ANTHROPOGENIC AND NATURAL ANALOGUES FOR THE DEVELOPMENT OVER TIME OF MIXTURES OF WOOD-BASED ASH AND ACTIVATED SEWAGE SLUDGE

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SUMMARY: There are considerable incentives in Sweden – from environmental, conservation and economic points of view – to utilize ash and activated sewage sludge in covers for landfills. The Tveta Recycling Plant thus uses mixtures of these residues in the protection layer and ash in the seal. Others, however, use mixtures of ash and sewage sludge also for the latter, and this has prompted questions as to whether the Tveta Recycling Plant is actually complying with the requirements on best available technology. Times of hundreds and thousands of years are not accessible to ordinary experiments. Therefore, alteration of organic material over time has been studied in to historical sources (anthropogenic analogue) and in marine sediments according to modern sources (natural analogue). For the case of the Tveta Recycling Plant, neither alkalinity nor salinity was identified to protect a seal of ash and activated sewage sludge in the long term, and results on availability of oxygen were inconclusive. The anthropogenic analogue of Roman cements and similar supports the selection of solely ash as the best alternative for the seal.

1. INTRODUCTION

Many landfills in Sweden are subject to closure within the next ten years. It is estimated that the total area to be covered amounts to some 20 square kilometres and that the total volume of material may exceed 50 million tonnes.(Hedenstedt and Rihm, 2003) As a result of the recurrent glaciations, the soil cover in Sweden is generally very thin and it would indeed be difficult to find the magnitude of suitable material needed domestically.

According to the Swedish Environmental Code, ”*reuse and recycling, as well as other management of materials, raw materials and energy are encouraged*”. As a consequence of the cold climate in Sweden, in combination with the ambition to use recycle and natural cycle fuels, considerable volumes (≈ 1 Mtonne) of wood-based ash are generated every year. Thus, there is a considerable incentive from an environment and conservation as well as an economical point of view to attempt to use at least some of these ashes in covers for landfills. In 2001 the Landfill Site at the Tveta Recycling Plant was one of the first that was permitted to use waste as cover material.(Tham and Ifwer, 2006) The generic design used is shown in Figure 1.

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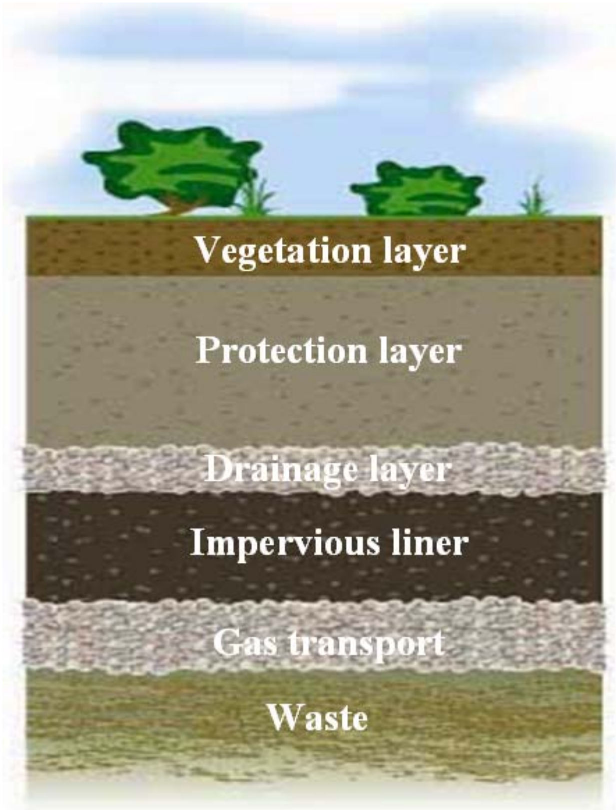

Figure 1. The generic design for covers at the Landfill Site at the Tveta Recycling Plant.
The *vegetation layer* comprises mainly compost. Typical thickness ≥ 0,3 m.
The *protection layer* comprises activated sewage sludge mixed with fine slag fraction of ash. Typical thickness ≥ 1,5 m.
The *drainage layer* comprises slag (bottom ash). Typical thickness ≥ 0,3 m.
The *impervious liner* comprises compacted ash, typically with a high content of reactive fly ash. Typical thickness ≥ 1,0 m.
The *gas transport layer* comprises bed sand. Typical thickness ≥ 0,3 m.
The *waste* is mainly domestic waste.
The total thickness of a cover is 3,5 – 4 m.
The fuel giving rise to the ash was a mixture of wood, plastic and corrugated fibreboard but also peat and wood chips.

The perhaps most crucial issue in this regard was to show that a seal made of ash is sufficiently impervious to water in order to meet the Swedish implementation of the European Union Landfill Directive (Tham and Andreas, 2008). Another task was to determine the health and environment related properties of the ash. (Adler et al, 2004, Sjöblom et al, 2006a and 2006b; Sjöblom, 2009).

Most of the characterization has been carried out at the University of Luleå, Sweden, and this work has defined the physical and chemical parameters of interest. The results have shown that due to initially high pH, adjustment of the moisture and mixing to obtain a suitable grain size distribution it was possible to obtain a liner built of ash within the cover such that it complies with the Swedish criteria for imperviousness. The results from the test area of four hectares show that a combination of bottom and fly ash can meet the requirements on a tight cover, corresponding to 50 litres per square metre and year for a landfill for non-hazardous waste and 5 litres per square metre and year for a landfill for hazardous waste. The Swedish Environmental Court has granted a long term permission to utilize ash and other waste materials such as contaminated soil, activated sewage sludge, foundry sand, e t c.

It is important that a cover is able to maintain its protective and sealing function over a long time. The Swedish Environmental Protection Agency has stated (Swedish Environmental Protection Agency, 2004) that a landfill for non-hazardous waste must maintain its intended function for several hundred years, and a landfill for hazardous waste for thousands of years.

Consequently, the ageing of ash and possible long term behaviour has been studied (Tham and Andreas, 2008; Sjöblom et al, 2006a; Sjöblom, 2009). The results indicate that ash cures with time to a hard but somewhat deformable solid and that the minor elements such as transition metals become incorporated into the phases formed by the major elements such that they become inaccessible to the pore water.

Comparison has also been made with Roman cements – an anthropogenic analogue for the behaviour over long times – and with coal ash, see (Sjöblom, 2008). These results also support the selection of only ash as the sealing material.

Other types of impervious liners have, however, also been tested in Sweden. An overview of the situation is presented in (Tham and Ifwer, 2006) where eleven landfills were investigated in order to describe different cover systems. The report shows that most covers are made of primarily ash and activated sewage sludge. It was also found that many of the seals comprise an intimate mixture of the two.

In view of this situation, questions have been asked whether a mixture of ash and activated sewage sludge might be a preferable option also for the Tveta Recycling Plant. This issue relates not only to what might be most appropriate from a business point of view, but also to whether the Tveta Recycling Plant actually complies with the requirements in the Swedish Environmental Code on use of best available technology (BAT).

It was soon found that the crucial question is whether a homogenous mixture of ash and activated sewage sludge might have a more positive prognosis regarding the long term functioning as compared to a seal made of only ash.

Times of hundreds and thousands of years are not accessible to ordinary experiments.

Therefore, natural as well as anthropogenic analogues that resemble the real system can be used in combination with efforts to understand the mechanisms involved.

Waste consisting of faeces, ash and food residues has been generated by civilisations for millennia. Thus, historical landfills and latrine pits (which contained ash as well) constitute an *anthropogenic analogue*.

Similarly, various residues including organic matter sediment onto the bottom of the sea. The surface of the sediments is contacted by oxygenated salty water. This constitutes a *natural analogue*, at least for the case of not very high pH-values.

These analogues have been studied in order to find a basis for the assessment of the feasibility of seals consisting of a homogenous mixture of ash and activated sewage sludge for the case of the Tveta Recycling Plant. The details of this work are being published in the report series of Ångpanneföreningen's Foundation for Research and Development, see (Sjöblom and Tham, 2009). The work has included compilation of information from a number of historical sources that are not generally available. They have therefore to a large extent been included in the appendices of the report. Almost all of this material is in Swedish.

The purpose of the present paper is to make the most important information in this report available to the international community.

2. LINER SEALS USING A HOMOGENOUS MIXTURE OF ASH AND ACTIVATED SEWAGE SLUDGE

Extensive research and development work has been carried out on utilization of activated sewage sludge for landfill covering purposes in Sweden, and a total of sixteen unique[1] reports are referenced in (Sjöblom and Tham, 2009), a couple of which are in English (Hermann, 2006; Hermann et al, 2006). The designs used are similar to that in Figure 1.

The most widespread use is in the protection layer. In some cases, sludge is used for the seal as well. Landfill sites having access to sludge from paper mills often use that while others use activated sewage sludge. Paper mill sludge and activated sewage sludge are chemically quite different and the present paper is limited to activated sewage sludge, which is generally available.

The investigations made show that tight seals can be obtained provided that the mixing is efficient. A number of tests have also been carried out on the long term properties, and it has been found that they are good in the time span available, i e in a decade perspective.

It has also been found appropriate that the fraction of ash is high (25 – 30 % figured on dry weights) such that the pH in the mixture is high, as well as the salt content and the buffer capacity.

Sundberg et al, 2002, have put forward that oxygen from the air cannot be more accessible to the activated sludge in the seal than what corresponds to the solubility in water in combination with the percolation. At an oxygen saturation degree of 10 %, only 0,1 grams organic material (figured as carbon) can become decomposed per square metre and year. This is compared with the seal, which has a thickness of 0,5 metres and contains 30 kg organic material (figured as carbon) per square metre.

It is also maintained that the high pH and the high buffer capacity of the ash gives rise to conditions where non-biological deterioration might dominate. In such cases, complexing agents might form. It is concluded, however, that their influence on any transport will be insignificant since the permeability is very low. (Wikman et al, 2005)

3. HISTORICAL SOURCES ALTERATION OF ORGANIC MATTER AND ASH OVER TIME ACCORDING TO

Decomposition of organic matter in soil is almost always biological in nature and takes place in such a manner that a metabolite from one organism is the food of another. The rate is strongly dependent on the chemical composition. It has been assessed that about two thirds of fresh dead matter becomes decomposed within one season, while the average age of humus in soil may vary between 250 and 1 900 years. (Stevenson, 1994)

It can thus be expected that alterations have taken place over time in historical landfills and similar, and that historical sources might be consulted for details. Consequently a number of historical sources have been investigated, and the results are documented in (Sjöblom and Tham, 2009). Most of these sources are in Swedish, but sources in other languages include (Berzelius 1808; Lavoisier, 1777a and 1777b; Leconte 1862; Stahl, 1748).

Mentioning of personal hygiene aspects appears sporadically in historic sources and there exist modern literature in which also such aspects of human culture are summarized. It is thus known that at least up to the 19th century, most of the human faeces fell into pits in the ground from where they might have been removed at a later stage for fertilization or saltpetre beneficiation purposes. Although ash was a valued raw material for a large number of uses, there

[1] In some cases the same report is issued by different publishers.

are reasons to assume that much of it was deposited together with the latrine in the towns.

It was discovered in the mid 19th century that cholera and other plagues might spread from latrine pits and into adjacent wells for drinking water (Heyman 1877). This gave rise to the introduction of fresh water through pipes together with water closets as well as a certain amount of literature on the topic.

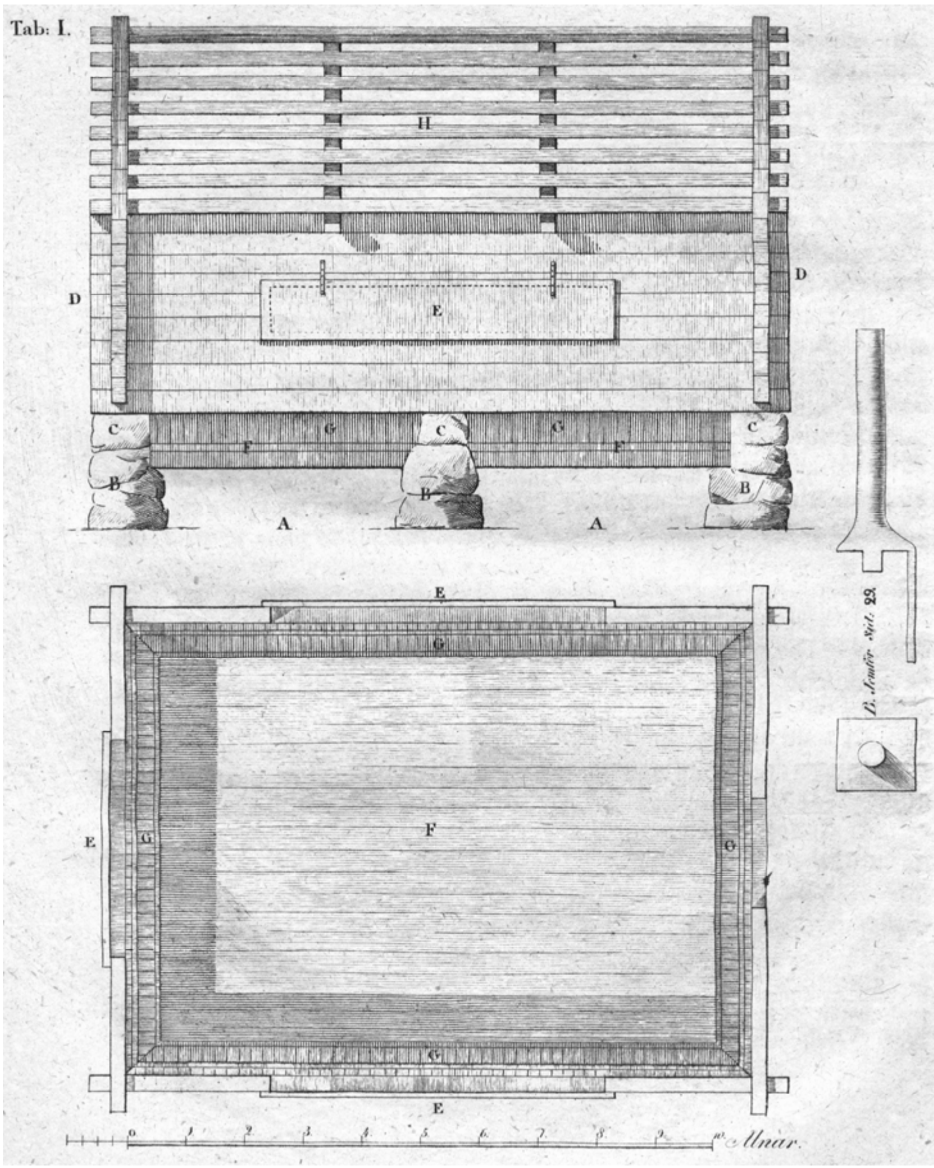

Figure 2. A saltpetre barn according to (Edman 1813). The barn was filled to a height of around one metre with a mixture of faeces, ash, mortar and organic structure material. Urine was recurrently poured over the bed. In this way, organic nitrogen was added at the same time as the bed was kept constantly wet. The bed was turned a few times every summer season. After five years or somewhat less, the process was completed. At this stage, the material might contain on the order of 6 kilograms saltpetre per cubic meter. The saltpetre was beneficiated by fractional recrystalization in which it was utilised that the desired product, potassium nitrate, had certain solubilities in water at low and at high temperature.

Gunpowder was introduced in Sweden already in the 15th century, and records from the early 16th century provide details on how saltpetre might be beneficiated from soil onto which animals had urinated. A century or two later, guns had become the dominating weapon in war, and the associated preparation of saltpetre from faeces, ash and what we today might call structure material had become one of the most important activities for a state in order to defend its integrity against the neighbouring enemies. Saltpetre, potassium nitrate, is the major ingredient in gunpowder (75 %).

Initially, saltpetre was beneficiated by leaching soil from under barns and stables, and by evaporation and fractional counter current crystallization of the leacheate. During the course of the 18th century this practice was shifted to dedicated saltpetre barns containing a mixture of faeces, ash, mortar and organic structure material, see Figure 2.

Ash (or ash in the refined form of potash) is an essential ingredient if the powder is to be used for more than fireworks. Sodium nitrate is deliquescent (hygroscopic) and must be avoided.

Sodium chloride can also constitute a problem since it has about the same solubility in water at low temperature as potassium nitrate. One important prerequisite for obtaining a sufficiently pure potassium nitrate was to add sufficient amounts of ash. Ash is namely rich in potassium, and also has a high potassium to sodium ratio.

Addition of ash was also essential to provide pH buffer capacity. Generation of nitrate is favoured by alkalinity while low pH-values instead favour the formation of dinitrogen oxide and nitrogen.

The literature generally indicates that nitrate is formed when organic matter and ash is left for any considerable time. This is the case also for "constructions" having dimensions on the order of meters, at least if a number of decades or a decennium or two are allowed for the process to take place.

Typically, oxidizing conditions in soil appear as a direct result of venting of air through open porosity. The compilation above shows that for cases in which such venting is constrained, flow of nitrate-rich water, or diffusion of nitrate ions, might instead be the dominating mechanism for transfer of oxygen equivalents.

4. ALTERATION OF ORGANIC MATTER IN MARINE SEDIMENTS ACCORDING TO MODERN SOURCES

The fate of organic matter in sea sediments is of great importance in geochemistry and geology.

The topic has been studied extensively, and excellent compilations of current knowledge are available, see e g (Burdige, 2006). Qualitative descriptions of the mechanisms involved are given in Figure 3.

The oxygen concentration in water is generally not higher than what corresponds to the partial pressure of oxygen in air at atmospheric pressure. Such oxygen is partially "converted" to nitrate in the decomposition process. The concentration of nitrate in terms of equivalents per volume sediment can become higher than that of oxygen, thus giving rise to a higher concentration gradient and a more rapid diffusion into the bed.

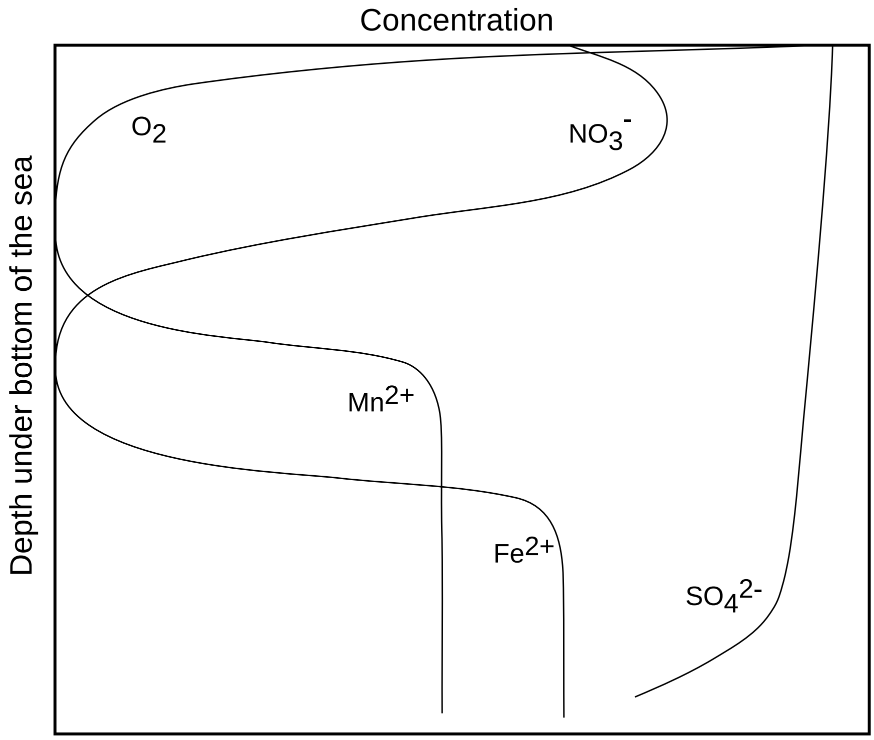

Figure 3. Oxidation of sea sediments. The bacteria usually prefer free oxygen. Nitrate formed in the decomposition can however diffuse further into the seabed than the oxygen (cf text). In the absence of oxygen, manganese may be reduced, and at lower redox potential also iron. At the lowest redox potentials, sulphate can become reduced in which case sulphur or hydrogen sulphide is formed. After (Burdige, 2006).

5. DISCUSSION AND CONCLUSIONS

The findings above will now be analysed with regard for relevance for the case of the Tveta Recycling Plant. Some general conclusions are also presented.

5.1 The significance of the alkalinity of the ash

At this plant, ash has been kept for a number of years in a heap, thus allowing for ageing of the ash. In 2002, drilling was carried out to investigate the state of the store and in order to characterize the material. At that time, the size of the store was estimated to about 600 thousand tonnes. Four holes were drilled at a total length of about 80 meters. Composite samples were taken every metre. It was found that the pH of the leach water was generally between 8 and 10, and the highest value was 11,3. Ten percent of the values exceeded 11 and twenty-five percent exceeded 10.

This might be compared with the pH-value obtained for leacheate of fresh ash which is typically somewhat below 12,7 which value corresponds to a saturated solution of calcium hydroxide in water. It is generally thought that lowering of pH in leacheate waste is associated with carbonation of the ash. A large number of measurements were made of the actual carbonate content of the samples and it was found that little carbonation had taken place. Consequently, carbonation could only marginally be responsible for the lowering of the pH. The lowering of the pH must instead have been caused by ageing phenomena. This issue is elaborated on further in (Sjöblom, 2009).

It is generally thought that microorganisms are largely dormant at pH-levels above about 10. Cases are known, however, where fungi may be active in soda-type soils up to pH 11 (Elíades, 2006). It thus appears that the pH provided by the ash at the Tveta Recycling Plant is insufficient to ensure any long-term durability of a liner with a mixture of ash and activated sewage sludge.

5.2 The significance of the salt content

Modern ash, including the one at the Tveta Recycling Plant contains not only the “traditional” salts in ash such as sodium and potassium chloride and carbonate, but also substantial amounts of sulphate from the flue gas cleaning.

The tolerance among microorganisms to salt is highly varying. The salt content in the Great Salt Lake in the United States varies between 5 and 27 % depending on location and season. Certain microorganisms as well as insects are accommodated to this environment.

Salts may inhibit biological activity by osmosis effects as well as effects of tightening the microstructure in the soil material. It cannot be concluded, however, that the salt in ash is sufficient to hinder biologic activity.

Instead, it is found that the sulphate present offers a high redox puffer capacity although the rate of reaction may be moderate and the incubation time perhaps relatively long.

5.3 The significance of the imperviousness of the seal

It has been put forward (Sundberg et al, 2002), as mentioned above, that a seal consisting of a mixture of ash and activated sewage sludge is so impervious, the penetration of water so slow, and the content of oxygen in the water so low that the oxidation of the sludge in the seal will be very slow and can be neglected.

This mechanism is, however, not identical with the mechanisms found and observed in seabed sediments and may not be in full concordance with the historical data on decomposition of organic material in mixtures with ash.

The present report is therefore inconclusive on this issue.

5.4 The significance and consequences for the Tveta Recycling Plant

When the best available technology (BAT) is to be selected, it is a requirement on a method that it will achieve the intended results with a reasonable degree of assurance. This requirement includes the behaviour in the long term.

For the case of the Tveta Recycling Plant, with the ash that is currently available, it appears daunting to attempt to accumulate sufficient evidence to prove the long-term feasibility of mixtures of ash and activated sewage sludge in a seal. It should be kept in mind that best available technology is a relative concept, i e comparison must be made with existing other options. Tveta Recycling Plant has accumulated considerable evidence to prove the concept of a seal made only of ash. This is relevant for the ash available to the Tveta Recycling Plant and may not be applicable to some other facility.

5.5 General conclusions

Behaviour of covers of landfills over long times is in general not accessible by ordinary experiments. Therefore, natural as well as anthropogenic analogues that resemble the real situation may be utilized in combination with theory. The present paper provides examples on such use of analogues for assessments of long-term behaviour.

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