Pergamon

Wat. Sci. Tech. Vol. 39, No. 2, pp. 103–109, 1999.

0273–1223/99 $19·00 + 0·00

PII: S0273-1223(99)00013-X

MINERAL OIL BIO-DEGRADATION WITHIN A PERMEABLE PAVEMENT: LONG TERM OBSERVATIONS

C. J. Pratt*, A. P. Newman** and P. C. Bond*

* *School of The Built Environment, Coventry University, Coventry, CV1 5FB, UK*
** *School of Natural and Environmental Sciences, Coventry University, Coventry, CV1 5FB, UK*

ABSTRACT

The paper reports on the first 300 days of a research project conducted at Coventry University, which has focused on the ability of a permeable pavement, reservoir structure to retain and treat petroleum-derived pollutants through *in situ* microbial bio-degradation. The research has required the construction of a full-scale model permeable pavement in the laboratory, which has been subjected to prolonged low-level hydrocarbon contamination, representative of typical loadings to urban surfaces such as highways and car parks. Water quality and bio-degradation indicators have been monitored over several months so that the capability of the permeable pavement to maintain a viable and effective microbial population could be assessed. The research has demonstrated that the structure can be used as an effective *in situ* aerobic bio-reactor.

KEYWORDS

Permeable pavement; reservoir structure; nutrients; bio-degradation; aerobic bio-reactor.

INTRODUCTION

Traditionally, stormwater runoff from impermeable surfaces has been intercepted and discharged swiftly to sewer systems and watercourses. Rapid growth of urban and industrial areas has resulted in an associated increase in impermeable surfaces such as roofs, highways and paved surfaces. This continual expansion and infilling is placing an increased burden on existing drainage networks and urban watercourses. During periods of heavy rain, large volumes of runoff may exceed the capacity of sewer systems, resulting in risks of flooding to property and to human health. In addition, pollutants deposited on impermeable surfaces may be entrained by stormwater flow, concentrated within sewer systems, and discharged to aquatic ecosystems with little or no treatment. Urban stormwater can contain toxins, such as heavy metals, oil and other hydrocarbons, whilst average levels of suspended solids may exceed that of untreated sewage (Imhoff, 1989). The majority of pollution in urban stormwater originates from non-point or diffuse sources. These are notoriously difficult to locate and quantify and may include wet and dry atmospheric deposition from industrial and domestic properties; traffic emissions; decomposed litter; de-icing salts; vegetative residues; pet faeces; and soil losses.

The construction of permeable highways as an alternative to impermeable surfaces has been shown to be an effective method of stormwater source control. The main design criterion for infiltration systems has usually been the reduction of peak discharge through the retention of stormwater flow. To date, little thought has been directed towards the use of such systems for the treatment of retained pollutants. Previous research has demonstrated the ability of a permeable pavement to retain both suspended solids and mineral oil (Pratt *et al.*, 1995, 1996).

In situ bio-remediation (microbial degradation) has been shown to be a potent technique for the breakdown of contaminants into less harmful forms, particularly in the fields of contaminated land remediation and oil-spill clean-up. Research initiated at Coventry University has been targeted towards using this technology to degrade petroleum-derived hydrocarbons within the sub-base of full-scale, laboratory model permeable pavement structures (Pratt *et al.*, 1996). To date, studies have indicated that the use of the permeable pavement structure as an *in situ* aerobic bio-reactor for the breakdown of petroleum-derived hydrocarbons is feasible. Flourishing microbial populations have been established for a period of approximately 30 days after dosing laboratory model pavement structures with high loadings of mineral oil, previously seeded with commercially available microbe and nutrient mixes (Brownstein, 1997).

The aim of the ongoing research described in this paper is to investigate whether a microbial population, subjected to chronic, low level hydrocarbon contamination will persist long-term within the structure under simulated field conditions. Research has concentrated on the effects on microbial survival of nutrient and water availability within the structure and their means of supply. It is hoped that the system will offer a long-term solution for the reduction of automobile-derived hydrocarbons on urban paved surfaces, which are washed into watercourses untreated via separate, storm sewers.

APPARATUS AND METHODS

The study was carried out in the laboratory, where environmental conditions could be controlled and indicators of microbial activity monitored. A representative section of the pavement was housed in a glass tank having a plan area of 610 mm x 610 mm, and a height of 780 mm (Brownstein, 1995; Pratt *et al.*, 1996). The pavement comprised pre-formed concrete blocks bedded on clean gravel, with vertical drainage provided through gravel-filled inlets between the blocks (Figure 1). A geotextile membrane (Exxon Terram 1000) separated the block bed from the underlying sub-base, comprising 600 mm depth of washed 20-50 mm granite. The entire structure rested on an additional geotextile underlay, supported by a stainless steel mesh, allowing effluent to flow into a collection funnel located at the base of the tank.

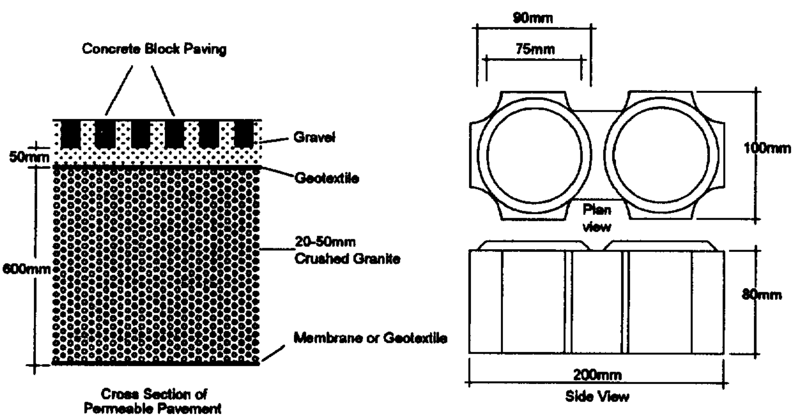

Figure 1. Cross section through permeable pavement, and detail of concrete blocks.

An oil dripper, comprising a covered glass funnel with Tygon-tubing stem and fishing-line insert, allowed oil to be dripped onto the surface of the pavement over a period of approximately 10 hours, simulating crank-case leakage. A compressor-driven rain-maker allowed rainfall events to be simulated using distilled water, whilst an outlet valve at the base of the rig permitted effluent to be removed periodically for analysis (Figure 2). Water quality was monitored by means of oil and grease concentration, chemical oxygen demand (COD), and pH.

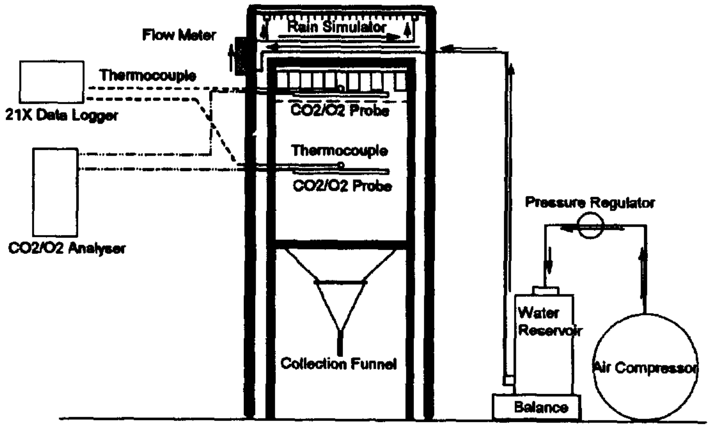

Figure 2. Rainfall simulation system and monitoring components.

Analysis of oil and grease in the effluent was performed to American Society for Testing and Materials method D 3921-85 (ASTM, 1985), a procedure adopted within the UK. The procedure involves liquid-liquid extraction of the effluent with carbon tetrachloride and analysis of CH_2 and CH_3 bond absorbance by infra-red spectrometry. The method was modified slightly, in that the extract was immersed in liquid nitrogen for ten minutes and then thawed in a water bath to break down the emulsion resulting from the lysis of bacterial cells. The solution was then filtered and dehydrated in the normal way prior to analysis. The average recovery was found to be 84.85% (SD 5.75, n=12), and all results have been recovery-corrected.

The COD of the effluent was evaluated using the ASTM Micro Spectrophotometric Procedure (ASTM, 1988). The method utilises digestion in a screw-top vial with pre-prepared reagents (supplied by Hach Europe, Belgium) and a spectrophotometer to measure absorbance at a wavelength of 420 nm.

The pH of the simulated rainfall and the effluent were measured using a Jenway (UK) 3071 pH meter, with a Fisons (UK) glass and Ag/AgCl electrode.

Microbial activity was evaluated by dipslide counts of colony forming units (CFU) (nutrient agar dipslides supplied by Abinghurst, UK). An indication of microbial respiration was obtained through the use of carbon dioxide, oxygen and temperature probes positioned mid-depth in both the pea-gravel bedding layer and the granite sub-base (Brownstein, 1995).

RAINFALL, OIL, NUTRIENT AND SAMPLING REGIMES

The weekly rainfall regime comprised two random 2121 ml events lasting 3 hours and 33 minutes each. This provided an annual rainfall of 594 mm, the average yearly rainfall occurring in London (Wallén, 1970). In addition the frequency of rainfall (an average of once every 3.5 days) and the intensity of rainfall (1.6 mm/h) compared favourably with London figures (once every 3.2 days at 1.4 mm/h).

A literature review on the quantity of oil and grease deposited on urban surfaces derived from concentrations in run-off, indicated an average loading of 178 mg/m^2/week. However, as there is a dearth of information available on the percentage of deposited hydrocarbons reaching drainage systems, it was estimated that this loading represented only one percent of the total amount of oil and grease settling on urban surfaces. Recent research has indicated that this was a close approximation, with typical oil retention values for tarmac and concrete surfaces being 97.7 and 98.2 percent, respectively (Newman *et al.*, 1998). Oil additions of 3.3 g occurred twice-weekly to the permeable pavement, i.e. 100 times the average value derived from the literature review. Positions and times of oil additions on the surface of the model pavement were selected randomly.

Rain simulation was commenced on Day 0, whilst oil application was started on Day 19: both inputs continued throughout the study. On Day 48, 250 ml of oil degrading microbe mixture and 100 ml of a proprietary liquid fertiliser (both supplied by Bio-Logix Limited, UK) were added to the surface of the pavement. This constituted a nitrogen: phosphorus: potassium (NPK) addition of 2.5 g : 0.5 g : 0.04 g, respectively. No further microbial seeding of the structure was made throughout the study. Further 100 ml applications of liquid fertiliser took place on Days 118 and 183. On Day 237 18 g of Osmocote, slow-release fertiliser (NPK 14:13:13, produced by Scotts Europe B.V., The Netherlands), was added to the surface of the structure in 25 plastic baskets. This addition constituted the same amount of nitrogen present in one application of liquid fertiliser.

Effluent samples were obtained after each rainfall event; values for pH and CFU were determined; and samples were then acidified and refrigerated prior to COD and oil and grease analysis. Carbon dioxide and oxygen measurements were taken at intervals of four hours, whilst temperature data was recorded every fifteen minutes. Both sets of data were continually downloaded to a PC for analysis.

RESULTS

After the first addition, subsequent applications of liquid fertiliser were associated with reductions in COD and oil and grease concentrations in the effluent (Figure 3). Nutrient additions were reflected by elevated carbon dioxide and depressed oxygen levels within the structure. Figure 4 illustrates delta values (i.e. measured values minus ambient values) for these gases over the study period. Increased microbial activity was only maintained for 30 to 60 days following the addition of liquid fertiliser. Biological activity appeared to be rejuvenated by each application of the liquid fertiliser. A gradual reduction in nutrient availability due to dilution and leaching, was assumed to be responsible for the observed decrease in metabolic indicators over time.

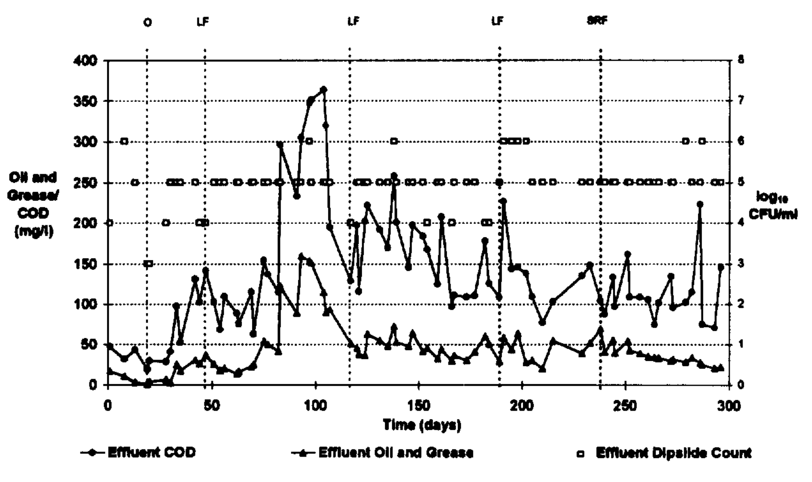

Figure 3. Variations in COD, total oil and grease, and CFU over the study period.

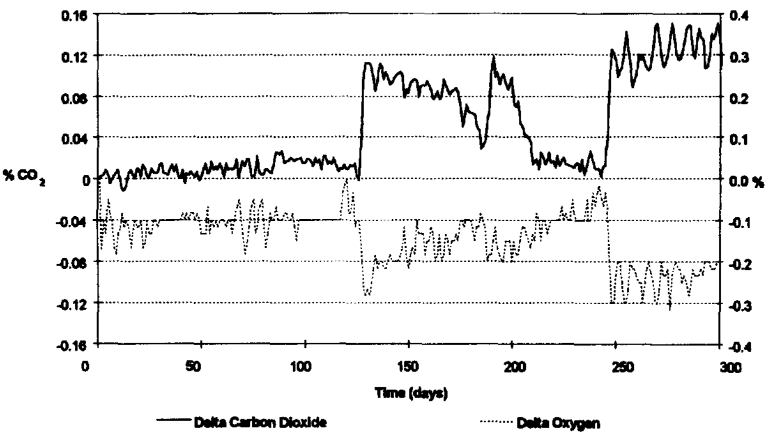

Figure 4. Delta gas levels recorded for the sub-base of the permeable pavement.

Slow-release fertiliser, used successfully in oil spill clean-up operations, was used in an attempt to counteract this 'tailing off' of biological activity. It was hoped that with the application of Osmocote on day 237, a stable oil-degrading microbial population could be maintained for a period of up to 6 months. Initial observations have been encouraging with elevated carbon dioxide (average 0.12% above ambient) and depressed oxygen (average 0.25% below ambient) remaining constant throughout the period.

From day 237, oil and grease levels decreased consistently from 69 to 22 mg/l. COD, although more variable possibly in response to 'sloughing off' of biofilm from the structure, decreased from 10^4 to 73 mg/l. CFU/ml averaging consistently about 10^6 suggested a stable biomass within the structure. A noticeable rise in structure temperatures, particularly within the sub-base, was evident after day 237 and was interpreted as indicating increased bacterial metabolic activity associated with the addition of the slow-release fertiliser.

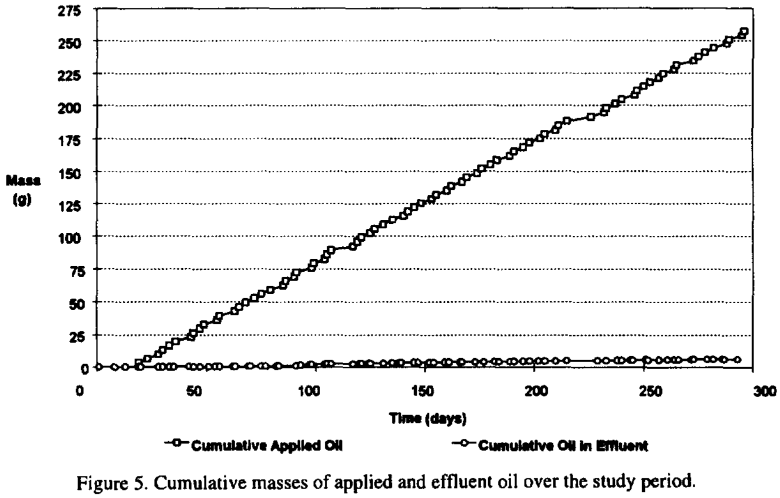

Figure 5. Cumulative masses of applied and effluent oil over the study period.

A total of 257.4g of oil was added to the structure during the study period, with only 6.3g recovered in the effluent, which represented an interception/retention efficiency of 97.6% (Figure 5). Although a percentage of this figure will be due to retention by the pavement construction materials, data from gas, temperature and dipslide observations suggest active breakdown of hydrocarbons was occurring, concentrated within the sub-base. Levels of oil and grease and COD in the effluent reached maxima of 150 mg/l and 350 mg/l at day 98, subsequently, the concentrations declined steadily to 22 mg/l and 73 mg/l, respectively. Analysis of infra-red spectra from effluent extracts of oil and grease indicate that after the third addition of liquid fertiliser a marked decrease in the ratio of CH_2 to terminal CH_3 groups occurred; further evidence for the biological breakdown of long-chain hydrocarbons into shorter molecules. pH analysis of mildly acidic simulation rainfall (pH 4.5-6.5) and effluent indicated that the construction materials were having a buffering effect, maintaining an effective pH of about 7.0, which is beneficial to microbial growth.

CONCLUSIONS

The development of permeable pavements as pollution treatment devices offers a potential solution to the problem of uncontrolled discharge of stormwater-associated contaminant loadings. Traditionally, stormwater runoff conveyed by separate surface water systems has been regarded as relatively clean and thus has been discharged directly to watercourses with little or no treatment. This has resulted not only in aesthetically displeasing and environmentally damaging oil sheens on water bodies, but also in the introduction of carcinogenic compounds, such as benzopyrene and benzanthracene, into food chains. By breaking down petroleum-derived hydrocarbons at source, both the health of aquatic organisms and that of fish and, hence possibly humans could benefit.

The research has shown that the permeable pavement has performed as an effective *in situ* aerobic bio-reactor, reducing petroleum contamination in the effluent to 2.4% of the oil applied to the pavement: this represents an effluent discharge equivalent to $22g/m^2$/year from an oil deposition on the pavement surface of some $900g/m^2$/year. Nutrient supply appears to be a limiting factor in the efficiency of the structure to degrade oil. Liquid fertiliser may be rapidly diluted and leached from the structure, leading to a reduction in microbial activity. The use of slow-release fertiliser has resulted in a constant, low level supply of nutrients to the biomass, which has promoted sustained oil-degradation within the structure. The recent development of oleophillic slow-release fertilisers for the oil-spill clean-up industry may also show promise. It is hoped that their future use on the pavement might allow nutrients to be released proportionately to contamination, extending their residence time within the structure and reducing effluent concentration. Effective use of the nutrients must be ensured, otherwise there is the danger that high levels in the effluent may cause eutrophication problems in receiving waters. Research continues in order to identify the proportion of the retained oil which is degraded, as compared with that which is simply retained long-term within the pavement construction.

So far, only unused, clean motor oil has been applied to the structure in the laboratory studies reported. Clean motor oil contains very low levels of poly-aromatic hydrocarbons (PAH), however, high temperatures and pressures within automobile engines result in high levels of PAH being released in the emissions. Further studies will be conducted utilising used mineral oil.

The structure will continue to be monitored to determine the effective duration of a single application of slow-release fertiliser. Provisional results are encouraging, with microbial activity maintained in excess of 12 months from one application of slow-release fertiliser (Pratt *et al.*, 1998). It is hoped that ultimately such a system may be widely used to ameliorate the input of petroleum-derived hydrocarbons into urban watercourses, from highways, car-parks and oil-handling facilities.

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