



FINAL REPORT

EVALUATION OF WASTE CONCRETE ROAD MATERIALS FOR USE IN OYSTER AQUACULTURE-PHASE III

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Mark Bundy Ph.D., PEARL, Morgan State University
Anastasia E. M. Chirside, Ph. D., University of Delaware,
Dong Hee, Kang, Ph.D., Research Associate, Morgan State University

Prepared by:
Organization: Morgan State University
1700 E. Cold Spring Lane,
Baltimore, MD 21251-0001

Prepared for:
Virginia Center for Transportation Innovation and Research
530 Edgemont Road
Charlottesville, VA 22903

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16. Abstract <p>This project is the third phase of a three-phase project. The primary objective of the research is to determine the suitability of recycled concrete from road projects as bottom conditioning material for on-bottom oyster aquaculture in the Chesapeake Bay. This phase of the project evaluated the RCA for petroleum byproducts and provided methodologies for SHA evaluation of materials. The results of this project showed that using RCA as a base material for oyster reefs did not contain any hydrocarbon chemicals and did not detect any water extractable SVOC from recycled concrete from EPA 1316 extraction. Low anthracene, fluorine, phenanthrene and pyrene of samples collected from Machado Construction Co., Inc. and The Recycling Center were detected. However, the concentrations of State of Maryland Department of the Environment cleanup standard for soil and groundwater are much higher than detected concentrations. This is at least 100 times below the considered hazardous level of Protected Groundwater Standard. The result concluded that RCA gives no concern for hydrocarbon component releasing into the Chesapeake Bay watershed, if RCA is using as a bottom conditioning material for oyster aquaculture.</p>			
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Research Professor Mark Bundy passed away on June 1st (2016) at the age of 69. Dr. Bundy was an Associate Research Professor at Morgan State University Patuxent Environmental & Aquatic Research Laboratory (PEARL) and has been the Director of Environmental Programs here at the PEARL for the past 8 years. Mark was involved in the strategic planning of all of the PEARL'S programs, including PEARL's commercial oyster support programs, and Power Plant Environmental Training Program. Every day friends and colleagues here can feel his impact on the Patuxent River. Mark brought to PEARL a much-needed view of the importance of environmental science and working with natural resource managers. Under Mark's influence the PEARL began to expand its program; below are some examples:

- Aquaculture – Mark initiated and developed the Oyster Aquaculture program at the PEARL. He provided guidance to the program managers, secured funding and facilitated relationships with management and industry.
- Transportation – Brought in private sector partners to develop a program focused on recycled highway materials for use in aquaculture.

Mark came to the PEARL from the Maryland Department of Natural Resources, where he served as the Assistant Secretary for Chesapeake Bay Programs. He was responsible for the overall administration of the Department's activities related to the State's policies on the restoration of the Chesapeake Bay. His career was spent protecting Maryland's environment through policy and research. Professionally and personally, Mark was a lover of the outdoors. He had a passion for sport fishing and photography. Mark lived his life unapologetically and we loved him for it. He will be truly missed.

Disclaimer

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1. INTRODUCTION

As part of their Environmental Stewardship Program, SHA intends to increase the use of recycled materials and use their products in an environmentally responsible manner. One way of meeting these goals is to recycle materials in an environmentally responsible project. As roads and bridges are resurfaced, old concrete is removed and usually discarded, which places a burden on society to absorb the waste concrete in landfills or other disposal sites. Instead of discarding this material, it would be in the best interest of the environment to recycle it into an alternative use, such as to condition portions of the Chesapeake Bay bottom to support spat-on-shell oyster restoration or aquaculture projects (SHA, 2013, and 2015).

Native oyster populations in the Chesapeake Bay are at less than 1% of historic levels due to two protozoan diseases (MSX; Multinucleated Sphere Unknown, a disease caused by *Haplosporidium nelsoni*, and Dermo disease caused by *Perkinsus marinus*), overharvesting, and pollution (CRC, 1999). This tremendous decline in the oyster population has dramatically changed the bay's ecosystem and the oyster industry. Individual oysters filter 4-34 liters of water per hour, removing phytoplankton, sediments, pollutants, and microorganisms from the water column (CERP, 2007). Historic oyster populations of Chesapeake Bay could filter excess nutrients from the estuary's entire water volume every three to four days. Today, that would take nearly a year.

Spat-on-shell is the most ecologically friendly and traditional method of culturing oysters in the Chesapeake region. To make new areas ready for on bottom spat-on-shell aquaculture, the barren Bay bottom needs to be built-up with a hard material that will support the spat-on-shell (a process known as bottom conditioning) and prevent it from sinking into soft muddy bottoms. Historically, old oyster shell was used for this purpose. However, the decline of the Chesapeake Bay region's oyster industry has led to the scarcity of available oyster shells and using them for bottom building is no longer practical. A lack of availability of oyster shell in the Bay is necessitating the investigation of alternative materials.

This is a three-phased study to examine the use of recycled concrete aggregate (RCA) from road construction to spat-on-shell. RCA is crushed concrete material created from the removal and milling of old concrete pavement / road infrastructure. The material is processed and sorted for reuse as base, sub-base, fill material for embankments, and in new concrete mix. For RCA to be used within the aquatic setting of the Chesapeake Bay, state agencies must be assured that it has no direct negative environmental impacts, that it is practical to use in this application and it meets regulatory criteria. In the Phase I study, the chemical behavior of RCA under saturated conditions was evaluated. That resulted in either no leaching of adverse materials or leaching at a rate that is orders of magnitude below regulatory levels. Further, the introduction of RCA did not raise pH above the minimum threshold for introduction in Maryland waters. In Phase I and II, the experiments performed in the laboratory and the field showed that there was no difference in RCA and oyster shell on oyster recruitment, survival, or growth, nor was there an effect on the associated community of organisms (SHA, 2013 and 2015)

The Federal Highway Administration (FHWA) and the U.S. Environmental Protection Agency (EPA) encourage beneficial use of recycled materials, including pavement materials. However,

some of these recycled materials may contain toxic substances, such as heavy metals and potentially hydrocarbon chemicals, which could leach when inundated with water, impact neighboring aquifers or streams, and impair ecological health and function. Therefore, the recycled concretes' leaching characteristics must be assessed before they are used (Kang et al., 2011). Understanding this behavior will ultimately help determine the suitability of RCA for supporting oyster aquaculture.

1.1 Summary of Previous Work

Evaluation of Waste Concrete Road Materials for Use in Oyster Aquaculture (MD- 13-SP109B4E) (SHA, 2013)

The primary objective of the Phase I study was to determine the suitability of recycled concrete from road projects as conditioning material for on-bottom oyster aquaculture in the Chesapeake Bay. The testing was designed to evaluate the impact on water chemistry from the introduction of RCA and evaluate the effect of RCA on the survivorship and growth of oyster spat. The results of this project showed that using RCA as a base material for oyster reefs did not adversely affect oyster spat growth and survival, or the surrounding environment. None of the metals leached at a rate that exceeded the EPA drinking water standards. This standard is more stringent than the current EPA total maximum daily loads (TMDLs) for Chesapeake Bay waters. There was no statistical difference between shell and RCA on the growth, survivorship, average length, or recruitment of young oysters. Initial pH was slightly higher for the RCA (8.20 to 8.36) than the oyster shell control (8.0 to 8.2), but pH stabilized to around 7.6 to 7.8 for all treatments after seven days. Based on the findings of this study, the recommendation is to initiate a second phase that places RCA on test plots in the Chesapeake Bay to validate the laboratory tests in situ.

Evaluation Of Waste Concrete Road Materials For Use In Oyster Aquaculture – Field Test (MD-15-SHA-MSU-3-12) (SHA, 2015)

The primary objective of this Phase II study was to determine the suitability of RCA from road projects as bottom conditioning material for on-bottom oyster aquaculture in the Chesapeake Bay. The testing was designed to evaluate the potential introduction of organisms attracted to the RCA pile in situ that may be potential predators of oyster spat, determine potential impacts on or disruptions to the use of traditional harvesting gear on aquaculture areas conditioned with RCA, and identify regulatory or administrative structures that oversee the use of RCA and challenges within those structures. Three substrates were tested for their effect on benthic communities: RCA, RCA with a veneer of oyster shells and oyster shells. There was no difference in population or community parameters among the three substrates. The number and type of species were the same among the substrates as were their absolute and relative abundances. Oyster spat settlement was the same among the three substrates as well. Waterman tonguing on RCA found it heavier and more difficult to work than tonguing on oyster shells. They recommended that RCA be used either with a veneer of oyster shells or in applications where tonguing was not anticipated. Overall the findings support the use of RCA in select applications. However, the regulatory structures presently in place do not include a mechanism for the acceptance of a novel material. Moving forward with RCA or any new material requires an application for a reef project. The acceptance of the project is then a de facto acceptance of the material. Adopting a criteria for materials used in reef construction will provide agencies with a basis for supporting choices on

materials used and the private sector with a basis to develop products to meet restoration and aquaculture needs.

1.2 Reclaimed Concrete Aggregate (RCA)

Disposal in landfills has historically been the most common method of managing concrete materials. However, recycling has become a more attractive option as an aggregate substitute in pavement construction. In the case of Mn/DOT, the concrete removed is permitted for use as recycled concrete aggregate and used almost 100% for road construction. Recycled concrete will be reducing landfill disposal material, fuel consumption and preserving natural aggregate resources. RCA consists of sand and various sizes and shapes of gravel, and there is a growing interest in substituting alternative aggregate materials due to the increasing amount of aggregate consumption that will be reached, which is expected to be 2.5 billion tons by 2020 in USA (FHWA, 2004). Portland cement concrete can be reclaimed during demolition of roads, pavements, airfield, runways, buildings, and other sources as a coarse granular material that can be used in various applications for aggregate. The RCA may include 10 to 30 percent subbase soil and asphalt pavement during excavation. Therefore, the RCA is a mixture of concrete, soil, and small quantities of bituminous concrete (FHWA, 2008).

RCA has rougher surface texture, lower specific gravity, higher water absorption, and lower specific gravity than natural aggregates. As a particle size decreases, specific gravity decrease and increase in absorption, due to the higher mortar proportion. Specific gravity of RCA reported from 2.0 to 2.5 (ACPA, 1993). RCA is generally more permeable than natural aggregate. Coarse RCA has favorable mechanical properties for aggregate use, including good abrasion resistance, good soundness characteristics, and bearing strength. Los Angeles Abrasion loss (%) is 25 to 40 which are somewhat higher than those of conventional aggregates. California Bearing Ratio (CBR) values are 94 to 148, and Percentage of magnesium sulfate soundness for a 6-year Long Island, New York, study for use as a granular subbase or base is 3.8 (ACPA, 1993; Hanks and Magni, 1989).

Leachability of contaminants and pH changes by RCA are significant environmental considerations about its use in road construction. The cementitious materials stem heavy metals by leaching. Leaching test of Portland cement concrete through Toxicity Characteristic Leaching Procedure (TCLP) using acetic acid showed that Arsenic, Beryllium, Cadmium, Chromium, Mercury, Lead, and Selenium are detected (Kanare and West, 1993). The long-term potential leachable trace toxic metals of Portland cement concrete showed that arsenic (19.9 mg/kg), beryllium (1.4 mg/kg), chromium (72.7 mg/kg), lead (75.3 mg/kg), Nickel (72.0 mg/kg) and vanadium (44.1 mg/kg) were detected (Sangha et al., 1998) and significant levels of calcium and aluminum were also detected (Nelson et al., 2000). In the case of Minnesota where deicing salts are extensively used, recycled concrete may contain relatively high levels of chlorides associated with corrosion of steel. Cement paste consists of calcium-aluminum-silicate compounds. The pH of RCA-water mixtures often exceeds 11 due to calcium hydroxide that could have an adverse effect on the environment.

1.3 Recycled concrete aggregate types

Recycled concrete aggregates are produced by breaking down concrete and crushing concrete into pieces of the desired size. The resulting recycled aggregate comes in the following 4 basic sizes.

1. RC-6 Recycled Concrete ($< 1 \frac{1}{2}$ ""): RC-6 can be used as a sub-base for parking lots and roadways, residential driveways, walkways and select back-fill.
2. RC-2 Recycled Concrete ($1 \frac{1}{2}$ " to $2 \frac{1}{2}$ ""): RC-2 is frequently used for construction entrances, drainage and erosion control, stabilization of wet areas, under-cutting, etc.
3. RC-Surge Recycled Concrete (3" to 8"): RC-Surge can be used for erosion control, bulkheads and slope protection.
4. RC-57 Recycled Concrete ($\frac{3}{4}$ " – $1 \frac{1}{2}$ ""): RC-57 is used primarily as a subgrade for concrete slabs, base material for driveways, or erosion prevention fill.



A) RC-6



B) RC-2



C) RC-Surge



D) RC-57

Figure 1 Recycled concrete type by sizes (<http://www.laneyrecycling.com>)

1.4 Type of petroleum product

Petroleum byproducts contain primarily hydrocarbons, and generally are classified into two major component categories: hydrocarbons and nonhydrocarbons. The hydrocarbon constituents can be grouped into saturated hydrocarbons (Aliphatic such as Butane and Isobutane, Alicyclic), unsaturated hydrocarbons (Alkenes/Olefins such as Ethylene 1-Butene and Alkynes /Acetylenes such as Acetylene 1-Butyne), and aromatics which are common environmental contaminants.

Aromatic compounds are a special class of unsaturated hydrocarbons. These compounds are based on the benzene ring, which contains six carbons. There may be two or more aromatic rings fused together resulting in polycyclic aromatic hydrocarbons (PAHs). Even though petroleum hydrocarbons that can be measured by the total petroleum hydrocarbon (TPH) method are not generally regulated as hazardous wastes, measured TPH values suggest the relative potential for environmental exposure and ecological health effects. Leaking underground storage tanks (LUST) are the most frequent causes of federal and state governmental involvement in petroleum hydrocarbon problems, especially groundwater contamination. TPH test results may require action on local or state governments to remove or reduce the TPH problem.

Table 1. Summaries for the indicator compounds and appropriate carbon ranges for the TPH determination depended on the source of the petroleum product (TPHCWG 1997a and 1998)

Indicator Compound/ TPH Fraction	Gasoline	Kerosene Jet Fuel Diesel	Diesel, light fuel oils	heavy fuel oils	crude oil	Highly Refined Base Oils	Used Motor Oil, Lubricatin g	Unknown
Aliphatics > C6 - C8	X				X			X
Aliphatics > C8 - C10	X	X	X		X			X
Aliphatics > C10- C12	X	X	X		X			X
Aliphatics > C12- C16		X	X		X	X		X
Aliphatics > C16- C28			X	X	X	X	X	X
Aromatics > C8 - C10	X	X	X		X			X
Aromatics > C10 - C12	X	X	X		X			X
Aromatics > C12 - C16		X	X		X	X		X
Aromatics > C16 - C121			X	X	X	X		X
Aromatics > C21 - C35				X	X	X	X	X

Petroleum products are complex mixtures of hundreds of hydrocarbon compounds. The exact composition of petroleum products varies depending upon the source of the crude oil and the refining practices which are separated into fractions having similar boiling points. Automotive gasoline is a mixture of low-boiling hydrocarbon compounds suitable for use in spark-ignited internal combustion engines (ATSDR 1995). Automotive gasoline typically contains about 150

hydrocarbon compounds, though nearly 1,000 have been identified (ATSDR 1995). Typical hydrocarbon chain lengths range from C₄ through C₁₂ with a general hydrocarbon distribution consisting of 4-8% alkanes, 2-5% alkenes, 25-40% isoalkanes, 3-7% cycloalkanes, 1-4% cycloalkenes, and 20-50% aromatics (IARC 1989a). However, these proportions vary greatly. Unleaded gasolines may have higher proportions of aromatic hydrocarbons than leaded gasolines. Other petroleum products are Stoddard solvent (C₇-C₁₂), which is a petroleum distillate widely used as a dry cleaning solvent and as a general cleaner and degreaser, jet fuels (C₄ - C₁₆) which are light petroleum distillates, fuel oil #1 (C₁₁-C₂₀) which is used in atomizing burners, #2 (C₁₁-C₁₉) which is used primarily for home heating and industries, and #6 which can be blended directly to heavy fuel oil or made into asphalt, and mineral oils which are often lubricating oils, but they also have medicinal and food uses. The indicator compounds and appropriate carbon ranges for the TPH determination are dependent upon the source of the petroleum product and are summarized in Table 1.

1.5 Physical parameters

A large range of behavior in environmental media is governed by their physicochemical properties. Volatilization, biodegradation, partitioning, oxidation, photo-degradation, etc. also change these mixtures. As a result of these characteristics, the assessment of petroleum hydrocarbon mixtures is difficult. The more soluble or volatile compounds will migrate to other locations. The mostly non-mobile components are left behind at the release site. Therefore, location of release, length of time between the release and exposure, media of exposure, etc. can also contribute to these difficulties. The physical parameters for hydrocarbon fractions are listed in Table 2.

Solubility is one of the key factors in determining the impact of a chemical in the environment. The greater the solubility, the higher the dissolve rate in water and migration away from the contaminated area. Solubility generally decreases with increasing molecular weight of the hydrocarbon compounds. For compounds having similar molecular weights, the aromatic hydrocarbons are more water soluble and mobile in water than the aliphatic hydrocarbons (ASTM 1985) and branched aliphatics are less water-soluble than straight-chained aliphatics (ATSDR, 1999).

Volatility is a tendency of a chemical to migrate as a vapor and primarily a function of the vapor pressure of the compound and estimated using Henry's law, since hydrocarbon fractions characterized by relatively high vapor pressures tend to volatilize and enter the vapor phase. In general, compounds having a vapor pressure in excess of 10⁻² mm Hg are more likely to be present in the air phase than in the liquid phase. Compounds characterized by vapor pressures less than 10⁻⁷ mm Hg are more likely to be associated with the liquid phase. Compounds possessing vapor pressures that are less than 10⁻² mm Hg, but greater than 10⁻⁷ mm Hg, will have a tendency to exist in both the air and the liquid phases (Knox, 1993).

Chemical mobility can be determined based on the organic carbon-water partition coefficient (K_{oc}). If the chemical is strongly associated with the substrate, the chemical is relatively immobile and will not be leached or transported great distances from the area of the release. A compound that is strongly sorbed to the organic carbon in the substrate is less available and less

likely to be volatilized or biodegraded. Koc values <50 L/kg, 50-150 L/kg, and 150-500 L/kg are considered to be very mobile, mobile, and intermediate in mobility, respectively (Dragun, 1988).

Table 2. Physical parameters for hydrocarbon fractions (TPHCWG, 1997b)

Composite of Fractions	Solubility (mg/L)	Vapor Pressure (mmHg)	Henry's Law Constant(cm ³ /cm ³)	log Koc	Chemical Type
Aliphatic					
C ₅ -C ₆	36	266	47	29	Volatile
C ₆ -C ₈	5.4	47.88	50	3.6	Volatile
C ₈ -C ₁₀	0.43	4.788	55	4.5	Volatile
C ₁₀ -C ₁₂	0.034	0.4788	60	5.4	Volatile
C ₁₂ -C ₁₆	0.000076	0.05776	69	6.7	Extractable
C ₁₆ -C ₃₅	0.0000025	0.000836	85	8.8	Extractable
Aromatic					
C ₅ -C ₇	220	83.6	1.5	3.0	Volatile
C ₇ -C ₈	130	26.6	0.86	3.1	Volatile
C ₈ -C ₁₀	65	4.788	0.39	3.2	Volatile
C ₁₀ -C ₁₂	25	0.4788	0.13	3.4	Extractable
C ₁₂ -C ₁₆	5.8	0.03648	0.028	3.7	Extractable
C ₁₆ -C ₂₁	0.65	0.000836	0.0025	4.2	Extractable
C ₂₁ -C ₃₅	0.0066	3.34E-07	0.000017	5.1	Extractable

1.6 Toxic substance criteria

Polycyclic Aromatic Hydrocarbons (PAHs) are nonpolar and lipophilic and composed of multiple aromatic rings. Most PAHs are not soluble in water and persist in the environment. Aqueous solubility of PAHs decreases approximately logarithmically as molecular mass increases (Choi et al., 2010). PAHs are formed during the incomplete burning of coal, oil and gas. The sources of PAHs have included vehicle exhaust, weathered material from asphalt roads, lubricating oils, gasoline, diesel fuel, and tire particles (Takada et al., 1990). There are more than 100 different PAH compounds. The EPA has characterized 16 PAHs as priority pollutants. The U.S. Department of Health and Human Services has determined that many PAHs may be considered carcinogenic (ATSDR, 1999). Despite the large number of hydrocarbons, only a relatively small number of the compounds are well characterized for toxicity. Health assessment of the risks from environmental media are difficult due to the lack of adequate knowledge about the movement of components in soil and lack of knowledge about the toxicity of the components (Heath et al. 1993). However, the health effects of some fractions can be well characterized, based on their representative compounds. In the case of Maryland, Code of Maryland Regulations, Title 26 which is Department of Environment, Subtitle 08: Water Pollution, Chapter

2: Water Quality(COMAR 26.08.02.03-2) provide the numerical criteria for toxic substance in surface water which is listed in Table 3.

Table 3 COMAR 26.08.02.03-2 Numerical Criteria of Toxic Substances in Surface Waters

Substance CAS*		Human Health for Consumption of: (Risk Level = 10 ⁻⁵) (µg/L)		
		Water + Organism	Organism Only	Chemical Group
Acenaphthene	83329	670	990	PAH
Anthracene	120,127	8,300	40,000	PAH
Benzo(a)Anthracene	56553	0.038	0.18	PAH
Benzo(a)Pyrene	50328	0.038	0.18	PAH
Benzo(b)Fluoranthene	205992	0.038	0.18	PAH
Benzo(k)Fluoranthene	207089	0.038	0.18	PAH
Bis(2-Ethylhexyl) Phthalate	117817	12	22	Phthalates
Butylbenzyl Phthalate	85687	1,500	1,900	Phthalates
Chrysene	218019	0.038	0.18	PAH
Dibenzo(a,h)Anthracene	53703	0.038	0.18	PAH
Diethyl Phthalate	84662	17,000	44,000	Phthalates
Dimethyl Phthalate	131113	270,000	1,100,000	Phthalates
Di-n-Butyl Phthalate	84742	2,000	4,500	Phthalates
Fluorene	86737	1,100	5,300	PAH
Hexachlorobenzene	118741	0.0028	0.0029	Organic Compounds
Hexachlorobutadiene	87683	4.4	180	Organic Compounds
isophorone	78591	350	9,600	Organic Compounds
Pentachlorophenol (PCP)**	87865	2.7	30	Pesticides and Chlorinated
Pyrene	129000	830	4,000	PAH

* Chemical Abstracts Service (CAS)

** Pentachlorophenol (PCP) has numerical criteria for aquatic life (ug/L); acute is 19 ug/L and chronic is 15 ug/L for fresh water, acute is 13ug/L and chronic is 7.9ug/L for salt water.

1.7 Research Objectives:

While it is generally recognized that RCA materials do not present a great risk to human health or the environment, a better characterization of the amount and type of chemicals that leach in the environment will help provide a better means to correctly manage this material. This project is the third phase of a three-phase project. The primary objective of the research is to determine

the suitability of recycled concrete from road projects as bottom conditioning material for on-bottom oyster aquaculture in the Chesapeake Bay. The leaching performance of recycled concrete materials potentially contaminated by toxic organic substances, especially petroleum residues and PAH, was investigated through a series of laboratory experiments that included batch and tank leaching experiment. This phase of the project will:

1. Evaluate the RCA for toxic organic substances, especially petroleum byproducts and PAHs.
2. Provide methodologies for SHA evaluation of materials

2. MATERIALS AND METHODS

2.1 Sample collection

The first task was to select different RCA sources throughout the state of Maryland. Three of the samples were collected from three concrete dumping sites for recycling concrete materials. Recycled concrete sampling site information is listed in Table 4. The samples were collected in two 250ml wide mouth glass jars with Teflon-lined screw caps from crushed RCA stockpiles at the plants

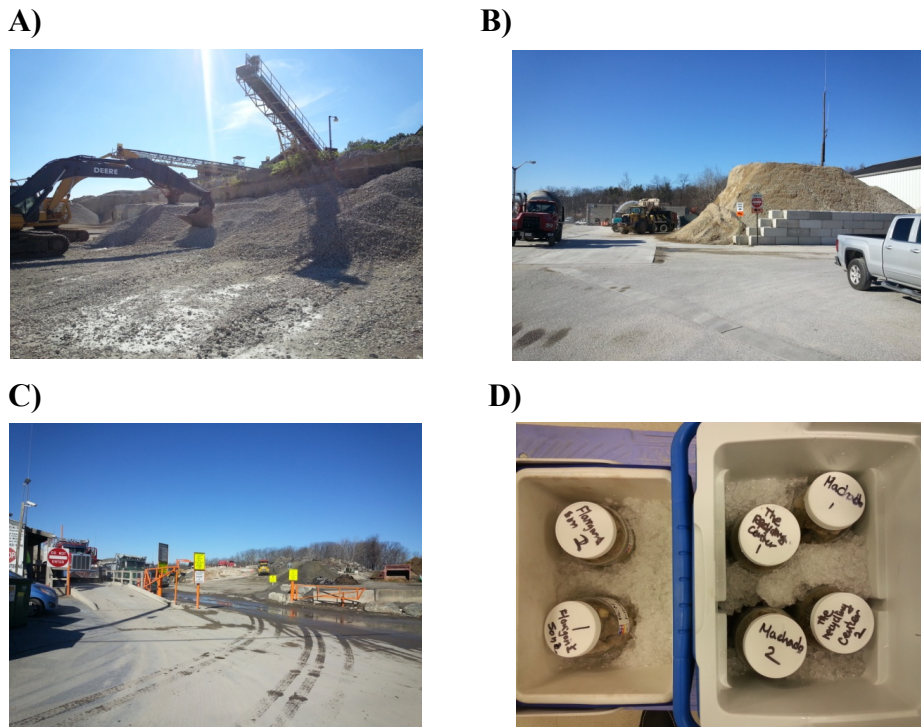


Figure 2 Sampling sites, A) Flanigan & Sons, Inc. B) Machado Construction Co., Inc. C) The Recycling Center and D) sample reserved in cooler after sampling

RC-Surge recycled concrete (3” to 8”) will be used for oyster conditioning materials; however, smaller material sizes were collected from each plant for extraction convenience. Bigger sizes required additional process for extraction such as crushing or grinding which may generate inaccurate results. Therefore RC-6 (< 1 1/2”) was collected from Machado Construction Co., Inc. and The Recycling Center, and RC-2 (1 1/2” to 2 1/2”) was collected from Flanigan & Sons, Inc., where RC-2 was the smallest size.

Table 4 Recycled Concrete Sampling Sites in Maryland

Company	Flanigan & Sons, Inc.	Machado Construction Co., Inc.	The Recycling Center
Address	2444 Loch Raven Road Baltimore, MD 21218	2930 Hammonds Ferry Road, Baltimore, MD 21227	14852 Old Gunpowder Road, Laurel, MD 20707
Phone	410-467-5900	410-247-2662	410-792-2999
Sample Type	RC-2	RC-6	RC-6

The collected samples were cooled to 4±2°C immediately after collection. Collected RCA samples were extracted within 48 hours of collection, and analyzed with one week of extraction. Two separate samples were collected at the same time and placed under identical circumstances and managed the same throughout field and laboratory procedures. Analyses of field duplicates give a measure of the precision associated with sample collection, preservation and storage, as well as laboratory procedures.

2.2 Moisture content measurement

In order to provide the dry mass equivalent of the "as-tested" material, the solids content of the subject material should be determined. Place a collected recycled concrete samples into a dish and then dry the sample to a constant mass at 105 ± 2 °C .



Figure 3 Moisture content measurement of RCA

To check for constant mass by returning the dish to the drying oven for 24 hours, the dishes were cooled to room temperature in a desiccators and then re-weighed. The moisture contents of collected recycled concrete samples from Flanigan & Sons, Inc., Machado Construction Co., Inc., and The Recycling Center are $5.63 \pm 1.56\%$, $6.59 \pm 0.65\%$, $8.51 \pm 1.35\%$, respectively.

2.3 Extraction method

Volatile organic compounds (VOCs) were not expected to be a major concern with regard to leaching from RCA. Because of their volatility, the compounds such as butane, propane, benzene, toluene, ethylbenzene and xylene in gasoline are preferentially volatilized (Bauman, 1988) and most of these compounds would tend to evaporate quickly when milled on the roadway or at structure demolition sites. RCA are mixed source and would be expected to be stockpiled for a while. Even given that, volatile organic compounds in the leachate from various extraction test (Toxicity Characteristic Leaching Procedure, (TCLP), Synthetic Precipitation Leaching Procedure (SPLP), Deionized Water Leaching Procedure, and Column Leaching Procedure) of recycled asphalt pavement (RAP) were not found above the detection limit. All results were below the detection limit (BDL) (Timothy, 1998). Therefore, VOCs were not considered to be analyzed in this experiment.

2.3.1 Microscale Solvent Extraction (MSE)-Method 3570

EPA test method 3570 method was applied to extract petroleum chemical to determine the mobility of petroleum analytes presented in the recycled concrete. Method 3570 is a procedure for extracting organic compounds from wastes, especially semivolatile organic compounds (SVOCs). Collected recycled concrete samples are prepared by shake extraction for at least 4 hours with methylene Chloride (CH_2Cl_2) in sealed extraction tubes. Sample extracts are collected, dried by sodium sulfate (Na_2SO_4), and concentrated using a modification of the Kuderna-Danish concentration method. All solid samples are kept cold during the extraction procedure by storing them in a cold room (4°C). All samples are transferred from the cold room only for as long as necessary to remove the sample aliquot. As much as possible, the sample container is kept tightly capped. The extract is transferred to a 2 mL vial fitted with a PTFE lined screw cap, and the vial is capped and stored in the freezer until analysis.

2.3.2 Liquid-Solid Partitioning (LSP) as a Function of Liquid-to-Solid Ratio Using a Parallel Batch Extraction Procedure-Method 1316

This method (Liquid-solid partitioning as a function of liquid to solid ratio in soil materials using a parallel batch procedure) is suitable for assessing the leaching potential of a wide range of solid materials such as industrial wastes, soils, sludge, combustion residues, sediments, stabilized materials, construction materials, and mining wastes. A tank-leaching test is used to assess the potential and speed of leaching of recycled concrete over the long-term period. This method consists of five parallel extractions of a particle size-reduced solid material in reagent water over a range of L/S values from 0.5 to 10 mL eluent/g dry material. The bottles are tumbled in an end-over-end fashion for over 24 hours of contact time based on the maximum particle size ($<0.5\text{mm}$)

of recycled concrete. At the end of the contact interval, the liquid and solid phases are roughly separated via settling. The bulk of the eluate is clarified by filtration in preparation for constituent analysis.

2.3.3 Semivolatile Organic Compounds (SVOCs) by GC/MS, EPA 8270B

EPA method 8270D was used to determine the concentration of semivolatile organic compounds in extracts prepared from RCA. The semivolatile compounds are introduced into the GC/MS by injecting the sample extract into a GC equipped with a narrow-bore fused-silica capillary column. The length is 30 m, inside diameter is 0.25 mm, and 0.25 μm film thickness silicone-coated fused-silica capillary column (Perkin Elmer, MA) was adopted for the analysis. The GC column is temperature-programmed to separate the analytes, which are then detected with an MS connected to the GC. Commercial semivolatile and PAH mixtures were purchased from Perkin Elmer (MA). The stock standard solutions were transferred into bottles equipped with PTFE lined screw caps. Each 1-mL sample extract undergoing analysis was spiked with 10 μL of the internal standard solution. Five calibration standards for semivolatile and PAH mixtures were prepared at different concentrations. 2-fluorophenol, nitrobenzene-d5, and 2-fluorobiphenyl were used for surrogate solution which was purchased from Perkin Elmer (MA).

GC/MS operating condition are as follows:

- Mass range: 35-500 amu
- *Scan* time: ≤ 1 sec/scan
- Initial temperature: 35 $^{\circ}\text{C}$, hold for 4 minutes
- Temperature program: 35-320 $^{\circ}\text{C}$ at 10 $^{\circ}\text{C}/\text{min}$
- Final temperature: 300 $^{\circ}\text{C}$, hold
- Injector temperature: 250-300 $^{\circ}\text{C}$
- Transfer line temperature: 250-300 $^{\circ}\text{C}$
- Sample volume: 1 μL
- Carrier gas: helium at 30 cm/sec.

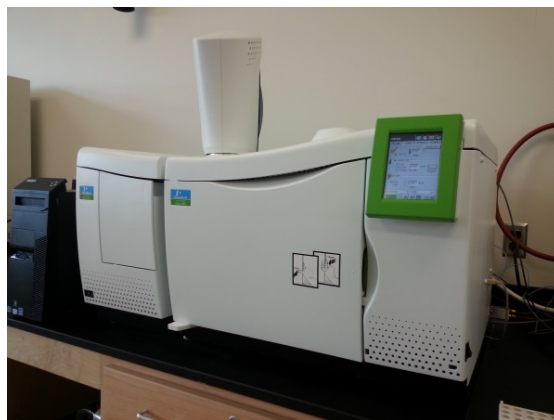


Figure 4 Clarus SQ 8 GC-MS

3. RESULTS AND DISCUSSION

Task 1. Evaluate the RCA for hydrocarbons

The determination of whether RCA could be used in oyster aquaculture depends on the character of the material, the possible pathways of risk, and appropriate environmental agency's regulations and policies. The hydrocarbon components leaching risk for RCA was the focus of this study. Vehicles through fluid spills, accidents, general wear and tear on a vehicle, etc., could possibly have spread the contamination encountered. The direct exposure to humans through inhalation, ingestion, or dermal contact and leaching to ecosystems are two major risk pathways. RCA stockpiles at recycling facilities are restricted, and concern about direct exposure at these facilities by workers is much more likely to target air emissions from the RCA crushing process. Therefore, direct exposure to humans was not addressed. The evaluation of leaching risk is performed by determining the concentration of a pollutant and comparing that concentration to the applicable guidance concentration. Hydrocarbon component leaching risk for RCA doesn't exist in most current policies; therefore the State of Maryland Department of the Environment cleanup standard for soil and groundwater was adopted to compare hydrocarbon concentrations leached from RCA (Table 5).

RCA samples from three sites were used in this study. The samples were extracted as described above and aliquots of the extracts were analyzed using GC/MS. The GC/MS results for the 3 samples are presented in Table 6 which was extracted by water and 7 which was extracted by solvent. In the case of EPA 1316 extraction, all results of organic chemical concentration in RCA were BDL (Below Detection Limit) (Table 6).

In the case of EPA 3570 extraction, all results of organic chemical concentration in RCA were BDL (Below Detection Limit) for all results of Flanigan & Sons, Inc. except anthracene, fluorine, phenanthrene and pyrene of samples collected from Machado Construction Co., Inc. and The Recycling Center where RC-6 size of RCA sample were collected. RC-6 is the smallest RCA product in recycling facilities. The concentrations of the State of Maryland Department of the Environment cleanup standard for soil and groundwater are much higher than detected concentrations. This was at least 100 times below the considered hazardous level of Protected Groundwater Standard (Table 7).

Even though, the results are not comparable with general rules of environmental fate of hydrocarbon compounds, the higher molecular weight compounds tend to sorb to the soil and persist at the site of release. Lighter petroleum products such as gasoline contain constituents with higher water solubility and volatility and lower sorption potential than heavier petroleum products such as fuel oil that has relatively low water solubility, low volatility and high sorption capacity (Stelljes and Watkin 1991).

Table 5 State of Maryland Department of the Environment cleanup standard for soil and groundwater

Semivolatile Organic Compounds (SVOCs)	Groundwater Standards		Soil Standards		
	Type I and II Aquifers*		Residential Clean-up Standard	Non-Residential Clean-up Standard	Protection of Ground water
	(ug/L)	(ug/L)	(mg/kg)	(mg/kg)	(mg/kg)
Acenaphthylene	37	0.037	470	6100	100
Anthracene	180	0.18	2300	31000	470
Benz(a)anthracene	0.2	0.0002	0.22	3.9	0.48
Benzo(b)fluoranthene	0.2	0.0002	0.22	3.9	1.5
Benzo(g,h,i)perylene	18	0.018	230	3100	680
Benzo(k)fluoranthene	0.3	0.0003	2.2	39	15
bis-(2-Ethylhexyl)adipate**	400	0.4	53	240	--
bis(2-Ethylhexyl)phthalate**	6	0.006	46	200	2900
Chrysene	3	0.003	2.2	390	48
Dibenz(a,h)anthracene	0.2	0.0002	0.022	0.39	0.46
Diethyl phthalate	2900	2.9	6300	82000	450
Di-n-butyl phthalate	370	0.37	780	10000	5000
Fluorene	24	0.024	310	4100	140
Hexachlorobenzene**	1	0.001	0.4	1.8	0.052
Hexachlorocyclopentadiene**	50	0.05	47	610	1800
isophorone	70	0.07	670	3000	0.41
Pentachlorophenol**	1	0.001	5.3	24	--
Phenanthrene	180	0.18	2300	31000	470
Pyrene	180	0.018	230	3100	680

* Type I aquifer means an aquifer having a transmissivity greater than 1,000 gallons/day/foot and a permeability greater than 100 gallons/day/square foot, and for natural water with a total dissolved solids concentration less than 500 milligrams/liter.

Type II aquifer means an aquifer having either:

A) a transmissivity greater than 10,000 gallons/day/foot, a permeability greater than 100 gallons/day/square foot and natural water with a total dissolved solids concentration of between 500 and 6,000 milligrams/liter; or

B) a transmissivity between 1,000 and 10,000 gallons/day/foot, a permeability greater than 100 gallons/day/square foot and natural water with a total dissolved solids concentration of between 500 and 1,500 milligrams/liter.

** Type I aquifer cleanup standards are same as EPA drinking water standard for organic chemicals.

Table 6 organic chemical concentration in RCA extracted by EPA 1316

Semivolatile Organic Compounds (SVOCs)/PAH	EPA 1316		
	Flanigan & Sons, Inc.	Machado Construction Co., Inc.	The Recycling Center
	mg/kg	mg/kg	mg/kg
Acenaphthylene	BDL	BDL	BDL
Anthracene	BDL	BDL	BDL
Benz(a)anthracene	BDL	BDL	BDL
Benzo(a)pyrene	BDL	BDL	BDL
Benzo(b)fluoranthene	BDL	BDL	BDL
Benzo(B)triphenylene	BDL	BDL	BDL
Benzo(g,h,i)perylene	BDL	BDL	BDL
Benzo(J)fluoranthene	BDL	BDL	BDL
Benzo(k)fluoranthene	BDL	BDL	BDL
bis-(2-Ethylhexyl)adipate	BDL	BDL	BDL
bis(2-Ethylhexyl)phthalate	BDL	BDL	BDL
Butylbenzyl phthalate	BDL	BDL	BDL
chlorobenzilate	BDL	BDL	BDL
Chrysene	BDL	BDL	BDL
Dibenz(a,h)anthracene	BDL	BDL	BDL
Diethyl phthalate	BDL	BDL	BDL
Di-n-butyl phthalate	BDL	BDL	BDL
Fluorene	BDL	BDL	BDL
Hexachlorobenzene	BDL	BDL	BDL
Hexachlorocyclopentadiene	BDL	BDL	BDL
isophorone	BDL	BDL	BDL
Pentachlorophenol	BDL	BDL	BDL
Phenanthrene	BDL	BDL	BDL
Pyrene	BDL	BDL	BDL

* Below the Detection Limit

Table 7 organic chemical concentration in RCA extracted by EPA 3570

Semivolatile Organic Compounds (SVOCs)/PAH	EPA 3570		
	Flanigan & Sons, Inc.	Machado Construction Co., Inc.	The Recycling Center
	mg/kg	mg/kg	mg/kg
Acenaphthylene	BDL*	BDL	BDL
Anthracene	BDL	2.78	1.97
Benz(a)anthracene	BDL	BDL	BDL
Benzo(a)pyrene	BDL	BDL	BDL
Benzo(b)fluoranthene	BDL	BDL	BDL
Benzo(B)triphenylene	BDL	BDL	BDL
Benzo(g,h,i)perylene	BDL	BDL	BDL
Benzo(J)fluoranthene	BDL	BDL	BDL
Benzo(k)fluoranthene	BDL	BDL	BDL
bis-(2-Ethylhexyl)adipate	BDL	BDL	BDL
bis(2-Ethylhexyl)phthalate	BDL	BDL	BDL
Butylbenzyl phthalate	BDL	BDL	BDL
chlorobenzilate	BDL	BDL	BDL
Chrysene	BDL	BDL	BDL
Dibenz(a,h)anthracene	BDL	BDL	BDL
Diethyl phthalate	BDL	BDL	BDL
Di-n-butyl phthalate	BDL	BDL	BDL
Fluorene	BDL	1.65	0.3
Hexachlorobenzene	BDL	BDL	BDL
Hexachlorocyclopentadiene	BDL	BDL	BDL
isophorone	BDL	BDL	BDL
Pentachlorophenol	BDL	BDL	BDL
Phenanthrene	BDL	0.95	0.28
Pyrene	BDL	1.40	0.06

* Below the Detection Limit

Task 2. Provide testing protocol for SHA evaluation of materials

Hydrocarbon species can enter the environment from a number of sources. The origin of the contaminants has a significant behavior upon the species present and hydrocarbons are

generally not applied to the environment for a purpose; thus, hydrocarbon contamination results almost entirely from accidents. The source of hydrocarbon in this study is unknown.

During the course of this project, a variety of tests and procedures was employed to ensure that the use of RCA would not impact the water quality, local ecosystem or any of the life states of the oysters themselves. The test and procedures employed were standard laboratory and field methodologies that are applicable to any similar material. As other new materials are presented to the SHA for use in marine environments, the SHA needs a testing protocol that can be applied to assess the potential impacts of these proposed new materials on the marine environment. This listing will provide the SHA with a standard set of protocols that can be used by other researchers to test the suitability of RCA in the marine environment.

1. Planning Stage: Preliminary evaluation for RCA

RCA sampling and preservation: RCA samples must be extracted within 14 days of collection, and analyzed within 40 days of extraction. Alternatively, samples may be frozen (- 10°C) in the field or in the laboratory. Samples frozen in the laboratory must be preserved at $4 \pm 2^\circ \text{C}$ from the time of sampling and frozen within 48 hours. A summary of sample collection, preservation, and holding times is provided in Table 8. Smaller RCA size such as RC-6 is more appropriate for extraction convenience than bigger sizes such as RC-2, RC-57 and RC-Surge which required an additional process.

Table 8 Holding times and preservatives for RCA Samples (MADEP, 2003)

Container	Preservation	Holding Time	
		Extraction	Analysis
250 mL, amber glass jar with Teflon-lined screw cap	Cool to $4 \pm 2^\circ \text{C}$	14 days	40 days
250 mL, amber glass jar with Teflon-lined screw cap, Filled to only 2/3 capacity to avoid breakage	Freeze at - 10°C	14 days	40 days

At least two separate sample collections at the same time and place under identical circumstances and managed the same throughout field and laboratory procedures are required to get presentative results. All analysis should be duplicated at least three times..

Moisture content measurement for RCA sample: Moisture content will affect the result of hydrocarbon concentration. RCA samples should be collected during several days after rain events.

Extraction and analysis: Various extraction and analysis are organized by EPA. EPA 1316 and 3570 for extraction and EPA 8270D were applied in this project, but the extraction and analysis method could be adapted based on targeted components.

Data interpretation: The results need to compare with the regulation to obtain the permission of field application. However, only a small number of the compounds are well characterized for toxicity. In this research, the results were compared with COMAR 26.08.02.03-2 which provides the numerical criteria for toxic substance in surface water (Table 3). In the case of a water sample, the results could be compared to EPA drinking water standards. The following table is an example quoted from an EPA drinking water standard related to petroleum industry activity.

Table 9 EPA Drinking Water Standard related to petroleum industry activity-

<http://www.epa.gov/your-drinking-water/table-regulated-drinking-water-contaminants>

Chemical	MCLG* (mg/L)	MCL** or TT*** (mg/L)	Potential Health Effects from Long-Term Exposure Above the MCL	Sources of Contaminant in Drinking Water
Ethylbenzene	0.7	0.7	Liver or kidneys problems	Discharge from petroleum refineries
Ethylene dibromide	zero	0.00005	Problems with liver, stomach, reproductive system, or kidneys; increased risk of cancer	Discharge from petroleum refineries
Toluene	1	1	Nervous system, kidney, or liver problems	Discharge from petroleum factories
Xylenes (total)	10	10	Nervous system damage	Discharge from petroleum factories; discharge from chemical factories

Definitions:

*Maximum Contaminant Level Goal (MCLG) - The level of a contaminant in drinking water below which there is no known or expected risk to health. MCLGs allow for a margin of safety and are non-enforceable public health goals.

**Maximum Contaminant Level (MCL) - The highest level of a contaminant that is allowed in drinking water. MCLs are set as close to MCLGs as feasible using the best available treatment technology and taking cost into consideration. MCLs are enforceable standards.

***Treatment Technique (TT) - A required process intended to reduce the level of a contaminant in drinking water.

2. Field Application Stage

RCA selection: Based on the result of EPA 3570, RC-2 (1^{1/2}” to 2^{1/2}”) collected from Flanigan & Sons, Inc. was not releasing any hydrocarbon components, but the result of RC-6 (< 1^{1/2}”) which is smaller in size than RC-2 collected from Machado Construction Co., Inc. and The Recycling Center showed that several hydrocarbons components were detected. Fine material has higher sobbed area and more potential to containing harmful components. Thus, bigger RCA size such as RC-Surge is recommended.

RCA handling: The RCA origin/source information will be helpful to remove any concern for potential hydrocarbon source. Selected RCA should be sitting in the stockpile after crushing for at least 14 days to evaporate VOSSs.

RCA application: Washing RAC is recommended to remove fine particles on RCA surface. Fine particle has higher sorbed surface and higher potential of containing pollutants than that of bigger size RCA. When applying RCA to the field, RCA should be gently poured into water to minimize disturbing water system.

After RCA application: Periodical water sampling and analysis plan are recommended to monitoring any detrimental effect to Chesapeake Bay watershed.

4. CONCLUSIONS

The purpose of this study was to determine if the use of RCA from the surrounding roadway or building demolition would be appropriate for use as bottom conditioning material for oyster aquaculture. The major concern was that accidental spills onto the road surface from vehicles could contaminate the pavement and the surrounding road site. This possible contamination could make the material unsuitable for use in the bottom applications under water.

The hydrocarbons that are most strongly sorbed onto soil organic matter will be most resistant to loss or alteration by the other processes. Conversely, the more volatile/soluble hydrocarbons will be the most susceptible to change by volatilization/reaction/leaching/biodegradation. Thus, volatile components were not chosen because volatile compounds spilled onto the roadway would evaporate quickly. RCA will be placed in the recycling plant before the crushing process and also will sit for awhile in the stockpile before application. In almost all cases of hydrocarbon contamination, the compounds of interest were the semivolatile components included in PAHs.

The resulting water chemistry from the following leaching tests were used to evaluate acute and chronic water quality necessary for protecting marine and estuarine life based on the water quality standard of Maryland (COMAR 26.08.02.03-2).

RCA samples from three sites were used in this study. All results of organic chemical concentration in RCA were BDL (Below Detection Limit) (Table 6 and 7) for both extraction methods of Flanigan & Sons, Inc. Anthracene, fluorine, phenanthrene and pyrene were detected at EAP 3570 extraction for the samples collected from Machado Construction Co., Inc. and The Recycling Center. However, the concentrations were at least 100 times lower than COMAR 26.08.02.03-2. The result concluded that RCA should give no concern for hydrocarbon components releasing into the Chesapeake Bay watershed, if RCA is used as a bottom conditioning material for oyster aquaculture.

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