedish oil shale processing plant in Sillamäe was totally destroyed during WW II. The plant was rebuilt as a uranium extraction and processing facility (also called Industrial Combine No. 7; Facility No. 7; Oil Shale Processing Plant; enterprise P.O.B. P-6685; Sillamäe Metallurgical Plant; now *Silmet*).

The purpose of the plant was to produce uranium from the local Dictyonema shale. The first batch of Estonian uranium was produced in the town of Narva at the pilot plant (later known as the Cloth Dyeing Factory) during the winter of 1944/45. Construction of the Sillamäe plant began in 1946 and uranium production started at the end of 1948.

Everything that went on there was a strictly kept military secret. The word 'uranium' was a taboo and different names were used instead, such as product A-9, metal, tar, silicon, tin, lead, aluminum, etc. The names of the technological processes were changed as well (for example, uranium ore was named sand, filtration – separation, enrichment – moistening, etc.) and the

* Some aspects of this Report were presented at the NATO Advanced Research Workshop in Tallinn, Oct. 5–9, 1998.

chemicals (soda, sulfuric acid, ammonia, etc.) were named products 1, 2, 3, etc., and these numbers were often changed.

In 1946, together with the Narva pilot plant and the Sillamäe uranium facility, mining of the Dictyonema shale began. The first underground mine (Mine No. 1) began production in 1947 and continued to operate for five years (Fig. 1, striped area). In 1952, the Sillamäe underground mine was shut down because better ores were found elsewhere. The total quantity of the Dictyonema shale mined in 1948-1952 was 271,575 tons.

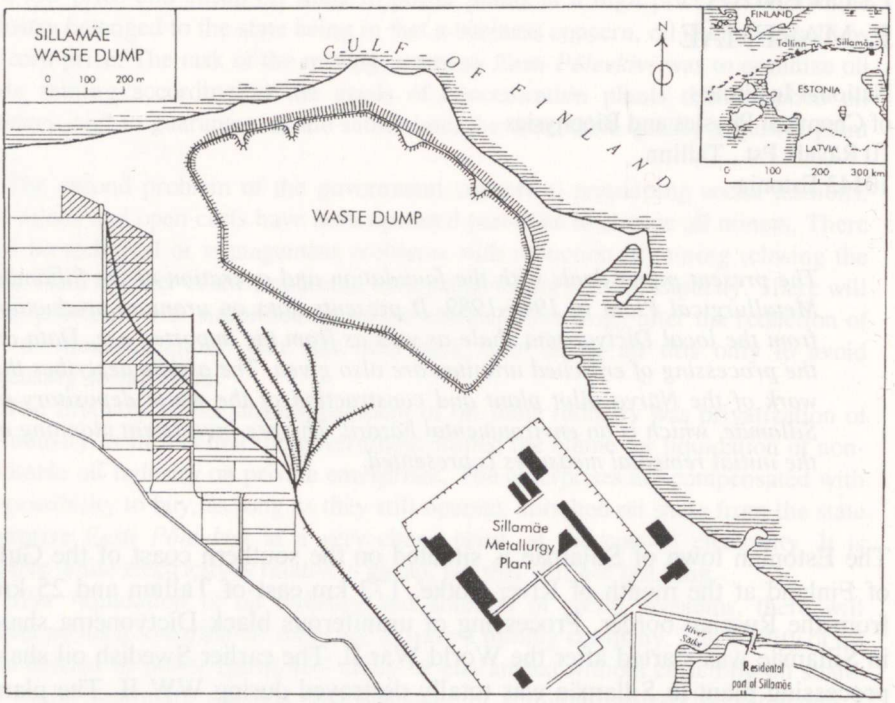

Fig. 1. Sillamäe waste dump and underground mine (striped area)

Uranium production from this shale was 22.5 tons of elemental uranium (while the final product was 40 % concentrate). Later production at the Sillamäe facility utilized richer uranium ores and concentrates from Central Asia, Hungary, Romania, Poland, and particularly Czechoslovakia and East Germany. Uranium production from imported uranium ores and concentrates (mostly as U_3O_8) was enormous; an amount equivalent to about 98,681 tons of elemental uranium was produced in 1950-1989. In 1982, processing of enriched uranium fuel (2 to 4.4 % ^{235}U) and its reconditioning into UO_2 was started. Altogether 1354.7 tons of enriched uranium was processed. The production and processing of uranium from ores and concentrates at the Sillamäe Facility are shown in Tables 1-5.

Table 1. **Uranium Production from the Imported Ore at the Sillamäe Metallurgical Plant in 1950-1970 according to Accounting Data**

	Year									
	1950	1951	1952	1953	1954	1955	1956	1957	1958	1959
Quantity of processed dry ore, 10^3 t	21.9	39.0	68.2	102.9	133.9	142.6	162.8	185.2	224.3	237.1
Average uranium content in ore, %	0.176	0.181	0.242	0.389	0.616	0.620	0.639	0.668	0.630	0.509
Uranium contained in 40 % (1950-1952) and 80 % (1953-1970) chemical concentrates, produced from the ore, t	25.7	53.6	127.7	303.3	711.0	760.2	903.8	1085.6	1249.6	1091.5

	Year										
	1960	1961	1962	1963	1964	1965	1966	1967	1968	1969	1970
Quantity of processed dry ore, 10^3 t	238.0	241.8	252.3	258.1	386.8	389.1	266.9	194.4	139.3	102.8	81.58
Average uranium content in ore, %	0.510	0.520	0.510	0.511	0.359	0.362	0.523	0.703	1.01	1.402	1.831
Uranium contained in 40 % (1950-1952) and 80 % (1953-1970) chemical concentrates, produced from the ore, t	1133.1	1171.9	1203.5	1241.5	1300.5	1341.2	1340.1	1320.0	1357.5	1408.9	1465.4

Table 2. **Uranium Production from the Imported Ore and Imported Concentrate at the Sillamäe Metallurgical Plant in 1971-1977 according to Accounting Data**

	Imported ore							Imported concentrate						
	1971	1972	1973	1974	1975	1976	1977	1971	1972	1973	1974	1975	1976	1977
Quantity of imported raw material, t	21394	15309	18111	26788	33523	33354	10648	2918	3503	4318	4956	5942	6359	7771
Average uranium content of raw material, %	2.64	3.71	3.14	2.5	2.31	2.42	2.31	51.93	48.29	46.90	53.1	50.1	49.9	50.2
Uranium in U_3O_8 concentrate, produced from the raw material, t	542.2	557	558	650	766	799.3	253.4	1492.2	1683	2016	2612	2964	3178.9	3875.4

Table 3. **Uranium Production from Chemical Concentrates from the then German Democratic Republic and Czechoslovakia at the Sillamäe Metallurgical Plant in 1978-1989 according to Accounting Data**

	1978	1979	1980	1981	1982	1983	1984	1985	1986	1987	1988	1989
Uranium in the processed chemical concentrate, 10^3 t	4425.5	4587.8	4810.5	4844.1	5183.8	4689.9	4860.4	5064.6	4627.5	4527.2	4390.1	4407.0
Uranium in U_3O_8, produced from the imported chemical concentrate, t	4417.2	4574.3	4796.8	4822.6	5170.5	4656.4	4850.0	5055.0	4608.0	4514.0	4367.0	4306.0

Table 4. **Enriched Uranium Dioxide UO_2 Processing at the Sillamäe Metallurgical Plant in 1983-1989 according to Accounting Data**

Product	1983	1984	1985	1986	1987	1988	1989	1983-1989
2.0 % ^{235}U	34.623	79.109	98.606	53.489	103.547		40.119	
2.4 % ^{235}U	0	0	0	0	74.036		150.569	
3.0 % ^{235}U	13.190	25.504	0	0	40.347		66.082	
3.3 % ^{235}U	68.025	52.056	71.492	88.342	22.571		0	
3.6 % ^{235}U	0	0	0	19.026	0		0	
4.4 % ^{235}U	0	0	0	0	0	(planned 7.0)	0	
Uranium in enriched UO_2, t	115.838	156.669	170.098	160.857	244.501	255.900	256.770	1354.669

Table 5. **Total Uranium Production at the Sillamäe Metallurgical Plant in 1948-1989**

Production of uranium	Duration of production	Production	
		planned	actual
Uranium in 40 % chemical concentrate, produced from local Dictyonema shale, t	1948-1952	34.7	22.451
Uranium in 40 % chemical concentrate, produced from imported ore, t	1950-1952	187.0	207.0
Uranium in U_3O_8, produced from imported ore, t	1953-1977	23628	24514.5
Total from imported ore	1950-1977	23815	24721.5
Uranium in U_3O_8, produced from imported chemical concentrate, t	1971-1989	73591	73959.3
Total uranium produced from Dictyonema shale, imported ore, and imported concentrate, t	1948-1989	97440.7	98703.251
Uranium in enriched uranium dioxide UO_2 (2 to 4.4 % ^{235}U), produced from various enriched raw materials, t	1982-1989	1476.9	1354.669
Total uranium produced and processed, t	1948-1989		100054.920

Such large-scale production naturally led to considerable pollution and waste formation, the extent of which was further increased through extraction of niobium and tantalum from imported loparite ore starting in 1970.

Loparite, mined on the Kola Peninsula, contains niobium, tantalum and the rare earths; considerable quantities of thorium (5-6 kg/t) and uranium (0.2 kg/t); and mesothorium and radium. During early processing of loparite ore there was no market for the rare earths. These were dumped into a waste depository that turned into an unusual and ecologically ill-studied polluter.

The pollution of the depository with enriched uranium (^{235}U) is curious. This isotope is not quite as poisonous as ^{239}Pu, but is still a significant pollutant. During uranium extraction from the concentrate the allowed losses were only 0.15 %, the losses accepted during enriched uranium dioxide preparation, however, amounted to 0.74 % (later 0.60 %).

This inconsistency is further accentuated by the circumstance that during December 1989, immediately before uranium production closed, these losses reportedly fell abruptly to 0.004 % without any improvements in technology. Still more confusing is the split between the dominant forms of losses. During 1988 a total of 255.9 tons of enriched UO_2 was produced at Sillamäe with an average enrichment of 2.4 %. This is equivalent to 6.1 tons of pure metallic ^{235}U. In a report about plant effluent air pollution, a loss of 3.44 Ci of ^{235}U is stated. Regeneration filters, reported to have 98.98 % efficiency, leave 0.035 Ci or more than 16 kg of ^{235}U released to the air as fine UO_2 dust.

In order to evaluate the ecological and nuclear non-proliferation-related dangers through possible sidetracking of fissionable enriched uranium, scientists have some work to do. The situation is further exacerbated by the factory-reported pollution data, whereby 3×10^{14} Bq (7×10^3 Ci) of highly radioactive radium ^{226}Ra is in the waste depository. Seven thousand Ci of radium weighs 7 kg, but the corresponding amount of natural uranium amounts to only 21,900 metric tons, which is far less than the amount actually produced. Possibly most of the radium was already extracted in the then German Democratic Republic.

In December 1989, both above-mentioned uranium production lines were shut down, and uranium processing was discontinued at Sillamäe as a result of political developments in the Baltic States.

Construction of the Sillamäe waste depository began in the mid-fifties. Until this time problems of waste handling both in Narva and Sillamäe were unsolved. The Narva pilot plant had no provisions for waste storage during its operating years. At least until the end of 1949, all processing wastes were stored in the closed territory of the plant, situated within the boundaries of the town. Later the processing tailings (solid residues) were trucked to Sillamäe.

But in Sillamäe, in the early years of plant operations, the Dictyonema shale tailings were discharged directly into the sea - into the Gulf of Finland.

In 1948 a two-section washing launder was used which delivered solid wastes as well as process waters into the sea. In 1950 the solid wastes were mixed with seawater and the mother liquors in agitators and pumped into the sand box from where the diluted pulp was pipelined into the sea. In the same year, due to the usage of imported ores, a waste depository was planned. The Leningrad Projection Office, later Moscow Projection Office, broadened the existing storage area and existing construction of dams. The waste depository was sited near the sea, lying at 500 m from the industrial site and 1200 m from the residential part of Sillamäe.

Construction of the present waste depository began in the mid-fifties by filling a nearly 1/3-km^2 area of the existing storage area with oil shale ash and processing waste. The waste dump and sediment reservoir are shown in Fig. 2.

In May 1959, construction of the surrounding and intermediate dams of the "A" and "B" reservoirs was begun. The dams were built of local sand, gravel, production tailings, limestone scrap, building refuse, and trash. In the course of construction, the embankment of the dams was not impervious. Pumping of waste into reservoir "A" stopped in 1962 when the height rose to 12.35 m above sea level. Filling of the "B" reservoir continued until spring 1964 when it was stopped and filling of reservoir "A" began again. In August 1964, filling of reservoir "B" continued and by November construction of the reservoir "C" dam was started. It is possible that some waste was also dumped in the old Dictyonema shale mines (see Fig. 1).

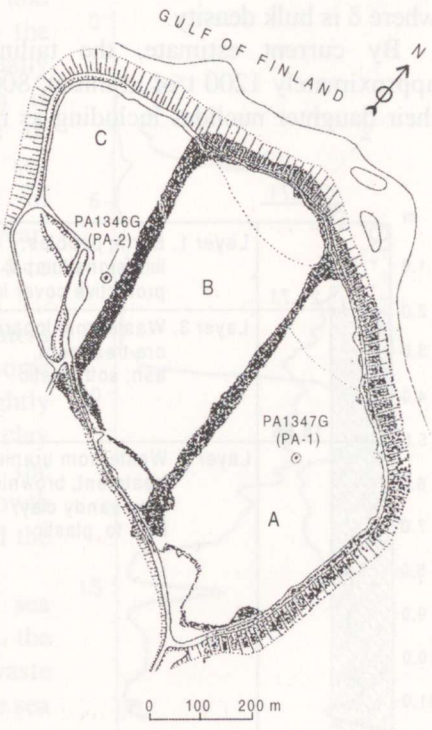

Fig. 2. Sillamäe waste dump and sediment reservoir

Up to 1996 the height of the dams was extended to 25 ± 0.5 m above sea level. The lower part of the dams consists of sand, gravel and ash layers that are cemented as the result of water filtration. The upper part consists mainly of gravel, building refuse and broken stones. Distribution of weak sandy clay and ash in the embankment is very irregular. By now the projected dam stability reserve has fallen to as low as 20 % in some sections and is thus nearly exhausted. There is a real probability of a dam failure that could be triggered by a small earthquake or explosion, in which case a large amount of

radioactive waste might be swept into the Gulf of Finland. Sillamäe is situated in a seismically active area. The last earthquake (about magnitude 3 in Richter units) has been registered near Narva in 1881 [1].

Three depositional layers of waste are differentiated in the waste depository. They are (Fig. 3):

Layer 3 – loparite ore processing waste consisting of grey sandy clay mixed with oil shale ashes from local power stations.

Layer 5 – uranium ore processing waste consisting of brown sandy clay.

Layer 6 – pebbles consisting of oil shale ashes and grey sandy clay.

The capacities of the three layers were calculated using the *Terra Modeler* and *Terra Quantity* programs. According to this layer capacities expressed in tons are as follows:

Layer 3	($\delta = \sim 1.4$ t·m^{-3})	4,000,000 t
Layer 5	($\delta = \sim 1.55$ t·m^{-3})	4,052,000 t
Layer 6	($\delta = \sim 1.7$ t·m^{-3})	648,000 t

where δ is bulk density.

By current estimate, the tailings in the waste depository contain approximately 1200 t of uranium, 800 t of thorium, and up to 90-120 kCi of their daughter nuclides including as much as 12 kg of ^{226}Ra. The γ-radiation level on the dump surface amounts to 100-1700 μR/h, in places as high as 2000-3000 μR/h.

The locations of the boreholes PA-1 (1347 G, depth 20.4 m, ∅ 89 mm) and PA-2 (1346 G, depth 16.5 m, ∅ 89 mm) are shown in Fig. 2. Borehole PA-1 was drilled in the sediment reservoir "A", the filling of which was started in 1959. Borehole PA-2 was drilled within sediment pool "C" which began to be filled at the end of 1964. Gamma-ray logging with the CK-1-74 well logging station was carried out in the boreholes. PCKY well logging apparatus with NaI (Cs) single crystal scintillation detector was used as a calibra-

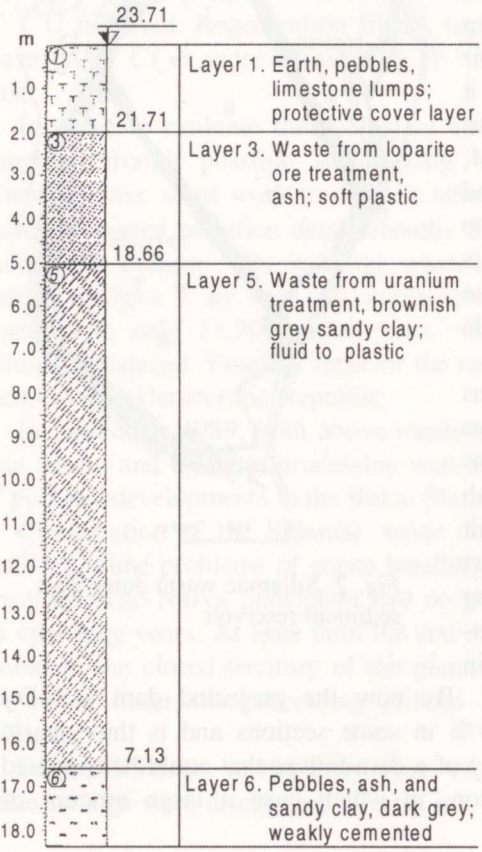

Fig. 3. Depositional layers of waste ore in Sillamäe waste depository

tion device, metrologycally checked and adjusted according to geophysical studies requirements in force in Estonia.

Only γ-radiation from the waste products surrounding the boreholes was recorded. Measurements for boreholes PA-1 and PA-2 are shown in Fig. 4, expressing the γ-radiation exposure rates (μR/h) in relation to recorded depth. The γ-radiation level in PA-1 reaches 7500-9000 μR/h at a depth of 19 m and in PA-2, 5000-7000 μR/h at 5-7 m depth.

The drill cores were taken to the laboratory [2] where they were sawn into pieces of 12 cm lengths. The drill core was colloidal sandy clay saturated with water that liquefied if subjected to vibration. The samples of drill core were placed and kept in 1.0 l hermetically sealed plastic containers.

The floor of the waste depository (layer 6 or, in its absence, layer 5) consists of natural gravel and pebbles underlain by a watertight layer of Cambrian blue clay. The clay layer dips 2.5-4.0 m/km southward.

The waters of the waste depository drain through the layer of gravel and pebbles into a ditch that empties into the sea. The amount of water flowing daily from the waste dump into the sea is estimated as 1600 m^3. The amount of dissolved nitrogen carried to the sea constitutes 1500 tons a year, that of mineral matter – 6000-12000 tons a year. Total dissolved solids ranges from 10.4 to 21.2 g/l and averages about 15 g/l.

Earlier studies show that waters percolating from the tailings carry some dissolved natural radionuclides that slightly contaminate beach sand and the blue clay layers. The high nitrogen content of the waste dump water has increased the growth of seaweeds in the coastal water behind the dam.

At Sillamäe, the currents of the sea move predominantly eastward along the coast. This is why the dissolved waste products carried from the dump into the sea or wind-blown towards the town are polluting the town of Sillamäe as well as its eastern beaches. Preliminary data show that the waste depository of the *Silmet* Metallurgical Plant is a real threat to the Gulf of Finland as well as for the town of Sillamäe and its environs for several reasons:

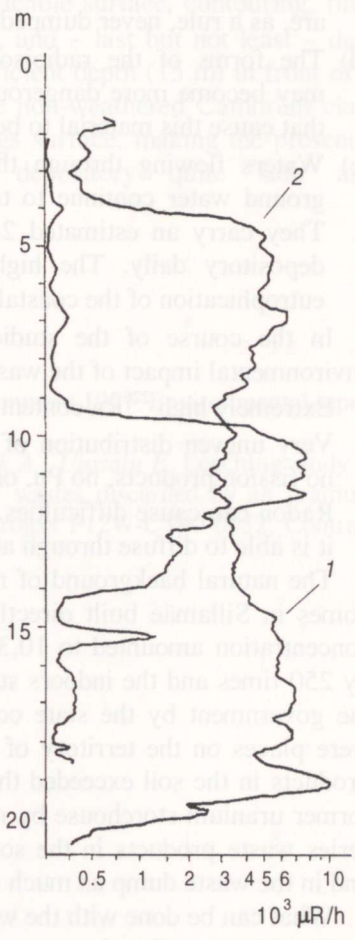

Fig. 4. Measurement results for boreholes PA-1 and PA-2

(a) About 6 million m^3 of radioactive (90 to 120 kCi) tailings lie behind the dam, the stability reserve of which is practically nil. Taking into consideration the potential for age flow and thixotrophy of the waste material, an explosion or earthquake could rupture the dam and release wastes to the sea.

(b) The tailings contain about 1200 tons of uranium and a large quantity of varied natural radionuclides. Radionuclides could spread into the sea-, surface- and ground waters or disperse as wind-blown dust into the surrounding environment, thus endangering people and the environment in general.

(c) Besides radionuclides, the tailings contain numerous other toxic compounds in the processed uranium and niobium ores and concentrates, such as arsenic and water-soluble rare earth salts. No data are available on the environmental impact of the latter, because these valuable salts are, as a rule, never dumped.

(d) The forms of the radioisotopes present, particularly radium isotopes, may become more dangerous with time through biochemical interactions that cause this material to become more soluble.

(e) Waters flowing through the tailings and carried out to the sea with ground water continue to transport pollutants into the Gulf of Finland. They carry an estimated 24 tons of mineral matter leached out of the depository daily. The high level of nitrogen in dump water causes eutrophication of the coastal seawater.

In the course of the studies, essential information for estimating the environmental impact of the waste was established. Among them:

- Extremely high ^{226}Ra content (12 kg in $RaSO_4$) in the depository
- Very uneven distribution of γ-radioactive nuclides in the depository, but no fission products, no Pu, or other transuranium elements were found
- Radon can cause difficulties. The half-life of ^{222}Rn is less than 4 days, but it is able to diffuse through at least 5 m of topsoil

The natural background of radon is about 40 Bq/m^3. In the basements of homes in Sillamäe built directly on Dictyonema shale, the measured radon concentration amounted to 10,300 Bq/m^3, exceeding the natural background by 250 times and the indoors standard by 70 times. The information given to the government by the state commission founded in 1989 stated that there were places on the territory of Sillamäe where the content of radium waste products in the soil exceeded the natural background by 85 times, and in the former uranium storehouse by more than 2000 times. The content of thorium series waste products in the soil exceeded natural background by 73 times, and in the waste dump as much as 160 times.

What can be done with the waste dump and its surroundings?

The highly polluted area extends about 300 meters from the waste dump dam. The polluted soil could be partially removed and the whole area covered with clean soil. The whole surface of the waste dump should be

paved or covered with special covering. Because the present operation of the plant is accompanied by hazardous waste production, these wastes should not be dumped into the old waste depository. Possible radioactive elements in the waste should be precipitated as insoluble substances, such as melted into glass or concrete blocks, and buried in special waste depositories as it is done elsewhere in the world.

In 1996 the Estonian government allocated funds for the preliminary reinforcement of the waste dump. The dam was heightened and the waste dump was reinforced on the seaward side with granite blocks, concrete, and limestone. The estimated cost of proper reinforcement of the dam is, of course, much higher and it is currently being planned under PHARE Multi-Country Environmental Programme.

The pre-investment planning of the initial remedial measures is described in Report PPEESD-9802 (Chemnitz 30.03.1999). It includes removal of free pond water, interim covering creating a trafficable surface, contouring, final covering of the consolidated tailings surface, and – last but not least – dam stabilization by placing a pile grillage of sufficient depth (15 m) in front of it and penetrating for 1 to 2 m into the stable non-weathered Cambrian clay. The final covering of the consolidated tailings surface, making the presently dangerous radioactive Sillamäe waste depository quite safe and environment-friendly, is foreseen in 2005.

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