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Effects of Water Flow Rate and Temperature on Leaching From Creosote-Treated Wood

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Abstract

Creosote has a long history of use as a preservative, particularly in industrial wood products. However, its use has come under increasing scrutiny as a result of concerns about potential effects on aquatic and terrestrial non-target organisms. Despite the long use of creosote, there is relatively little data on the rates of creosote loss in many exposures, including aquatic applications. To address this concern, the Federal Highway Administration has funded a series of studies to evaluate the environmental impact of creosote-treated wood used in timber bridges. In the study reported here, we investigated the leaching of creosote from rough-sawn Douglas-fir lumber under simulated river flow conditions. Treated wood samples were contained in a metal tank, and deionized water was passed through the box at three predetermined flow rates and temperatures. The water was periodically sampled for the concentration of five major creosote components. The leach rates were highly variable, but in general they tended to increase with both flow rate and temperature. The possible onset of turbulent flow in the tank may have been responsible for the high leach rates observed at high flow rates. In general, the rates of leaching determined in this study were greater than those previously reported for creosote-treated piling. Longer exposures may be needed to better predict creosote release rates during decades of in-service use.

Keywords: Creosote, leaching, solid phase microextraction, acenaphthalene, dibenzoquinone, fluorene, fluoranthene, phenanthrene, pollution.

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Introduction

Creosote, one of the oldest wood preservatives, is a mixture of hundreds of compounds. It typically contains 85% polycyclic aromatic hydrocarbons (PAHs), 10% phenolics, and 5% N-, S-, and O-heterocyclics (Nestler 1974). Certain PAHs are known or suspected to be toxic, carcinogenic, or teratogenic, and have been listed as toxic substances by the Environmental Protection Agency (EPA) in the United States. Creosote has been extensively used to treat utility poles, railway crossties, posts, marine pilings, and bridge timbers, providing excellent protection to wood against fungal, insect, and marine borer attack.

Creosote may migrate into the environment through emission, water leaching, or direct soil contact from creosoted wood products in service (Ingram and others 1982, 1984; Behr and Baecker 1994; Bergqvist and Holmroos 1994; Gevao and Jones 1998; Kohler and others 2000). Despite the long history of industrial use, there is relatively little data on migration of PAHs from creosote into the surrounding environment, such as might occur from creosoted timber bridges. It is generally assumed that the quantities of PAHs entering the environment from treated wood in service are relatively small. The available evidence, however, is limited, inconclusive, and sometimes conflicting (Ingram and others 1982, Lebow and Morrell 1988). In comparing creosoted Southern Pine pilings exposed to marine borer attack in the sea for 9.5 years with unexposed treated wood, Lorenz and Gjovik (1972) found that creosote in exposed pilings had a composition similar to that of the original creosote, even for the lower boiling constituents. Miller (1972, 1977) examined three Douglas-fir pilings immersed in estuarine water of Yaquina Bay, Oregon, through 8 ensuing years. Creosote content in two of the three pilings remained constant, while the third piling lost as much as 20% creosote from the outer 1.25-cm zone after 8 years of exposure. In contrast, significant losses in the creosote component after various periods of marine exposure have also been reported (Stasse 1959, Stasse and Rodgers 1965, Gjovik 1977). The absence of reliable and systematic data makes it difficult to accurately assess or model the risk of creosote leaching to environments. With increasing environmental concerns from the public, it is likely that creosote, together with other wood preservatives, will be put under closer scrutiny by regulatory agencies. It is therefore critical to find a means for reliable determination of the suitability of using creosote.

The use of wood treated with chemical preservatives for bridge construction is widespread in the United States and Canada. One great concern has been the potential for migration of creosote components from treated wood exposed over or adjacent to surface waters, and its impact on non-target aquatic organisms (Brooks 1995). To address these concerns, the Federal Highway Administration (FHWA) has funded a series of research projects to evaluate the environmental impact of creosote-treated wood used in timber bridges. The focus of this study is to characterize the rate of release of creosote from submerged lumber. However, it is impractical to obtain data on creosote leaching from the timber in service in river conditions.

An alternative and more economic approach to direct quantitative analysis is to model the leaching of creosote components into the environment. Brooks (1997) developed mathematical models for assessing the environmental risks associated with the use of creosote-treated pilings in aquatic environments. The models were designed to provide a worst case assessment, and they might have overestimated sediment and water column concentrations of PAHs derived from creosoted wood by a factor of 1.2 to 2.0. The models were developed based primarily on data obtained by Ingram and others (1982) in a tank leaching study where samples were placed in seawater in a sealed stainless steel tank and stirred continuously. Ingram and others (1982) also observed different leaching rates at 20°C and 40°C with small wood samples immersed in still seawater in a 4-L beaker.

To use the models developed by Brooks for predicting creosote leaching in river conditions, a potentially important variable, water flow rate, should be incorporated in the models. In addition, the temperature range for river water can be much wider than that for seawater. To provide reliable information for modeling, we investigated the effects of water flow rate and temperature on creosote leaching.

Materials and Methods

Wood Samples

Douglas fir (*Psuedotsuga menziesii* (Mirb.) Franco) lumber (50 by 150 mm by 2.4 m long) was treated with P1/P13 creosote (AWPA 1999) to a target retention of 192 kg/m^3 using a full-cell process at a commercial treatment facility in Eugene, Oregon. The treatments were performed in accordance with the best management practices of the Western Wood-Preservers' Institute (WWPI) (WWPI 1996). Eight boards were cut into 300-mm-long sections, excluding the end 50 mm of each board, taking care to exclude knots. The samples were randomly mixed, wrapped with plastic, and stored indoors until needed. Before use, samples were end-sealed twice with epoxy resin to minimize end-grain effects.

Water

Deionized water was used for all the leaching tests. A bacterial control agent, BULAB 6001 (Buckman Laboratories Inc., Memphis, Tennessee), was added to the water at 40 ppb to limit the potential for biodegradation of the creosote components.

Gas Chromatography Analysis

Gas chromatography (GC) analysis was performed using modifications of procedures described by Langenfeld and others (1996). Solid-phase microextraction (SPME) was performed using 7-µm film thickness poly (dimethylsiloxane) fiber (Supelco Corp., Bellefonte, Pennsylvania). An aliquot of creosote-contaminated water (20 mL) was placed in a 20-mL screw-cap vial with a Teflon-lined septum. The vial septum was pierced with a needle. The needle, with the fiber withdrawn inside, was then pushed through the pierced opening. Once the needle penetrated the septum, the plunger on the fiber holder was depressed to expose the fiber to the water sample for 30 min, while the water was continuously agitated by a magnetic stirrer. Once the extraction was completed, the fiber was withdrawn back inside the fiber holder, removed from the water sample, and analyzed immediately.

A Hewlett–Packard 5890 series II gas chromatograph (Hewlett–Packard Corporation, Palo Alto, California) equipped with a splitless injection port and a flame ionization detector (FID) was used to analyze the extracted components in the fiber. The injector and detector were maintained at 300°C and 330°C, respectively. The column temperature program was 60°C for 3 min, ramped at 10ºC/min to 320ºC, and held at 320ºC for 1 min. The column was a 30-m DB-5 (0.25 mm I.D., 0.25 µm film thickness) capillary column (Supelco Corp., Bellefonte, Pennsylvania). A stock standard containing naphthalene (4 mg/mL), 2-methyl naphthalene (1 mg/mL), 1-methyl naphthalene (1 mg/mL), acenaphthene (1 mg/mL), dibenzofuran (0.5 mg/mL), fluorine (0.3 mg/mL), phenanthrene (0.3 mg/mL), and fluoranthene (0.1 mg/mL) was prepared in methanol for standard curve calibrations. Concentrations of the eight chemicals were adjusted to ensure the linearity for each chemical within the tested concentration ranges. A series of standard solutions were prepared by spiking 1, 2, 4, 10, 15, and 20 µL of stock standard into 20 mL deionized water.

Whole Creosote Baseline Sample

A whole creosote sample was taken from the treating plant prior to treatment. A stock solution was prepared by dissolving 0.1 g in 10 mL methylene chloride (10 mg/mL). Four concentrations of this stock (2, 5, 10, and 20 µL) were spiked into 20 mL deionized water and analyzed by SPME GC. Concentrations of chemicals were determined by the best dilutions that gave peaks within the standard curve ranges.

Tank Leaching System

The tank leaching system contained a 59-L plastic water tank and an aluminum sample box (Fig. 1). The water temperature in the tank was controlled by a thermostat connected to both a heater and a chiller. Hot or cold water was pumped through a 15.2-m copper tubing (0.9-cm I.D.) coil to maintain a tank temperature of 5°C, 20°C, or 35°C. A layer of vinyl foam weatherstripping was used along the edge of the cover to minimize evaporation. The tank was wrapped with reflective foil insulation to reduce radiative heat loss.

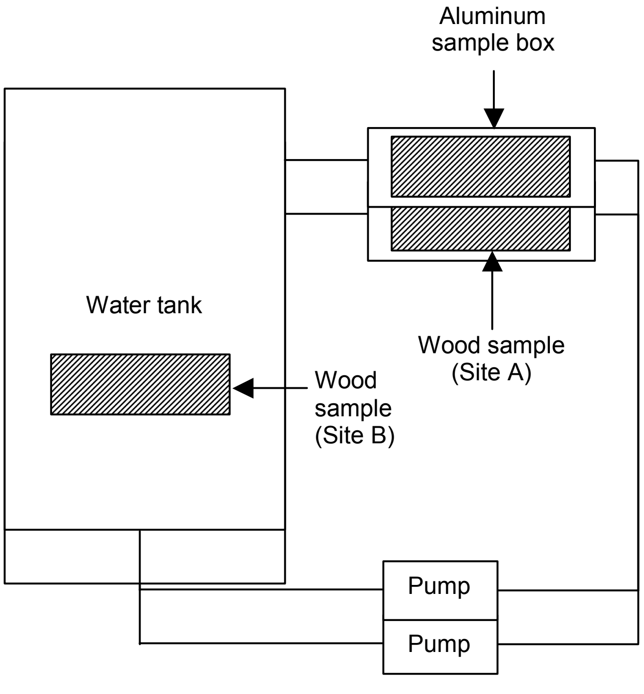

Figure 1—Tank leaching apparatus. Wood sample is placed in water tank for trials with no flow (site A) and in aluminum sample box for trials at non-zero flow rates (site B).

Three water flow rates were tested: 0, 4, and 8 cm/s. For testing leaching at zero flow rate, the wood sample was kept immersed in water using a plastic tube supporter and a lead weight. For testing leaching at 4 and 8 cm/s flow rate, the wood sample was kept in the middle of the aluminum sample box by a few aluminum supporters glued to the walls. The cover was tightly screwed to the box with a layer of rubber gasket in-between. An air outlet valve was installed at the end of sample box to ensure that the sample box was always full of water. Aliquots of 60 mL of leachate water were removed at 0, 1, 2, 4, 8, and 24 h after the creosoted sample was immersed in the system. After 72 h, the leach water was drained and the tank refilled with fresh deionized water. Twenty-five liters of water was used for each run, and two or three runs were performed for each sample. The collected leachate was filtered through 2-µm glass filter paper prior to analysis by SPME GC. Most samples were analyzed on the same day when they were taken; some samples taken at 24 h were analyzed after 1 or 2 days.

While this experimental apparatus allows for the investigation of leaching at high flow rates, it also presents some limitations. Since the water is recycled through the leaching apparatus, the concentration of leachate increases with time in the circulating solution. This may give rise to interaction factors not present in the field, since the leachate is not diffusing into an effectively infinite volume, as it might be in a river. The concentrations in the apparatus were observed to level off with time at levels well below their solubility limits. In an ideal situation, the levels should be determined by the distribution coefficients for the individual components between the aqueous and organic phases. To estimate the worst case scenario for modeling purposes, this experimental design thus required modeling the concentration of leachate with time and extrapolating to find the leach rate at zero time. It was then assumed that the leach rate at zero time was equal to the leach rate at infinite dilution.

The apparatus also allows the study of leaching at high flow rates without a wasteful consumption of water. However, to keep the pump size manageable, the annular region between the wood sample and the walls of the sample container was kept at 12.5 cm. This small clearance gave rise to turbulent flow at the high (8-cm/s) flow rate, which produced a large effect on the leach rate (see Results).

Experimental Design and Statistical Analyses

Experimental design was done with the aid of Design-Expert 6 (Stat-Ease, Inc., Minneapolis, Minnesota). Three-level factorial design was applied with the center point replicated. Specifically, there were two factors, water flow rate and temperature. Each factor had three test levels that were evenly spaced (flow rate 0, 4, and 8 cm/s; temperature 5°C, 20°C, and 35°C). Typical leaching patterns were found to be in hyperbolic forms when plotted as concentration as function of leaching time. A hyperbolic function, $Y = aX/(b + X)$, where Y is chemical concentration and X leaching time, was used to model the data using SigmaPlot 2000 for Windows Version 6 (SPCC Inc., Chicago, Illinois). The leaching rate at the starting point was found to be a/b by taking derivative dY/dX at $X = 0$. Multiple linear regression (MLR) was applied to the tank leaching data to investigate the effects of water flow rate and temperature on creosote leaching using Design-Expert 6.

Results and Discussion

Whole Creosote

Eight PAH compounds, which constituted 40.54% of whole creosote, were used as references to assess creosote migration into the water (Table 1). These PAHs were selected because they are major components of whole creosote and they encompass a range of chemical structures and volatility.

Tank Leaching

Results of initial studies showed high variations in three low-boiling fractions: naphthalene, 2-methyl naphthalene, and 1-methyl naphthalene (Xiao and others 1999). These compounds were either not detectable or presented at highly variable concentrations. This variability was likely due to the relatively high vapor pressure of these components, which allowed them to volatilize from the leachate water. These three compounds were excluded from further analysis. The remaining five compounds constituted 29.23% of the whole creosote, still providing a good representation of medium- and high-boiling fractions.

A typical leaching pattern consisted of a steep increase in the first few hours, followed by a slower increase and finally a plateau (Fig. 2). Wood exposed to river water is surrounded

Table 1—Percentage of selected chemicals in whole creosote

Creosote component	Proportion of whole creosote (%)	95% confidence interval (%)
Naphthalene	7.35	4.8–9.7
2-methyl naphthalene	2.35	1.6–3.1
1-methyl naphthalene	1.41	0.5–2.2
Acenaphthene	5.87	4.99–7.23
Dibenzofuran	3.64	3.15–4.15
Fluorene	5.42	4.52–6.44
Phenanthrene	11.44	8.4–12.7
Fluoranthene	3.06	2.44–4.4
Total	40.54	—

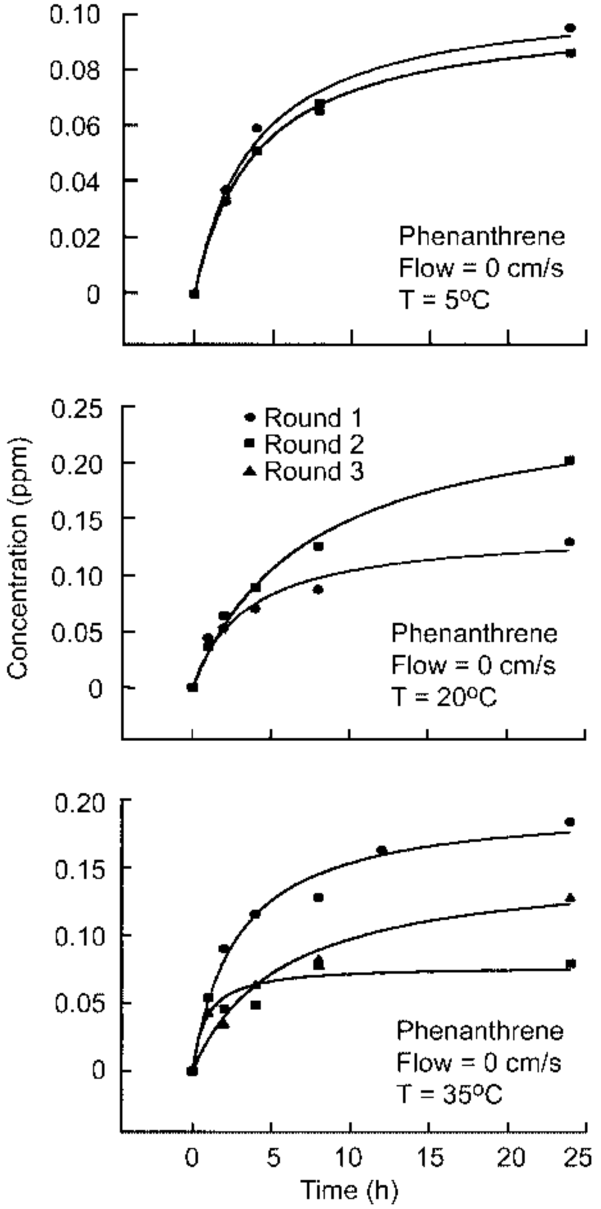

Figure 2—Phenanthrene concentrations in leachate over time.

constantly by fresh water. Therefore, the leaching rate at the start point of each run best represented an actual leaching situation. The hyperbolic model ($Y = aX/(b + X)$) was found to fit the data satisfactorily, yielding an average adjusted R^2 of 0.94. The leaching rate at the start point equals the initial slope, which is the derivative dY/dX at $X = 0$. Leaching rates from two runs for each trial were averaged, and the value was used as the leaching rate for that trial. Although for some trials the difference between the runs might be significant (Xiao and others 1999), averaging between the two leaching rates may help to reduce the error due to the high variations of wood material.

The leaching rates at 8 cm/s were dramatically different than those obtained at 0 and 4 cm/s (Table 2), showing higher variation and unpredictability. This observation led us back to some basics for water flow and to the retrospective discovery of turbulent flow at 8 cm/s other than the assumed laminar flow. Sometimes the sample box was only partially filled with water at 8 cm/s flow, which may have been due to the insufficient power of the water pump to maintain this flow rate. This might have contributed to the observed high variation in leaching rate.

Transition to Turbulent Flow

The extreme increase in leaching at the 8-cm/s flow led us to consider the possibility that the flow in the leaching apparatus had transitioned to the turbulent regime. The Reynolds number (N_{Re}) was calculated from

$$N_{\mathrm{Re}} = \frac{Lv\rho}{\mu}$$

where

L is characteristic length,

v fluid velocity,

ρ fluid density, and

μ fluid viscosity.

Using standard reference values for the density and viscosity of water, and defining the characteristic length as

$$L = \frac{4(a_w \cdot b_w - a_s \cdot b_s)}{2(a_w + b_w + a_s + b_s)}$$

$N_{\mathrm{Re}} = 254v$. Thus for $v = 4$, $N_{\mathrm{Re}} = 1016$, and for $v = 8$, $N_{\mathrm{Re}} = 2032$. The transition to turbulent flow is generally regarded to commence around $N_{\mathrm{Re}} = 2000$. Thus, it is reasonable to conclude that turbulent flow in the leach apparatus is at least partially responsible for the large increase in leach rates observed at the 8-cm/s flow rate.

The leaching rates for all five creosote components, which constituted about 29% of the whole creosote, are summed in Table 2. It cannot be assumed that these leaching rates are representative of creosote as a whole, because components differ in solubility and volatility.

The most thorough study of creosote in the literature is probably that of Ingram and others. (1982). This study reported concentrations in the leach water of the five components studied here ranging from 0.97 to 1.67 lb/min. This is comparable to the results of our study, with the exception of

Table 2—Leach rates for five components of creosote-treated lumber at various flow rates and temperatures

Temperature (°C)	Water flow rate (cm/s)	Leach rate (μg-cm^{-1}-day^{-1})					
		Acenaphthene	Dibenzofuran	Fluoranthene	Fluorene	Phenanthrene	Sum for five components
5	0	27.3	17.1	26.0	8.0	14.7	93.2
20	0	39.9	17.2	13.6	8.3	20.8	99.8
35	0	55.4	12.7	16.2	7.1	34.0	125.4
5	4	149.7	150.3	72.5	59.8	140.6	572.9
20	4	145.6	178.8	57.7	77.5	182.7	727.8
35	4	236.8	584.2	35.0	110.2	529.8	1,496.0
5	8	249.5	147.8	81.9	127.9	392.5	999.6
20	8	1,212.8	191.0	99.1	520.9	538.6	2,562.4
35	8	828.5	566.9	171.1	758.4	1,267.7	3,592.6

the high flow and temperature region. However, the leach rates at infinite dilution ($t = 0$) were very high and are not sustainable. For example, the sum of the leaching rates for the five components at 20°C and 4-cm/s flow is 728 ug/cm^2-day. At the 192-kg/m^3 target retention, the average amount of whole creosote in the sample is 192 mg/cm^3. If one ignores diffusion rates, evaporation, retention, distribution, and similar mitigating variables, and assumes a leach rate of whole creosote of 1 mg/cm^2-day, it appears that all the creosote would be lost from the outer centimeter of wood in only 192 days. Yet, creosote treatments last for decades; most of the creosote remains in the wood. Thus, the initial leach rates observed here must decrease with time, but on a longer scale than the experimental times used in this study.

Conclusions

Acenapthene, dibenzofuran, fluoranthrene, fluorine, and phenanthrene move from creosote-treated wood into fresh water at rates influenced by both temperature and flow rate of the water. In a fixed volume of water, the concentration of these components approaches an equilibrium state of constant concentration. The increase in concentration of each component with exposure time was empirically modeled using a hyperbolic equation. Leach rate at infinite dilution was then derived from the model. The highest leach rates generally occurred at the highest temperature and flow rate, although it appears that turbulence at the highest flow rate may have greatly accelerated leaching. It is apparent that the leaching rates determined in this study are greater than those that occur over very long exposures. Longer leach times may be needed to more accurately assess the rate of release from creosote-treated wood after many years in service.

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