

## THERMALLY TREATED COAL TAR-CONTAMINATED SOIL AS A CONCRETE AGGREGATE

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### ABSTRACT

*The current practice of landfilling thermally treated and untreated coal tar-contaminated soil is expensive, and it is a waste of a potentially useful material and landfill space. A less expensive and possibly more practical alternative is to use coal tar-contaminated soil as an aggregate in concrete. Because little work has been done in this area, the present study was undertaken to evaluate the effects of coal tar-contamination and thermal treatment on the physical properties of soil and the strength of concrete containing contaminated soil as an aggregate. Also assessed was the leachability of coal tar constituents (specifically, polycyclic aromatic hydrocarbons (PAHs)) from contaminated soil, concrete containing contaminated soil, and concrete containing thermally-treated contaminated soil.*

*Mechanical sieve analyses and direct shear tests showed no significant change in grain size distribution or internal friction angle for contaminated and uncontaminated soil treated at 250 °C and 550 °C. Compressive and splitting tensile strength tests performed on concrete specimens showed a slight decrease in strength with the degree of incorporation of contaminated aggregate. Concrete compressive strength, elastic modulus, and tensile strength also decreased as the aggregate treatment temperature increased, especially at temperatures in excess of 450 °C. However, the strength decreases were small (less than 15%), and the concrete strength was still within typical values for structural concrete. The leachability tests showed that as treatment temperature was increased, levels of PAHs in the soil significantly decreased. From the leachability tests performed on samples of crushed concrete that contained contaminated soil, it appears that only low molecular weight PAHs (i.e., less than or equal to 166 g/mol) were stabilized by the concrete. The poor stabilization of higher molecular weight PAHs may be an artifact of the sample preparation method which required that the concrete be crushed prior to leaching.*

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## INTRODUCTION

Between the mid-1800s and the 1960s, manufactured gas plants (MGPs) were widely used in the U.S. to produce gaseous fuel from available coal, coke, oil and other fossil fuels (Hatheway, 1997). A by-product of coal gasification was coal tar, a dense, non-aqueous phase liquid that was often disposed of on-site in wells, pits and lagoons. Coal tar was also introduced into the subsurface environment due to leaks and spills from tanks and piping networks, as well as the dismantling of plants taken out of service (Luthy et al., 1994). The total number of sites in the U.S. contaminated with coal tar is estimated to be more than 32,000 (Hatheway, 1997). Coal tar contamination is of particular concern because it contains a wide range of hazardous chemicals, including polycyclic aromatic hydrocarbons (PAHs), many of which are known or suspected carcinogens (IARC, 1983).

Coal tar-contaminated soil is commonly placed in landfills or treated using thermal processes such as thermal desorption, incineration, and coburning. However, treated soils often contain residual contamination and consequently must be disposed of in solid waste landfills. In a review of 200 coal tar-contaminated sites at which remediation activities have been performed, Owen and Unites (1999) found that 55% of these sites landfilled contaminated soils and 49% employed thermal treatment (desorption or coburning). Reusing contaminated soil or treated soil containing residual contamination may be an alternative to disposal in landfills. One possible use for this soil is as an aggregate in concrete. Incorporating coal tar-contaminated soil into concrete may help to stabilize residual contaminants, preventing them from leaching out of the soil. Also, the possibility of marketing soil with residual contamination for use as aggregate in concrete provides a possible financial incentive for remediation.

To date, little work has been done to determine whether thermally treated coal tar-contaminated soil is a suitable concrete aggregate. Several studies have reported relatively high (25% to 98%) stabilization of pure organic compounds (not compounds associated with contaminated soils) in cement (Owens et al., 1996; Faschan et al., 1996; Conner, 1995; Hebatpuria et al., 1999). However, only one study was found in which soils contaminated with organics were incorporated into concrete. In this study it was found that the effective diffusivity of benzene was reduced by as much as five orders of magnitude when contaminated sand was incorporated into concrete (Ezeldin and Vaccari, 1995). Because the investigators did not report on the physical properties of the concrete (i.e., tensile and compressive strength, elastic modulus, etc.), applications for concrete containing contaminated aggregate are not known.

This paper presents an evaluation of some of the physical and chemical properties of thermally treated coal tar-contaminated soil and of concrete incorporating this soil as an aggregate. The specific objectives of this study were to determine the effects of thermal treatment on the physical characteristics of natural and coal tar-contaminated soil, evaluate the strength characteristics of concrete containing thermally treated natural and coal tar-contaminated soil as aggregate, and assess the leachability of PAHs from concrete containing coal tar-contaminated soil as aggregate.

## **LABORATORY TEST METHODS**

### **Soil Mixture Preparation and Geotechnical Testing Procedure**

The soil used in this study was a gap-graded mixture of tan to brown sub-angular to sub-rounded sand and gravel. This type of soil is typical of many former MGP sites (Luthy et al., 1994). The contaminated soil was prepared by mixing coal tar sludge obtained from a former MGP site in New Bedford, Massachusetts with the air-dried sand and gravel mixture. The final contaminated soil was mixed to contain approximately 3,000 ppm PAHs by weight.

A furnace was used to thermally treat 10 kg batches of the sand and gravel mixtures. The furnace was preheated to a specified temperature between 250°C and 650°C before each treatment. The soil was heated for one hour, then promptly removed and spread in a large metal pan to cool. The residence time of one hour was meant to approximate actual practice and temperatures of 250°C to 650°C represent the range commonly employed during thermal desorption and incineration. However, the laboratory method did not employ mixing by rotary kiln during thermal treatment or cooling by quenching with water, as is commonly performed in thermal treatment processes.

Mechanical sieve tests (ASTM D422) and direct shear tests (ASTM D3080) were performed to characterize the physical properties of the natural and contaminated soil before and after thermal treatment. Two sieve tests were used in order to characterize the full range of particle sizes. A coarse sieve analysis, with sieve sizes between 26.67 mm and 2 mm was initially performed. The portion of soil retained on the 2 mm sieve and in the pan of the coarse sieve analysis was set aside for a fine sieve analysis using sieve sizes between 2 mm and 0.075 mm.

The direct shear tests were performed using normal loads of 36 kg and 66 kg during shear. Based on ASTM specifications and the shear mold dimensions, 6.5 cm diameter and 5 cm height, the maximum particle size appropriate for the mold should be 6.5 mm. The soil particle size in this investigation was reduced using a 9.42 mm sieve. Soil was compacted into the shear mold by tamping three equal lifts each with ten blows using a Harvard miniature compactor (290 g hammer dropped 38 cm). The thermally treated soil was also mechanically sieved after direct shear to evaluate particle breakage using the fine sieve analysis.

### **Concrete Mix Design and Strength Testing Procedure**

The concrete mix design used in this study combined cement, water, coarse aggregate, and sand in the proportions 1 : 0.52 : 3 : 2, respectively, by weight. Table 1 shows the actual mix proportions for the 13 batches of concrete prepared for this study. From each batch, three 10.2 cm x 20.3 cm cylinders were prepared for compression tests, and three 10.2 cm x 15.2 cm cylinders were prepared for splitting tension tests. Elastic modulus, Poisson's ratio, and compressive strength were determined following the procedures of ASTM C369 and ASTM C39. The indirect tensile strengths of the concrete were determined by performing Brazilian splitting tensile tests as per ASTM C496. All specimens were cured in a humid room for 21 days prior to testing.

**Table 1. Concrete Batches.**

Batch Characteristics				Batch Proportions as kg/m <sup>3</sup> of Concrete					
Batch Name	Batch Aggregate Replacement Description	Type of Aggregate Replacement <sup>1</sup>	Replacement of Aggregate by Weight (%)	Cement	Water	Fine Aggregate	Thermally Treated Fine Aggregate	Coarse Aggregate	Thermally Treated Coarse Aggregate
U-0%	standard (no replacement)	U	0.0	353.9	181.5	763.2	0.0	1053.3	0.0
U-40%-250	40% replacement with 250* C treated soil	U	40.4	353.9	181.3	470.2	244.8	611.9	489.7
U-40%-375	40% replacement with 375* C treated soil	U	40.0	353.9	181.4	521.4	242.3	568.3	484.6
U-40%-450	40% replacement with 450* C treated soil	U	40.0	353.9	181.4	521.4	242.3	568.3	484.6
U-40%-550	40% replacement with 550* C treated soil	U	40.4	353.9	181.3	470.2	244.8	611.9	489.7
U-40%-650	40% replacement with 650* C treated soil	U	40.0	353.9	181.4	521.4	242.3	568.3	484.6
U-40%-550CA	40% replacement with 550* C treated gravel (coarse)	U	41.0	353.9	181.5	763.2	0.0	309.0	744.2
U40%-550FA	40% replacement with 550* C treated sand (fine)	U	40.8	353.9	181.5	21.2	742.0	1053.3	0.0
U-100%-550	100% replacement with 550* C treated soil	U	100.0	353.9	181.0	0.0	764.8	0.0	1052.2
C-10%	10% replacement with untreated contaminated soil	C	10.0	353.9	181.4	751.3	0.0	1065.3	0.0
C-10%-250	10% replacement with 250* C treated contaminated soil	C	10.0	353.9	181.4	690.7	60.6	944.1	121.2
C-40%	40% replacement with untreated contaminated soil	C	40.0	353.9	181.4	763.7	0.0	1052.9	0.0
C-40%-250	40% replacement with 250* C treated contaminated soil	C	40.4	353.9	181.3	470.2	470.2	611.9	489.7

1. 'U' indicates uncontaminated, 'C' indicates contaminated.

## Chemical Analysis of Soil and Concrete

EPA Method 1311-Toxicity Characteristic Leaching Procedure (TCLP) was used to assess the mobility of selected PAHs from coal tar-contaminated soil and concrete samples. This laboratory procedure is not meant to extract all contaminants from the solids, rather, it is designed to approximate leaching conditions present in the environment given slightly acidic precipitation and groundwater conditions. The TCLP method requires size reduction of the soil and concrete samples so that all particles are smaller than 1 cm. Concrete samples, which were gathered from failed cylinders after strength testing, were crushed using a rubber mallet, then passed through a 1-cm sieve.

Based on preliminary evaluations outlined in the TCLP test, the extraction fluid used for all samples consisted of 5.7 mL glacial acetic acid and 64.3 mL 1N NaOH diluted to 1 liter with distilled water. This solution had a pH of  $4.93 \pm 0.05$ . For each extraction, 50 g of solid sample (soil or concrete) was placed in a 1 liter jar with 940 mL extraction fluid. The jars were then placed in a rotary agitation device and rotated at 30 rpm for 18 hours at ambient temperature ( $23 \pm 2$  °C). Following the extraction procedure, the solid sample was separated from the liquid extract by vacuum filtering through a 0.7  $\mu$ m glass fiber filter. Next, the TCLP extract was liquid-liquid extracted using 500 mL of methylene chloride (DCM) for 24 hours. The DCM solution was then separated from the water-based TCLP extract solution using a separatory funnel. The Kuderna-Danish method was used to concentrate the DCM to less than 8 mL. Just prior to analysis, this extract was further concentrated by evaporation using pressurized nitrogen gas to less than 1 mL, then approximately 10  $\mu$ L of benzene was added (to provide a solvent with a lower vapor pressure than DCM) and the final solution was concentrated to a volume appropriate for analysis. All glassware was washed with Alconox detergent, then double-rinsed with distilled water, methanol, and DCM.

Concentrations of 15 PAH compounds in the TCLP extract were quantified by gas chromatography (GC) with mass selective detection (MSD) using a selective ion monitoring (SIM) method. The GC/MSD system consisted of a Hewlett Packard (HP) 6890 GC and a HP 5973 MSD. Data acquisition and analysis were conducted using HP MS ChemStation software. The GC system was equipped with a 30 m HP-5 5% phenyl methyl siloxane capillary column that had an inside diameter of 0.25 mm and a 0.25 $\mu$ m film thickness. The GC oven was programmed as follows: (i) after injection hold for 1.5 min at 50°C, (ii) ramp to 310°C at 8°C/min, and (iii) hold for 10 min at 310°C. The flow of the carrier gas (He) was maintained at 1 mL/min; the transfer line temperature was maintained at 280°C. Samples were injected in 1.4  $\mu$ L volumes in a split/splitless injector operated in the splitless mode. The mass range scanned was 50 to 450 amu in the electron impact mode (70 eV); the scanning rate was 1.8/s. Quantification of PAH compounds was done by comparing peak areas of compounds detected in extract samples with those of known standards. Calibration curves were developed with authentic standards for 15 compounds. TCLP extract samples and standards were injected in triplicate.

RESULTS AND DISCUSSION

Physical Properties of Soil

After being mixed with coal tar sludge, the formerly tan to brown sand and gravel mixture changed to a dark black-brown and had a distinct odor. After being treated for one hour at 250°C, the contaminated soil had lightened somewhat in color to a medium brown-gray. This treated soil still had a strong odor that diminished as it cooled. However, the odor never completely abated. The contaminated soil that was treated for one hour at 550°C came out of the furnace with a dark pink-brown color and only a slight odor.

Results of the mechanical sieve analyses for all soil groups, as shown in Figure 1 and in Table 2, indicated that thermal treatment had no significant effect on the particle size distribution or distribution parameters of the gap-graded soil.

Results of the direct shear tests performed on densely packed (dry density range of 19.4 kN/m<sup>3</sup> to 20.1 kN/m<sup>3</sup>) untreated and thermally treated soils indicated that thermal treatment did not significantly affect shear strength. As shown in Figure 2, the effective internal soil friction angles ( $\phi'$ ) for all five soils were between 39° and 42°, with an average of 40.6°. Mechanical sieve analysis performed on the thermally treated soils after direct shear testing showed that no significant particle breakage had occurred.

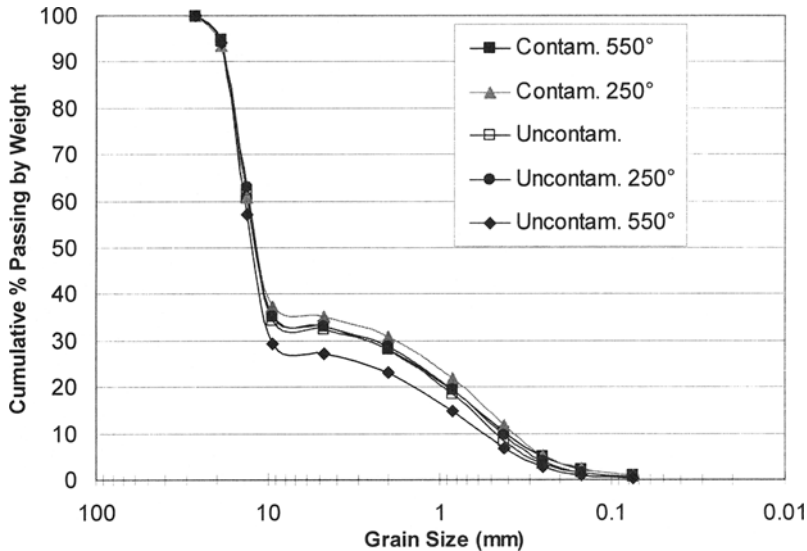
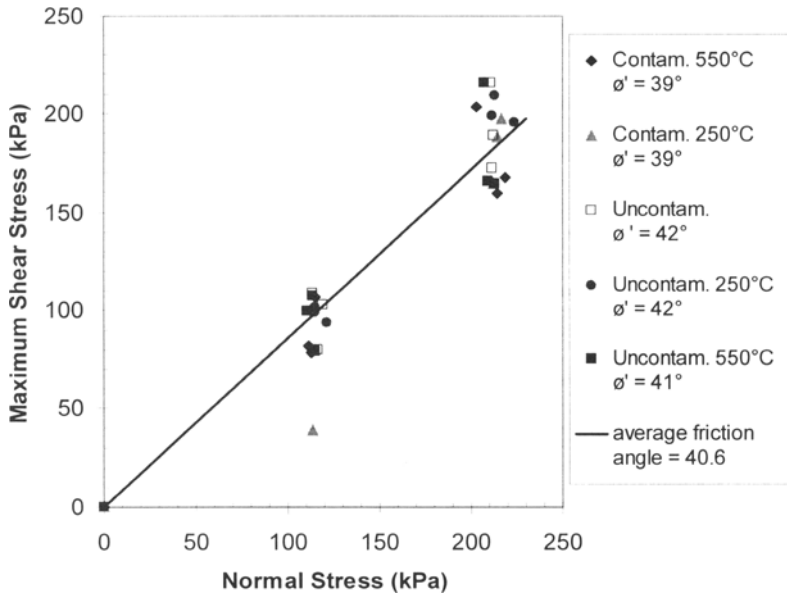


Figure 1. Comparison of Average Grain-Size Distribution of Soil.

**Table 2. Summary of Particle-Size Distribution Parameters.**

Soil Type	Coefficient of Uniformity, $C_u$	Coefficient of Gradation, $C_c$
Uncontaminated Soil		
Gravel Portion	1.53 +/- 0.05	0.99 +/- 0.03
Sand Portion	3.71 +/- 0.02	0.98 +/- 0.04
Uncontaminated Soil, 250°C		
Gravel Portion	1.52 +/- 0.04	1.00 +/- 0.02
Sand Portion	3.73 +/- 0.02	0.96 +/- 0.01
Uncontaminated Soil, 550°C		
Gravel Portion	1.53 +/- 0.01	0.98 +/- 0.01
Sand Portion	3.72 +/- 0.22	0.98 +/- 0.06
Contaminated Soil, 250°C		
Gravel Portion	1.59 +/- 0.01	1.03 +/- 0.07
Sand Portion	4.07 +/- 0.50	0.97 +/- 0.11
Contaminated Soil, 550°C		
Gravel Portion	1.58 +/- 0.05	1.01 +/- 0.04
Sand Portion	4.01 +/- 0.48	1.03 +/- 0.03



**Figure 2. Comparison of Maximum Shear Stress vs. Normal Stress for Soil.**

### **Physical Properties of Concrete**

Distinct changes in the physical properties of concrete were observed as the treatment temperature of the aggregate was increased. Concrete cylinders containing aggregate treated at temperatures less than 450°C generally failed with the cracks going around the coarse aggregate. In the concrete that contained aggregate treated at higher temperatures, fracturing occurred through the coarse aggregate, causing the aggregate to split into pieces. Results of the compression tests (Figures 3a and 3b) show little change in compressive strength or elastic modulus with aggregate treatment temperatures up to 450°C. However, decreases in both compressive strength and elastic modulus were apparent in concrete samples incorporating aggregate treated at temperatures greater than 450°C. At an aggregate treatment temperature of 650°C, compressive strength and elastic modulus were decreased by 9% and 14%, respectively, as compared to concrete containing untreated aggregate. Results of split cylinder tests on uncontaminated soil, shown in Figure 4, demonstrate a general decrease in splitting tensile strength as the treatment temperature of replaced aggregate is increased. The splitting tensile strength decreased on average by approximately 1.3% per 100°C increment in treatment temperature. It is expected that at treatment temperatures greater than 250°C, the splitting tensile strength of concrete containing contaminated soil would likewise decrease.

The results also show that when the amount of untreated contaminated aggregate substitution is increased from 10% to 40%, the splitting tensile strength of the concrete decreases. This is most likely due to coal tar coating the surface of aggregate and preventing bonds from forming between aggregate and cement. At a treatment temperature of 250°C, the splitting tensile strengths of concrete containing contaminated aggregate are similar to concrete containing uncontaminated aggregate, suggesting that the negative effects regarding aggregate-cement bonds have been eliminated by thermal treatment.

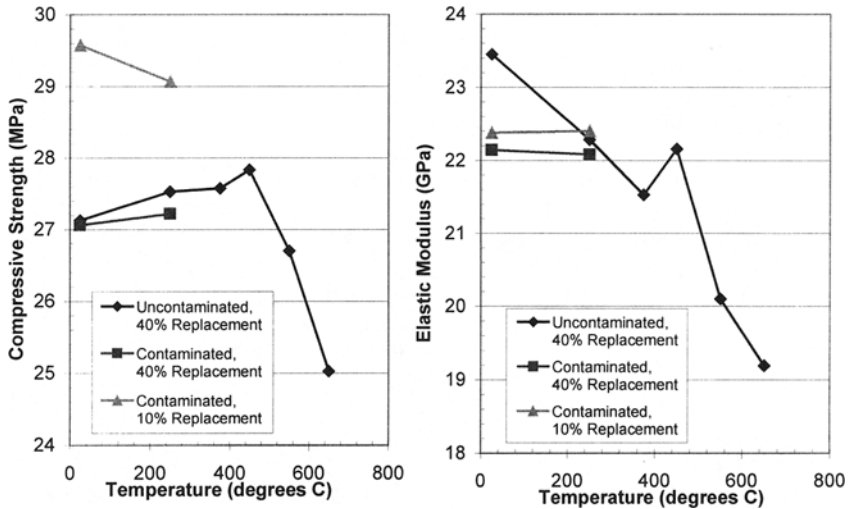
Although changes in strength parameters of concrete due to thermal treatment of aggregate are clear, the small decreases in compressive strength, elastic modulus and splitting tensile strength may not be critical, as even the reduced values remain within typical range for structural concrete.

### **Chemical Analysis of Soil TCLP Extracts**

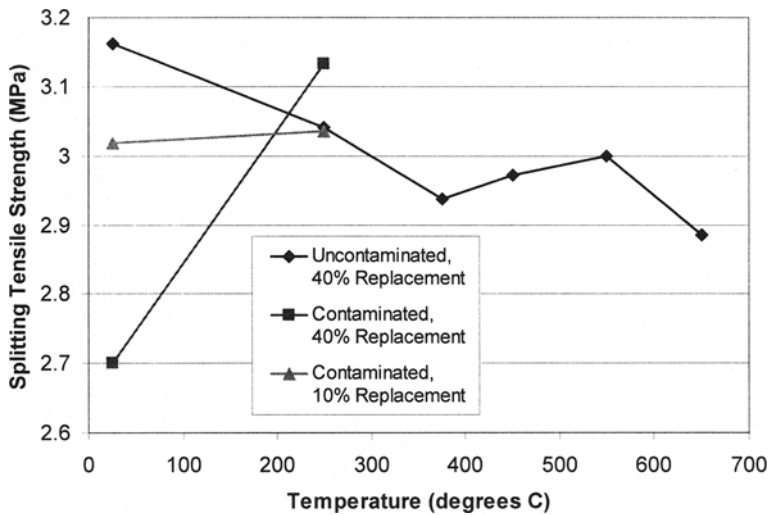
After the 18 hour extraction process, the uncontaminated soil, untreated contaminated soil, and 250°C-treated contaminated soil TCLP solutions appeared cloudy from the tan-colored fines that remained in suspension after agitation was completed. The 550°C-treated contaminated soil TCLP solution was completely opaque and dark brown, indicating a greater degree of suspended fine particles. The pH of the TCLP extract was  $4.93 \pm 0.05$  both before and after extraction.

The results in Table 3 show that, as expected, the amounts of leachable PAHs in soil decreased as treatment temperature increased. Compared to the TCLP extract of the untreated contaminated soil, the total amount of PAHs in the extracts of the soils treated at 250°C and 550°C decreased by 73% and 98%, respectively. For the 250°C-treated soil extract, the concentrations of pyrene, benz(a)anthracene, chrysene, benzo(b)fluoranthene and benzo(a)pyrene were not significantly different





**Figure 3. a) Temperature of Aggregate Treatment vs. Compressive Strength and b) Temperature of Aggregate Treatment Elastic Modulus of Concrete**



**Figure 4. Temperature of Soil Treatment versus Tensile Strength of Concrete Specimens.**

Table 3. Concentrations of PAHs in Soil TCLP Extracts.

MW <sup>1</sup> (g/mol)	Compound	Untreated Uncontaminated Soil		Untreated Contaminated Soil	250° C Treated Contaminated Soil		550° C Treated Contaminated Soil	
		Concentration in TCLP extract <sup>2</sup> (ppb)	Concentration in TCLP extract <sup>2</sup> (ppb)	Concentration in TCLP extract <sup>2</sup> (ppb)	Concentration in TCLP extract <sup>2</sup> (ppb)	% Reduction <sup>3</sup>	Concentration in TCLP extract <sup>2</sup> (ppb)	% Reduction <sup>3</sup>
128	Naphthalene	0.014 +/- 0.001	205 +/- 21.6	8.41 +/- 0.266	4.18 +/- 0.982	96 %	4.18 +/- 0.982	98 %
152	Acenaphthene	0.019 +/- 0.004	32.9 +/- 9.19	2.58 +/- 0.278	0.042 +/- 0.003	92 %	0.042 +/- 0.003	100 %
152	Acenaphthylene	0.005 +/- 0.004	82.4 +/- 6.77	4.23 +/- 0.642	0.079 +/- 0.010	95 %	0.079 +/- 0.010	100 %
166	Fluorene	nd (0.001)	53.8 +/- 6.38	9.03 +/- 1.35	0.115 +/- 0.013	83 %	0.115 +/- 0.013	100 %
178	Phenanthrene	0.039 +/- 0.002	101 +/- 9.55	76.6 +/- 14.7	2.66 +/- 0.972	24 %	2.66 +/- 0.972	97 %
178	Anthracene	nd (0.001)	19.7 +/- 4.05	7.23 +/- 1.61	0.136 +/- 0.018	63 %	0.136 +/- 0.018	99 %
202	Fluoranthene	nd (0.001)	23.1 +/- 3.12	22.0 +/- 5.82	1.48 +/- 0.652	5.1 %	1.48 +/- 0.652	94 %
202	Pyrene	nd (0.001)	10.3 +/- 2.04	14.1 +/- 4.61	0.702 +/- 0.084	0 %	0.702 +/- 0.084	93 %
228	Benz(a)anthracene	nd (0.001)	0.493 +/- 0.080	0.526 +/- 0.163	0.063 +/- 0.023	0 %	0.063 +/- 0.023	87 %
228	Chrysene	nd (0.001)	0.276 +/- 0.038	0.327 +/- 0.128	0.045 +/- 0.014	0 %	0.045 +/- 0.014	84 %
252	Benzo(b)fluoranthene	nd (0.008)	0.217 +/- 0.034	0.235 +/- 0.047	0.034 +/- 0.023	0 %	0.034 +/- 0.023	84 %
252	Benzo(k)fluoranthene	nd (0.001)	nd (0.009)	nd (0.008)	nd (0.001)	-	nd (0.001)	86 %
252	Benzo(a)pyrene	nd (0.001)	0.115 +/- 0.020	0.124 +/- 0.012	0.021 +/- 0.022	0 %	0.021 +/- 0.022	82 %
276	Indeno(1,2,3-cd)pyrene	nd (0.001)	nd (0.009)	nd (0.008)	nd (0.001)	-	nd (0.001)	86 %
276	Benzo(g,h,i)perylene	nd (0.001)	nd (0.009)	nd (0.008)	nd (0.001)	-	nd (0.001)	86 %
<b>TOTALS</b>		<b>0.076</b>	<b>530</b>	<b>145</b>	<b>9.56</b>	<b>73 %</b>	<b>9.56</b>	<b>98 %</b>

1. "MW" indicates molecular weight.

2. "nd(0.005)" indicates concentration below detection limit (detection limit indicated in parentheses).

3. Percent reduction calculated as: 100 - (100 x concentration in treated contaminated soil/ concentration in untreated contaminated soil).

4. Note: All samples were extracted in duplicate and each extract was analyzed in triplicate (n=6).

than those in the untreated soil extract. In fact, for these compounds, the concentrations in 250°C treated soil actually exceeded the concentrations in untreated soil. While these differences in concentrations are not statistically significant, the results indicate that at this temperature there was no net removal of higher molecular weight (i.e., greater than 202 g/mol) PAHs. In the 550°C - treated soil extract, the removal efficiencies for lower molecular weight PAHs (i.e., less than or equal to 202 g/mol) were greater than 90%, while for higher molecular weight PAHs the removal efficiencies were greater than 80%.

Factors related to how the thermal treatment was accomplished may explain the lower than expected reductions of PAHs. In this study we used a furnace to perform thermal treatment. The soil samples were placed in bowls inside the furnace for one hour at the desired temperature and then removed to cool. In rotary kiln incinerators, used widely in treating contaminated soils, the contaminated soil is constantly mixed by the rotation of the furnace bed. These conditions greatly favor the volatilization and thermal destruction of PAHs in contaminated soils. It is expected that the reduction efficiencies achieved with a rotary kiln would be significantly higher than what was observed under laboratory conditions.

### **Chemical Analysis of Concrete TCLP Extracts**

After the 18 hour extraction process, TCLP extracts of the concrete samples appeared opaque and gray in color. Upon examining the filtered solids, it was apparent that the concrete chunks had partially disintegrated, leaving behind loose aggregate and fine cement powder. During extraction, the pH of the extraction fluid changed from 4.93 ( $t = 0$ ) to 12.13 ( $t = 18$  hrs) due to the alkaline nature of the concrete.

The concentrations of PAHs measured in the concrete TCLP extracts are shown in Table 4. The results for the C10% and C40% concrete indicate that incorporation of contaminated soil into concrete caused some stabilization of PAHs. The total concentration of 15 PAHs measured in the C10% and C40% extracts was on average 82% lower than what was expected had no stabilization occurred. Stabilization appeared to be greatest for the PAHs with molecular weights less than or equal to 178 g/mol. For higher molecular weight PAHs, the measured concentrations actually exceeded expected concentrations based on the mass of contaminated aggregate added to the concrete. While these increases in concentrations are not statistically significant, the results provide clear indication that the higher molecular weight PAHs were not well stabilized in the concrete.

Results similar to these were obtained for the concrete containing soil treated at 250 °C. Lower molecular weight PAHs were relatively well stabilized in both C10%-250 and C40%-250 concrete, while the higher molecular weight compounds were not. In contrast to the results for C10% and C40% concrete, the total concentrations of 15 PAHs measured in the C10%-250 and C40%-250 extracts were comparable to what was expected in the absence of stabilization. Driven largely by high levels of phenanthrene, fluoranthene, and pyrene, the total concentration of 15 PAHs measured in the C10%-250 extract was actually somewhat higher than expected in the absence of stabilization. Similarly, the total PAH concentration for the C40%-250 extract was only slightly lower than what was expected in the absence of stabilization. It is not clear why the higher molecular weight PAH were not better

Table 4: Concentrations of PAHs in Concrete TCLP Extracts.

MW <sup>1</sup> (g/mol)	Compound	C10%			C10%-250			C40%			C40%-250		
		Expected in absence of stabilization <sup>2</sup> (ppb)	Measured <sup>3</sup> (ppb)	% stab. <sup>4</sup>	Expected in absence of stabilization <sup>2</sup> (ppb)	Measured <sup>3</sup> (ppb)	% stab. <sup>4</sup>	Expected in absence of stabilization <sup>2</sup> (ppb)	Measured <sup>3</sup> (ppb)	% stab. <sup>4</sup>	Expected in absence of stabilization <sup>2</sup> (ppb)	Measured <sup>3</sup> (ppb)	% stab. <sup>4</sup>
128	Naphthalene	15.7	<b>0.057</b> +/- 0.015	100	0.614	<b>0.043</b> +/- 0.025	93	62.7	<b>0.378</b> +/- 0.136	99	2.57	<b>0.210</b> +/- 0.046	92
152	Acenaphthene	2.5	nd (0.001)	100	0.167	<b>0.012</b> +/- 0.027	93	10.0	<b>0.061</b> +/- 0.026	99	0.767	<b>0.197</b> +/- 0.038	74
152	Acenaphthylene	6.29	<b>0.044</b> +/- 0.019	99	0.317	<b>0.052</b> +/- 0.005	84	25.2	<b>0.394</b> +/- 0.108	98	1.30	<b>0.169</b> +/- 0.020	87
166	Fluorene	4.11	<b>0.026</b> +/- 0.008	99	0.690	<b>0.085</b> +/- 0.013	88	16.4	<b>0.406</b> +/- 0.134	98	2.79	<b>0.232</b> +/- 0.023	92
178	Phenanthrene	7.63	<b>0.256</b> +/- 0.173	97	5.77	<b>6.91</b> +/- 1.63	0	30.8	<b>3.93</b> +/- 0.925	87	23.6	<b>16.2</b> +/- 3.51	31
178	Anthracene	1.51	<b>0.238</b> +/- 0.142	84	0.553	<b>0.570</b> +/- 0.144	0	6.03	<b>1.82</b> +/- 0.556	70	2.23	<b>1.51</b> +/- 0.157	33
202	Fluoranthene	1.71	<b>3.73</b> +/- 1.28	0	1.62	<b>5.61</b> +/- 1.28	0	7.02	<b>9.18</b> +/- 1.27	0	6.73	<b>10.2</b> +/- 2.75	0
202	Pyrene	0.732	<b>3.40</b> +/- 1.51	0	1.02	<b>3.15</b> +/- 1.05	0	3.10	<b>9.46</b> +/- 1.33	0	4.31	<b>8.13</b> +/- 2.23	0
228	Benz(a)anthracene	0.023	<b>0.199</b> +/- 0.119	0	0.025	<b>0.228</b> +/- 0.041	0	0.136	<b>0.511</b> +/- 0.098	0	0.148	<b>0.404</b> +/- 0.139	0
228	Chrysene	0.009	<b>0.155</b> +/- 0.077	0	0.013	<b>0.205</b> +/- 0.067	0	0.073	<b>0.380</b> +/- 0.070	0	0.089	<b>0.311</b> +/- 0.129	0
252	Benz(b)fluoranthene	0.017	<b>0.060</b> +/- 0.024	0	0.018	<b>0.084</b> +/- 0.013	0	0.066	<b>0.106</b> +/- 0.021	0	0.073	<b>0.091</b> +/- 0.045	0
252	Benz(k)fluoranthene	-	nd (0.001)	-	-	nd (0.002)	-	-	nd (0.001)	-	-	nd (0.001)	-
252	Benz(a)pyrene	0.003	<b>0.020</b> +/- 0.013	0	0.003	<b>0.029</b> +/- 0.002	0	0.029	<b>0.044</b> +/- 0.012	0	0.032	<b>0.028</b> +/- 0.013	15
276	Indeno(1,2,3-cd)pyrene	0.001	nd (0.001)	-	0.001	nd (0.002)	-	0.000	nd (0.001)	-	0.001	nd (0.001)	-
276	Benz(g,h,i)perylene	0.001	nd (0.001)	-	0.001	nd (0.002)	-	0.000	nd (0.001)	-	0.001	nd (0.001)	-
TOTALS		40.2	<b>8.19</b>	<b>80</b>	10.8	<b>17.0</b>	<b>0</b>	162	<b>26.7</b>	<b>83</b>	44.6	<b>37.7</b>	<b>16</b>

1. "MW" indicates molecular weight.

2. Concentration of PAHs expected in the absence of stabilization, calculated from percent contaminated soil in concrete by weight and PAH concentrations measured in TCLP analysis for each soil.

3. "nd(0.005)" indicates concentration below detection limit (detection limit indicated in parentheses).

4. "% stab." indicates percent stabilized and is calculated as: 100 - (100 x expected concentration - measured concentration) / expected concentration.

5. Notes:

- C10% and C10%-250 concrete contain 7.6% contaminated aggregate by weight. C40% and C40%-250 concrete contain 31% contaminated aggregate by weight.
- All samples were extracted in duplicate and each extract was analyzed in triplicate (n=6).
- All concentrations were blank corrected. Concentrations in plain concrete blanks ranged from nd(0.001) to 0.085 ppb (Phenanthrene).

stabilized in the concrete. It is possible that by crushing the concrete and subjecting the pieces to rotary extraction, which caused further particle breakage, the surface area of the concrete was sufficiently increased to allow near complete dissolution of higher molecular weight PAHs. But if this were the case, it would also be expected that less than 166 g/mol PAHs should also have been poorly stabilized. That our results indicate otherwise may reflect unaccounted losses (rather than stabilization) of lower molecular weight (less than 166 g/mol) PAHs during concrete preparation, curing, TCLP extraction, or chemical analysis.

Additional factors that may have affected the results have to do with the TCLP extraction methods used. Prior to extraction, it was necessary to break the concrete into small pieces, and this often resulted in the fracture of gravel-sized aggregate. As a result, the TCLP extracts of the concrete represent leachable PAHs on both the sand and gravel whereas the TCLP extracts of the soil represent leachable PAHs only on particles passing a 1 cm sieve. Since smaller soil particles have a greater specific surface area than gravel sized particles, a given amount of contaminated sand would be expected to contain a greater mass of PAHs than the same amount of contaminated sand and gravel. Consequently, this method may have led to an overestimate of the expected concentrations and percent stabilization of lower molecular weight PAHs in concrete. Also, during rotary extraction, the samples of concrete partially disintegrated, which may have caused fine particles to remain in TCLP extract even after filtration through a 0.7  $\mu\text{m}$  glass fiber filter. Since PAHs tend to sorb on to particulate matter, this may have resulted in increased concentrations of higher molecular weight PAHs in the filtered extract. It is likely that the degree of concrete disintegration and PAH leachability would be decreased if the concrete was not crushed prior to TCLP extraction.

## SUMMARY

The results of this study show that thermally treated coal tar-contaminated soil may be useful as a concrete aggregate. Geotechnical tests indicated that no significant changes in shear strength or grain size distribution occur for soils subjected to thermal treatment up to temperatures of 550°C. Decreases in concrete strength parameters associated with incorporation of contaminated and thermally treated aggregate were small, and in all cases, concrete strength remained within typical values for structural concrete. Treating aggregate at 250°C appeared to eliminate the strength changes associated with coal tar contamination, and at treatment temperatures as high as 650°C strength decreases did not exceed 10%. Leachability results suggest that incorporating coal tar-contaminated soil into concrete may stabilize PAHs with molecular weights less than or equal to 166 g/mol. However, additional work is needed to better characterize this stabilization and to quantify losses of lower molecular weight PAHs (e.g. due to volatilization) during TCLP extraction and from the concrete during mixing and curing. The relatively high leachability of PAHs with molecular weight greater than or equal to 166 g/mol may be a result of the leaching method used.

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