Meeting summary: Environmental conditions for coal ash utilization in infrastructures and paving

A discussion of the scientific – professional team attended by international experts
Workshop on the environmental aspects of coal ash utilization
Tel-Aviv 16.12.2009

Participants (lecturers quoted herein): Dr. Yaacov Nathan (chairman), Prof. Rami Keren, Prof. Uri Mingelgrin, Guy Reshef, Dr. Meni Ben-Hur, Dr. Pinchas Fine, David Weinberg, Yoetz Deutsch, Dr. Ariel Metzger, Dr. Hans A. Van der Sloot

Background documents: Discussion platforms - 16.12.2009
Summaries of workshop's lectures
Presentations of workshop's lectures

Discussion topics:

Coal ash contains, among other things, trace elements including oxy-anions, posing risk to man and the environment. These elements are released from the ash when it comes in contact with water. However, their availability is usually very low, firstly due to their limited solubility resulting from the chemical bonds created in the ash under the high combustion temperatures in the furnace and the glassy structure of the grains, and secondly due to their low concentration in the ash. Therefore, coal ash is not defined as a hazardous or toxic material by all the countries and environmental organizations throughout the world.

However, concern over the possible contamination of water sources and soils by toxic elements originating from the ash, and their intake by agricultural crops, brought the Ministry of Environmental Protection to define limitations and conditions on using ash as a paving and filling material in infrastructure and agriculture applications. Environmental control is expressed by defining maximum concentrations of contaminants allowed in the ash in order to fall under the classification "Usable Ash". The environmental conditions are derived by assessing the potential risk to the environment in the various applications, and take into consideration the mechanisms whereby toxic elements are released from the ash. Processes which affect the elements' leaching in every application and their behavior in the soil, (assuming a use of ashes which fall under the definition of "Useable Ash") are also considered.

The proposed environmental conditions have undergone a re-evaluation and re-formulation. For this purpose, studies sponsored by the National Coal Ash Board were used as a basis, and the conditions were presented during the workshop[1] which focused on the following main topics:

Risk assessments:

[1] Summary of discussions based on the transcript of the workshop's second day.

A. The behaviour of fly ash in road embankments – the sealing process and changes over time of water permeability.
B. The leaching of trace elements out of fly ash (viewed as a monolith) in road embankments, and out of the interface between the fly ash and the cover layer in the embankment.
C. A field-applicable environmental evaluation of the results of fly ash leaching tests done in the laboratory, using currently available methods.

Recommendations for regulatory actions, based on risk assessments:

A. Standardization – environmental standards to which fly ash, in its various applications, should conform.
B. Environmental conditions for utilizing fly ash in infrastructures located in different groundwater susceptibility zones.
C. Designing a laboratory method for testing fly ash as a monolith, and a formula for adapting test results to the ash's expected behaviour in actual road embankments.

Following is a summary of the main discussions that took place during the workshop (given in the order at which the presentations were given)[2]:

On the behaviour of coal ash in a road embankment

Prof. Rami Keren (Volcani Center for Agricultural Research)

Examinations of a fly ash embankment along road 6 (Revadim) that was compacted as required, showed that the layer was very hard, nearly as hard as stone. It was very difficult to dig into it (even using heavy machinery). In addition to its hardness, the ash embankment was characterized by a decrease over time in permeability to water. This occurred due to changes that took place inside the ash layer; its particles interacted with carbon dioxide that was brought with the water, and pozzolanic processes were also involved. These effects are critical, since the ash particles, after being compacted, are very dense; the greater the density of a material, the smaller the voids that are subsequently sealed due to the processes described here.

Similar findings were found in testing the water permeability in another fly ash embankment situated near the access road to Jisr a-Zarka. There, the embankment near the asphalt layer, after 3 years, was entirely sealed. In the case of the embankment near Revadim, it was the combination of the clay and sand layers above the ash, the vegetation that consumes water and the little precipitation in the region that precluded, from the start, the possibility of any rain water reaching the ash layer. Therefore, it is not necessary to debate the issue of regulations there, since nothing comes out of the ash layer, and even if water will reach the ash layer over time, it will be incapable of exiting, since by then the

[2] The discussion's protocol appears in appendix 1. Original presentations were in English and/or Hebrew, transcripted from audio recordings by Dan Shriki.

surfaces of the layer shall have been completely sealed, effectively preventing water from draining downward.

Dr. Hans A. Van der Sloot (ECN, The Netherlands)

From the available information and the input of Rami Keren and Yoetz Deutsch (that the drilling equipment in the ash embankment at Jisr a-Zarka broke) regarding the material strength, I can say that the material has considerable strength, and I do not foresee any significant changes occurring to the environment.

Dr. Yaacov Nathan (Geological Survey of Israel)

After 8 years of monitoring the groundwater at Zikim, where a bottom ash embankment was placed, no changes were recorded. This confirms our supposition on bottom ash, and we should follow the same principles in fly ash applications, since we consider it a monolith; we also need to monitor the quality of the water passing through it, since this will be the only valid way to determine the effects, if any, on that water – whether they be good, bad or non-existent.

Yoetz Deutsch (Geological Survey of Israel)

The Jisr a-Zarka site is effectively a large scale laboratory for examining the behaviour of coal ash throughout time. Three drills were done at a distance of approximately 1 meter from the road pavement above the embankment, in the cover soil. The third drill, which was done 8 years after the embankment was created, was disrupted on account of the resistance of a hard layer of coal ash. Its resistance was so great, that it simply tore the drilling rods and head; these components, it should be remembered, are made of steel and have a diameter of a few inches. This corroborates what was said earlier, that we have obtained a monolith, something as strong as a boulder, not merely a sparse layer of ash. We also found that a new phase was created (the mineral known as Stilpnomelane) and that the material condensed into a mass that is much stronger than it was earlier. These findings lead to the conclusion that the embankment is active, but not active as pertains to environmental issues.

Prof. Uri Mingelgrin (Volcani Center for Agricultural Research)

As discussed earlier, the ash in an embankment acts as a monolith. Water barely permeates it. This proves that the release of metals will be very slow.

Dr. Pinchas Fine (Volcani Center for Agricultural Research)

There is a process of re-mineralization and re-crystallization of the material (Yoetz Deutsch demonstrated this). So in actuality the minerals will be less soluble and the release of trace elements and heavy metals will decrease more than it is already now.

On the leaching of trace elements from a fly ash embankment to the environment

Prof. Rami Keren (Volcani Center for Agricultural Research)

In light of the findings given above on the behaviour of ash in road embankments, we are dealing here, in most cases, with a water impermeable mass, and one that obtains this status in a relatively short time. Most of the fly ash used in such infrastructures is covered by asphalt, so water will probably not reach it in any case, no matter how much time passes. As a result, all trace elements will remain within this mass. Even if we ignore these facts, which in themselves are sufficient to prove our point, even if a leak does somehow develop, we are looking at a preferential flow or motion of these elements toward the groundwater, and there is a long delay in their dispersion in the ground. So the elements transition rate is dynamic, as is the flow of groundwater through the aquifer. In light of this, the dilution effect of groundwater will in fact ensure that no change will occur to water quality. This is corroborated by the examination of groundwater in the area of Jisr a-Zarka embankment, where there were no changes to the concentration of heavy metals in the water, even though the water is shallow there and the distance to it is short. Hence, in order to calculate the dilution factor of trace elements, and subsequently apply proper regulations, it will be necessary to obtain several parameters, namely the depth and quality of the groundwater in the area of the ash application, the distance from it to water sources below and on the surface, the groundwater's direction of flow, and the concentration of trace elements in the groundwater.

Dr. Hans A. Van der Sloot (ECN, The Netherlands)

Characterization tests we conducted on Israeli fly ash (in the range of pH=7.8-12) showed that, in the case of Molybdenum and Selenium for example, the concentration of the leachate from "aged" ash (simulation of ash in an embankment, one year after placing) was lower, and in any case, the concentrations measured in all types of ashes were below the Dutch Soil Quality Decree threshold for insulated application (i.e. covered by asphalt). So things look well from this aspect.

Dr. Yaacov Nathan (Geological Survey of Israel)

Bottom ash leaching results indicate that the material is inert; in practical terms there is no constraint in utilizing it. Leaching from fly ash indicate that the material is non hazardous. The only question that remains is whether the ash caused a change in the composition of groundwater. In this regard, analyses of groundwater beneath the ash embankment and beneath the control area (i.e. no ash) at Jisr a-Zarka showed that the groundwater beneath the ash embankment were actually cleaner. That is, groundwater was not affected by rainwater that fell down on the embankment.

Dr. Meni Ben-Hur (Volcani Center for Agricultural Research)

The exposure of an embankment's fly ash to rainwater can be avoided by covering the ash with a layer of soil, as was done in the Jisr a-Zarka embankment. The surface runoff on the soil layer was greater than on the control area with no ash (gravel). The concentration of Boron in this runoff was higher in the ash area than that of the control area, And there are several explanations for that: 1. Capillary movement from the ash to the soil during the dry periods, and subsequent accumulation there; 2. Mixing of soil and ash due to subterranean fauna activities; 3. Adsorption of soil's boron (it is concentrated in the leaves, and with their decay they enrich the soil with it) by the roots of plants which reach down to the ash layer. And indeed, the concentrations of boron, and also molybdenum and barium, were much higher in plants growing in the ash area than in those growing in the control area. However, this can be due to the high solubility of these elementse in the fly ash. This situation was reversed for manganese and nickel, possibly because of their low solubility in the ash. This may be the reason for the elements' different behaviour.

Yoetz Deutsch (Geological Survey of Israel)

We tested 21 trace elements in the fly ash extracted from the drills at Jisr a-Zarka embankment, done in 2000, 2003 and 2007, in a similar location as possible. Two elements are presented here: Be and V, and no significant difference was exhibited in the concentration profile along the drill between the various years, although towards the edges of the drill section near the surface and in the depth, the elements content decreases as one approaches the soil.. The remaining elements behave more or less the same.

The leaching of elements from the fly ash was done using TCLP method. We can see that excluding Se, B and Mo, the others were barely leachable. The concentration of Se and Mo was very low from the start (less than 10 mg/kg), and in fact B alone that had a high concentration and ranges from 200 to500 mg/kg. Whether the embankments are inert or not, it can not be deduced, despite the fact we found no indication for cations or oxy-anions mobility.

Prof. Uri Mingelgrin (Volcani Center for Agricultural Research)

In the matter of the release of heavy metals to the environment, the critical parameter is their rate of release. Suppose we examine over time a site containing high levels of such elements. If their release rate is sufficiently slow, then even if in the distant future all metals reach the groundwater, it will be necessary to find just how much they were diluted because the water contained in the aquifer is not immobile. Will the metals reach the groundwater after one year, or after one million years? This question has great impact on public health issues.

Guy Reshef (Israel Water Authority)

We examined the leaching potential of trace elements in configurations of Colombian fly ash columns (alone), in natural soil (alone) and in a mixture of ash and soil (using a homogenous mixture and separate strata, i.e. the ash layer placed above the soil layer). We also examined the pH of the solutions. The pH, which is a factor that have a great effect on the solubility and concentration of trace elements in leachates, was lower in the mixed columns' leachate than that of the ash, due to the adsorption of hydroxyl groups to clay and aluminium and iron oxides. That also explained the fact that the leached amount for boron, vanadium and selenium in the mixed configuration was high compared to their combined amount in the soil columns and ash columns. This amount was low for chromium, molybdenum (oxy-anions) and lithium. For all these elements, the leached amount from the layered configuration was lower than that of the soil column and ash column combined. The amount of elements leached from the ash/soil columns (whether mixed or in layers) was higher than that of the soil only columns. The conclusion is that the addition of fly ash to the soil causes changes to its chemistry, and these may increase the solubility of trace elements contained in the ash.

Concerning the test method

Prof. Rami Keren (Volcani Center for Agricultural Research)

The TCLP method is unsuitable for characterization of the ash. Another method should be found, one that relates to the application of ash at the site and correctly reflects the effects the ash application has on the environment. It is according to such a method that regulations should be based, since in the ash application in the field it is incorrect to treat the ash as is; rather it should be considered as in a system that behaves in a manner unique to its location. Based on how the ash behaves in the embankment, it should be considered a monolith, not an aggregate. The test method for selecting the standard to be used in the fly ash embankment application should be chosen accordingly: leaching from a monolith, not from a granular material. In testing monoliths, it is necessary to know the geometric surface area of the mass, but this parameter is not relevant in the matter of elements solubility. The correct parameter is the true surface area, which is much greater than the geometric surface. Using the BET method (Brunauer-Emmett-Teller) will yield a good estimate of that surface area.

Dr. Hans A. Van der Sloot (ECN, The Netherlands)

Results obtained using a certain leaching test, whether TCLP, EN 12457, column test or pH dependent test, cannot be translated verbatim to the actual conditions in the site. So it is useless to compare such concentrations with the water quality criteria, since each particular element passes through a certain medium, such as a soil layer or an aquifer, on its route to the compliance point taken as a reference basis by the regulator in determining groundwater quality.

A key issue here is that if the fly ash used in road embankments hardens due to chemical reactions, it should be tested using the tank leaching test for monoliths, whereby the contaminant release rate can be calculated. This should be done jointly with a pH stat characterization test, which provides information on chemical changes in the material resulting from various factors. The question is where is the transition point between a granular medium and a monolith? The monoliths you created crumbled during their testing, and this shows that exposure conditions used in the tank test are in a certain regard extreme, since the material is exposed to great quantities of water. An alternative method is to test the material using the compacted granular leaching test (CGLT[3]); this test is designed for materials that behave as monolith in embankment applications, since water will not penetrate them, but will rather run along their surface. The material is tested as a compact granular material, and leaching is conducted from one side only, the side defined as the exposed side. The stages of this test are

[3] A brief overview of the method is given in appendix 2.

identical to those of the tank test, with the exception that the material will not crumble during the test. I am not in favour of the BET test; it behoves us to remain with the geometric surface area. Even if the material's surface is somewhat irregular, this phenomenon has no significant effect on test results. In the CGLT method, the compacted granular material is placed inside a water tank, and a sheet of glass beads (0.5cm in size) is placed upon its upper surface. Water is replaced according to the leaching steps (similar to the tank test), and care is taken to ensure that the sample is not disturbed. This preserves the gradient that develops with time. This method is flawless, and the results it yields are very consistent.

Yoetz Deutsch (Geological Survey of Israel)

A question was raised here, whether coal ash behaves as a monolith or as a powdery medium in embankment or other applications. If it behaves like a monolith, then it should be tested as such. Our routine tests were TCLP. Lately we made a transition to the European method EN 12457 for testing granular materials. It was suggested to test fly ash as a granular and monolith mass using the EA NEN 7375 method. If these methods show a measurable correlation, we will need to find their transfer factor. For this purpose we prepared cylinder masses of fly ash from 5 different sources. These were cured for 2-3 months, with the intention of having them harden. The Colombian and Indonesian ash crumbled within hours of placing them in water (the leaching solution), while the other three managed to maintain integrity. Of these three, the South African and Russian remained quite stable. The cracks that developed in the cylinders prohibited us from measuring their geometric surface area, since the areas inside the cracks were also in contact with the water.

Concerning susceptibility zones

Prof. Uri Mingelgrin (Volcani Center for Agricultural Research)

In light of the issues raised by Guy Reshef (susceptibility of zone B similar to that of zone A; quoted below), it may be necessary to change the recommendation for the proposed environmental conditions in application of fly ash within the various susceptibility zones. If I understand Dr. Van der Sloot correctly, he proposes that risk assessment and environmental conditions rely on the susceptibility of the specific area into which ash is to be introduced, not according to the general definition of susceptibility zones.

Dr. Pinchas Fine (Volcani Center for Agricultural Research)

We do not need to invent the wheel all over. Israel's most vulnerable conditions might be less vulnerable that those of the Netherlands, since the groundwater here is much deeper, the soils are mostly of limestone etc. In the Netherlands the soil is generally more acidic and sandy, and the groundwater is shallower. Therefore, if they consider it safe to use fly ash in infrastructures[4], it surely

[4] In the Netherlands, use of fly ash in infrastructures is not permitted. Only recently has a sample fly ash embankment been created and the environmental conditions pertaining to such applications brought under consideration.

should be safe to do so here. We just need to establish proper regulations, not invent something from scratch.

Guy Reshef (Israel Water Authority)
Zone A refers to the mountain aquifer, where the groundwater is very deep, around 300 meters below the surface. Zone B refers to the coastline aquifer, where the groundwater is much shallower and there is a risk of its contamination. Therefore, it is considered as zone A. This might affect the recommendations for required environmental conditions.

Summary

- **On the matter of evaluating the risks involved in fly ash applications – risks to the environment may be summarized as follows:**

  1. A road embankment made of fly ash that was compacted as required, hardens over time until it finally becomes impermeable to water. This occurs due to Pozzolanic processes and interaction with carbon dioxide in the air. In addition, in most cases the fly ash layer is covered by a layer of soil, and above that an asphalt layer. So in practical terms water will be incapable of reaching the ash.

  2. Assuming that water does permeate the ash layer, the above mentioned processes and re-mineralization processes occurring within the stratum will significantly reduce the ash's solubility and the release of trace elements from it.

  3. Assuming that water permeates the ash layer and manages to exit from it (a very improbable scenario), the water will move throughout the ash stratum in preferred paths, so most of the coal ash will not come into contact with the flowing water. As a result, the amount of toxic elements released from the ash will be smaller than if the water had flown throughout the entire ash matrix (uniform flow where the contact between coal ash particles and water flowing through the pores is maximal). Moreover, upon passing through the soil medium located beneath the ash layer, some of the elements that were released from the ash will be adsorbed; this will slow their motion towards the groundwater.

  4. If, against odds, water has passed through the ash layer and indeed reached the groundwater, the groundwater's dilution (due to the water's continuous motion) will effectively prevent any significant deterioration of their quality.

- **On the matter of finding the correct method for testing fly ash, for evaluating its behaviour in the environment:**

  Due to the difficulty in measuring the geometric surface area required in the tank leaching test (EA NEN 7375), utilization of the CGLT test (NEN 7347) will be considered. This test should provide a solution to the problem of packing the tested sample in a manner that prevents its crumbling, and by that make it possible to measure its geometric surface area (which in this method is considered the relevant parameter for risk assessment).

- **On the matter of defining regulations for the use of fly ash according to susceptibility zones**

  It was proposed that environmental conditions for fly ash use will be formulated after accepting the definition of zone B as equivalent to zone A.

The following conclusion was drawn on the behaviour of ash in road embankment applications – coal ash used in such applications has no significant effect on the environment. Coal ash treated according to standard road embankment specifications[5] and used as a structural filler in such applications (moistened, compacted and covered by soil and asphalt layers), does not release to the environment pollutants at concentrations bearing a potential risk, due to both the ash layer's imperviousness to water and chemical processes (pozzolanic and others) occurring inside the embankment, which reduce the ash's solubility. This conclusion is based on many years of research and monitoring of sites that were initiated by the National Coal Ash Board for the purpose of examining the environmental hazards involved in using coal ash as a filling material in road paving. The studies presented in the workshop can serve as a basis for the proposed change of the definition of fly ash used as a filling material in road embankments, from "Non-hazardous" to "Inert".

[5] The specification of the Israel National Road Company, extracted from chapter 51 of the general road construction specification: coal ash as a filling medium used in groundwork. http://www.coal-ash.co.il/docs/CalAsh_ConstructionWorks_MifratMaatzChap51_July2008.pdf

Appendix 1

Sections from the protocol on fly ash usage in infrastructure and paving

U. Mingelgrin (Chairman of the meeting, the participants of which were involved in the application of ash in infrastructure, paving and agriculture and pollutants monitoring procedures): The meeting will revolve around upgraded propositions for regulating potential pollution in fly ash applications in infrastructure and agriculture. Specific attention shall be given to the recommended maximum levels of leachable trace elements contained in the ash, as a function of its application.

R. Keren: Regulations and standardization need to take into account the physical-chemical properties of coal ash. One cannot ignore this, since the ash is a dynamic system that undergoes different stages; regulations should relate to those elements that might potentially harm the environment. In estimating the release of micro-elements from the ash, it is important to select the most appropriate testing method in order to set the appropriate standards. The correct method can redefine the fly ash as hazardous, non-hazardous or inert. For example consider the changing of the classification of a material from “Hazardous” to “Non-hazardous” when the heavy metal concentrations in the leachate are lower than their concentrations in the material. Therefore it is critical to conduct tests using the proper method, a method that will reflect the environmental conditions expected in the specific fly ash application. It is also necessary to know the solubility mechanism whereby micro-elements get released from the ash in order to obtain risk assessment evaluation from these elements in the different media. Additional data required for regulation concerns the depth and quality of the groundwater in the site of the ash application, the distance from water sources- subterranean (wells) and on the surface, the direction of flow of groundwater which is very important and the concentration of micro-elements in the water, in order to obtain their dilution factor.

Current regulations are based on information that is comprised in part of “Rules of thumb” and in part on the TCLP testing method. I believe that this method is the worst one, and that another method should be selected and adopted. Standards and regulations should be established based on testing methods appropriate for the designated use, and on the environmental conditions in which the system will exist.

In fly ash applications, the ash should not be taken as is, but rather as a part of a system, a system whose behaviour is dictated by its location and the surrounding conditions.

H. A. Van der Sloot: Results obtained from leaching tests, whether TCLP, EN 12457, column test or pH dependence test, cannot be translated one to one to the actual conditions at the site. So we cannot directly compare the received concentrations to ground water/ drinking water quality criteria, because that would leave a missing link: what was released from the material. Whether it is applied inside the soil, an embankment or the road, the element must pass through a certain medium, such as a soil stratum or an aquifer, in order to reach that point of compliance which the regulator considers a base of reference for determining the quality of groundwater or drinking water situated near the well.

Characterization leaching tests of different types of Israeli fly ash show consistent behaviour of Molybdenum, its leachate concentration using “old” ash being lower. The pH of the fresh ash is around 12, while the lowest pH is around 7.8; this range constitutes the range for a given application, as set forth by the Dutch Soil Quality Decree. All results yielded are below the upper threshold (which represents the Dutch maximal threshold), so things look good for you. Selenium behaves similarly.

I believe that the key issue in considering infrastructure for a road or a road embankment is that, if the fly ash hardens in its application stratum on account of chemical reactions, we must test it as a monolith, and in doing so try to perform a pH stat test as well. The combination of these two will provide us with a chemical characterization of the material, while the tank leaching test will tell us the actual release rate of contaminants. This will help us arrive at a trustworthy conclusion about the true influence on the environment.

Y. Nathan: Leaching tests on bottom ash indicate that the material is inert. Leaching tests conducted on fly ash indicate a non-hazardous material. There are practically no limitations on bottom ash utilization. We need to reconsider the restriction radius (for coal ash used in infrastructures: the maximal distance from water wells), since the monitoring of groundwater in a bottom ash application site at Zikim showed that elements released from the bottom ash had no effect on the groundwater.

G. Reshef: I will present the results of the work we did in 1996, along with Meni Ben-Hur and Rami Keren. I think this was one of the first attempts done in Israel to evaluate the effects of fly ash and soils on trace elements leaching. We tested Colombian fly ash with three soil types: Loess, Red loam and Vertisol. We used these different packages: soil only, soil and ash in equal amounts in two different configurations – in one, we mixed them in a homogenous manner and packed them in columns, while in the other we took a layer of ash and placed a layer of soil beneath it. Then we tested: pH, EC and trace elements content in the leachate after using 2.5 litres of water for leaching.

These are the pH test results, a parameter that is most critical in controlling solubility and concentration of trace elements in a leachate. An intermediate pH was found in the mixed (ash-soil) columns, since that pH was first of all based on the fly ash’s pH, but lower due to the attachment of hydroxyl groups to the clay and aluminium and iron oxides.

I will now present two slides showing the concentration of elements in the leachates from the different configurations. One shows the elements lithium (which is a cation), chromium and molybdenum (which are oxy-anions). A sharp decline is originally evident in the elements’ concentration in the leachate. Only after leaching with 200 ml, the decline became more gradual, with a concentration of less than 2mg/liter (for leachates of all configurations). The second slide shows the elements boron, vanadium and selenium, and a different picture emerges – an increase in concentration at the start of the leaching and then a decrease to the same levels exhibited by the columns containing layered configurations. Vanadium behaved differently from the other two (at the time we did not look into the reasons for this).

We also calculated the masses of elements leached in the different column configurations. Then we translated these results into an enrichment factor, which we calculated as well. The mass leached from the layered configuration column was lower than the total mass leached from the soil and ash columns combined (in percentage). In the mixed configuration, boron, vanadium and selenium showed an increase in the leached mass (relative to the soil column and ash column combined). For lithium, molybdenum and chromium in the mixed configuration columns, the mass of the leached elements was lower (again, in comparison to the above).

Conclusions: Adding fly ash to a soil will naturally alter its chemistry, including pH; these changes will increase the solubility of trace elements originating from the ash. The amount of microelements leached from the ash/soil columns (whether mixed or layered) was higher than that from the soil only columns. We found that boron, vanadium and selenium in the mixed columns were positively enriched; this is due to the relatively low pH of the solution surrounding the ash particles in the mixed columns. Conversely, we found a negative enrichment factor for chromium, molybdenum and lithium in the mixed columns. As for the layered columns, we found negative enrichment factors for all the elements tested.

M. Ben-Hur: One way of preventing the exposure of fly ash in embankment applications to rain is to cover it with a soil layer (as was done at Jisr a-Zarka). Surface runoff over the fly ash-loaded area was higher than over the control area (implemented using gravel). The concentration of boron in surface runoff over the covered ash was higher than in the control area. The high concentration of boron in the layer above the ash may be explained in several ways: for one, the capillary movement from the ash to the soil during dry periods, and subsequent accumulation there; two, the mixing of soil and ash as a result of subterranean fauna activity; and three, the collection of Boron by the roots of plants, its subsequent transition to their leaves, and finally its release to the soil when the leaves die and decompose, mixed with the soil and enrich it with boron. To confirm this argument we tested the leaves and stems of plants taken from the areas and found that boron, molybdenum and barium concentrations were indeed much higher in plants taken from the fly ash area than they were in plants taken from the control area. However, the situation was reversed for manganese and nickel. We know that boron, molybdenum and barium are very soluble in fly ash, and this may be the reason why their concentration in the plants was so high. We know that Manganese and Nickel have low solubility in fly ash, which may be why these elements behaved differently.

R. Keren: This is the tractor that exposed the fly ash layer during the field test conducted along road 6 (Revadim) in which the embankment's permeability to water was measured. I did not measure water permeability at this point. As you can see in the photograph, the tractor was incapable of penetrating the ash layer due to its extreme hardness, akin to that of stone. We even heard the noise, as if we were driving on asphalt, not on some soft material. The road was paved according to the standard specification. The fly ash was packed, moistened and compacted according to the contractor's recommended proctor value. Thus, its density was rather high from the start.

There was a decrease in water permeability rate, due to changes that occurred in the fly ash layer. These changes resulted from interactions with carbon dioxide along with the water, and due to pozzolanic processes. The greater is a material's density, the less free space is found within it that may be subsequently sealed.

I measured the water permeability rate along the Jisr a-Zarka embankment's slope, and found that after 3 years, the embankment was totally sealed (just like stone) near the paved asphalt layer. As I descended down the slope, the water permeability rate increased; it seems that this is due to faulty compacting of the fly ash layer's edges along the slope.

The embankment's relatively high water permeability rate found in a test conducted 4 years after its paving is in seeming contrast to the nearby area's general tendency toward total sealing. This is due to several reasons: the absence of water inside the layer of ash (the water could not penetrate this layer due to the presence of the heavy clay and sand layer above), the meager amount of rainfall in the area, and the vegetation that grew in the area, consuming significant amounts of water. This in essence protects us from the risk of contaminants leaching from the ash there. For if the water does not penetrate the fly ash layer, there is no need at all for regulation there, since nothing will leave that layer. And even if the water penetrates the ash layer, it will remain sealed there due to the solidification of the layer. So I feel that we are on the safe side.

In selecting the method for choosing the standard for road embankments, if the system at hand is a mass that is impervious to water, then it should be treated as a monolith, not an aggregate.

Y. Deutsch: The Jisr a-zarka site is effectively a large scale laboratory for examining the behaviour of coal ash throughout time. Three drills were done at a distance of approximately 1 meter from the road pavement above the embankment, on the soil cover. The third drill, which was done 8 years after the embankment was created, was disrupted on account of the resistance of a hard layer of coal ash. Its resistance was so high, that it simply tore the drilling rods and head; these components, it should be remembered, are made of steel and have a diameter of a few cm. The engine supplied sufficient drilling power, but the metal was insufficiently strong. This corroborates what was said earlier, that we have obtained a monolith, something as strong as a rock, not merely a sparse layer of ash. These findings lead to the conclusion that the embankment is active, but not active as pertains to environmental issues, but to become more inert.

We tested a total of 21 chemical elements in three drills, trying always to drill in the same area. This was to ensure that a faithful representation of the elements' status would be obtained. I presented two elements here, Be and V, and it is evident that they exhibited no change in concentration through time. It is true that towards the edges of the section profile, we see near the surface and deep beneath it, that the elements content decreases as one approaches the soil, but there is no significant change in concentration between the different drills. If we take the element Be in the road embankment, we see a certain concentration in the year 2000 and a slightly smaller concentration in 2007 – i.e. a gradual decrease. In contrast, in the bridge embankment, which is comprised of fly ash, the concentration in 2000 is in the middle of the range of concentrations measured then in 2003 it is nearly the same or a bit smaller than in 2007. Effectively there is no change in concentration. The same results were found for V. The remaining elements act more or less the same, showing some slight variation but nothing more.

The extent of the elements' leaching from the fly ash was tested in a TCLP test. We can see that 3 elements were actually leached: Se, B and Mo. The concentration of Se and Mo was 8 ppm and 10 ppm in the bridge embankment, and 3 ppm and 5 ppm at the road embankment, respectively, these are low values from the start. It was the element B alone that had a high concentration, between 200 and 500 mg/kg. We also discovered a new mineral that was created in the embankment – Stilpnomelane. Are the embankments totally inert? I do not know. But to summarize, there was no mobility of cations and oxy-anions, a new phase was created and the material hardened.

A question was raised here, whether coal ash behaves as a monolith or as a powdery material in embankment or other applications. If it behaves like a monolith, then it should be tested as such. Throughout the years our regular tests were done for a powdery ash according to TCLP. Lately we made a transition to the European method EN 12457 in testing granular materials, following the recommendation of the professional scientific team. It was suggested to test fly ash as a monolith using the EA NEN 7375 method. If these methods show a measurable correlation, we will need to find their transfer coefficient and use it in the simple test for powdery ash as representative of a monolith. The granular method (EN 12457/2) is fairly straightforward: place it, shake it, remove it and measure it. In the monolith testing method, which lasts between 4 to 64 days, this procedure is repeated 4 or 8 times. The fly ash samples were compacted into cylinders at one of the Israeli Institute of Standards' laboratories, using the standard method (56 percussions) and were cured for 28 days under fixed humidity conditions (in order to quicken the pozzolanic hardening process). For two additional months the cylinders stood in the open air inside one of the Israel Geological Survey laboratories. Finally, cylindrical slices were cut from the masses. Two of the samples, the Colombian and Indonesian ashes, crumbled within hours of placing them in water. The other three, South African, Russian and Australian, managed to maintain integrity. But the Australian sample suffered some crumbling and cracking. The fissures that developed in the cylinders caused some problems: A) What is the true surface area of the samples? We could not measure this, since the areas inside the fissures were also in contact with the water. B) Some ash samples lacked stability and crumbled, even after a long period of curing.
In comparing the concentrations yielded in the granular method (microgram/liter, ppb) and the monolith method (mg/m^2), no consistent correlation was found.

R. Keren: I wish to comment on the monolith testing method. Firstly, I should define the system; monoliths may be characterized by two kinds of surface areas: one, a geometric area, calculated based on the radius – but this is not the relevant surface area for elements solubility. The necessary parameter is the true surface area, which shows "hills and valleys" when observed under a microscope. This true surface area is much greater than the geometric area. We should remember this in evaluating the surface area and its applications in the real world. Earlier we discussed an alternate method for determining surface area – the BET (Brunauer-Emmett-Teller) method. I believe this method could give us better results.

H. A. Van der Sloot: From the available information and the input of Rami Keren on the hardness of the ash embankment and that of Yoetz Deutsch on the breaking of the drilling equipment, I can say that the material has considerable strength, and one would not expect any significant interaction with the environment.

The key issue here should be to establish whether the material in question is a monolith or not. The question is where is the transition point between a granular material and a monolith? And coal fly ash can be a tricky material in this regard. If you state that the monoliths you created crumbled during their testing, this can serve as an indication to the transition point. You no doubt learned that the exposure conditions used in the tank test are in a certain regard extreme, since the material is exposed to great quantities of water. An alternative method is to test the material using the compacted granular leach test (CGLT); this test is designed for materials that act as monoliths in application, since water will not penetrate them, but will rather run along their surface downward. Hence, the material can be tested as a compact granular monolith, and leaching, in this case, should be conducted from one side only, the side defined as the exposed side. The stages of this test are identical to those of the tank test: it has the same leaching process, everything is virtually identical, and the material does not crumble. I am not in favor of the BET test; let us stay with the geometric surface area. This issue was debated in length. Even if the material's surface is somewhat irregular, this phenomenon has no significant effect on test results.

R. Keren: I whole heartedly agree with you on this point, but our objective is to compare between different types of fly ash. If we take several granular materials that are compacted in different ways, we will receive wide ranging test results, and, accordingly, their true surface areas will differ. So in comparing between ash types, the difference between the true and geometric surface areas could be a critical factor.

H. A. Van der Sloot: In the CGLT method, the granular material, clay for example, is placed in a water tank, and then compacted within its packing configuration. Then a layer of glass beads (0.5 cm each) is put on top of it. Water is replaced like in the tank test (as concerns the leaching steps), and care is taken to ensure that this layer remains undisturbed. In this manner we maintain the gradient developing over time. This works perfectly and provides very consistent results.

D. Weinberg: First of all, I should say that we have seen here many things and learned quite a lot. However, the issue of utilizing coal ash in various environmental applications should be examined. That is, if we compare its utilization in quarry land filling, quarry renovations, or in other infrastructure or agricultural applications, we should remember that there are, aside from the economical aspect, environmental and health management aspects to take into consideration. We should measure all applications using these criteria. Sooner or later, trace elements will reach the environment. So every atom of Molybdenum and Cadmium will reach the environment. The question of applications rises – if we consider water well drillings, then the issue of restriction radii is entirely irrelevant, since they are based on the mortality of micro-organisms within the soil. It was said that the metals in question originated from the ground and will return to the ground, but Cadmium reaching the ground will remain there, and eventually, during my time or during the time of my descendants, reach the well water. For me, it is important that well water quality is preserved.

On the other hand, if fly ash is applied in agriculture, massive quantities of the trace metals will reach the environment, and will also be contained in dust – inhalable dust. Such issues involve serious health aspects that must be taken into consideration. I think that before we discuss this solution or that, we first need to examine in depth the long term aspects, and in particular the long term health aspects. This will ensure that we preserve a good and healthy environment.

U. Mingelgrin: I wish to refer to the assertion that great quantities of metal are introduced into the environment. The issue here is not only that the metals came from the environment and are returning to it. The critical parameter is their release rate. It is true that in infrastructure applications we introduce large quantities into the site. But the aquifer water is not stagnant. If the release rate is sufficiently slow, even if in the distant future all the metals reach the groundwater, the matter of dilution must still be considered. Whether it takes a million years for the metals to reach the groundwater or a single year, has a major effect on public health. I think that you did not address this point.

D. Weinberg: The topics discussed here are ambiguous. The studies presented did not render a clearly defined picture of the level of safety, and certainly not a nation-encompassing picture portraying the effects such applications may have on the environment.

U. Mingelgrin: Two lectures demonstrated that the applications involve a monolith that is impervious to water. So it is certain that there will be no considerable diffusion from most of the ash. And, I wish to emphasize again that I am referring to road infrastructure. Other applications could indeed be very different. So the very fact that the ash behaves like a monolith and water can barely pass through it, proves that release of metals will be very slow.

R. Keren: Ash used in infrastructure applications can be considered, for all practical purposes, a virtual container from which nothing will leak. In most cases it becomes impervious in a relatively short period. Anything coming with the water should leave with it, in any porous medium. In this case there are two effects: one, the non-permeability of the material, which indicates no water motion and, consequently, the retaining of all microelements within the mass. Second, if there is indeed a leak, it will be a flow through preferred paths of the elements toward the groundwater, characterized by a protracted delay in the dispersion of the metals in the soil. The elements transition rate is dynamic, as is the flow of groundwater in the aquifer. Hence the dilution effect will be strong and no real change in water quality will be evident. In the Jisr a-Zarka embankment, where the groundwater is very shallow, tests conducted by Dr. Yaacov Nathan and Dr. Ariel Metzger showed there were no changes in the groundwater’s heavy metals concentration. Ergo, all hitherto accumulated test results are proof that there is no risk of health hazards, as David Weinberg claims.

P. Fine: Additionally, Yoetz Deutsch demonstrated the re-mineralization and re-crystallization processes that are taking place in the embankment. So in essence the minerals there will be less soluble and the release of trace elements, heavy metals and others will be even smaller.

Y. Nathan: It is not only the introduction of fly ash into a system into which water cannot enter. The question is will the water’s new composition be identical to its former one? Will it be of higher or lesser quality? This is the real issue; it will not suffice to examine whether certain elements are introduced into the water, even if those elements are toxic. How much water (precipitation) is added to the system? What is the amount of the elements that are added to the water? Will their concentration in the water change, or not? These are the questions that should be asked, for we are dealing here not with a fixed system, but rather with a dynamic one, and the only question is: what are the concentrations of the elements in the groundwater? We presented analyses of groundwater samples taken from beneath the Jisr a-Zarka embankment and from the control well; these showed that the water from beneath the embankment were actually purer than unaffected groundwater. I will not jump to conclusions here. All I am trying to say as that we need to calculate not only the amount of toxic elements or others in the groundwater, but also the amount of rainwater that penetrates the embankment.

R. Keren: Most of the fly ash used in road embankments is covered by asphalt, so water will not penetrate there in any case.

D. Weinberg: But are you looking at short term effects only, or long term effects as well?

R. Keren: In any range.

H. A. Van der Sloot: I think that the problem you are trying to solve is one met by all regulators: what is acceptable? And the key question, which I believe most are familiar with by now, is that the value “zero” does not exist, but that there is a certain level that does not present a risk. We should compile the data we have in order to justify the implementation of certain applications. In Europe, and especially in the Netherlands, we have developed a regulation for the usage of alternative materials in construction. Europe is moving in this direction as a whole, using CEN/TC 351 activities (the European Technical Committee for regulating the release of contaminants from construction products to the environment) for the essential requirement no. 3 (of the CPD directive for construction products, dealing with environment, health and hygiene). This regulation looks 100 years ahead, since such processes do not cease after a year or two. We have much to learn from each other, and from the experience accumulated. I think the regulating authorities will benefit from seeing the status of things, their ongoing development and the subsequently developed approaches.

G. Reshef: I am talking about the differences between susceptibility zones A, B and C. How do you quantify the parameters you mentioned, such as soil adsorption and climate conditions, or any other factors that can affect the ash’s physical-chemical properties? Zone A refers mainly to the mountainous aquifer, were groundwater is very deep. Zone B refers to the coastal aquifer, but we consider it as zone A. This might lead to your changing your recommendations on environmental conditions.

U. Mingelgrin: We might need to change this recommendation in light of what you raise, Dr. Reshef. You propose that as far as adding fly ash, zone B might be more problematic?

G. Reshef: Yes. Zone B, which is the coastal area, has a greater potential for groundwater contamination. In contrast, the groundwater in zone A is 300 meters below surface.

H. A. Van der Sloot: I have a comment on the classification of materials in a certain category. Classification as a rule is based on the results of tests conducted on the material. Materials should not be classified under rigid categories. Every test result can produce viable applications, taking into consideration those applications’ criteria. One type of quality allows implementations under defined conditions, and another under other conditions.

U. Mingelgrin: If I understand you correctly, you are suggesting that conditions be entirely site-specific. In other words, no more referencing of conditions to general definition of susceptibility zones, but rather using a certain model to forecast the effects of each specific site. And these results will serve as a basis for the determination what is allowed and what is not. Is this correct?

H. A. Van der Sloot: Yes.

Y. Nathan: We have in our possession the data of 8 years monitoring at Zikim site, where a bottom ash layer was placed upon a sandy soil stratum. The results are unmistakable: there is no change in the groundwater at all. This shows that placing bottom ash at a certain site does not affect the groundwater there. Like we assumed in the past, I think we should adopt this in other fly ash applications, (since we consider it a monolith) and only by measuring the quality of the water that passes through this monolith. This will be the only way to ascertain the effect on the water – whether it will be good, bad or neutral – in comparison with other applications lacking fly ash monoliths.

P. Fine: As concerns the susceptibility zones discussed by Mr. Reshef, I think we do not need to invent the wheel all over. Israel's most vulnerable conditions might be less vulnerable that those of the Netherlands, since the groundwater here is much deeper, the soil is dominantly calcareous etc. In the Netherlands the soil is generally more acidic and sandy, and the groundwater is shallower. Therefore, if it is considered safe to use fly ash in infrastructures there, it surely should be safe to do so here. We just need to establish proper regulations, not invent anything new.

U. Mingelgrin: Yes, but I am not sure the groundwater in the Netherlands used as drinking water. What if their drinking water comes from the surface?

H. A. Van der Sloot: No.

P. Fine: I have no doubt that their concern for the environment is no less that ours.

U. Mingelgrin: I would like to summarize this meeting and note that we have here several important statements that may serve as the beginning of another discussion. We will need to use what we learned here as a basis for further deliberations, before we can finalize our recommendations in the scientific advisory committee.

Appendix 2

Extraction of granular material using NEN 7347 method
CGLT: Compacted granular leach test
General review[6]

This test is used to determine the leaching behavior of inorganic compacted granular materials under conditions where the leaching is mainly diffusion controlled. The test provides the result of the cumulative emission from the effective area exposed to leaching in mg/m^2. The purpose of the test is to simulate the leaching mentioned above, from a compacted granular building and waste materials in an aerobic environment, as a function of time over a period of 64 days. The test is based on the same principles and procedures as described in the diffusion test for moulded and monolithic materials according to NEN 7375. Generally, a test conducted on ash, is recommended after a curing period of it, in order for taking place of sealing and aging processes, which represent the long term effects.

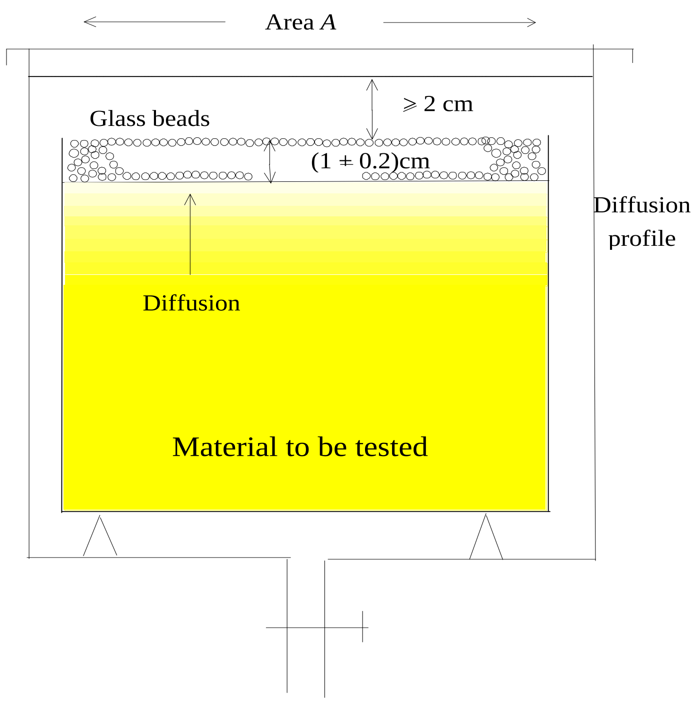
The sample is compacted in a plastic (inert) cylinder vessel while saturating it with water, and then exposing it on one side to the leaching solution (demineralized pH-neutral water with a conductivity of max. 1 μS/cm). This inner vessel is inserted into an outer plastic vessel and filling it with the solution, which is replenished in a predetermined time periods (see below- leaching steps). The sample is compacted to an approximately of a distance of 1 cm below the inner vessel upper edge, and covering it with a layer of glass beads in order to avoid disturbing the compacted bed when changing the solution between extraction steps. The outer vessel must be fitted at the bottom with a drainage point in such a way that the eluate can be drawn off without disrupting the diffusion profile in the sample.

As is done for monolith test according to EA NEN 7375, the CGLT method is also carried out in leaching steps: eight for characterization test over a total cumulative time of 64 days and four for compliance test over 4 days, at a temperature that may vary between 18°C and 22°C. at the end of each step the eluate is drawn off and checked for trace elements, pH and conductivity. The value of the pH says something about the alkalinity of the sample. The course of the pH during the test gives an indication of the stability of the sample. Great variations in the pH of the eluates indicate that the sample is not yest in equilibrium, or is not yet stable. The conductivity gives information on the salt load in the eluates, which is important for the analysis. After drawing off the eluate the vessel is

[6] Based on the document: NEN 7347. Netherlands standard: Leaching characteristics of solid earthy and stony building and waste materials-leaching test – Determination of the leaching of inorganic components from compacted granular materials.

refilled with water with the same volume, while maintaining an accuracy of 15 min the refreshment times.

In the next page a schematic diagram of the test set-up:



For each element the measured leaching per fraction is calculated with the formula:

$$E_i^* = \frac{C_i \times V}{f \times A}$$

in which:

E^*_i is the measured leaching of a component in fraction *i*, in mg/m^2;

c_i is the concentration of that element in fraction *i*, in µg/l;

V is the volume of the eluates, in l;

f is a conversion factor (f = 1000), in µg/mg;

A is the effective area of the sample exposed to leaching, in m^2. The area *A* of the sample is equal to the area of the internal diameter of the inner vessel.

For each element to be tested the measured cumulative leaching ε is calculated in each of the periods $n = 1$ to N, where the period $n = 1$ lasts from the start of the test to the first refreshment time (includes the fraction i = 1), period $n = 2$ from the start of the test to the second refreshment time (includes the fractions 1 + 2), etc. this calculation is carried out according to:

$$\varepsilon_n^* = \sum_{i=1}^{n} E_i^*$$

for $n = 1$ to N

in which:

ε^*_n is the measured cumulative leaching of a component for period n comprising fraction $i = 1$ to n, in mg/m^2;

E^*_i is the measured leaching of that component in fraction i, in mg/m^2;

N is the number of periods, equal to the number of specified refreshment times ($N = 8$).