THE VOLUMES OF SPENT OIL SHALE FROM ESTONIAN OIL-SHALE PROCESSING UNITS IN 1921–2002

R. VESKI[*]

Department of Oil Shale Research,
Tallinn University of Technology
5 Ehitajate Rd., Tallinn 19086, Estonia

It is suggested that there are about 86 million t (at Kiviõli 12.5, Kohtla-Järve 70.7 and Narva 3.0 million t) oil-shale semi-coke and coke deposited in the Estonian landfills since 1921. However, there is no explanation how these volumes were calculated and no data about the volumes of semi-coke formed at Vanamõisa, Kohtla-Nõmme and Sillamäe. According to the calculations based on operation of processing units, 74.7% of dry spent oil shale deposited in Estonia is located at Kohtla-Järve. These deposits (83.22 million t) consist of semi-coke and coke, 48.75 and 34.47, respectively. At Kiviõli mostly semi-coke is deposited – 19.2 million t (17.2%). Ash-like spent shale is deposited at Narva (4.34 million t, 3.9%), the landfills of other locations at Vanamõisa, Kohtla-Nõmme and Sillamäe hold 2.77, 1.09 and 0.7 million t of SC (2.5, 1.0 and 0.7%), respectively. This paper presents a method for calculation the volume of spent shale.

Introduction

The main processing units for shale oil production have been: various vertical gas generators (retorts), tunnel ovens, Davidson horizontal rotary retorts, and after the 1950s also large vertical generators (retorts), solid-heat-carrier (SHC) units and chamber-ovens, all of them of different throughput rate, number of similar apparatus, start-up and cease times. Most of them use(d) lump oil shale, but SHC units and Davidson horizontal rotary retort – shale fines as feed. In this paper the term 'spent oil shale' (SS) is used for semi-coke (SC), coke and processed shale from SHC units as a generalizing term only. Where possible, more exact terms are used.

Depending on operation of units the volume and properties of SS varied to a very great extent. For example, SHC units give spent shale almost free of organic carbon, as SC formed during producing shale oil was further

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[*] E-mail *rein.veski@mail.ee*

burned to obtain hot solid heat carrier for the next stage of raw shale processing.

The SS landfills (called in Estonia also 'semi-coke mountains', 'ash mountains' or 'ash hills') are classified according to § 8 of Waste Act (RT I, 26.02.2004, No. 9, P. 52; Register: RT I, 12.01.2004, No. 23, P. 15) as environmentally hazardous causing the largest environmental impact in Estonia.

It is generally suggested that there is about 70 million t of oil-shale SC and coke deposited in the Kohtla-Järve landfill in use (comprising also sulphur slimes, oil sludge and some other wastes) and three hills in Kiviõli containing the total of 14–15 million t of the SC. So, the total amount of SC and coke has to be approximately 70–80 million t [1], also 85 million t [2], more precisely 86,126,097 t (at Kiviõli 12.5, Kohtla-Järve 70.7 and Narva 3.0 million t) [3]. Sometimes the number of 100 million t of SS is mentioned [4]. There is no data about SC formed at Vanamõisa, Kohtla-Nõmme and Sillamäe. There is also no explanation how the referred volumes of SS were calculated.

As the estimated cost of closure of SS landfills is very high, approximately 3 billion EEK [2], it seems quite appropriate to calculate the volumes of SS formed in Estonian oil-shale processing plants since 1921 more precisely. The calculation is also important because SC and coke may serve as a potential raw material to various products or be used to produce heat or/and power. The amount of SS in deposits is calculated according to equation

$$SS = SS_{formed} - SS_{used} - SS_{exported} + \\ + H_2O_{chemically\ bound\ with\ SS} + CO_2{}_{chemically\ bound\ with\ SS}$$

There are only a few primary sources dealing with data needed to calculate the amount of SS formed or used, and no sources which deal with all components of the equation above.

Shale Oil Production

Data about the volume of oil shale used in Estonian oil shale processing industry for the period since 1921 and shale oil yields were needed to calculate the volumes of SS. The data published by Kattai [5] based mostly on Yefimov and his co-authors [6, 7]. In this study data published by Kattai were used. In cases when they differ from data by Yefimov, or Yefimov's data differ from those published earlier [8: year 1921] or in yearbooks [9: years 1922, 1923, 1925, 1929, 1931, 1932–1934; 10: years 1926–1929; 11: years 1930; 12: years: 1931–1933], earlier data were preferred. In most cases the preference was made to avoid approximation made in later sources.

Before World War II the volume of oil shale processed rose rapidly but stayed low. No data was available for the year 1945. The maximum volume

of oil shale processed was 4.499 million t in 1966, the volume of shale oil produced 563,200 t in 1961, and the highest yield of oil from oil shale processed according to data as given in statistical reviews or collected by V. Yefimov was 19.9% in 1937 (Fig. 1).
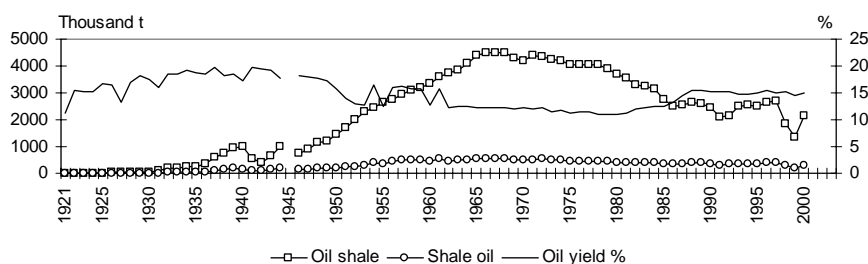

Fig. 1. Volume of oil shale processed, shale oil produced and oil yield in the oil shale processing industry during 1921–2000

Figure 1 reflects the progress in construction of processing units, in number of different units and changes in quality of feed shale processed. The period of long boom of Estonian oil shale processing industry is characterized also as the period of the lowest oil yield from oil shale due to chamber ovens (operated in 1948–1987) which produced from oil shale domestic gas to supply Leningrad and later Tallinn.
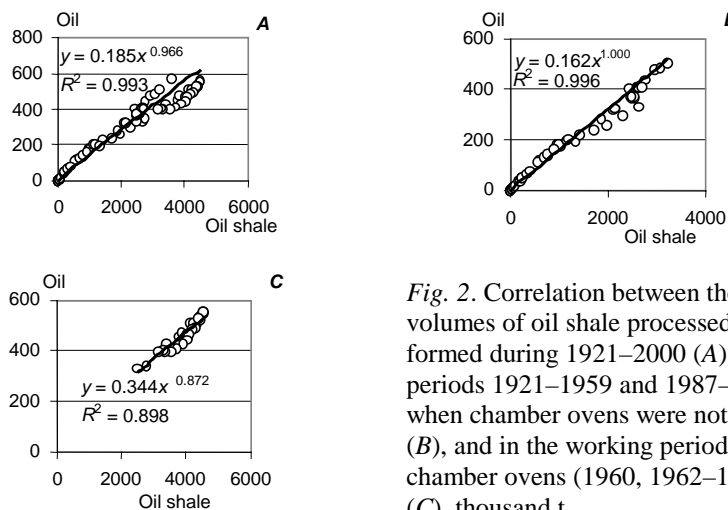

Fig. 2. Correlation between the volumes of oil shale processed and oil formed during 1921–2000 (*A*), in two periods 1921–1959 and 1987–2000 when chamber ovens were not in use (*B*), and in the working period of chamber ovens (1960, 1962–1986) (*C*), thousand t

Correlations between the volumes of oil shale used and oil produced in Estonia since 1921 (Fig. 2) turned out to be statistically good (R^2 0.993) (see Fig. 2*A*), correlation was especially good for two periods before and after operating of chamber ovens (R^2 0.996). For the time range when chamber ovens were operated (see Fig. 2*B*) the correlation was not so strong

(R^2 0.898), as the share of chamber ovens giving mainly gas and units giving mainly shale oil was changed as the new ovens were launched during the first period, and old ones were closed at the end of the period. Linear correlation was a little weaker compared with the power one, R^2 being 0.951, 0.983 and 0.896, respectively.

In spite of different manufacturing processes used in Estonian oil shale processing industry and changeable quality of feed oil shale, the volume of oil (y, t) is truthfully expressible according to the equation $y = 0.185x^{0.966}$ (see Fig. 2), or by linear equation $y = 0.129x$, R^2 0.938, where x is the volume of oil shale used, t.

We have to stress that in the Soviet period (1940–1991) and after 1991 no official statistical data about oil production was published. Calculations above became possible only thanks to V. Yefimov and V. Kattai. Since 1993 the official data about the volume of oil shale used in processing units is available from Energy Balances published by the Statistical Office of Estonia [13] as consumption of oil shale "for conversion to other forms of fuels". The 'other forms' mean shale oil which is used only for energy purposes and does not represent the volume of total oil produced in oil shale enterprises (Fig. 3*A*), as there is almost no correlation between the yields of fuel oil (Energy Balances) and oil (Yefimov) (Fig. 3*B*), R^2 being only 0.019.
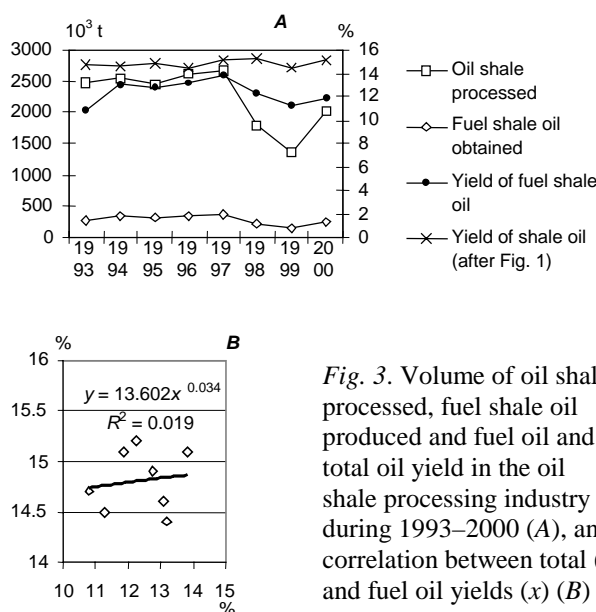

Fig. 3. Volume of oil shale processed, fuel shale oil produced and fuel oil and total oil yield in the oil shale processing industry during 1993–2000 (*A*), and correlation between total (*y*) and fuel oil yields (*x*) (*B*)

Semi-Coke: Two Scenarios of Calculations

There is no official or even non-official data available about the volume of SS formed annually before 1991. According to "The National Waste Management Plan" [2] and reviews of Estonian waste management in 2002 and 2003 [14, 15], the volume of SC formed in Estonian shale oil enterprises in 1993–2003 varies within 0.63 (in 1999) and 1.35 million t (1997) annually (Fig. 4*A*) as the amount of shale oil production was changing annually depending greatly on fuel oil price on the free market.
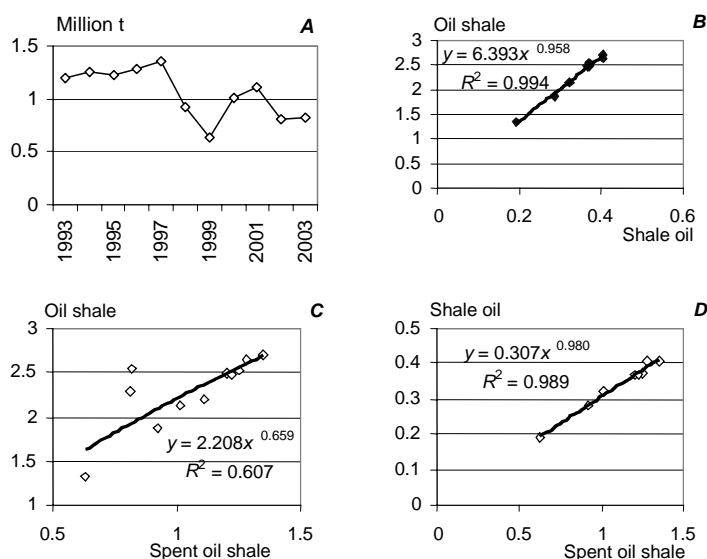
Fig. 4. Volume of SS produced in 1993–2003 (*A*), correlations between oil shale processed and shale oil produced (*B*), oil shale processed and SS obtained (*C*), and shale oil produced and SS obtained (*D*), million t

The correlation between shale oil and oil shale (Fig. 4*B*) represents the end of the correlation Fig. 2*B* with almost the same value of R^2. Unfortunately, the correlation between oil shale (volumes from Fig. 1) and SS (volumes from Fig. 4*A*) turned out to be not so strong (R^2 0.607) to justify the calculations of SS formed in the two periods (1921–1959 and 1987–2000) when chamber ovens were not in use. Fortunately, the correlation between the volumes of shale oil (volumes from Fig. 1) and SS (volumes from Fig. 4*A*) was quite good (R^2 value is 0.989), giving a strong argument to calculate the volume of SS for the periods chamber ovens were not used.

Table 1. **Oil Production Volumes and Yields, Yields and Properties of Dry SS Calculate Volumes of Oil Shale and SS by Units. Locations and Properties Units in 1921–2002 (after [5–7])***

Location/Unit	Shale oil 10^6 t	Oil % from		Spent oil shale			A^d %	OM %
		oil shale	Fischer retort oil	From shale %	MJ/kg	CO_2, %		
1	2	**3**	4	5	6	7	8	9
Kohtla-Järve								
Vertical retorts	13.161	16.4	74	58	*3.17*	*19*	*72.9*	*8.1*
Tunnel ovens	0.537	20.1	95					
Chamber ovens	2.736	4.9	22.6	63	5.8	10.6	73.8	15.6
Total	16.434							
Kiviõli								
Vertical retorts	2.314	16.5	74.5	58	4.18	17.7	70.3	12.2
Tunnel ovens	3.002	20.8	97.5	64.8	2.68	25.5	65.3	9.2
SHC units	0.296	13.3		48	*1.26*	*11.6*	*85.7*	*2.7*
Total	5.612							
Narva								
SHC units	0.999	13	77.4	53	1.26	11.6	85.7	2.7
Sillamäe								
Tunnel ovens	0.141	18.3						
Kohtla-Nõmme								
Davidson retorts	0.218	15.5	88.8	65	3.98	25.4	63.4	11.2
Vanamõisa								
Retort "Fusion"	0.6							
Total Estonia	24.004							

N o t e s : the figures in *italics* in the first 14 columns as compilation from other sources cited, in most using the values of this table, analogous units from the same table.

Disposed from Processing Units (first 14 columns of the Table), and Ratios to of Dry SS (the remaining columns) Formed at Estonian Oil Shale Processing

Spent oil shale contains, %				Reference (additional)	Shale/oil	Spent oil shale/oil	Oil shale 10^6 t	Dry SS from processing units 10^6 t			Spent oil shale PJ
Oil	C	H	S^d					Exit volume	Incl. OM	Incl. Oil	
10	11	12	13	14	15	16	17	18	19	20	21
Kohtla-Järve											
1.4	*6.7*		*1.8*	[16–19]	6.1	3.54	80.27	46.6	3.77	0.64	147.7
					4.98	3.23	2.67	1.73	*0.16*	*0*	*4.6*
0	*14*	*0*	2.4	[19–21]	20.41	12.9	55.84	35.18	5.49	0	204
							138.8	83.51	9.42	0.65	356.3
Kiviõli											
1.5	6.24		1.94	[19]	6.06	3.51	14.02	8.12	0.98	0.12	33.9
0.45	7.4	*0*	1.69	[19, 21]	4.81	3.12	14.44	9.37	0.85	0.04	25.1
0					7.52	3.61	2.23	1.1	*0.03*	0	1.3
							30.69	18.59	1.86	0.16	60.3
Narva											
0					7.69	4.08	7.68	4.08	0.11	0	5.1
Sillamäe											
					5.46	3.54	0.77	0.5	*0.04*	*0*	*1.3*
Kohtla-Nõmme											
0.75	7.72	0.3		[22]	6.45	4.19	1.41	0.91	0.1	0.01	3.6
Vanamõisa											
					6.45	*4.19*	3.87	2.51	*0.27*	0	10
							183.2	110.1	11.8	0.82	436.6

cases these figures as averages, the figures in the 2nd part of the Table (columns 15–19) are calculated

Correlations-based calculation of the volume of SS originates from the correlation between the volumes of SS and shale oil (Fig. 4*D*) and unit-based calculation of coke from chamber ovens. Shale oil production in 1921–2002 was 24.004 million t (Table 1), that of chamber ovens gave only 2.736 million t, so oil from other processing units was 21.268 million t and SS formed in other units than chamber ovens is according to the equation from Fig. 4*D* ($y = 0.307x^{0.98}$) approximately 70 million t.

The oil yield of chamber ovens was as low as 4.9% (see Table 1): to process one ton of oil 20.41 t of shale was needed and 12.86 t of coke was formed (63% from initial shale). So, for production of 2.736 million t oil 55.8 million t oil shale was used, and 35.3 million t coke formed. This gives the total volume of SS formed in Estonian oil shale processing units, which is, according to the mostly correlation-based calculation (70 + 35.3), 105.3 million t.

The unit-based calculation of SS is more precise as the initial data for calculation was mostly available. Unit-based calculation takes into consideration the specific characteristics of all processing units. Vertical retorts (common name in Estonia is gas generators) and tunnel ovens with oil yield from oil shale 16.4–20.8% (the highest, 97.5% from laboratory Fischer retort yield), respectively (see Table 1) were and still are the main oil producing units. Oil yield from oil shale in chamber ovens 4.9% corresponds only to 22.5% of Fischer retort yield, in our case it is important to stress that OM content of chamber oven coke is as high as 15.6%. The OM content of SC was lowest in SHC units – 2.7%, with no residual oil left in spent shale.

The highest oil content, according to Table 1, is 1.5% in SC from vertical retorts. The residual oil in SC from vertical retorts was sometimes as high as 6.5% (for example [23]). It is considered that oil content higher than 2% in SC of vertical retorts indicates serious disturbances in the technological process of retorts [24].

The yield of SS from feed oil shale (48–65% in Table 1), calorific value (1.26–5.8 MJ/kg), CO_2 content (10.6–25.5), ash yield (63.4–85.7%) and the other characteristics depend on the characteristics of the technological unit, as well as on the properties of feed shale. Lower CO_2 content of SS is typical both to chamber oven coke and SHC unit spent shale in different reasons: chamber ovens gave real coke which differs from SC of low-temperature processing by higher calorific value, and SHC units gave mostly ash-like spent shale with very low calorific value, lower CO_2 content in SS from both units was caused by higher processing temperatures and corresponding higher decomposition rates of carbonates compared with SC.

Despite great variations in composition of feed oil shale and rates of oil shale/shale oil (5.46–20.41) or SS/shale oil, the total volume of oil shale turned out, by our controlling calculation, to be almost the same (183.2 t, see row 17 in Table 1) compared with 178.2 million t after [5–7]. The difference in values calculated and collected is only 2.8%. Therefore, calculation of the volume of SS 110.1 t, and the content of OM and oil components in SS by the unit-based method (see Table 1) is justified. Volume of SS by unit-based calculation is higher than that obtained by the correlation-based method (105.3 million t). Both values calculated are higher than 85 or rarely referred 100 million t of SS [2–4].

Spent Shale Deposited

The volume of SS in landfills should be smaller than that formed in oil shale processing units, as some portion of it is used in Estonia or exported, and higher due to chemical reactions of hot SS with water and CO_2.

Oil shale coke was mostly used in production of mineral wool, and SC, to a smaller extent, to produce cement and compost. The total volume of mineral wool produced in Estonia from oil shale coke and waste bricks from

1959 (51,100 m^3) to 1985 (292,700 m^3) was 5,275 million m^3 [25–27] or 0.79 million t, as the average mass of mineral wool produced was 150 kg/m^3. In our calculations we proceeded from the 70% share of coke in feed and calcination loss of coke during mineral wool processing due to dissociation of remaining carbonates and burning of its OM (loss of ignition 32.3%) [28] giving (0.79 × 0.7)/0.677 = 0.83 million t of chamber ovens coke in feed.

According to [29], 9,366 t in 2001 and 10,013 t in 2002, totally 19,379 t SC were used for cement production at *Kunda Nordic Tsement* Ltd. There are some reports about the use of SC as an ingredient of composts (approximately 50%) in mixture together with pig slurry and peat [2].

According to Estonian newspaper "Äripäev" (09.10.1995, 08.01.1996, 17.04.2000 and 27.11.2000) also about 16,000 t compost was produced in 1994–1995, 30,000 t compost sold, and about 100 and 120 t sent to Finland, maximum total 23,110 t of compost SC component. Compost is also called soil improver. So, total use of SC in cement and compost (soil improver) making is 42,489 t. Total volume of spent shale to landfills was 110.10 – 0.85 – 0.02 = 109.23 million t (Table 2, 3rd column).

We have to stress that the data available about the composition of non-reacted SS (see Table 1) give only an idea of its CO_2 and OM content, but no direct information on the content of oxides and hydroxides. Dry spent oil shale disposed from processing units is commonly cooled down with water or transported to deposits by wet ash removal. So, to the oxides of SS exited mass of H_2O and/or of CO_2 must to be added to elucidate the final mass of dry SS in landfills. For example, according to the equation $CaO + H_2O = Ca(OH)_2$, the mass of calcium hydroxide yielded is 132% of the mass of calcium oxide, and that of calcium carbonate already 178% of calcium oxide. The mass of SS increases depending on the quantity of free oxides present in SS. As the share of oxides in SS is commonly not high, the increase in its mass is not high.

For example, there are some investigations made with the aim to reduce the degree of carbonate decomposition in vertical retorts to 20–25% [17]. The degree was 23–78% according to [30], but in the case of tunnel ovens only 5%. According to Table 1, SC of tunnel ovens was characterized by the lowest degree of carbonate decomposition, and so by the highest CO_2 content in spent shale – 25.5%, in the case of no decomposition of carbonates, CO_2 percentage in spent shale should be 26.8%. Other units gave SC with CO_2 content of 10–19%. If oxides in mineral matter of SS from all processing units will react only with H_2O, the mass of dry spent shale in landfills should be higher by only 0.21 million t, and when spent shale reacts with CO_2 completely, the mass of dry SS will increase by 1.09 million t, and is **111.32** million t (see Table 2).

Table 2. **Volumes of SS in Landfill Formed at Estonian Oil Shale Processing Units 1921–2002, million t**

Location	Dry not reacted (1)	Used (2)	(1)–(2)	When reacted with H_2O	When reacted with CO_2	With moisture (av.34.4%)	Share by locations, %
Kohtla-Järve	83.51	0.85	82.66	82.72	83.22	126.86	74.7
incl. chamber ovens	35.18	0.83	34.35	44.41	34.47	52.54	30.9
Kiviõli	18.59	0.02	18.57	18.65	19.20	29.27	17.2
Narva	4.08		4.08	4.13	4.34	6.61	3.9
Vanamõisa	2.51		2.51	2.51	2.77	4.22	2.5
Kohtla-Nõmme	0.91		0.91	0.91	1.09	1.66	1.0
Sillamäe	0.50		0.50	0.50	0.70	1.15	0.7
Total	110.10	0.87	109.23	109.44	111.32	169.77	100.0

The moisture content of SC, transported to landfills and/or cement factory, is also to be taken into account. SC transported from *AS Viru Keemia Grupp* (*Viru Chemistry Group* Ltd) to *Kunda Nordic Cement* Ltd contains 26–38% moisture (average 34.4%) [29]. So, the volume of moist SS deposited in landfills reaches approximately 167 (correlation-based calculation) or 169.77 million t by the unit-based calculation.

There are no investigations about the burning or composition of burned SC and coke in landfills, and almost no interest to solve this problem, although the phenomenon is well known to permanent residents of Kohtla-Järve [31, 32], while self-ignition of oil shale tailings and Dictyonema oil shale in Maardu dumps was a matter of scientific research. Nevertheless, inner burning of landfills may cause a partial or complete loss of the combustible part of the SC and coke and decomposition of carbonates in their mineral part as it occurs in SHC units (for composition, see Table 1).

In spite of the incompleteness of data to calculate the volumes of dry SS formed and deposited in landfills earlier, we have more or less trustworthy data about the volumes of SS by processing units, locations of deposited SS, and a scenario to do more accurate calculations in the future, if more precise initial data will be available, from archives, for example.

SC taken from landfills contains, according to special investigations, as much as 0.6–5% oil residue [33] (our data in Table 1, column 10: 0–1.5%). Therefore, the real oil content of SC in landfills may be much more than 0.82 million t (see Table 1, column 20). So, there is an urgent need to carry out a thorough study of SC landfills as man-made organic-rich geological formations.

Landfills of SS, especially at Kohtla-Järve, contain also other hazardous wastes, such as oil sludge (tar residue containing shale oil, water and solid particles) formed at oil shale processing, sewage sludge, sulphur slimes,

mineral oil wastes and acid tars. The volume and composition of these wastes may be a matter of further studies.

Conclusions

1. Correlation between the volumes of oil shale processed, shale oil produced and SS, and ratios of shale/oil and SS/oil by processing units were calculated, and two routes of calculations of volume of SS from 1921 until to 2002 were developed: (1) correlation-based and (2) unit-based calculation. These routes of calculations give an opportunity to control the results of the study and may be useful to specify the volumes of SS in the future if more precise initial data become available.

2. Dry spent shale (not reacted with water or CO_2) formed in oil shale processing units from 1921 until 2002 was 110.1 million t, 0.87 million t of which was used to produce mineral wool, cement and compost (soil improver), 109.23 million t was deposited in the landfills. The volumes of deposited dry spent shale after reaction with H_2O and CO_2 is 109.44 and 111.32 million t, respectively, with moisture content 34.4% – 169.77 million t.

3. Most of dry SC and coke formed in Estonia (with an allowance for reaction of oxides with atmospheric CO_2) is deposited at Kohtla-Järve (83.22 million t, 74.7% of spent shale deposited). 34.47 million t (30.9%) of this amount is coke from oil shale chamber ovens.

4. The next larger SS location is Kiviõli with its 19.2 million t of SC (17.2%) with some minor portion of ash-like spent shale from the SHC pilot plant. Ash-like spent shale is deposited also at Narva 4.34 million t (3.9%).

5. The other locations Vanamõisa, Kohtla-Nõmme and Sillamäe hold 2.77, 1.09 and 0.7 million t of SC (2.5, 1.0 and 0.7%), respectively.

6. The scenario used gives opportunity to calculate more accurate volumes of spent shale if more precise initial data will become available.

Acknowledgements

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