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# Environmental impact of highway construction and repair materials on surface and ground waters: Case study: crumb rubber asphalt concrete

Mohammad F. Azizian, Peter O. Nelson\*,  
Pugazhendhi Thayumanavan, Kenneth J. Williamson

*Department of Civil, Construction, and Environmental Engineering, Oregon State University,  
Corvallis, OR 97331-2302, USA*

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## Abstract

The practice of incorporating certain waste products into highway construction and repair materials (CRMs) has become more popular. These practices have prompted the National Academy of Science, National Cooperative Highway Research Program (NCHRP) to research the possible impacts of these CRMs on the quality of surface and ground waters. State department of transportations (DOTs) are currently experimenting with use of ground tire rubber (*crumb rubber*) in bituminous construction and as a crack sealer. Crumb rubber asphalt concrete (CR-AC) leachates contain a mixture of organic and metallic contaminants. Benzothiazole and 2(3H)-benzothiazolone (organic compounds used in tire rubber manufacturing) and the metals mercury and aluminum were leached in potentially harmful concentrations (exceeding toxic concentrations for aquatic toxicity tests). CR-AC leachate exhibited moderate to high toxicity for algae (*Selenastrum capricornutum*) and moderate toxicity for water fleas (*Daphnia magna*). Benzothiazole was readily removed from CR-AC leachate by the environmental processes of soil sorption, volatilization, and biodegradation. Metals, which do not volatilize or photochemically or biologically degrade, were removed from the leachate by soil sorption. Contaminants from CR-AC leachates are thus degraded or retarded in their transport through nearby soils and ground waters.

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## 1. Introduction

Assessment of the environmental impact of highway construction and repair materials (CRMs) has been a challenge to many of the regulatory agencies involved in the management of water resources that receive runoff from highway environments. These materials are often waste by-products from other industries, such as foundries and incinerators, or recycled products, such as rubber tires or roofing shingles. During the wet seasons, there is a potential for leaching of the chemical constituents in these materials and the possibility of transport to adjacent surface and subsurface water bodies. The leachates from highway CRMs are typically chemically complex, often containing a combination of

both organics and inorganics in solution. Many factors influence the release of specific constituents from a construction material, including major element chemistry, pH, redox (reduction/oxidation) conditions, chemical complexation, soil sorption, and biological activity. Many of the studies found in the literature do not specifically address the environmental impacts of the construction materials themselves. Rather, the focus has been with runoff contaminants caused by road use, erosion from construction activities, and other factors.

The scope of this study was to apply a validated chemical and toxicity evaluation methodology to assess the leaching behavior of crumb rubber asphalt concrete (CR-AC) in highway environments and the removal of toxic constituents in the leachate by environmental removal/reduction/retardation (RRR) processes.

A study reported by US EPA (1973) discusses methods for controlling pollution from construction activity. This report indicates that the construction materials

\* Corresponding author. Tel.: +1-541-737-6835; fax: +1-541-737-3099.

E-mail address: peter.nelson@orst.edu (P. O. Nelson).

used, such as asphalt, may themselves contaminate runoff, and should be controlled. This report also focuses on the prevention of such pollution through erosion control practices, for example, but does not discuss the impact of construction materials on the environment. Other reports (Scheidt, 1967; Gupta et al., 1981; Driscoll et al., 1989) detail the typical constituents of highway runoff, their possible sources, and the potential impact on the surrounding environment. The reported contaminants result from engine exhaust, deicing chemicals, herbicides, and grease and oil, among other sources. None of the studies found in the literature discuss the impact of the highway material on water quality.

Studies have compiled information about leaching tests to evaluate the use of solid wastes in highway construction (Bauw et al., 1991; Environment Canada, 1990, 1991; de Groot and van der Sloot, 1991; Fallman and Hartlen, 1994; Mulder, 1991). In general, these studies have shown that leaching behavior within and between materials is controlled by geochemical characteristics. Leaching tests using a variety of leaching solutions have been used for characterizing CRMs, for regulatory compliance, and for simulating on-site effects (van der Sloot et al., 1994; Harrington and Kins, 1966). Because of the slow kinetics of such leaching, several projects have focused upon the development of accelerated leaching tests to predict long term leaching behavior (Caldwell et al., 1994; Meij and Schaftnaar, 1994; van der Sloot et al., 1989, 2000; de Groot and van der Sloot, 1992; Eighmy et al., 1997).

Mainprize et al. (1976) reported that various benzothiazoles are likely to be present in the effluents from factories manufacturing certain rubber additives, particularly benzothiazole itself (BTH), 2-hydroxybenzothiazole (BTOH), 2-mercaptobenzothiazole (MBT) and benzothiazole-2-sulphonic acid (BTSA). Reddy and Quinn (1997) found that benzothiazole, 2-hydroxybenzothiazole, and 2-(4-morpholino) benzothiazole were the major organic compounds that leach from rubber and asphalt containing 1–3% rubber.

Significant research has indicated that laboratory toxicity testing is a useful tool for predicting the effects in the field if carried out in an appropriate manner. Specific support for the use of toxicity testing and the overall rationale for applying laboratory results to field conditions are reported in literature (Mount et al., 1986; Thomas et al., 1986; Peterson et al., 1985). A primary advantage of toxicity tests over the exclusive use of chemical analyses of CRM leachates is the ability to better assess the effects of unidentified chemical constituents, and hence their presence.

In this study, two types of organisms were chosen: algae (*Selenastrum capricornutum*) and water fleas (*Daphnia magna*). *Selenastrum capricornutum* represents plant species while *D. magna* represents animal species. It was considered of critical importance that both a

plant and an animal species be selected for testing of the highway CRMs. Plants and animals have biological differences that would cause them to react differently (or not at all) to different chemicals. Algae was used most often due to its sensitivity and ease of use. *Daphnia* testing was conducted for most samples but does not provide a high level of sensitivity compared with algae. The organisms chosen for this study were selected by taking into account ease of use, expense, accuracy, and sensitivity.

## 2. Experimental approach

A methodology has been developed that uses a series of laboratory tests to evaluate important processes that affect the chemical composition, aquatic toxicity, and fate of leachates from CRMs in highway applications. Leaching from the highway surface is applicable to both the permeable and impermeable highway reference environments as shown in Fig. 1 a–f (Nelson et al., 2001). The leaching tests provide information on the mobility of constituents in CRMs under a range of conditions thought to provide reasonable estimates of expected leachate chemical concentrations. RRR process tests provide information on the removal of leachate constituents by natural processes (Fig. 2).

### 2.1. Batch leaching test

The batch leaching test generally consists of solids (crushed and sieved to  $\leq 1/4$  inch) extracted by distilled water (1:4 solid:liquid mass ratio) in a sealed 2-l polycarbonate bottle, tumbled at room temperature, and filtered to remove solids. Solid: liquid ratio of 1:4 was adapted based on an EPA standard protocol for preparation of batch leachate (US EPA, 1989a).

The short-term batch leaching test lasts for 24 h, while the long-term leaching test continues until the solution concentrations (as measured chemically) reach a plateau (up to several days). Samples may be taken during the batch leaching test in order to quantify leaching kinetics. Chemical and toxicological analysis of the leachate during the batch leaching process provides the chemical composition and toxicity data needed for initial evaluation of the test material (Nelson et al., 2000).

### 2.2. Flat plate leaching test

The flat plate leaching test is based on the reference environments of impermeable highway surface (Fig. 1a). The flat plates are prepared by compacting the CR-AC material into standard 4-inch diameter  $\times$  4-inch cylinders (Marshall method of specimen preparation, AASHTO, 1995). The test cylinder, sealed such that the top flat surface is exposed, is immersed in a 2 l glass beaker containing 1 l distilled water and mixed above the flat

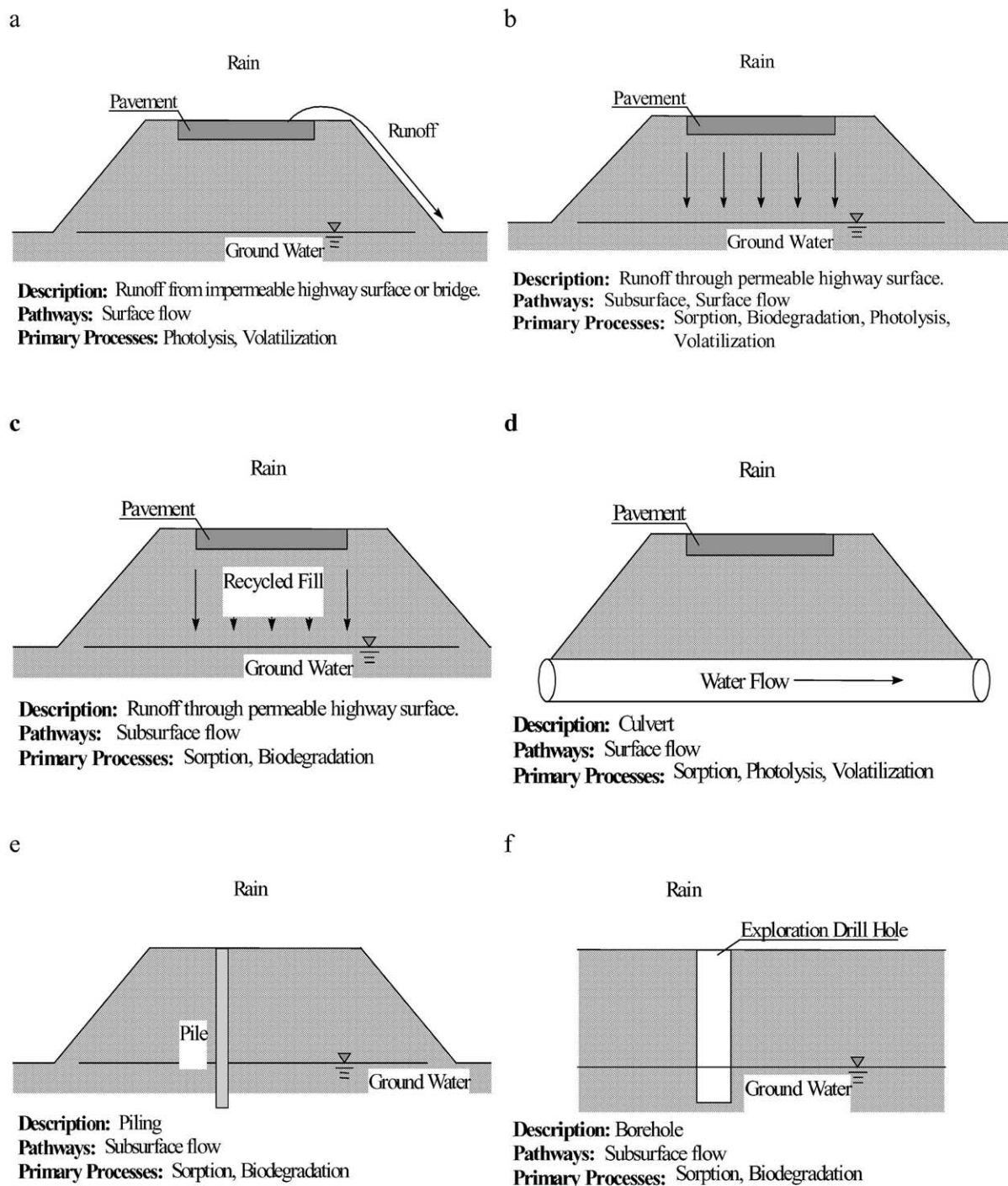


Fig. 1. Leaching from both the permeable and impermeable highway reference environments.

plate with a paddle stirrer. The test continues for 10 days with samples taken at 1,3,5,7, and 10 days to determine leaching kinetics and flux rates for compounds of interest.

### 2.3. Soil sorption test

The soil sorption test is based on the reference environments of permeable highway surface (Fig. 1b, c, and

d). The 24-h batch sorption tests are conducted in a reactor with soil-plus-leachate suspensions using the two test soils. The ratio of leachate to soil concentration is varied to evaluate the capability of environmental soils to adsorb available contaminants at equilibrium. The soil particles must be fully dispersed within the aqueous phase to achieve complete adsorption (typical soil/leachate ratios of 50, 100, 150, 200, and 250 g/l) (Steketee et al., 1997).



#### 2.4. Photolysis test

Photolysis is induced by exposing a test material leachate to “solar radiation” from a Xenon-arc lamp. Through photolysis, light can affect the chemical composition and toxicity of organic compounds. The photolysis test is based on the reference environments of impermeable highway surface (Fig. 1a). This study exposes the leachate to a Xenon arc lamp to mimic solar radiation at about 30 Watts/m<sup>2</sup> in a 20 °C constant temperature room. This light intensity is about one tenth the intensity of ambient sunlight. For the photolysis test, 24-h batch leachate aliquots were placed in 2-l Pyrex brand borosilicate glass bottles with stoppers and exposed to the Xenon arc lamp light. An initial sub-sample of the leachate is removed (time = 0) followed by additional sub-sampling with time to determine the photolysis degradation rate.

#### 2.5. Volatilization test

The volatilization test is based on the reference environments of both permeable and impermeable highway surface (Fig. 1a). The volatilization tests are conducted with 24-h batch leachate placed into 1-l glass beakers. The beakers were placed in a 20 °C controlled temperature room and the test solutions were continuously sparged with air at a flow rate of 250 ml/min. Samples were taken daily over the 7-day test period and analyzed for chemical content and toxicity.

#### 2.6. Biodegradation test

The biodegradation test is based on the reference environments of permeable highway surface (Fig. 1b–f). Biodegradation testing is conducted in the laboratory using 24-h batch leachate seeded with organisms from a mixed culture known to degrade a wide variety of aliphatic and aromatic hydrocarbons. The solutions are spiked with adequate nutrients to promote microbial growth and incubated at 20 °C and continuously aerated to supply oxygen. Samples are taken at intervals beginning at 2 h to test algal chronic toxicity to determine whether toxicity had been removed.

#### 2.7. Toxicity tests

Toxicity testing involves the use of standard test organisms in laboratory exposures to samples or solutions leached from highway CRMs. Ninety-six-h *Selenastrum capricornutum* algal chronic toxicity tests (US EPA, 1991a) and 48-h *Daphnia magna* (water flea) acute mortality tests (US EPA, 1991b) were performed on leachates. A definitive test is conducted to determine the concentration (percent of full-strength leachate) producing an EC<sub>50</sub> or LC<sub>50</sub>. The EC<sub>50</sub> is the leachate concentration that

inhibits algal growth to 50% relative to a control. LC<sub>50</sub> is the leachate concentration that causes 50% mortality of *Daphnia* relative to a control. Lower EC<sub>50</sub> and LC<sub>50</sub> values indicate greater toxicity, because the material is toxic even at low concentrations (high dilutions). A series of sub-samples is prepared from the original sample to be tested. With whole-effluent toxicity tests the original water sample is diluted in different ratios to produce the range of doses to which the organisms are exposed.

### 3. Test materials

Asphalt-based materials were either prepared in the laboratory or were obtained from field projects. CR-AC was obtained pre-mixed directly from Massachusetts Department of Transportation (MS DOT) (Nelson et al., 2001). In preparation of the asphalt-based materials for laboratory testing, Oregon Department of Transportation (ODOT), 1993 mix design was used to estimate the expected optimum binder content (5.4% of total mix by mass). Locally available aggregates were used in asphalt assemblages. Aggregate and waste material gradations were determined using ASTM procedure C136 (ASTM, 1994). After mixing, the asphalt assemblages were aged following the procedure established by the Strategic Highway Research Program (AASHTO, 1994).

Two representative test soils were selected to determine the effect of soil adsorptive capacity on the reduction of toxicity in leachates prepared from CR-AC. The selected soils were Woodburn soil (*Mollisol*), typically described as montmorillonitic with moderate organic content and distributed throughout the Ohio and Upper Mississippi Valleys, and Sagehill soil (*Aridisol*), typical of alkaline coarse-grained soil with free CaCO<sub>3</sub> of arid climate conditions and found in the southwest deserts. The soils were prepared from air-dried samples, sieved through a 1/4 inch screen.

### 4. Analytical methods

Metal analyses were performed with an inductively coupled plasma atomic-emission spectrometry (ICP-AES, Varian Liberty 150). Extraction of organic chemicals from the leachate is performed according to EPA methods 1624 and 1625 (Gas Chromatography-Mass Spectrometry Methods for Analysis of the Volatile and Semi-volatile Organic Priority Pollutants) (US EPA, 1989b, c). Briefly, a measured volume of the CR-AC leachate (1 l), was serially extracted in a separatory funnel three times with methylene chloride. The extracts were concentrated to a volume of 1 ml by Kuderna-Danish evaporation. A Hewlett-Packard (HP6890 plus) gas chromatograph with a flame ionization detector

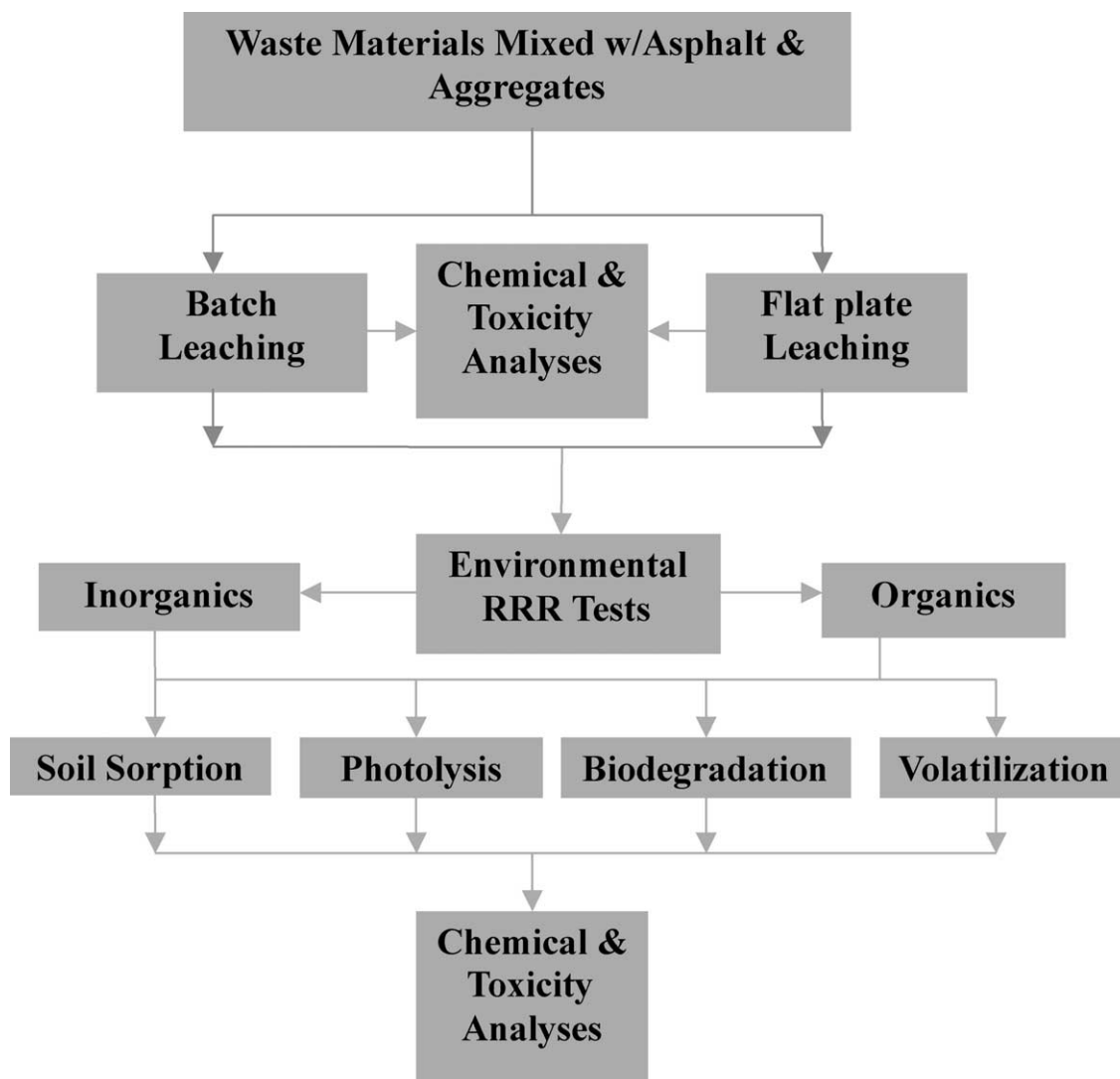


Fig. 2. Evaluation methodology.

(FID) and mass spectrometer (MS 5973 detector) and a Reztec fused silica capillary column 30 m in length, 0.32 mm i.d., were used for target analytes identification and quantification.

## 5. Results and discussion

### 5.1. Batch leaching tests

#### 5.1.1. Chemical testing

Batch leaching tests indicated that the maximum leaching rate occurred within 24 h as exemplified by the data for Al and Hg (Fig. 3). The leaching occurred rapidly, about 50% of the final concentration was reached within the first 10 h. The leaching rates during the first 10 h probably well represent the case of first-flush chemical releases from new construction materials; typical leaching rates of weathered materials probably

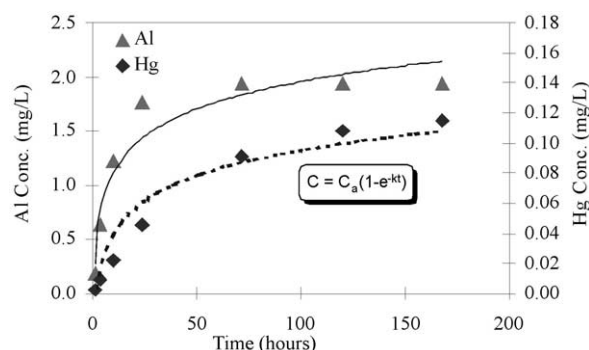


Fig. 3. CR-AC batch leaching experimental data and model fit for aluminum and mercury.

are best described by the slower loss rates observed after several days of leaching. A suite of 20 metals in CR-AC 24-h leachate was determined by ICP-AES. Of these, seven elements (Al, Ca, K, Mg, Sr, P, and Hg) were determined to be above instrumental (ICP-AES) detection

limits and 13 (V, Zn, As, Ba, Ni, Co, Fe, Cr, Cu, Sb, Pb, Cd, and Se) were below detection limits. Of particular significance are the higher than expected concentrations of mercury (Hg) and aluminum (Al).

To further investigate the sources of Hg and Al, 24-h leachates were generated for the following samples: asphalt mixed with aggregate, Mississippi #30 asphalt, Mississippi #20 asphalt, and three aggregate size fractions (1/2-in minus, 3/4-in minus, and 4-in minus). Asphalt samples were prepared as coatings on sand prior to leaching. Mercury and aluminum results on these leachates revealed that the likely source of Hg was Mississippi #30 asphalt and of Al was aggregate in the CR–AC leachate (Table 1). A literature search was conducted to determine background concentrations of Hg and other elements in crude oils. Concentrations of the elements S, V, Cr, Fe, Co, Ni, Zn, Se, Br, I, Ag, and Hg were reported in 45 crude oil samples of 17 oil fields originating from nine different Mesozoic or tertiary reservoir rock formations (Filby, 1987; Hitchon et al., 1987; Smith et al., 1987; Hirner et al., 1985). The Hg concentration ranges are from 0.02 to 0.33 ppm and 0.11 to 1.43 ppm in crude oils and in the asphalt fractions, respectively.

Organic compounds and their chemical formulae in the 24 h CR–AC leachate were determined from GC/MS analysis and are listed in Table 2. Benzothiazole ( $C_7H_5NS$ ) and 2(3H)-benzothiazolone ( $C_7H_5NOS$ ) were the main peaks of possible significance in the GC–MS spectrum and were separately quantified. Concentrations of benzothiazole and 2(3H)- benzothiazolone were determined to be 0.447 and 0.012 mg/l, respectively. Batch leaching tests indicated that the maximum benzothiazole leaching rate occurred within 10 h (Fig. 4), and it follows a first order rate expression of the form  $C = C_a(1 - e^{-kt})$ . Because of the relatively high benzothiazole concentration in the CR–AC leachate, it is considered to be a potentially toxic compound and was determined throughout the soil sorption, volatilization, biodegradation, and photolysis experiments.

### 5.1.2. Toxicity testing

An overall increase in toxicity of the leachates with leaching time was observed from both *S. capricornutum*

Table 1

Aluminum and mercury concentrations in 24-h leachates of CR–AC and subfractions

Sample ID	Al mg/l	Hg mg/l
Crumb rubber-modified asphalt	1.870	0.023
Asphalt mixed with aggregate	1.540	0.017
Asphalt # 30 Miss.	0.014	0.027
Asphalt # 20 Miss.	0.017	BDL
Aggregate 1/2 inch	1.653	BDL
Aggregate 3/4 inch	1.751	BDL
Aggregate manuf. sand	1.030	BDL

BDL, concentration below detection limit.

Table 2

Organic compounds identified in 24-h CR–AC leachate using GC–MS

Organic compound	Chemical formula
Pyrido [1,2-alazepine-6,6,8,9-tetracarboxylic acid, 10-(benzyloxy)-6,7-dihydro-, tetramethyl ester	$C_{25}H_{23}NO_{10}$
Phthalic anhydride	$C_8H_4O_3$
Benzothiazole	$C_7H_5NS$
Decanoic acid, silver (1+) salt	$C_{10}H_{20}O_2$
Benaldehyde, 3-hydroxy-4-methoxy	$C_8H_8O_3$
Glycine, N-methyl- N-(1-oxododecyl)	$C_{15}H_{29}NO_3$
Quinoline, 2,3-dimethyl	$C_{11}H_{11}N$
[1,1-Biphenyl]-2-ol	$C_{12}H_{10}O$
2(3H)-Benzothiazolone	$C_7H_5NOS$
Hexadecanoic acid	$C_{16}H_{32}O_2$

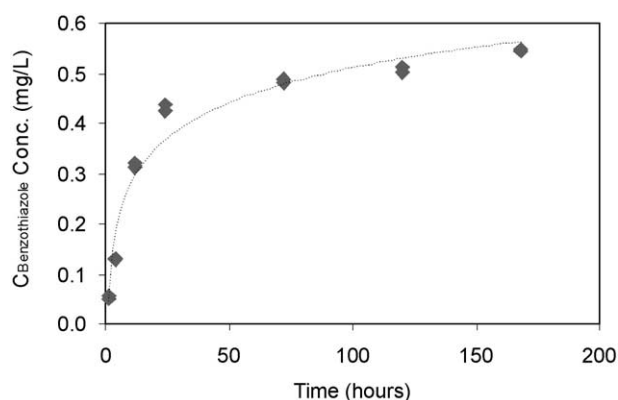


Fig. 4. CR–AC batch leaching experimental data and model fit for benzothiazole.

and *D. magna* test results. CR–AC batch leachates exhibited sharp (about three-fold) increase in algal toxicity between 1- and 24-h leachates. A mean  $EC_{50}$  value of 58% indicating moderate toxicity in 1-h leachate and a mean  $EC_{50}$  value of 18% indicating high toxicity in 24-h leachate was observed from algal test. However, only a small increase in algal toxicity was observed between 24-h and 168-h leachates with the 168-h leachate exhibiting an  $EC_{50}$  value of 15%. For *D. magna*, CR–AC batch leachates exhibited a similar pattern in their toxicity response between 1-h leachate and 24-h leachate. While non-toxic response was observed in 1-h leachate the toxicity of the leachate for *Daphnia* increased exhibiting a mean  $LC_{50}$  value of 44% indicating a moderate toxicity. Again, only small increase in *Daphnia* toxicity was observed between the 24-h leachate and 168-h leachate with the 168-h leachate indicating a mean  $LC_{50}$  value of 42%. It is pertinent to note that for 24-h batch leachates *S. capricornutum* exhibited a mean  $EC_{50}$  value of 18% and *D. magna* exhibited a mean  $LC_{50}$  value of 44%. This marked difference in their toxicity values is due to their published difference in sensitivity towards metals and organics (Biesinger et al., 1986).

Gradual increase in toxicity with time for *S. capricornutum* test organisms was associated with progressive release of aluminum, mercury, and benzothiazole in the leachates. Comparison with chemical analysis indicated good correspondence between toxicity and concentrations of aluminum, mercury, and benzothiazole present in the CR–AC leachates. Aluminum, mercury, and benzothiazole were present at concentrations that were higher than or closer to their  $EC_{50}$  values (0.6 mg/l Al; 0.059 mg/l Hg; and 0.65 mg/l benzothiazole) for *S. capricornutum* and  $LC_{50}$  values (0.005 mg/l Hg) for and *D. magna*. These toxic actions could be simply additive or even synergistic depending on the interaction of toxicants involved. Thus, it is possible that combined action of these chemicals might well have contributed to the toxicity for *S. capricornutum* and *D. magna*.

### 5.2. Flat plate leaching tests

Results for Al and Ca concentrations in leachate from the CR–AC specimens in the flat plate experiments are shown in Fig. 5. A power function was used to represent the flat plate leachate data. Based on linear regression of the flat plate data, aluminum concentrations in flat plate leachates are given as:  $C_{Al} = 5 \times 10^{-4} (t)^{1.11}$ , where concentration ( $C$ ) is in mg/l and time ( $t$ ) is in hours. Knowing the average surface area of the CR–AC flat plate and the volume of the leaching solution, the flux,  $F$ , of aluminum from CR–AC was calculated as  $F = (V/A) dC/dt$ ; where  $V$  is the leachate volume ( $L$ ) and  $A$  is the surface area ( $cm^2$ ). For a volume of 1 l, surface area of  $78.5 cm^2$ , and release rate,  $dC/dt = 5.55 \times 10^{-4} mg/l-h$  (using eq.  $C_{Al} = 5 \times 10^{-4} (t)^{1.11}$ ), the resulting flux for flat plates leachate is:  $F = (1/78.5 cm^2) \times 5.55 \times 10^{-4} mg/l-h = 7.07 \times 10^{-6} mg/cm^2-h$  for aluminum.

### 5.3. CR–AC leachate soil sorption

Benzothiazole is used as a toxic compound to measure CR–AC leachate sorption for the potentially toxic organic compounds. Use of surrogate chemicals is clearly to be desired, since the chemical can be mixed

and toxicity determined from its relationship to the mixed surrogate concentration. In most cases, total organic compound (TOC) could be used as a surrogate for the toxic organic compounds (Nelson et al., 2000). Detoxification of CR–AC–asphalt leachate by Woodburn and Sagehill soils was evaluated for the leachates after soil sorption using the two toxicity tests. Complete removal of toxicity for both *S. capricornutum* and *D. magna* was observed at 50 g/l soil:solution ratio of both soils (Table 3). Significant removal of benzothiazole (approximately 78% reduction in concentration when compared with control leachate) and complete removal of mercury accompanied this detoxification of CR–AC leachate at 50 g/l of Woodburn soil. Approximately 58% decrease in benzothiazole and 100% removal of mercury at 50 g/l Sagehill soil was observed. At higher soil concentrations ( $> 50 g/l$ ) no toxic effect was observed in the leachates for both test organisms, as expected. A concomitant pattern of removal of metals and organics was associated with toxicity removal in the soil sorption tests.

### 5.4. Photolysis of CR–AC leachate

The results of the photolysis degradation of benzothiazole in the CR–AC leachate showed no changes in benzothiazole concentrations over the 7 days of photolysis test. This is consistent with the fact that both benzothiazole and 2(3H)-benzothiazolone adsorb sunlight very weakly at wavelengths above 290 nm (Lang, 1961; Fife et al., 1975). Reddy and Quinn, 1997 concluded that benzothiazole and 2(3H)-benzothiazolone leachate from CR–AC materials are not directly photolyzed. Detoxification was investigated by testing the leachate toxicity before and after photolysis. Toxicity of the leachate for *S. capricornutum* retained its high toxicity level during that seven-day photolysis period. Similarly, *D. magna* values remained approximately the same after seven days of photolysis, when compared with the control leachate.

### 5.5. Volatilization of CR–AC leachate

Volatilization caused significant removal of benzothiazole from CR–AC leachate as shown in Fig. 6. The flux across the water-air interface is a first-order process, commonly assumed for environmental conditions, such that:  $C = C_0 e^{-k_v t}$ , where:  $C_0$  = initial concentration of constituent (mg/l),  $C$  = concentration of constituent at time  $t$  (mg/l), and  $k_v$  = first-order volatilization rate constant (1/h). The first-order volatilization coefficient,  $k_v$ , determined by linear regression (Fig. 6), equals 0.04/h or 0.96/day. It was concluded that benzothiazole is volatile with a half-life of about 1 day. Volatilization may contribute to significant environmental loss of these organic compounds if exposure times in surface environments are sufficient. Toxicity results indicated that a 76% reduction in benzothiazole was accompanied by complete removal of

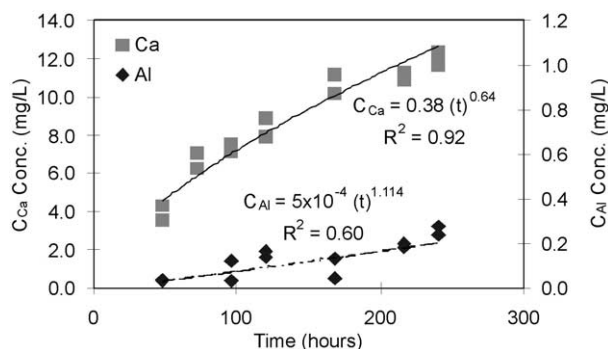


Fig. 5. CR–AC flat plate experimental data and model fit for aluminum and calcium.



Table 3

*S. capricornutum* %EC<sub>50</sub> and *D. magna* %LC<sub>50</sub> values for CR–AC leachates before and after sorption to 50 g/l soils. Each value is mean from duplicate tests

Test	<i>S. capricornutum</i> %EC <sub>50</sub>	<i>D. magna</i> %LC <sub>50</sub>	Mercury (mg/l)	Benzothiazole (mg/l)
Crumb rubber–asphalt control leachate	18%	42%	0.018	0.45
Sagehill soil (50g/l)	NTE	NTE	BDL	0.26
Woodburn soil (50g/l)	NTE	NTE	BDL	0.13

NTE, no toxic effect. BDL, concentration below detection limit.

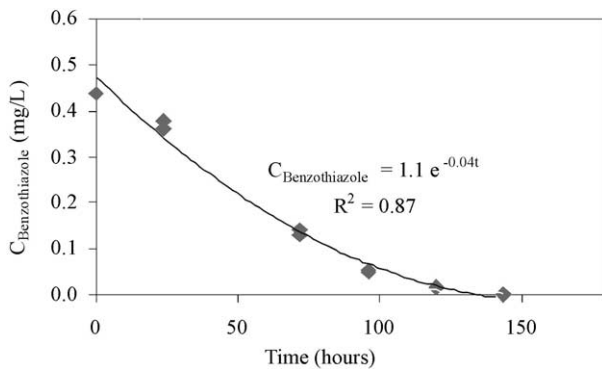


Fig. 6. CR–AC volatilization experimental data and model fit for benzothiazole.

toxicity for *S. capricornutum* after 3 days. Thus, the effectiveness of volatilization in reducing toxicity might in part derive from removal of benzothiazole and related compounds in the leachate.

### 5.6. Biodegradation of CR–AC leachate

Biodegradation testing of CR–AC leachate was conducted in the laboratory using 24-h batch leachate solutions and following the method described in the

experimental approach section. Samples were taken at intervals beginning at 2 h to test algal chronic toxicity to determine whether toxicity had been removed. *S. capricornutum* toxicity data indicated no toxic effect after 12 h of biodegradation reaction time. It was found that by adjusting the leachate pH to 7 with the nutrient buffer solution required for the biodegradation test, aluminum was precipitated and removed from the solution by filtration. Mercury also disappeared from the solution due to adsorption on the aluminum precipitate. Benzothiazole concentration was almost the same after 12 h biodegradation reaction time. The toxicity of benzothiazole to *S. capricornutum*, was determined for a standard solution of benzothiazole. Toxicity test results indicated that benzothiazole was highly toxic to *S. capricornutum* (EC<sub>50</sub> = 0.68 mg/l). Although there was about 0.45 mg/l benzothiazole in the 24-h CR–AC leachate, no toxic effect was observed, as aluminum and mercury had been removed by precipitation in the buffer solution. Thus, toxicity observed in the CR–AC leachate might well be due to the joint effects of mercury, aluminum and benzothiazole. Removal of one or more of these constituents resulted in complete or partial removal of toxicity in the CR–AC leachates during biodegradation testing.

Table 4

Summary of batch leaching and RRR processes results for CR–AC

Test	Conditions	Potential toxicant	Toxicant conc. (mg/l)	Algal %EC <sub>50</sub>	<i>D. magna</i> %LC <sub>50</sub>
Short-term batch leaching	24 h	Benzothiazole	0.45	17	44
		Aluminum	1.5		
		Mercury	0.02		
Long-term batch leaching	7 days	Benzothiazole	0.54	25	42
		Aluminum	1.81		
		Mercury	0.116		
Flat-plate leaching	Init.–day 10	Aluminum	0.04–0.24	NTE	NTE
		Benzothiazole	0.42–0.040		
		Mercury	0.02–0.016		
Volatilization	Init.–day 7	Benzothiazole	0.44–0.437	49–NTE	38–NTE
		Aluminum	1.67–1.65		
		Mercury	0.019–0.018		
Photolysis	Init.–day 7	Benzothiazole	0.44–0.019	9–5	31–34
		Aluminum	1.62–0.04		
		Mercury	BDL		
Biodegradation	Init.–day 10	Benzothiazole	0.44–0.019	21–NTE	NTE
		Aluminum	1.62–0.04		
		Mercury	BDL		

NTE, no toxic effect. BDL, concentration below detection limit.

### 5.7. Summary of results

Results for batch leaching tests, RRR process tests, and toxicity tests for CR–AC are summarized in Table 4.

## 6. Conclusions

Batch leaching tests indicated that about 50% of the final leachate concentration was reached within the first 10 h. Leaching rates during the first 10 h probably well represent the case of first-flush chemical releases from new construction materials; typical leaching rates of weathered materials probably are best described by the slower loss rates observed after several days of leaching. Leaching from the flat plate test showed significant mass transfer limitations as compared to the batch leaching tests, with no toxic effect being measured in the flat plate leachate. In batch leaching, the benzothiazole concentration was about 0.54 mg/l after seven days. Benzothiazole is a highly toxic compound with an EC<sub>50</sub> of 0.68 mg/l. Aluminum and mercury also were detected in the leachate at concentrations of 1.5 and 0.02 mg/l, respectively. Reduction in the concentrations of benzothiazole, aluminum, and mercury after sorbing to Woodburn and Sagehill soils rendered the leachate completely nontoxic. Volatilization and biodegradation reduced benzothiazole concentration by about 90% to 0.04 mg/l with a subsequent removal of toxic effect. Photolysis did not affect the benzothiazole concentration and toxicity was not changed. This suggests that benzothiazole is a volatile and biodegradable compound, but resists the photolysis oxidation process. Contaminants from CR–AC leachates are thus degraded or retarded and should not be transported into nearby soils and groundwaters.

The methodology is limited to environmental impacts on the aquatic organisms tested in this study, although it could be extended to any desired target organism with additional laboratory work. The methodology only applies to the near-highway environment, i.e. on the order of a few meters from the roadway. Environmental impacts at further distances must be evaluated using other modeling and analytical techniques. The methodology is limited to construction and repair materials and does not account for operational aspects of highways, such as automobile-related pollutants or de-icing activities. However, new materials may be similarly evaluated following the protocols developed during this study (Nelson et al., 2000).

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