

Environmental impact from an alum shale deposit, Kvarntorp, Sweden – present and future scenarios

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Abstract During the Second World War it was decided to produce oil through pyrolysis of alum shale giving rise to waste products (coke and ash). Waste was deposited in the open pits and in a waste deposit. Due to the high remaining energy in the waste materials the waste deposit still today has significantly elevated temperatures (above 500 °C). Remaining pyrite in the waste material has also led to ARD with elevated trace metal concentrations. The waste deposit is no great environmental problem today but as soon as the waste pile cools off both the volumes of drainage and concentrations of trace metals will increase dramatically.

Key Words alum shale, trace metals, kerogene, temperature

Introduction

During the Second World War (WW2) there was a fuel shortage in Sweden and it was decided to produce oil through pyrolysis of alum shale in Kvarntorp, Kumla. The alum shale contains approximately 20% kerogene (organic matter) and 12% pyrite. Prior to pyrolysis the alum shale was crushed and the finer fraction (below 5 mm) was discarded (approximately 20%) to avoid sintering during the pyrolysis. Depending on the process used the waste products were either coke or ash. During the years 1942–1966 waste was deposited in the open pits but also in a waste deposit (fig. 1) close to the industrial area. It is estimated that this waste deposit consists of finer fraction (3 Mt), coke (2 Mt) and ash (23 Mt) with a total volume of 40 million m³ (total height around 100 m).

Combination of hot ash, pyrite and organic matter (kerogene) has led to significantly elevated temperatures (>500 °C) in the waste deposit and the processes are still active 44 years after the closure of the waste pile. Oxidation of pyrite can increase the temperature to 70–100 °C, followed by ignition of kerogene giving rise to temperatures close to 1 000 °C (Bharati et al. 1995; Puura 1998).

The aim of the present study was to study the temperature evolution of the waste pile, characterize the leachates from the pile and try to predict the future environmental impact on the surroundings.

Methods

Several methods, both geotechnical and chemical, were used in order to study the evolution of leachates from the waste deposit. Sampling of solid samples was performed through drilling in combination with installation of shallow ground water wells. Ground and surface water samples were also sampled.

Solid samples were analyzed for trace elements as well as major constituents. Leaching was performed in parallel at (i) pH 5, (ii) oxidizing conditions and (iii) reducing conditions in order to determine the potential leaching in the future.

Ground and surface waters were analyzed for trace elements as well as basic chemical parameters. Temperature measurements were performed in deep (20 m) steel pipes (n 21) on the waste deposit and through helicopter thermography (surface temperature).

Results and discussion

Solid phase

From the total concentrations of major elements a mineralogical composition was estimated (tab. 1) for both the original shale and the ash formed after pyrolysis. The major difference between the shale and the ash is that it is assumed that all pyrite has been converted into hematite and the organic matter kerogene has been driven off or combusted. It is also possible that some calcite has been converted into CaO due to the high temperatures during the pyrolysis. Neutralisation potential from calcite compared to the acid producing potential (pyrite) is to low (tab. 1). However,

Table 1 Calculated mineralogical composition of the alum shale and the ash from the process using old data from the literature, chemical analysis and a method developed by Puura 1998

	Alum shale	Ash
Calcite (CaCO_3) (%)	1.6	2.1
Pyrite (FeS_2) (%)	12	-
Hematite (Fe_2O_3) (%)	-	10
Illite (%)	31	42
K-feldspars (%)	13	17
Quartz (%)	23	29
Kerogene (%)	18	-

illite and other clay minerals will also take part in the neutralisation process (Puura and Neretnieks 2000). Due to the heterogeneous composition of the waste pile a whole range of different waters can thus be expected (from acid to alkaline).

Groundwater

There is a large span in chemistry in the 15 shallow ground waters around the waste deposit (tab. 2). Low pH waters are found in the areas where non processed shale has been deposited while high pH waters are found in areas where calcite has been converted to CaO . This large span in chemistry is expected from the heterogeneity of the waste deposit with different materials in different parts of the heap.

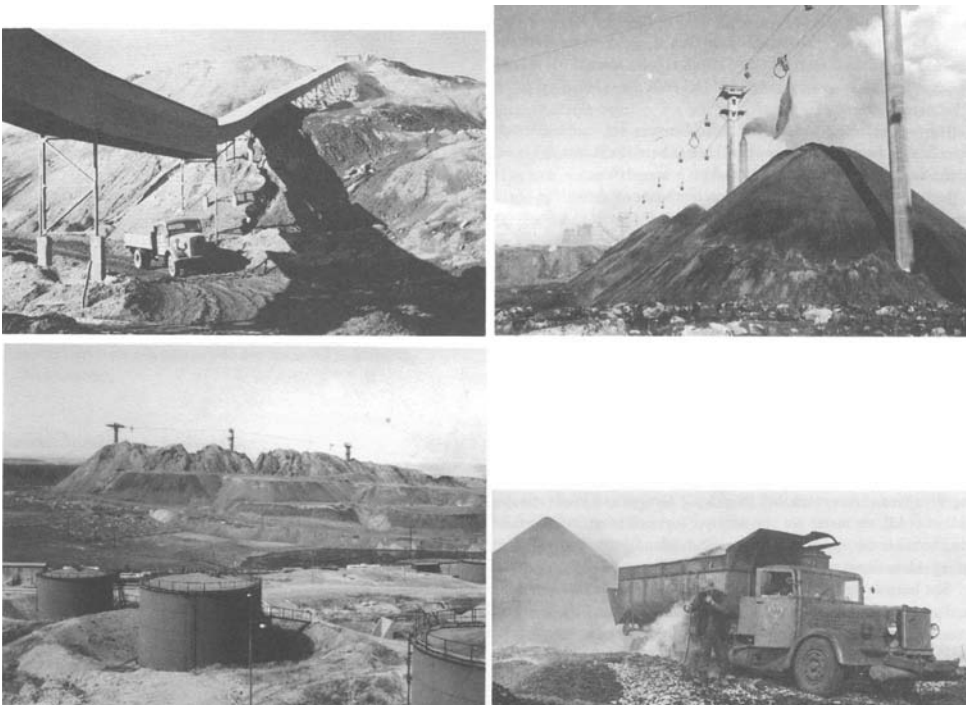


Figure 1 Overview of the waste deposit as it grew during the years 1942–1966. Methods for waste transportation varied through the years

Table 2 Ground water composition around the waste deposit. Three ground waters are presented of 15 representing the large span in chemistry

	0402 Acidic	0406 Neutral	0404 Alkaline		0402 Acidic	0406 Neutral	0404 Alkaline
Ca (mg/l)	484	511	698	As (µg/l)	2.00	113	2.09
Fe (mg/l)	328	0.01	0.00	Cd (µg/l)	4.02	0.31	0.24
K (mg/l)	237	105	213	Co (µg/l)	244	0.78	0.05
Mg (mg/l)	409	192	0.09	Cr (µg/l)	1.02	0.50	0.50
Na (mg/l)	92.5	26.4	66.3	Cu (µg/l)	6.50	1.00	1.00
Si (mg/l)	36.4	3.31	0.28	Mo (µg/l)	1.00	935	142
Mn (µg/l)	23 300	710	1.00	Ni (µg/l)	1 190	6.95	8.12
Al (µg/l)	19 800	13.8	2.00	Pb (µg/l)	1.50	0.20	0.30
Sr (µg/l)	288	1 480	1 890	Tl (µg/l)	0.13	0.31	0.03
Ba (µg/l)	10.1	22.0	97.7	U (µg/l)	129	1 760	0.50
pH	3.20	7.70	12.2	V (µg/l)	1.47	12.3	0.77
El cond. (µS/cm)	5 510	3 050	6 690	Zn (µg/l)	2 170	2.26	1.00
HCO ₃ (mg/l)	1.00	130	1 000				
SO ₄ (mg/l)	4 260	2 100	656				

Leaching

Results from the leaching tests indicate an increased leaching of several trace elements if the chemical conditions are changed (tab. 3). Probably will pH decrease slightly when water starts to infiltrate resulting in higher leachability for the trace elements. This behaviour is also confirmed in the ground waters with higher concentrations in the more acidic water even though many of the oxyanions (As, U, Mo and V) were found at their highest concentrations in the neutral ground water (tab. 2).

Temperature

Approximately 50% the temperature measurements performed in the waste pile had a flat profile with depth where the temperature did not exceed 50 °C. The other 50% had similar profiles, but with temperatures between 50 and 100 °C. However, a couple of profiles showed temperatures exceeding 500 °C (maximum range for the thermometer), indicating sustained kerogene oxidation (fire) at a depth around 15 m below the surface. These profiles agreed well with the ones performed in the early 1980s (700 °C at a depth of 5 m). It is, however, possible that the zone of very high temperatures today is found at greater depths than nearly 30 years ago (15 m today compared with 5 m in the early 1980s). It is suspected that the temperature interval below 100 °C is partly generated by pyrite oxidation while higher temperatures are generated by kerogene oxidation (open fire). Heat activity is still noted at the surface of the waste pile. For instance, during the spring of

Table 3 From the leaching tests estimated maximum leachable concentrations (mg/kg dw) and leachable amounts (tons) assuming lowering of pH and mainly oxidizing conditions. This is compared with the leaching at the moment at natural pH. The numbers should be regarded as rough estimates

		Ni	U	As	V	Cd	Tl	Mo
Leachable now	mg/kg dw	2.7	0.61	1.6	1.0	0.28	0.22	1.6
	tons	76	17	46	29	7.8	6.2	46
Max leachable	mg/kg dw	11	29	15	71	1.8	2.3	28
	tons	310	820	410	2 000	51	63	780

2010 large areas had to be dug out and treated to lower the surface temperatures. It has earlier been shown that the worst temperatures, from a local environmental perspective, are found in the range 40–100 °C, where the rates of pyrite and kerogene oxidation are high at the same time as there is plenty of water in the system (Puura 1998).

Assessment of future temperatures in the waste deposit is extremely hard since there is no good data on (i) the remaining heat in the entire deposit and (ii) the remaining chemical energy (in pyrite and kerogene) in the entire deposit. There is, in other words, no easy way to determine how long it would take for the deposit to cool down. Assuming the present heat energy (rough estimate from the temperature measurements) and no remaining chemical energy (conservative estimate) only cooling through heat conductance to the air, it would take approximately 100 years to reach below 100 °C (on average) and several hundred years to reach ambient temperature. Since this simple model doesn't take into account the remaining chemical energy the cooling time is probably much longer.

Future scenarios

Today the waste deposit has an elevated temperature and minimal runoff is formed compared to the evaporation. No significant runoff will be formed until the waste deposit reaches lower temperatures (below 70 °C on average) in approximately hundred years counted from 1966. It has been estimated that the volume of runoff will increase from 0.5 L/s today to 5 L/s when the deposit is cool. Chemistry will also change when the temperature has decreased. Higher rates of oxidation of pyrite and kerogene will take place (Puura 1998) in the presence of more water. This will lead to somewhat lower pH and increased (orders of magnitude) leaching of many of the trace elements (tab. 3). Increased runoff will increase the yearly mass flow with an additional order of magnitude.

Conclusions

Today the environmental impact from the waste deposit is relatively small and local. However, in 100–150 years the waste deposit will cool off and start to leach significantly higher loads of trace elements to the surroundings. Further studies on both the cooling and the chemical leaching are needed in order to be able to handle the probable increased trace element loadings in the future.

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