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# Environmental Evaluation of Materials Used for Cold Recycled Mixes

Final report

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Coordinator: Czech Technical University in Prague (CTU)
Partner 1: University of Kassel (UK)
Partner 2: University College Dublin (UCD)
Partner 3: Laboratório Nacional de Engenharia Civil, I.P. (LNEC)
Partner 4: Wirtgen GmbH

# CEDR Call2012: Recycling: Road construction in a post-fossil fuel society

# CoRePaSol

# Characterization of Advanced Cold-Recycled Bitumen Stabilized Pavement Solutions

# Environmental Evaluation of Materials Used for Cold Recycled Mixes

Final report

## Deliverable 5.1

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**Author(s) of this deliverable**:

Amir Tabaković, University College Dublin

Ciaran McNally, University College Dublin

Aoife Quinn, University College Dublin

Fatima Batista, Laboratório Nacional de Engenharia Civil

Anabela Maia, Laboratório Nacional de Engenharia Civil

Jan Valentin, Czech Technical University in Prague

Michael Engels, Wirtgen GmbH

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# Executive Summary

The use of cold-recycled materials as part of road management strategies is becoming increasingly important due to environmental, legislative and financial pressures. The CoRePaSol project is seeking to develop solutions that will promote cold recycling technologies by focussing on key technical challenges, such as material conditioning and testing, life cycle analysis etc. This report offers guidance on the environmental evaluation of materials used for cold recycled mixes.

It is widely known that for a significant period of time, coal tar was the dominant binder used in road construction. This practice continued across Europe until the 1990's, at which point it was prohibited for environmental and health reasons. The most significant of these is the presence of polycyclic aromatic hydrocarbons (PAHs) in the coal tar and the carcinogenic nature of these compounds. This has led to the introduction of bituminous binders for new road constructions, but there still exists many European roads where tar is the active binder. These have often been overlain with new constructions, but the original tar material is still present. These materials cannot be hot-recycled as they will generate fumes which violate health and safety directives. A key challenge for the CoRePaSol project is to assess the ability of the cold-recycling process to stabilise tar containing materials by encapsulating the tar material within a new bituminous matrix.

Similarly, there is increasing pressure for the road construction sector to provide a waste management solution for other sectors by incorporating industrial by-products within the road construction. A challenge for CoRePaSol is to assess whether materials such as crumb rubber or fuel ashes can be used in cold-recycled mixes without any adverse environmental impacts.

When making these assessments, the research must also allow for the practical nature of the cold-recycling industry. It is intended that these results will be taken on by industry, and as such it is important that industry-recognised testing processes are used. The environmental assessment has sought to make use of established test procedures where possible.

To address these challenges a number of specific actions were initiated:

- A review of the available test methods was conducted and the most suitable test methods were assessed. Key parameters in this assessment included the ability to assess leaching of organic/inorganic contaminants; test duration; sample preparation; status of test method. It was found that there were no ideal tests that meet all of the requirements and instead a series of promising test methods were used. These were:
  - Batch test for granular waste materials
  - Up-flow percolation test
  - Tank diffusion test for monolithic samples
- A series of samples materials were identified and these were used to manufacture cold-recycled samples for assessment. The primary focus was on the leaching of PAHs from tar and materials were sampled from the Czech Republic and a site in Germany where tar was cold-recycled over 20 years ago. Material was also obtained from a current cold-recycling project in Ireland. Assessment of leaching arising from the use of crumb rubber and fly-ash was also included.

- Materials were tested before and after the cold-recycling process. This represents a significant challenge in the material assessment as before recycling, the aggregate is a loose granular material. After cold-recycling the material has been transformed to a compacted state, with many of the finer particles closely held together by a bitumen-cement matrix.
- Eluates from the leaching tests were then analysed for leaching of key contaminants. PAH concentrations were assessed using GC-MS, whereas inorganics were assessed using ICP-AES and/or ICP-MS.

Based on these various testing programmes, a number of comments were made on current state of the art with respect to environmental assessment of cold-recycling practices. The most significant issue was found to relate to the absence of a suitable testing methodology for testing cold-recycled materials. It was found that there was no single test that satisfied all of the criteria, which creates uncertainty in interpreting results.

It was however observed that there was an encapsulating effect associated with the cold recycling process. The reliability of this effect could not however be confirmed due to the nature of the testing methods - some element of sample crushing is usually required. The absence of a suitable test that meets all requirements has been found to be a significant shortcoming and the development of such a test should be a focus of future research work.

Samples were obtained from a German project that involved the cold recycling of a tar containing material in 1991. Leaching data was obtained and this was compared to the current leaching behavior. The leaching levels are largely similar to what was observed in 1991 and significantly lower than the assessment of tar material (prior to recycling). This suggests that the encapsulation effect has been effective.

It was observed that the highest levels of PAH leaching were associated with the Irish recycling site, where tar was not expected. For this particular site there was no information available on the material being recycled, which highlights the value of preliminary screening in advance of any recycling work. This may involve the use of relatively simple tools such as the PAK marker etc.

The percolation test was found to be useful in identifying PAH leaching, but it was also found that after the specified L/S ratio of 10 l/kg had been achieved, the tests in some cases may have not yet reached equilibrium. This may be due to the high hydraulic conductivity of the samples, which leads to reduced contact time between the sample and the leaching water. This suggests that, depending on the material, longer testing times may be required (currently about 28 days). Nevertheless in some countries shorter testing durations might be required.

This tank test was also employed to assess leaching associated with the use of fly-ashes. It was found that the tank test is sensitive to changes in the material composition and suggests that it is suitable for assessing the leaching of inorganic compounds in cold recycled applications. It was also found that selection of appropriate fly-ashes is of importance; in this study the addition of biomass and municipal waste to the coal resulted in an ash with higher leaching levels.

# 1 Introduction

In the asphalt recycling process, the environmental properties of recovered materials must be considered, in order to ensure that there are no potentially harmful constituents present in the mix (no leaching of toxic constituents, dust particles which might cause air emission concerns, etc). Concerns over potential pollutant leaching from reclaimed asphalt (RA) come from various sources (Brantley and Townsend, 1999). Bitumen is a derivative of petroleum containing different types of hydrocarbons. During its service life, it comes in contact with many chemicals generated from traffic, such as vehicle exhaust, gasoline, lubricating oils and metals from tyres brake lining wear. The major chemicals typically contained in asphalt pavement are heavy metals (Cd, Cr, Cu, Ni, Pb and Zn) and Polycyclic Aromatic Hydrocarbons (PAHs) (Legret et al., 2005; Brantley and Townsend, 1999). Traffic related sources of PAHs include vehicle exhaust, lubricating oils, gasoline and tyre particles (Sadler et al., 1999; Takada et al., 1990). Legret *et al.* (2005) studied the leaching effects of heavy metals and PAHs from RA. They reported that pollutant leaching is weak and that the concentration of leaching remained below EU limit values for drinking water. However, the study also showed that in comparison with new conventional asphalt, the concentrations of total hydrocarbons and some PAHs were higher in leachate from RA. Legret *et al.* concluded that environmental tests, such as batch and percolation test (EN 12457-4:2002; CEN/TC351 N 0272), can be used for assessing pavement materials. They did however suggest that field experiments should also be considered to allow for scaling and hydrological conditions such as infiltration.

Another significant cause for concern is the presence of tar which was for a period used as a binder in pavement construction. That practice is for at least two decades prohibited across Europe due to the presence of increased concentrations of harmful PAHs and coal tar is classified in the CERCLA (Comprehensive Environmental Response, Compensation and Liability Act) National Priority List (NPL) of Hazardous Substances (CERCLA 2007). Despite this there are still potential sources of tar within the European roads network. This might by particularly the case in Eastern Europe and Germany where tar was used into the beginning of 1990’s, and also in countries with legacy road issues. Nevertheless it might be also an important issue for the most developed countries, where tar was used widely in the past (1960’s and 1970’s) and is contained in older pavement structures which sooner or later have to be rehabilitated or modernized – this is a case of Germany, the Czech Republic etc. In these cases, there is often uncertainty as to what materials have been employed in previous road constructions, therefore there exists often a potential for presence of tar.

This study focused on investigation of the leaching effect from various RA materials including tar containing materials. The batch, percolation and tank leaching tests were employed in the investigation. Four different materials were tested, they are: i) Recycled Asphalt (RA) from Czech Republic, ii) combined RA and gravel from Ireland, iii) RA containing tar from Germany and iv) RA containing bitumen or mix modified by Crumb Rubber (CR) from Portugal. The effect of encapsulation of contaminants by foamed bitumen and/or bituminous emulsion in place cold recycling procedures was investigated.

# 2 Testing Procedures

## *2.1 Leaching test methods*

One of the main issues in assessing the leaching potential of pavement materials is the selection of appropriate test methods. Although the reclaimed asphalt should be seen as a product and not a waste, the methods for the assessment of release of dangerous substances from construction products are still under development (Enell *et al.*, 2012). This has long been recognised as a difficult issue and in 2005 CEN formed the Technical Committee TC351, entitled "Construction products: Assessment of release of dangerous substances". However this committee has still not finalised its work and the challenge of assessing leaching behaviour remains. To address this, a number of leaching tests were considered, including:

1. Batch test for granular waste materials, as per EN 12457-4
2. Batch test for soils, as per ISO/TS 21268-1
3. Up-flow percolation test, as per the method currently being developed by CEN TC 351 and per CEN/TS14405
4. Tank test, conducted on monolithic samples
5. Aggregates leaching test, as per EN 1744-3
6. Re-circulating column test, as used in the Re-Road project

In assessing the suitability of these tests, a number of parameters were considered. These include:

- Suitability for assessing organic leaching. A key interest in the project is the potential for the cold-recycling process to prevent future PAH leaching. As such, any selected test method should be capable of supporting this. This is a complex issue, as PAHs will attach themselves to the surface of many materials commonly used in test equipment set-up. To overcome this, wherever possible, the sample and leaching water should only come in contact with materials such as aluminium, glass, stainless steel or some specific types of plastics. In this respect it is recommended to follow the requirement stated by the Commission Regulation No. 836/2011 (19.08.2011) which states in C.2.2.2 related to specific procedures for polycyclic aromatic hydrocarbons that: “The analyst shall ensure that samples do not become contaminated during sample preparation. Containers shall be rinsed with high purity acetone or hexane before use to minimise the risk of contamination. Wherever possible, apparatus and equipment coming into contact with the sample shall be made of inert materials such as aluminium, glass or polished stainless steel.” It is however recognised that this is not always possible to achieve using standard test configurations.
- Test duration. Leaching is a diffusion driven process and as such there is the possibility that some tests will be quite long and unsuitable for a project of this duration.
- Equipment required. Some tests require expensive, non-standard equipment, and may not be suitable for wider testing programmes.
- Number of analyses required. Determination of PAH concentrations is usually obtained by gas chromatography – mass spectrometry (GC-MS) analysis which is

potentially expensive. Tests with a high number of analyses required are potentially very expensive.

- Particle size. Many of the available tests were originally developed for soils, and are designed for materials with a small particle size. Given that cold recycled materials can have aggregate particles larger than 30mm, some level of sample processing is often required and this will inevitably influence test behaviour.
- Standard test. Preference should be given to tests that are linked to European standards.

To differentiate between the various available test methods, the characteristics of each test were summarised. These are presented in Table 1. As these methods are assessed, it becomes clear that none are ideal.

**Table 1: Summary of potential test methods**

| Test Method | EN Test? | Max Particle Size (mm) | Duration (days) | L/S Ratio | No of Analyses Required |
|---|---|---|---|---|---|
| Batch Test: EN 12457-4[1] | Yes | 10 | 1 | 10 | 1 |
| Batch Test: ISO/TS 21268-1[2] | No | 4 | 1 | 2 | 1 |
| Percolation Test:[3]<br>- method currently being developed by CEN TC 351<br>- CEN/TS 14405 | No | 10 | ≈ 28-30 | 10 | 7 |
| Tank Test[4] | No | Monolith | 64 | - | 8 |
| EN 1744-3[5] | Yes | 32 | 1 | 10 | 1 |
| ER-H [6] | No | - | 7 | 2 | 1 |

1. The batch test according to EN 12457 Part 4 is a test that is developed for waste granular materials and requires the aggregates to be processed to a maximum particle size of 10 mm. While the specification of a particle size is useful, the maximum value is not representative of the recycling process. The test duration of 24 hrs is useful, but this is achieved by rotating a glass bottle filled in which the sample is held. This is not

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[1] Batch test: "Characterisation of waste — Leaching — Compliance test for leaching of granular waste materials and sludges"

[2] Batch Test used in Re-Road: "ISO/TS 21268-1:2007. Soil quality - Leaching procedures for subsequent chemical and ecotoxicological testing of soil and soil materials - Part 1: Batch test using a liquid to solid ratio of 2 l/kg dry matter"

[3] Percolation Test used in Re-Road: "CEN/TC351-N0272, 2010, Draft Generic horizontal up-flow percolation test for determination of release of substances from granular construction products. N0272"

[4] EA NEN 7375:2004 "Leaching characteristics of moulded or monolithic building and waste materials: Determination of leaching of inorganic components with the diffusion test"

[5] "Tests for chemical properties of aggregates —Part 3: Preparation of eluates by leaching of aggregates"

[6] Recirculating column test used in Re-Road; Danish Pre-standard; further info in: Gamst, J, Kjeldsen, P & Christensen TH. "Determination of solute organic concentration in contaminated soils using a chemical-equilibrium soil column system." *Water, Air, and Soil Pollution* 183.1-4 (2007): 377-389.

representative of the leaching practice in the field. Finally, the test can be used for assessment of organics as the all parts can be obtained from either glass or Teflon.

2. The batch test to ISO/TS 21268-1 was originally developed for soils and also has many of the disadvantages associated with the EN batch test above (unrepresentative leaching process). This test was also used during the Re-Road project (Enell et al, 2012) where concern was expressed over the small particle size and the potential for the leaching of PAH-H to be overestimated.
3. The percolation test, both according to the method that is being developed by TC351 and the method of CEN/TS 14405, represents a more realistic testing regime, in that the water flows through the sample. However the sample must be in granular form with a maximum particle size of 10 mm.

   The percolation test being developed by TC351 is suitable for organics, but the columns required are not commonly available. These must be made of either stainless steel or blown glass, and as such are quite expensive. The test duration is quite long (28 days) and requires a large number of GC-MS analyses (7 eluate samples per column, 4 replicates/columns).

   The percolation test according to CEN/TS 14405 (issued by TC292) is similar to the above described. This test was developed for waste granular materials. In this test the water up-flows through the specimen as well, which in many ways simulates field conditions (i.e. landfills) quite closely. For certain conditions (depending, among others, on the bulk density of the material) the test can also require a long period (about 30 days). It requires a large number of analyses of the constituents of the eluate samples (7 eluate samples per column and 2 replicates/columns) as well. It should also be noted that the indicated timing is a maximum duration, since the test could take less days depending on the permeability of the material (CEN/TS 14405:2004).
4. The tank test is the only test method that allows for monolithic asphalt samples. However the test duration is long (64 days) and a large number of samples are required (8 samples). The most significant drawback is that the test set-up is not suitable for organics, as some parts cannot be made using glass, stainless steel or other specific types of plastics.
5. The aggregate leaching test allows a large particle size and gentle stirring. It is assumed that this is due to the relatively fast leaching that takes place with granular aggregates. It is however unlikely that this will happen as quickly for reclaimed asphalt where the PAHs are required to diffuse into the water. As such, it is highly questionable if equilibrium is reached in this timeframe.
6. The re-circulating column test was also used within the Re-Road project (Enell et al. 2012). It was also developed for soils testing and has no requirement on aggregate particle size. The testing equipment is also quite specialised and not available within the project consortium.

From the above discussion it is clear that there is no ideal test available, and as such a compromise testing programme was developed. It was decided that:

- The principle test for use in this project would be the batch test to EN 12457-4. It is performed to a European Standard and has the capacity to assess leaching of organics.

- Some testing would be conducted using the percolation test as developed by CEN/TC 351 and CEN/TC 292, but this would be limited due to issues with the long time usually required and the costs associated with the high number of samples required for PAH and metals (heavy metals) analysis.
- The tank test would also be investigated. The use of monolithic samples is a major advantage, but similarly the absence of organic assessment represents a drawback. Nonetheless, this test was considered worthy of investigation, as it is the one that offers the closest set-up to real conditions.

## 2.2 Materials used

A number of materials were selected for use within this project. These were:

1. A reclaimed asphalt containing tar from the Czech Republic.
2. A reclaimed asphalt with some underlying gravel milled from an Irish recycling project. The original material was very mixed and derived from a number of sources. The sample used was from a single location, but trial pits have shown significant variation in composition at different site locations.
3. Core samples taken from a previous cold recycling of tar material in Germany in 1991. Only the layer containing the cold recycled tar material was selected for analysis and a sample core is shown in Figure 1. In this particular case the encapsulation effect of the previous cold recycling process can be investigated. Leaching data is available that describes the original material, and this can be used to benchmark the measured leaching behaviour.
4. A reclaimed asphalt containing Crumb Rubber (CR) from Portugal. It is worth to note, that in Portugal two main types of asphalt rubber (AR) mixtures produced with bitumen modified by high content of crumb rubber (bitumen-rubber) are used in road construction: GG-AR – gap-graded mixtures (produced with 0/12.5 mm aggregates, with a bitumen rubber content from 8 % to 9 % of the total weight of the mixture), placed as structural overlays and/or as wearing courses, with an air void content generally between 4.5 % and 6.5 %; and OG-AR – open-graded mixtures (produced with 0/10 mm aggregates, with a bitumen rubber content from 9 % to 10 %), placed as wearing courses, and with a void content generally between 12 % and 18 %. Taking into account that in the present case, the main objective was to assess the environmental behaviour of cold recycled mixes containing reclaimed asphalt with rubber modified bitumen, it was chosen for leaching testing the mixture where higher contents of rubber are used (OG-AR). In addition, the maximum dimension of the aggregates used in this type of mixture (10 mm) would be more suitable for the leaching tests to be carried out (batch and percolation tests), probably without need for crushing oversized materials.
5. Within on-going studies conducted at CTU in Prague, various types of waste fillers have been tested. In general the term waste filler refers to fines that originate during crushing of aggregates. Similarly these fines can also be formed by back-filling during heating of aggregates on asphalt mix plants. These materials in general have different mineralogy depending on the type of stone that is processed in a quarry. Some of these fines can have pozzolanic potential and therefore their milling and mechanical

activation can lead to a kind of '*active filler*' or '*intelligent stabilizer*'. These materials have been used as alternative fillers for cold recycled mixes; however leaching tests have not yet been conducted on mix options with such fillers.

6. Experiments were also conducted on the use of fly-ash originating from fluidized combustion. This material usually contains relatively high content of pozzolanic active components, which potentially allow using this material in civil engineering for several applications. For this reason it is possible to develop suitable approaches how exploit this by-product for preparation of binders/stabilizing agents and increase the raw material basis. In the Czech Republic several heat power plants and generation plants can be found which use fluidized combustion cauldrons since 1996. These facilities have a combustion effectiveness of 85% to 88%. The cauldrons performance is based on the principle of coal combustion at temperatures between 850°C and 950°C in a circulating fluidized layer which is composed by milled coal, desulphurization additive (limestone) and additive for stabilizing the fluidized layer (inert sand). The fluidized layer is formed by a dispersive system, which is created by gas flow in required amount in the bed of particles loose below the fluidized furnace grate. During the dissociating process the $SO_2$ released from the coal relate to $CaSO_4$ what has quite important environmental benefit. If it would be to the contrary, large amounts of $SO_2$ emissions would be released to the air causing significant pollution. This is one of the reasons why this type of combustion is used. Additionally lower combustion temperature also reduces the generation of $NO_x$ emissions. Resulting product can be defined as a mix of ash from the original fuel, untreated desulphurizing agent (CaO with eventual residue of $CaCO_3$), calcium sulphate ($CaSO_4$) and products of reaction between ash particles and CaO, as well as non-burnt fuel. With respect to the combustion temperature, which is lower for the fluidized process than for the granular combustion technique, the untreated CaO is presented in the form of so called softly calcined lime (about 30%). On the other hand fluidized combustion fly-ashes contain relatively higher amount of $SO_3$ (7-18%), which can cause in the binder formation of ettringite. For fluidized combustion fly-ashes absence of hot melt is typical as well. [20]. Important are as well chemical compounds containing aluminium. In fly-ashes from granular combustion (> 950°C) usually aluminium is mostly fixed in mullite, which is unable for any reaction during different types of high-speed milling. By contrast in fly-ashes from fluid combustion the aluminium seems to be present in roentgen-amorphous conglomerates (probably in metakaolinite). Therefore the hydration reaction is much faster comparing to regular pozzolanic reaction.

   To get a highly effective fly-ash based binder it is necessary to apply physical (mechanical) activation of this material. Fluidized combustion fly-ash with content of $SiO_2 + Al_2O_3$ larger than 50 % by mass is milled up to the maximum particle size 200 μm using a high-speed disintegrator equipped with different sets of rotors moving in opposite direction with radial velocity >160 $m.s^{-1}$. Material milled by this process is on its own reactive and it is not necessary to use another additive for stimulating hydration process. The first generation of this newly originated loose inorganic binder is called Dastit®, patented in the Czech Republic. Potential benefits of physical activation are savings in form of reduced energy demand necessary for production of traditional hydraulic binders.

## Grading curves

Each of the materials used in the study was investigated for grading, the results of which are shown in Figure 2, except for the reclaimed asphalt rubber from the Portuguese National Road, which is shown in Figure 3, since this type of material presented quite different characteristics (gap-graded mixture containing crumb-rubber, with a reduced maximum "original" dimension of the aggregates).

It can be seen that the RA material containing tar from the Czech Republic was of a very fine grading. In this case, the binder content was also determined as per EN 12697-39: 2012, and was found to be 6.1%.

The Irish RA sample was obtained from a road recycling project in Ireland. The depth of recycling was 300 mm and the bituminous component was approximately 2/3 of this (i.e. typically 200 mm depth). It can be seen that the material is coarser than the Czech RA, but still within the Wirtgen design grading envelope. The binder content in the material was not investigated, as for the recycling project it was assumed that the binder within the RA would remain inactive.

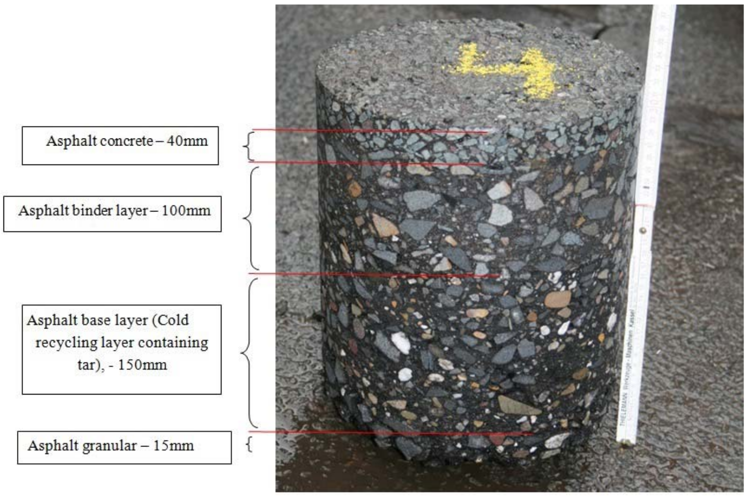

**Figure 1: Core sample with tar containing cold recycled layer - base layer**

The RA containing tar was obtained from cores extracted from a site in Germany. The cores were 250 mm in diameter and typically 290 mm deep. The cores typically contained a 40 mm surface course, a 100 mm binder course layer, over a 150 mm cold recycled layer. This is the layer of main interest as the original material contained tar before being cold recycled in 1991. The various core layers were separated by cutting with a circular saw. The fourth layer (asphalt granular) could not be cut off as it was very thin and loose; it was therefore removed by scraping the loose material with a straight edge. The core binder and base layers containing the tar were heated to 100 °C and hand crushed. The binder content and tar content were not further analysed for health and safety reasons associated with the binder burn off. Two material mixtures were required for the leaching tests:

- The cold recycled tar base course.
- The combined binder course (containing tar) and cold recycled base course (ratio 40:60).

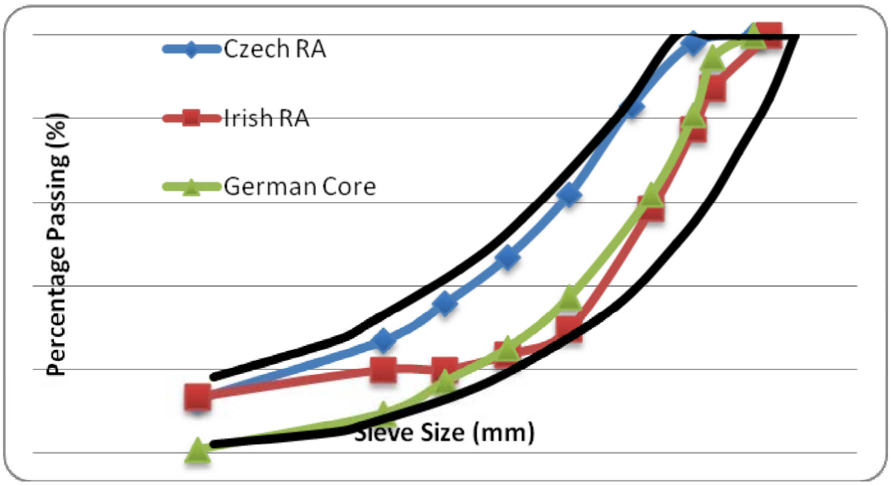

**Figure 2: Grading curves for the materials used**

The grading for the first of these materials is included in Figure 2. It can be seen that the grading is much coarser than the other RA materials; this is due to the fact that the other materials where processed using a milling machine, whereas the core was processed using laboratory equipment.

The RA containing crumb rubber (RAR) was obtained from milling the wearing course of a Portuguese National Road (EN 14), were an open-graded asphalt rubber mixture (OG-AR) was placed in a layer about 3-4 cm thick, during rehabilitation works in 2006. This Asphalt Rubber mixture was originally produced with 0/10 mm aggregates (Figure 3), with a bitumen rubber content of 9 % of the total weight of the mixture. The used bitumen rubber binder was produced by the wet process, i.e. bitumen modified with high rubber content (20 % of crumb rubber and 80% of 50/70 penetration grade bitumen).

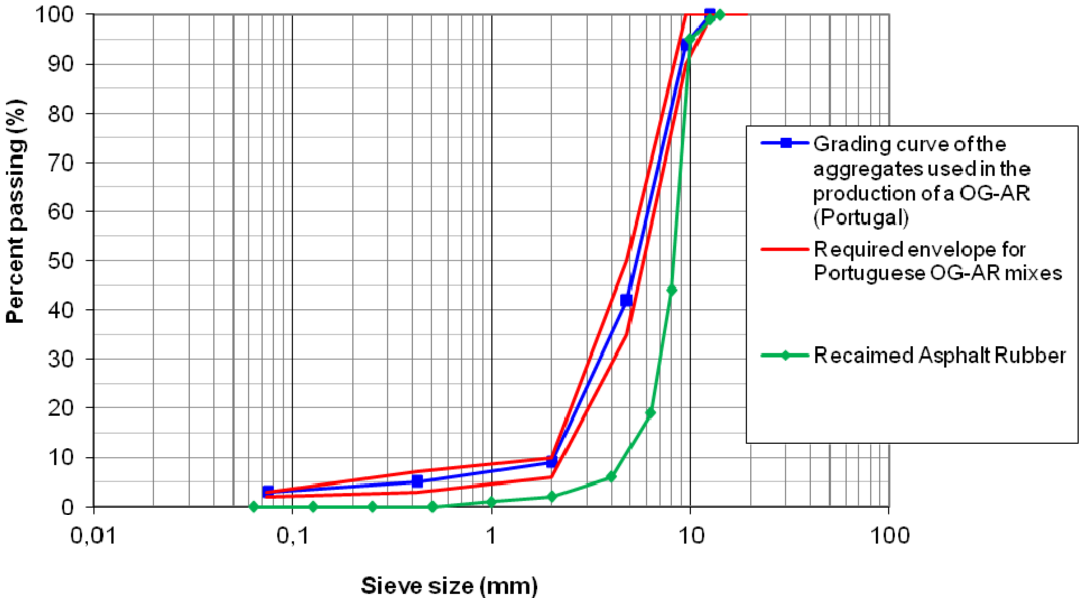

**Figure 3: Grading curve of the aggregates originally used in the production of OG-AR wearing course of a Portuguese National road**

In order to prepare suitable material for leaching tests (i.e. agglomerations of RAR not larger than 10 mm), samples of collected Reclaimed Asphalt Rubber were loosened and separated into smaller dimensions by placing them in an oven, at a temperature around 80ºC, and later manually handled and properly separated.

The processed reclaimed asphalt rubber (RAR) material used in the study was investigated for grading, the results of which are shown in Figure 3. Sporadic agglomerations exceeding 10 mm of larger dimension (in a percentage below 5 %) were rejected for further testing.

As it can be inferred by Figure 3, the Reclaimed Asphalt Rubber material used in the leaching tests can be considered as mono-granular material. Thus, compacted samples of this type of bituminous material would have a very high void content, even more if they are compacted at room temperature (about 20 ºC). In this case, it is very likely that they also would present a very high hydraulic conductivity.

In the study done in the Czech Republic using diffusive tank test application of selected coal combustion by-products (CCB) was assessed if used as binder or active filler in cold recycled mixes as an alternative to more common variations with bituminous binders and cement. In this case, the fly-ash from fluidized combustion is used as a substitute for the hydraulic binder. The fly-ash tested was subjected to mechanical activation in high-speed disintegrators while the parameters of the mix under scrutiny reflected the impact on the strength and deformation parameters of the mix; last but not least the water susceptibility indicator was also monitored. From the environmental perspective of practical applicability of the mixes a chemical analysis and some simple leaching tests have been done. More details to mix designs and evaluations done for fly-ash are given in Report D1.2. It should be only pointed out that the fly-ashes from the Hodonin power plant and the Plzen heating plant contain unusually small quantities of free lime; such fly-ash loses its self-binding ability and an addition of a certain quantity of lime or cement would be highly recommended in a certain stage of compacted mix production.

# 3 Preparation of Cold Recycled Samples

In assessing the impact of binder encapsulation on leaching behaviour, two different binder types were used: bituminous emulsion and foamed bitumen.

For the set of mixtures tested in UCD (Czech sourced RA, Irish sourced RA, German core), the target binder content for all mixtures was 3% and the moisture content was 4%. No cement was used in the manufacture of the samples as this could confuse the results of the leaching tests. These mix parameters were used as they were quite typical of what is currently used in day-to-day practice in Ireland. Using this process it was felt that test samples could be produced that would reflect site practices.

In the case of the leaching tests carried out at LNEC, the Reclaimed Asphalt Rubber was cold stabilised by adding to the mixture 5% of C60B5 bitumen emulsion (former C60B7 REC), which corresponds to approximately 3% of residual “new” bitumen and 2% of water.

## *3.1 Mixing procedures*

Two different mixing procedures were used depending on the material mix type, one for cold recycled mixes containing bituminous emulsion and one for cold recycled mix containing foamed bitumen.

### *3.1.1 Bitumen emulsion mixing procedure*

The cold recycled mixes containing bituminous emulsion were mixed using a drum mixer at a maximum speed of 30 RPM. The reason for the slow speed is that at higher mixing speeds the bituminous emulsion breaks down and the binder separates from the water, thus inhibiting adequate coating of the aggregates.

The following steps were taken during the mixing procedure:

i. dry aggregates are pre-mixed for about 1 minute (applicable only for combined compositions, e.g. combined binder course containing tar and cold recycled base course, in a 40:60 ratio);
ii. half of the water is added into the mix (when applicable);
iii. the emulsion is gradually added to the mix;
iv. the remaining water is added to the mix (when applicable).

In order to avoid breaking the water/binder bond in the bituminous emulsion, the cold recycled mixes were mixed for several minutes.

### *3.1.2 Foamed bitumen mixing procedure*

The cold recycled mixes containing foamed bitumen were mixed using a Wirtgen WLB10S foam mixing table. The mixing procedure took place in the Atlantic Bitumen (AB) laboratory, in Oranmore, Co. Galway. All constituents were prepared in the UCD laboratory prior to being brought to the AB laboratory.

The following steps were taken during the mixing procedure:

i. dry aggregates are pre-mixed for about 1 minute;
ii. half of the water is added into the mix;
iii. the foamed bitumen (produced with 70/100 bitumen) is added to the mix;
iv. the remaining water is added to the mixture.

After the mixing procedure, the mixes were placed into trays and placed into an oven for curing for 3 days at 50 °C. This curing procedure was adopted as none of the samples contained hydraulic binder (cement or lime); this is also implemented in other work packages (WP 1 and 2) where mixtures did not contain hydraulic binders.

Finally, some tank tests were conducted using different Czech sourced material (heterogeneous reclaimed asphalt, fly-ash, waste filler). A significant issue with the tank test is that it is designed to measure leaching of inorganic compounds, and cannot be used in its current form for assessing leaching of organics. This is due to the presence of materials in the test apparatus that can adsorb PAHs to their surface. As such, the focus was on leaching of heavy metals. Three materials were selected for this aspect of the testing, all of which were bitumen emulsion bound. They included a mixture with 3.5% emulsion and 3.0% cement CEM II B32.R according to EN 197-1; a mixture with 3.5% emulsion and 10% mechanically activated fly-ash from the Hodonin power plant and a mixture with 3.5% emulsion and 10% mechanically activated fly-ash from the Plzen heating exchanger plant.

# 4 Laboratory Testing

In order to determine the extent of potential environmental benefits associated with the use of cold-mixed bitumen stabilized materials by cold-recycling technique, namely their effect on leaching to local groundwater and subsoil and their long-term environmental compatibility, a full range of materials for testing was prepared, which are listed in Table 2. Three leaching tests were used for the determination of environmental effect, they are as follows:

- Batch leaching test;
- Percolation test;
- Tank test.

**Table 2: Summary of leaching tests conducted**

| Sample No | Sample Description | Partner Responsible |
|---|---|---|
| **Batch Testing** | | |
| B1 | Czech RA | UCD |
| B2 | Irish RA | UCD |
| B3 | German cold recycled tar base | UCD |
| B4 | German cold recycled tar base with binder course | UCD |
| B5 | Portuguese reclaimed asphalt rubber (RAR) | LNEC |
| B6 | Foam mix containing Czech RA | UCD |
| B7 | Foam mix containing Irish RA | UCD |
| B8 | Emulsion mix containing Irish RA | UCD |
| B9 | Foam mix containing German cold recycled tar base | UCD |
| B10 | Emulsion mix containing German cold recycled tar base | UCD |
| B11 | Emulsion mix containing Portuguese reclaimed asphalt rubber (RAR-BE) | LNEC |
| **Percolation Testing** | | |
| P1 | German cold recycled tar base | UCD |
| P2 | Portuguese reclaimed asphalt rubber (RAR) | LNEC |
| P3 | Emulsion mix containing Portuguese reclaimed asphalt rubber (RAR-BE) | LNEC |
| **Tank Testing** | | |
| T1 | Emulsion mix from Czech job site | CTU |
| T2 | Emulsion mix from Czech job site with 10 % mechanically activated fly ash (Plzen) and 3.5 % bitumen emulsion | CTU |
| T3 | Emulsion mix from Czech job site with 10 % mechanically activated fly ash (Hodonin) and 3.5 % bitumen emulsion | CTU |

It is worth mentioning which procedures were used for each of the selected leaching tests, as follows:

- Batch leaching tests were conducted both at UCD and LNEC according to EN 12457-4.
- Up-flow percolation tests were performed at UCD according to the method currently being developed by CEN/TC 351, and were conducted at LNEC according to the method of CEN/TS 14405. It should be noted that this Technical Specification has been developed in order to assess the leaching behaviour of granular waste, and allows the analysis of different elements of eluates samples. Furthermore, Annex C (chapter D.9 – Beneficial use of test results) states that "Results from the up-flow percolation test may be comparable to those obtained by carrying out the batch compliance tests parts 1 to 4 (..)", advising however that they may lead to different end results. Taking this into account, at LNEC, eluate samples collected from both leaching tests (batch and percolation) were analysed for the same elements (PAH and metals) for comparison of their results.

- Tank test were conducted at CTU, on monolithic samples, according to the principles defined for diffusive test in EA NEN 7375:2004. This test is used for monitoring of long-term water impact on test specimens with respect to chemical (content of analytes in the leachate) and mechanical leaching (content of loosen particles from the compacted test specimen). It is a static test to avoid the influence on natural diffusion and the deterioration of specimen's surface. The aim of the test is to simulate leaching of different compounds from a monolithic sample without avoidance of air ingress. The leaching is analyzed as a time function for a period of 64 days. The test set the character and properties of the material if stored in leaching fluid (demineralized water with neutral pH). The leaching fluid is supplemented in set time periods. The concentration of compounds which are leached to the water in particular consecutive parts of leaching and in time periods after 6 h, 24 h, 54 h, 4 days, 9 days, 16 days and 36 days. The pH value at which the leaching was done is influenced by the tested material. Based on the results of the diffusive test it is possible to calculate for each analyte's content which was leached per square unit.

## *4.1 Preparatory treatment of laboratory samples for leaching tests*

Before conducting both batch and percolation tests, materials were riffled to produce samples of approximately 1 kg and 10 kg, respectively for each type of the referred leaching tests. Each sample was then passed through a 10 mm sieve.

In the case of the materials tested at UCD, the aggregate that did not pass 10 mm sieve was placed in a freezer at -25 °C for 24 hours. At this point the material was fed though a jaw crusher and again sieved through 10 mm sieve in order to ensure that appropriate aggregate size was achieved. Four samples of each material were prepared for testing, three material samples and one blank (a control sample that only contained water).

In the case of the materials tested at LNEC (Batch tests: B5 and B11; Percolation tests: P2 and P3), the oversized material was below 5 % for both samples, allowing that sporadic agglomerations aggregate that did not pass 10 mm sieve were rejected for further testing. Therefore no further crushing was needed to be performed either in the "untreated" reclaimed asphalt rubber material (B5 and P2) or after being cold stabilised with bituminous emulsion (B11 and P3).

It should be noted that, prior to testing, samples of each material (B5, B11, P2 and P3) were collected and its water content was determined, being obtained, for all samples, 0.1 %.

In the case of the diffusive tank test the test specimens were prepared according to the requirements defined in technical specifications TP208 (see e.g. report D1.1 or report D1.2). Cylindrical test specimens of 100±2 mm diameter and 63±5 mm height were compacted and cured at room conditions and relative humidity of 40-70 % for at least 28 days. Before the tank test started, the volume of the test specimens is always calculated and the bulk density registered.

## 4.2 Leaching tests procedures

### 4.2.1 Batch leaching test (EN 12457-4:2002)

**Batch leaching tests conducted at UCD**

Approximately 100 g of each material per test sample is weighed and poured into the test bottle. The test bottle itself is glass with Polytetrafluoroethylene (PTFE) caps to reduce the risk of PAHs being lost to the surface of the bottle. A leaching solution containing 1 litre of deionised water and 0.01% $CaCl_2$ and 0.01% $NaN_3$ was prepared and poured into the test bottle. The $CaCl_2$ was used so that the leachate had an ionic strength mimicking natural rainfall and $NaN_3$ was used to inhibit microbial degradation of PAHs in of the test samples; this was deemed necessary in order to prevent water bacteria from damaging the pollutant levels within the test sample. Figure 4 (a) shows a test bottle with test sample and leachate. Due to the highly dangerous leaching substance sodium azide ($NaN_3$), a laboratory safety protocol was adopted in order to prevent any accidents and to ensure safety of the laboratory and its users.

**Figure 4: (a) Batch test leaching test bottle containing test sample and leachant; (b) Batch test rotating table**

The bottles were placed onto the rotating table as shown in Figure 4 (b) and the solution was agitated for 24 hours at speed of 12 RPM. As can be seen from Figure 4 (b), the bottles were placed into specially made plastic tubes in order to prevent spillage or lab contamination in case of bottle breakage during the test. A large cylinder was attached to the safety tubes in order to bring the speed of rotation into line with the test requirements.

At the completion of the test, samples for PAH analysis were stored at 5 °C, before being extracted with 20 ml of hexane using C18 extraction disks within three days. Following extraction into hexane, samples were sent to the laboratory for PAH analysis where concentrations in the extracts were determined by gas chromatography – mass spectrometry (GC-MS). The concentration of metals, such as: copper, zinc, nickel, manganese, cobalt, cadmium and lead is determined using technique such as inductively coupled plasma mass spectrometry (ICP-MS), inductively coupled plasma atomic emission spectroscopy (ICP-AES) or atomic absorption spectrometry (AAS).

### Batch leaching tests conducted at LNEC

Batch leaching test conducted at LNEC, at the Concrete, Stone and Ceramics Unit of the Materials Department, were performed according to EN 12457-4:2002, as follows:

- A test portion of 90 ± 5 g of dry mass was placed inside of a polypropylene bottle and ultra-pure water was added until a liquid to solid ratio of 10 l/kg was established;
- The capped bottle was rotated at the maximum speed indicated in the standard, 10 rpm, for 24 h using the equipment shown in Figure 5;
- After solids settling, the eluate was filtered over a 0,45 µm membrane filter and the conductivity and pH were measured;
- Two replicates per sample and a blank test were performed.

As will be further described, eluate samples for metal concentration assessment were acidified with ultra-pure $HNO_3$ prior to analyses.

**Figure 5: Batch leaching test (LNEC)**

### Preparation of collected eluates for further analysing

Both for the tests conducted at UCD and at LNEC, at the completion of the tests, eluate samples for PAH and metals analyses were prepared according to the procedures described in section 4.3, and later sent to the laboratory for analysis.

## *4.2.2 Percolation test (CEN/TC351 N 0272 and CEN/TS 14405)*

The percolation test is a more complex test and due to the extended duration (usually up to 28-30 days); this is focussed on specific materials. As with the batch test, the maximum particle size is 10 mm and the test sample is prepared by carefully placing it in custom built glass or specific type of plastic (e.g. PMMA) columns. The material samples are then packed with a stainless steel ram in a standardized manner, using five separate layers, and with three sub-layers in each.

### Tests conducted at UCD (CEN/TC351 N 0272)

Figure 6 shows the percolation test set-up for the percolation tests carried out at UCD.

A 0.01% solution of $CaCl_2$ and $NaN_3$ in deionised water was pumped upwards at a continuous linear velocity of 15 ± 2 cm/day (measured through an empty column) though four glass columns in parallel. The $NaN_3$ and $CaCl_2$ were used for same reason as described above. Three of the columns were filled with the 10 mm RA sample, while one remained empty (blank column) for control purposes. All columns were protected from light during the test to prevent degradation of the PAHs. The test duration was of approximately 30 days, concluding once a terminal, cumulative liquid-to-solid (L/S) ratio of 10 l/kg was achieved. Seven eluate samples are taken from each column for analysis. The first is taken at an L/S ratio of 0.1 l/kg and the last at a cumulative L/S ratio of 10.0 l/kg; 5 other samples are taken at discrete intervals throughout the test.

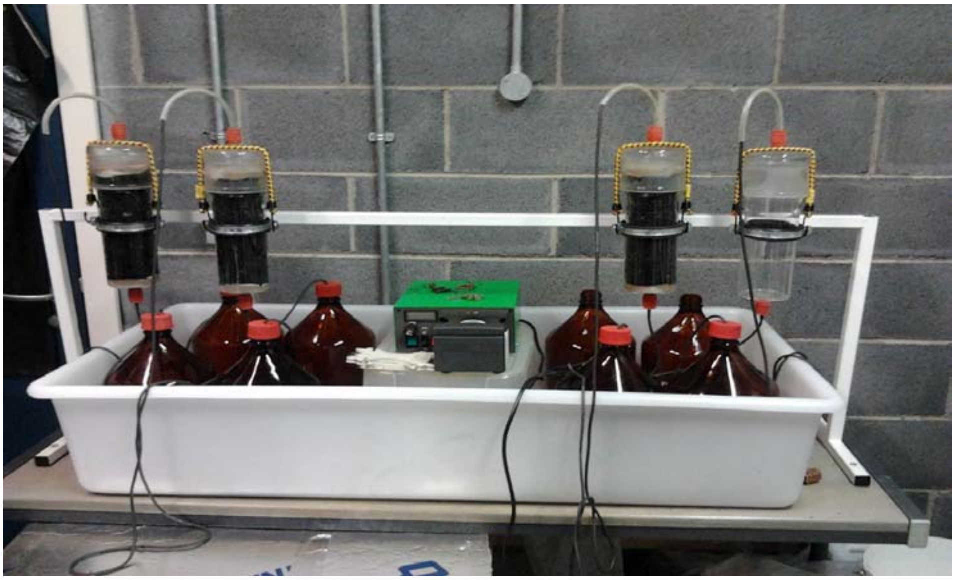

**Figure 6: Percolation test set-up at UCD**

**<u>Percolation tests conducted at LNEC(CEN/TS14405: 2004)</u>**

Percolation tests conducted at LNEC, at the Environmental and Engineering Geology Unit of the Geotechnical Department, were performed according to the procedure established on CEN/TS 14405:2004.

Approximately 3 kg of each (dry) material per test sample was prepared and carefully placed in columns in a standardized manner, using five separate layers. For both materials (P2 and P3 test specimens) it was only possible to test one column. Figure 7 shows the percolation test set-up at LNEC.

An ultra-pure water was up-flowed through the specimens using hydrostatic pressure (another possible is to use a pump). After saturating both specimens, and before start dynamic percolation test, those were left for a period of three days. Due to the high hydraulic conductivity of the specimens (related with the grading of the material, as referred in section 2.2) the used hydraulic gradient (0.05 m/m) was very low in both tests. Even in these conditions, the duration of the dynamic percolation test, after the initial three days in static conditions, was much lower than the usual: about 90 min. in both cases (P2 and P3 test specimens). As in the tests conducted at UCD, all columns were protected from light during the test to prevent degradation of the PAHs. Once a terminal cumulative liquid-to-solid (L/S) ratio of 10 l/kg was achieved, the tests were stopped. Seven eluate samples were taken from each column for analysis. The first was taken at an L/S ratio of 0.1 l/kg and the last at a cumulative L/S ratio of 10.0 l/kg. Five other samples were taken at discrete intervals throughout the test.

**Figure 7: Percolation test set-up at LNEC: (a) Colum with P2 (RAR) and column with P3 (RAR-BE); (b) Running percolation test on P3 (RAR-BE column)**

### *4.2.3 Tank test (*EA NEN 7375:2004*)*

The tank test is a diffusion test and is conducted according to EA NEN 7375:2004; its purpose is for the monitoring the long-term effect of water on the leaching behaviour. The diffusion test is carried out statically to avoid influencing the natural diffusion and, at the same time, to avoid disturbing the sample surface. The nature of the test is to assess the properties of the material in its used state, with the entire sample (a monolithic cylindrical specimen containing RAP, 10% fly-ash and 3.5% bituminous emulsion compacted according to the static compaction method used in the Czech Republic and described in Project report D1.1) placed in the leaching liquid (demineralised water of neutral pH). Additions of the leaching liquid take place at certain time intervals for the period of 64 days, and the quantity of leaching per surface area is determined. The qualitative evaluation of the leaching is based on limits stipulated in regulations concerning construction materials, in this case specified by the Dutch Ministry of Housing, Spatial Planning and the Environment (1999). The leachates were compared to the limits for pavement base courses (Building Materials Decree BMD, 64 days). They were also compared to the Landfill Regulation Amendment for England and Wales no. 1640 of 2005 for waste deposited at landfills – leaching after 64 days.

## 4.3 Eluates analysis

Eluate samples were taken at a range of liquid/solid ratios, depending on the performed leaching test and were analysed for evaluation of PAH and heavy metals released.

### *4.3.1 Detection of polycyclic aromatic hydrocarbons (PAH)*

It is worth mentioning, that there is still not an extended consensus on the best procedure for analysis of eluates for organic substances (Sloot & Dijkstra, 2013). As so, the procedure to preserve eluate samples for this type of analysis must be selected in accordance with the recommendations of the laboratory where the analysis will be performed, and taking into account, among others, the period between the samples collection and its analysis.

At UCD, at the completion of the leaching tests, samples for PAH analysis were stored at 5°C, before being extracted with 20ml of hexane using C18 extraction disks within three days. Following extraction into hexane, samples were sent to the laboratory for PAH analysis where concentrations in the extracts were determined by GC-MS.

At LNEC, eluate samples from leaching tests (batch test, L/S=10 l/kg; percolation test, L/S=0.1-10 l/kg) were collected to dark glass recipients (in order to protect from light), stored at temperatures less then 8 ºC and sent to the analysis, according to laboratory recommendations. PAH compounds were determined by GC-MS as well.

### *4.3.2 Detection of metals*

The concentration of metals, such as: copper, zinc, nickel, manganese, cobalt, cadmium and lead was determined using techniques such as inductively coupled plasma mass spectrometry (ICP-MS), or inductively coupled plasma atomic emission spectroscopy (ICP-AES), depending on the laboratory where the analysis of eluates was performed.

At LNEC, eluate samples from leaching tests (batch test, L/S=10 l/kg; percolation test, L/S=0.1-10 l/kg) were collected to specific plastic recipients and fixed by nitric acid addition prior to be sent to analysis, in accordance to the recommendations of the laboratory. It is worth mentioning that, in the case of batch tests, eluate samples were analyzed for metals detection by ICP-AES (in an internal laboratory of LNEC), and some of the metals were selected to be analyzed by ICP-MS (in an external laboratory) in order to allow for further comparisons. In the case of percolation tests, all eluate samples were analyzed by ICP-MS (in an external laboratory) depending on the type of metal element.

At CTU, special plastic recipients were used for the leaching test containing a plastic grid. Before the test the plastic recipients and grids were subjected for 24 hours to a 1 % solution of nitric acid. After a thorough flush by distilled water the water was tested for presence of required analytes (distilled water was kept for 24 hours in the recipient). After this initial testing test specimens (monoliths) were placed to the recipient on grids. The distance to the face of the recipient has to be at least 20 mm. The recipients with test specimens were filled with distilled water whereas the specimens have to be at least 20 mm below the water level. In the performed test the water volume was 3 Litres. The impact of the environment was monitored by a blind reference without any test specimen. All recipients were during the test covered by a plastic sheet and the room temperature was kept at 20±2 °C.

Particular leaches were taken after 6, 24, 54, 96, 168, 336, 816 and 1536 hours from the recipients with test specimens as well as from the blind reference. At each time the test specimens were removed by latex gloves and put on cleaned and rinsed PE plateau. The leaches were decanted to sample-tubes and pH value as well as the conductivity was determined. Then defined elements or compound were assessed.

During the leaching test it was recognized that there could be material peel off from the specimens. Arisen mass detriments were collected for 1-8 level of leaches decanting. After the last level the solid content of loose material was filtered and the content of undiluted particles was determined after drying at 105 °C. In general the weight loss was 0.005 g to 0.02 g material per one test specimen. Eluate samples were analysed for metals detection by ICP-AES (analyses done by Water Management Research Institute of T.G. Masaryk).

# 5 Test Results and Discussion

To allow comparison of the test results, the data is divided by testing methodology. In the following sections discussion of gained results arising from the conduced tests is summarized.

## 5.1 Batch test results

Summaries of the results for tests carried out on the various materials are presented in this section. Full details of the batch test data are available in the Appendix to this report.

### 5.1.1 Evaluation of the presence of PAH

**Table 3: Test data for Czech RA materials**

| Compound | Detection Limit (μg/l) | Quantity Leached (μg/l) | |
|---|---|---|---|
| | | B1: Czech RA | B6: Foam Mix with Czech RA |
| Naphthalene | <0.014 | 0.095 | 2.078 |
| Acenaphthylene | <0.013 | <0.013 | <0.013 |
| Acenaphthene | <0.013 | 0.025 | 0.106 |
| Fluorene | <0.014 | 0.016 | 0.082 |
| Phenanthrene | <0.011 | 0.029 | 0.234 |
| Anthracene | <0.013 | <0.014 | <0.015 |
| Fluoranthene | <0.012 | 0.037 | 0.110 |
| Pyrene | <0.013 | 0.037 | 0.060 |
| Benzo(a)anthracene | <0.015 | 0.008 | 0.018 |
| Chrysene | <0.011 | 0.008 | 0.018 |
| Benzo(bk)fluoranthene | <0.018 | 0.021 | 0.021 |
| Benzo(a)pyrene | <0.016 | 0.013 | 0.007 |
| Indeno(123cd)pyrene | <0.011 | 0.008 | <0.011 |
| Dibenzo(ah)anthracene | <0.01 | <0.01 | <0.01 |
| Benzo(ghi)perylene | <0.011 | 0.013 | <0.011 |
| **PAH 16 Total** | **<0.195** | **0.26** | **2.73** |

Table 3 shows the data obtained for the tar containing Czech RA material. From observing the sum of the 16 PAHs present, two comments can be made:

1. The levels of PAH present within the RA material (sample B1) are relatively low;
2. The sample corresponding to the foam mix that utilises the Czech RA material (sample B6) has shown increased leaching. The overall leaching level however remains quite low.

**Table 4: Test data for Irish RA materials**

| Compound | Detection Limit (µg/l) | Quantity Leached (µg/l) | | |
|---|---|---|---|---|
| | | B2: Irish RA | B7: Foam Mix with Irish RA | B8: Emulsion Mix with Irish RA |
| Naphthalene | <0.014 | 10.593 | 0.904 | 4.508 |
| Acenaphthylene | <0.013 | 0.286 | 0.266 | 0.089 |
| Acenaphthene | <0.013 | 18.387 | 5.419 | 5.507 |
| Fluorene | <0.014 | 7.133 | 2.225 | 2.127 |
| Phenanthrene | <0.011 | 18.950 | 8.535 | 3.975 |
| Anthracene | <0.013 | 2.498 | 1.978 | 0.436 |
| Fluoranthene | <0.012 | 11.605 | 9.557 | 0.793 |
| Pyrene | <0.013 | 9.415 | 8.888 | 0.516 |
| Benzo(a)anthracene | <0.015 | 3.289 | 4.130 | 0.047 |
| Chrysene | <0.011 | 3.375 | 4.577 | 0.066 |
| Benzo(bk)fluoranthene | <0.018 | 5.868 | 10.884 | 0.052 |
| Benzo(a)pyrene | <0.016 | 4.203 | 7.414 | 0.038 |
| Indeno(123cd)pyrene | <0.011 | 2.551 | 6.401 | 0.019 |
| Dibenzo(ah)anthracene | <0.01 | 0.246 | 0.510 | <0.01 |
| Benzo(ghi)perylene | <0.011 | 2.881 | 5.987 | 0.028 |
| **PAH 16 Total** | **<0.195** | **101.26** | **77.68** | **18.20** |

The data presented in Table 4 correspond to reclaimed material sampled from an Irish cold recycling site. In this case there are a number of very interesting points to note:

1. The levels of PAH associated with the reclaimed material (sample B2) are very high; for comparison, they are 2 orders of magnitude higher than the sample corresponding to the Czech tar containing RA (sample B1). This would strongly indicate that tar was present within the original road construction. For this particular site there were no records available that describe the materials used, and in this scenario it is clear that tar containing material can be used without knowledge.
2. The levels of PAH associated with foam recycling (sample B7) are reduced in comparison to the data presented for the untreated reclaimed material.
3. The levels of PAH associated with the emulsion recycling (sample B8) are significantly reduced in comparison to the untreated reclaimed material.

**Table 5: Test data for German core materials**

| Compound | Detection Limit (µg/l) | Quantity Leached (µg/l) | | | |
|---|---|---|---|---|---|
| | | B3: German Cold Recycled Tar base | B4: As B3, with Binder Course | B9: Foam Mix containing B3 as aggregate | B10: Emulsion Mix containing B3 as aggregate |
| Naphthalene | <0.014 | 3.811 | 0.999 | 0.066 | <0.014 |
| Acenaphthylene | <0.013 | 0.188 | <0.013 | 0.078 | <0.013 |
| Acenaphthene | <0.013 | 5.115 | <0.013 | 0.157 | 2.030 |
| Fluorene | <0.014 | 2.923 | <0.014 | 0.072 | <0.014 |
| Phenanthrene | <0.011 | 10.170 | 1.027 | 0.707 | 14.604 |
| Anthracene | <0.013 | 0.321 | 0.263 | 0.353 | 2.371 |
| Fluoranthene | <0.012 | 4.927 | 2.046 | 3.923 | 19.498 |
| Pyrene | <0.013 | 2.786 | 1.829 | 4.322 | 13.088 |
| Benzo(a)anthracene | <0.015 | 1.328 | 1.535 | 2.624 | 10.990 |
| Chrysene | <0.011 | 1.302 | 1.529 | 3.045 | 12.248 |
| Benzo(bk)fluoranthene | <0.018 | 1.972 | 2.854 | 6.855 | 18.916 |
| Benzo(a)pyrene | <0.016 | 1.086 | 1.524 | 3.811 | 9.864 |
| Indeno(123cd)pyrene | <0.011 | 0.582 | 1.115 | 2.506 | 6.295 |
| Dibenzo(ah)anthracene | <0.01 | 0.129 | <0.01 | 0.640 | 0.793 |
| Benzo(ghi)perylene | <0.011 | 0.394 | 0.976 | 1.858 | 6.050 |
| **PAH 16 Total** | **<0.195** | **37.03** | **17.04** | **31.02** | **116.75** |

The data presented for the German core samples is also very interesting. This tar-containing material was cold-recycled in 1991; as such, the levels of PAH remaining within the sample are an important parameter. From assessing data in Table 5, following comments are made:

There are still significant levels of PAH within the cold recycled tar layer (sample B3) and the combined cold recycled tar & binder course layer. This suggests that even though the material has been in the ground for almost 25 years, the PAHs have not yet been fully leached. It should be noted that the used bitumen emulsions do not contain PAH, and that any increase in leaching behaviour is associated with the recycled material. Comparison with original sample data is required for further interpretation of this point.

The foam mix material containing the cold recycled tar (sample B9) showed a slight decrease in leaching levels. However the emulsion mix material containing the cold recycled tar (sample B10) showed a significant increase in leaching levels. This behaviour is in significant contrast to what was observed for the Irish material (presented in Table 4).

The data presented in Table 6 corresponds to the results from PAH analyses in eluate samples from batch tests carried out at LNEC.

**Table 6: Test data for Reclaimed Asphalt Rubber (RAR) and RAR after being cold stabilised with bitumen emulsion (RAR-BE) from Portugal (Batch tests: EN 12457-4/Method of analysis: GC-MS)**

| **Polycyclic Aromatics Hydrocarbons** | **QL (GC-MS)** (µg/l) | **B5 (RAR) leachate** (µg/l) | **B11 (RAR-BE) leachate** (µg/l) |
|---|---|---|---|
| Naphthalene | 0.100 | <0.100 | <0.100 |
| Acenaphthylene | 0.010 | <0.010 | <0.010 |
| Acenaphthene | 0.010 | <0.010 | <0.010 |
| Fluorene | 0.020 | <0.020 | <0.020 |
| Phenanthrene | 0.030 | <0.030 | 0.032(a) |
| Anthracene | 0.020 | <0.020 | <0.020 |
| Fluoranthene | 0.030 | <0.030 | <0.030 |
| Pyrene | 0.060 | <0.060 | <0.060 |
| Benz(a)anthracene | 0.010 | <0.010 | <0.010 |
| Chrysene | 0.010 | <0.010 | <0.010 |
| Benzo(b)fluoranthene | 0.010 | <0.010 | <0.010 |
| Benzo(k)fluoranthene | 0.010 | <0.010 | <0.010 |
| Benzo(a)pyrene | 0.020 | <0.020 | <0.020 |
| Indeno(1.2.3.cd)pyrene | 0.010 | <0.010 | <0.010 |
| Benzo(g.h.i)perylene | 0.010 | <0.010 | <0.010 |
| Dibenz(a.h)anthracene | 0.010 | <0.010 | <0.010 |
| Sum of 16 PAH | 0.370 | <0.370 | <0.370 |
| Sum of PAH (MoE) | 0.19 | <0.19 | <0.19 |
| Sum of 6 PAH (WHO) | 0.090 | <0.090 | <0.090 |
| Sum of 4 PAH | 0.040 | <0.040 | <0.040 |

**Legend:**
QL – Quantification Limit
GC-MS - Gas Chromatography - Mass Spectrometry
(a) Measurement uncertainty = ±26,0%

The concentrations of PAH compounds in eluate samples from B5 (RAR) e B11 (RAR-BE) are below the quantification limits, with exception for Phenantrene in B11 leachate, whose concentration nevertheless remains very close to the quantification limit. This insignificant variation on the Phenantrene concentration from B5 to B11 leachates, is probably only related with the variability of the samples and the uncertainty of the tests.

### *5.1.2 Evaluation of the presence of metals*

The laboratory analyses of the eluates collected from the batch tests carried out at LNEC on B5 (RAR) and B11 (RAR-BE) test specimens, were performed for seven elements (Cd, Co, Cu, Pb, Mn, Ni and Zn) by ICP-AES. In view to compare the results from batch and percolation tests, two metals (Lead and Nickel) were selected for analysis also by ICP-MS. In Table 7 the obtained results are presented.

**Table 7: Test data for Reclaimed Asphalt Rubber (RAR) and RAR after being cold stabilised with bitumen emulsion (RAR-BE) from Portugal (Batch tests: EN 12457-4 / Method of analysis: ICP-AES & ICP-MS)**

| Metals | DL (ICP-AES) | QL (ICP-MS) | B5 (RAR) leachate | | | | B11 (RAR-BE) leachate | | | |
|---|---|---|---|---|---|---|---|---|---|---|
| | | | Analysed by ICP-AES | | Analysed by ICP-MS | | Analysed by ICP-AES | | Analysed by ICP-AES | |
| | (mg/l) | (µg/l) | (mg/l) | (mg/kg) | (µg/l) | (mg/kg) | (mg/l) | (mg/kg) | (µg/l) | (mg/kg) |
| Cadmium, Cd | 0.002 | 0.50 | <0.002 | <0.02 | - | - | <0.002 | <0.02 | - | - |
| Cobalt, Co | 0.020 | 0.50 | <0.020 | <0.20 | - | - | <0.020 | <0.20 | - | - |
| Copper, Cu | 0.013 | 1.0 | <0.013 | <0.13 | - | - | <0.013 | <0.13 | - | - |
| Lead, Pb | 0.009 | 1.0 | <0.009 | <0.09 | <1.0 | <0.01 | <0.009 | <0.09 | <1.0 | <0.01 |
| Manganese, Mn | 0.013 | 0.50 | <0.013 | <0.13 | - | - | <0.013 | <0.13 | - | - |
| Nickel, Ni | 0.006 | 3.0 | <0.006 | <0.06 | <3.0 | <0.03 | <0.006 | <0.06 | <3.0 | <0.03 |
| Zinc, Zn | 0.006 | 2.0 | <0.006 | <0.06 | - | - | 0.026 | 0.26 | - | - |

**Legend:**

DL – Detection Limit; RL – Quantification Limit

ICP-AES - Inductively Coupled Plasma - Atomic Emission Spectrometry

ICP-MS - Inductively Coupled Plasma - Mass Spectrometry

With respect to B5 (RAR) eluate, all the concentration values are below the detection/quantification limits. Considering B11 (RAR_BE) eluate, only the concentration of Zinc element is above the detection limit.

For analysing the results, they were compared in Table 8 to the required values of the Council Decision 2003/33/EC (19.12.2002), that establishes criteria and procedures for the acceptance of waste at landfills pursuant to Article 16 of and Annex II to Directive 1999/31/EC: Limit values for waste acceptable at landfills for inert waste.

**Table 8: Evaluation of the results from the presence of metals (Method of analysis: ICP-AES) in B5 (RAR) and B11 (RAR-BE) leachates from batch tests (EN 12457-4)**

| **Metals** | **Limit values for waste acceptable at landfills for inert waste (L/S=10 l/kg)** (mg/kg dry substance) | **B5 (RAR) leachate** | | **B11 (RAR-BE) leachate** | |
|---|---|---|---|---|---|
| | | **Analysed by ICP-AES** (mg/kg) | **Evaluation** | **Analysed by ICP-AES** (mg/kg) | **Evaluation** |
| Cadmium, Cd | 0.04 | <0.02 | ✓ | <0.02 | ✓ |
| Cobalt, Co | - | <0.20 | (n/a) | <0.20 | (n/a) |
| Copper, Cu | 2 | <0.13 | ✓ | <0.13 | ✓ |
| Lead, Pb | 0.5 | <0.09 | ✓ | <0.09 | ✓ |
| Manganese, Mn | - | <0.13 | (n/a) | <0.13 | (n/a) |
| Nickel, Ni | 0.4 | <0.06 | ✓ | <0.06 | ✓ |
| Zinc, Zn | 4 | <0.06 | ✓ | 0.26 | ✓ |

**Legend:**
ICP-AES - Inductively Coupled Plasma - Atomic Emission Spectrometry
✓ Concentration below the EC Directive limit for waste acceptance at landfills for inert waste
(n/a) – not applicable

From Table 8 it can be concluded that all detected metals contents are below the maximum limit values required in the referred Council Decision, allowing inferring that the analysed bituminous materials are similar to inert wastes with respect to the analysed metals.

## *5.2 Percolation test results*

### *5.2.1 Evaluation of the presence of PAH*

The German cold tar sample B3 was tested using the percolation test and a summary of the results are presented in Table 7. In this case the results are presented in the form of µg of leached PAH per kg of sample. The reason for this is that the water in contact with the sample is varying throughout the test, and serves to make comparison with the batch test more difficult. This represents an important distinction between percolation and batch tests:

- For batch tests, the same water is used throughout the test, and the chemical composition will change as leaching progresses. This means that even though the test conditions can be more aggressive, the test set-up allows PAHs to accumulate within the leaching water and can have the effect of inhibiting further diffusion.
- For percolation tests, the reverse is the case. Test conditions are more representative of real life, and diffusion is not inhibited by the presence of PAHs within the water.

What is however clear from the test is that for most PAHs, the quantity leached is increasing with test duration. This can be observed graphically from Figures 5 through 8. This suggests that the test has not yet reached equilibrium, and there is no indication as to whether the peak in leaching behaviour has yet been observed. This suggests that the percolation test, currently being developed by CEN TC 351 for “determination of the release from granular construction products” is not of sufficient duration for the leaching of PAH from reclaimed road materials. The data produced from this test will be compared to other test data in the next section.

**Table 7: Summary of the percolation test data obtained on sample B3**

| Compound | Detection Limit (µg/l) | Quantity Leached (µg/kg) | | | | | | |
|---|---|---|---|---|---|---|---|---|
| | | 1st Extraction | 2nd Extraction | 3rd Extraction | 4th Extraction | 5th Extraction | 6th Extraction | 7th Extraction |
| Naphthalene | <0.014 | 0.522 | 0.956 | 6.769 | 2.967 | 10.237 | 23.463 | 59.396 |
| Acenaphthylene | <0.013 | 0.044 | 0.057 | 0.331 | 0.127 | 0.226 | 0.503 | 0.598 |
| Acenaphthene | <0.013 | 0.192 | 0.419 | 2.873 | 1.369 | 4.655 | 10.751 | 26.005 |
| Fluorene | <0.014 | 0.086 | 0.222 | 1.507 | 0.721 | 2.364 | 5.508 | 13.592 |
| Phenanthrene | <0.011 | 0.257 | 0.643 | 4.030 | 2.083 | 6.391 | 14.716 | 40.697 |
| Anthracene | <0.013 | 0.029 | 0.083 | 0.503 | 0.280 | 0.812 | 1.752 | 4.999 |
| Fluoranthene | <0.012 | 0.082 | 0.185 | 1.223 | 0.659 | 1.902 | 3.654 | 9.281 |
| Pyrene | <0.013 | 0.047 | 0.090 | 0.588 | 0.330 | 0.939 | 1.785 | 4.516 |
| Benzo(a)anthracene | <0.015 | 0.009 | 0.011 | 0.063 | 0.043 | 0.117 | 0.179 | 0.396 |
| Chrysene | <0.011 | 0.009 | 0.012 | 0.070 | 0.043 | 0.118 | 0.211 | 0.471 |
| Benzo(bk)fluoranthene | <0.018 | 0.006 | 0.003 | 0.012 | 0.010 | 0.042 | 0.050 | 0.146 |
| Benzo(a)pyrene | <0.016 | 0.004 | 0.003 | 0.008 | 0.007 | 0.029 | 0.027 | 0.086 |
| Indeno(123cd)pyrene | <0.011 | 0.001 | bdl | bdl | bdl | bdl | bdl | bdl |
| Dibenzo(ah)anthracene | <0.01 | 0.000 | bdl | bdl | bdl | bdl | bdl | bdl |
| Benzo(ghi)perylene | <0.011 | 0.001 | bdl | bdl | bdl | bdl | bdl | bdl |

Legend: bdl – below detection limit

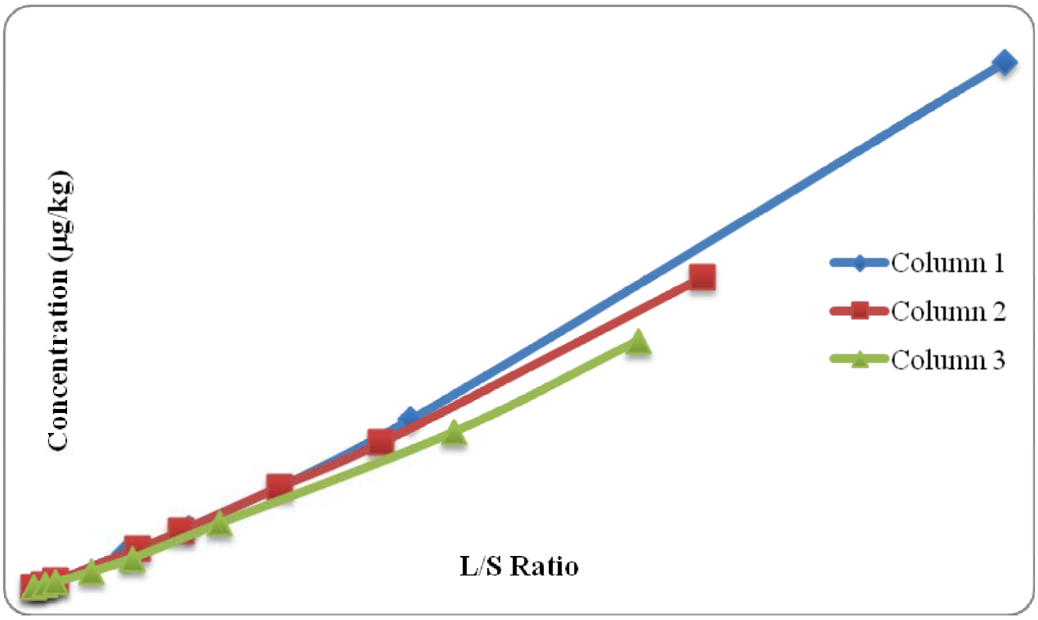

Figure 8: Cumulative naphthalene leaching from German recycled cold tar

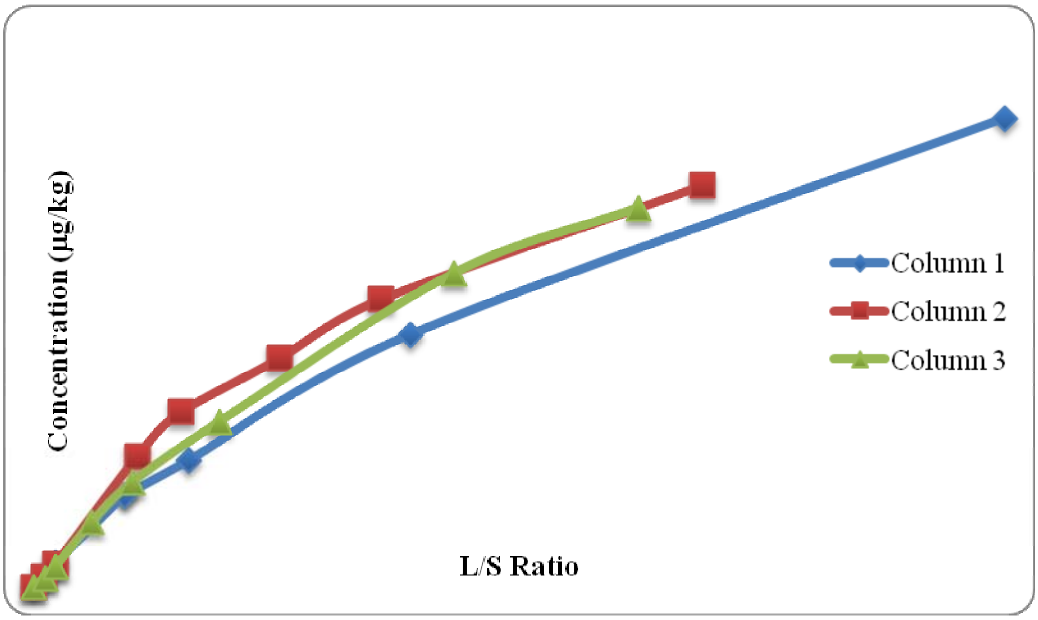

Figure 9: Cumulative acenaphthylene leaching from German recycled cold tar

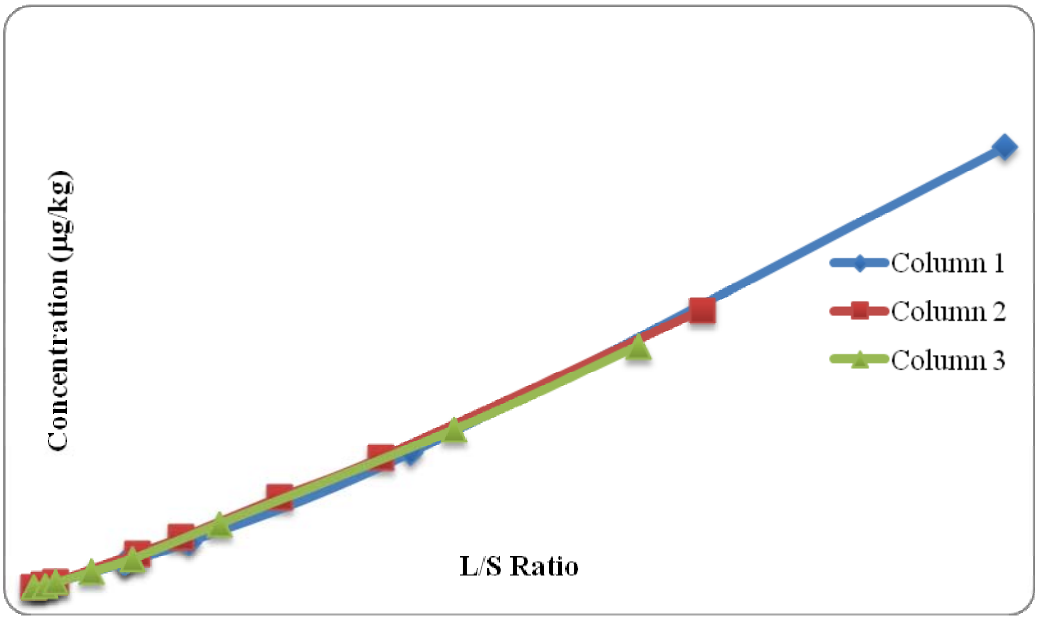

Figure 10: Cumulative acenaphthene leaching from German recycled cold tar

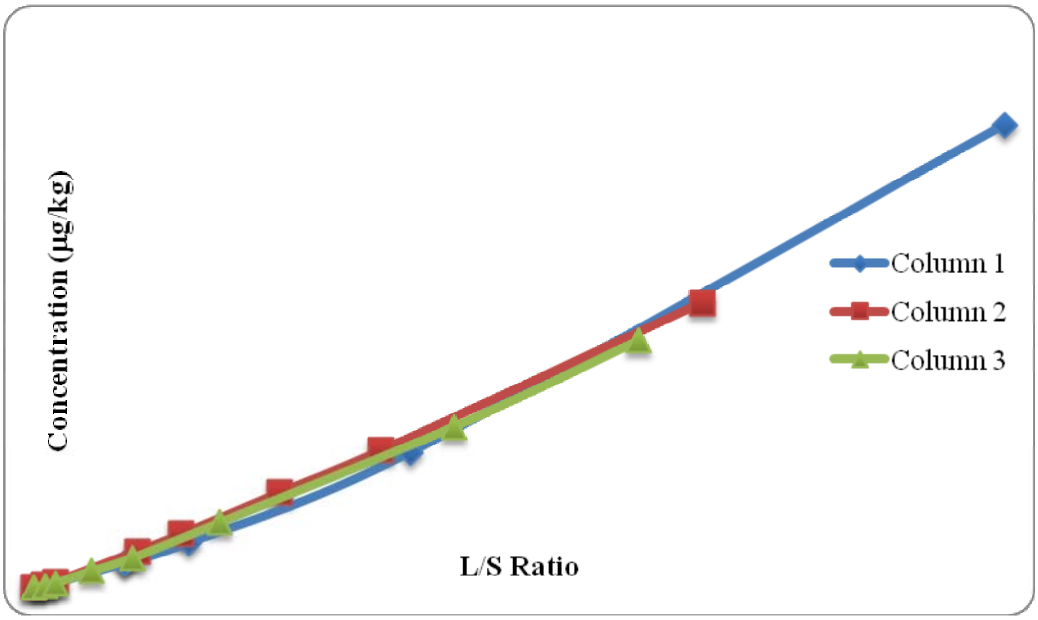

Figure 11: Cumulative fluorene leaching from German recycled cold tar

Table 9 shows the results of PAH concentrations detected on the first extraction of eluate samples (L/S=0.1 l/kg) from percolation tests conducted at LNEC.

**Table 9: Test data for Reclaimed Asphalt Rubber (RAR) and RAR after being cold stabilised with bitumen emulsion (RAR-BE) from Portugal (Percolation tests: CEN/TS 14405 / Presence of PAHs - Method of analysis: GC-MS)**

| **Polycyclic Aromatics Hydrocarbons** | **QL by GC-MS** (µg/l) | **P2 (RAR) leachate** **1st fraction volume (on a total of seven); (L/S=0.1 l/kg)** (µg/l) | **P3 (RAR-BE) leachate** **1st fraction volume (on a total of seven); (L/S=0.1 l/kg)** (µg/l) |
|---|---|---|---|
| Naphthalene | 0.100 | <0.100 | <0.100 |
| Acenaphthylene | 0.010 | <0.010 | <0.010 |
| Acenaphthene | 0.010 | <0.010 | <0.010 |
| Fluorene | 0.020 | <0.020 | <0.020 |
| Phenanthrene | 0.030 | <0.030 | <0.030 |
| Anthracene | 0.020 | <0.020 | <0.020 |
| Fluoranthene | 0.030 | <0.030 | <0.030 |
| Pyrene | 0.060 | <0.060 | <0.060 |
| Benz(a)anthracene | 0.010 | <0.010 | <0.010 |
| Chrysene | 0.010 | <0.010 | <0.010 |
| Benzo(b)fluoranthene | 0.010 | <0.010 | <0.010 |
| Benzo(k)fluoranthene | 0.010 | <0.010 | <0.010 |
| Benzo(a)pyrene | 0.020 | <0.020 | <0.020 |
| Indeno(1.2.3.cd)pyrene | 0.010 | <0.010 | <0.010 |
| Benzo(g.h.i)perylene | 0.010 | <0.010 | <0.010 |
| Dibenz(a.h)anthracene | 0.010 | <0.010 | <0.010 |
| Sum of 16 PAH | 0.370 | <0.370 | <0.370 |
| Sum of PAH (MoE) | 0.19 | <0.19 | <0.19 |
| Sum of 6 PAH (WHO) | 0.090 | <0.090 | <0.090 |
| Sum of 4 PAH | 0.040 | <0.040 | <0.040 |

**Legend:**

QL – Quantification Limit

GC-MS - Gas Chromatography - Mass Spectrometry

Taking into account that, in the first fraction volume (L/S = 0.1 l/kg) of percolation test, all PAH values were below the quantification limits, similarly to the results obtained in the eluate samples collected from batch tests on the same type of materials (RAR and RAR-BE), it was considered that no further analysis was necessary or relevant for the remaining extractions.

### *5.2.2 Evaluation of the presence of metals*

The laboratory analyses of the leachate collected from the percolation tests performed at LNEC (CEN/TS 14405) were carried out for nine elements (As, Cd, Cr, Co, Cu, Pb, Mn, Ni and Zn) in the first fraction volume (L/S = 0.1 l/kg). In Table 10 the obtained results are presented.

In the first fraction volume (L/S = 0.1 l/kg) of the leachate from P2 (RAR), Cadmium and Chromium concentrations were below the quantification limits and from P3 (RAR-BE), Cadmium, Chromium and Arsenic concentrations were below those limits as well. Among the analysed metals above the quantification limit, two metals were selected for further analysis in the other six fractions volumes, Lead and Nickel, considering their classification as priority substances in the aim of Directive 2013/39/EU before mentioned.

**Table 10: Test data for Reclaimed Asphalt Rubber (RAR) and RAR after being cold stabilised with bitumen emulsion (RAR-BE) from Portugal (Percolation tests: CEN/TS 14405 / Presence of metals - Method of analysis: ICP-MS)**

| Metals | QL (ICP-MS) (µg/l) | P2 (RAR) leachate | | | | | | | P3 (RAR-BE) leachate | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | | Fraction volume (on a total of seven) | | | | | | | Fraction volume (on a total of seven) | | | | | | |
| | | 1st L/S=0.1 l/kg | 2nd L/S=0.2 l/kg | 3rd L/S=0.5 l/kg | 4th L/S=1 l/kg | 5th L/S=2 l/kg | 6th L/S=5 l/kg | 7th L/S=10 l/kg | 1st L/S=0.1 l/kg | 2nd L/S=0.2 l/kg | 3rd L/S=0.5 l/kg | 4th L/S=1 l/kg | 5th L/S=2 l/kg | 6th L/S=5 l/kg | 7th L/S=10 l/kg |
| | | (µg/l) | | | | | | | (µg/l) | | | | | | |
| Arsenic, As | 1.0 | 6.0 | - | - | - | - | - | - | <1.0 | - | - | - | - | - | - |
| Cadmium, Cd | 0.50 | <0.50 | - | - | - | - | - | - | <0.50 | | | | | | |
| Chromium, Cr | 5.0 | <5.0 | - | - | - | - | - | - | <5.0 | - | - | - | - | - | - |
| Cobalt, Co | 0.50 | 8.00 | - | - | - | - | - | - | 2.74 | - | - | - | - | - | - |
| Copper, Cu | 1.0 | 55.7 | - | - | - | - | - | - | 145 | - | - | - | - | - | - |
| Lead, Pb | 1.0 | 25.2 | 11.8 | 5.1 | 2.4 | 1.4 | <1.0 | <1.0 | 23.2 | 10.2 | 6.0 | 2.1 | 1.4 | 1.0 | <1.0 |
| Manganese, Mn | 0.50 | 137 | - | - | - | - | - | - | 95.3 | - | - | - | - | - | - |
| Nickel, Ni | 3.0 | 308 | 36.7 | 30.2 | 26.4 | 20.2 | 8.0 | <3.0 | 220 | 28.4 | 29.8 | 26.2 | 17.8 | 5.0 | <3.0 |
| Zinc, Zn | 2.0 | 254 | - | - | - | - | - | - | 619 | - | - | - | - | - | - |

**Legend:**

QL – Quantification Limit

ICP-MS - Inductively Coupled Plasma - Mass Spectrometry

Figure 12 graphically presents Lead and Nickel content evolution from first fraction volume (0.1 l/kg) to seventh fraction volume (10.0 l/kg) of percolation test performed at LNEC.

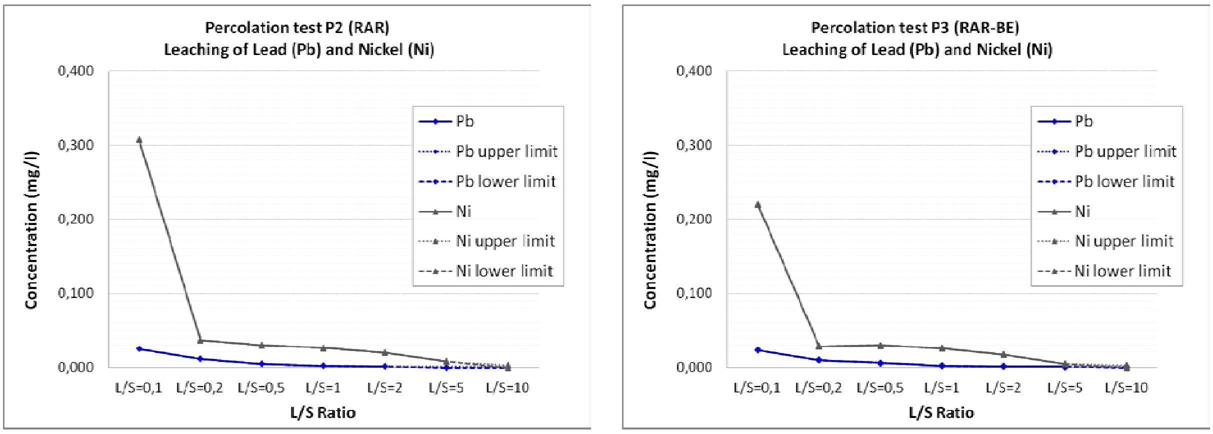

**Figure 12: Percolation test: Lead and Nickel content evolution with each fraction volume**

For further analysing of the above presented results, they were also compared to the required values of the before mentioned Council Decision 2003/33/EC of 19 December 2002 (Table 11).

**Table 11: Evaluation of the results from the presence of metals (Method of analysis: ICP-MS) in P2 (RAR) and P3 (RAR-BE) leachates from percolation tests (CEN/TS 14405)**

| Metals | Limit values for waste acceptable at landfills for inert waste $C_0$ (L/S=0.1 l/kg) (mg/l) | P2 (RAR) leachate | | P3 (RAR-BE) leachate | |
|---|---|---|---|---|---|
| | | 1st fraction volume (on a total of seven) (L/S=0.1 l/kg) Analysed by ICP-MS (mg/l) | Evaluation | 1st fraction volume (on a total of seven) (L/S=0.1 l/kg) Analysed by ICP-MS (mg/l) | Evaluation |
| Arsenic, As | 0.06 | 0.0060 | ✓ | <0.0010 | ✓ |
| Cadmium, Cd | 0.02 | <0.00050 | ✓ | <0.00050 | ✓ |
| Chromium, Cr | 0.1 | <0.0050 | ✓ | <0.0050 | ✓ |
| Cobalt, Co | - | 0.00800 | ✓ | 0.00274 | ✓ |
| Copper, Cu | 0.6 | 0.0557 | ✓ | 0.145 | ✓ |
| Lead, Pb | 0.15 | 0.0252 | ✓ | 0.0232 | ✓ |
| Manganese, Mn | - | 0.137 | ✓ | 0.0953 | ✓ |
| Nickel, Ni | 0.12 | 0.308 | × | 0.220 | × |
| Zinc, Zn | 1.2 | 0.254 | ✓ | 0.619 | ✓ |

**Legend:**
GC-MS - Gas Chromatography - Mass Spectrometry
✓ Concentration below the EC Directive limit for waste acceptance at landfills for inert waste
× Concentration above the EC Directive limit for waste acceptance at landfills for inert waste

According to the referred restrictive criteria, only the Nickel metal is above the limit value required in the referred Council Decision for the first fraction volume of the leachate, but its content strongly decreases from the first fraction volume of leachate to the following ones (Figure 12).

The results obtained for Nickel detected concentrations in the first fraction volume of the leachate and in the following ones eventually help to predict the long-term performance of RAR and RAR-BE materials, once it suggests that after first “leaching actions” (e.g. first rains) a great decrease of Nickel concentrations would occur.

For further analysis, cumulative values of Lead and Nickel concentrations during the percolation tests were determined (Figure 13). These results will be compared to the correspondent ones obtained in the batch test at L/S= 10l/kg (see paragraph 5.4.1).

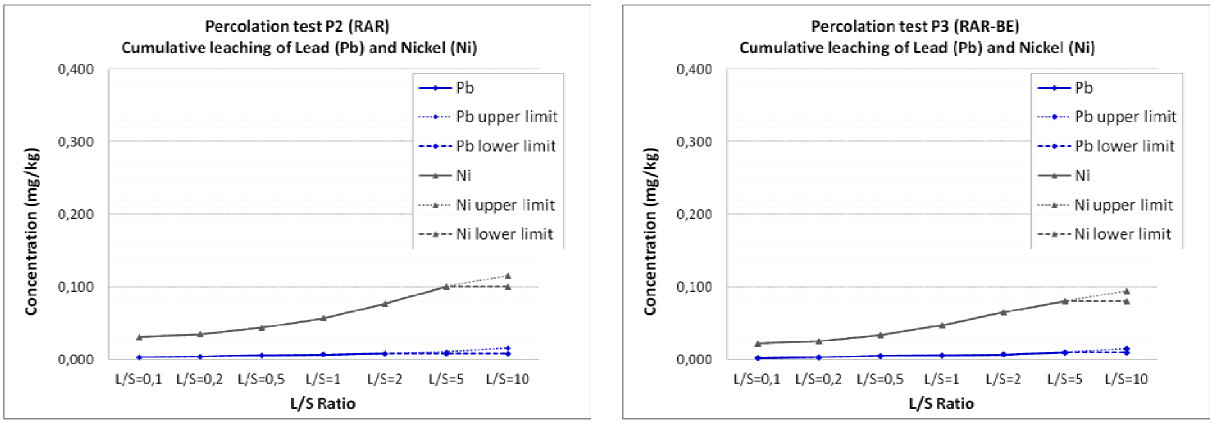

Figure 13: Cumulative Lead and Nickel leaching, from LNEC tests on P2 (RAR) and P3 (RAR-BE) test specimens

Regarding the potential effect of binder emulsion (Figure 14), three of the elements analysed decreased their concentration in the leachate (Cobalt, Manganese and Nickel) from P2 (RAR) to P3 (RAR-BE) samples. In fact, from Figure 12 and Figure 14, it can be clearly inferred that Nickel content decreases about 30% when the reclaimed material is involved by bituminous emulsion, suggesting the possibility that this metal is partially encapsulated/stabilized by the binder.

On the other hand, two of the metals analysed increased their concentration in collected leachates (Copper and Zinc) from P2 (RAR) to P3 (RAR-BE) samples, probably due to the emulsion composition added. Nevertheless, it must be stressed that, in both cases (P2 and P3 leachates), the detected Copper and Zinc concentrations were well below the maximum limit values for waste at landfills for inert waste (Council Decision 2003/33/EC of 19 December 2002).

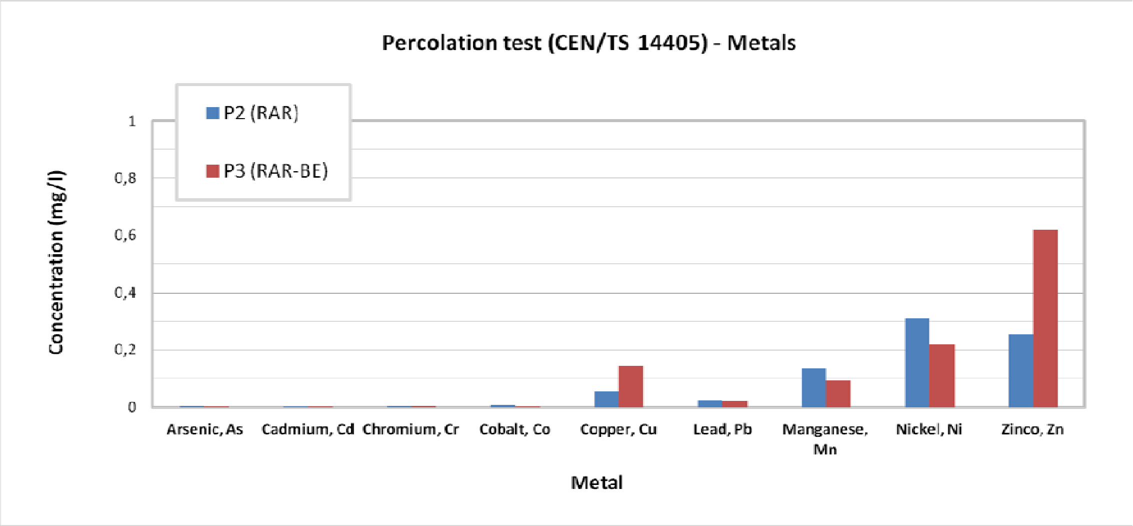

Figure 14: Comparison between metals concentrations on the first fraction volume of leachate from P2 (RAR) and P3 (RAR-BE) test specimens

## 5.3 Tank test results

The results from the diffusive tank test of the Czech RA material is shown in Table 10. In this case it should be noted that the sample T1 is from a Czech emulsion mix, and that samples T2 and T3 are similar, but contain 10% mechanically activated fly ash.

**Table 10: Effect of cold recycling on tank test results**

| Leaching of analysed elements (mg/m$^2$) | Sample T1 | Sample T2 | Sample T3 | BMD 64 days | Leaching from Landfilled waste(*) (64 days) |
|---|---|---|---|---|---|
| Chloride | 2 810 | 2 553 | 2 516 | - | 10 000 |
| Sulfate | 2 390 | 71 397 | 126 308 | 27 000 | 10 000 |
| Arsenic | 0.45 | 9 | 2.48 | 41 | 1.3 |
| Cadmium | 0.09 | 0.086 | 0.085 | 1.1 | 0.2 |
| Chromium | 0.6 | 0.6 | 0.6 | 140 | 5 |
| Copper | 4.3 | 4 | 3.6 | 51 | 45 |
| Mercury | 0.03 | 0.03 | 0.02 | 0.4 | 0.1 |
| Nickel | 0.9 | 0.9 | 0.8 | 50 | 6 |
| Lead | 0.2 | 0.31 | 0.3 | 120 | 6 |
| Zinc | 8.3 | 5.2 | 6.1 | 200 | 30 |

**Legend:**
BMD - Building Materials Decree (Dutch Ministry of Housing, Spatial Planning and the Environment, 1999).
(*) Landfill Regulation Amendment for England and Wales no. 1640 of 2005 for waste deposited at landfills.

From observing the data above, we can see that for most heavy metals, the levels of leaching do not increase with the addition of the mechanically activated fly ash. The exception to this however is sulphate and arsenic, both of which show significant increases in leaching with the addition of fly ash. It can also be seen that the leaching associated with these materials after 64 days is significantly higher than that associated with a landfill at the same point in time.

It should be noted that the Tank Test is the only test that has allowed the samples to be tested in the same manner in which they are used in service, i.e. as a monolith. In this particular case where leaching of PAHs is not a concern, the test appears to have performed well. It should however be noted that the increased levels of leaching are worthy of further investigation.

## 5.4 Discussion of results

A significant amount of data has been presented in the preceding sections and this will be further assessed now. Key points worthy of discussion include the validity of the test methods, the implications of current practice and recommendations for further usage.

### *5.4.1 Influence of test method*

It was noted in section 2 that there are no ideal test methods available for assessing the leaching associated with cold recycled materials. Some tests are unsuitable for assessing leaching of PAHs, while others require significant processing of the aggregates. This means that some level of compromise was required, and in this study the choice was to use the batch test, besides percolation and tank tests. This allowed the assessment of PAH leaching associated with tar and crumb rubber materials, but did require some degree of size reduction (e.g. sample crushing) to produce the test specimen in the case of batch and percolation leaching tests.

In the case of RA containing tar, it is proposed that this has led to some unusual trends, including:

- Sample B1 produced low leaching levels, and a small increases was observed when the material was used in a foam mix;
- Sample B2 produced high levels of leaching and a moderate decrease was observed when the material was used in a foam mix. A significant reduction was observed when the material was used in an emulsion mix.
- Sample B3 produced quite high leaching levels. A moderate decrease was observed when the material was used in a foam mix, but a significant increase was observed when the material was used in an emulsion mix.

These results are quite contradictory and suggest that there are other issues involved, most notably the size reduction required. To produce a sample that meets the batch test requirement (max particle size of 10mm), some level of crushing of the cold-recycled material is needed. A potential side effect of this is that this can expose fresh, tar-containing surfaces within the material, thus destroying the encapsulation effect. This was not consistently observed – it is postulated that the cases where decreases in leaching were observed were due to the encapsulating effect being disrupted to a lesser extent.

Regarding RA containing crumb rubber (before or after being cold stabilized with bituminous emulsion), a comparison between the results obtained for the same materials in batch tests and in percolation tests was also performed.

In what concerns PAH detected concentrations, they were below the quantification limits for both leaching tests, with exception of Phenanthrene substance in eluate samples collected from batch test on B11 (RAR-BE) specimens. However, even in this case, the concentration was very low and very close to the quantification limit.

The results obtained both on batch tests (Table 7) and percolation tests (Table 9) were somewhat expected, since the materials tested on those leaching tests – RAR material having in its constitution, as binder, an aged bitumen (50/70 pen grade) modified with crumb rubber (reclaimed asphalt rubber pavement about 8 years in service); and RAR-BE material, thus having in its constitution not only aged bitumen-rubber, but also “new” bitumen from the bituminous emulsion (C60B5) used as binder in the cold stabilisation of the RAR material – are supposed to present insignificant concentrations of PAH, thus not being reported as hazardous materials by the List of Hazardous Waste adopted by Commission Decision 2001/118/EC.

With view to compare metals detected concentrations, the results of percolation tests for the cumulative values at the end of each test (L=10 l/kg) were compared with the correspondent values obtained in batch tests on same materials (RAR & RAR-BE).

Table 12 shows the results obtained for Lead and Nickel concentrations on the eluate samples from batch and percolation leaching tests.

**Table 12. Comparative analysis of Lead and Nickel concentrations detected between batch and percolation tests on RAR samples (B5 and P2 test specimens) and RAR after being cold stabilised with bitumen emulsion (RAR-BE) (B11 and P3 test samples)**

| **Metals** | **B5 / P2 (RAR) leachate** | | | **B11 / P3 (RAR-BE) leachate** | | |
|---|---|---|---|---|---|---|
| | **Batch tests: EN 12457-4** | | **Percolation tests: CEN/TS 14405** (a) | **Batch tests: EN 12457-4** | | **Percolation tests: CEN/TS 14405** (a) |
| | Analysed by ICP-AES | Analysed by ICP-MS | Analysed by ICP-MS | Analysed by ICP-AES | Analysed by ICP-MS | Analysed by ICP-MS |
| | (mg/kg) | (mg/kg) | (mg/kg) | (mg/kg) | (mg/kg) | (mg/kg) |
| Lead, Pb | <0.09 | <0.01 | 0.008 – 0.016 (b) | <0.09 | <0.01 | 0.010 – 0.015 (b) |
| Nickel, Ni | <0.06 | <0.03 | 0.101 – 0.116 (b) | <0.06 | <0.03 | 0.08 – 0.095 (b) |

**Legend:**

ICP-AES - Inductively Coupled Plasma - Atomic Emission Spectrometry

ICP-MS - Inductively Coupled Plasma - Mass Spectrometry

(a) Cumulative values (L/S = 10 l/kg)

(b) Since last extractions were below detection limit, lower limit and upper limit were calculated in accordance to CEN/TS 14405

From the comparison between the results obtained for the same materials in batch tests and in percolation tests, the following considerations could be inferred:

- With respect to Lead detected concentrations both tests seem to be in accordance;
- With regard to Nickel concentrations, slightly higher levels were obtained in the percolation tests than in batch tests, but even so, both values can be considered of the same order of magnitude, taking into account not only the variability of the samples (reclaimed asphalt rubber pavement) but also the uncertainty of the tests (leaching tests and analysis of the eluates).

### *5.4.2 Behaviour of previously recycled tar material*

It was previously noted that the German tar material was previously cold recycled in 1991; at that time a series of tests were also carried out on the material, including a form of batch test. This particular form of the batch test (known as S4) involved the sample bottle being shaken end-over-end for a period of 24 hrs. The L/S ratio used was also 10.0. The results for the S4 tests in 1991 are compared to the batch test conducted on sample B3. The full results are not available for the 1991 test data, but instead some key parameters. These include:

- PAH 16 (sum of 16 PAHs as identified by EPA)
- PAH 6 (sum of 6 PAHs as previously used by TVO; these include Fluoranthene, Benzo(b)fluoranthene, Benzo(k)fluoranthene, Benzo(a)pyrene, Indinopyrene and Benzo(ghi)perylene),
- Benzo(a)pyrene (used alone as it is highly carcinogenic).

The results for this comparison are given in Table 11.

**Table 11: Comparison of leaching data to that obtained from a report in 1991**

| **Parameter** | **Quantity Leached (µg/l)** | | | | | |
|---|---|---|---|---|---|---|
| | Sample 1 | Sample 2 | Sample 3 | Sample 4 | Sample 5 | Average |
| **Original Coal Tar Asphalt** | | | | | | |
| PAH 16 | 98.7 | 119.4 | 194.9 | 112.0 | 102.8 | 125.6 |
| PAH 6 | 66.3 | 70.0 | 110.0 | 64.0 | 58.3 | 73.7 |
| Benzo(a)pyrene | 18.0 | 20.0 | 29.0 | 17.0 | 17.0 | 20.2 |
| **Cold Recycled Tar (conducted in 1991)** | | | | | | |
| PAH 16 | 14.2 | 23.6 | 37.2 | 45.7 | 17.1 | 27.6 |
| PAH 6 | 7.9 | 11.8 | 18.6 | 23.6 | 9.2 | 14.2 |
| Benzo(a)pyrene | 2.0 | 2.8 | 4.0 | 4.6 | 1.8 | 3.0 |
| **Cold Recycled Tar (conducted in 2014)** | | | | | | |
| PAH 16 | 38.71 | 35.18 | 37.21 | | | 37.03 |
| PAH 6 | 9.50 | 8.32 | 9.06 | | | 8.96 |
| Benzo(a)pyrene | 1.18 | 0.97 | 1.11 | | | 1.09 |

A comparison of the data shows that the original recycling process had the effect of significantly reducing the levels of PAH leached, suggesting that the encapsulation process was successful. The levels of leaching obtained from the current testing programme and that conducted 23 years ago are largely similar, suggesting that the PAHs have not leached from the cold recycled material in the interim. While the issue of size reduction will inevitably lead to some uncertainty in the results, the similarity levels are very encouraging and promote confidence in the results.

It is also interesting to note that in the original report, the conclusion was also made that the batch test is not fully satisfactory in assessing the encapsulation effect in cold recycled mixes. The authors have also recommended a form of tank test known as the ‘Trog test’, which was also conducted on monolithic samples. That test, which does not appear to be available today, showed a significant encapsulation effect associated with the cold recycling process.

### *5.4.3 Application of cold recycling process*

The final point which has not yet been discussed is the issue of where the recycling process is used in the road. Earlier in the report it was noted that Legret et al (2005) recommended field studies to assess the effect of parameters such as infiltration. The schematic shown in Figure 9 is a representation of the numerous routes that water may take in an asphalt pavement.

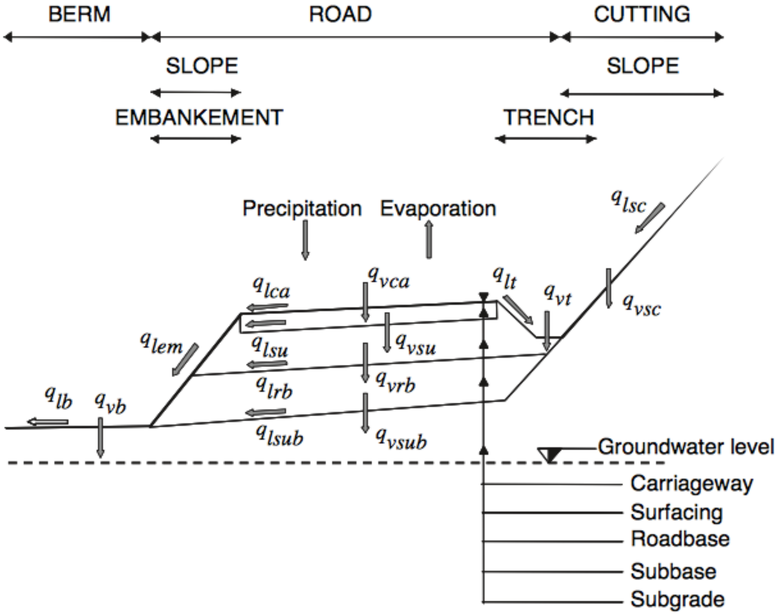

**Figure 9: Schematic of water-flow in an asphalt pavement, taken from Erlingsson et al (2009)**

The importance of this point is that when the cold recycled material is used as a base layer, it is typically overlain by a significant depth of asphalt, and that material will often feature some form of camber. The effect of this is that the majority of the water that meets the road surface is lost to run-off, and that a relatively small amount will interact with the base layer in the form of leaching. This significantly reduces the risk associated with cold recycling in comparison to using reclaimed materials in surface layers. A quantification of that risk has not been included as it was outside the scope of this project, but the point is worthy of attention.

# 6 Conclusions

Based on the data presented above, some conclusions and observations may be made:

- It was observed that the highest levels of PAH leaching were associated with the Irish recycling site, where tar was not expected. For this particular site there was no information available on the material being recycled, which highlights the value of preliminary screening in advance of any recycling work. This may involve the use of relatively simple tools such as the PAK marker etc.
- The batch test data showed that the encapsulating effect associated with the cold recycling process was present, but could not show the reliability of this effect. This is due to the nature of the test method in that some element of sample crushing is usually required.
- The percolation test data also showed relatively high PAH leaching levels for tar containing reclaimed asphalt material. The suitability of the test is also questionable as the data showed that, for this type of reclaimed/recycled asphalt material, the test cannot be of sufficient duration, i.e. after 28 days the tests may not have reached equilibrium. Besides, for other types of asphalt material, as such reclaimed asphalt from open-graded asphalt layers (being almost “monogranular”) that are of very low compactability for the type of compaction applied according to the standard test procedure, may lead to very high hydraulic conductivity of test samples, and thus, to reduced duration of dynamic percolation test.
- Samples were obtained from a German project that involved the cold recycling of a tar containing material in 1991. Leaching data was obtained and this was compared to the current leaching behavior. The leaching levels are largely similar to what was observed in 1991 and significantly lower than the assessment of tar material (prior to recycling). This suggests that the encapsulation effect has been effective.
- Difficulties remain in assessing the effectiveness of cold recycling as a means of encapsulating tar (and other contaminants). The absence of a suitable test that meets all requirements has been found to be a significant shortcoming. The development of such a test should be a focus of future research work.
- With respect to leaching associated with reclaimed asphalt material containing crumb rubber, the results obtained in batch test (EN 12457-4) and in percolation tests ( CEN/TS 14405) were quite consistent, as follows:
    - PAH, leaching levels were very low (generally, below the quantification limit), in accordance to which was expected, since tar was not used in the original asphalt rubber mixtures. Besides, the leaching test data showed that any increase in PAH levels related to vehicle exhausts, lubricating oils or gasoline in the surface of the pavement, were not significant, in spite of the reclaimed asphalt rubber had been extracted from the wearing course of a pavement with about 8 years in service.;
    - Heavy metals (As, Cd, Cr, Co, Cu, Pb, Mn, Ni and Zn) leaching levels were, in general, relatively low. In eluate samples collected from batch tests (L/S=10 l/kg), only B11 (RAR-BE) leachate showed a Zinc concentration above the detection limit, but even so, quite far away from the maximum limit value for

waste acceptable at landfills for inert waste (Council Decision 2003/33/EC) . In the case of eluate samples collected from percolation tests, only Nickel concentration in the first fraction volume of the leachate (L/S=0.1 l/kg) was found to exceed the limit value required for the first extraction by the referred Council Decision. Nevertheless, it was observed that its concentration decreased significantly after the first fraction volume of the leachate, remaining in very low values in the following extractions. Thus, from the comparison between the results of percolation tests for the cumulative values at the end of the test (L=10 l/kg) with the correspondent values obtained in batch tests on same materials (RAR & RAR-BE) could be concluded that they were of the same order of magnitude ,

Still with regard to Nickel concentration, the decrease in about 30 % from RAR material (P2 samples) to RAR-BE material (P3 samples), suggests that Nickel metal may be partially encapsulated by cold recycling using bituminous emulsion.

- From the tank test it was observed that when the cold recycled mix was manufactured using RAP, cement and bituminous binder, the leaching performance fulfilled the requirements set by Dutch Ministry of Housing, Spatial Planning and the Environment or by the Landfill Regulation Amendment for England and Wales. As the RAP does not contain any tar materials, it can be expected that PAH content should be similar to a standard asphalt mix.
- When tank testing was used to assess the replacement of cement with fly-ash from two different sources, different behaviour was observed. In one case the threshold for arsenic was exceeded, while both materials also significantly exceeded the threshold for sulfates. It is considered that this problem originates from the raw materials used to produce the ash, in that the coal was partly replaced by biomass and municipal waste. This clearly shows the importance of carefully selecting the source of ash used for cold-recycling applications. This work also shows that the tank test is sensitive to changes in the material composition and suggests that the tank test is suitable for assessing the leaching of inorganic compounds in cold recycled applications.

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# Appendix I: Details of leaching test results – PAHs

| Compound | Detection Limit (µg/l) | Quantity Leached (µg/l) | | | |
|---|---|---|---|---|---|
| | | B1 Sample 1 | B1 Sample 2 | B1 Sample 3 | Blank |
| Naphthalene | <0.014 | 0.195 | 0.038 | 0.051 | bdl |
| Acenaphthylene | <0.013 | bdl | bdl | bdl | bdl |
| Acenaphthene | <0.013 | 0.023 | 0.026 | 0.025 | bdl |
| Fluorene | <0.014 | 0.023 | bdl | 0.025 | bdl |
| Phenanthrene | <0.011 | 0.023 | 0.026 | 0.038 | bdl |
| Anthracene | <0.013 | bdl | bdl | bdl | bdl |
| Fluoranthene | <0.012 | 0.023 | 0.026 | 0.063 | bdl |
| Pyrene | <0.013 | 0.023 | 0.026 | 0.063 | bdl |
| Benzo(a)anthracene | <0.015 | bdl | bdl | 0.025 | bdl |
| Chrysene | <0.011 | bdl | bdl | 0.025 | bdl |
| Benzo(bk)fluoranthene | <0.018 | bdl | bdl | 0.063 | bdl |
| Benzo(a)pyrene | <0.016 | bdl | bdl | 0.038 | bdl |
| Indeno(123cd)pyrene | <0.011 | bdl | bdl | 0.025 | bdl |
| Dibenzo(ah)anthracene | <0.01 | bdl | bdl | bdl | bdl |
| Benzo(ghi)perylene | <0.011 | bdl | bdl | 0.038 | bdl |
| Benzo(b)fluoranthene | <0.01 | bdl | bdl | 0.051 | bdl |
| Benzo(k)fluoranthene | <0.01 | bdl | bdl | 0.013 | bdl |
| **PAH 16 Total** | **<0.195** | **0.310** | **bdl** | **0.481** | **bdl** |

| **Compound** | **Detection Limit (µg/l)** | **Quantity Leached (µg/l)** | | | |
|---|---|---|---|---|---|
| | | **B2 Sample 1** | **B2 Sample 2** | **B2 Sample 3** | **Blank** |
| Naphthalene | <0.014 | 10.758 | 10.903 | 10.118 | bdl |
| Acenaphthylene | <0.013 | 0.283 | 0.301 | 0.273 | bdl |
| Acenaphthene | <0.013 | 16.505 | 21.602 | 17.055 | bdl |
| Fluorene | <0.014 | 6.424 | 8.320 | 6.655 | bdl |
| Phenanthrene | <0.011 | 17.182 | 22.495 | 17.173 | bdl |
| Anthracene | <0.013 | 2.596 | 2.951 | 1.945 | bdl |
| Fluoranthene | <0.012 | 10.758 | 13.602 | 10.455 | bdl |
| Pyrene | <0.013 | 8.768 | 10.942 | 8.536 | bdl |
| Benzo(a)anthracene | <0.015 | 2.919 | 3.728 | 3.218 | bdl |
| Chrysene | <0.011 | 3.101 | 3.786 | 3.236 | bdl |
| Benzo(bk)fluoranthene | <0.018 | 5.192 | 6.330 | 6.082 | bdl |
| Benzo(a)pyrene | <0.016 | 3.717 | 4.592 | 4.300 | bdl |
| Indeno(123cd)pyrene | <0.011 | 2.283 | 2.680 | 2.691 | bdl |
| Dibenzo(ah)anthracene | <0.01 | 0.222 | 0.252 | 0.264 | bdl |
| Benzo(ghi)perylene | <0.011 | 2.657 | 2.951 | 3.036 | bdl |
| Benzo(b)fluoranthene | <0.01 | 3.737 | 4.553 | 4.382 | bdl |
| Benzo(k)fluoranthene | <0.01 | 1.455 | 1.777 | 1.700 | bdl |
| **PAH 16 Total** | **<0.195** | **93.333** | **115.437** | **95.000** | **bdl** |

| **Compound** | **Detection Limit (µg/l)** | **Quantity Leached (µg/l)** | | | |
|---|---|---|---|---|---|
| | | **B3 Sample 1** | **B3 Sample 2** | **B3 Sample 3** | **Blank** |
| Naphthalene | <0.014 | 3.060 | 2.510 | 3.340 | bdl |
| Acenaphthylene | <0.013 | 0.150 | 0.120 | 0.170 | bdl |
| Acenaphthene | <0.013 | 4.120 | 3.460 | 4.360 | bdl |
| Fluorene | <0.014 | 2.350 | 1.990 | 2.480 | bdl |
| Phenanthrene | <0.011 | 8.330 | 7.090 | 8.270 | 0.026 |
| Anthracene | <0.013 | 0.240 | 0.160 | 0.360 | bdl |
| Fluoranthene | <0.012 | 3.970 | 3.340 | 4.190 | bdl |
| Pyrene | <0.013 | 2.270 | 1.890 | 2.340 | bdl |
| Benzo(a)anthracene | <0.015 | 1.090 | 0.890 | 1.120 | bdl |
| Chrysene | <0.011 | 1.070 | 0.870 | 1.100 | bdl |
| Benzo(bk)fluoranthene | <0.018 | 1.680 | 1.260 | 1.670 | bdl |
| Benzo(a)pyrene | <0.016 | 0.920 | 0.690 | 0.930 | bdl |
| Indeno(123cd)pyrene | <0.011 | 0.480 | 0.380 | 0.500 | bdl |
| Dibenzo(ah)anthracene | <0.01 | 0.10 | 0.09 | 0.11 | bdl |
| Benzo(ghi)perylene | <0.011 | 0.360 | 0.240 | 0.320 | bdl |
| Benzo(b)fluoranthene | <0.01 | 1.21 | 0.91 | 1.20 | bdl |
| Benzo(k)fluoranthene | <0.01 | 0.47 | 0.35 | 0.47 | bdl |
| **PAH 16 Total** | **<0.195** | **30.190** | **24.980** | **31.260** | **bdl** |

| Compound | Detection Limit (µg/l) | Quantity Leached (µg/l) | | | |
|---|---|---|---|---|---|
| | | **B4 Sample 1** | **B4 Sample 2** | **B4 Sample 3** | **Blank** |
| Naphthalene | <0.014 | 1.026 | 1.014 | 0.958 | bdl |
| Acenaphthylene | <0.013 | bdl | bdl | bdl | bdl |
| Acenaphthene | <0.013 | bdl | bdl | bdl | bdl |
| Fluorene | <0.014 | bdl | bdl | bdl | bdl |
| Phenanthrene | <0.011 | 1.115 | 0.849 | 1.116 | bdl |
| Anthracene | <0.013 | bdl | bdl | 0.789 | bdl |
| Fluoranthene | <0.012 | 2.064 | 1.685 | 2.389 | bdl |
| Pyrene | <0.013 | 1.859 | 1.534 | 2.095 | bdl |
| Benzo(a)anthracene | <0.015 | 1.500 | 1.178 | 1.926 | bdl |
| Chrysene | <0.011 | 1.526 | 1.356 | 1.705 | bdl |
| Benzo(bk)fluoranthene | <0.018 | 2.846 | 2.274 | 3.442 | bdl |
| Benzo(a)pyrene | <0.016 | 1.500 | 1.219 | 1.853 | bdl |
| Indeno(123cd)pyrene | <0.011 | 1.128 | 0.932 | 1.284 | bdl |
| Dibenzo(ah)anthracene | <0.01 | bdl | bdl | bdl | bdl |
| Benzo(ghi)perylene | <0.011 | 0.910 | bdl | 1.042 | bdl |
| Benzo(b)fluoranthene | <0.01 | 2.051 | 1.644 | 2.474 | bdl |
| Benzo(k)fluoranthene | <0.01 | 0.795 | bdl | 0.968 | bdl |
| **PAH 16 Total** | **<0.195** | **15.474** | **bdl** | **18.600** | **bdl** |

| **Compound**<br>Analysed by GC-MS | **Reporting Limit (µg/l)** | **Quantity Leached (µg/l)** |
|---|---|---|
| | | **B5 Average values (*)** |
| Naphthalene | 0.100 | <0.100 |
| Acenaphthylene | 0.010 | <0.010 |
| Acenaphthene | 0.010 | <0.010 |
| Fluorene | 0.020 | <0.020 |
| Phenanthrene | 0.030 | <0.030 |
| Anthracene | 0.020 | <0.020 |
| Fluoranthene | 0.030 | <0.030 |
| Pyrene | 0.060 | <0.060 |
| Benz(a)anthracene | 0.010 | <0.010 |
| Chrysene | 0.010 | <0.010 |
| Benzo(b)fluoranthene | 0.010 | <0.010 |
| Benzo(k)fluoranthene | 0.010 | <0.010 |
| Benzo(a)pyrene | 0.020 | <0.020 |
| Indeno(1.2.3.cd)pyrene | 0.010 | <0.010 |
| Benzo(g.h.i)perylene | 0.010 | <0.010 |
| Dibenz(a.h)anthracene | 0.010 | <0.010 |
| **Sum of 16 PAH** | **0.370** | **<0.370** |
| **Sum of PAH (MoE)** | **0.19** | **<0.19** |
| **Sum of 6 PAH (WHO)** | **0.090** | **<0.090** |
| **Sum of 4 PAH** | **0.040** | **<0.040** |

* The laboratory hasn't provided individual samples values.

| **Compound** | **Detection Limit (µg/l)** | **Quantity Leached (µg/l)** | | | |
|---|---|---|---|---|---|
| | | **B6 Sample 1** | **B6 Sample 2** | **B6 Sample 3** | **Blank** |
| Naphthalene | <0.014 | 2.065 | 2.082 | 2.087 | bdl |
| Acenaphthylene | <0.013 | bdl | bdl | bdl | bdl |
| Acenaphthene | <0.013 | 0.120 | 0.102 | 0.098 | bdl |
| Fluorene | <0.014 | 0.087 | 0.082 | 0.076 | bdl |
| Phenanthrene | <0.011 | 0.283 | 0.224 | 0.196 | bdl |
| Anthracene | <0.013 | bdl | bdl | bdl | bdl |
| Fluoranthene | <0.012 | 0.141 | 0.102 | 0.087 | bdl |
| Pyrene | <0.013 | 0.076 | 0.061 | 0.043 | bdl |
| Benzo(a)anthracene | <0.015 | 0.033 | 0.020 | bdl | bdl |
| Chrysene | <0.011 | 0.033 | 0.020 | bdl | bdl |
| Benzo(bk)fluoranthene | <0.018 | 0.033 | 0.031 | bdl | bdl |
| Benzo(a)pyrene | <0.016 | 0.022 | bdl | bdl | bdl |
| Indeno(123cd)pyrene | <0.011 | bdl | bdl | bdl | bdl |
| Dibenzo(ah)anthracene | <0.01 | bdl | bdl | bdl | bdl |
| Benzo(ghi)perylene | <0.011 | bdl | bdl | bdl | bdl |
| Benzo(b)fluoranthene | <0.01 | 0.022 | 0.020 | bdl | bdl |
| Benzo(k)fluoranthene | <0.01 | bdl | bdl | bdl | bdl |
| **PAH 16 Total** | **<0.195** | **2.891** | **2.724** | **2.587** | **bdl** |

| **Compound** | **Detection Limit (µg/l)** | **Quantity Leached (µg/l)** | | | |
|---|---|---|---|---|---|
| | | **B7 Sample 1** | **B7 Sample 2** | **B7 Sample 3** | **Blank** |
| Naphthalene | <0.014 | 0.841 | 0.835 | 1.034 | 0.025 |
| Acenaphthylene | <0.013 | 0.216 | 0.308 | 0.276 | 0.025 |
| Acenaphthene | <0.013 | 5.193 | 5.582 | 5.483 | bdl |
| Fluorene | <0.014 | 2.114 | 2.297 | 2.264 | 0.025 |
| Phenanthrene | <0.011 | 7.364 | 8.978 | 9.264 | 0.037 |
| Anthracene | <0.013 | 1.682 | 1.989 | 2.264 | bdl |
| Fluoranthene | <0.012 | 8.045 | 10.637 | 9.989 | bdl |
| Pyrene | <0.013 | 7.443 | 10.187 | 9.034 | bdl |
| Benzo(a)anthracene | <0.015 | 3.170 | 4.736 | 4.483 | bdl |
| Chrysene | <0.011 | 3.727 | 5.648 | 4.356 | bdl |
| Benzo(bk)fluoranthene | <0.018 | 8.205 | 13.527 | 10.920 | bdl |
| Benzo(a)pyrene | <0.016 | 5.830 | 9.527 | 6.885 | bdl |
| Indeno(123cd)pyrene | <0.011 | 4.761 | 7.923 | 6.517 | bdl |
| Dibenzo(ah)anthracene | <0.01 | 0.386 | 0.648 | 0.494 | bdl |
| Benzo(ghi)perylene | <0.011 | 4.727 | 7.198 | 6.034 | bdl |
| Benzo(b)fluoranthene | <0.01 | 5.909 | 9.736 | 7.862 | bdl |
| Benzo(k)fluoranthene | <0.01 | 2.295 | 3.791 | 3.057 | bdl |
| **PAH 16 Total** | **<0.195** | **63.705** | **90.022** | **79.299** | **bdl** |

| **Compound** | **Detection Limit (µg/l)** | **Quantity Leached (µg/l)** | | | |
|---|---|---|---|---|---|
| | | **B8 Sample 1** | **B8 Sample 2** | **B8 Sample 3** | **Blank** |
| Naphthalene | <0.014 | 4.800 | 4.514 | 4.211 | bdl |
| Acenaphthylene | <0.013 | 0.086 | 0.097 | 0.085 | bdl |
| Acenaphthene | <0.013 | 5.614 | 5.597 | 5.310 | bdl |
| Fluorene | <0.014 | 2.157 | 2.208 | 2.014 | bdl |
| Phenanthrene | <0.011 | 3.943 | 4.181 | 3.803 | bdl |
| Anthracene | <0.013 | 0.400 | 0.486 | 0.423 | bdl |
| Fluoranthene | <0.012 | 0.771 | 0.806 | 0.803 | bdl |
| Pyrene | <0.013 | 0.500 | 0.528 | 0.521 | bdl |
| Benzo(a)anthracene | <0.015 | 0.057 | 0.042 | 0.042 | bdl |
| Chrysene | <0.011 | 0.057 | 0.069 | 0.070 | bdl |
| Benzo(bk)fluoranthene | <0.018 | 0.043 | 0.069 | 0.042 | bdl |
| Benzo(a)pyrene | <0.016 | 0.043 | 0.042 | 0.028 | bdl |
| Indeno(123cd)pyrene | <0.011 | 0.029 | 0.028 | bdl | bdl |
| Dibenzo(ah)anthracene | <0.01 | bdl | bdl | bdl | bdl |
| Benzo(ghi)perylene | <0.011 | 0.029 | 0.028 | 0.028 | bdl |
| Benzo(b)fluoranthene | <0.01 | 0.029 | 0.056 | 0.028 | bdl |
| Benzo(k)fluoranthene | <0.01 | bdl | 0.014 | bdl | bdl |
| **PAH 16 Total** | **<0.195** | **18.529** | **18.694** | **17.380** | **bdl** |

| Compound | Detection Limit (µg/l) | Quantity Leached (µg/l) | | | |
|---|---|---|---|---|---|
| | | B9 Sample 1 | B9 Sample 2 | B9 Sample 3 | Blank |
| Naphthalene | <0.014 | 0.051 | bdl | 0.080 | 0.026 |
| Acenaphthylene | <0.013 | 0.077 | bdl | 0.080 | bdl |
| Acenaphthene | <0.013 | 0.167 | bdl | 0.147 | bdl |
| Fluorene | <0.014 | 0.077 | bdl | 0.067 | bdl |
| Phenanthrene | <0.011 | 0.667 | 15.629 | 0.747 | bdl |
| Anthracene | <0.013 | 0.333 | 7.048 | 0.373 | bdl |
| Fluoranthene | <0.012 | 3.846 | 82.790 | 4.000 | 0.026 |
| Pyrene | <0.013 | 4.218 | 82.145 | 4.427 | 0.026 |
| Benzo(a)anthracene | <0.015 | 2.462 | 76.855 | 2.787 | bdl |
| Chrysene | <0.011 | 3.064 | 75.403 | 3.027 | bdl |
| Benzo(bk)fluoranthene | <0.018 | 6.923 | 156.242 | 6.787 | bdl |
| Benzo(a)pyrene | <0.016 | 3.795 | 90.855 | 3.827 | 0.026 |
| Indeno(123cd)pyrene | <0.011 | 2.359 | 56.790 | 2.653 | bdl |
| Dibenzo(ah)anthracene | <0.01 | 0.667 | 12.290 | 0.613 | bdl |
| Benzo(ghi)perylene | <0.011 | 1.744 | 41.694 | 1.973 | bdl |
| Benzo(b)fluoranthene | <0.01 | 4.987 | 112.500 | 4.880 | bdl |
| Benzo(k)fluoranthene | <0.01 | 1.936 | 43.742 | 1.907 | bdl |
| **PAH 16 Total** | **<0.195** | **30.449** | **697.742** | **31.587** | **bdl** |

| **Compound** | **Detection Limit (µg/l)** | **Quantity Leached (µg/l)** | | | |
|---|---|---|---|---|---|
| | | **B10 Sample 1** | **B10 Sample 2** | **B10 Sample 3** | **Blank** |
| Naphthalene | <0.014 | bdl | bdl | bdl | bdl |
| Acenaphthylene | <0.013 | bdl | bdl | bdl | bdl |
| Acenaphthene | <0.013 | 3.110 | 2.981 | bdl | bdl |
| Fluorene | <0.014 | bdl | bdl | bdl | bdl |
| Phenanthrene | <0.011 | 10.868 | 15.893 | 17.052 | 0.021 |
| Anthracene | <0.013 | bdl | 3.398 | 3.714 | bdl |
| Fluoranthene | <0.012 | 10.868 | 23.612 | 24.013 | bdl |
| Pyrene | <0.013 | 7.011 | 16.019 | 16.234 | bdl |
| Benzo(a)anthracene | <0.015 | 5.670 | 13.728 | 13.571 | bdl |
| Chrysene | <0.011 | 6.495 | 14.874 | 15.377 | bdl |
| Benzo(bk)fluoranthene | <0.018 | 9.692 | 23.184 | 23.870 | bdl |
| Benzo(a)pyrene | <0.016 | 4.736 | 11.777 | 13.078 | bdl |
| Indeno(123cd)pyrene | <0.011 | 3.165 | 7.864 | 7.857 | bdl |
| Dibenzo(ah)anthracene | <0.01 | bdl | 2.379 | bdl | bdl |
| Benzo(ghi)perylene | <0.011 | 3.099 | 7.699 | 7.351 | bdl |
| Benzo(b)fluoranthene | <0.01 | 6.978 | 16.689 | 17.182 | bdl |
| Benzo(k)fluoranthene | <0.01 | 2.714 | 6.495 | 6.688 | bdl |
| **PAH 16 Total** | **<0.195** | **64.714** | **143.408** | **142.117** | **bdl** |

| **Compound**<br>Analysed by GC-MS | **Reporting Limit (µg/l)** | **Quantity Leached (µg/l)** |
|---|---|---|
| | | **B11 Average values (*)** |
| Naphthalene | 0.100 | <0.100 |
| Acenaphthylene | 0.010 | <0.010 |
| Acenaphthene | 0.010 | <0.010 |
| Fluorene | 0.020 | <0.020 |
| Phenanthrene | 0.030 | 0.032 (a) |
| Anthracene | 0.020 | <0.020 |
| Fluoranthene | 0.030 | <0.030 |
| Pyrene | 0.060 | <0.060 |
| Benz(a)anthracene | 0.010 | <0.010 |
| Chrysene | 0.010 | <0.010 |
| Benzo(b)fluoranthene | 0.010 | <0.010 |
| Benzo(k)fluoranthene | 0.010 | <0.010 |
| Benzo(a)pyrene | 0.020 | <0.020 |
| Indeno(1.2.3.cd)pyrene | 0.010 | <0.010 |
| Benzo(g.h.i)perylene | 0.010 | <0.010 |
| Dibenz(a.h)anthracene | 0.010 | <0.010 |
| **Sum of 16 PAH** | **0.370** | **<0.370** |
| **Sum of PAH (MoE)** | **0.19** | **<0.19** |
| **Sum of 6 PAH (WHO)** | **0.090** | **<0.090** |
| **Sum of 4 PAH** | **0.040** | **<0.040** |

* The laboratory hasn’t provided individual samples values.

(a) Measurement uncertainty = ±26,0%

**Percolation test data obtained on sample P1: Column 1**

| Compound | Detection Limit (µg/l) | Quantity Leached (µg/kg) | | | | | | |
|---|---|---|---|---|---|---|---|---|
| | | 1st Extraction | 2nd Extraction | 3rd Extraction | 4th Extraction | 5th Extraction | 6th Extraction | 7th Extraction |
| Naphthalene | <0.014 | 0.512 | 1.082 | 7.853 | | 7.590 | 31.384 | 103.644 |
| Acenaphthylene | <0.013 | 0.049 | 0.059 | 0.306 | | 0.157 | 0.572 | 0.980 |
| Acenaphthene | <0.013 | 0.183 | 0.394 | 2.881 | | 2.986 | 12.960 | 44.411 |
| Fluorene | <0.014 | 0.073 | 0.208 | 1.423 | | 1.497 | 6.551 | 23.763 |
| Phenanthrene | <0.011 | 0.211 | 0.506 | 3.208 | | 3.873 | 16.364 | 68.981 |
| Anthracene | <0.013 | 0.026 | 0.060 | 0.398 | | 0.427 | 1.860 | 8.478 |
| Fluoranthene | <0.012 | 0.059 | 0.138 | 0.854 | | 1.001 | 3.233 | 14.188 |
| Pyrene | <0.013 | 0.030 | 0.066 | 0.420 | | 0.487 | 1.516 | 6.863 |
| Benzo(a)anthracene | <0.015 | 0.005 | 0.008 | 0.050 | | 0.070 | 0.143 | 0.519 |
| Chrysene | <0.011 | 0.005 | 0.010 | 0.050 | | 0.070 | 0.172 | 0.634 |
| Benzo(bk)fluoranthene | <0.018 | 0.003 | 0.002 | bdl | | 0.035 | 0.057 | 0.231 |
| Benzo(a)pyrene | <0.016 | 0.003 | 0.002 | bdl | | 0.017 | 0.057 | 0.115 |
| Indeno(123cd)pyrene | <0.011 | bdl | bdl | bdl | | bdl | bdl | bdl |
| Dibenzo(ah)anthracene | <0.01 | bdl | bdl | bdl | | bdl | bdl | bdl |
| Benzo(ghi)perylene | <0.011 | bdl | bdl | bdl | | bdl | bdl | bdl |

**Percolation test data obtained on sample P1: Column 2**

| Compound | Detection Limit (µg/l) | Quantity Leached (µg/kg) | | | | | | |
|---|---|---|---|---|---|---|---|---|
| | | 1st Extraction | 2nd Extraction | 3rd Extraction | 4th Extraction | 5th Extraction | 6th Extraction | 7th Extraction |
| Naphthalene | <0.014 | 0.517 | 1.041 | 9.325 | 5.180 | 12.930 | 12.645 | 48.042 |
| Acenaphthylene | <0.013 | 0.045 | 0.058 | 0.487 | 0.201 | 0.246 | 0.265 | 0.517 |
| Acenaphthene | <0.013 | 0.215 | 0.471 | 4.146 | 2.287 | 5.855 | 5.723 | 21.421 |
| Fluorene | <0.014 | 0.101 | 0.264 | 2.233 | 1.217 | 3.016 | 3.032 | 10.745 |
| Phenanthrene | <0.011 | 0.280 | 0.743 | 6.203 | 3.539 | 7.885 | 8.363 | 33.095 |
| Anthracene | <0.013 | 0.032 | 0.094 | 0.705 | 0.481 | 0.950 | 1.074 | 4.443 |
| Fluoranthene | <0.012 | 0.084 | 0.209 | 1.939 | 1.061 | 2.370 | 2.299 | 8.059 |
| Pyrene | <0.013 | 0.042 | 0.101 | 0.906 | 0.530 | 1.150 | 1.124 | 3.857 |
| Benzo(a)anthracene | <0.015 | 0.007 | 0.011 | 0.092 | 0.071 | 0.117 | 0.114 | 0.413 |
| Chrysene | <0.011 | 0.007 | 0.012 | 0.109 | 0.071 | 0.141 | 0.126 | 0.482 |
| Benzo(bk)fluoranthene | <0.018 | 0.002 | 0.002 | 0.025 | 0.018 | 0.059 | 0.038 | 0.207 |
| Benzo(a)pyrene | <0.016 | 0.002 | 0.002 | 0.017 | 0.013 | 0.035 | 0.025 | 0.103 |
| Indeno(123cd)pyrene | <0.011 | bdl | bdl | bdl | bdl | bdl | bdl | bdl |
| Dibenzo(ah)anthracene | <0.01 | bdl | bdl | bdl | bdl | bdl | bdl | bdl |
| Benzo(ghi)perylene | <0.011 | bdl | bdl | bdl | bdl | bdl | bdl | bdl |

**Percolation test data obtained on sample P1: Column 3**

| Compound | Detection Limit (µg/l) | Quantity Leached (µg/kg) | | | | | | |
|---|---|---|---|---|---|---|---|---|
| | | 1st Extraction | 2nd Extraction | 3rd Extraction | 4th Extraction | 5th Extraction | 6th Extraction | 7th Extraction |
| Naphthalene | <0.014 | 0.537 | 0.744 | 3.129 | 3.721 | 10.192 | 26.360 | 26.504 |
| Acenaphthylene | <0.013 | 0.039 | 0.053 | 0.199 | 0.181 | 0.275 | 0.672 | 0.297 |
| Acenaphthene | <0.013 | 0.177 | 0.393 | 1.592 | 1.821 | 5.123 | 13.572 | 12.184 |
| Fluorene | <0.014 | 0.085 | 0.194 | 0.866 | 0.946 | 2.578 | 6.940 | 6.270 |
| Phenanthrene | <0.011 | 0.279 | 0.679 | 2.680 | 2.709 | 7.415 | 19.420 | 20.016 |
| Anthracene | <0.013 | 0.029 | 0.095 | 0.406 | 0.358 | 1.058 | 2.323 | 2.077 |
| Fluoranthene | <0.012 | 0.103 | 0.209 | 0.878 | 0.917 | 2.336 | 5.429 | 5.597 |
| Pyrene | <0.013 | 0.068 | 0.102 | 0.437 | 0.460 | 1.179 | 2.714 | 2.828 |
| Benzo(a)anthracene | <0.015 | 0.015 | 0.012 | 0.047 | 0.058 | 0.165 | 0.280 | 0.257 |
| Chrysene | <0.011 | 0.014 | 0.015 | 0.051 | 0.058 | 0.143 | 0.336 | 0.297 |
| Benzo(bk)fluoranthene | <0.018 | 0.014 | 0.005 | 0.012 | 0.012 | 0.033 | 0.056 | bdl |
| Benzo(a)pyrene | <0.016 | 0.008 | 0.003 | 0.008 | 0.008 | 0.033 | bdl | 0.040 |
| Indeno(123cd)pyrene | <0.011 | 0.003 | bdl | bdl | bdl | bdl | bdl | bdl |
| Dibenzo(ah)anthracene | <0.01 | 0.001 | bdl | bdl | bdl | bdl | bdl | bdl |
| Benzo(ghi)perylene | <0.011 | 0.002 | bdl | bdl | bdl | bdl | bdl | bdl |

### Percolation test data obtained on sample P2: Column 1

| **Compound**<br>Analysed by GC-MS | **Reporting Limit (µg/l)** | **Quantity Leached (µg/l)** |
|---|---|---|
| | | **1st Extraction** |
| Naphthalene | 0.100 | <0.100 |
| Acenaphthylene | 0.010 | <0.010 |
| Acenaphthene | 0.010 | <0.010 |
| Fluorene | 0.020 | <0.020 |
| Phenanthrene | 0.030 | <0.030 |
| Anthracene | 0.020 | <0.020 |
| Fluoranthene | 0.030 | <0.030 |
| Pyrene | 0.060 | <0.060 |
| Benz(a)anthracene | 0.010 | <0.010 |
| Chrysene | 0.010 | <0.010 |
| Benzo(b)fluoranthene | 0.010 | <0.010 |
| Benzo(k)fluoranthene | 0.010 | <0.010 |
| Benzo(a)pyrene | 0.020 | <0.020 |
| Indeno(1.2.3.cd)pyrene | 0.010 | <0.010 |
| Benzo(g.h.i)perylene | 0.010 | <0.010 |
| Dibenz(a.h)anthracene | 0.010 | <0.010 |
| **Sum of 16 PAH** | **0.370** | **<0.370** |
| **Sum of PAH (MoE)** | **0.19** | **<0.19** |
| **Sum of 6 PAH (WHO)** | **0.090** | **<0.090** |
| **Sum of 4 PAH** | **0.040** | **<0.040** |

### Percolation test data obtained on sample P3: Column 1

| **Compound**<br>Analysed by GC-MS | **Reporting Limit (µg/l)** | **Quantity Leached (µg/l)** |
|---|---|---|
| | | **1st Extraction** |
| Naphthalene | 0.100 | <0.100 |
| Acenaphthylene | 0.010 | <0.010 |
| Acenaphthene | 0.010 | <0.010 |
| Fluorene | 0.020 | <0.020 |
| Phenanthrene | 0.030 | <0.030 |
| Anthracene | 0.020 | <0.020 |
| Fluoranthene | 0.030 | <0.030 |
| Pyrene | 0.060 | <0.060 |
| Benz(a)anthracene | 0.010 | <0.010 |
| Chrysene | 0.010 | <0.010 |
| Benzo(b)fluoranthene | 0.010 | <0.010 |
| Benzo(k)fluoranthene | 0.010 | <0.010 |
| Benzo(a)pyrene | 0.020 | <0.020 |
| Indeno(1.2.3.cd)pyrene | 0.010 | <0.010 |
| Benzo(g.h.i)perylene | 0.010 | <0.010 |
| Dibenz(a.h)anthracene | 0.010 | <0.010 |
| **Sum of 16 PAH** | **0.370** | **<0.370** |
| **Sum of PAH (MoE)** | **0.19** | **<0.19** |
| **Sum of 6 PAH (WHO)** | **0.090** | **<0.090** |
| **Sum of 4 PAH** | **0.040** | **<0.040** |

# Appendix II: Details of leaching test results – Metals

### Batch test data obtained on sample B5

| **Compound**<br>Analysed by ICP-AES | **Detection Limit (mg/l)** | **Quantity Leached (mg/l)** | | |
|---|---|---|---|---|
| | | B5 Sample 1 | B5 Sample 2 | **B5 average** |
| Cadmium, Cd | 0.002 | <0.002 | <0.002 | **<0.002** |
| Cobalt, Co | 0.020 | <0.020 | <0.020 | **<0.020** |
| Copper, Cu | 0.013 | <0.013 | <0.013 | **<0.013** |
| Lead, Pb | 0.009 | <0.009 | <0.009 | **<0.009** |
| Manganese, Mn | 0.013 | <0.013 | <0.013 | **<0.013** |
| Nickel, Ni | 0.006 | <0.006 | <0.006 | **<0.006** |
| Zinc, Zn | 0.006 | <0.006 | 0.006 | **<0.006** |
| **Parameter** | - | B5 Sample 1 | B5 Sample 2 | **B5 average** |
| pH | - | 6.60 | 6.65 | **6.63** |
| Conductivity (µS/cm) | - | 7.19 | 7.18 | **7.19** |

| **Compound**<br>Analysed by ICP-MS | **Reporting Limit** (µg/l) | **Quantity Leached** (µg/l) |
|---|---|---|
| | | **B5 average (*)** |
| Lead, Pb | 1.0 | <1.0 |
| Nickel, Ni | 3.0 | <3.0 |

* The laboratory hasn't provided individual samples values.

**Batch test data obtained on sample B11**

| **Compound** Analysed by ICP-AES | **Detection Limit (mg/l)** | **Quantity Leached (mg/l)** | | |
|---|---|---|---|---|
| | | B11 Sample 1 | B11 Sample 2 | **B11 average** |
| Cadmium, Cd | 0.002 | <0.002 | <0.002 | **<0.002** |
| Cobalt, Co | 0.020 | <0.020 | <0.020 | **<0.020** |
| Copper, Cu | 0.013 | <0.013 | <0.013 | **<0.013** |
| Lead, Pb | 0.009 | <0.009 | <0.009 | **<0.009** |
| Manganese, Mn | 0.013 | <0.013 | <0.013 | **<0.013** |
| Nickel, Ni | 0.006 | <0.006 | <0.006 | **<0.006** |
| Zinc, Zn | 0.006 | 0.026 | 0.026 | **0.026** |
| **Parameter** | - | B11 Sample 1 | B11 Sample 2 | **B11 average** |
| pH | - | 6.60 | 6.65 | **6.63** |
| Conductivity (µS/cm) | - | 7.19 | 7.18 | **7.19** |

| **Compound** Analysed by ICP-MS | **Reporting Limit** (µg/l) | **Quantity Leached** (µg/l) |
|---|---|---|
| | | **B11 average (*)** |
| Lead, Pb | 1.0 | <1.0 |
| Nickel, Ni | 3.0 | <3.0 |

* The laboratory hasn't provided individual samples values.

## Percolation test data obtained on sample P2: Column 1

| **Compound** Analysed by ICP-MS | **Reporting Limit (µg/l)** | **Quantity Leached (µg/l)** | | | | | | |
|---|---|---|---|---|---|---|---|---|
| | | 1st Extraction | 2nd Extraction | 3rd Extraction | 4th Extraction | 5th Extraction | 6th Extraction | 7th Extraction |
| Arsenic, As (a) | 1.0 | 6.0 | - | - | - | - | - | - |
| Cadmium, Cd | 0.50 | <0.50 | - | - | - | - | - | - |
| Chromium, Cr | 5.0 | <5.0 | - | - | - | - | - | - |
| Cobalt, Co (a) | 0.50 | 8.00 | - | - | - | - | - | - |
| Copper, Cu (a) | 1.0 | 55.7 | - | - | - | - | - | - |
| Lead, Pb (a) | 1,0 | 25.2 | 11.8 | 5.1 | 2.4 | 1.4 | <1.0 | <1.0 |
| Manganese, Mn (a) | 0.50 | 137 | - | - | - | - | - | - |
| Nickel, Ni (a) | 3.0 | 308 | 36.7 | 30.2 | 26.4 | 20.2 | 8.0 | <3.0 |
| Zinc, Zn (a) | 2.0 | 254 | - | - | - | - | - | - |

(a) Measurement uncertainty = ±10,0%

## Percolation test data obtained on sample P3: Column 1

| **Compound** Analysed by ICP-MS | **Reporting Limit (µg/l)** | **Quantity Leached (µg/l)** | | | | | | |
|---|---|---|---|---|---|---|---|---|
| | | 1st Extraction | 2nd Extraction | 3rd Extraction | 4th Extraction | 5th Extraction | 6th Extraction | 7th Extraction |
| Arsenic, As | 1.0 | <1.0 | - | - | - | - | - | - |
| Cadmium, Cd | 0.50 | <0.50 | - | - | - | - | - | - |
| Chromium, Cr | 5.0 | <5.0 | - | - | - | - | - | - |
| Cobalt, Co (a) | 0.50 | 2.74 | - | - | - | - | - | - |
| Copper, Cu (a) | 1.0 | 145 | - | - | - | - | - | - |
| Lead, Pb (a) | 1,0 | 23.2 | 10.2 | 6.0 | 2.1 | 1.4 | 1.0 | <1.0 |
| Manganese, Mn (a) | 0.50 | 95.3 | - | - | - | - | - | - |
| Nickel, Ni (a) | 3.0 | 220 | 28.4 | 29.8 | 26.2 | 17.8 | 5.0 | <3.0 |
| Zinc, Zn (a) | 2.0 | 619 | - | - | - | - | - | - |

(a) Measurement uncertainty = ±10,0%