

Leaching of Heavy Metals and Polycyclic Aromatic Hydrocarbons from Reclaimed Asphalt; Application of an Up-flow Percolation Test

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Abstract

Reclaimed asphalt (RA) is a valuable secondary material which is currently greatly under-utilized. The optimum use of RA is in a road surface layer as this uses the engineering properties of the material to its full capacity. Uncertainties exist concerning the variable nature of the material and its potential to contribute to the pollution both of water bodies and air. Among the contaminants which could be released from RA are heavy metals and polycyclic aromatic hydrocarbons (PAHs). The widespread use of RA has many benefits, both financial and environmental. Environmental benefits include a reduction in landfill demand due to material from the demolition and re-surfacing of roads and a reduction in the demand for virgin aggregates.

A series of leaching tests on RA were conducted so that the material can be environmentally characterised and potential threats to the environment from its use identified. Draft CEN/TC 351 N0272 has been selected as the testing method following a review of suitable tests. This comprises an up-flow percolation test on size-reduced material. Seven samples are collected at specified liquid-to-solid (L/S) ratios over the course of the test from each column. The set-up comprises four columns and the test duration is approximately thirty days, terminating when a cumulative L/S ratio of 10 l/kg is reached.

Results from the test are reported as released quantity of a constituent per quantity of sample for analysis in the eluate fraction and the cumulatively released quantity of contaminants released in all of the eluate fractions. Examination of test results will also enable the identification of the different leaching mechanisms involved and their relative importance in leaching from RA.

Keywords: Reclaimed asphalt; leaching; heavy metals, PAHs

1. Introduction

Reclaimed asphalt is a valuable resource which is not currently used to its full technical potential. Much of the material is still either disposed of in landfills or used in low level applications such as in lower unbound road layers or as construction fill. More efficient use of RA involves using the material in applications for which it was originally designed i.e. re-using material removed from road surface layers in new surface layers. Higher rates of RA usage reduce the demand for landfill space and for virgin aggregates as well as lowering CO₂ emissions during the asphalt production process. However, in order to ensure that unnecessary restrictions are not placed on its usage due to lack of available data and that its use does not pose an environmental risk, the material must first be characterized. This research forms part of the European FP7 Re-Road project (2009 – 2012) which aims to fully environmentally characterize RA.

There are three potential sources of contamination associated with RA; firstly contaminants in the bitumen, additives and aggregates; secondly, contaminants which built up on the surface of the road during its previous pavement life/lives and thirdly, those contaminants which build up during the life of

the newly laid pavement containing RA. These contaminants may then be available for leaching into road runoff or water infiltrating the road surface or they can be blown from the surface by wind. Leaching tests, performed according to Draft CEN/TC 351 N0272 (CEN/TC351, 2010), a full characterisation test, were conducted on RA to assess the material. Here we present results for two of the materials tested; a tar-containing Swedish RA and an Irish RA from a mixed source stockpile. We refer to the samples simply as ‘TCA’ and ‘MSS’.

2. Experimental Method

2.1 Sample preparation

The MSS sample was prepared via the process of cryo-crushing. Material taken from the mixed source stockpile was roughly sorted so that large pieces of RA which were too big to be crushed were heated in the oven for approximately 1 hour at 80 °C - 100°C and crumbled by hand into smaller pieces. Once cool this material was combined with the rest of the material sourced from the stockpile. All material was then frozen at -20°C for 12 hours and then passed through a jaw crusher to achieve a particle size of 10mm. The size-reduced RA was then packed into glass columns with a stainless steel rammer in a standardized manner, using five separate layers with three sub-layers in each.

2.2 Test procedure

Figure 1 shows the percolation test set-up. 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 run in parallel. NaN_3 was used to inhibit microbial degradation of PAHs in the columns and was deemed necessary due to the long test duration. CaCl_2 was used so that the leachant had an ionic strength mimicking natural rainfall. Three of the columns were packed with 10mm RA particles and one remained empty (blank column). All columns were protected from light during the test. 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.

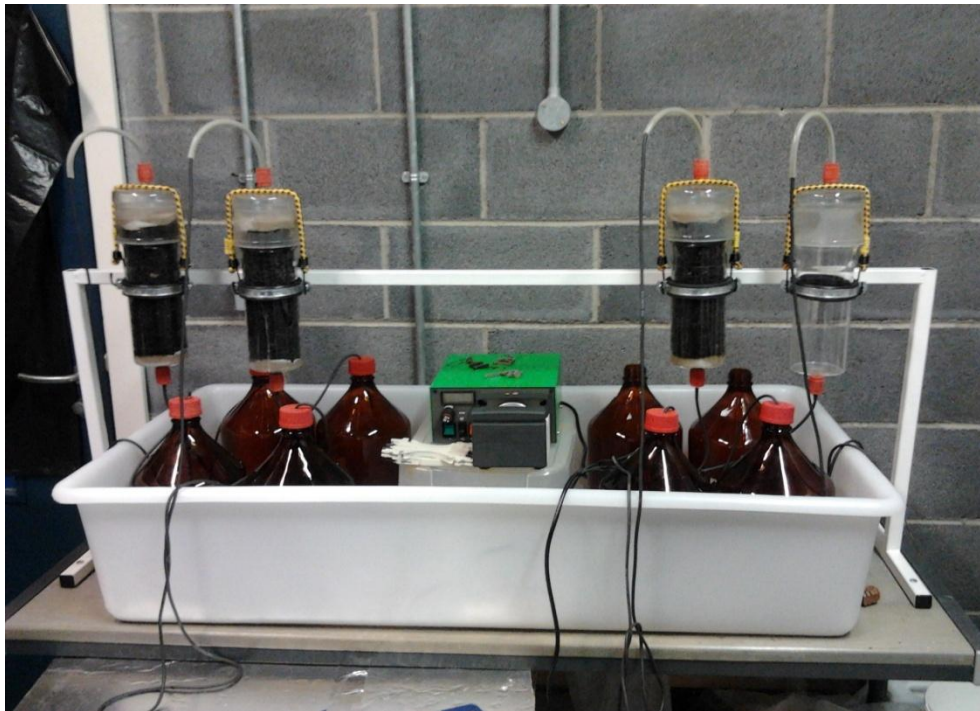


Figure 1. Percolation test set-up

A sample was collected from each of the three packed columns at the following cumulative (L/S) ratios; 0.1, 0.2, 0.5, 1.0, 2.0, 5.0 and 10.0 l/kg. Two samples were taken from the blank column; one at the beginning and one nearing the end of the test period. Approximately 100ml of each sample was acidified to pH < 2 for heavy metals analysis. For samples taken from L/S ratio 0.1 – 1.0 l/kg, the remainder of the sample was collected for PAH analysis. For fractions 5 – 7, corresponding to L/S ratios of 2.0 – 10.0 l/kg, a sub-sample was taken for PAH analysis due to the large sample volumes. Samples for PAH analysis were stored at 5°C and extracted with 20ml of hexane using C18 extraction disks within three days of collection.

2.3 Sample analysis

Following extraction into hexane, samples for PAH analysis were sent to a commercial laboratory where concentrations in the extracts were determined by gas chromatography – mass spectrometry (GC-MS). Atomic absorption spectrometry (AAS) with an air-acetylene flame was used for the inorganics analysis. Copper, zinc, nickel, manganese, cobalt, cadmium and lead concentrations in the samples were determined in this way.

3. Results

The concentrations of the heavy metals leached throughout the percolation test are shown below. Table 1 lists the quantities of metals in each sample fraction while Figure 2 shows the cumulative heavy metals leached from the TCA and MSS samples in µg/kg. The concentrations of all metals were similar in both leachates except for copper and manganese which were higher in the ‘TCA’ sample.

Table 1. Metal leaching from ‘TCA’ & ‘MSS’ samples

		Fraction						
Metal (µg/kg)	Sample	1	2	3	4	5	6	7
Copper	TCA	0.020	0.016	0.035	0.029	0.054	0.119	0.350
	MSS	0.004	0.000	0.024	0.013	0.063	0.100	0.122
Zinc	TCA	0.007	0.009	0.005	0.013	0.017	0.109	0.099
	MSS	0.010	0.012	0.018	0.003	0.031	0.055	0.015
Nickel	T	0.063	0.065	0.073	0.095	0.090	0.435	0.489
	MSS	0.013	0.012	0.045	0.067	0.145	0.374	0.661
Manganese	TCA	0.585	0.146	0.502	0.164	0.411	0.395	0.317
	MSS	0.004	0.003	0.009	0.011	0.021	0.055	0.108
Cobalt	TCA	0.007	0.005	0.008	0.025	0.064	0.198	0.248
	MSS	0.009	0.007	0.022	0.030	0.078	0.174	0.336
Cadmium	TCA	0.020	0.067	0.013	0.082	0.033	0.198	0.054
	MSS	0.005	0.004	0.011	0.014	0.026	0.082	0.124
Lead	TCA	0.050	0.054	0.108	0.161	0.342	1.317	1.734
	MSS	0.047	0.041	0.111	0.199	0.433	1.056	1.867

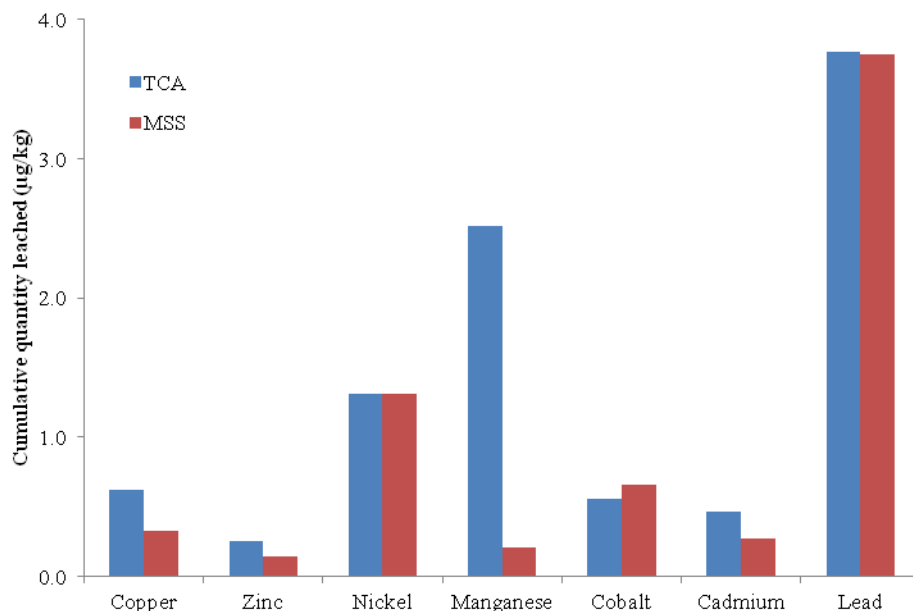


Figure 2. Cumulative heavy metals leached during percolation test

Figure 3 shows cumulative leached amounts of PAHs as the average of the sum of the three columns for each of the seven fractions for both the 'TCA' and 'MSS' samples in µg/kg. The concentration of the majority of the low and medium weight PAHs were comparable in both leachates except for naphthalene which is significantly higher and phenanthrene which is higher in the 'TCA' leachate and acenaphthene which is higher in the 'MSS' leachate. The concentration of higher molecular weight PAHs was below detection limits in many of the samples from both leachates but all of those measured were present in higher concentrations in the 'MSS' leachates.

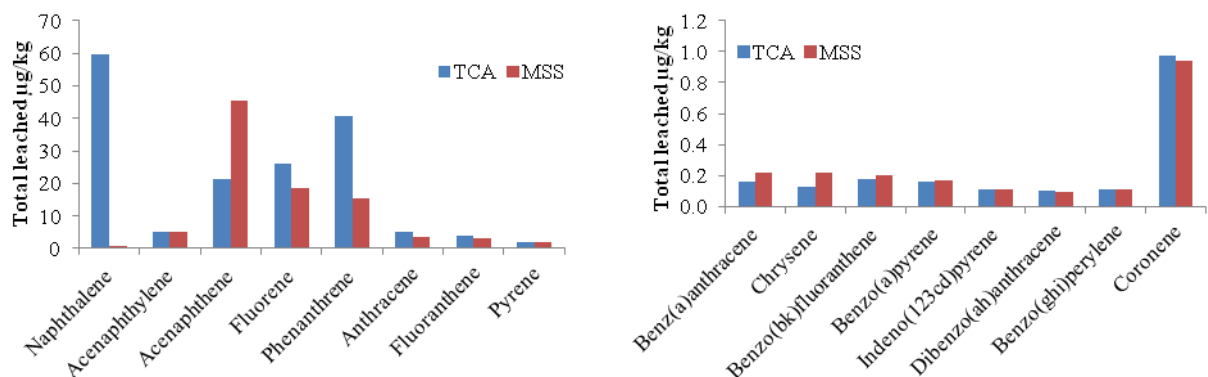


Figure 3 Total cumulative leached amounts of PAHs in percolation test eluate

4. Discussion

The 2011 CERCLA list of priority hazardous substances (Agency for Toxic Substances and Disease Registry, 2011) is generated using the algorithm given below, taking into account the frequency of occurrence of substances at US National Priorities List hazardous waste sites and facilities, substance toxicity, concentration in the environment and the potential for human exposure.

TOTAL SCORE=NPL FREQUENCY+TOXICITY + POTENTIAL FOR HUMAN EXPOSURE
 (1,800 max. points) (600 points) (600 points) (300 conc. pts.) + (300 exposure pts.)

Table 2. Position of relevant PAHs on the CERCLA list of hazardous substances

Relevant PAH	CERCLA Ranking
Benzo(a)pyrene	8
Benzo(b)fluoranthene	10
Benzo(a)anthracene	37
Naphthalene	80
Fluoranthene	133
Chrysene	142
Acenaphthene	168
Indeno(1,2,3-cd)pyrene	173
Pyrene	253

Based on this, it is possible to rank key PAHs of interest when assessing reclaimed asphalt as a pavement material. These are given in Table 2 above and provide a useful context for assessing the results of the leaching test.

4.1 Leaching behaviour: PAHs

Figure 4 shows naphthalene and fluoranthene leaching as a function of cumulative L/S ratio as both the number of µg leached per kg of dry mass of the RA in the column and the cumulative % of the total quantity leached at each L/S ratio.

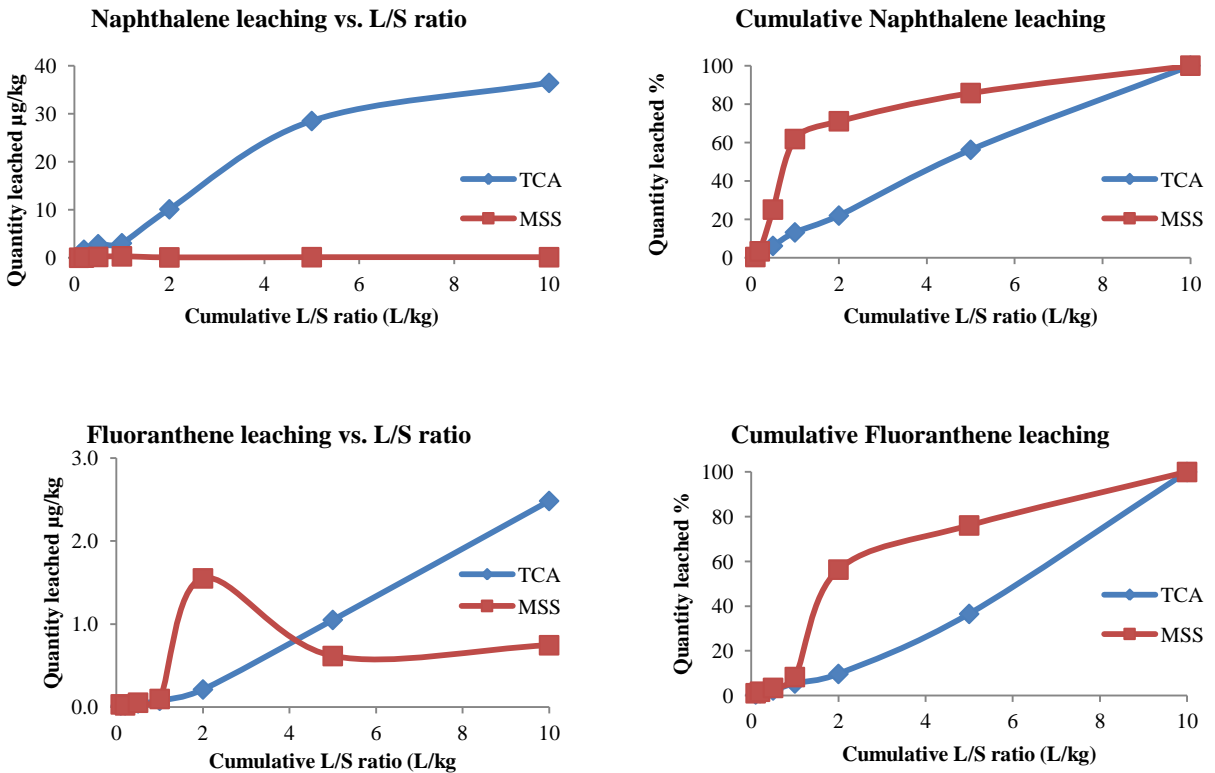


Figure 4. PAH leaching trends

Leaching appears to have occurred at a reasonably constant rate throughout the duration of the test from the 'TCA' sample, whereas there is a spike in concentration in the 'MSS' eluate which occurs approximately at L/S ratio of 2 l/kg for both compounds. This spike in concentration may be due to a deviation in prescribed leaching conditions caused by a pump malfunction and leakages from the columns, resulting in a longer contact time of the leachate with the sample due to a lower linear velocity through the columns.

The results give only a tentative indication of the leaching behavior of RA – further testing on 5 additional RA samples are planned as well as comparison of the 'TCA' leaching data with the Swedish Geotechnical Institute (SGI) who performed a duplicate test.

4.2 Leaching behaviour: Heavy metals

The heavy metals selected for study rank as follows on the CERCLA list of priority hazardous substances (Agency for Toxic Substances and Disease Registry, 2011); lead 2nd, cadmium 7th, cobalt 52nd, nickel 57th, zinc 75th, Copper 125th and manganese 140th. Figure 5 shows copper and zinc leaching trends from both samples in µg/kg per fraction and cumulatively released quantities per fraction as a percentage of the total leached amounts. The concentration of most of the heavy metals in both leachates generally increased with increasing L/S ratio. This is in contrast to the PAH trend observed with higher concentrations generally detected at the lower L/S ratios or in the earlier eluate fractions. Whilst the concentrations of heavy metals observed are low, it may be appropriate to increase the percolation test duration to further assess their leaching potential given that the concentrations generally increased with increased leaching time.

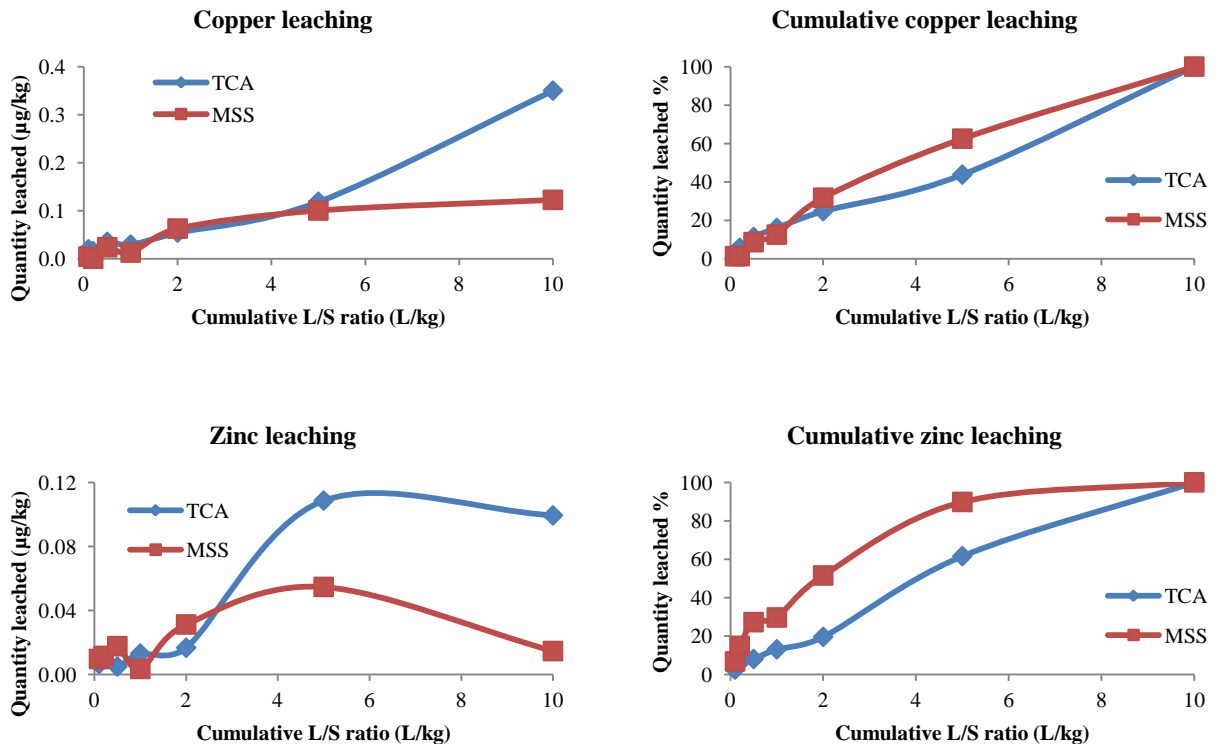


Figure 5 Copper and zinc leaching trends

Acknowledgement

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