

MILJÖRIKTIG ANVÄNDNING AV ASKOR

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Program area ENVIRONMENTALLY CORRECT USE OF COMBUSTION RESIDUES

## The Swedish Ash Programme 2002-2008

**Biomass, wastes, peat - any solid fuel but coal**

Henrik Bjurström and Roger Herbert



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A synthesis of the Ash Programme in English

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**Abstract**

In Sweden, producers of combustion residues have since 2002 implemented a collaborative applied R&D programme aimed at the utilisation of combustion residues (ash). The fuels are biomass, wastes, peat – any solid fuel but coal. In this report, the main lines of the programme are described: ash as a geotechnical material in civil works, in landfill construction and closure as well as recycling plant nutrients in wood ash. Technical and environmental questions have been addressed, with a slight emphasis on environment issues as non-technical issues are important.



## Executive summary

The research programme "Environmentally correct use of combustion residues" at the Swedish Thermal Engineering Research Institute (Värmeforsk) was initiated in 2002 in order to develop the knowledge needed to attain a utilization of combustion residues in Sweden. The programme, also called Ash Programme, has been extended twice, first 2006-2008 and presently 2009-2011.

The present report is a synthesis of work performed and results obtained during the second period, 2006-2008. In order to reach out into Europe, where the prerequisites for utilisation of waste are defined to an increasing extent, this synthesis has been written in English. For the sake of completeness, it was extended so as to include also achievements during the first period, where progress has already been described in a synthesis in Swedish.

In the world at large, combustion residues are often taken to be coal combustion by-products. Municipal waste incineration is the second industry producing such residues. Swedish characteristics are:

- Coal is not a commonly used fuel, partly because of the conversion of the Swedish energy system to a sustainable bio-fuel based system
- There are many types of combustion residues, resulting from a large number of fuel fractions, diverse technique of combustion (a.o., fluidised bed furnaces are more common than abroad) and by the small size of combustion plants from an international perspective, yielding small quantities of residues at each site

In 2006, approximately 1.3 million tons of combustion residues were produced, out of which 79 % were utilised. The largest volume is in civil works, mainly as a construction material in landfills.

An important Swedish use is compensating soils for the harvest of biomass in forests. For the use of woody solid biofuels to be sustainable, forestry must be sustainable. This implies a.o. that the mineral nutrients removed when harvesting biomass should be recycled to forest soils, e.g. as ash. The availability of nutrients for a continued good growth is important, but even more important is the buffering capacity of the soils: forestry is acidifying and even more so if all biomass is removed, as in whole-tree-harvesting.

Work within the Ash Programme has followed a few main lines, besides being complemented with investigations of chemical problems and dissemination of information:

- Covers for landfills and mine tailings
- Civil works, e.g. road-buildings, where both geotechnical and environmental questions have been addressed
- Cement and concrete applications
- Compensating soils for removing biomass and the mineral nutrients in the biomass

The emphasis of the Programme is on environmental questions, even if technical questions have been treated. The time perspective in this context is much longer than the 3-5 years that are usual in an applied R&D programme, i.e. decades after ash has been placed on a site, e.g. in a road, or spread to forest soil. New test fields have been created in the programme and old test fields have been evaluated in order to gather available information.

### **Combustion residues as a barrier against penetration of water**

A technical function provided by combustion residues is to prevent the percolation of water through the body of a landfill or a heap of mine tailings, where percolation could lead to dispersal of undesirable substances. In addition to having suitable functional properties, combustion residues can replace the virgin natural materials that are commonly used to this purpose. Fly ashes with their binding properties are the primary materials. Two types of sealing layers have been investigated:

- FSS, i.e. digested sewage sludge stabilised using fly ash; fly ash is mixed with the sewage sludge in order to raise the pH of the sludge and to prevent degradation of biological material in the sludge
- Ash as a monolithic sealing layer, in which the reactive binding properties of the ash are important

The Ash Programme has built upon preparatory work and prepared test surfaces in pilot-scale experiments on four landfills in the 2002-2005 period. In the second period of the programme the test surfaces have been monitored. The permeability of the layers is low, but monitoring needs to be continued. Guidelines for carrying out FSS sealing layers have been compiled.

Sealing layers that consist of combustion residues only are being used by *a.o.* Tveta Återvinning since 2000. Building these layers and monitoring programmes have been documented within the Ash Programme. The sealing layer fulfils the functional requirements through diffusional processes: particle size distribution, moisture content and reactivity of the combustion residue contribute to the minimization of the pore volume and consequently to percolation of water into the body of the landfill.

### **Combustion residues in a mining environment**

The need to seal a body of waste exists also for tailings impoundments and rock dumps in mining environments. In this case, entrance of both water and oxygen into the waste must be prevented: oxygen reacts with sulphidic ores to produce sulphuric acid that dissolves heavy metals; the leachate is often called Acid Mine Drainage.

The 80 ha impoundment at Gillervattnet contains waste sand from sulphidic ores. A test field has been established on 3 ha. In this field, the waste is sealed with a layer of digested sewage sludge on top of a layer of fly ash, or a layer of sludge and ash, in order to prevent atmospheric oxygen to reach the tailings sand. The role filled by the combustion residues is to raise the pH value and stabilize the layer against penetration by plant roots that are established in the sludge layer. Fresh ash is toxic to plant roots

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and a layer of hardened ash is too hard for the roots to penetrate. Mixed layers of sludge and ash are too soft for that. One should avoid plants with roots that produce carbohydrates, as there are indications that these carbohydrates attack the ash layer.

On the other hand, some rock dumps are cultural remains to be preserved and, in some instances, may not be capped. In experiments where high-pH fly ash was injected into rock waste, leaching could be reduced to 1 % of the leaching from an untreated heap. Combustion residues were also shown to be useable as reactive filters that neutralise leachates.

### **Combustion residues in civil works – technical questions**

In decision processes when selecting materials for civil works, alternative (i.e. waste-based) construction materials for civil works have a handicap in comparison with natural materials: their technical properties are different. However, designs using alternative materials and those using natural materials are functionally equivalent, and sometimes the former are better.

If one examines when combustion residues are produced, when roads are built, and the difficulty of delivering large quantities of combustion residues to works in a short time, roads are probably not the most interesting area of use. However, the body of functional requirements developed for roads makes the discussion of material properties and specifications as well as functional requirements easier.

At the inception of the Ash Programme, the properties that needed to be known in order to predict function of the design were defined. Primarily two types of combustion residues are interesting: fly ash, preferably from solid biofuels, in e.g. unsurfaced roads, and bottom ash in surfaced roads. The properties have been determined for a few sample residues. The programme of tests is quite comprehensive and also includes tests of leaching properties in order to determine their environmental impact. Evaluation of the properties is not an easy task, as there is in general very little experience of the behaviour of residues in nature and of the impact of ageing.

The key to the future is therefore establishing and monitoring pilot-scale and full-scale experiments. New roads have been built in the Ash Programme and old roads have been evaluated.

The utilisation of fly ash in non-surface gravel roads developed in Finland has been imported to Sweden<sup>1</sup>. In successive investigations, fly ash has been characterised, recipes for materials have been developed and a few sample roads have been built. The main results are that the bearing capacity and the freeze-thaw properties have been improved and the somewhat increased leaching of salts from one of the roads, caused by the presence of fly ash, is temporary. In order to make best use of the attractive properties of the fly ash and in order to save on ash resources, they should be used to stabilise poor or worn out road materials.

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<sup>1</sup> It should be mentioned though that Vattenfall in Uppsala had already been building non-surfaced roads using fly ash before the start of the Ash Programme.

MSWI bottom ash, i.e. aged bottom ash from municipal waste incineration, has been used in three experimental civil works before the Ash Programme was conceived: the Törringe road in Malmö, Vändöra within the premises of the Linköping utility and within the Dåva premises in Umeå. These sites have been monitored within the Ash Programme in order to collect experience for designing roads with combustion residues, e.g. determining the stiffness by deflectometry. MSWI bottom ash has 70 % of the bearing capacity of crushed rock, but also 70 % of the latter's bulk density, which makes packing easier.

Crushed rock is a very common material in road designs. Rock needs to be crushed in several steps for the particle size distribution to become optimal. However, MSWI bottom ash has a particle size distribution that supplements that of crushed rocks. A composite material, such as those studied in one project may yield savings in energy as well conservation of natural resources.

### **Combustion residues in civil works – environmental questions**

The most important barrier to the use of waste materials in civil works is not technical; it is the evaluation of environmental impact. That is also the main reason for the creation of the Ash Programme. The problem was that there wasn't any specific regulation and that guidelines and limit values for contaminated soil and landfills were used.

The first stage of work with a proposal for guidelines was reviewing the relevant regulatory framework. This applies irrespective of whether materials are waste or virgin materials. The status of waste leads to difficulties in the permitting and management steps, where difficulties could perhaps be avoided if the material, i.e. combustion residues, could benefit from an end-of-waste procedure. When end-of-waste has been reached, the material meets specifications and its quality is known on the market. Equally important is the fact that it no longer is a poorly defined waste. "End of Waste" discussions are now being pursued within the EU.

The intrinsic hazardous properties of a material are only part of the information required in an assessment of the risks associated with using it. Next step in the Ash Programme is an analysis of the risks for health and for environment. Two cases were studied by Bendz et al. using the methodology developed for contaminated soil:

- A non-surfaced forest road (a gravel road) with a rather shallow layer of ash
- A surfaced road with MSWI bottom ash in the subbase, i.e. a rather deep layer

All paths for dispersal from the body of the road and exposure for man or for environment have been described. The model is conservative, i.e. the scenarios used in the analysis are reasonable worst case and yield rather large safety margins. For example, a person is supposed to live all its life within 20 m from the road, to eat 30 % of its vegetables grown in the vicinity of the road, to drink water from its own well, walk on the road and breathe dust. When the road is not in use anymore, it is used as a recreation area by adults and children.

The results from the calculations indicate that the risks are dominated by dust dispersal from the road, not by leaching as it is usually assumed. The critical parameter is the arsenic content in the combustion residues which may not exceed 15 mg/kg in a decommissioned road. If account is taken only of leaching from combustion residues, the guideline values for content of e.g. heavy metals are significantly larger and should not present any barrier to use of residues in civil works.

Work with guidelines has been supplemented on several issues:

- Heavy metal uptake by vegetation has been investigated – the experimental material is lysimeters invaded by trees, a.o. birch, and grass, established in 1993 at SGI in Linköping
- The Swedish Radiation Protection Authority has issued an ordinance limiting the impact of caesium 137 on well water and recipient surface waters. A guideline value of 2 kBq/kg has been calculated for combustion residues back-tracking from the regulation
- Molybdenum and antimony, which occur as easily-leached oxo-anions, were not included in the first project. Guideline values were computed also for them
- The regional risk has been studied. If all MSWI bottom ash produced in a catchment area is used, the previously derived values must be revised
- Dispersal of dust from roads has been investigated in order to avoid starting from worst case assumptions

These calculations are always conservative, starting from simplified computational models in view of obtaining safety margins in assessments. To assess how large these margins are, real-life cases must be used to calibrate models. A trench was dug in the Vändöra road and the materials were studied. Below the wearing course, the combustion residues are almost unaffected after more than ten years. The most affected materials are those at the edges of the road: salts have been leached out by water and oxygen has penetrated. In a follow-up study, the chemical processes have been simulated. Some results are available but interpretation is difficult for e.g. anions.

Other roads (Törringe, Dåva) have also been monitored as well as the large surface at the Svågertorp shopping centre in Malmö.

### **Ash in cement and concrete**

Combustion residues have for a long time been used in cement and concrete. There are standards though only for silica-rich coal ashes, with a small admixture of ash from other fuels, which does not facilitate the introduction of bio-ash in these types of use.

However, fly ash from solid biofuels may be used as fines in simpler types of concrete when natural aggregates are being replaced with other materials, such as crushed rock. Some fly ashes have been tested in laboratory investigations. There is some loss of consistency, but this can be avoided by reducing the proportion of fly ash.

Ash may also be used in a mining environment: cavities are back-filled with tailings stabilised with cement. One can replace part of the cement with ash. Successful feasibility tests were carried out at the Zinkgruvan mine.

### **Ash to forest soils**

A prerequisite for a sustainable use of woody solid biofuels is that forestry is sustainable. This implies a.o. that the mineral nutrients that were removed during an intensive harvest (the stem wood of conventional forestry and the logging residues for fuel) should be returned to the forest soils, e.g. as ash. Even more important than sustaining a good biomass yield is buffering the soil: forestry is in itself acidifying and it is even more acidifying if all biomass is removed.

This recycling of bio-ash is regulated and details are found in the recommendations from the National Forest Agency regarding harvesting logging residues and recycling ash. However, recycling is not carried out to the extent which is desirable. Only parts of the surfaces where logging residues have been collected are receiving any compensation with ash. This is largely a matter of communication.

The National Forest Agency has intensified information campaigns, a.o. within the EU-Life project RecAsh, in order to try to reach all stakeholders. The Ash Programme has chosen to supplement this project as well as all the research that has been carried out, and still is carried out, for the National Energy Agency by focusing on growth: the resistance to ash recycling is to a large extent caused by the knowledge that spreading ash will not lead to tangible immediate effects. If it can be shown that ash may lead to increased growth on some soils, yielding a comparatively rapid economic return, this could contribute to an increase in the quantities recycled.

One should distinguish between mineral soils, for which the recommendations have mainly been issued, and peat lands. The conditions are different, which should be reflected in discussions. Investigations of the effects of spreading ash on mineral soil have not focused on growth but on possible negative effects on the environment. On the other hand, it is known that spreading ash on peat lands usually leads to an increased production of biomass, but at the same time one does not know its effect on environment as well, e.g. production of greenhouse gases or effects on water.

There is a controversy regarding the possible effect of applying ash on the growth of biomass on mineral soil. Results from older investigations and older experimental fields have been revisited. Whether growth can be seen to increase or decrease depends on the fertility of the soil. If fertility is low, growth decreases; if fertility is high, growth increases, but reservations should be made: the results are not always statistically significant because results are so variable. According to one school of thought, the increased growth is caused by ash releasing nitrogen which then becomes available to vegetation. According to the other school, increased growth is caused by a better nutrition status, primarily with respect to phosphorus.

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Are there enough peat lands for ash fertilisation to be interesting? In a prestudy, the surfaces where ash could lead to growth cashable in the near future (ten to fifteen years) were estimated to be ca 190 000 ha, out of the 1 million ha drained and afforested peat lands in Sweden.

In older experiments as well as newly established experiments, the effect of a dose of ash on the water chemistry, on the production of greenhouse gases – principally methane and nitrous oxide, on biodiversity, and on the microbial mass in drained and afforested peat land has been investigated. The result is that ash does not have any significant negative effects. The production of methane is unaffected. Production of carbon dioxide and nitrous oxide seems to decrease unexpectedly after applying ash. An effect on growth could be observed in the older experiments, but not in the more recent ones, which may be due to the time lapse of a few years between treatment and response.

Ash has then a fertilising effect on peat lands. A mire that was drained and afforested in 1983 was simultaneously fertilised with ash and with phosphorus. The effect was probably not very long-lived, primarily because of the circumstances: the iron and sulphide content of the peat is high and the dose of 26 t/ha of ash was not enough to keep the pH value high during the following years. Most of the phosphorus has been adsorbed on iron and aluminium oxides and thus been immobilised.

### **Knowing ash materials better**

Combustion residues are complex materials. In the course of work with the utilisation of combustion residues, it became apparent that there were gaps in the knowledge of their properties. When that was the case, projects were started in order to fill the gaps: unburned matter, hydrogen evolution, antimony, bio-availability of arsenic, zinc.

Combustion residues are principally inorganic and contain very little organic content. This has been confirmed in a few projects, although knowledge of organic constituents is not exhaustive. In a first project, the different methods of analysis for unburned matter were studied. Loss on ignition or LOI is the most common one but yields an answer that also includes a.o. bound water. The TOC<sup>2</sup> method yields a more satisfactory measure of the combustible unburned matter, i.e. mostly carbonised fuel.

Attention to organic content has hitherto been focused on trace compounds such as the dioxins. In a follow-up to the above-mentioned project, the content of organic trace substances was investigated through a commercially available semi-quantitative screening of semi-volatile substances. Very few compounds were found to be present with concentrations exceeding 0.1 mg/kg. Without any doubt, more results should be obtained by using more advanced methods and instruments, but special measures should then also be taken to minimise contamination of samples. Many of the substances that were identified and quantified do actually not belong to ash, but are most probably the result of contamination.

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<sup>2</sup> TOC is usually taken to be Total Organic Carbon, but as carbon in combustion residues mostly consist of elemental carbon, TOC should be understood as Total Oxidisable Carbon

After an explosion in an underground storage facility, the potential for hydrogen evolution from combustion residues was investigated. It is largest for residues from combustion of wastes, negligible for clean solid biofuels. The cause for hydrogen evolution is mainly the metallic aluminium in the fuel that is not oxidised during its passage through the furnace.

Antimony is another problem that is limited to waste fuels. As antimony and its compounds are volatile, it occurs mainly in fly ash, but antimony leaches mostly from bottom ashes. However, leaching tests indicate that most of it is not available, bound to the solid phases by adsorption or through solid solutions.

As arsenic is the critical component in the proposal for environmental guidelines for combustion residues, it is important to know the extent of the safety margins. The oral bio-availability of arsenic and a few other elements was determined in one investigation. Unfortunately, arsenic is 80 – 100 % available, i.e. arsenic is dissolved in a liquid simulating intestinal fluids after treatment with saliva and gastric juices. However, the test does not yield any answer as to the extent to which arsenic and the other investigated elements permeate the intestine walls, liver and other cell walls to accumulate in the cells, or to the extent to which it is excreted with faeces.

A fresh ash in contact with water is converted rather rapidly to a less aggressive material. Experiments with ash placed in water, in 1 m<sup>3</sup> large containers, confirm this fact, although the access to carbon dioxide needed for carbonation and stabilisation is limited. Depending on the exact composition of the combustion residue, a large part of the stabilisation is affected through reactions between acid and basic oxides.

When waste is classified as hazardous or non-hazardous according to the Waste Directive, zinc is the critical element. In the methodology developed within the Ash Programme, a plausible but conservative choice of chemical bonding was made for the elements in the assessment of ecotoxicity, property H14. The compound that was chosen for zinc was its oxide. International work with environmental risks has since then resulted in zinc oxide being classified as ecotoxic, which upends the results from the methodology developed within the Ash Programme. This problem can be solved only by determining the actual compounds in which zinc occurs. The results of an investigation of several ash samples with X-ray absorption spectroscopy, EXAFS, indicate that zinc actually occurs as a silicate or ferrite, which are far less soluble compounds than the oxide. It is probable that zinc is bound also by adsorption on solid surfaces or as solid solution in e.g. the silicates.

Even if ecotoxicity may be computed using the templates in the laws on chemicals, an actual determination in tests is the most accurate. A battery of tests with organisms living in slightly saline waters was investigated using several ash samples in one project: all tests should be included in a determination of ecotoxicity as none of the tests was the most sensitive in all cases. Ecotoxicity could not be predicted in calculations using literature data. Most important was that tests showed false positive responses: the ecotoxicity could often be ascribed to substances such as potassium, aluminium or

nitrate/nitrite. MSWI bottom ash that has been aged properly was shown not to be ecotoxic in those tests.

### **Improving the properties of combustion residues**

The prerequisite for improving the properties of ash or a combustion residue is that it is a *sine qua non* requirement for an utilisation with well-defined criteria. In the absence of such criteria, investing into improvements or purification is seldom relevant, and large-scale endeavours have not been initiated.

There was one initial need: stabilisation of ash that is spread onto forest soils with respect to leaching, primarily of alkali. The technical possibilities have been investigated in two projects. An addition of silica-rich ash may stabilise calcium-rich ash through the reactions between basic and acid oxides to the same properties as silica-rich ash. However, the effect on leaching of alkali is quite limited. In order to be effective, coatings with *e.g.* fine silica dust should be quite thick, which increases the costs significantly. The same conclusion was reached in a project where combustion residues were coated using foam bitumen.

The no-improvement policy adopted at the inception of the Ash Programme does not prevent the Programme from surveying available techniques. Carbonation of ash, purification by heating (*e.g.* vitrification) and purification by washing with water or acids have been examined in a few projects.

### **Information, dissemination of results**

The primary sources of information on work carried out within the Ash Programme are the technical reports. Approximately a hundred reports have been published and are available on the Internet site of Värmeforsk.

In order to better reach out with the results, the Ash Programme has taken additional steps to disseminate information:

- Newsletters that have been distributed to many stakeholders, including permitting authorities
- Seminars at the end of a period and workshops on selected questions
- Handbooks on relevant utilisations: fly ash in roads, fly ash stabilised sewage sludge as cover for landfills, MSWI bottom ash in a composite material and alternative construction materials in landfills
- A database on the properties of combustion residues, Allaska, is available on the web site of the Ash Programme
- Contributions to developing a university level textbook or compendium on utilisation of secondary materials

As the Ash Programme is an applied R&D programme, publishing in international scientific journals has not been a priority. However, a few studies with scientific value have been published by the performers of some projects.

A particular outlook on the international stage is the gathering of information performed in two studies. In the first study, the conditions for a successful utilisation have been investigated in six European countries, *i.e.* essentially the cooperation between different stakeholders independently of the national legislation on residues. In the second project, the influence of legislation was studied in a few countries.

In order to demonstrate the advantages that reusing waste materials may offer, system analyses or life cycle analyses have been carried out. The goal was to provide a collected picture weighing different targets.

Keywords: combustion residue, ash, utilisation, applied R&D, environmental impact

## Sammanfattning

Programmet "Miljöriktig användning av askor" vid Värmeforsk inleddes 2002 i syfte att ta fram den kunskapsbas som behövdes för att få till stånd ett nyttiggörande av förbränningsrester i Sverige. Programmet, som även kallas för Askprogrammet, har förlängts två gånger, först 2006-2008 och nu 2009-2011.

Föreliggande rapport utgör en syntes av arbetet och resultaten i den andra perioden, 2006-2008. För att nå ut till Europa, där förutsättningarna för nyttiggörandet av avfall bestäms i allt högre grad, har syntesen författats på engelska. Den har utsträckts till att omfatta även den första perioden, vilken redan beskrivits på svenska, för att framställningen skall vara så fullständig som möjligt.

Med askor menas i den stora världen oftast kolaskor i första hand. I andra hand avses askor från förbränningen av hushållsavfall. Utmärkande för Sverige är:

- Dels att kol knappast förekommer som bränsle, delvis till följd av omställningen av det svenska energisystemet till ett uthålligt biobränslebaserat system
- Dels att det finns ett flertal typer av askor, vilket beror på att bränslefraktionerna är många, att förbränningstekniken är varierande (bl a är fluidbäddpannor vanligare än utomlands) samt att anläggningarna ofta är små efter internationella mått och därmed askmängderna små vid varje ort

I Sverige producerades 2006 knappt 1,3 miljoner ton förbränningsrester per år, varav ca 79 % kommer till användning. Den stora volymen finns i geotekniska användningar, huvudsakligen som konstruktionsmaterial på deponi.

En viktig svensk användning är kompensation för bortförel av biomassa från skogen. Uthålligheten i användningen av träbaserade biobränslen kräver att skogsbruket är uthålligt. Det innebär bl a att de minerogena näringsämnen som bortfördes vid skörd bör återföras till skogsmarken, t ex som aska. Än viktigare än näringsämnen för fortsatt god tillväxt är markens basmättnadsgrad: skogsbruk är försurande och än mer om all biomassa bortförel.

Arbeten inom programmet har utförts i några huvudlinjer samt kompletterats med undersökningar av kemiska frågor och informationsinsatser:

- Användningar vid sluttäckning av deponier och upplag av gruvavfall
- Anläggningsbyggande, t ex vägar, både geotekniska frågor och miljöriktlinjer
- Cementbundna användningar
- Näringskompensation till mark för uttaget av biomassa

Tyngdpunkten i programmet ligger på miljöfrågor, även om tekniska spörsmål avhandlats. Tidsperspektivet är mycket längre än de 3-5 år som är normal i tillämpade program, nämligen flera tiotal år efter att askan lagts ut, t ex i en väg, eller spridits till skogsmark. I projekten har därför såväl nya provobjekt anlagts som erfarenheten samlats in från äldre objekt.

## Askor som barriär mot vatteninträngning

Den barriärfunktion som skall fyllas är att förhindra att vatten sipprar genom en deponikropp eller ett upplag av gruvavfall och föra med sig ut miljöskadliga ämnen. Förutom att de har goda funktionsegenskaper kan askor ersätta de jungfruliga naturmaterialen som normalt används till detta. Det är i första hand flygaskor med deras bindande egenskaper som kommer i fråga. Två typer av tätande skikt är aktuella:

- FSA, d v s flygaskstabiliserat avloppsslam; flygaska blandas in i rötat avloppsslam för att höja pH och förhindra nebdrytning av det biologiska materialet i slammet
- Aska som tätskikt, där de bindande egenskaperna hos den reaktiva askan utnyttjas

Askprogrammet har byggt vidare på tidigare förarbeten och lagt ut ytor i pilotskaleförsök på fyra deponier under perioden 2002-2005. Under den andra programperioden har försöken följts upp. Skikten har låg permeabilitet men fortsatt uppföljning behövs. En manual för genomförandet av FSA-tätskikt har sammanställts.

Tätskikt som består enbart av askor används bl a av Tvetå Återvinning sedan 2000. Anläggandet av tätskikt och uppföljningen har dokumenterats inom Askprogrammet. Tätskiktet uppfylla de funktionskrav som ställs på en sluttäckning genom diffusionstyrda processer: kornstorleksfördelning, fukthalt och askans reaktivitet bidrar till minimering av porvolymen och därmed till att vatteninträngningen minskar.

## Askor i gruvsammanhang

Behovet att täta en avfallskropp finns även för upplag av gruvavfall, t ex varphögar eller sandmagasin. I detta fall är det både syre och vatten som måste förhindras att tränga in: syre reagerar med sulfidmalmen till svavelsyra som lakar ut tungmetaller, motsvarande det s k AMD eller Acid Mine Drainage.

Gillervattnet är ett magasin på 80 ha för anrikningssand från sulfidmalmer. På 3 ha har ett försöksfält anlagts. Här också tätas magasinet med ett lager avloppsslam ovanpå ett lager flygaska eller av slam och flygaska i syfte att hindra luftsytet från att tränga ner till den svavelrika sanden. Askans roll är att höja pH och stabilisera skiktet mot inträngning av rötter från vegetationen som etableras i slamskiktet. Färsk aska är toxiskt för växtrötterna och ett härdat lager av aska är för hårt för att rötterna skall kunna tränga ner. Blandningar av slam och aska är för mjuka för att hindra rötterna. Växter med rötter som producerar sockerarter skall undvikas då det finns indikationer på att de fräter på askskiktet.

Varphögar har däremot ett kulturhistoriskt värde och får inte täckas. I de försök som gjordes med injicering av basisk flygaska kunde utlakningen minskas till ca 1 % av vad lakas ut från en obehandlad hög. Aska visades också kunna användas som reaktivt filter som neutraliserar lakvattnet.

## Askor i anläggningsbyggen – tekniska aspekter

Det handikapp som alternativa, dvs avfallsbaserade anläggningsmaterial har gentemot naturmaterial i beslutsprocesser är att de har andra geotekniska egenskaper än de konventionella materialens. Emellertid är konstruktioner med alternativa material och med konventionella material funktionsmässigt likvärdiga och ibland är de förra bättre.

Ser man till när askor produceras och när vägar bygg, tillsammans med svårigheterna att leverera stora mängder askor på kort tid till byggen är större vägar troligen inte den mest intressanta användningen för askor. Emellertid finns där en utvecklad kravprofil som underlättar diskussionen av materialegenskaper och -specifikationer samt funktionskraven för en konstruktion.

I inledningen av Askprogrammet bestämdes därför vilka egenskaper som behöver vara kända för konstruktionens funktion och askor karakteriserades. Framför allt två typer av askor är intressanta: flygaskor, gärna från biobränslen i bl a grusvägar och bottenaskor i asfalterade vägar. En grundläggande karakterisering av ett antal askor har utförts. Dessa tester är ganska omfattande och inkluderar också laktester för bedömningen av miljöpåverkan. Det är dock inte alldeles lätt att utvärdera egenskaperna då det till stora delar saknas erfarenhet av askornas beteende i fält och åldringens inflytande.

Nyckeln till framtiden är alltså anläggningen och utvärderingen av pilotskale- och fullskaleförsök. Inom Askprogrammet har såväl nya vägar byggts som äldre vägar utvärderats.

Användningen av flygaskor i grusvägar som utvecklades i Finland har tagits över till Sverige<sup>3</sup>. I successiva projekt har flygaskor karakteriserats, materialrecept har provats fram och några provvägar har byggts. I korthet är resultaten att vägarnas bärighet och tjältålighet ökats och att den något ökad utlakningen av salter från en av de anlagda vägarna, vilken beror på närvarons av flygaska, är övergående. För att flygaskornas goda egenskaper skall kunna utnyttjas optimalt och för att askorna skall räcka till längre sträckor bör de användas för att stabilisera dåliga eller utjänta vägmaterial.

Slaggrus, dvs mognade bottenaskor från avfallsförbränningen, har använts i tre provbyggen före Askprogrammets tid: Törringevägen nära Malmö, Vändöra inom Tekniska Verkens område i Linköping och inom Dåva kraftvärmeverks område i Umeå. Dessa objekt har följts upp inom Askprogrammet i syfte att hämta in den erfarenhet som behövs för konstruktionen av vägar med aska, bl a har styvheten bestämts med fallande vikt deflektometri. Slaggrus har ca 70 % av bärigheten hos ett bergkross, men även 70 % av bergkrossens skrymdensitet vilket gör den lättare att packa.

Ett mycket vanligt naturmaterial i vägbyggen är bergkross. Materialet behöver krossas i flera steg för att kornstorleksfördelningen skall passa kraven. Slaggrus är emellertid en finare fraktion som kompletterar bergkrossfraktioner. Ett kompositmaterial som de som

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<sup>3</sup> Det bör dock nämnas att flygaska från Vattenfall Värme pannor i Uppsala användes i grusvägar redan innan Askprogrammet kom till.

undersökts i ett separat projekt inom Askprogrammet ger möjligheten att spara energi i nedkrossningen av berg och att spara bergmaterial.

### **Askor i anläggningsbyggen – miljöfrågor**

Det största hindret för användning av restmaterial i anläggningsbyggen är inte teknik utan bedömningen av miljöpåverkan. Det är också ett av huvudincitamenten för att skapa ett Askprogram. Problemet i det sammanhanget är att det inte fanns något specifikt regelverk för ändamålet utan att riktlinjer och riktvärden för förorenad mark alternativt deponier användes.

Arbetet med ett förslag till riktlinjer inleddes med en genomgång av den relevanta lagstiftningen. Denna gäller oavsett om materialet är jungfruligt eller ett avfall. Status som avfall medför dock svårigheter vid tillståndsgivningen och hanteringen, vilka kunde kanske undvikas om materialet, d v s aska, kunde produktifieras. Med detta menades att materialet svarar mot specifikationer och håller en känd kvalitet på marknaden. Minst lika viktigt är att det inte längre är ett illa definierat avfall. Produktifiering anknyter till de ”End-of-Waste” diskussioner som nu förs på EU-nivå.

Den s k inneboende farligheten hos ett material är endast en del av informationen som behövs för att bedöma de risker som dess användning innebär. Arbetet inom Askprogrammet fortsatte därför med en analys av riskerna för människohälsan och för miljön. Två fall undersöktes av Bendz m fl med den metodik som tagits fram för bedömning av riskerna från förorenad mark:

- En skogsbilväg, d v s en grusväg, med ett relativt grunt asklager
- En asfalterad väg med slaggrus i förstärkningslagret, d v s relativt djupt beläget

Alla spridningsvägar från vägkroppen och exponeringsvägar för människan eller för miljön har beskrivits. Modelleringen är konservativ, d v s analysen utgår från ett rimligt värsta fall (worst case) scenario och ger tämligen stora säkerhetsmarginaler. T ex antas en människa bo hela sitt liv 20 m från vägen, att 30 % av de grönsaker som denna förtär odlats i närheten av vägen, dricka vatten från egen brunn, gå på vägen och andas stoft från vägen. När vägen inte brukas är den ett friluftsområde för vuxna och barn.

Ur resultaten från beräkningarna framgår att riskerna domineras av damningen från vägen, inte av lakningen som det hittills har varit vanligast att anta. Den kritiska storheten är arsenikhalten i askan som inte får överskrida 15 mg/kg i kvarlämnat askmaterial. Om hänsyn tas endast till lakning från askmaterialet blir riktvärden för innehållet av t ex tungmetaller ganska mycket högre och borde inte innebära något hinder för användningen av askor i anläggningsbyggen.

Arbetet med riktlinjer har sedan kompletterats på ett antal punkter:

- Upptaget av tungmetaller i växter som vuxit på aska har undersökts – försöksmaterialet är lysimeter vid SGI i Linköping anlagda 1993 som invaderats av träd, bl a björk, och gräs
- För kontamineringen med radioaktivt cesium finns en föreskrift från SSI, numera Strålsäkerhetsmyndigheten, som begränsar påverkan på brunnsvatten och på en

recipient. I ett projekt har ett riktvärde på ca 2 kBq/kg räknats ut för aska utgående från föreskriften

- Molybden och antimon, vilka bildar lättlakade oxoanjoner, ingick inte i det första riktlinjeprojektet. Riktvärden har beräknats också för dessa
- Risken på en regional nivå, inom ett avrinningsområde, har undersökts. Om all slaggrus används till vägar måste de tidigare beräknade riktvärdena sänkas.
- Spridningen av damm från vägarna har undersökts i syfte att bättre beskriva den, i stället för att utgå från värsta fall antaganden

Dessa beräkningar är alltid konservativa och utgår från förenklade beräkningsmodeller för att ge säkerhetsmarginaler vid bedömningen. För att kunna bedöma hur stora marginalerna är måste verkliga fall utnyttjas för att justera modellernas förutsägelser. Ett schakt grävdes i Vändöravägen och materialerna undersöktes. Under asfalten är aska i stort sett opåverkad efter mer än tio år. Mest påverkade är de material som ligger närmast väggkanten: salterna har lakats ut av vatten och syre har trängt in. I en uppföljning har de kemiska förloppen simulerats med matematiska modeller. Vissa resultat föreligger men tolkningen för t ex anjoner är osäker.

Även andra vägar (Törringe, Dåva) har följts upp, liksom den stora planen vid Svågertorp stormarknad i Malmö.

### **Aska i cementbundna applikationer**

Askor används sedan länge i cement och betong. Standarder finns dock endast för kiselrika kolaskor med viss inblandning av andra askor, vilket försvårar introduktionen av t ex bioaskor i dessa användningsområden.

Flygaskor från biobränslen är dock användbara som finmaterial i enklare typer av betong, t ex markbetong, när naturgrus ersätts med andra material som krossat material. Laboratorieundersökningar har utförts för ett antal olika flygaskor. Det sker en viss förlust av konsistensen, men den går att undvika vid lägre inblandningsgrader.

Askor kan också användas i ett gruvsammanhang: utbrutna hålrum fylls igen med anrikningssanden stabiliserad med cement. Aska kan ersätta en del av cementen. Genomförbarhetsförsök utfördes vid Zinkgruvan med goda resultat.

### **Aska till skogsmark**

Uthålligheten i användningen av träbaserade biobränslen kräver att skogsbruket är uthålligt. Det innebär bl a att de minerogena näringsämnen som bortfördes vid den intensiva skörd som helträdsuttag innebär (stamveden i det konventionella skogsbruket plus avverkningsresterna som bränsle) bör återföras till skogsmarken, t ex som aska. Än viktigare än näringsämnen för fortsatt god tillväxt är markens basmättnadsgrad: skogsbruk är försurande i sig och än mer försurande om all biomassa bortförs.

Formerna för denna återföring av bioaska har reglerats, närmast av Skogsstyrelsens rekommendationer vid uttag av avverkningsrester och askåterföring. Denna verksamhet sker dock inte i den utsträckning som är önskvärt. Endast en del av de arealer från vilka

skogsbränslen hämtas kompenseras med aska. Till stor del är detta en fråga om kommunikation.

Skogsstyrelsen har bl a genom EU-projektet RecAsh intensifierat spridningen av information för att försöka nå ut till alla parter. Askprogrammet har valt att komplettera detta projekt samt den forskning som utförts och fortsätter utföras på uppdrag av Energimyndigheten genom att fokusera på tillväxtsfrågan: till stor del beror motståndet mot askåterföring troligen på att effekten av att sprida aska inte är påtaglig. Kan det visas att aska ger en ökad tillväxt på vissa marker och därmed en jämförelsevis snabb ekonomisk nytta kan detta bidra till att öka de mängder som sprids.

Man måste skilja mellan fastmark för vilken rekommendationerna utfärdats av Skogsstyrelsen och torvmark. Förutsättningarna är olika, vilket bör återspeglas i diskussionen. De undersökningar av effekterna av en spridning av aska på fastmark som genomförts har inte betonat tillväxteffekter utan risken för negativa miljöeffekter. Å andra sidan vet man att en spridning av aska på torvmark brukar leda till en ökad produktion av biomassa, men samtidigt vet man inte lika mycket om påverkan på miljö som t ex produktionen av växthusgaser eller påverkan på vattnet.

Åsikterna går isär vad gäller askspridningens möjliga inverkan på biomassans tillväxt på fastmark. Resultaten från äldre undersökningar och äldre försök har omprövats. Om en ökad eller minskad tillväxt kan ses efter spridningen beror på markens produktivitet. Är produktiviteten sämre minskar tillväxten, är den god ökar tillväxten, med reservation för variationerna i resultaten som leder till att slutsatser inte alltid är statistiskt säkerställda. Enligt ena skolan beror den ökade tillväxten bara på att askan frigör kväve som blir tillgängligt för växterna. Enligt andra skolan beror tillväxten på en bättre näringsstatus, framför allt med avseende på fosfor.

Finns det tillräckligt mycket torvmark för att askgödsling skall vara intressant? I en förstudie uppskattades den areal där aska kan leda till en tillväxt som kan realiseras på kort sikt (tio till femton år) till ca 190 000 ha ur den miljon hektar dikade beskogade torvmarker som finns i Sverige.

I såväl äldre försök som nyanlagda försök har undersökts inverkan av spridningen av aska på vattenkemi, på produktionen av växthusgaser – framför allt metan och lustgas -, på den biologiska mångfalden samt på den mikrobiella massan i dikad och beskogad torvmark. Resultaten är att askan inte har några betydande negativa effekter. Produktionen av metan är opåverkad. Oväntat är att produktionen av både lustgas och koldioxid förefaller minska efter en tillförsel av aska. Effekt på skogsväxten kunde observeras i de äldre försöken, men inte i de nyare, vilket kan bero på att det tar några år innan någon sådan effekt kan iakttas.

Aska har således en gödslande effekt på torvmark. En dikad och beskogad torvmosse gödslades 1983 med aska och med fosfor. Effekten varade troligen inte så länge, mest beroende på omständigheterna: torvens sulfid- och järninnehåll är stort varför askgivan

på 26 t/ha inte räckte för att hålla pH uppe under åren. Det mesta av fosfor fastnade på järnoxider och aluminiumoxider, varför den är mindre tillgänglig för växtligheten.

## Bättre kunskap om askmaterialen

Askor är komplexa material. Under arbetet med användningar blir man varse luckor i kunskapen om deras egenskaper. Då behoven uppstått har kunskapen utvecklats i flera projekt: oförbränt, vätgasutveckling, antimon, arseniks biotillgänglighet, zink m fl.

Askor är huvudsakligen oorganiska och innehåller mycket lite organiskt. Detta bekräftades i några projekt, men kunskapen om det organiska innehållet är inte uttömmande: i ett första projekt utreddes betydelsen av det mått på oförbränt som lika analysmetoder ger. Glödförlusten är den vanligaste metoden, men utöver brännbart består svaret också av annat som bundet vatten. Metoden TOC<sup>4</sup>, ger ett mer rättvist mått på det brännbara oförbränt, som består mest av kol, d v s förkolnat bränsle.

Uppmärksamheten kring det organiska innehållet har hittills fokuserats mest på organiska spårämnen som de s k dioxinerna. I ett uppföljningsprogram bestämdes innehållet av organiska spårämnen i några askprov med en förutsättningslös semi-kvantitativ screening av semi-flyktiga ämnen. Mycket lite med halter över 0,1 mg/kg kunde hittas. Utan tvivel kan mer identifieras och kvantifieras med bättre analysmetoder, men då måste även särskilda steg vidtas för att minimera kontaminering av proven. Många bland de ämnen som hittades i screeningen hör normalt inte till askan, utan tillkom under hanteringen.

Efter en explosion i ett berglager undersöktes askors potential att producera vätgas. Denna är störst för avfallsaskor, obetydlig för rena biobränslen. Orsaken är huvudsakligen det metalliska aluminium som finns i bränslet och som inte hinner oxideras under färden genom pannan.

Antimon är också ett problem som är begränsat till avfallsaskor. Då antimon och dess föreningar är lättflyktiga förekommer den framför allt i flygaskor, men lakning sker mest från bottenaskor. Emellertid indikerar lakegenskaperna att det mesta av antimon är otillgängligt, bundet genom adsorption eller som fast lösning i resten av askan.

Då arsenik är den kritiska komponenten i miljöriktlinjerna för askor är det viktigt att veta vilka marginaler som finns. Den orala biotillgängligheten hos arsenik och några andra grundämnen bestämdes i ett projekt. Tyvärr är arsenik 80 - 100 % biotillgänglig, d v s arsenik löses upp i en vätska som motsvarar tarmvätska efter behandling med mun- och magvätskor. Testet säger dock inget om i vilken grad arsenik och de andra undersökta ämnena passerar tarmväggar, lever och andra cellväggar för att ackumuleras i cellerna, ej heller hur mycket som förs ut ur kroppen med fekalier.

En färsk aska omvandlas tämligen snabbt i kontakt med vatten till ett mindre aggressivt material. Försök med aska som placeras under vatten i 1 m<sup>3</sup> stora behållare bekräftar

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<sup>4</sup> TOC brukar användas som förkortning för Totalt Organiskt Kol, men då kolet i askor mest är elementärt kol bör TOC uppfattas som Totalt Oxiderbart Kol.

detta, även med en begränsad tillgång till den koldioxid som annars anses vara nödvändig för karbonatiseringen och stabiliseringen. Beroende på askans exakta sammansättning sker en inte oväsentlig del av stabiliseringen genom reaktioner mellan de de basiska och sura oxiderna.

I klassningen av avfall som farligt eller icke-farligt enligt Avfallsförordningen är det zink som är kritiska ämnet. I den klassningsmetodik som utvecklades i Askprogrammet gjordes ett trovärdigt men konservativt val av förekomstform för de aktuella grundämnena i bedömningen av ekotoxiciteten, egenskap H14. Den förening som valdes för zink var zinkoxid. Det internationella arbetet med miljörisker ledde sedermera till att zinkoxid klassades som ekotoxiskt, vilket kullkastade resultaten från Askprogrammets klassningsmetodik. Det problemet kan lösas endast genom en bättre karakterisering av zinks bindningsformer i askan. Resultaten från en undersökning av flera askor med EXAFS, Röntgenabsorptionsspektroskopi, kunde tolkas som att zink egentligen förekom som zinksilikat eller möjligen zinkferrit, vilka är betydligt mindre lösliga än oxiden. Troligen är zink dessutom bundet antingen genom sorption till partikelytor eller i fast lösning i t ex silikaterna.

Även om ekotoxicitet, fara H14 i Avfallsförordningen, kan beräknas enligt mallar i kemikalielagstiftningen är en bestämning i tester a priori den mest rättvisande. Ett batteri av tester med brackvattenorganismer undersöktes för flera askor i ett projekt: alla tester bör ingå i en bestämning då ingen av testerna alltid var känsligast. Ekotoxiciteten i testerna kunde heller inte förutsägas genom beräkningar från litteraturdata. Testerna uppvisade framför allt falska positiva resultat då ekotoxiciteten ofta kunde härledas till ämnen som kalcium, kalium eller ammonium/nitrit. Väl lagrat slaggrus visade sig inte vara ekotoxiskt i dessa tester.

### **Förbättringar av askans egenskaper**

Förutsättningen för att en askas egenskaper skall behöva förbättras är att det skulle vara ett absolut krav för en användning med väl definierade kriterier. I avsaknaden av sådana upplevs satsningar på förbättringar föga relevanta, och större insatser har inte genomförts.

Det fanns inledningsvis ett behov: att stabilisera aska som sprids till skogsmark med avseende på utlakningen, framför allt av alkali. Möjligheten att åstadkomma detta har undersökts i två projekt. Kalciumrika askor kan stabiliseras med en tillsats av kiselrika askor genom reaktioner mellan sura och basiska oxider till samma egenskaper som kiselrika askor. Däremot är effekten på utlakningen av alkali begränsad. För att vara effektiv bör drageringen med t ex finfördelad silika vara betydligt tjockare, vilket gör metoden alltför kostsam. Samma slutsats nåddes i ett annat projekt där aska belagts med skumbitumen.

Policyn att inte förbättra askor hindrar dock inte orientering om de möjligheter som står till buds. Karbonatisering av aska, rening genom upphettning (bl a förglasning), rening genom tvättning med vatten eller syror har granskats i olika projekt.

## Information, resultatspridning

Den primära källan till informationen om de arbeten som utförts inom Askprogrammet är forskningsrapporten. Ett hundratal projektrapporter finns tillgängliga på Värmeforsks webbplats.

För att bättre nå ut med resultaten har Askprogrammet gjort ytterligare informationsåtgärder:

- Nyhetsbrev till många organisationer, inklusive myndigheter
- Seminarier vid avslutningen av en programperiod och workshops om aktuella frågeställningar
- Handböcker om aktuella användningar: flygaskor i väg, flygaskstabiliserat avloppsslam som täckmaterial, slaggrus i ett sammansatt obundet material samt alternativa konstruktioner på deponier
- En databas ”Allaska” med kvantitativ information om askors egenskaper, tillgänglig på Askprogrammets webbplats
- Bidrag till utvecklingen av material för en universitetskurs om användningen av restmaterial

Eftersom Askprogrammet är ett tillämpat forskningsprogram har inte publicering i internationella vetenskapliga tidskrifter prioriterats. Ett antal arbeten med ett mer vetenskapligt värde har dock publicerats av projektens utförare.

En särskilt internationell utblick utgörs av den inhämtning av information som utfördes i två projekt. I det första projektet utreddes villkoren för en framgångsrik användning i sex europeiska länder, huvudsakligen samspelet mellan olika aktörer oberoende av den nationella lagstiftningen. I det andra projektet undersöktes lagstiftningens inverkan i några länder.

För att reda ut de fördelar som användning av restmaterial som aska kan ge har systemanalyser eller livscykelanalyser utförts genom några projekt. Målet var att ge en samlad bild där flera mål kan vägas samman.

Nyckelord: aska, förbränningsrester, nyttiggörande, tillämpad forskning, miljöpåverkan



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# 1 Introduction

## 1.1 Background

The Swedish cooperative RD&D Programme “Environmentally correct use of combustion residues”, or Ash Programme in short, was started in 2002 in order to provide a knowledge basis for uses of combustion residues. It is co-financed by utilities, industry and the state through the Swedish Energy Agency. The Programme was extended into a second period 2006-2008, and at the end of 2008, into a third period ending 2011.

The Ash Programme is fundamentally a technical RD&D program dominated by environmental questions. The trigger was the need for a common national understanding of environmental impacts, especially/particularly in the context of permitting. A requirement for this understanding is *a.o.* an improved knowledge of material properties. As work and discussions with stakeholders have progressed, the Programme has also overflowed into less technical aspects, such as acceptance and perception of waste and benefit.

The keyword or key concept for the Ash Programme is “sustainable management of resources”. Perhaps this wasn’t what the Programme was perceived to represent in the beginning, but this is what it evolved into as the less technical aspects developed.

Combustion residues are usually thought of as waste, not resources, and there surely is nothing sustainable about waste? Waste is something to be managed, of course, but it is not a resource or one wouldn’t have to discard it. A resource benefits one, but a waste is a cost or an obligation. This dichotomy between waste and resource has to be resolved if combustion residues are to find a place in a sustainable civilisation.

The recipe for sustainability is somewhat similar to the waste strategy, *i.e.* refrain from using, reuse and recycle materials. Additional rungs on the waste strategy ladder are extract energy or, as a last resort, discard. Actually, natural resources may be used but only if utilisation does not exceed the natural replenishment of these resources. Discarding is not supposed to be an option unless priority is given to other considerations.

Combustion residues or ash are the mineral residues from combustion (extraction of energy) and from a narrow point of view there isn’t much in this recipe that fits them as materials. Sustainable management should rather be understood in a wider sense, as finding the place for ash in several uses of resources, one of them being the sustainable use of solid biofuels. Ash also has a place in refraining from using, as e.g. when they are used in civil engineering works instead of natural materials that would have been quarried or when they are diverted from ultimate disposal in landfills.

Using combustion residues is practised in several countries and Sweden is not unique in this area. However, context, acceptance both by the broader public and the experts,

legislation, supply of natural materials and alternative materials as well as entrepreneurship differ though from country to country. The fuels differ too: combustion residues are often taken to mean coal ash or ash from municipal waste incineration. The situation in Sweden regarding fuel (a large share of solid biofuels, a well-developed incineration of municipal waste) and limited use of combustion residues as materials in civil works may in this sense be purely Swedish.

## 1.2 Production of ash in Sweden

If all types of ash from all large combustion plants are taken into account, somewhat more than one million tons of ash is produced yearly in Sweden. The figures reported in the following tables are from a survey 2006 by Svenska Energiaskor.

Table 1-1. Ash production in Sweden during 2006, figures rounded off to the nearest 1000 t DS per year

Type of furnace	Fuel	Bottom ash	Fly ash and APC residues	Combined bottom and fly ash
Grate furnaces and others	Municipal solid waste, industrial waste	445 000	93 000	
	Solid biofuels (wood chips, logging residues)	13 000	10 000	77 000
	Solid biofuels and sludge from the pulp and paper industry	12 000	11 000	10 000
	Mixed fuels	35 000	25 000	
	Wood chips and peat	2 500	1 000	2 000
	Coal	3 000	2 000	
Pulverised fuel furnace	Coal and peat	7 000	24 000	
	Peat, wood, etc	18 000	31 000	
Fluidised bed furnaces (CFB, BFB)	Municipal solid waste, industrial waste	45 000	57 000	
	Solid biofuels	15 000	14 000	5 000
	Mixed fuels	31 000	88 000	14 000
	Peat and wood chips	11 000	27 000	
	Pulp and paper industry	21 000	78 000	10 000
	Coal (PFBC)	5 000	44 000	
Subtotals		660 000	500 000	120 000
<b>Grand total</b>	<b>1 280 000</b>			

Table 1-2. Utilisation of ash in Sweden during 2006, quantities rounded off to the nearest 1000 t DS

Type of utilisation	Quantities	Fuel / process
Recycling to forest soils	28 000	Solid biofuels / CFB fly ash and combined ash from grate furnace
Agricultural uses	2 000	Solid biofuels /bottom ash from grate furnace
Soil amendment	4 000	Solid biofuels
Cement clinker	2 000	Solid biofuels / fly ash from grate furnace or bottom ash from FB furnace
Civil works (e.g. roads)	130 000	Peat and wood / CFB fly ash
Parking places and other surfaces	35 000	MSWI bottom ash
Scrap metal	40 000	MSWI bottom ash
Back-filling decommissioned oil storage caverns	18 000	Waste fuels / CFB fly ash
Neutralisation of waste acids in Norway	32 000	Hazardous waste / APC residues and fly ash
Stabilisation of APC residues from MSWI	49 000	Coal / PFBC
Construction materials on landfills	650 000	MSWI bottom ash and other residues
Covering mining waste deposits	6 000	Solid biofuels / CFB fly ash
Others, storage pending decisions	12 000	
<b>Grand total</b>	<b>1 009 000</b>	

In 2006, approximately 79 % of the produced combustion residues (ca 1 million tons) were utilised. The rest is land filled. The largest use is as construction material on landfills, 650 000 tons, as fill or as capping material. The economic incentive is waiving the tax on waste sent to landfill. Residues which would anyhow have been land filled replace virgin materials in these functions and allow conservation of natural resources. Large volumes of materials are needed today but this usage will not last for ever. The present landfills will be closed within the next 10 to 15 years.

One may note that the quantity 28 000 tons recycled to forest soil as compensation is far from the target. As logging residues are extracted from ca 30 000 ha each year, the need for ash as compensation (see next section and chapter 7) is closer to 60 to 90 000 tons yearly.

### 1.3 Recycling ash to forest soils

A Swedish particularity is the recycling of wood ashes to forest soils in order to preserve their production capacity in a long time perspective. The present debate in the EU and elsewhere on sustainable use of bioenergy focuses on liquid or gaseous biofuels for transportation (Fehrenbach et al., 2008). The Swedish concern with ashes for sustainability is until now focused rather on maintaining a sustainable production of solid woody biofuels.

Forests are a natural resource producing biomass as raw materials for many commodities such as paper, timber for building. Any removal of biomass from the forest removes resources that would have been available for next generation: carbon, nitrogen,

mineral nutrients. There is a tipping point beyond which removal will not be compensated by natural processes of photosynthesis for carbon, nitrogen fixation or weathering of minerals followed by uptake in the plants.

In particular, removing slash and logging residues intensifies the extraction of biomass resources. It has been feared that this intensification may deplete soils in the long run (or even in the short run). Another effect is a further accentuation of forestry's natural acidification of soils when cationic nutrients taken up in plants are replaced with hydrogen ions in the soil. In addition to the impact of intensive biomass harvesting, acid rain from the use of fossil fuels all over Europe causes a further acidification of forest soils in southern and western Sweden.

The buffering capacity of soils is reduced, nutrients are leached from them and surface waters become more acid. Liming lakes and rivers may be effective, but only for a short time, and it has to be repeated at time intervals of a few years. It has been hoped that liming the soils, upstream of the surface waters, would have a more lasting effect. A recent examination of soil status many years after spreading lime or ash has, however, not fully validated this opinion (Hjerpe et al., 2008).

The content of mineral nutrients and cations buffering the forest soils against acidity are 1.5 to 4 times larger in slash than in the tree stems used in pulp and paper or timber industries. However, these nutrients are found in the mineral combustion residues. Recycling the latter to forest soils is then a suitable compensation for the nutrients that were removed in the first place.

Retaining productivity of the forest soils is though a little too easily interpreted as increasing the production. True, the fertilising effect of wood ash on the growth of pine trees has been demonstrated by the Finnish Forest Research Institute already in the 1930's. Some areas of drained peat land suffer from an imbalance of nutrient supply, being deficient in potassium and phosphorous as well as nitrogen-rich. The fertilizing effect of wood ash lasts longer than that of conventional fertilisers, up to 30 to 40 years.

However, investigations within the several research Programmes funded by the state between early 1990's and 2007 have returned a more varied picture for mineral soils (Dahlberg et al., 2006). On highly productive mineral soil in southern Sweden, positive effects of ash spreading on growth have been demonstrated in several studies. The probable cause for this effect is that ash increases the availability of nitrogen to vegetation in this region. Furthermore, much of this nitrogen originates from atmospheric deposition (acid rain). Removing surplus nitrogen by whole-tree harvesting and returning ash with the other mineral nutrients may contribute to reducing this problem.

In those areas where the soil is less productive, e.g. the northern parts of Sweden, growth is often limited by the supply of nitrogen. The deposition of nitrogen from acid rain is also much lower. Spreading ash will have a limited impact on short-term growth or even a negative one as alkaline ash tends to mobilize nitrogen.

## 1.4 Large volumes of combustion residues

It is appropriate to return wood ash to forest soils, because its chemical composition reflects the solid biofuels' content of elements, which content would over time have been available to vegetation if wood hadn't been harvested and removed. However, this can't be said about other combustion residues, as their chemical composition does not reflect that of fuels from the biosphere.

These other combustion residues represent a very large volume, at least for the producers, and the only market able to swallow it is that for construction materials, *e.g.* as secondary aggregates. This market is ca 100 million tons per year. Combustion residues total ca 1 million tons per year for the whole country and they are produced in many small plants: a production rate of 10 000 tons per year is rare. They are not rapidly available in volumes sufficient for an investment in infrastructure and will be of marginal importance. Furthermore, the permitting procedure has been so time-consuming and difficult to predict that contractors prefer to use other materials. Permitting is an important non-technical barrier.

Combustion residues have been considered a replacement for natural sand or gravel, natural aggregates, as these materials are the target of conservation efforts being a finite natural resource. However, untreated they cannot replace these natural materials on a one-to-one basis because the technical properties are not equivalent. Using residues to conserve natural resources is then more subtle and involves finding uses where they present a technical or economic advantage.

A few such uses have been found. Today, they are predominantly used as construction materials in landfill closure. There is a huge need extending a few years into the future. Conservation here is refraining to use natural materials in a location that anyhow is labelled "waste" by replacing them with residues, *a.o.* combustion residues. Finding ash a use on a landfill exempts them from the landfill tax, which tax is actually aimed at steering reusable materials away from landfills.

Even when the landfills with organic or combustible waste have been closed, there will still be landfills. Resource conservation is here considered to also mean saving space and volume for other wastes by refraining to place on landfills materials that may otherwise be used, *i.e.* combustion residues. Of course, some residues may present such risks that it may not be desirable to place them anywhere else than in controlled sites, but as many as possible should find a use.

The key to a future general use of combustion residues in civil works is resolving all aspects of the permitting procedure. It is easy to focus on one aspect only to obtain a swift decision, but what is needed is a holistic approach where the residues are considered not as waste but as a material among others in a sustainable use of resources.

## **1.5 Goal and audience**

The purpose of this synthesis is to provide an overview of the progress achieved by the Ash Programme during the first two periods of activities, from 2002 to 2008. The targeted audience is international, not only domestic as usual.

## 2 Ash as a barrier against water penetration

The percolation of water through landfills commonly leads to the mobilization of soluble compounds in the deposited material and to the subsequent generation of a leachate containing these compounds. In order to avoid undesirable environmental impacts from leachate release, soil covers are installed on landfills with the purpose of greatly reducing the percolation of water into the deposited material. According to regulations currently included in the European waste directive and previously in the Swedish waste ordinance (ordinance 2001:512, §31; see also Naturvårdsverket, 2004), the final cover on a landfill should be constructed so that water percolation does not exceed 5 litres per square meter and year ( $5 \text{ l m}^{-2} \text{ y}^{-1}$ ) for covers over hazardous waste, and does not exceed  $50 \text{ l m}^{-2} \text{ y}^{-1}$  for nonhazardous waste. According to recommendations from the Swedish Environmental Protection Agency (Naturvårdsverket, 2004), a complete cover on a landfill consists of five different layers (vegetation layer, protective cover, drainage layer, sealing layer, and a levelling layer), each having its own specific function (see Figure 2-1). Low percolation rates can be achieved by installing a sealing layer, within the soil cover, with a low hydraulic conductivity.

There are a number of materials that fulfil the functional requirements of a sealing layer and are suitable for this application. Natural geological materials such as clay or clay-rich glacial till (in the Nordic countries) have been traditionally selected as sealing layer materials, but these are natural resources and not readily available in many locations and their excavation may prove to be aesthetically undesirable. Instead of using limited natural resources, there has been an increased focus on the use of alternative materials in sealing layers. Boiler and incinerator fly ash is a material that, after hardening, acquires a low hydraulic conductivity and is therefore a suitable candidate for sealing layer construction. The application of boiler and incinerator fly ash in sealing layers on landfills has therefore been the focus of a number of studies and demonstration projects within the Ash Programme.

An early study (Tham and Ifwer, 2006) within the Ash Programme investigated the current use of secondary construction material in waste management, with a focus on incineration ash. Information from eleven landfill sites was compiled and included the practical experience of using ash as the primary material in landfill cover. Tham and Ifwer (2006) describe the function of ash in the different layers and discusses the advantages or disadvantages of the techniques applied. The overall results show that incineration ash is a suitable material for use in cover constructions, either alone, or mixed with sewage sludge. Data on water percolation below existing covers has indicated that the covers can meet permeability requirements.

In consideration of the growing need to find alternative materials for landfill construction and the need for guidance in the use of these alternative materials, specifically with regard to liner and cover layer materials, Rihm et al. (2009) have written a handbook providing pertinent information for the construction of landfills. The handbook discusses the goals and legislation influencing the design of construction on a

landfill, functional and environmental requirements of construction, general conditions for construction work on a landfill site, capping construction design, material behaviour, methods for assessing the environmental behaviour of the construction, description of some plausible materials, and quality assurance systems for construction and construction materials.

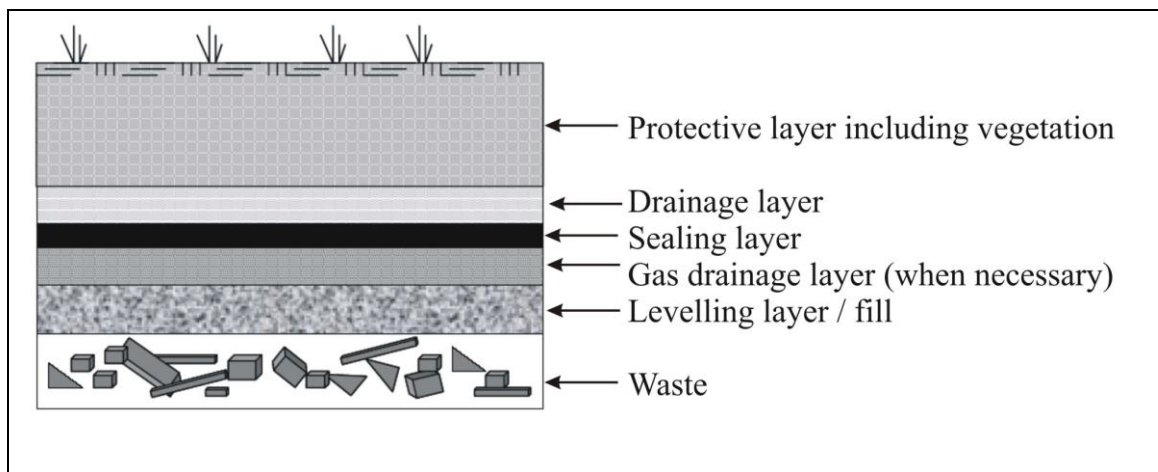


Figure 2-1. General design for construction of soil cover on a landfill (Naturvårdsverket, 2004).

## 2.1 FSA pilot-scale demonstrations

During the two programme periods, fly ash stabilized sewage sludge (FSA, from Swedish flygaskastabiliserat avloppsslam) has been investigated as a sealing layer material on a number of municipal landfills. As a result of these studies (see below), an instruction manual (Carling et al., 2007) was written with the purpose of providing advice in the manufacture and installation of landfill sealing layers, based on FSA, which fulfil functional demands. The manual contains a description of geotechnical and environmental demands that need to be met, including requirements for the composition of the FSA mixture, storage of the material prior to installation, installation of the sealing layer, and monitoring of the cover installation.

The Ash Programme has been actively involved in the installation and monitoring of two pilot-scale demonstrations of the FSA concept on municipal landfills (see below). In addition to these two projects, a full-scale FSA-based cover was installed on the Galgberget mine tailings deposit in Falun, with completion in 2002. Although this FSA cover is not associated with the Ash Programme, it demonstrated the successful design and implementation of a landfill cover composed of FSA (Hallberg et al., 2005).

### 2.1.1 Dragmossen landfill

The Dragmossen landfill in Älvkarleby, Sweden has been used for a pilot-scale demonstration of a fly ash – sewage sludge sealing layer. Initial laboratory studies (Mácsik et al., 2003) were used to establish an appropriate ash – sludge ratio, based on

the resulting hydraulic conductivity, undrained shear strength, and durability. The results of the tests indicated that appropriate mixes ranged from 40% - 60% sewage sludge, with the remaining fraction as fly ash. Material properties that particularly affected layer performance were water content, pH, and CaO content. The study demonstrated that water loss from the fly ash – sewage sludge mixture should be avoided, as increases in hydraulic conductivity may result. The ash-sludge sealing layer has about the same price as traditional liner materials (e.g. bentonite mats) if material costs are not accounted for. However, if it is considered that the ash and sludge are not deposited in a landfill and that there are not associated waste taxes and deposition costs, then fly ash and sewage sludge are a more economical alternative to traditional liner materials.

In the pilot-scale field demonstration at the Dragmossen landfill approximately 1 500 tonnes FSA have been used, covering an area of 2 400 m<sup>2</sup> at a thickness of 0.55 m (Mácsik et al., 2005). The FSA mixture contained 45 – 50 % fly ash. The demonstration project investigated 1) the manufacturing and construction of the sealing layer, 2) the permeability, stability, settling, and leachability of the sealing layer, and 3) the long-term durability of the construction. Practical issues involved in sealing layer construction were evaluated in the report, such as time limits on the storage of materials prior to construction and odorous ammonia emissions during construction.

A subsequent study (Mácsik et al., 2007) conducted at the Dragmossen landfill consisted of studying various material properties in the cover, sampling and analysis of water and gas samples, and a laboratory study of FSA's resistance to degradation under anaerobic conditions. In the field, water and gas lysimeters were used for acquiring water and gas samples at various levels below the sealing layer, the fill layer, and the protective cover. FSA samples from the sealing layer were obtained in order to determine the material's permeability, bulk density, and water content. Results from the water lysimeters demonstrated that the percolation rate through the sealing layer, two years after installation, was < 30 mm/yr. This indicates that the FSA has maintained a low hydraulic conductivity and has not exhibited a decline in performance since installation. The leaching of dissolved compounds from the sealing layer, measured as electrical conductivity, was relatively constant during the three year period that sampling was conducted; the total amount of leached DOC, total nitrogen, and total sulphur was correlated with percolation rate.

A considerable amount of practical experience was acquired as a result of the pilot study (Mácsik et al., 2007), in terms of construction, transport, and spreading of the sealing layer material. In particular, it was observed that improper mixing of the FSA resulted in material with a higher permeability than desirable.

### **2.1.2 Tekniska verken, SRV återvinning and Renova**

In a different study (Carling et al., 2006; Ländell et al., 2009), FSA was studied in field tests at three landfills: Tekniska Verken in Linköping, SRV Återvinning in Huddinge and Renova in Gothenburg. These pilot projects were preceded by feasibility studies supported by Swedish Waste Management (*Avfall Sverige*) and the R&D department of

the Swedish Water & Wastewater Association (*Svenskt Vatten Utveckling*). The initial field tests (Carling et al., 2006) were performed with the aim of studying operational methods, properties and effectiveness associated with using sludge and ash on a larger scale; later field tests (Ländell et al., 2009) studied the sealing layer function during a longer time period. The project consisted of initial laboratory investigations of different ash-sludge mixtures, construction of experimental areas, and the monitoring of the operational effectiveness of the test areas. In addition, studies investigated the durability of the material mixtures, the management of ammonia emissions from the mixtures (Carling et al., 2006), the permeability of the FSA mixture, and leaching from the sealing layer (Ländell et al., 2009).

In general, the laboratory tests (Carling et al., 2006) showed that the properties of ash, sludge and the mixtures varied considerably. This underlines the great importance of characterizing both the raw materials and the mixtures prior to using them on a larger scale in the field.

At the Tekniska Verken and SRV Återvinning landfill sites, the FSA mixture was deposited in two 20 – 25 cm layers with the first layer being packed before the second layer was deposited. Construction took place in 2004 and 2005, respectively. At the Renova landfill, one 40 cm layer was emplaced. During the construction of the sealing layers, water lysimeters, gas probes, subsidence meters, and groundwater tubes were installed. The performance of the sealing layer construction at Tekniska Verken was initially studied for a period of one year after installation (Carling et al., 2006). Hydraulic conductivity measurements and seepage into the lysimeters indicated a low percolation rate ( $12 \text{ l m}^{-2} \text{ yr}^{-1}$ ). Gas measurements indicated the presence of elevated concentrations of methane in the sealing layer and overlying layers, suggesting that a certain degree of degradation occurs in the sealing layer; the pH of the mixture, one year after installation was in the range 8.0 – 8.6, and was probably not high enough to inhibit the activity of methanogenic bacteria in the ash-sludge mixture.

For a three year period following installation of the FSA sealing layers in Gärstad (Tekniska Verken) and Sofielund (SRV Återvinning), the function of the sealing layers was studied (Ländell et al., 2009). The covers were monitored with lysimeters both above and below the sealing layer, gas probes, groundwater wells, and subsidence meters. Water flow to the lysimeters indicate a low rate of percolation through the FSA sealing layer, with measured flow rates of  $5 - 18 \text{ l m}^{-2} \text{ yr}^{-1}$  in Gärstad; the percolation rate above the sealing layer was much greater, in the range  $118 - 157 \text{ l m}^{-2} \text{ yr}^{-1}$ . The low percolation rate can be partially attributed to the presence of a drainage layer over the sealing layer. Gas measurements indicate the production of methane and carbon dioxide, most likely from the degradation of organic matter in the sealing layer, although the rate of this process is assessed as being low. Runoff from the sealing layer contained relatively high concentrations of nutrients and metals, compared to guideline values for surface water. Although the concentration of nitrogen and some metals were observed to decrease with time in the runoff, it was not possible to determine if the runoff could be eventually released to surface water recipients with treatment, or if treatment would be needed in the long term.

## 2.2 Alkaline degradation of cellulose and its impact on metal leaching

The application of a mixture of ash and cellulose – rich materials, e.g. activated sewage sludge, as sealing layers on landfills has been investigated in the Ash Programme. In theory, this combination of materials produces a layer with low hydraulic conductivity, high pH, and a high capacity for binding metals. Researchers within the Ash Programme have, however, raised a number of concerns regarding the degradation of cellulose in the sealing layer, where the major concern was that cellulose degradation will lead to the production of organic molecules that can bind metals that occur in the ash and result in increased metal leaching and mobility. This issue of increased metal mobility is also relevant in cases when ash and organic wastes are co-deposited in landfills, if ash is injected into older landfills, and the use of ash and sewage sludge as construction material in roads.

Cellulose degradation in the presence of fly ash has been reviewed in a literature study (Wikman et al., 2003c) and in laboratory and field studies (Wikman et al., 2003c, 2005). The literature study indicated that biological degradation would be inhibited at the potentially high pH ranges that develop in contact with the alkaline ash. Under these conditions, the abiotic dissociation of cellulose may be an important albeit slow degradation process, with isosaccharinic acid (ISA) as a major by-product. Slow degradation under alkaline conditions promotes the long-term stability of a sealing layer, and is a positive result of mixing fly ash and stabilized sewage sludge. However, metal – ISA complexes may form that thereby increase the mobility of metals (Svensson et al., 2007).

The laboratory experiments (Wikman et al., 2003c) indicated that the degradation product ISA resulted in an increased content of lead and zinc in the leachate from fly ash. When the experimental conditions were adjusted to correspond to a covered deposit after 250 years the leaching, lead leaching increased from 31 to 39 % and the leaching of zinc from 1.8 to 2.3 % when the content of ISA was increased 20 times. Wikman et al. (2003c) concluded, however, that cellulose degradation under alkaline conditions will be slow. Slow cellulose degradation, combined with a low percolation rate through the cover, would therefore result in only minor metal releases from a sealing layer to the underlying landfill.

Laboratory experiments (Wikman et al., 2005) were conducted in order to investigate the long-term stability of fly ash – sewage sludge mixtures. The main focus for the studies was on the leaching of organic material (as TOC in the solid phase) from compacted ash-sludge mixtures and on the development of hydraulic conductivity with time. The experiments were conducted as 2<sup>7</sup>-1 reduced factor experiments with seven factors (ash-sludge material, ash-sludge ratio, biological activity, freezing, drying, initial water content, and compaction) studied in 72 permeameters. Factor values were chosen for extreme conditions in a sealing layer. The results indicated varying degrees of TOC leaching over the course of several experimental cycles and under abiotic conditions, supporting the conclusion that TOC loss occurred primarily through the leaching of organic material from the volume. For low conductivity mixtures (< 10<sup>-9</sup> m/s), TOC leaching rapidly attained quasi-steady state conditions, and did not have an adverse

effect on permeability; instead, a decrease in conductivity occurred as a consequence of increased compaction. The study stresses the importance of maintaining moist conditions in an ash-sludge liner or sealing layer and that adequate compaction and homogeneity is required for low leaching of organic material and for maintaining low permeability.

### **2.3 The monolithic behaviour of ash in landfills**

Because of its high pH and calcium content, moist ash tends to harden upon drying and carbonatation to a concrete-like substance. A continuous layer of ash in a landfill could thus be expected to perform as a monolith under certain conditions (*e.g.* water content, differential subsidence, and composition). Such monolithic behaviour is desirable in that it would promote increased stability and decreased water percolation.

The Tveta landfill has been the focus of a number of studies on ash use in landfills. The injection of ash for the stabilisation of landfills and to prevent differential subsidence was investigated in an early study (Wikman et al., 2003a). A pilot-scale study was conducted where ca. 100 tons of fly ash slurry was injected at a depth of three to nine meters below the ground surface into the landfill body. Very little pressure was needed to inject the slurry, and normally no back-pressure developed during the injection. After the completion of the project, the landfill was excavated in one region where injection occurred so as to ascertain the degree of spreading of the injected ash slurry. However, during the excavation, only a smaller fraction of the total injected ash volume was recovered, and it appeared that the distribution of ash in the subsurface was random. The study was inconclusive with regard to the effect of ash injection on increased ground stability and decreased subsidence since the injected ash volume was very small compared to the effected landfill volume. Nevertheless, the injection of fly ash was successful despite complications (*e.g.* clogging of the injection equipment), demonstrating that the technical difficulties were surmountable. However, the final project costs for fly ash injection were relatively high, such that the stabilisation method is too expensive to be considered as an option for landfill stabilisation.

In 2000, Telge Återvinning Company received permission to install a final cover consisting of ash and other secondary construction materials from waste products on a 4 ha municipal waste landfill at the Tveta waste recycling centre. Tham and Andreas (2008) described the basic characterization of the sealing layer material, the construction of the final cover, and results from field experiments. The landfill sealing layer was constructed using a combination of different materials, including fly ash, bottom ash, Friedland clay, and various slag products. Laboratory studies were used to study such material properties as ash hardening and carbonatation. These two properties are of great importance for the long-term functionality of a sealing layer. The study indicates that the ash material appears to be able to adapt to local subsidence in the municipal landfill by maintaining a certain degree of elasticity over a period by remaining moist. Hardening of the ash continues over the course of many years, and the authors claim that the ash slowly transforms to clay minerals, which is advantageous for the long-term function of a sealing layer.

At the study site, six test areas were established in flat and steep areas with different cover materials. Lysimeters were installed under the sealing layer at each test area, and indicated a percolation of 1 – 22 l m<sup>-2</sup> yr<sup>-1</sup>; the percolation rate was always much less than the requirement of 50 l m<sup>-2</sup> yr<sup>-1</sup>. Leachate and drainage water samples were analyzed for nitrogen compounds, chloride, and various metals and non-metals.

In accordance with literature data, the field observations show that a cover constructed only with ash material can under certain conditions form a monolithic structure.

### 3 Ash as a barrier in mining environments

Certain waste materials are reactive in contact with oxygen and water. Sulphide-rich mining waste is one such material, where oxygen will react with sulphide minerals in the waste, producing sulphuric acid and releasing metals bound in the sulphide minerals. Water percolating through the deposit will transport metals and acidity into the environment. Therefore, the long-term disposal of oxidizable wastes requires the installation of a sealing layer that prevents both the diffusion of oxygen and the penetration of water into the waste deposit.

Mine waste deposits, both tailing impoundments and waste rock deposits, are traditionally covered in Sweden with a low permeability sealing layer composed of clay or clay-rich glacial till. This layer is in turn covered with a protective cover (often glacial till) to prevent the erosion and desiccation of the sealing layer. Large amounts of clay or glacial till are needed in the construction of a sealing layer, and these may be limited resources in the geographic area in which the mine waste is located. In addition, the extraction of clay and till changes the landscape in a way that may be unacceptable in certain areas. Instead, alternative materials may be used in the construction of a sealing layer, such as ash.

In addition to their function as a sealing layer, alkaline combustion residues can be used for the stabilization of mine wastes and in reactive barriers for the neutralization of acidic leachate from mine wastes. Metals may be preferentially retained in such barriers under alkaline conditions.

This chapter presents projects conducted within the Ash program that have investigated ash as a component in sealing layers over mine waste, for mine waste stabilization, and as a reactive component in barrier systems. In terms of the covers, it should be noted that in many cases these covers do not greatly differ from covers installed on municipal landfills (see Chapter 2). However, for municipal landfills, the function of the cover is primarily for the prevention of water infiltration and hence for reducing leachate generation, while for mine waste deposits this function also includes a barrier to oxygen diffusion and the prevention of sulphide oxidation. As shown in many studies, low permeability, water-saturated sealing layers with a perched water table is effective barriers to both water and oxygen penetration; such sealing layers are appropriate for both municipal landfills and mine waste deposits.

#### 3.1 Ervalla

Between 1995 and 2002, the Ervalla deposit near Örebro was covered with sealing layer consisting of 1-2 meters of fly ash, which was covered with ca. 20 cm of sewage sludge mixed with fly ash (20 % by volume; Bäckström and Johansson, 2004). Infiltration through the sealing layer is estimated to 40 – 80 l m<sup>-2</sup> yr<sup>-1</sup>, based on measurements of hydraulic conductivity ( $2.6 \times 10^{-10}$  m s<sup>-1</sup> –  $5.2 \times 10^{-10}$  m s<sup>-1</sup>). Compared with contemporary tailings deposits in which the tailings thickness is often in the order of tens of meters, the Ervalla tailings were deposited in a thin layer of 20 – 40 cm.

Findings from 2003 and 2004 (Bäckström and Johansson, 2004; Bäckström and Karlsson, 2006) indicate that covering the deposit led to a decrease in the leaching of some metals, whereas the leaching of other metals increases. Samples collected from surface water draining from the deposit, from before and after the installation of the cover, demonstrate an increase in surface water pH, a decrease in the concentrations of iron, nickel, cobalt and lead, and an increase in the concentrations of arsenic, barium, chromium and copper. Arsenic was most likely derived from the fly ash, as arsenic concentrations were higher in the fly ash than in the mine waste. Groundwater samples collected under the sealing layer indicated a higher pH and greater concentrations of total organic carbon, potassium, calcium and sodium, suggesting that the sealing layer was relatively permeable, allowing the percolation of these constituents from the sealing layer into the underlying mine waste. Leaching tests on the weathered mine tailings indicate that a large fraction of many elements are bound to a reducible fraction, such as to iron or manganese oxides (Bäckström and Karlsson, 2006). There is thus a risk that a high water table and anoxic conditions under the sealing layer may result in the mobilization of metals bound to mineral phases that are soluble at low redox potentials. Long-term monitoring is necessary in Ervalla in order to determine if the ash/sludge cover has led to an improvement in water quality near the deposit.

### **3.2 Gillervattnet – establishment of vegetation and root penetration**

Pilot-scale field studies with fly ash and sewage sludge have been conducted at the Gillervattnet tailings deposit in Boliden (80 ha) and the Western tailings deposit in Garpenberg (Greger et al., 2006; Greger et al., 2009). In Boliden, the pilot-scale application of fly ash, sewage sludge, and an ash/sludge mixture were investigated as materials in a sealing layer in three plots with a size of 0.3 – 1 ha (Figure 3-1). Vegetation was later established in a protective cover over the sealing layer. The primary focus of these studies was on the function of the sealing layer and the ability of different plant species to penetrate the sealing layer with their roots, and the subsequent impact on drainage water quality. In addition to the field study, laboratory experiments were conducted that studied plant growth and root penetration. The sealing layer test cells in Boliden were constructed in 2003-2005, while fly ash was applied to test cells in Garpenberg in 1998, prior to the start of the studies by Greger et al. (2006, 2009).

Greenhouse experiments (Greger et al., 2006) indicated that a sealing layer composed solely of fly ash was preferable to an ash-sludge mixture, since plant roots have a much lower capacity for penetrating a compacted ash layer. These results were confirmed in the field study (Greger et al., 2009). In addition, ash is generally toxic to plant roots because of high pH, high alkalinity, and high levels of potentially toxic substances, such as heavy metals. In the greenhouse experiments, concentrations of many elements were however higher in drainage water from mine waste covered with fly ash compared to drainage water from mine waste covered with sludge or mixtures of ash and sludge; sewage sludge thus seems to have a preventive effect on metal leaching caused by the ash.

Root penetration was studied under field conditions in both Boliden and Garpenberg, while nutrient and metal leaching was also studied in the Boliden test plots (Greger et

al., 2009). The study indicated that root penetration can be inhibited if the sealing layer is constructed with a penetration resistance greater than 2.5 MPa. The addition of sewage sludge to the sealing layer is not advisable, as this lowers the layer's penetration resistance. Greenhouse experiments (Neuschütz and Greger, 2008) indicate, however, that sewage sludge in the sealing layer may promote the retention of metals in the sealing layer. Regardless of the composition of the sealing layer, the release of nutrients and metals decreases with time.

The plant species established in a protective cover are relevant for the degree of root penetration through a sealing layer. The roots of reed canary grass may weaken a hardened ash layer by decreasing pore water pH and consuming water, thereby desiccating the sealing layer. The risk for root penetration may therefore increase with time. Coltsfoot and birch are plant species with a lower ability for penetrating hardened ash layers. The energy crop reed canary grass was observed to reduce the leaching of nutrient elements from sewage sludge and fly ash, partly by decreasing the amount of drainage water, but also by changing the geochemical conditions in the substrate (e.g. pH and redox potential; Greger et al., 2006). Reed canary grass, however, has the capacity to weaken and penetrate even hardened fly ash sealing layers with a penetration resistance of ~5 MPa; the study suggests that the secretion of saccharides by plant roots may contribute to this process (Greger et al., 2009).

While the sealing layer constructed in Boliden (Greger et al., 2006, 2009) covered an area of less than three hectares, the total area of the Gillervattnet tailings impoundment is 80 ha, which is typical for this type of deposit. There is thus a great potential for the application of large volumes of ash in covers on mine tailings deposits. A major issue in this case is the transport of boiler and incinerator ash from urban areas where it is produced to more remote areas where many base metal mines are located.

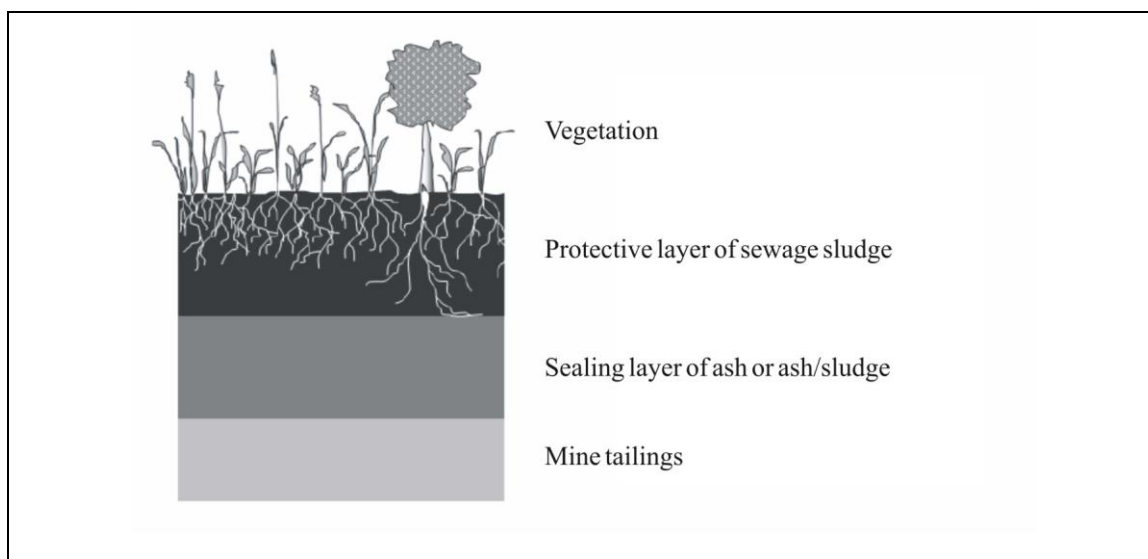


Figure 3-1. Schematic picture of a soil cover containing fly ash and sewage sludge (from Greger et al, 2006).

### 3.3 Mine waste stabilization and leachate treatment

In the Bergslagen region of central Sweden, there are large quantities of waste materials from ore and metal processing dispersed in deposits of varying size throughout the region. Estimates indicate (Bäckström et al., 2009) that there are ca. 3 270 000 m<sup>3</sup> waste rock, 14 300 000 m<sup>3</sup> tailings, and 1 600 000 m<sup>3</sup> slag that require remediation to prevent future environmental impact (for example, see Figure 3-2). While some of these deposits may be treated with traditional soil covers to minimize oxygen and water intrusion (see above), many of these deposits are of historical value, residing in a cultural setting where there are strong interests in preserving the rock piles (i.e. waste rock dumps). However, many of these waste rock dumps produce an acidic leachate which may be detrimental to the local environment. Therefore, with this in consideration, Bäckström et al. (2009) have studied the stabilisation of the mine waste with alkaline combustion products and other residues, in order to determine if stabilisation may be an effective solution for leachate mitigation.

In the study by Bäckström et al. (2009), laboratory and pilot-scale field experiments were conducted. The results from the laboratory experiments indicated that waste dump samples treated with fly ash have a significantly higher pH compared to the samples treated with industrial residues containing carbonates (lime mud and green liquor dreg; in Swedish *mesakalk*, *grönlutsslam*, respectively) (pH 10 compared to pH 8). However, the alkalinity levels show the opposite trend, with significantly higher alkalinity in the samples treated with carbonate materials compared to the samples treated with materials dominated by hydroxides (fly ash). Trace metal concentrations in the leachates were reduced by > 99% in the samples treated with carbonate materials, regardless of amount added. For fly ash, an addition of at least 10% fly ash (90% weathered waste rock, by weight) was needed to reach a 99% reduction in metal concentrations. According to Bäckström et al. (2009), the dominating chemical mechanisms behind the reduction of trace metals were most likely sorption and precipitation.



Figure 3-2. Rock waste deposit at Ljusnarsberg mine site (left) and close – up photo of weathered, acidic rock waste that requires treatment.

The pilot-scale stabilization experiments (Bäckström et al., 2009) were conducted in 1.5 m x 0.8 m (length x diameter) columns filled with weathered mine waste from the Ljusnarsberg mine. In the stabilisation experiments, the mine waste was interlayered with fly ash, lime mud and green liquor dregs. The results of the stabilisation experiments indicated that the leachate pH in the treated systems (pH 5.8 – 6.8) had increased between 0.8 and 1.8 pH units, compared to the untreated reference system. The increase in pH resulted in a significant decrease (on average 98%) in trace metal concentrations. The main mechanism behind the reduction is suspected to be metal sorption and, to a lesser degree, precipitation. Stabilisation with aged fly ash results in the best performance in terms of pH increase and metal concentration decrease.

For the treatment of acidic, metal-rich mine leachate, Bäckström et al. (2009) developed a reactive filter system. A pH increase to  $> 6$  was achieved with systems containing either fresh fly ash or lime kiln dust as the initial neutralisation agent. While copper and lead exhibited a reduced mobility in most treatment systems, pH levels below 6 did not result in an effective removal of zinc and cadmium; the higher pH levels maintained in the filter systems with fresh fly ash and lime kiln dust were required to remove a substantial fraction of dissolved zinc and cadmium. The results (Bäckström et al., 2009) indicate that the filter systems that contained alkaline material dominated by hydroxide alkalinity performed best. Bäckström et al. (2009) concluded that the use of alkaline by-products to neutralise acidic mine waste and acid mine drainage from historical mine sites may give rise to both environmental and economical benefits and should therefore be encouraged as a sustainable remediation method.

## 4 Ash in roads – studies from a geotechnical perspective

Considerable amounts of raw materials are used annually in Sweden for the construction of roads. Crushed rock is a traditional road construction material in Sweden, and is generally not considered a limited resource. However, excavation and transportation costs can be high, depending on the site location for road construction, and rock excavation with associated forest clearing may leave a long-lasting imprint on the local environment. The selection of alternative materials, such as bottom ash, in road design is therefore a method of reducing the amount of excavated rock needed.

The selection of materials for road construction is based on a number of criteria, including geotechnical properties, availability, and cost. Geotechnical properties such as the bearing capacity and durability of unbound materials in a road can be improved through stabilization, i.e. the mixture of a binding agent with a material to improve their composited geotechnical properties. Cement, Merit<sup>5</sup> and lime are examples of common binders for the stabilization of roads, but fly ash from solid biofuels and coal combustion is an alternative material which has been used on smaller roads and surfaces in Sweden and Finland. In Sweden, the Swedish Road Administration is the governmental authority responsible for issuing technical requirements for the design and dimensioning of roads and road drainage. If an alternative material is to be accepted in road construction, it must pass the criteria established by the Swedish Road Administration.

It may be noted that the Ash Programme has focused on the use of combustion residues in roads; other geotechnical applications (e.g. parking lots, industrial surfaces) have only been studied to a lesser extent, despite predictions that the use of ash as aggregates in other types of pavement applications will be of increasing importance in the future. Indeed, because of the seasonal availability of boiler and incinerator combustion residues and the relatively small volumes produced at individual district heating plants and incinerators, the application of combustion residues to large-scale road construction projects may be limited. Instead, the temporal and spatial availability of combustion residues may make them more suitable for smaller applications such as parking lots, industrial surfaces, and local roads.

An underlying goal for all projects that have focused on the application of combustion residues in road construction has been to increase the level of confidence for the material properties of combustion residues, in terms of both geotechnical and environmental properties, thereby improving the basis for decision making. Advantages associated with the use of combustion residues as a binding agent in the base and sub-base layers of roads is emphasized in many projects. In addition, the potential environmental impact of combustion residues in roads has been a critical aspect in many

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<sup>5</sup> “Merit 5000” is a certified product from the Swedish steel industry, produced from dried, air-cooled blast furnace slag, a waste product. This material is commonly used in Swedish studies of alternative construction materials because the material has passed a certification process, is available on the market, and has a number of advantageous properties.

studies (see Chapter 5), especially in terms of the long-term fate of combustion residues after the decommissioning of roads (i.e. after tens to hundreds of years).

## **4.1 Geotechnical properties of combustion residues**

In order for combustion residues to be accepted as alternative materials in road construction, it is necessary to establish the geotechnical properties of these materials and how these materials fulfil the requirements of the Swedish Road Administration. Several studies in the Ash Programme, including von Bahr et al. (2004), have investigated functionality requirements and testing methods.

### **4.1.1 Fly ash**

Early studies during the program period investigated the properties of combustion residues and their potential use in geotechnical applications, such as road construction. Mácsik et al. (2004) studied nine different fly ash products and grouped the materials according to technical properties and potential applications. Properties such as grain-size distribution, water content, void ratio, loss on ignition, compaction properties, and compressive strength development were investigated. See also Lahtinen et al. (2005c). A critical criterion for the use of fly ash was the stability of the material in the presence of water and after repeated freeze-thaw cycles. The results of this preliminary study indicated that the fly ash products could be grouped into three categories: 1) fly ash with low CaO content and with no compressive strength development that could not be used in the subbase of roads without binder additives such as cement and Merit; 2) fly ash with a medium-high content of CaO and which demonstrated an adequate compaction and compressive strength development; 3) fly ash with a significant compressive strength development and which can be used without the addition of other binders like cement. Fly ash from the last two groups are appropriate for application as landfill sealing layers, the subbase and road-base in gravel roads with low traffic volume, and as stabilizing agents in soils.

### **4.1.2 Bottom ash**

An inventory was conducted (von Bahr et al., 2004) of the functional requirements and associated testing methods for bottom ash in road construction and other applications. This project was motivated by the need for developing testing methods that are valid for combustion residues; traditional testing methods have been adapted for standard materials (e.g. gravel, crushed rock) and are not necessarily suitable for non-standard materials (i.e. combustion residues). Testing methods must therefore be developed that investigate the functional properties of combustion residues in a roadway. Functional requirements identified in this study for a variety of uses, including use in the road-base and subbase layers of roads, are: 1) the material must have acceptable bearing capacity and stability, 2) the material must have an acceptable degree of settling and compression, 3) the material must have acceptable frost, mechanical and chemical durability, 4) the material must not cause unacceptable frost heaving, 5) the material must not contribute to an increased risk for slippery road surfaces, 6) the material must have good drainage, 7) the material should not be difficult to handle. These functions and corresponding indicator properties are illustrated in Figure 4-1.

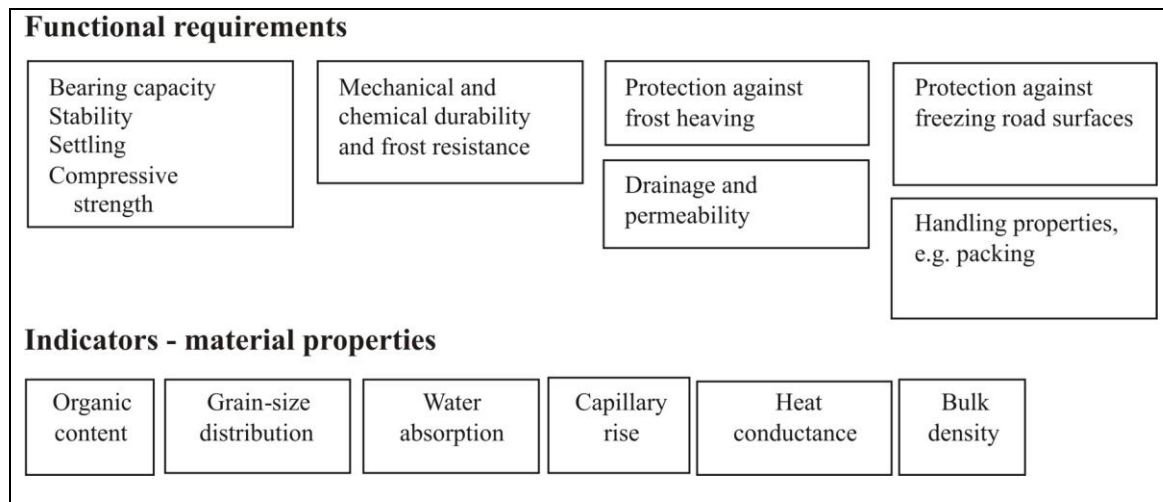


Figure 4-1. Functional requirements and related material properties to be considered in the evaluation of combustion residues for road construction. Adapted from von Bahr et al. (2004).

As a continuation of von Bahr et al. (2004), von Bahr et al. (2006) investigated three bottom ash materials in terms of a variety of mechanical parameters that are related to the functionality of combustion residues in road construction (cf. 0). This study indicated that certain properties exhibit a high degree of variability among bottom ash materials, including bulk density, water absorption, and grain-size distribution. In addition, many properties (e.g. capillary rise, modified compressibility, evaluation of bearing capacity and stability from cyclic load triaxial tests) are difficult to evaluate since there is a lack of test data from combustion residues in relation to field performance. A thorough characterization of combustion residues for geotechnical uses was recommended by von Bahr et al. (2006); such a characterization is necessary for the construction application and would provide valuable information for future planning.

There have been a number of studies within the Ash Programme that have studied the properties of combustion residues mixed with other alternative materials. As discussed previously (see section 2.1), fly ash mixed with activated sewage sludge is used in sealing layers on landfills. In another study (Lahtinen et al., 2005b), a mixture of fly ash from energy production and fibre sludge from the paper industry was investigated as a suitable material mixture for geotechnical applications. Laboratory tests indicated that specific mixtures were suitable for construction purposes because of the materials' elasticity and resistance to permanent deformation. This leads to a significantly greater frost resistance than conventional materials. In certain cases, the addition of a binding agent such as cement is necessary to prevent a loss in compressive strength after freeze – thaw cycles. A further benefit from the use of such mixtures was the very limited dust production during handling.

As indicated above, a number of geotechnical properties (e.g. bulk density, water absorption, grain-size distribution) are often investigated in combustion residues prior to their use in construction. Hemström et al. (2009a) point out, however, that the capillary


properties of combustion residues, often described in water retention curves, are rarely determined. An understanding of a material's capillary properties is highly relevant in the evaluation of leaching during the use of combustion residues in construction, the dimensioning of sealing layers consisting of combustion residues, and in the assessment of the long-term properties of waste. Work is in progress.

Experience with the use of alternative materials in road construction has indicated that it is difficult to predict the long-term evolution of such properties as stability and leachability. Processes such as carbonatation and reduction in grain-size occur during the ageing of alternative products, such as MSWI bottom ash, which lead in turn to changes in material stability and leachability. Arm et al. (2008b) have addressed these concerns in a study that has focused on providing a better understanding of the long-term properties of alternative materials and on proposing laboratory methods for accelerated ageing. The aim with developing such methods is to provide a method for the prediction of the long-term properties of newly-produced alternative materials, prior to use in a geotechnical application. In the study by Arm et al. (2008b), steel slag and bottom ash were collected from the subbase of roads constructed with these materials in 1996 (in Smedjebacken municipality) and 1997 (Törringevägen, see section 4.3.2), respectively. With regard to the bottom ash, the analysis of the aged material indicated that bottom ash from the pavement edge was more aged than bottom ash from the road centre, as suggested by differences in electrical conductivity and soluble salts leached from the subbase material. These differences were inferred to be caused by differences in water exposure, and not carbonatation reactions, as no difference in pH was found. Thus, Arm et al. (2008b) concluded that the leaching properties of bottom ash had not changed significantly during its ten years of residence in the subbase of the road. This implies that long-term leaching properties can be studied with percolation tests, although geotechnical properties such as stiffness and stability must be studied in artificially aged materials.

A method was successfully developed (Arm et al., 2008b) for the accelerated ageing of steel slag, with a goal of achieving the degree of carbonatation and leaching that was observed in the subbase of the road (Smedjebacken). The best result was achieved with exposure to carbon dioxide for seven days at moderate moisture content and 40°C. For the accelerated ageing of bottom ash, where the goal was to achieve the pozzolanic reactions that were observed during the analysis of samples from Törringevägen by scanning electron microscopy, treatment (with hydroxide addition) was not able to replicate the observed leaching properties in the field samples. The increased leaching of soluble salts from the aged material was likely the result of the decrease in grain size that occurred as a result of treatment. The authors suggest that future research efforts for accelerated ageing should involve other methods than a pH increase in order to achieve metal oxidation, glass reactions, and clay mineral formation.

### Road design

In general, a road can be divided into four layers, as illustrated below.

Wearing course	
Road-base	
Subbase course	
Subgrade	

The *wearing course* or surface course is the top layer of the road pavement and is generally designed to be impervious to water penetration, be durable, and is chosen so as not to deform under the weight of traffic appropriate to the road. The *road base* provides additional load distribution and contributes to drainage. This layer can be both stiff and impervious under a poor asphalt wearing course so as to prevent water penetration into the road fabric. The wearing course and base course can be grouped together and termed *surfacing*.

The *sub-base course* is a load-spreading layer, especially during construction, carrying site traffic and protecting the subgrade. The *subgrade* is the layer of naturally occurring material the road is built upon, or it can refer to the imported fill material that has been used to create an embankment upon which the road pavement is constructed.

Material adapted from [www.highwaysmaintenance.com](http://www.highwaysmaintenance.com)

## 4.2 Preparatory studies prior to demonstration projects

Svedberg et al. (2008) investigate applications where binders are used for the stabilization of unbound layers in the road-base and subbase of roads. Two different applications for a full scale demonstration are developed: one for use in gravel roads, and the other for use in paved roads, where a highway bypass around the city of Sala is chosen as a reference location; fly ash, cement, and Merit are chosen as binders. An evaluation is presented of the technical and environmental properties of the binders and ballast in different road layers, including a description of the production of the materials, road construction, and economical aspects. The goal of the developmental work is that the report will provide a basis for evaluating the potential applications of fly ash as a binder. According to the report, various properties (bearing capacity, frost heaving, risk for frozen road surface, drainage) properties are listed which should be investigated in laboratory investigations during project planning.

It is proposed that material mixtures should be studied in a three-stage programme that includes the characterization of the material, identification of appropriate mixtures, and a detailed investigation of selected mixtures. In the second stage, it is suggested that material mixtures should be evaluated based on requirements for compressive strength, the development of shear strength, and frost resistance, while in the third stage the mixtures are further investigated in terms of quality and production considerations, frost heaving, and leachability.

The results of the project, using fly ash and ballast acquired within the Sala region, indicate that stabilization of unbound layers will improve the bearing capacity of the

road construction significantly, even though the total depth of the structure is reduced. The suggested applications (*i.e.* binders mixed with crushed rock in the road base) do not exhibit frost heaving although fly ash exhibits a particle size distribution like silt. The durability against frost and thaw cycles has been assessed and is expected to be acceptable. The fly ash used will need addition of cement and Merit to perform well in freeze and thaw tests; this has been concluded in other studies as well (Lahtinen, 2001). Applying both materials will result in a reduced depth of the structure and the thickness of the subbase layers can be greatly reduced.

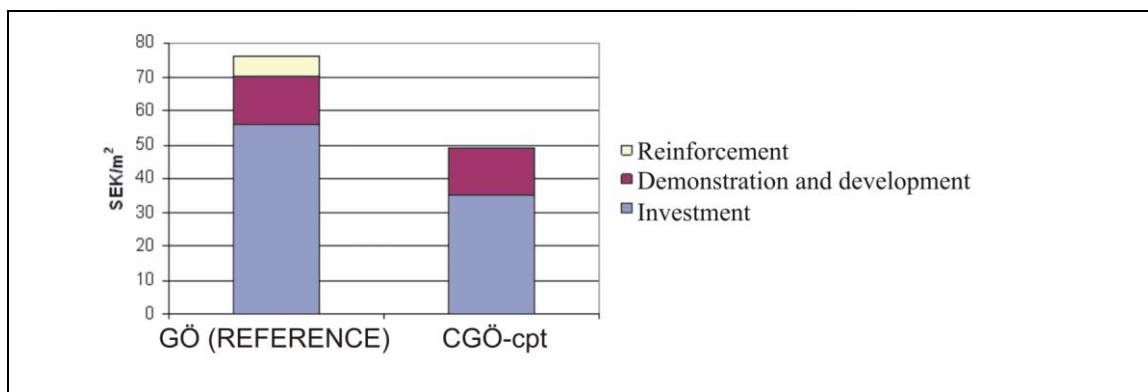


Figure 4-2. Results from the LCC-analysis shown as today's value in a 40 year life cycle (from Svedberg et al., 2008)

Not only does this project consider the geotechnical requirements for road construction, it also considers the economics of the construction project using life cycle cost (LCC) analysis. The LCC calculations indicate that the investment costs for a road using a stabilized layer are slightly higher than the investment costs for the reference construction. This is probably due to the relative short sections to be used in the proposed demonstration sections (Sala city bypass). However, the calculations indicate that the life cycle cost is likely to be 15 – 25 % lower for the fly ash applications than the reference over a considered time span of 40 years and a interest rate at 4 % (Figure 4-2), and that the construction project is therefore economically feasible over this timeframe.

### 4.3 Pilot scale demonstrations

A number of pilot-scale demonstration projects have been conducted, and are summarized in Table 4-1, below. A brief description of the various projects is presented in the following section.

Table 4-1. Summary of demonstration projects for road construction with combustion residues.

Site	Date of completion	Road use	Combustion residue application
Norberg	2003	Forest road	Fly ash in base and subbase course
Börje	2002	Forest road	Fly ash + gravel in base and subbase course
Hallstavik	2004	Forest road	Fibre sludge fly ash
Ofärne	2008	Forest road	Bark fly ash (+ green liquor sludge + cement + gravel) in base course
Törringevägen (Malmö)	1998	Road on outskirts of village Käglinge	Bottom ash, stored for > 2 years
Dåvamyran (Umeå)	2001	Transport road within Umeå Energi's facilities	Bottom ash, stored for 4 – 7 months
Gumpekulle – Vändöra (Linköping)	1987	Small cloverleaf outside of sewage treatment plant	Bottom ash

#### 4.3.1 *Norberg and Börje*

Prior to work in Sweden, it was shown in Finland (Lahtinen, 2001) that fly ash – stabilized gravel roads have a greater bearing capacity and a longer projected life span than gravel roads constructed without fly ash amendments. In order to investigate the effects of fly ash stabilization on the geotechnical properties and environmental impact of a road, two different pilot tests were conducted (Mácsik, 2006): one in Norberg and one in Börje (near Uppsala). Both road sections with related reference sections were investigated during a two year period. Only fly ash was used in the bearing layer in Norberg and fly ash mixed with gravel was used in Börje.

The results of the study (Mácsik, 2006) indicate that stabilization with fly ash increased the bearing capacity of the roads. However, stabilization is adversely affected by the effect of water and frost, and adequate drainage is therefore vital for long-term stability. Fly ash – stabilized roads did not exhibit a greater level of metal and non-metal leaching than the reference sections, with the exception of sodium, potassium and sulphate leaching, which was greater in the stabilized sections.

#### 4.3.2 *Törringe and Dåva*

In order to increase confidence in the use of combustion residues in road construction, more data is needed on the long-term performance of these roads, both in terms of the evolution of geotechnical properties and the long-term release of soluble compounds from the road materials. Such data was acquired in two follow-up studies (Arm, 2005) during 2004, conducted on roads constructed with MWSI bottom ash in 1998 (Törringe, outside Malmö) and in 2001 (Dåvamyran, near Dåva power station, Umeå). The bottom

ash, acquired from the combustion of household and industrial waste, was used in the subbase of both roads, instead of gravel (see Figure 4-3). The material at the Dåva site had been stored half a year prior to use, while the Törringe material had been stored for more than two years. Both test roads were compared with reference roads, and falling weight deflectometry measurements were used to study the layer elastic moduli for different layers and the surface modulus for the whole road structure.

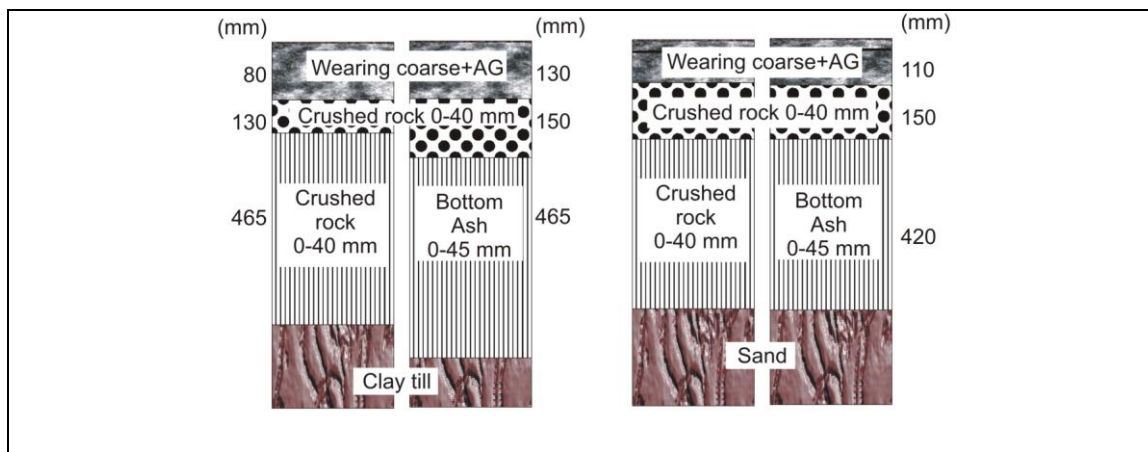


Figure 4-3. Illustration of materials used in road construction in Törringe (left) and Dåvamyrän (right). For each site, the reference structure is shown on left, test structure on right. Bottom ash is used as a subbase course in the test structure, overlain by crushed rock in the base course. AG = asphalt-bound gravel

The result shows that in Törringe, the stiffness of the MSWI bottom ash did not change compared with the results from earlier measurements. However, in Dåva, the stiffness of the test structure was lower in 2004 than in 2002. In both test roads, the test structure had lower stiffness than the reference structure. In Törringe, the ratio between the stiffness of the test structure and the stiffness of the reference structure was unchanged. In Dåva, however, the stiffness ratio had increased. This increase in stiffness may be related to the shorter storage time before use for the older bottom ash.

The study by Arm (2005) illustrates the need to identify geotechnical parameters that reflect the true performance of a road construction with alternative materials (*e.g.* combustion residues). The parameter “stiffness” is currently used by the Swedish National Road Administration, and MSWI bottom ash has been shown to have a lower stiffness than crushed rock with a larger particle size. However, the parameter “bearing capacity” may be more appropriate for applications with combustion residues, as this reflects a materials resistance to permanent deformation / settling during loading.

In a subsequent study (Arm et al., 2008a), conducted 10 years after the construction of Törringevägen and 7 years after road construction at Dåvamyrän, it was shown that bottom ash used as the sub-base material retains its strength after this period of time (Figure 4-4). At the Dåva facility, the measurements revealed that strength initially decreased with time, but then stabilised. As in previous studies (Arm, 2005), the strength of the bottom ash was about 70% of that of the crushed rock, which has to be taken into

account in the design phase. According to the strength properties, Arm et al. (2008a) report that bottom ash is suitable for use as sub-base material if the road is properly designed. It can also be used as a filling material, in embankments and as a capping layer. This result confirms the conclusions from previous studies.

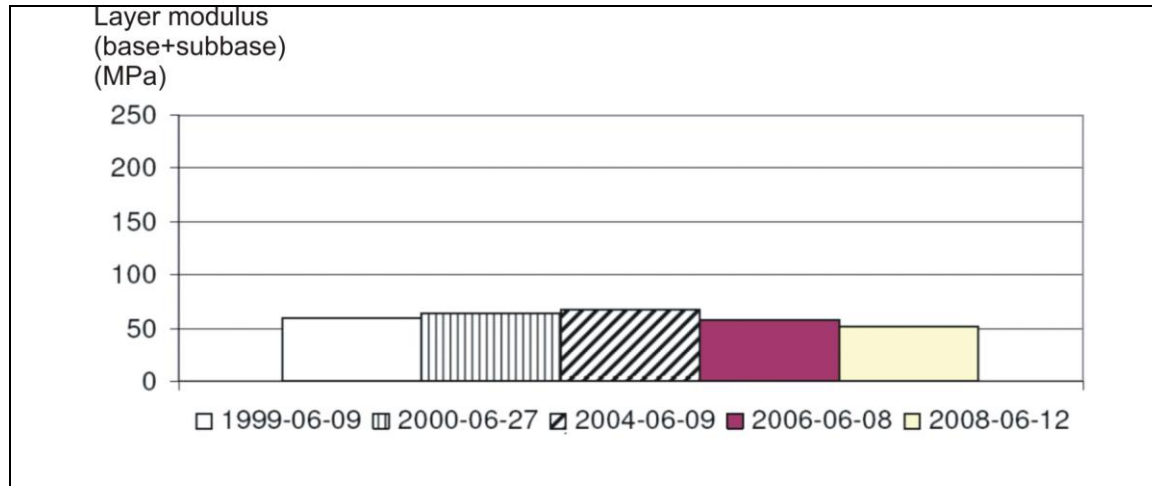


Figure 4-4. Results from falling-weight deflectometry tests on Törringevägen test road, showing stiffness change in test structure. Layer moduli for the combined layer of base and sub-base are evaluated with falling-weight deflectometry measurements. Mean value of ten points. Test structure designed according to Figure 4-3.

### 4.3.3 Hallstavik

Older gravel roads are often in need of renovation when their bearing capacity declines. To increase bearing capacity and long-term durability, binding materials (e.g. cement) can be added to the gravel. In a study by Mácsik and Svedberg (2006), fibre sludge (paper mill) fly ash was compared with other binder materials (cement, Merit 5000, lime) for later use in a full-scale test in Hallstavik. The materials were tested in terms of compressive strength development, durability to freeze – thaw cycles, and thawing resistance. The results of the laboratory study indicated that fly ash / cement and fly ash / Merit mixtures with a 3% cement or Merit addition had a positive effect as a binder, since there was an increase in the durability to freeze – thaw cycles. As a single component binder material, the addition of fly ash to gravel increased the mixture's stability, with the greatest stabilization effect (improved freeze-thaw and frost durability) measured at a 30% fly ash addition.

Following the preliminary laboratory investigation (Mácsik and Svedberg, 2006), the results were used for the renovation of a road in 2004 in Hallstavik. This use of fly ash as a binder material demonstrated the applicability of fly ash and also conferred a great amount of practical experience in handling fly ash. For the full-scale test, the road was divided into sections, including a reference section. For one of the sections, fly ash was first laid out and then covered with gravel, whereupon both materials were mixed together with a road scraper. In another section, the gravel was first laid out and covered with fly ash prior to mixing. Water needed to be added to the fly ash prior to mixing in

order to obtain optimal compaction. Measurements by falling weight deflectometry from before and 12 months after road renovation indicate an increase in the road's bearing capacity, as reflected by improvements in surface modulus and stiffness. Water collected from lysimeters installed below the road bed indicated a greater leaching of soluble compounds (sodium, sulphate) in the fly ash – stabilized road sections, compared with the reference section, but these concentrations were expected to decrease with time. After five years, the lysimeters show that pH and electrical conductivity as well as contents of sodium, sulphate, and other soluble compounds decrease

#### **4.3.4 Ofärne**

In the Ofärne road construction project (Mácsik et al., 2009) fly ash and green liquor sludge from Iggesund Paperboard were used as binders in the stabilization of a gravel road. The aim of the project was to improve the bearing capacity of a gravel road, mainly during the thawing period in the springtime. An initial laboratory investigation identified an appropriate mixture of binders (fly ash, green liquor and cement), based on the mixtures' packing properties, bulk and dry density, compressive strength and strength reduction after a number of freezing cycles. The results of the laboratory tests demonstrated that a mixture consisting of 16% fly ash (by weight), 3.6% green liquor sludge, 0.4% cement and 80% gravel would improve a gravel road's stability and bearing capacity, and also improve the frost resistance of the base course.

During the spring of 2008, two road sections near Ofärne (ca. 10 km west of Iggesund), about 2 km in length, were stabilized with a base course using 150 mm of the fly ash mixture. The stabilized road was investigated during autumn 2008 with regard to bearing capacity and environmental impact. Deflection measurements performed during October 2008 showed some improvement of the stabilized road's bearing capacity compared to the reference section. A significant improvement was that the left and right wheel track had a more homogeneous surface modulus than in the reference section. Surface water samples did not show any impact from the stabilized road section. The road section will be further investigated during spring 2009, after the snow has melted, before it is evaluated.

#### **4.4 Manuals for road construction with fly ash and bottom ash**

One barrier to the use of combustion residues in geotechnical applications is the lack of experience with road construction companies in using such materials, and the lack of "good examples" that lead the way for others to follow. Therefore, two manuals (Munde et al., 2006; Tyllgren, 2008) have been written in order to facilitate the use of combustion residues in primarily road construction.

In order to surmount such obstacles, a manual (Munde et al., 2006) was written in order to facilitate the use of fly ash in road construction, with the manual written primarily for road administrators, environmental authorities, and industry (fly ash producers). In this context, fly ash is used in the base and subbase of gravel roads. The manual emphasizes the advantages of using fly ash in road construction, including improvements in

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stiffness, stability and freeze-thaw durability relative to traditional road construction using sand or gravel. The report lists primary and secondary properties of the raw materials that should be considered in determining the viability of material choices (e.g. compressive strength, dry density, frost durability, water ratio). In this context, a material's compressive strength is a primary parameter that is used to evaluate the material's applicability for a specific function, and is evaluated using either a one-dimensional or triaxial stress test, where the triaxial stress test is preferred. A number of case studies are given as "good examples" in the report, including the use of fly ash in forest roads (Norberg, Hallstavik), in gravel roads (Börje; Luopionen, Finland), and as filler material and in surfaces for bearing heavy loads (Librobäck, Mälardalen).

In order to increase the use of waste products that occur in large volumes and to thereby reduce the deposition of these products in landfills and the extraction of natural resources, a handbook has been written for the use of bottom ash from municipal and industrial waste incineration in grate furnaces (Tyllgren, 2008). The use of bottom ash in combination with crushed rock is a concept where the strengths of each material are accentuated when the products are used as a composite. For example, bottom ash has relatively low compressive strength, but the compressive strength increases considerably as a composite with crushed rock. A critical factor affecting the properties of bottom ash is the degree of combustion / sintering. The rock material provides structural strength while the bottom ash functions as a filler and binder. For the unbound road-base in a road, the crushed rock no longer needs to be ground to the grain-size of sand in order to fulfil the proper functionality; a larger grain-size is sufficient when combined with bottom ash. The handbook presents a structured and quality assured production process, which is needed in order for a bottom ash - crushed rock composite material to be used on a large scale. The intention with the manual is that experienced manufactures and distributors of ballast material can produce and utilize a composite product with information in the manual. Based on the descriptions in the manual, material safety data sheets and building product declarations can be developed as part of certification and quality assurance.

In general, the production of a bottom ash - crushed rock composite material can be divided up into the following steps: 1) production of bottom ash, 2) sorting and storage of bottom ash to permit ageing and carbonatation, 3) stabilization, when appropriate (e.g. with crushed asphalt), 4) washing to decrease silt content and reduce salt content in leachate water, 5) choice of components for composite material, 6) manufacture of road material, 7) choice of proportions of various materials based on end-use. The manual presents three case studies where mixtures of bottom ash, crushed rock, and reclaimed asphalt were tested and used in roads.

## **5 Ash as a construction material – studies from an environmental perspective**

Because of their nature and origin, the composition of combustion products reflects the original composition of the ignited materials. Since most of the organic matter in fuels will be released as carbon dioxide and water vapour, volatile elements and compounds will concentrate in fly ash, while non-volatile elements and recalcitrant compounds will concentrate in bottom ash. In using combustion residues as construction materials, the occurrence, chemical form, and mobility of compounds in combustion residues, combined with an assessment of the relative risks posed by the compounds to humans and the environment, are central issues when determining their applicability and appropriateness in construction. Various types of leaching tests combined with geochemical / hydrogeological modelling are the primary tools used to evaluate the potential mobility of compounds from combustion residues into the environment; it is the interpretation of these leaching tests, combined with model results, that provides a basis upon which decisions can be made on the use of combustion residues in a geotechnical application (e.g. road).

The Ash Programme has supported research that can potentially provide a foundation for the implementation of large – scale construction projects using combustion products as construction materials. Therefore, it is crucially important to evaluate the potential impact of combustion residue use on humans and the environment. A series of studies has focused on the environmental legislation (Håkansson et al., 2004) that affects combustion residue use and on developing environmental guidelines (Bendz et al., 2006c) that reflect the potential environmental impact (e.g. dust dispersal; Gustafsson et al., 2006) of combustion residue use in primarily road construction. The use of bottom ash in district heating culverts and the leaching properties of such materials has also been the subject of study (Pettersson et al., 2004).

### **5.1 Legislation and the use of combustion products**

Håkansson et al. (2004) have reviewed the existing legislation in Sweden in terms of the regulation of combustion product use in construction. The potential use of combustion residues as construction material is influenced by environmental legislation in Sweden, covered primarily through the Swedish Environmental Code (*Miljöbalken*). The code does not specifically address the use of alternative materials but regulates the risks that could result from their use; the general considerations that must be taken into account regarding the environment are the same no matter if conventional materials are to be used or if combustion residues are used. The same concern for the environment is applicable irrespective of whether the combustion residues are regarded as waste, chemical products or a commodity.

For waste, including material for recycling, procedures and criteria for handling and deposition are regulated in the Landfill Directive and in the Swedish Waste Ordinance, but only regarding risk. When it comes to the benefits of combustion product use, there are wordings supporting a use in the Swedish Waste Ordinance, but only in general terms. The code for chemical products is very comprehensive and contains detailed

requirements on environmental and health investigations, product information, packaging and storage. Generally, these requirements concern only hazardous products, i.e. products that have properties corresponding to hazardous waste.

For use in construction, the status of combustion residues as waste or product will strongly affect the outcome of a permitting process. The End-of-Waste concept<sup>6</sup> is discussed in depth by Håkansson et al. (2004). Examples are given where alternative waste-derived or secondary materials have been allowed in construction according to the current legislation. The authors request clear and definitive decisions from the environmental authorities, defining the End of Waste criteria for a material or criteria for the use of a material when it is considered a waste. At the time of this report, there are no national guidelines that direct the use of combustion products in construction and there is therefore a considerable degree of interpretation involved when applying the Environmental code and other regulations.

## 5.2 Establishing environmental guidelines for ash use in construction

In a second phase to the project detailed in Håkansson et al. (2004), Bendz et al. (2006c) propose an environmental assessment method to be implemented when combustion residues are used in construction (*i.e.* roads). Furthermore, general environmental guidelines are determined for metal concentrations in combustion residues, with the intention of providing a unified basis for the future evaluation of combustion residue use in construction.

A risk-based model for exposure to metals and non-metals in combustion residues is used by Bendz *et al.* (2006c) for the determination of environmental guidelines. Exposure in this model is calculated using the same principles (*e.g.* total concentration – based exposure pathways) and calculations as used for determining the guidelines for contaminated soils (Elert et al., 1997). The establishment of guidelines can be divided up into three parts in terms of the description of model parameters and the calculation of 1) guidelines based on health risks and impact on soil quality from the dispersal of ash particles, 2) guidelines based on health and environmental risks from leachable content of combustion residues, and 3) concentrations in combustion residues that can potentially pose health risks, based on emissions from the construction after it is decommissioned. A transport model has been used to investigate the dispersal of compounds from the construction to the local environment.

Model calculations indicate that health risks during the operation of a road are dominated by exposure to particulate combustion residues that are dispersed from the road body. Calculated guidelines are presented in Table 5-1. Exposure through the consumption of edible plants that are exposed to particulates is in many cases (*e.g.* for arsenic, lead, mercury) the determining factor for the guideline level. Assumptions inherent in the calculation of these guidelines, such as the assumption that 30% of a

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<sup>6</sup> As a result of having been processed physically or chemically, the waste material loses the properties that make it a waste and becomes a product that can be traded as any industrial material.

person's vegetable consumption is derived from within 20m of a gravel road for the person's entire life, leads to conservative calculation of guideline values. According to Bendz et al. (2006c), a large safety margin is contained in the guideline values.

Bendz et al. (2006c) have produced guidelines for health and environmental risks based on the leachable content of combustion residues (Table 5-2); these guidelines apply for exposure through drinking groundwater and environmental impacts on nearby surface water. Three different emission scenarios have been modelled:

- Scenario I: Infiltration through base layer in non-surfaced road (gravel road)
- Scenario II: Groundwater and surface water penetration into the road, with and without surfacing
- Scenario III: Infiltration in the edges of a surfaced road (asphalt cover)

A major difference between the scenarios is the mode of transport. While transport in the first two scenarios is modelled as water flow through the road body, transport in the third scenario is dominated by diffusion. Many parameters in the model are chosen conservatively (e.g. leachate production, large concentration gradients at the roadsides which maximize diffusion, no dilution from road to well, high hydraulic conductivity in aquifer) such that transport to groundwater and nearby surface water is not likely underestimated. Other parameters, such as the distribution coefficient ( $K_d$ ), have been obtained from previously published values and are conservatively set so as not to underestimate transport. Distribution coefficients are very sensitive to pH and other local conditions and there is therefore a considerable degree of uncertainty in the  $K_d$  values used in the model. The model calculations based on the leachability of combustion residues indicates that many Swedish combustion residues have a lower metal leachability (as L/S 10) than the proposed guidelines (Table 5-2). However, for some combustion residues in gravel roads, the leaching of soluble compounds such as sulphate and chloride can pose more than an insignificant risk of impact on groundwater quality.

As a concluding remark, it may be noted that these guidelines have not yet been implemented in road construction using combustion residues. Despite the development of these environmental guidelines, it is apparent in Sweden that the environmental authorities use a different line of reasoning when considering the application of waste in construction, where the long-term fate of combustion residues after road decommissioning is a primary concern.

*Table 5-1. Guideline values for content of substances (mg/kg DS) based on the risk for the dispersal of combustion residues as particulates from a road. Values are calculated in order to correspond with “insignificant” environmental risk when used as a construction material in a 10m wide road without any additional safety measures. Adapted from Bendz et al. (2006c).*

Substance	Risk <sup>1</sup>	Non-surfaced road	Surfaced road
Antimony <sup>2</sup>		-	-
Arsenic	H	110	290
Lead	H	2 70	5 300
Cadmium	H	520	1 100
Copper	E	26 000	200 000
Chromium	E	31 000	240 000
Mercury	H	150	290
Nickel	H	1 400	2 700
Selenium <sup>2</sup>			
Zinc	E	64 000	500 000
Fluoride <sup>3</sup>			
Chloride <sup>3</sup>			
Sulphate <sup>3</sup>			
Naphthalene	E	2 600	20 000
Benzo(a)pyrene	E	64	500

Footnotes to Table 5-1:

<sup>1</sup> The risk is a sanitary risk (H) or an environmental risk (E)

<sup>2</sup> A guideline value could not be computed as there are no data on background level

<sup>3</sup> Dispersal of these substances as dust is deemed to be insignificant, no guideline value was computed

Bendz *et al.* (2006c) performed a risk assessment for the case when a road construction is decommissioned and the road material remains in place. Guideline values are depicted in Table 5-3. In the scenario used by Bendz *et al.* (2006c), the road is forgotten by the local population, children and adults use the area for recreation, and vegetables are grown in the soil that contains combustion residues.

Table 5-2. Guideline values for health and environmental risks based on the leachable content of combustion residues. "S" is the leachable amount of the substance, based on the model results, for a liquid-to-solid ratio of 10. "C<sub>0</sub>" is the initial concentration of the leachable substance that is produced in a leaching test. Table adapted from Bendz et al. (2006c).

Substance	Risk <sup>1</sup>	Scenario I and II		Scenario III	
		S (L/S=10) (mg/kg)	C <sub>0</sub> (mg/l)	S (L/S=10) (mg/kg)	C <sub>0</sub> (mg/l)
Antimony	H	0.84	0.24		
Arsenic	H	6.5	0.92	10	960
Lead	H	11	7.2	120	80
Cadmium	E	0.4	0.54	3	6.1
Copper	H/E	15	11	100	240
Chromium	E	33	15	120	750
Mercury	E	0.35	0.054	2	7.0
Nickel	E	1.7	1.2	25	8.9
Selenium	H	1.2	1.1	1.2	310
Zinc	H	64	45	250	1 300
Fluoride	H	120	65	500	7 900
Chloride	H	1 800	2 500	25 000	62 000
Sulphate	H	2 800	2 200	50 000	32 000
Naphthalene <sup>2</sup>		10		10	
Benzo(a)pyrene <sup>3</sup>		0.25	-	0.25	-

Footnote to Table 5-2:

<sup>1</sup> The risk is a sanitary risk (H) or an environmental risk (E)

<sup>2</sup> The transfer time is long enough for naphthalene to be fully degraded before it reaches a water supply well; therefore S is only based on transport to the soil underlying the road body

<sup>3</sup> Because of the high value of the distribution coefficient K<sub>d</sub>, the plume does not reach a water supply well in a relevant length of time; therefore S is only based on transport to the soil underlying the road body

Using this scenario, the calculations indicate that the arsenic concentration in combustion residues may pose more than an "insignificant" risk for exposure through dust production (particulate dispersal) from the road body. In other words, the average total arsenic concentration in Swedish combustion residues is greater than 15 mg/kg arsenic. As with the other model calculations (see above), the system boundaries and parameters are chosen with a large safety margin. The model parameters that have the greatest significance on the risk assessment are number of days exposure per year and annual consumption of vegetables that were grown near the road; the values in Table 5-3 have been calculated for an annual exposure of 40 days and an annual consumption of 1 kg vegetables. A system boundary that greatly affects the outcome of the calculations is that the combustion residues are completely exposed and can be freely spread with winds.

Table 5-3. Calculated guideline values for the content of toxic substances in combustion products in the surface layer of a road after decommissioning that could present a sanitary risk. Adapted from Bendz *et al.* (2006c).

Substance	Computed guideline value for content (mg/kg DS)
Arsenic	15
Lead	1 500
Cadmium	60
Copper	Not relevant (> 5 %)
Chromium (III)	Not relevant (> 5 %)
Mercury	60
Nickel	300
Zinc	Not relevant (> 5 %)
Naphthalene	300
Benzo(a)pyrene	5

The risk assessment provided by Bendz *et al.* (2006c) took only limited consideration of the potential additive effects of other contamination sources that may have an impact on human health and the environment. In consideration of this limitation, Wik (2009) provides an analysis of the composite risk posed by the large scale use of alternative road construction materials (*e.g.* combustion residues) on a regional scale. The catchment areas for Motala ström and the Emån river were used as test cases. The study calculated material flows for mercury, cadmium, and lead, which indicated that the large scale use of combustion residues in road construction in the catchment areas would lead to an insignificant increase in the emission of these metals in the catchments, compared with the natural flow of these metals and the base case where conventional ballast is used instead. For comparison, Wik (2009) also calculated the annual emission of cadmium, mercury and lead from roadways, assuming that the concentration of these metals in the road leachates equals the guideline values determined by Bendz *et al.* (2006c; *cf.* Tables 5-1 and 5-2). A comparison of these two calculations indicated that the latter emissions are much greater than the emissions calculated using material flows. Wik (2009) concluded that the guideline values proposed by Bendz *et al.* (2006c) pose a much greater risk than "insignificant risk" when examined with a regional material flow analysis. This is the case if all combustion residues that are produced in the region are used in construction.

If ash is left in the road after the road has ceased to be used, there is a risk that, among other compounds, heavy metals will be taken up by the vegetation that will have invaded the disused road. A lysimeter with wood ash was installed in 1993 at the SGI laboratory. After the immediate experiments were completed, the lysimeter, as well as other lysimeters in the test field were left exposed to the weather and to precipitation. During the 15 years since it was built, this lysimeter has been invaded by vegetation: birch, willow, various weeds. An analysis was performed of these plants' uptake by Hemström *et al.* (2009b):

- For birch and willow that have grown in wood ash or in crushed rock, there isn't any significant difference in metal content, except for arsenic which is higher in trees growing in wood ash. Willow has accumulated more arsenic, cadmium,

copper, nickel, lead, antimony and zinc than birch when both have grown on wood ash. In the case of crushed rock, only cadmium has accumulated more in willow

- The zinc content in willow leaves, both on wood ash and on crushed rock, is larger than the tolerable daily intake for grazing animals
- The availability of metals is lower in ash than in crushed rock
- The leachability at L/S=2 was less than 1 % in both lysimeters and it is slightly larger in crushed rock than in wood ash.

### 5.3 Caesium 137

Contamination of ash with the radioactive isotope caesium-137 ( $^{137}\text{Cs}$ ) is regulated in Sweden by an ordinance from Strålsäkerhetsmyndigheten<sup>7</sup> (SSMFS 2008:16). Combustion residues containing 0.5 to 10 kBq/kg of caesium-137, so-called contaminated ash, may be used for civil works provided that the content in a near-by well does not exceed 1 Bq/l and provided that the increase in a near-by recipient producing edible fish does not exceed 0.1 Bq/l.

The connection between caesium content and radiological impact in well and recipient is not immediately apprehended. Sjöblom (2009a) has assessed the migration of caesium from combustion residues through soils and back-calculated an admissible content of caesium in the residues. Taking into account attenuation, migration of caesium and potassium as well as their sorption in soil, it seems unlikely that the maximum regulatory values will be reached. However, in an initial period precautions are required and a monitoring programme should be undertaken in order to validate the conclusions. One should refrain from using ash in the immediate vicinity of surface waters or wells and the caesium content should be at most 2 kBq/kg. At the low activities required, 1 Bq/l, direct measurement of caesium-137 is not feasible as this is the background radiation level from naturally occurring potassium-40 ( $^{40}\text{K}$ ). Caesium may, however, be monitored in wells by using potassium as a surrogate, for which there is a limit of 12 mg/l in drinking water.

### 5.4 Bottom ash use in the subbase course

#### 5.4.1 Vändöra

The "Vändöra" project (Bendz et al., 2006a) is a demonstration of the use of combustion residues in road construction, where the long-term environmental and geotechnical properties of bottom ash are investigated. The roadway, a small cloverleaf turnabout at the entrance of Linköping's sewage treatment plant, was studied 16 years after construction. This is therefore a relatively unique project since there are very few roads constructed with combustion residues that have been studied so long after construction; previous results from this road were published in Flyhammar and Bendz (2006).

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<sup>7</sup> The Swedish Radiation Protection Authority (SSI) and the Swedish Nuclear Power Plant Inspectorate (SKI) have been merged into one authority per 1<sup>st</sup> of January, 2009. The ordinance SSMFS 2008:16 replaces the original SSI ordinance, SSIFS 2005:1.

Two different stretches of road were investigated, where one possessed a subbase layer consisting of 42 cm bottom ash from municipal waste combustion, and the other possessed a subbase layer with gravel, the traditional material. The aims of the project were to study 1) the cumulative effects of loading and age on the ash's geotechnical properties, 2) the cumulative effects of leaching after a longer period of ash use in a subbase layer, and 3) the conditions for the recycling of bottom ash from existing roads.

The road segments were studied with a number of geotechnical methods, including test loads with falling weight for determination of the road material's stiffness, and dynamic triaxial tests on undisturbed samples from the subbase layer in order to determine mechanical properties (e.g. stiffness, stability) of the material. In addition, the effect of water movement through the road body was investigated with leaching tests on bottom ash from the subbase layer and on underlying soils. A total of 187 samples were collected from the reference and test road segments.

The environmental investigation of metal mobility in the road has produced results that contribute to a better understanding of the geochemical processes occurring in bottom ash used in roads. Evidence for water intrusion into the subbase layer near the edges of the road is indicated by lower electrical conductivities in these peripheral areas (Figure 5-1). In addition to water movement, oxygen diffuses into the road body from the sides of the road not covered by asphalt. According to Bendz *et al.* (2006a), this results in an oxygen gradient through the bottom ash, producing regions of different oxidation – reduction (“redox”) potential in the subbase layer (i.e. different redox zones). The mobility of manganese, arsenic and chromium can be explained by this redox zonation since the mobility of these elements is dependent on redox potential. For example, manganese is less soluble under oxidizing conditions close to the edges of the subbase layer (see Figure 5-2), so concentrations of leachable manganese are lower here but higher toward the middle of the road where oxygen diffusion is limited and manganese may occur in a more soluble form.

In a subsequent study (Bendz *et al.*, 2009), water flow and geochemical transformations in the test road were simulated using the models LeachXS/Orchestra and Hydrus 2-D. Constant pH experiments (pH 2 – 12) were conducted with samples from the test road; these results were then simulated by assuming equilibrium with a variety of potential solid phases and atmospheric carbon dioxide, aqueous complexation with dissolved organic carbon, and allowing for surface complexation with hydrous ferric oxides. The simulation results suggested that macroelement concentrations in the test road leachates were controlled by equilibrium with such elements as calcite, laumontite, aluminium hydroxide, ferrihydrite, and dolomite. The interpretation of anion data was difficult because of the lack of thermodynamic data on the reaction of anions with common minerals in bottom ash, such as ettringite.

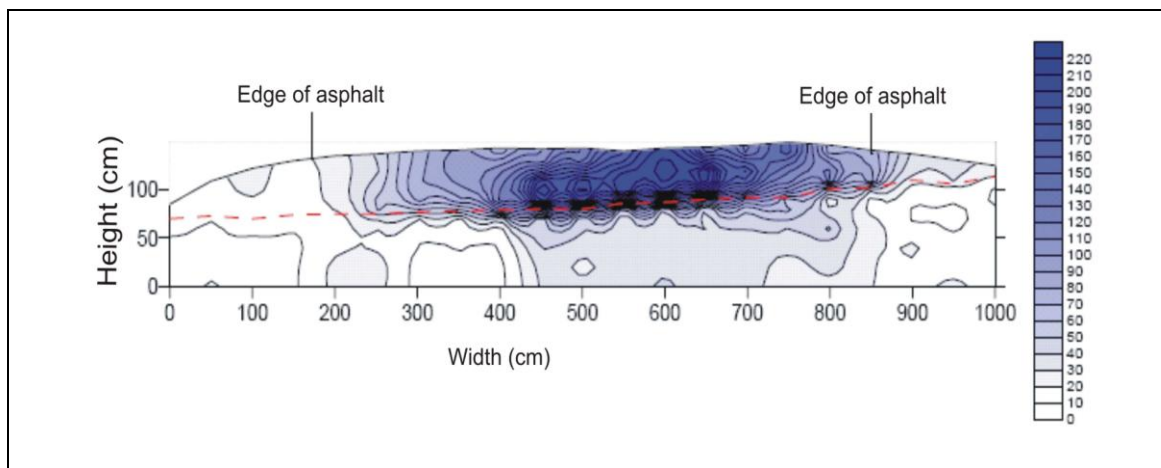


Figure 5-1. Example of variations in electrical conductivity (mS/m) in road body, after samples are suspended in water at a 1:5 mass/volume ratio. The dashed line marks the boundary between bottom ash and the underlying soil. Asfaltskant = edge of asphalt; bredd = width; höjd = height. From Bendz et al. (2006a).

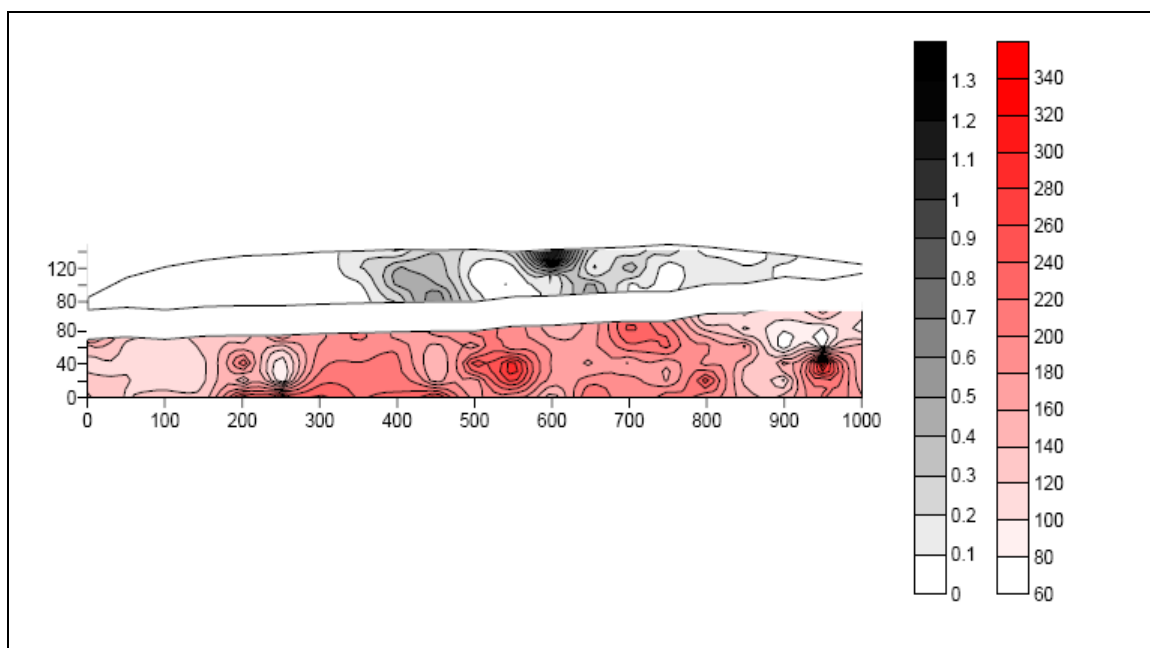


Figure 5-2 Distribution of leachable amounts (mg/kg) of manganese in the subbase (gray) at  $L/S=10$  and extractable amounts (mg/kg) in the subgrade layer (red). X and Y axes marked in centimetres. From Bendz et al. (2006a).

### 5.4.2 Dåvamyran

A test road was constructed in 2001 within the Dävaverket facilities in Umeå, using aged bottom ash in the sub-base layer (Arm, 2005). For comparison, a reference road was also established. The environmental impact of this structure was first reported in Lind et al. (2005), and later in an additional follow-up by Arm et al. (2008a), seven years after road construction. In the latter study, groundwater samples were collected close to the road, and leachate was collected from lysimeters placed under the road and in the road slope.

Leachate analyses from the lysimeters indicated that the accumulated leaching of calcium and sulphate increased during the study period. Concentrations of copper, chromium, chloride, sodium and TOC were initially greater than levels from the reference road, but these concentrations decreased greatly with time, with the exception of chromium, which exhibited less of a decrease (Figure 5-3). Similar behaviour was observed for a number of organic compounds. Leaching of organic compounds was initially greater from the test road during the first few years, but concentrations decreased to similar levels as leached from the reference road after 5 – 6 years. However, it should be noted that several compounds (fatty acids) were leached from the reference road at higher concentrations than from the test road with bottom ash.

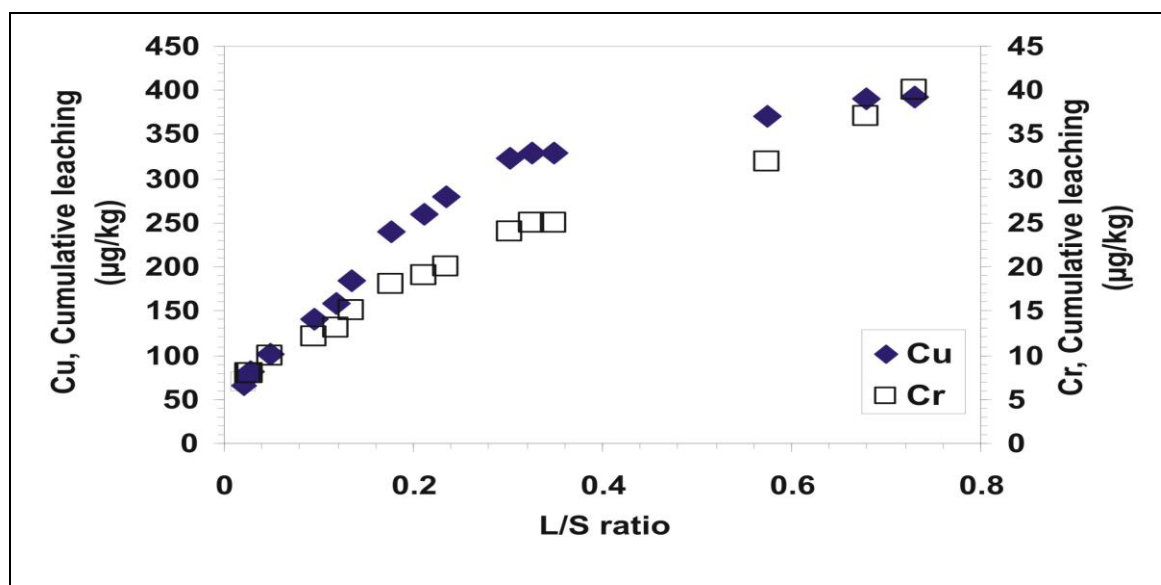


Figure 4 – 3. Calculated accumulated amount of leached copper (above) and chromium (below) collected in the road slope lysimeter under the Dåvamyran test road. Conditions for the calculations are discussed in Arm et al. (2008). Values for  $L/S < 0.4$  are from Lind et al. (2005). From Arm et al. (2008).

A comparison of results from previous laboratory leaching tests (Nilsson et al., 2002) with proposed guideline limits for the use of waste in construction (Naturvårdsverket, public consultation 2007/08), and with guideline limits for deposition as inert waste, indicated that it is primarily sulphate, chloride, copper, and chromium (initially) that are

critical compounds in terms of leaching to the environment. However, Arm et al. (2008a) indicated that leaching from the road body itself, composed of bottom ash as well as crushed rock, shows a significantly different pattern compared with leaching under laboratory conditions using bottom ash alone. Such aspects should be considered when the use of bottom ash in construction is assessed.

### **5.4.3 Svågertorp**

In Svågertorp near the city of Malmö, MSWI bottom ash was used in the subbase layer of a parking lot and connecting roadways (Flyhammar, 2009). The focus of the study was on the migration of mobile ions such as chloride and sulphate, because these substances can be used as early indicators of the migration of contaminants such as heavy metals. The results of a field study indicated a certain level of chloride and sulphate leaching to the underlying clay till. However, this formation has a low permeability, and Flyhammar (2009) concludes that the leaching of sulphate and chloride from the road should have no significant impact on groundwater quality in the bedrock aquifer that underlies the clay till.

## **5.5 Dust dispersal from roads**

As indicated in section 5.2 dust emissions from roads and subsequent deposition on local soils is a critical process in determining environmental guidelines for using combustion residues in roads. In response to the importance of this process, two projects (Gustafsson et al., 2006, 2009) address this issue. In Gustafsson et al. (2006), dust emissions were studied from a gravel road where the road base had been stabilized with fly ash, through measurements of deposition and particle concentration of PM<sub>10</sub> and PM<sub>2.5</sub>. The aim was to provide data for the assessment of the environmental effects of ash particle emissions. The study was conducted during one day when a truck passed over the road 165 times and a light duty vehicle 103 times. The road had a thin wearing course of gravel. Over the course of the day, the relative air humidity was 60-90 %, and wind speed was 2-4 m/s parallel to the road (Gustafsson et al., 2006).

The dust emissions were calculated to be 2.4 g/m<sup>2</sup> road surface. About 90 % of the emitted particles were deposited within 2-20 meters of the road edge. Many of the elements present in the deposited fly ash particles are easily leached from the material. The sampling and leaching methods used in this study (Gustafsson et al., 2006) resulted in L/S ratios of 10<sup>3</sup>-10<sup>4</sup> and the mobilization of readily soluble elements (e.g. sulphur, calcium, phosphorus and magnesium).

Dust emissions from the road and dispersion to the surroundings are affected by a number of factors where traffic characteristics and meteorology probably are the most important. The results from this study indicated that dust emissions from the studied road did not seem to pose an excessive environmental risk. However, the authors recognize that fly ash with greater concentrations of undesirable components may pose a greater risk.

The dispersion of ash particles was further investigated in Gustafsson et al. (2009), which 1) described appropriate models for calculating dust emission and dispersal during road construction, 2) evaluated a new method for investigating the significance of moisture content on dust dispersal, and 3) investigated ash properties that distinguish ash from other airborne particles. The results of the study show that currently available modelling tools cannot adequately describe the emissions from all stages of road construction, and that certain existing models lack key variables. The laboratory tests conducted within the scope of the project demonstrated that dusting from the ash samples was reduced significantly at moisture contents above ~15%. The study indicated that the morphological or mineralogical properties of ash particles are not adequate indicators of the presence of ash in particle samples, while the presence of sulphur, mercury, cadmium, and the Mg/Al ratio are better indicators. The most appropriate indicator elements depend, however, on the specific combustion residue used.

## **5.6 Bottom ash as filler material in district heating pipe culverts**

In addition to its use in road construction, bottom ash from circulating fluidized bed boilers has been investigated in order to determine if it is an appropriate material as filler in district heating pipe culverts (Pettersson et al., 2004). This study focused on both the geotechnical and leaching properties of the material. The geotechnical analyses show that the bottom ash fulfils the requirements for filler material used in district heating pipe culverts. In such applications, the bottom ash should be only lightly compacted so as to avoid crushing the material, which may result in an increased amount of fine material and thereby a greater leachability. Leaching tests on bottom ash, from both the combustion of biofuels and from the combustion of municipal and industrial waste, were compared. According to Pettersson et al. (2004), biofuel-based bottom ash had a lower content of many heavy metals. The leachate from fresh bottom ash contained a higher concentration of leachable components than aged ash, and the mobility of metals such as lead, copper, chromium and zinc was greater in the fresh ash. In contrast, aged bottom ash exhibited a greater leachability of antimony, molybdenum and sulphate ion,  $\text{SO}_4^{2-}$ .

## 6 Utilisation in cement and concrete

As presented previously in Chapter 4, fly ash and bottom ash from combustion of solid biofuels have been evaluated as a binding agent in road construction, primarily for stabilization of the base layer. Other applications of ash utilize this material as a replacement for cement, such as in panel stope mining and concrete construction below the ground surface. In general, the compressive strength of combustion residues in an application and the development of the compressive strength with time, are key parameters to be investigated when replacing cement with, for example, fly ash.

### 6.1 Use of fly ash in concrete

In an effort to identify potential concrete – related applications where combustion residues can replace concrete as a binder, Nordström and Thorsell (2003) have compiled regulations and standards that apply to concrete applications. The study indicated that fully developed regulations are only available for concrete used as a structural building material. In other applications, the regulations are formulated in such a fashion so that an opportunity is provided for the use of alternative materials. Although an original objective of the project was to identify acceptable variations in key parameters for which guidelines are available, but it was later shown that this was not possible since guidelines are rarely available.

Nordström and Thorsell (2003) identified several potential concrete applications where combustion residues (not including coal combustion residues) can be applied. Important factors influencing the application of specific combustion residues include type of combustion residue, availability, storage, local market, stability in fuel mixtures, and personal interest. Identified areas of use include use as the following:

- filler material in concrete with crushed aggregates and in self-compacting concrete;
- mining applications;
- road construction material with improved frost resistance;
- ground stabilisation with lime/cement columns and in cement-stabilised gravel;
- injection grouting for stabilisation of soil and landfills.

The authors noted that the application of combustion residues hinges on the general acceptance of these materials. Many concrete manufacturers were noted to be sceptical to the use of fly ash in concrete. However, an increased desire to reduce CO<sub>2</sub> emissions from, for example, concrete manufacturing, would favour the use of fly ash from solid biofuels as an amendment in concrete-based applications.

## 6.2 Fly ash as filler in concrete applications

As crushed rock has become an increasingly important replacement for natural gravel in concrete, there is an increased need for fine-grained binders and filler material that improve concrete's rheological properties during pumping. Fly ash is a material that has been previously used as a substitute for concrete, and Sundblom (2004) examined the potential of using Swedish fly ash as filler material in concrete and if this ash material fulfills the requirements for a filler. Cement with fly ash as filler may currently be used in the construction of houses and in industrial applications; however, since European standards have not yet been developed for fly ash applications in construction, fly ash may only be used in cases where there are no formal requirements on the water – cement ratio.

Sundblom (2004) investigated a number of fly ash materials from different fuel mixes and boiler types. Previous experience from Sydsten's concrete production indicated that the total amount of chloride in the concrete should not be higher than 0.1% and LOI (Loss on ignition) must be less than 10 %. The different ash analyses showed that the fly ash from fluidised bed furnaces and pulverized fuel furnaces fulfilled all the chemical requirements, but the fly ash from grate furnaces had difficulties to fulfil the LOI requirement. Results from tests of the rheological properties of different fly ash materials indicated an increased water consumption of fly ash from paper mills compared with the other ash from other fuel mixes (*i.e.* biofuel/domestic waste combustion), which was probably affected by the morphology of the ash grains.

The experiments (Sundblom, 2004) showed that all fly ash contributed to the final strength of the hardened concrete. For concrete mixtures containing an ash quantity of  $60 \text{ kg m}^{-3}$ , a loss in consistency was observed to varying degrees during the first hour after mixing. When the ash addition was reduced to  $30 \text{ kg m}^{-3}$ , only the paper mill fly ash mixture exhibited a loss in consistency. Sundblom (2004) indicated that the reasons why certain ash mixtures create a greater loss of consistency than others were not fully understood. Contrary to the laboratory results, a field demonstration in Malmö using fly ash from a pulverized peat-fired furnace as filler ( $40 \text{ kg m}^{-3}$ ) showed favourable results with no loss in consistency.

As a continuation to the study by Sundblom (2004), Sundblom (2006) sought to ascertain the components in a fly ash that contribute to the initial loss in consistency in a concrete containing a fly ash filler, and to thereby propose a method for evaluating various fly ash materials as filler material in concrete. This is necessary so as to be able to select an appropriate fly ash for a concrete application. Three different fly ash materials were selected for this study (Hallstavik paper mill, Mälarenergi Västerås, Vattenfall Uppsala). The tested materials (Sundblom, 2006) showed qualities that were desirable in filler materials, such as improving the stability and the rheological behaviour of the concrete.

The results from Sundblom (2006) indicated that 1) the compressive strength of concrete with a paper mill fly ash filler increases with free CaO content, which depends on the combustion temperature in the boiler. The use of fly ash as a filler generally leads to an increase in the compressive strength of the concrete (Figure 6-1); 2) there is a higher content of reactive SiO<sub>2</sub> and free CaO in fly ash from circulating fluidised bed (CFB) boilers compared with pulverized fuel fired boilers; 3) the higher water demand by paper mill fly ash (CFB boiler) may have been the consequence of a coarser grain size; 4) physical parameters such as grain shape and grain-size distribution influence the behaviour of the fly ash filler. Fly ash from pulverised fuel fired boilers exhibited similar rheological behaviour as traditional limestone filler.

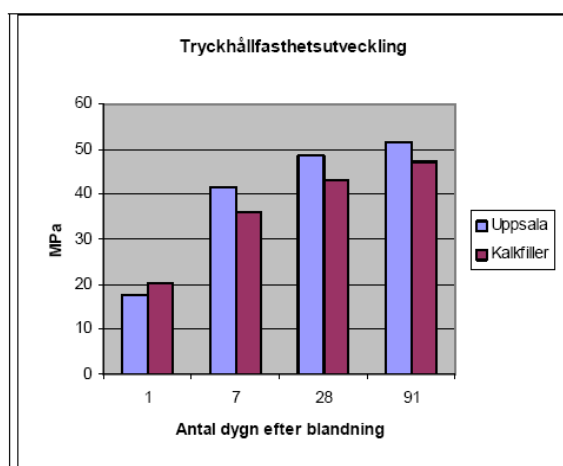


Figure 6-1. Compressive strength of concrete containing fly ash filler from Uppsala district heating plant, compared with concrete containing a traditional limestone filler (kalkfiller in Swedish). Development of compressive strength shown after 1, 7, 28, and 91 days.

Sundblom (2006) proposed a testing scheme (below) that would eventually lead to the certification of fly ash as filler in concrete, based on EN 450 in Sweden:

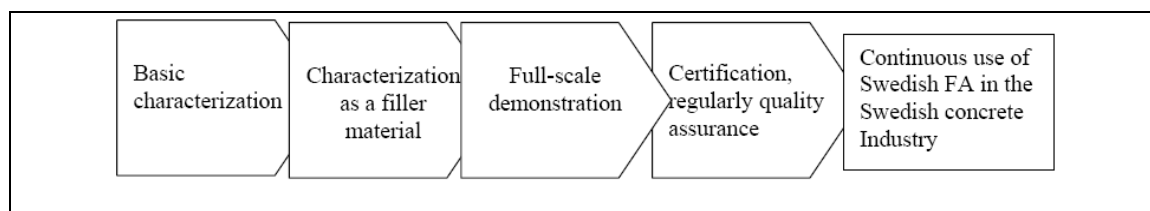


Figure 6-2. Proposal for a testing scheme to be used in the certification of fly ash as a filler.

According to the flowchart and the work by Sundblom (2006), certification is the next step in ensuring that fly ash partially replaces traditional fillers in concrete applications.

### 6.3 Panel stope mining

Laboratory tests and a full-scale demonstration at the Zinkgruvan mine (Nordström et al., 2004) have been used to evaluate the potential for replacing cement in panel stope mining with fly ash. As shown in Figure 5-1, panel stope mining implies the removal of large blocks or "stopes" of ore by boring into the rock mass from above and below and then detonating explosives in the many boreholes. When the stope is being removed, it is traditionally filled with a cement – tailings – water mixture for stabilization. For primary stopes (Figure 6-1), a compressive strength of several MPa is required, while a lower compressive strength (0.15 – 0.20 MPa) is sufficient to maintain water saturation and prevent hydraulic failure in secondary stopes (Figure 6-3). The study by Nordström et al. (2004) has investigated if fly ash is sufficiently reactive so as to be able to partially or completely replace the cement binder. Replacing cement with fly ash would lead to a greater profitability due to reduced binder costs.

Laboratory tests and a small-scale demonstration in the Zinkgruvan mine showed that it is possible to replace 50% of the cement with fly ash; this mixture provided sufficient compressive strength for its use in primary and secondary stopes. The fly ash used in this study was derived from the combustion of de-watered de-inked paper sludge. Ash with such a source material was chosen since a requirement for the application was that the ash should be available throughout the year (*cf.* fly ash from district heating that is primarily available during the winter months). Six month storage of the paper sludge fly ash led to considerable improvements in compressive strength, compared with results after 28 days of hardening.

An estimate, based on mining activity around 2003, indicated that 20,000 - 50,000 tons fly ash /year could be used in mines as a binder in conjunction with panel stope mining.

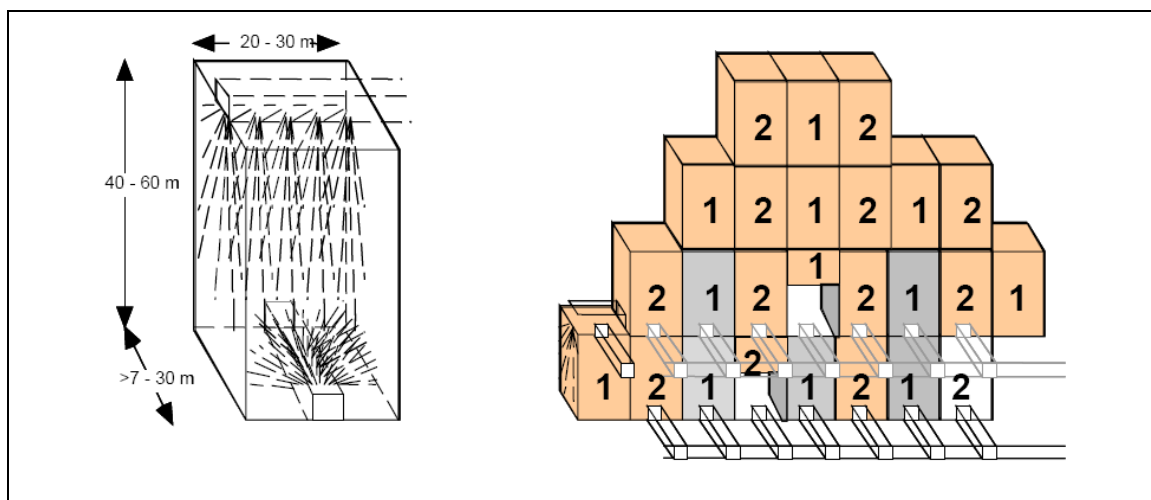


Figure 6-3. Example of the size of a stope (left) and an example of primary (1) and secondary stopes (2) (right). From Nordström et al. (2004).

## 7 Ash to forest soils

### 7.1 The need for compensating with ash

Sustainability of an intensive harvesting of biomass from forests, but also of agricultural residues and energy crops such as willow is a vital issue when tapping biomass as a renewable energy source. The base case in forestry being conventional harvesting of timber and pulpwood, the additional harvesting that calls for the word “intensive” is that of logging residues. The word commonly used is whole-tree-harvesting

Forest growth in itself has an acidifying effect on soil as vegetation picks up mineral nutrients, particularly cations from the soil, which form the incombustible ash of the biomass. In a virgin forest, these cations are returned to the soil when dead trees left in place decompose. In conventional forestry it is assumed that withdrawal of cations or nutrients (ash) in timber is compensated by input from weathering of minerals in the soil. Branches and other logging residues are left on the site and return their minerals to the soil.

Harvesting logging residues causes a considerable additional loss of mineral cations and other plant nutrients which loss in most cases is not fully compensated by weathering. Returning the combustion residues to the forest soil may compensate for this removal. These residues are to a large part the ash content of the wood fuel.

The acidifying effect of forestry is compounded by acid rain. Sulphur oxides and nitrogen oxides emitted by combustion of e.g. fossil fuels precipitate and lower the pH value of soils. Acid rain is at present much less of a problem than it was e.g. twenty years ago, as steps have been taken to reduce emissions, but it will take quite some time for soil to recover. One of the main effects is a lowering of the pH of surface waters. These have been limed for some time in order to counteract this increasing acidity. However, the effect of liming surface waters is rather short-lived. It is being thought that liming the soil in the catchment areas would provide a more stable and long-lived pH-raising effect as well as speed up the recovery. Liming only leads though to some deficiencies and wood ash provides a more balanced compensation with other mineral nutrients besides the base cations.

The questions of acidification, depletion of mineral nutrients and their effect on growth are imbedded in each other. A measure aiming at compensation or counteracting acidification is not necessarily a fertilizing measure even if it aims at maintaining long-term productivity.

The latest formal expression of this concern for sustainability and the condition of forest soils is the revised recommendations on whole tree harvesting and compensation with ash issued by the Swedish Forest Agency in 2008 (Skogsstyrelsen, 2008). They supersede an earlier text from (Skogsstyrelsen, 2001 and 2002), which in its turn replaced a first text on ash recycling issued in 1994 (Naturvårdsverket, 1994).

**Compensation, liming, fertilization**

Recycling ash or compensating with ash has been introduced as links in a technical cycle of mineral nutrients, *a.o.* base cations: those removed in the logging residues harvested are returned to the forest soil as ash. In popular terms, the purpose of this cycle is to prevent a possible depletion of nutrients several forest generations in the future. It may actually be more important to preserve the buffering capacity of the soil and mitigate downstream effects on surface waters.

The quantities of nutrients in the removed slash correspond to at most ca 2 t/ha of ash, and the maximum dose in the recommendations of the Swedish Forest Agency is 3 t/ha. The governing principle is balance between removal and restitution of both nutrients and trace elements.

“Liming with wood ash” is a measure aimed at counteracting acidification of forest soil, in particular in South-West Sweden. The role of both lime<sup>8</sup> and ash is to adjust the pH of soil, but wood ash contains other nutrients and compensates to some extent for the imbalance in nutrition that would have arisen with lime only. The aim is to return as soon as possible to healthy conditions.

Fertilizing implies a net addition of nutrients to forest soils in order to rectify a deficit in nutrition, which unchecked would have led to slower growth, the goal being a larger production. On mineral soil, nitrogen is very often the limiting nutrient and nitrogen is seldom found in ash. On drained peat land, several nutrients are often limiting; usually it is phosphorus and potassium. As wood or biomass ash contains most of these, it could be a suitable fertiliser.

The distinction between compensation and fertilization has an impact on the flow of potentially harmful trace elements. When compensating with ash, returning mineral nutrients to forest soils one returns also these trace elements. If there is a balance of nutrients, it is assumed that there will not be any net addition of trace elements. In other contexts such as fertilizing agricultural soils, the desired effect is a net addition of plant nutrients. In order to prevent a net addition of trace elements, *e.g.* cadmium and their accumulation in soils and crops requirements are placed on a maximum tolerable content in commercial fertilisers or in digested sludge.

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<sup>8</sup> In this context, lime is agricultural lime *i.e.* mainly limestone (calcium carbonate), not burnt lime or slaked lime.

These recommendations are based on results from comprehensive research Programmes starting in the 1980's. These research Programmes financed to a very large extent by government agencies as well as by industry since the 1980's have:

- Defined the treatment of ash preliminary to its spreading on forest soils in order to avoid short-term damage to vegetation and soil chemistry
- Verified that spreading ash in doses corresponding to the removal of nutrients does not harm the environment and has the desired effect on the acidity of the soil
- Verified that trace elements in wood, and consequently in ash, including undesirable elements as cadmium do not present a threat to forest ecology at compensatory doses
- Demonstrated acceptance by authorities of compensating measures and ash spreading activities in several parts of the country

For reviews of results from these Programmes, the interested reader is referred to two general surveys at a few years' interval:

- The Environmental Impact Assessment of whole-tree harvesting performed for the Swedish National Forestry Agency (Egnell et al. 1998, in Swedish)
- A synthesis of the most recent research Programmes, 1997-2004, (Dahlberg et al., 2006, also in Swedish, but a translation in English is being prepared)

## **7.2 Implementing compensation and the policy of the Ash Programme**

The first text on recycling of ash or compensation with ash was issued in 1994. With the usual delays between introduction of a new procedure and large-scale implementation, it can be expected that practice would take some years before picking up speed. However, implementation lags more than one would have expected. The percentage of clear-cuts with removal of logging residues that received a compensating dose of ash is increasing, but still quite low: in 2003, logging residues were extracted from ca 30 000 ha each year, but only 2 000 to 4 000 ha received compensation (Mellblom, 2006). This figure has improved since then, but it is still far from satisfactory.

A time delay in adoption of an innovation is natural, but the present delay has been deemed to reflect a reluctance from all involved. This may be related to several non-technical barriers:

- Information has not been made available or been assimilated
- A lack of understanding between ash producers (the heat or CHP plants), fuel providers and forest owners
- Regional differences in pre-conditions
- A lack of economic incentive for all stakeholders

**The project RecAsh**

In spite of a general positive attitude among all stakeholders, “Lack of Knowledge” and “Lack of plain regulation” were the main non-technical barriers to recycling wood ash to forest soils identified by the Swedish Forest Agency. An EU-LIFE project called RecAsh was initiated in order to overcome these information barriers.

The information gap has been addressed by seminars, hands-on demonstrations, presentation of case studies, a web site [www.recash.info](http://www.recash.info) as well as by completing an “Ash Recycling Handbook” (Emilsson, 2006).

**Differences between North and South Sweden**

There is a public acceptance of compensation with ash in the south, but there is little acceptance in the north.

Acid rain falls mainly over south Sweden, where it increases the total nitrogen load, the availability of nitrogen and the general acidification of soil and surface waters. Harvesting logging residues actually removes some of the nitrogen that would have become available, contributing thus to the recovery. At the same time, removing logging residues also removes cations in the ash and accelerates acidification.

Liming, with limestone, dolomite or ash, is a measure aimed at restoring the natural pH of forest soil and surface waters. Spreading ash or limestone to forest soils enhances nitrogen mineralisation, making it available to the established vegetation. Depending on conditions the net result could be a small additional growth on fertile soils or a temporary outflow of nitrogen.

Acid rain is not an issue in northern Sweden and mineral forest soils are poor. Under certain circumstances, spreading ash could yield a decrease in growth as ash appears to promote microbiological fixation of nitrogen. On the contrary, fertilizing with nitrogen is desirable as it would increase growth.

One of the perceived non-technical barriers is the lack of economic incentive. Treating ash, offering it and supplying it to land owners are costs for the ash producer. Spreading ash is a cost for the land owner. Land owners are not to expect an immediate increased growth, as would usually be expected after fertilisation. On the contrary, a short term growth reduction may be expected on many sites. Add to that uncertainty and there are many reasons for not doing anything.

The Ash Programme wished to prioritize actions leading to use of combustion residues, in this case the implementation of the desired recycling, complementing results from earlier research. An initial survey of the state-of-the-art, of achievements in recycling ash to forest soils and of remaining open issues was performed (Bjurström, 2002). This survey suggested that identifying these surfaces where ash does lead to increased growth and consequently to a tangible economic incentive, and prioritising these could contribute to a more rapid adoption of ash recycling. These areas may not necessarily be those in most need of compensation, but demonstrating profitable uses may accelerate general acceptance of compensation with ash.

### **7.3 Additional growth on mineral soils**

Almost no early field experiments with ash to forest soils can show a statistically significant effect on growth. There may be several reasons to that, one being of course that there is no effect, but others are that the ash doses are too small for effects to be detectable, the observation time too short, not enough data (too few studies on environmental impact did include growth parameters) etc. However, when summing up the results the general impression received is that together they tend to indicate an increased growth under certain conditions, and a reduced growth under other conditions.

There is a controversy as to what causes the increased growth. One school claims that ash, being a basic material, stimulates microbial mineralisation of soil organic matter, thereby mobilising nitrogen which becomes available to the plants. The other school claims that the mineral nutrients in ash, principally phosphorus, yield an increased growth.

According to Jacobson (Jacobson, 2003), spreading ash to a fertile plot could yield a growth increase of up to ca 10 %. Spreading to a less fertile mineral soil could actually decrease growth with up to ca 10 %. Individual figures are usually below the level of statistical significance.

Sikström revisited one of the field experiments initiated in the early 1990's (244 Åled) in order to quantify growth of the forest after a decade (Sikström and Jacobson, 2007). Data after five years were already available for this field with limestone, fertilisers and ash treatments. Although an increase of tree growth in the treated plots 15 to 25 % relatively to the untreated reference was documented, the difference is not statistically significant.

In a subsequent pilot study, Sikström *et al* have reviewed data from a number of experiments throughout the Nordic Countries, mostly liming experiments and a few ash spreading experiments (Sikström et al., 2009b). These plots were been treated 10 to 20 years earlier, and may yield information on long-term growth effects. The preliminary results are:

- Reduced growth on soils with low productivity
- No effect on soils with average productivity
- Increased growth on soils with high productivity

Overall these results confirm findings in the previous studies. Sikström et al could not find any dependence of growth response to lime or ash on C/N ratio in the humus layer. The study is now being deepened in a more comprehensive review.

The approach adopted by Thelin (Thelin, 2006) for assessing growth increase starts with an examination of nutrient status in the trees and particularly that of needles. He reviewed previously published as well as unpublished data from field experiments in beech and spruce forests in south-west Sweden, selecting 23 spruce test fields and 10 beech fields, including reference plots. Soil samples, soil water samples as well as samples of biomass were collected and tree growth was assessed.

Analysis of the results yields the following results for growth parameters:

- A growth increase of ca 14 % for ash-treated surfaces with spruce compared to reference untreated surfaces, significant within the study but difficult to compare with results from other studies as the experimental design did not include any replicates<sup>9</sup>
- This increase in growth was accompanied by an increase in phosphorus content in the spruce needles. There was no correlation between nitrogen content of the needles and relative growth
- On beech-planted surfaces, Thelin could not observe any difference in growth between treated and untreated surfaces
- The concentration of elements increased in spruce needles on ash-treated surfaces relatively to untreated surfaces, but it remains close to deficiency in *a.o.* nitrogen, phosphorus and potassium

Thelin interprets the results as a phosphorus limitation on growth that in several fields turns to a nitrogen limitation after treatment with ash. The phosphorus supply in ca 2 t/ha of ash is though not sufficient to bring the biomass to saturation with respect to phosphorus.

In 2008 and 2009, Thelin fertilised five experimental plots with either a conventional NPK fertiliser or with ash and nitrogen (Thelin, 2009). Some years will be required for the experiments to develop, but preliminary results after one year have been obtained. The observed increase of nitrogen and phosphorus concentration in leaves and needles in both cases will probably be followed by an increase in growth in the future.

## 7.4 Peat lands

In contrast to the mineral soils where growth is limited by nitrogen, on organic soils generally other mineral nutrients (phosphorus and potassium) are limiting growth. Besides increasing pH, ash is a needed amendment. Earlier experience with ash fertilization (Magnusson and Hånell, 1996; 2000) leads one to expect significant improvements in yield. However, one of the two newly afforested sites reviewed by

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<sup>9</sup> Growth increases of 15 to 25 % have also been found in several investigations with replicates, but they were not deemed significant by their authors

Magnusson and Hånell showed (2000) a clear difference in growth (established vegetation in fertilised plots, bare land in reference plots) and the other no effect from ash amendment (growth being equal in fertilised and non-fertilised plots).

From the ash producer's point of view, answers are needed to two questions. The first one is whether the surface of peat lands is large enough to justify investigations within the Ash Programme. The second one is what the environmental impact of amendment with ash is and if it is acceptable. Confirmation of the growth increase is also needed.

Peat lands are not only the present peat bogs, untouched or harvested lands, but also former peat lands that have been drained during a large part of the 20<sup>th</sup> century and afforested. Instead of ca 30 000 ha peat cutover areas to be restored or afforested in the future, the relevant total area is close to a million hectare of drained peat lands.

If drained and forested peat lands are to provide a showcase of profitability for spreading ash, harvesting should be done within the next fifteen to twenty years. The yield on these surfaces must have been reduced by a shortage of mineral nutrients<sup>10</sup>.

Hånell extracted from the National Forest Inventory 1997-2001 information on peat depth, drains, vegetation and age of forest throughout the country (Hånell, 2004, Hånell and Magnusson, 2005). Rejecting the unproductive sites and the shallow peat sites, selecting sites with drains in good condition and with vegetation dominated by "better shrubs" (*Vaccinium myrtillis*, *V. vitis idaea*, *Equisetum silvaticum*, and tall sedges) or "low sedge" (*Eriophorum vaginatum*, *Scirpus caespitosus*) areas and finally rejecting open areas, seedling stands and young forests he estimated the total area corresponding to this target to ca 190 000 ha, which figure consists of:

- 90 000 ha in the northernmost areas
- 30 000 ha in mid-Sweden
- 70 000 ha in southern Sweden

Of course, if a general benefit of increased growth is sought rather than a profit from harvesting this increased growth in the near future, a much larger proportion of the million hectares is suitable for fertilisation with wood or peat ash.

The peat cutovers that are to be restored are otherwise the peat lands that are first considered. In the near future, half of the 436 ha being closed may be considered for fertilisation with ash. In the coming decade, the total area may turn out to be 3 000 – 5 000 ha. It may also be interesting to fertilize them, in order to help the establishment of vegetation.

The potential profits from increased growth on peat lands must not be accompanied by negative environmental impacts of an ash amendment. In studies by Sikström et al the effect of applying ash on tree growth, water chemistry and emissions of greenhouse gases was investigated (Sikström et al., 2006). Two field experiments were initiated in

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<sup>10</sup> There are sites where there is almost no vegetation and spreading ash could help to establish trees on the site. However attractive this is, this will not yield a rapid economic return.

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2003 in southwest Sweden. The sites chosen were oligotrophic mires with forests dominated by Scots pine (*Pinus sylvestris* L.). The stands have been thinned and the soils drained at the end of the 1980's:

- 273 Anderstorp, a site with poor to fair fertility; a randomized block experiment with five treatments (one untreated reference, 3.3 t/ha or 6.6 t/ha of either self-hardened crushed ash<sup>11</sup> or pelletized ash) where growth and greenhouse gases were monitored
- 274 Bredaryd, an untreated reference and 3.1 t/ha of crushed ash, where water chemistry and greenhouse gases were monitored

The effect of treatments on growth was too small to be detected in 2005. This result could be expected considering that two growing seasons is too short a period for obtaining a response. However, Sikström et al. (2006) observed a statistically significant increase in potassium and boron concentration in needles, which increase might be linked with an increased growth.

In the beginning of the 1980's, another field experiment (168 Perstorp, a site with low fertility) was initiated in order to test different fertilising regimes including ash as one of seven treatments. Sikström revisited the plots in order to assess the effects 26 years later on growth (Sikström, 2008). There had been almost no growth on the reference, untreated plot. A dose of 2.5 t/ha ash or corresponding doses of phosphate and potassium chloride yielded equally large growths: 1.6 to 1.9 m<sup>3</sup> per hectare and year. Increased growth was seen as both taller trees and a larger number of trees. Adding nitrogen to the phosphorus or potassium fertilisers had no additional effect on growth, which indicates that the nitrogen status of this peat land was sufficient from the beginning. The results of chemical analysis of needles are that phosphorus and potassium concentration increased compared with the reference plots. In spite of the increase, these concentrations indicate severe nutrient deficiency, especially for phosphorus.

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<sup>11</sup> Crushed ash is ash mixed with water after extraction and left to harden or cement without any further treatment. After ageing, the ash agglomerates may be quite large and they must be crushed to a size distribution more suitable for spreading. Moistened ash that is pelletized before the binding reactions take place does usually not need to be crushed.



*Figure 7-1. Photographs of the 168 Perstorp site, when established (left picture), and on a surface treated with ash after 26 growing seasons (from Sikström et al., 2009)*

Two of these three sites, Perstorp and Anderstorp, and an additional one, 278 Skogaryd, a site with high fertility, were revisited in 2008 (Sikström et al., 2009). Skogaryd received 3.3 and 6.6 t/ha of crushed ash in 2006, i.e. as in Anderstorp. The results were:

- In Anderstorp the growth increased ca 10 % with 3.3 t/ha ash and ca 20 % with 6.6 t/ha over the five years after spreading ash.
- In Skogaryd no effect on growth could be observed two years after spreading ash, however this period of time is usually too short for such a potential effect to be detected.

Ash had no effect on carbon dioxide emissions in Anderstorp. In Skogaryd, the emissions were reduced. The emissions of methane were unaffected by ash in both cases. Nitrous oxide was mostly below the detection limit in Anderstorp, but decreased in Skogaryd, which decrease is assumed to be caused by the increase in soil pH.

The microbial biomass and its structure were unaffected by spreading ash. Ash reduced the net mineralization of nitrogen. Vegetation above ground was similarly mostly unaffected. In Perstorp, the least fertile site, coverage and diversity increased. In the most fertile site, Skogaryd, diversity increased and there was a slight increase of production in soil vegetation.

In run-off, there was a temporary peak in conductivity as well as concentration of several substances which lasted a few months after application of ash. Similar effects were observed in groundwater chemistry (Sikström et al., 2006). In Skogaryd, there was very little effect on water chemistry (Sikström et al., 2009).

One may conclude that amending a drained peat land with ash or PK-fertiliser may be beneficial to the establishment of a young forest. However, it is unknown how long the increase in growth will last.



Figure 7-2. Photographs of experimental sites: 273 Anderstorp in September 2005 (to the left) and 278 Skogaryd (to the right) in before ash was spread.

Peat extraction from the Flakmossen mire ended in 1945 and nothing was done until the beginning of the 1980's. In 1983 the field was drained and fertilised using 23 t/ha fly ash from a bark boiler and additional phosphorus fertiliser before it was afforested. The conditions were monitored in 1985 and 1987. Nilsson and Lundin collected peat samples in 2003 and analysed them for a number of elements, available phosphorus and base cations as well as pH (Nilsson and Lundin, 2008). The main results are as follows:

- The amendments with ash and phosphorus were enough to raise phosphorus and potassium concentrations in the peat to a level sufficient to sustain vegetation
- Amendment with ash raised the pH value temporarily as pH now is back to pre-treatment levels – this is attributed to the high sulphide content of the peat, which sulphides become oxidized when exposed to air, depressing the pH
- A large part of the phosphorus has been immobilised by iron and aluminium oxides in the peat and is not available to the trees
- Most of the potassium as well as manganese and magnesium have been leached away by run-off
- In a study that was not performed within the Ash Programme, Nilsson and Lundin concluded that the carbon stored in peat and vegetation (trees) had increased 10 % (Nilsson and Lundin, 2006). In the Ash Programme study, the carbon content of the peat was reported to have decreased 10 %

The 3 t/ha of ash allowed in the recommendations from the Swedish Forest Agency are not enough to raise pH in similar peat cutovers or to provide phosphorus and potassium for a continued growth during one generation.

## **7.5 Ash from agricultural residues to crop-bearing land**

As is the case for forest lands, exploiting the agricultural soils to produce biomass depletes the soil of mineral nutrients. This is today compensated for by using fertilisers. Returning ash from e.g. straw to the fields will also compensate for this depletion as well as reduce the use of mineral fertilisers, the resources of which are finite. Wheat straw ash is primarily a potassium fertiliser, with some liming and some phosphorus effect.

If all wheat straw from a field is used as fuel, ca 250 kg ash will be produced per hectare (Ottosson et al., 2009). This is rather too small a quantity to be spread with present machinery. Several solutions were explored by Ottosson et al in view of the impact of a large dose of potassium on run-off and the ability of vegetation to take up the nutrients made available. A bi-annual spreading of bottom ash is at present the most interesting policy, but fly ash would then be discarded. The actual uptake of potassium and phosphorus needs to be determined more accurately before a firm conclusion can be reached about a recycling policy. Cadmium is not a problem from a regional point of view, as what has been carried away with straw is returned as ash.

## 8 Material knowledge

### 8.1 Unburned Carbon and organic matter

If the furnace and the flue gas treatment are run efficiently, the unburned but combustible content of residues is small. It has been practical to bundle the imperfections of the process, *i.e.* unburned carbon in the predominantly mineral materials, into any of the measures used to describe this carbon in order to focus on the inorganic part of ash. However, relying on lumped parameters has caused difficulties in discussions on what actually caused unsatisfactory performance in various contexts: cement-like binding properties, landfill, leaching...

Furthermore, increased attention to organic trace components, *a.o.* PCDD/F's and PAH's<sup>12</sup>, has made it necessary to penetrate in more detail what unburned carbon actually consists of.

Knowledge what the methods of analysis in daily use actually yield could be retrieved from literature (see e.g. Vosteen and Beyer, 2000a and 2000b), but there was a need to demonstrate it practically for the types of ash found in Sweden. Twenty-one samples representing various types of furnaces, different fuels, different types of residues and different handling histories were subjected to the standard methods of analysis for unburned carbon (Bjurström and Suér, 2005), principally Loss on Ignition (LOI) at several temperatures and instrumental TOC determination. To separate the various reactions contributing to LOI the samples were also studied using a TGA instrument fitted with a DTA unit for heat flows and a mass spectrometer to analyse the substances released by the sample during heating.

The results confirmed that LOI methods yield more than unburned carbon: hydration water from *e.g.* hydrates or hydroxides at comparatively low temperatures, inorganic carbonates or semi-volatile salts such as potassium chloride at the higher temperatures. If combustible but unburned carbon is desired, then TOC is the method of choice. However, TOC shouldn't be understood as Total Organic Carbon, as it often is, but rather as Total Oxidizable Carbon as this carbon is elemental.

Unless the combustion is really inefficient, the organic compounds in ashes may be expected to be trace components. Present knowledge was reviewed shortly (Bjurström, 2006): PCDD/F, PCB and PAH are well-known, but almost nothing is known about other compounds, of unknown persistence and unknown adverse health and environment impact, that could be present.

In order to gain experimental knowledge of the organic content of combustion residues, three ash samples from woody fuels were submitted to the semi-quantitative<sup>13</sup> screening

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<sup>12</sup> PCDD/F, PolyChlorinated DibenzoDioxins and Furans, PCB, PolyChlorinated Biphenyls and PAH, Polycyclic Aromatic Hydrocarbons

of semi-volatile compounds performed on MSWI bottom ash from the Dåva experimental field (Lind et al., 2005). Commercially available services were used, rather than research laboratories. This analysis involved (Larsson et al., 2009):

- Preparation of the sample (milling in liquid nitrogen)
- Extraction of non-polar compounds using dichloromethane followed by separation in a gas chromatograph and detection, identification and semi-quantification using a mass spectrometer
- Acidification of the samples, extraction of polar compounds using ether, silylation in order to lower their boiling point followed by separation in a gas chromatograph and detection, identification and semi-quantification using a mass spectrometer

The chromatograms obtained show a small number of well-separated peaks: at most 45 compounds could be identified with certainty, concentrations upwards of 0.1 mg/kg, and approximately as many more tentatively in concentrations of the order of 0.01 mg/kg. While some of the substances could be expected from a chemical point of view (*e.g.* PAH's), the presence of others are most certainly due to contamination during sample extraction, handling and analysis. Identification of a substance is also uncertain at these low concentrations. On the other hand, aromatic internal standards in the analysis of silylated samples were only partially recovered. Substances which are usually found in smoke or ash from small-scale combustion of wood were absent.

The conclusion reached is that the organic content of a residue from large-scale combustion is quite low, even if the TOC is 10 % or more. There is no doubt that research methods may yield more precise and complete information, but there is no reason to expect fundamentally different results. By all means, a better attempt at determining the content of organic substances in ash should be made, but attention should be given to all possible paths of contamination of the samples for the results to be relevant.

The non-volatile organic content of ash consists of larger molecular aggregates that usually are described as humic substances. They are quite stable but will be found in leachates and are reported as DOC, Dissolved Organic Carbon. The total content of Dissolved Organic Carbon (DOC) does not change with maturation time, but its characteristics do: from a basic hydrophilic and neutral character it becomes similar to the fulvic acid found in natural waters (hydrophobic and hydrophilic acids) (Johansson, 2003).

Leachates from two bottom ashes matured for four months and natural soil water used as reference were investigated using the conventional analytic methods for humus (Olsson et al., 2006a). Fractionation yielded large differences: whereas soil water is dominated by hydrophobic acids and hydrophilic acids, leachates from the ashes have a larger proportion of neutrals and much fewer hydrophobic acids. Fitting the

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<sup>13</sup> The screening is semi-quantitative: internal standards were not used because it is impossible to (i) predict beforehand which substances are actually present and (ii) to add an internal standard for each detected substance

characteristics to available models for humic substances indicated that phenolic groups were much fewer than one would expect in natural humus.

The trigger for this investigation of DOC and humic substances was actually leaching of copper. By binding copper (coordination with acidic groups or adsorption), they contribute to a high copper content in the leachate. At times, this is a problem: limit value in the landfilling directive are exceeded. In this regard, the conclusion of the investigation was that these high-molecular compounds do indeed contribute to leaching of copper, but the actual concentration in leachates is also influenced by kinetics of dissolution of copper-containing phases as well as adsorption of copper on ash surfaces (Olsson et al, 2006a; 2007; 2009).

## 8.2 Properties of inorganic components

Projects in the Ash Programme have focused on a few specific questions raised in findings from other projects or questions that in the meantime gained actuality, *i.e.*:

- Hydrogen evolution caused by the presence of metallic aluminium
- Leaching of antimony
- Availability of arsenic
- Speciation of zinc in ashes
- Immobilization of elements when ash is submerged

### 8.2.1 Hydrogen evolution

A series of explosions at an underground storage site in January 2004 was attributed to hydrogen that evolved from alkaline APC residues from a recently commissioned fluidised bed combustor using a new waste fuel when these came into contact with the pool of water at the bottom of the cavern and the metallic aluminium that they contained started to corrode. The appropriate measures have since then been taken at that plant, but the accident and previous events at other plants pushed the topic of metallic aluminium to the forefront.

A set of 31 ashes from 14 combustion plants representing different fuels, different types of ashes and different types of combustion plants were gathered (Arm et al., 2006; Arm and Lindeberg, 2006)). Their potential for evolution of hydrogen was tested and the presence of metallic aluminium in some of the largest emitters was confirmed by SEM.

Ashes from solid biofuels showed very little potential for hydrogen evolution and those from wastes had the highest potential. Waste fuels have high aluminium content, but this content alone does not determine the actual emission, as aluminium can be contributed by bed materials in fluidised bed furnaces, soil, etc... In grate furnaces the bottom ash has the largest hydrogen evolution potential but in fluidised bed furnaces any of the fly ashes could be the largest emitter. Although this obviously reflects the particle size distribution of the fuel, the thin aluminium layer in light fuel fractions such as liquid cartons could be expected to be buoyant and found in a fly ash from a grate furnace. Leaving the ash to mature in air reduces the potential for hydrogen evolution.

The presence of metallic aluminium in combustion residues cannot be avoided by end-of-pipe measures alone if one wants to keep full reactivity of the ash in a utilisation. Means to burn out the small metal flakes were studied in a follow-up laboratory investigation external to the Ash Program (Backman et al., 2007). However, this attempt was not successful: metallic aluminium flakes are difficult to combust. In the first investigation, it was reported that the aluminium particles had not even melted although temperatures in the furnace largely exceed the melting point of the metal.

### **8.2.2 Antimony**

Leachate content of antimony turned out to be a key question in an early project (Pettersson et al., 2004). When bottom ashes from fluidised bed furnaces, actually spent bed material, were subjected to leaching tests antimony was the element, the concentration of which often exceeded the limit values in the criteria available at that time<sup>14</sup>. This was somewhat surprising as antimony had until then not been considered a priority pollutant and was therefore not included in the standard set of elements analysed. A similar experience was also made in another project (Bjurström et al., 2004). Antimony had the potential of being troublesome as it analogously to arsenic forms oxyanions that are difficult to stabilise.

To map the actual extent of the antimony problem, the 31 ash samples gathered for the hydrogen evolution project (representative of the Swedish combustion plants and fuels) were run through leaching tests (Bäckström, 2006). The mobility of antimony was deemed to be high when the guideline leachate values for landfilling waste were exceeded, which they were in the case of eight bottom ashes and three fly ashes (taken here as any ash from boiler ash to APC residue). Analysis of the results together with information on fuels and ash composition led to the following conclusions:

- Antimony is found predominantly in waste fuels, and consequently in their combustion residues at concentrations exceeding 100 mg/kg in fly ashes and between 50 and 100 mg/kg in bottom ashes
- Most of the antimony (75 %) is found in the fly ashes for a given combustion plant
- Leaching of antimony is a problem in bottom ashes, but not in fly ashes
- A minor part of the antimony is released in the standard leaching tests and most of it seems to be unavailable
- Antimony does not behave as an oxyanion, which would indicate that it is either adsorbed on minerals or bound in solid phases in the ash

### **8.2.3 Arsenic**

When a proposal for environmental guidelines on utilisation of ashes in civil engineering works had been developed, the values obtained were compared with the data on composition and leaching properties available for Swedish ashes (Bendz et al., 2006c). The parameter critical for human health turned out to be arsenic concentration.

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<sup>14</sup> The criteria for reception of waste at landfills, 2003/33/EC

The mechanisms that are critical for exposure are traffic dust settling on vegetables that are grown 20 m from the road as well as accidental intake when a child eats ash particles.

In all calculations of limit values, the availability for humans was taken as 100 %, which is a worst case approach. In order to test this assumption, the bioavailability of arsenic, antimony and several metallic elements was investigated using an in vitro method developed by RIVM<sup>15</sup> (Carlsson et al., 2008). The three leaching solutions simulate saliva, gastric juices and liquids in the gallbladder and intestine. Five ash samples were collected from combustion plants and three samples taken from actual field experiments.

The bioavailability of arsenic was the highest among the elements investigated: from ca 50 % to 100 % depending on the type of ash. Total availability is then a reasonable assumption. On the other hand, bioavailability of antimony was very low, mostly below 20 %. The availability of antimony is anyhow quite low in the conventional leaching tests, below 1 % at L/S=10 (Bäckström, 2006). Although arsenic and antimony are chemically very similar, their behaviour is quite different in this respect.

The bioavailability of the metallic elements decreases from cadmium (ca 60 %) through zinc (variable, but mostly 50 to 60 %), lead (mostly ca 50 %), copper (variable from 20 % to 70 %) and nickel (variable from 10 % to 50 %) to chromium (10 to 30 %) (Carlsson et al, 2008). The particle size doesn't have any major importance, although the bioavailability for arsenic, copper and chromium is slightly lower in the larger fractions. The type of ash (bottom or fly) seems to have an influence, but the trend is not the same for all elements.

### **8.2.4 Zinc**

As oxides are the main constituents of ash, it is natural to assume that the trace element zinc will also be present as an oxide. However, this tentative speciation lands the user as well as the ash producer in trouble. Zinc oxide is classified as ecotoxic and from a regulatory point of view, with the zinc concentrations that are usually found in various fuels, including uncontaminated wood, ash will very often be classified as ecotoxic.

Equilibrium calculations using *a.o.* leaching data may suggest alternative speciations such as a zinc silicate (willemite) or a zinc ferrite (franklinite) (Sjöblom, 2007; 2009b), but there is no better proof than direct experimental evidence. Zinc is not present in concentrations large enough, or in crystallites large enough, for an identification of phases using X-ray diffraction. Therefore, the EXAFS<sup>16</sup> technique was used in order to attempt calculation of the binding distances for the nearest neighbours of zinc atoms (Steenari and Norén, 2008). When these are known, they can be compared with those in relevant crystal structures.

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<sup>16</sup> EXAFS, Extended X-ray Absorption Fine Structure spectroscopy

Twenty-four ash samples representing seven combustion plants were gathered from ongoing research activities as well as collected from the plants (Steenari and Norén, 2008). They represent combustion of solid woody biofuels in grate furnaces as well as fluidised bed furnaces as well as of waste in grate furnaces and in a BFB. Reference EXAFS-spectra were recorded for relevant compounds: zinc oxide, zinc sulphide, zinc chloride, zinc aluminate ( $ZnAl_2O_4$ ) and franklinite ( $ZnFeO_4$ ).

Analysis of the results and modelling yielded zinc silicate, zinc aluminate and zinc hydroxide as the most probable speciation for zinc in most ashes. The chemical bond distances suggested in a few cases the presence of zinc chloride (fly ashes) or zinc sulphide (also fly ashes) in minor amounts. Zinc oxide or a spinel structure such as franklinite could not always be eliminated from possible structures. This is partly due to the noise level and to the low zinc concentration that makes it difficult to see secondary peaks. It may be noted that in some cases, e.g. fly ashes from waste incineration, zinc may be present as a substituent for other ions in a silicate structure. In other ashes, analysis suggested that the zinc compound is adsorbed on surfaces. This agrees with the observation that the zinc concentration in leachates decreases with increasing contact times: zinc initially dissolved is re-adsorbed on the available mineral surfaces (Steenari and Norén, 2008).

### **8.2.5 Submerged combustion residues**

Combustion residues are being utilised to fill underground caverns which were used to store fuel oil. In this case, the residues are submerged as these caverns mostly are below the water table level and ground water seeps in. There is a concern about the impact on environment: will dissolved salts migrate in the groundwater, especially objectionable trace elements?

Previous experience is that the pH value is initially high but falls rather rapidly and that the concentration of lead in the supernatant water falls by a factor of 20 to 50 within a month. The hypothesis is that carbonation of the mixture immobilises lead and lowers the pH. The process was investigated for three combustion residues in 1 m<sup>3</sup> plastic vats, two per combustion residue (von Brömssen et al., 2009). These residues were two APC residues from waste incineration and one fly ash from a bark boiler at a pulp and paper mill (recovered wood, DIP-sludge and water treatment sludge as fuels). The vats were filled with water to half the available volume and then a mixture of residue and water was added to full volume. One of the vats for a residue was periodically topped with water to replace evaporated water. In the other vat, the water phase was siphoned off periodically and replaced with fresh water.

The carbonate equilibrium was confirmed as the main factor for the lowering of the pH for both APC residues from waste incineration, although the availability of carbon dioxide is quite low, in the vats as in the caverns. The concentration of very soluble compounds such as potassium chloride decreases through dilution, but salts in the residue do not dissolve readily which is probably caused by the low permeability of the hardened residue. Calcium and sulphate concentration are determined by the solubility

of gypsum. The concentration of a significant number of elements in the solution is limited by secondary precipitates such as sulphates, hydroxides and carbonates.

### **8.2.6 Ecotoxicity H14**

When classifying a waste as hazardous or not hazardous according to the Directive on Hazardous Waste<sup>17</sup>, one of the fourteen criteria to be used is ecotoxicity, H14<sup>18</sup>. While legislation on chemicals provides a method to calculate whether a chemical compound is ecotoxic, it is not certain that this will adequately reflect the actual properties of complex materials such as combustion residues.

In the case of a solid material such as ash, it is leached before its eluate is tested on several aquatic organisms. The standard test batteries proposed in EU work utilise fresh water organisms. There is a risk that these will react primarily to the salinity of the eluates and less to potentially ecotoxic pollutants. Stiernström et al have tested a percolation method to prepare a water extract for ecotoxicity tests and evaluated a battery of tests (Stiernström et al, 2009). The species represent different trophic levels: the bacterium *Vibrio fisheri* (Microtox®), the micro algae *Pseudokirchneriella subcapitata* (growth development), the copepod *Nitocra spinipes* (larval development) and zebra fish *Danio rerio* (embryo toxicity).

All the test organisms responded with distinct concentration-responses when exposed to the leachates from different combustion residues. None of the tests was always the most sensitive to all leachates. The ecotoxicological effects couldn't be predicted from analysis of the samples and from literature data and the calculation method in the legislation on chemicals overestimates somewhat the ecotoxicity. False positive results were obtained in several cases as the toxicity could be attributed to aluminium, calcium, potassium, nitrite or ammonium.

An issue that needs to be taken into account is the maturation of samples before they are submitted to extraction and tests, as well as their preparation. The percolation leaching method is appropriate as the samples will not be affected as much by particulate as in a shaking extraction. The study has focused on inorganic substances and needs to be developed for organic substances.

## **8.3 References for leaching**

When assessing the use of ashes in civil engineering works, what should the environmental properties of ashes be compared with? In the absence of criteria specific to ashes or other waste materials in the uses considered, one takes whatever is available. Mostly, these are the leaching criteria for acceptance of waste at landfills (2003/33/EC) or before these were published, the Swedish concentration criteria for polluted soil

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<sup>17</sup> This Directive, 91/689/EEC, has been implemented together with other EU texts, notably decision 2000/532/EC establishing a list of wastes, in the Swedish Waste Ordinance 2001:1063.

<sup>18</sup> A fifteenth hazard, sensitizing, has been included as H13 in the recent revision, the Waste Framework Directive 2008/98/EC

(Elert et al., 1996). The Ash Programme has strived to develop a proposal for waste materials, which is described elsewhere.

Clearly, these criteria provide an upper limit value in discussions. It is though difficult to assess the natural contribution from surrounding materials or the manoeuvring margins that choosing between various materials would afford a constructor. In order to provide a reference, the leaching properties of natural aggregates were determined for both rock minerals (23 samples) and tills (15 samples) representative of conditions throughout Sweden using standard methods for total concentration and for leaching tests (Ekvall et al., 2006).

Work in this project produced a mass of numerical values, the vast majority of which were below the level of quantification (LOQ) for leaching. The obvious conclusion is that natural aggregates in Sweden leach very little. Only in rare cases the limit value was exceeded for an element in a set of criteria. Leaching of fluoride exceeded the limit values for inert waste in the waste acceptance criteria as well as for potable water in a few samples. In the case of mercury, LOQ is slightly higher than the limit values for inert waste or potable water.

#### **8.4 Quality assurance**

The quality assurance system proposed by Hartlén and Grönholm (2002) and its documents for bottom ash to civil works is an example of the extent of documentation that such a system should result in. The basis for the environmental data used in their report is an investigation by SYSAV on the maturation in heaps of its MSW IBA (Flyhammar et al., 2004a; 2004b).

These data were reviewed for the Ash Programme (Flyhammar, 2006), yielding the following observations.

- The low mobility of cadmium, chromium, nickel, lead and zinc makes them less critical than copper, molybdenum, sulphate and chloride on which the emphasis should be laid
- There is not any obvious connection between the different parameters – leachability, availability and pH
- The pH value, initially proposed as single key parameter when determining the end of the maturation period, is not enough as it does not alone reflect all changes in parameters
- Furthermore, it seems to guarantee that all IBA will reach a pH-value of 9 after six months of storage. As surface and heart of a heap are not equally exposed to weather, maturation is unequal
- Ageing does not reduce leaching of sulphates or antimony

Ash is inhomogeneous and its composition as well as its properties depends on many factors: fuel, contaminants and combustion characteristics which all vary with time<sup>19</sup>. On the scale of several thousand tons, variability from day to day (whatever may cause it) is evened out by the length of the period needed to produce the quantity required for e.g. a parking lot. The number of samples to be taken and mixed in order to have a representative collective sample increases with the volume, but this is primarily a sampling problem, not a problem of variability of properties.

Regulations may, however, require that the tests of properties are performed for smaller volumes. The recommendations from the Swedish Forest Agency (Skogsstyrelsen, 2001) state a test at least each 250 to 500 tons of ash ready to spread on forest soils. If there is a possibility that <sup>137</sup>Cs radioactivity exceeds a limit value of 10 kBq/t<sup>20</sup>, samples must be taken every 30 tons (Strålsäkerhetsmyndigheten, 2008).

The number of properties that should be determined in these texts is quite small. If one adopts the same frequency to ash for geotechnical uses, or day-to-day monitoring, a comprehensive test procedure would be quite expensive. The variability of bottom ash from combustion of recovered wood with shredded tyres as additional fuel was investigated in one project (Bjurström et al., 2004) with the intention of finding a suitable sub-set of parameters that could be used for monitoring ash fresh out of the furnace. The full set of parameters still has to be determined for the large volumes of matured ash in e.g. terracing.

The operation of the vibrating grate furnace was monitored for a couple of months, with one small sub-set of properties being determined daily for working days and a larger sub-set for weekly samples. The variability was actually rather small, without any obvious trends. The presence of outliers in composition data points to sampling problems at this frequency. As in Flyhammar's study (2006), there wasn't any generally valid correlation between the different parameters (composition, leachability, loss on ignition).

The loss-on-ignition or LOI<sub>550</sub> was rather high, between 11.5 and 36.5 %, yielding a rather brittle bottom ash. It was easily crushed during handling which affected its particle size distribution and geotechnical properties. Stiffness decreased with increasing LOI. Because of the poor quality of this ash it was decided to abandon testing after ageing.

Fuels are more often than not mixed in solid biomass furnaces, the reasons being *a.o.* local availability and desire not to depend on a single source. A sulphur-rich additional fuel such as shredded tires in the plant above is more an additive in order to mitigate corrosion in superheaters than a primary fuel. Generally, "clean" fuels will produce "clean" ash with respect to suggested limit values for certain potentially harmful

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<sup>19</sup> Other factors that influence ash properties are furnace design, the location where an ash stream is extracted from the furnace and the flue gas system, but these are system factors that do not vary in the same manner.

<sup>20</sup> For ash that is spread on agricultural land, the upper limit for <sup>137</sup>Cs is 500 Bq/t.

substances in ash, e.g. heavy metals. Recovered wood is not a clean fuel in this sense, but it is cheaper than logging residues. An important question is to what extent clean and dirty fuels may be mixed and still produce an ash clean enough for *e.g.* spreading in forest soils.

The conclusions from a series of experiments in a 24 MW<sub>th</sub> fluidised bed furnace is that only small amounts of dirty fuel may be mixed with clean fuel if the combustion residues are to be clean (Bjurström and Wikman, 2005). Recovered wood contains questionable elements such as heavy metals in much larger concentrations than clean logging residues and a small proportion of recovered wood is enough to ruin the ash.

If one abstains from mixing on a continuous basis but combusts recovered wood and logging residues after each other, one should take into account the reservoir that the bed of the furnace represents. When these fuels are switched, the composition of the combustion residues will not relax instantly to a composition corresponding to the new fuel. The rate at which *e.g.* content of arsenic relaxes is approximately the same as the rate of replacement of bed sand to compensate for losses. These losses are caused by bleeding over-large particles as bottom ash, or attrition of particles and extraction as boiler ash or fly ash or APC residue.

## 9 Improving ash

For a producer of a combustion residue to consider improvements or treatments there must be an utilisation for which it is profitable, or necessary, to modify the properties of the material. As long as there isn't any economic incentive for improvements, the policy of the Ash Programme is to first consider uses where combustion residues can be used with minimal treatment.

Such minimal treatment may consist of *e.g.* removing metallic objects, screening to a desired particle size distribution and ageing the residues. Agglomeration and ageing is required for wood ash before it is spread on forest soils. Expensive advanced treatment, *e.g.* vitrifying or chemical treatments such as extraction, is not top priority.

Improving usually implies a target that should be reached. Today, the acceptance criteria for waste being landfilled or the criteria for contaminated soil are used in permitting. While these criteria are often quantitative enough in themselves, they were not devised for this purpose and their use for assessments of waste utilization has been criticised. The target for the improvements may be a moving one.

APC residues from waste incineration need to be stabilised if one wants to landfill them: it is not certain at all that they will clear the acceptance criteria for leaching in the EU directive nr/nr/nr. However, there is for the time being an utilisation (neutralisation of vast quantities of waste acid and backfilling a quarry) where the content and availability of heavy metals is not a problem. A further reason for the Ash Programme not to prioritise such projects is that the aim of the Ash Programme is to develop uses and not to facilitate landfilling.

Consequently, projects carried out within the Ash Programme aim at:

- Addressing imperfections in present treatments
- Taking advantage of opportunities for low-cost improvements
- Gathering a basis for well-informed choices on treatment

### 9.1 Wood ash to forest soil

Before they are spread to forest soils, wood ash must be stabilised: combustion residues are strongly alkaline and could damage undergrowth. This can usually be achieved by agglomerating the residues and letting them age or mature. This takes care of the highly basic oxides that are converted into more stable species, the pH of the residue being lowered in the process. Calcium oxide may react with water, carbon dioxide in air, and other components in the ash to form a variety of compounds such as hydroxide, carbonate, calcium aluminium silicate hydrates (cement reactions) or ettringite.

However, agglomeration and ageing alone do not always lead to a product that is as stable as desired. Some wood ashes are known to be problematic materials. Organic content is often claimed to be the reason, but this is not very convincing as some of the problematic ashes are well burned-out. A case in hand is the fly ash from the Falun

cogeneration plant with a fluidized bed furnace: the ash does not aggregate into large chunks as others do and tends to crumble.

If one considers the elements that enter into cementitious compositions (calcium, silicon, aluminium, iron) clean wood ash consists mainly of calcium, with small amounts of other elements. Wood ash is usually diluted in the combustion residues by two materials: soil material picked up by logging residues when these are harvested or handled; and in FB furnaces the bed sand. The silica content in both materials is usually very high. With small additions, the composition of the combustion residue may become close to that of Portland cement: ca 75 % calcium oxide (CaO), 20 % silica (SiO<sub>2</sub>) and 5 % alumina (Al<sub>2</sub>O<sub>3</sub>). Other compositions also show cementitious properties. However, the right composition is not enough for a residue to have binding properties, the mineral composition must be right and the components must be reactive.

Two projects have investigated means to stabilize wood ashes, starting with different approaches, and came to similar results.

In the first project (Mahmoudkhani and Theliander, 2004; Mahmoudkhani et al., 2007), samples of fly ash were collected from two combustion plants, one being a grate-fired furnace and the other a circulating fluidised bed furnace. The fuel is woody biomass in both cases: wood chips and bark being the main fractions. The main difference in ash composition is the presence of bed material, mainly silica from the fluidisation sand, in the ash from the CFB plant. Three methods were tested: thermal treatment, adding potassium silicate and pelletizing. The products were assessed by determining their mechanical strength, pore structure and leaching properties.

In the second project (Fjällberg et al., 2005), samples were collected from several plants burning mainly woody biomass in grate furnaces as well as FB furnaces. Efforts were made to gather a spectrum of ash composition from calcium-rich and silica-poor residues to calcium-poor and silica-rich residues. The binding properties of the residues, alone or in pairs were investigated in cup tests and the mechanical strength and leaching properties were determined.

The results were:

- Residues with already comparatively high silica content were the most stable and their stability could not be increased
- The stability of residues with comparatively high calcium oxide content could be improved by mixing them with a high-silica ash, with silica dust or with potassium silicate
- Treatment has little effect on potassium leaching

Although wood ash should bind, it doesn't always do so because the components are not reactive (too low a temperature in the furnace or ash was slaked prematurely) or because silica is not available in a finely divided form to react to a calcium aluminosilicate hydrate or cement gel. Heat treatment improves the binding properties. Adding a silica-

rich material increases the availability of silica for these reactions, shifting the overall composition of binding ash materials to a more silica-rich one than Portland cement.

Agglomeration and ageing also do not reduce leaching of alkali from the residues. The side effect of ageing and binding reactions during stabilisation is that alkali is redistributed as soluble salts in the pores of the solids. Potassium is even more available and will leach out of the agglomerated ash to soil at the first rain, and then further from the soil to surface waters.

Closing pores when pelletizing will reduce leaching of alkali to some extent. An alternative is coating the pelletized residue with an impermeable layer which technique was tested in the second project. Pelletized ash was coated with small amounts of silica dust or water glass (sodium silicate solution), at most ca 10 % by weight which was felt to be economically justifiable (Fjällberg et al., 2005). The effect on potassium leaching was quite slim. Actually, this 10 % of additional material is not enough to provide an adequate barrier. Taking into consideration the size of the ash pellets and experience in industries where coating pellets is routine, the quantity of material that should have been added would have been of the order of 50 %. This implies a significant extra cost.

## 9.2 Opportunities

Carbonation of combustion residues has been studied in a recent PhD thesis, with the aim of stabilising them (Ecke, 2001). Carbonation lowers the pH of the residues and carbonates of heavy metals are generally less soluble than their chlorides or sulphates. To some extent, this is a natural process when the residues are exposed to air and moisture, but improving the yield and shortening the time scale of the process could lead to improved properties of the residues with respect to environmental criteria.

The experience with APC residues from municipal solid waste incineration in Ecke's thesis was that the mobility of critical elements (lead, zinc, chromium) was reduced after carbonation. The mobility of cadmium increased though in this investigation, and it is well-known that the mobility of oxy-anions is increased. Such a treatment could stabilise some components (heavy metals) and contribute to flushing out other (oxy-anions). In an exploratory project for the Ash Programme, a bottom ash from MSWI and a fly ash from combustion of wood waste were carbonated to test how far their properties could be modified (Svensson et al., 2005; Todorovic and Ecke, 2006; Todorovic et al., 2006).

The acceptance criteria at landfills were used as a reference for mobility, *i.e.* leaching properties. Besides chloride, the critical elements were antimony, chromium, copper and molybdenum for the untreated bottom ash, and chromium, lead and selenium for the untreated fly ash. After carbonation, leaching properties had changed, mostly in the desired direction, but not sufficiently for having consequences for the classification of these materials according to the Waste Directive or the acceptance criteria. It is though an efficient method to reduce the concentration and leaching of chlorides. The process

can certainly be developed further, but such an effort needs a more precise purpose to be initiated.

Foamed bitumen is used in some countries to stabilise granular materials in civil engineering works. It has been shown that the release of PAH's from soil is significantly reduced and that the mechanical properties of the material are improved by the cohesive properties of the bitumen surface layers. Foamed bitumen could then be a readily available technique to modify both mechanical and leaching properties of bottom ash from MSWI.

A sample of MSWI bottom ash that has been aged for twenty months outdoors was fetched from SYSAV and sub-samples were treated using different recipes: foamed bitumen, bitumen emulsion and cement (Bendz et al., 2006b). Mechanical properties determined are porosity, density, stiffness module and compressibility. Porosity and available surface (BET) were approximately halved when ash was coated with bitumen. Samples with 4.5 % bitumen had better mechanical properties than untreated samples. The particles attach to each other more easily and the treated ash produced less dust, which could be an advantage.

Leaching properties were determined using standardised tests:

- Batch leaching (EN 12457-4); no significant change was observed in the leaching properties, except for nitrate that decreased somewhat and for DOC that increased somewhat
- Column leaching (EN 14405)
- pH-static leaching (prEN 14997)
- Surface leaching (NEN 7345)

In general, the effect of a fresh bitumen coating on leaching properties was quite limited, besides the increase of dissolved organic carbon (DOC) that one can expect. The available contact surface between ash and liquid decreases, but dissolution is mostly determined by solubility at neutral or close to neutral pH-values. At high or low pH-values leaching depends more on available surface and a coating mostly decreases leaching. There are exceptions:

- Antimony and selenium leached less at all pH-values in the pH-static test: apparently their mobility is determined by surface properties, which agrees with the findings in the antimony project (Bäckström, 2006)
- In column tests leaching of several metallic elements (manganese, copper, zinc, nickel, cobalt, cadmium and mercury) increased in coated samples
- Surface leaching of copper and zinc decreased in coated samples

### **9.3 Well-informed choices**

A no-treatment policy does not preclude orienting oneself about price, performance and potential of more advanced treatments than one is willing to consider. A large number of stabilization techniques have been developed and described in literature or in press releases. They consist of:

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- Either heating up the ash and vaporising comparatively volatile compounds, with the side effect of sintering or vitrifying the residue (an additional stabilizing process)
  - Or extracting soluble compounds from the ashes using water or a water solution

These techniques have been reviewed in separate reports in the inception of the Ash Programme (Wikman et al., 2003b; Bjurström and Steenari, 2003). Thermal treatment yields stabilised residues with a low content of the volatile heavy metals, but it is expensive and the energy consumption is high (Wikman et al., 2003b). The cost of treating the concentrate of heavy metals is often not included in surveys. Wet treatment is more easy to work with, can be made small-scale, has a low energy consumption and usually treatment of all residue or waste streams is included in the process design (Bjurström and Steenari, 2003). On the down side, it is not as efficient as thermal treatment in separating heavy metals and the solid residue meant for utilisation does not have satisfactory mechanical properties.

A side glance was also cast at a more passive treatment: blending APC residues from waste incineration with 5 % compost and storing them under landfill conditions (Ecke and Bjurström, 2005). The original aim of its creators was to devise a means to reclaim landfilled combustion residues in the distant future. When it degrades biologically the compost produces both carbon dioxide and humic materials which both bind heavy metals as well as other organic substances. This process yields a residue with a different composition, but it is far too slow to be attractive under normal industrial conditions.

## 10 Information and communication of results

The Ash Programme has used a number of different distribution channels in order to spread information based on the results of its research and development projects, and so that these results reach a broad range of target groups. A primary interest is making the results available to construction contractors, the environmental authorities (local, regional, national levels), and the general public. It is of vital importance that the Ash Programme maintains a legitimate credibility and that the results build confidence in the use of combustion residues in society. It is the experience of the Ash Programme that information activities such as seminars and meetings, which are coupled to existing development projects (*e.g.* a newly constructed road that contains fly ash of solid biofuels), receive the best results in terms of information transfer. In general, good examples are the most successful method of encouraging further use of combustion residues.

Information about the results of research projects conducted within the Ash Programme are spread primarily through reports and fact sheets that are published on Värmeforsk's website<sup>21</sup>. In addition to reports, results from the Ash Programme are distributed in the form of guidance documents and handbooks. Technically speaking, these are also reports published by Värmeforsk, but in contrast to the research reports, these are instead written as instruction manuals with construction contractors as the primary target group.

### 10.1 Guidance documents

During the first few years of the Ash Programme (*ca.* 2002 – 2004), there was a strong need to identify appropriate areas of research and development on the use of combustion residues, and to illuminate the various forms of legislation that controlled the use of combustion residues in society and the environment. Several guidance documents have therefore been produced which focus on specific high-priority issues.

Bjurström (2002) suggested a course of action that could be adopted by Värmeforsk, within the Ash Programme, for the recycling of wood ash to forest soils. The underlying assumption in the study is that wood ash should be recycled (*i.e.* spread on forest soils) in order to compensate for the removal of mineral nutrients when whole trees are harvested. Bjurström (2002) suggested several measures that ash-producers might consider, including the following: 1) to establish a structured procedure to identify obstacles to the use of combustion residues, and propose actions to remove these obstacles; 2) to stimulate interest in recycling ash by identifying areas where a short term benefit for growth can be demonstrated; 3) to develop methods for quality assurance, including standardization, which would lead to a wood ash product with consistent and well-known properties.

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<sup>21</sup> [http://www.varmeforsk.se/english/bibliotek/biblo\\_index.html](http://www.varmeforsk.se/english/bibliotek/biblo_index.html), available 2008-12-16

Adler et al. (2004) addressed the difficulties associated with the classification of combustion and incineration residues according to the new Swedish waste ordinance of 2002, and proposed a method for classification. The proposed, simplified method implies the following: 1) Reference substances are selected for different inorganic and organic chemical substances in the residues. These reference substances are utilised as a basis for the classification against the properties H4 – H8 and H10 – H11. This leads to a very substantially reduced need for analyses. The reference substances were chosen conservatively so that the hazard for health and environment would not be underestimated; 2) the main principles of the regulation on chemical products are applied (e.g. the weighting of properties relative to each other); 3) the property H13 is assessed based on the properties H4 – H8 and H10 – H11; 4) the property H14 is assessed based on the rules for environmental hazards in the regulations of the National Chemicals Inspectorate.

An increase in the utilisation of combustion residues is commonly related to the need for increased recycling and reduced waste deposition in landfills. There are a number of driving forces behind these goals, including the 6<sup>th</sup> EU Environmental Action Plan, the national environmental goals *Good built environment* and *Non-toxic environment*, the general rules of consideration in the Swedish Environmental Code, and taxes on waste received at landfill sites. With these various driving forces in consideration, Wilhelmsson and Pajkull (2004) investigated legislation that applies in different phases of a construction project. Their report also discussed issues that need to be taken into consideration for the appropriate selection and use of waste.

The use of combustion residues in road construction has been hindered with several obstacles in a number of EU countries. Kärrman et al. (2004) presented a compilation of how different EU countries have dealt with and overcome these obstacles, and suggested a path of action for Sweden. The study focused on Belgium, Netherlands, France, England, Finland and Denmark. Typical obstacles for the use of recycled materials in construction were identified for Sweden as well as these EU countries: 1) ambiguous laws and regulations, 2) lack of experience, 3) weak economic incentives, 4) project planning (i.e. material-related decisions are made too late in project planning), and 5) lack of methods for showing the benefits of recycling. In terms of the final point, Kärrman et al. (2004) maintained that environmental system analysis (ESA) needs to be employed, considering both the road construction and the alternative management of combustion residues (e.g. recycling vs. landfill disposal).

In a later report on conditions within the EU, Jansson and Wilhelmsson (2008) show how the use of waste / alternative building materials for construction purposes is managed in eight different countries within the European Union. A short review is provided on national legislation and the effect of guidelines on supporting and restricting environmentally-responsible reuse. In general, regulations are based on the protection of water and soil resources and the protection of health and the environment, with the pronounced aim of supporting waste reuse for construction purposes. In most countries, regulations concerning the environmentally-controlled use of secondary building materials contain these elements:

- Inert wastes are often free for construction use
- Specific waste fractions are allowed
- Specified use is defined
- Different material categories might be used/prescribed for different purposes
- Combustion residues of different origin are an important source for secondary aggregates
- The quality control of materials and construction is essential
- Specific precautions or remediation are prescribed, according to the purpose
- Reporting or simplified permit processes are prescribed

In the EU, there are generally specific regulations concerning use of secondary materials. If used in other applications than prescribed, the use will normally be required to go through the standard environmental permitting process.

## 10.2 Handbooks

A series of handbooks has been produced by the Ash Programme, but these publications are always the result of collaboration between many organisations, with the goal of facilitating the use of combustion residues in various applications. The handbooks are summarised in Table 6-1, and have been discussed previously in Chapter 4.

It has been noted that an obstacle to the use of combustion residues in geotechnical applications (e.g. road construction) is that construction contractors, road administrators, governmental authorities, and industry often lack experience with such materials. Handbooks (manuals) are therefore written to bridge the gap by illustrating the potential uses of combustion products and providing detailed instructions for construction applications.

*Table 10-1. Handbooks describing construction with combustion residues.*

<b>Title (English / Swedish)</b>	<b>Reference</b>	<b>Other collaborating organisations</b>	<b>Report</b>
Manual: Fly ash in civil engineering, Gravel roads <i>Handbok: Flygaska i mark- och vägbyggnad, Grusvägar</i>	Munde et al., 2006		954
Instruction manual: Fly ash stabilised sludge (FSS) as liner material <i>Vägledning: Flygaskastabiliserat avloppsslam (FSA) som tätskikt</i>	Carling et al., 2007	Avfall Sverige, Svenskt Vatten Utveckling	1010
Slag gravel for combined unbound materials in roads and ground works: Handbook <i>Slaggrus för sammansatta obundna material i väg- och anläggningsbyggande: Handbok</i>	Tyllgren, 2008		1054
Alternative construction materials for landfills. Guide. <i>Alternativa konstruktionsmaterial på deponier. Vägledning.</i>	Rihm et al., 2009	Avfall Sverige	1097

It has been an experience of the Ash Programme that handbooks are introduced most successfully when they are quickly followed by a workshop on the same topic. This was

the case with the introduction of the Fly ash – stabilised sludge manual (Carling et al., 2007); the subsequent workshop provided an appropriate venue for spreading information about the manual.

### **10.3 International publication**

Few of the results produced by research and development projects within the Ash Programme have been published internationally in peer-reviewed journals, although a greater number have been presented at international conferences and appear in conference proceedings. The relatively low publication frequency reflects the distribution of funding resources by Värmeforsk: because of the applied nature of research and development projects conducted within the Ash Programme, a majority of the projects are awarded to industry representatives and consultancies that have direct knowledge and contact with the industrial processes producing combustion residues. Certain projects are highly applied and would hence be difficult to publish in scientific journals (e.g. evaluation of specific testing methods). University employed scientists, who are more inclined to seek international publication of their research findings, are a minority. Therefore, most projects are reported in Swedish and are available on the Internet, but are not readily available to an international, English speaking audience. A review of the programme has been presented at a WASCON international conference (Ribbing, 2007).

### **10.4 Environmental systems analysis**

The utilization of alternative materials in construction, including recycled products such as combustion residues, often requires motivating the use of these materials for construction contractors, the environmental authorities, and the general public. One tool that has been developed within the Ash Programme, for specific application to combustion residues, is environmental systems analysis. Kärman et al. (2006) presented a method for environmental systems analysis that considered the use of recycled materials in a broad perspective. Various scenarios for the beneficial use or the disposal of the residuals that occur in a region were analysed. The method considered emissions to air and water as well as the conservation of natural resources and energy. Two case studies were considered: Case study 1 dealt with municipal solid waste incineration (MSWI) bottom ash, where scenarios for the beneficial use of ash in drainage layers of landfill covers and road construction were compared. A third scenario was also included as a reference, where the MSWI bottom ash was landfilled. The result of case study 1 showed that the use of ash in road construction was the most beneficial alternative in terms of the conservation of natural resources and energy, and also in terms of the release of several metals. In case study 2, the beneficial use of fly ash from peat combustion was analysed, including the use of peat ash as a construction material in small county roads and the use of peat ash mixed with sewage sludge as a covering material on landfills. This case study also included a reference scenario in which the peat ash, generated in Uppsala County, was landfilled. The results from case study 2 indicated that the use of fly ash for county roads used largest amount of crushed rock, but was the most beneficial alternative with regard to metal release. In terms of energy

consumption, the landfill cover alternative was the most beneficial alternative when transporting combustion residues distances of up to 60 km. This study demonstrated that the developed method for environmental systems analysis has a potential to be a tool for strategic environmental assessments in regional natural resources plans, municipal planning and environmental impact assessments.

### **Systems analysis and Life cycle analysis**

Systems analysis is the procedure and methodology for producing a basis and guidance for a systematic analysis in decision making situations (Beck, 1997). Systems analysis has also been described as a multidisciplinary approach that was developed in order to deal with complex problems that arise in society as well as in companies and organisations (Miser and Quade, 1985). Environmental systems analysis is a type of systems analysis that is used in the comparison of different alternatives so as to reach a defined function from an environmental perspective.

A well-known method for environmental systems analysis is life cycle analysis (LCA), which is a method for analysing and valuing the environmental impact of a product, a material, or a service during its life cycle, “from cradle to grave”. Central concepts are the use of the life cycle perspective, that a system is defined, and that the system’s resource consumption and emissions to air and water are quantified. The results can be used for comparing different alternatives for producing the same function, and for identifying where the primary environmental impact occurs in the system (adapted from Kärman et al., 2006).

Further discussions of life cycle analysis were held in Kärman et al. (2008). In this study, the aim was to investigate and suggest how a life cycle perspective could complement decision support with regard to the use of combustion residues. In accordance with procedures established during the implementation of former environmental legislation in Sweden, potential combustion residue applications are evaluated only in terms of local impacts generated by the application and the definition of waste. However, the current Swedish Environmental Code supports the consideration of environmental impacts from global warming, acidification, eutrophication in the prioritization of natural resource use. According to Kärman *et al.* (2008), there is a clear potential for implementing a life cycle perspective in decisions regarding combustion residues, primarily in environmental goal planning, in regional and municipal planning or in permit applications. In environmental goal planning, the life cycle perspective is useful for supporting decisions on remediation measures and follow-up.

A life cycle perspective could also be employed when considering material supply (e.g. traditional ballast or combustion residues) in construction projects (Kärman et al., 2008). The life cycle perspective can be included by 1) including the secondary materials (e.g. combustion residues) in inventories of sources for material supply when developing material supply plans, 2) combining material supply plans with an analysis

of the environmental impact in a life cycle perspective, and 3) including construction with combustion residues in material supply plans in permit applications.

Within the Ash Programme, Environmental Systems Analysis (ESA) has been employed (Olsson et al., 2008) to compare the relative environmental impacts of combustion residue recycling to forest land with combustion residue use in road construction. A third alternative was the disposal of combustion residues in landfills. According to ESA, both the recycling of wood ash in forests and use of combustion residues for road construction have benefits in terms of conserving natural resources and energy, compared to disposal in landfills. Critical parameters were 1) the assumption that nutrient compensation is needed if wood ash is not used in forests and 2) the system boundaries chosen for assessing heavy metal leaching. If nutrient compensation is not considered as necessary, the influence of transport and maintenance on the results increases.

## 10.5 Allaska

The database Allaska (a term implying "all ash" in Swedish) has been established as a compilation of combustion residue properties; these properties have been determined in various research and development projects within the Ash Programme. Additional data from the material testing of combustion residues (primarily dynamic cyclic triaxial tests) have been provided by the Swedish National Road and Transportation Research Institute (VTI; Arvidsson and Loorents, 2005). The database has evolved since 2004, originally starting as a database with 22 combustion residue samples that were studied in eight R&D projects (Bjurström et al., 2004); the database has evolved over the past several years (see Bjurström et al., 2006; Bjurström and Øritsland, 2008; Bjurström et al., 2009). During the period 2006 – 2008 (Bjurström et al., 2009), two external datasets from *Framework Programme Recycling of Ash* and *Värmeforsk's Base Programme* were added to Allaska, bringing the total number of samples in Allaska to ca. 450 registered samples

The database is flexible and was created in Microsoft Access in order to store the data in an easy-to-access database; data is provided in both Swedish and English. The database is available on the Internet at [www.askprogrammet.com](http://www.askprogrammet.com) and has a user-friendly interface for the downloading of data. The information stored extends from the fuel and the design of the combustion plant to the specific properties determined for a given use, such as shear modulus or permeability. Attention has especially been paid to the fact that fuels are often mixed and that the combustion residues are often mixed with other residues or with other materials. The database has been expanded with regard to availability, database searches, data download, and single data tracking capabilities.

## 10.6 Newsletters and multimedia production

A newsletter, *Ash and the Environment (Askor & Miljö)* has been regularly published (3 times / year) with the aim of spreading information about activities and research in the Ash Programme. Among all the various distribution channels (e.g. research reports, seminars, courses), the newsletter reaches the largest interest group at municipality offices, county administrations, governmental authorities and private companies.

The short film *Ash – a road to success* has been produced by Svenska EnergiAskor, a non-profit organisation owned by 11 energy companies, in order to market the use of combustion residues in roads. The film has been shown at meetings with environmental and health inspectors, with the aim of illuminating questions regarding the environmentally correct use of combustion residues, and the implementation of construction projects with combustion residues.

## 10.7 Seminars and courses

Seminars have been used by the Ash Programme in order to spread research results and information about ash applications and demonstration projects. Several seminars have been held since the start of the Ash Programme, including *Waste products and the Swedish environmental goals* (2002; *Restprodukter och de svenska miljömålen*) and *Use of sludge and ash as sealing layers on landfills* (2005; *Användning av slam och aska som tätskikt på deponi*). It has been noted (Herbert, 2008) that seminars are especially successful when they are held in conjunction with the completion of a field demonstration, highlighting the application of combustion residues. Seminars have also been held by other organizations and authorities (e.g. Swedish forest agency, Swedish environmental protection agency, Swedish district heating association) where results from the Ash Programme have been presented for a broad audience.

An academic course has also been developed, *Environmental assessment of the use of ash from energy and heat production (Miljöbedömning vid användning av energiaskor)*, Tiberg et al., 2008). The course discusses the possibilities to reuse combustion residues in different applications, provide information on the environmental and geotechnical properties of various combustion residues from energy production, and informs about the availability of handbooks, guidance documents and reports concerning the use of combustion residues. The course is primarily oriented to municipal and county employees, but students and incinerator and boiler operators will also be able to register for the course.

## 11 Analysis and synthesis of the results

In the following section, an analysis is provided of the Ash Programme for the period 2002 – 2008. The Programme has addressed a relatively wide range of questions and issues that have arisen over the years, with the purpose of providing a solid scientific basis for assessing the implications of combustion residue use in society. Given the wide range of issues that have been covered in the Programme, there is a need to tie together the general findings of these investigations in a synthesis of the results. Such a synthesis, given below, examines the major factors that have influenced the direction of research in the Ash Programme over the years, and points to key issues that currently determine the use of combustion residues in society.

### 11.1 Regulatory changes

Since the inception of the Ash Programme in 2002, the playing field has changed continuously. In 2003, the Swedish Environmental Protection Agency declared<sup>22</sup> the following (free translation):

- The use of waste products and waste must be stimulated. The use of waste as a resource should therefore be considered if the risk of contamination is assessed as being “insignificant”.
- The environmental goal *A good built environment*<sup>23</sup> cannot be reached without attaining the environmental goal *A non-toxic environment*.
- Guaranteeing a non-toxic environment must be given priority. That is, environmental consideration is given a higher priority than resource conservation when the risk of contamination is assessed as “not insignificant”.

This declaration provided the Ash Programme with incentive to investigate combustion residue applications that had insignificant impacts on the environment and human health. Since the term “insignificant” was never truly defined, there has been, since 2003, considerable discussion as to which applications of combustion residues pose or do not pose an insignificant risk (*ringa risk*, Swedish).

Over the ensuing six years (2003 – 2008), governmental directives and legislation have been introduced that have had an impact on the use of combustion residues. These include the following:

- REACH (EU)
- Building product directive (EU)
- *End-of-waste* criteria (EU)

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<sup>22</sup> Declaration of intent that was presented at a seminar with the Swedish Geotechnical Association on 19 March 2003.

<sup>23</sup> The Swedish Riksdag adopted in 1991 a set of 16 Environmental Objectives, [www.miljomal.nu](http://www.miljomal.nu). This awkward English wording is a literal translation of the Swedish wording (“*God bebyggd miljö*”) of objective number 15, which is the responsibility of the National Board of Housing, Building and Planning (*Boverket*). It consists of a number of targets related to urban planning, energy efficiency, health and environment as well as aesthetic qualities.

- Guideline values for contaminated soil (Swedish EPA)
- Criteria for the utilisation of waste as construction material (Swedish EPA)

In general, these directives and guidelines have imposed greater restrictions on the use of combustion residues in construction than the directives and guidelines that were enforced at the start of the Ash Programme. The application of wood ash to forest soils has generally not been affected by these directives and guidelines.

## **11.2 Combustion residues in forestry**

With regard to ash as compensation to forest soils, the regulatory context was established before the Ash Programme was initiated. Ash from the combustion of logging residues, which has been extended to mean combustion residues from woody biomass, should be returned to the forest soils. The purpose is to counteract the additional acidification of forest soils when logging residues are removed as fuels on top of the harvesting of timber or pulpwood of conventional forestry.

There is a regulatory framework in place, favouring compensation with ash. Potential effects on the environment have already been investigated in a large number of studies within programmes financed by government agencies. There are very few discernible negative effects. The environmental risks of spreading ash on forest soils are well-known and acceptable.

Although there is a general acceptance of compensation with ash, actual full-scale compensation is not yet established. There are a few non-technical barriers, some of them due to the intrinsic difficulties of communication between different stakeholders; some of them are due to regional differences within the country. The lack of economic incentive doesn't make things easier. Recycling ash is an additional cost for the ash producer as well as for the land owner, and the latter is not to expect an immediate effect on growth.

Rather than duplicating forestry research already carried out, the Ash Programme has decided to attempt demonstrating positive economic effects, at least in some cases that hopefully will increase the general interest in compensating with ash.

It is well known that the application of ash on drained peat lands is often followed by a visible increase in growth, and consequently in revenue. However, negative environmental side effects are feared: increased production of greenhouse gases from decaying peat, leakage of nutrients with run-off. Results from the research projects carried out in the Ash Programme indicate that, on the contrary, the production of greenhouse gases decreases after ash has been spread. Leakage of nutrients does not seem to be a larger problem than on mineral soil. There is though an uncertainty in the positive effect on growth that is expected: the dose of ash that is permitted, 3 t/ha, may not be enough to last through a whole growth cycle.

The Ash Programme has also wished to show that compensation with ash may lead to an increase in growth on mineral soil. That was already known as a tendency, but in the public mind, this increased growth on some soils has been completely overshadowed by a reduction in growth on other soils. The key factor is the productivity of the sites: low productivity, reduction of growth (which is the message often retained<sup>24</sup>), high productivity, increase in growth.

Results from sites where ash or lime has been spread have been revisited. It is not easy to prove beyond doubts, but the tendency has been confirmed. However, there is some controversy about which mechanisms are responsible for the observed increase in growth.

A difficulty faced by the Ash Programme is that it is an applied Programme with a time-frame of three to five years to fruition. There is a time delay of approximately five years before any effect of ash spreading on growth can begin to be observed. The time-frame relevant to forestry is of the order of decades. Negative effects may be observed almost immediately.

### **11.3 Combustion residues in geotechnical applications**

One of the major issues that have been addressed in the Ash Programme is the long-term stability of structures constructed with combustion residues, such as landfill covers and roads. The environmental impacts of such structures and the associated environmental risks have been another major issue.

#### **11.3.1 Long-term assessments**

Since the use and performance of combustion residues in construction has been poorly documented in the past, there is little data on the development of geotechnical properties or environmental impact in applications (e.g. road construction) that are older than ca. 10 years. In general, assessments on the order of 25 years or longer are needed in order to predict long-term changes in chemical composition and geotechnical properties with a high degree of certainty. Thus, the Ash Programme has identified long-term assessments as being of vital importance for future combustion residue use.

There are currently a large number of landfills that need to be covered in Sweden, and the use of combustion residues for this purpose in landfills is generally accepted. The use of fly ash stabilized sewage sludge (FSS) in municipal landfill covers was first extensively tested in Sweden in 2003 – 2004, and thus long-term assessments of cover stability and permeability are not available. Percolation rates through FSS covers are generally low ( $< 30 \text{ l m}^{-2} \text{ yr}^{-1}$ ), although higher rates have been measured. The greatest concerns with FSS covers are related to material longevity (i.e. degradation of organic material in the cover) and the change in percolation rates with time, along with nutrient

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<sup>24</sup> This could be because the risk of a loss is very often ranked higher than the expectation of a profit.

leaching. The further evaluation of these issues should be addressed with both laboratory experiments and long-term field demonstrations.

Although the Ash Programme has only existed since 2002, and has thus had a limited amount of time for the assessment of new applications during the Programme period, participants in the Ash Programme have been able to assess older applications in collaboration with producers of bottom ash (*e.g.* district heating plants). In particular, the road construction projects of Törringevägen, Dåvamyran and the Gumpekulle cloverleaf have been investigated between 8 and 16 years after construction (Arm et al., 2008a; Bendz et al., 2006a). Geotechnical data from these studies indicate that roads with bottom ash in the subbase layer maintain a relatively constant compressive strength, and that bottom ash is a viable alternative to crushed rock in the subbase layer. In terms of environmental impact, the leaching of bottom ash in roads is governed by the presence of a wearing course composed of asphalt. With an asphalt cover, there is little water flow within the road body (Bendz et al., 2006a); this is indicated by studies (Arm et al., 2008b) that demonstrate that 10-year old bottom ash from a subbase layer maintains a similar leachability to fresh bottom ash. As water flow is very low below an asphalt cover, the leaching of soluble components from a subbase layer with bottom ash is comparable to the level of leaching with crushed rock (Arm et al., 2008a).

The results from the various studies with bottom and fly ash suggest that it is not the direct environmental impact caused by the leaching of combustion residues that pose that greatest risk; leaching can be controlled in the proper design of applications (*e.g.* roads). Instead, the greatest uncertainty and, hence, the greatest potential risk is associated with road construction (*i.e.* dust dispersal; Gustafsson et al., 2006, 2009) and with abandoned roads (Bendz et al., 2006c; Wik, 2009). The long-term fate of combustion residues in road bodies is of course associated with a high degree of uncertainty. Will the road still be in use in 50 years? Will society have forgotten about the bottom ash in a road after 200 years, and try to grow crops in this material? In order to be able to consider the implications of these questions, it is necessary to be able to assess the environmental risks posed by combustion residues in various applications.

### **11.3.2 Environmental risk**

The key to using combustion residues in geotechnical applications is in the production of risk assessments that provide a credible and realistic picture of environmental impact. Credibility is often damaged by what is considered subjectivity in these assessments: there is generally an incomplete understanding of the long-term material properties as well as the long-term effects (*e.g.* leaching) of combustion residue use in an application. This results in the assignment of parameter values and material properties that are highly conservative and will overestimate such processes as leaching. In order to reduce this uncertainty, long-term studies and continued material characterization are needed.

Demonstration projects with bottom and fly ash have indicated that leaching can be controlled with the proper design of applications (*e.g.* roads). However, on a regional scale, environmental systems analysis has shown that the metal flows arising from

bottom ash use in roads are *not* insignificant compared to the total flows from crushed aggregates used in roads (Wik, 2009). This is the case when bottom ash is used with elemental concentrations equal to the guideline values established in Bendz *et al.* (2006b). The findings by Wik (2009), in combination with the general scepticism shown by environmental authorities and construction contractors, suggest that additional measures need to be imposed for minimizing the potential environmental impact posed by combustion residue use. The feasibility of washing combustion residues (*cf.* Bjurström and Steenari, 2003) so as to remove soluble salts (primarily sulphate and chloride salts) should be re-evaluated, despite the high costs associated with treatment.

### **11.3.3 Non-technical barriers**

The Ash Programme has demonstrated that combustion residues can be used to replace traditional materials (e.g. crushed aggregates) in construction. However, the current demand for combustion residues in construction is limited and crushed aggregates are generally selected in preference to other, less well-known, alternatives. The limited use of combustion residues can be explained by non-technical barriers, including risk perception and attitudes. The one issue that needs to be addressed, based on the environmental goal *A good built environment*, is whether our society can afford to ignore a readily – available material that can replace a natural resource, when we are continually seeking sustainable solutions to our resource needs. At present, the statistics speak for themselves: the uncertainty inherent in the use of combustion residues has been sufficient to limit their application; the exploitation of traditional materials is preferred.

Although the application of combustion residues in forestry and in landfills is accepted and non-controversial, this tangible need for combustion residues does not currently appear to be relevant for geotechnical applications. If a parallel is to be drawn with ash use in forestry, support will be needed from the environmental authorities and the Swedish Road Administration (for use in roads) if specific use in construction is to increase. Furthermore, it is likely that legislation or economic incentives will be required to motivate increased use in construction.

## **11.4 European trends**

A study from the International solid waste association (Crillesen & Skaarup, 2006), based on data from year 2003, indicated that several European countries (Denmark, Netherlands, France, Germany) actively utilize MSWI bottom ash, or are starting to view utilization as a viable option (e.g. Belgium). The primary uses of MSWI bottom ash are as building and road construction materials in these countries, with less than 30% of bottom ash being deposited in landfills. This is in contrast to Switzerland where all bottom ash is sent to landfills. In the European countries with a high rate of utilization, national legislation exists for regulating MSWI bottom ash utilization. In addition, Denmark and the Netherlands have set governmental targets of 85% and ~100%, respectively, for the utilization of bottom ash.

In Sweden, where such national legislation and targets are lacking, the rate of bottom ash utilization is relatively low (see Chapter 1). The existence of regulations that clearly provide criteria for the use of combustion residues will also generally promote the use of combustion residues as uncertainties in the assessment process are reduced.

The criteria for utilization differ significantly in the Denmark, the Netherlands, France, and Germany, but an assessment of the risk for environmental impact is common to all. For example, in Denmark, the contribution of recycled materials (*e.g.* bottom ash) along with other pollution sources may not exceed the drinking water guideline values, based on the results from batch tests at an L/S ratio of 2. A common set of regulations have been developed for handling contaminated soil and inorganic residues (*e.g.* MSWI bottom ash, fly ash) in Denmark, which allows for a consistent utilization of recycled materials in construction.

In general, the strategic approach on legislation and management is different throughout Europe, which in turn is a barrier to utilization. Such barriers are tax policies, regulations on exporting bottom ash, different definitions of utilization or disposal, and complex regulations for utilization. As indicated by the ISWA report (Crillesen & Skaarup, 2006, p. 3), “managing MSWI bottom ash has the same concerns throughout Europe ... because the utilization and disposal practises are more or less the same, it is considered desirable to create a level playing field for the marketing, utilization and management of MSWI bottom ash in Europe”.

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