Efficacy of Pentachlorophenol in Biodiesel versus Diesel

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Abstract

Pentachlorophenol (PCP) is widely used as a wood preservative for utility poles and other wood products. It has been proposed that a modified PCP carrier system based on a diesel/biodiesel mixture should be used in place of the conventional diesel/KB3 carrier, but questions exist as to whether this modified carrier system can provide the same service life as wood products treated with PCP/diesel/KB3. The main objective of this research was to evaluate the comparative decay resistance of wood treated with carrier formulations containing either diesel/KB3 or diesel/biodiesel. A 2-year efficacy study using an accelerated soil contact decay test was initiated to compare the performance of southern yellow pine wood treated with the conventional diesel/KB3 carrier and a modified diesel/biodiesel carrier, both with and without PCP. The residual hydrocarbon levels, PCP reduction, toxicity, and leaching of PCP of the samples remained approximately at the same level for treatments with similar PCP retention values for both carriers. For wood treated with PCP in these two different carriers, there was no evidence of differences in the average modulus of elasticity. Overall, this study recommends long-term field stake tests to determine the practical significance of these results.

Preservative treatments can extend the life of wood products by 20 to 40 times compared with untreated wood (Morrell 2004). Pentachlorophenol (PCP) is a major wood preservative used in the United States and is effective against nearly all wood deteriorating organisms, including termites, beetles, fungi, and bacteria (Carswell 1939). There are more than 160 million utility poles in service in North America. Currently, PCP is used for treatment of 50 percent of utility poles in the United States. A survey in the western United States found that 800,000 utility poles are disposed of each year (Morrell 2004). Although these materials are not considered a hazardous waste, all wood preservatives have a level of toxicity and can present a challenge with environmental remediation (Chu and Kirsch 1972, Prewitt et al. 2003, Kao et al. 2005).

Petroleum-derived hydrocarbons such as diesels are used as PCP carriers for wood treatment. These carriers play a major role in the performance of treated wood products. The efficacy of carriers and their interaction with preservatives has been investigated by a number of authors (Gjovik and Gutzmer 1985, Nicholas 1988, Barnes et al. 2006). Studies indicate that efficacy is attributed partly to the characteristics of the carriers as well as to the role of these carriers in the depletion rate or movement of active chemicals (Vaughan 1947, Duncan 1957, Arsenault 1970, Arsenault et al. 1984, Nicholas 1988).

Petroleum-derived hydrocarbons can also be a problem in the disposal of wood wastes. Current interest is focused on the use of biodiesel as an alternative or additive to petroleum diesel or other petroleum-based products.

Previous studies have demonstrated that microorganisms are capable of degrading petroleum-based hydrocarbons (Mellor et al. 1996); therefore, bioremediation could be used for cleanup of the contaminated sites. Also, lower toxicity and faster biodegradability for non–petroleum-based diesels was observed (Miller and Mudge 1997, Zhang et al. 1998, Bonten et al. 1999, Mudge and Pereira 1999, Taylor and Jones 2001, Schleicher et al. 2009, KeshaniLangroodi et al. 2011, Langroodi et al. 2012). These properties could be advantageous for application in the wood-preservation industry because biodiesel could have less secondary impact on the environment. On the other hand, because the efficacy of wood preservatives is affected by the carriers, PCP formulated with biodiesel could have a different performance in service compared with conventional PCP

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treatment systems. Currently, there is limited published information on the possible effect of biodiesel on the efficacy of the treated wood and biodegradability of PCP and its carriers (Morrell and Freitag 2010).

The objective of this study was to evaluate the efficacy of PCP using different ratios of petroleum diesel/KB3 and biodiesel on southern yellow pine.

Methods

Sample preparation

In this experiment, 110 test decay sticks measuring 3 by 14 by 200 mm were cut from southern yellow pine sapwood. PCP is mostly used on Douglas-fir and chromated copper arsenate (CCA) on southern yellow pine, but southern yellow pine was used due to availability and for being a dominant species in Mississippi. These sticks were treated with fossil fuel diesel (Shell Company of Australia), KB3 (a ketone still-bottom petroleum by-product that is used as a cosolvent to enhance solubility and effectiveness of PCP), and biodiesel (BioPreserve Company, Erie, Pennsylvania) in different ratios with and without selected PCP concentrations. The number of replicates, treatment formulations, and PCP retention values are given in Table 1. It should be noted here that different types of biodiesel are produced with different properties that may impact the efficiency of treated wood. These chemicals were impregnated into the sticks by full cell process. Ten untreated sticks were kept as controls. A 50-mm section from each end was cut and reserved for further analysis.

For evaluation of the wood preservatives in soil contact, standard method AWPA E23-07 (American Wood Protection Association [AWPA] 2008) was used. Soil used in this experiment was collected from an undisturbed forested area. The soil was spread on a plastic liner and air dried under a fume hood for 24 hours and sieved through a number 6 mesh screen (3.35 mm) to separate clods, rocks, and other debris.

Two sticks were placed in each plastic cup, covered with soil, and placed in chambers set at 43 percent relative humidity at 28°C for 2 years. Experimental units were monitored weekly by adding deionized water to maintain an approximate 20 percent soil moisture content and a 40 to 60 percent moisture content of the wood sticks. Sampling and test schedules for this study are shown in Table 2.

Bending test

All samples were fully saturated with deionized water by a vacuum process prior to determining the initial modulus of elasticity (MOE) with the bending test apparatus by deflecting them 1.75 mm (AWPA 2008). Bending tests were run three times to establish an average for unexposed MOE value (AWPA 2008). Samples were removed from the cups bimonthly, wiped to remove excess soil, and submerged in deionized water for 1 hour for the MOE test. To prevent interchange between chemicals of different treatments, each wood stick within each treatment group was placed in a different plastic bag. The MOE test is a nondestructive test, and afterward the wood sticks were returned to the plastic cups for the next sampling period.

Methylene chloride extraction process

On day 0 and after 2 years, a composite wood sample (0.5 g) was made from ground wood from the center of each wood stick in the plastic cups. The composite wood sample (1 g) was then placed into a cellulose extraction thimble and extracted using 200 mL methylene chloride by soxhlet extraction according to US Environmental Protection Agency (US EPA) method 3540 (Brilis and Marsden 1990). The extracts were condensed to 3 mL and transferred to test tubes for further analysis.

Oil and grease concentration

Oil and grease concentration was determined using a modified Standard Method 5520-F (Clesceri et al. 1998). Methylene chloride extracts (2 mL) from above were put in preweighed 50-mL flasks and placed in a fume hood overnight to evaporate the methylene chloride. The difference between initial weight and final weight was calculated as the amount of the oil and grease.

Determination of PCP concentration

Condensed extract (500 μL) was placed in a 2-mL sample vial, and then 100 μL of *N,O*-bis(trimethylsilyl)trifluoroacetamide was added; the vial was left at room temperature for 2 hours. Hexane (400 μL) was then added to make the final volume. PCP concentration was determined according to EPA Method 8041 (US EPA 2007) using an Agilent 6890 gas chromatograph equipped with a ^{63}Ni electron capture detector and an Agilent Ultra II capillary column. The injector temperature was 250°C, the column temperature

Table 1.—Identification and treatments of wooden sticks for efficacy evaluation.[a]

	Components			
Sample no.	Stock solution (g)	Toluene (g)	PCP (%)	Target PCP retention (pcf)
1–10	A, 90:10	—	—	Solvent control
11–20	B, 70:30	—	—	Solvent control
21–30	A, 60	240	5	0.3
31–40	A, 40	260	5	0.2
41–50	A, 20	280	5	0.1
51–60	A, 10	290	5	0.05
61–70	B, 60	240	5	0.3
71–80	B, 40	260	5	0.2
81–90	B, 20	280	5	0.1
91–100	B, 10	290	5	0.05
101–110	—	—	—	Control

[a] Stock solution A = 90 g diesel and 10 g KB3 in 500 g toluene; stock solution B = 70 g diesel and 30 g biodiesel 500 g toluene; PCP = pentachlorophenol.

Table 2.—Sampling and test schedule for efficacy evaluation experiment.

Test	Testing days
Wood pentachlorophenol determination	Day 0 and final
Oil and grease	Day 0 and final
Soil microorganisms' population	Every 3 mo
Wood toxicity	Day 0 and final
Wood toxicity characteristics leaching procedure	Day 0 and final
Wood bending test (strength loss)	Every 2 mo

was 175°C, and the detector temperature was 315°C. The helium flow rate through the detector was 1.5 mL/min.

Microorganism population

To enumerate bacteria and fungi in the soil surrounding the samples, the serial dilution plate technique was used. Three grams of soil was taken from three different points around the sticks in each decay chamber unit and thoroughly mixed. One gram of soil from each of these samples was added to 9 mL of sterile deionized water and mixed for further dilutions. Samples from selected dilutions (250 μL) of the microbial suspension were spread on duplicate plates of nutrient agar for bacteria, nutrient agar amended with 5 ppm of PCP for PCP tolerant bacteria, and potato dextrose agar with antibiotics for fungi. Appropriate dilutions were made as needed. The plates were incubated for 2 to 4 days at 28°C, and colonies on each plate were counted and averaged.

Toxicity

Microtox was used to measure the toxicity of wood samples. The toxicity analyzer measures the concentration of toxic substance needed to decrease the light output of a luminescent bacteria, *Photobacterium phosphoreum,* by 50 percent (effective concentration, EC_{50}). Chopped wood (0.5 g) obtained from the center of the exposed and unexposed wood sticks was added to 9 mL of sterile water, centrifuged, its pH adjusted to 6.0 to 7.0, and then was assayed by the Abbreviated Assay Procedure found in the Beckman Instruments Instructions No. 015-555879 with a 15-minute incubation period at 15°C.

Toxicity characteristics leaching procedure

The leaching characteristics of the wood were determined using a modified version of the EPA toxicity characteristics leaching procedure (TCLP; US EPA 1986). This test was performed at the beginning and the end of the experiment. EPA method 8041 was used for evaluating the PCP leached levels by gas chromatography described in the ''PCP concentration'' section.

Statistical analysis

The mechanical and chemical results from this experiment were statistically analyzed by using a completely random design with 10 replications for mechanical test for each treatment and five replications for chemical analyses for each treatment. One-way analysis of variance and Tukey's multiple comparisons test were used to compare treatment mean differences at $P = 0.05$. Data were processed by Statistical Package for the Social Sciences (SPSS), Predictive Analytic Software (PASW) version 18.

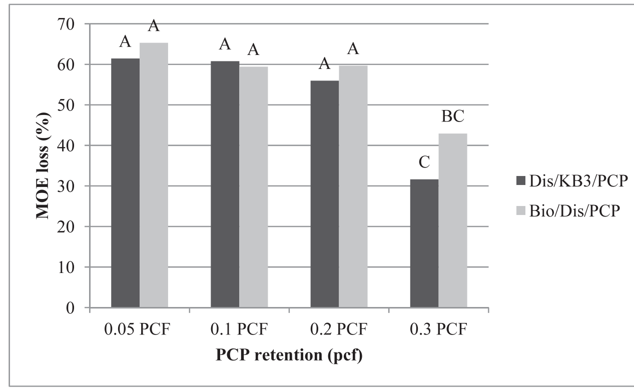

Figure 1.—Modulus of elasticity (MOE) losses for pentachlorophenol (PCP) treatments using diesel/KB3 and diesel/biodiesel formulations after 24 months of exposure. Note: Different letters indicate significant differences between treatments.

Results

Bending test

There were no significant differences in MOE loss between treatments containing biodiesel/diesel and diesel/KB3 with the same PCP concentration (Fig. 1). However, in treatments without PCP, there was a trend showing that over time MOE loss in biodiesel/diesel systems was higher than in diesel/KB3 systems. This can be observed by comparing results from sticks treated with biodiesel/diesel and diesel/KB3 for years 1 through 2 of this study (Fig. 2). Results show that for the first year, differences in MOE loss between wood sticks treated with biodiesel/diesel and diesel/KB3 were not significant; however, by 18 and 24 months, the differences in MOE loss between biodiesel/diesel- and diesel/KB3-treated sticks were significant (Fig. 2). The intrinsic decay resistance characteristics of the diesel/KB3 carrier compared with the biodiesel/diesel carrier over time should be considered the main reasons. However, there was significantly less loss in MOE in sticks containing diesel/KB3 alone compared with biodiesel/diesel alone (Fig. 2).

Oil and grease concentration

There was a large decrease in the oil and grease concentration by the end of this study (Figs. 3 and 4).

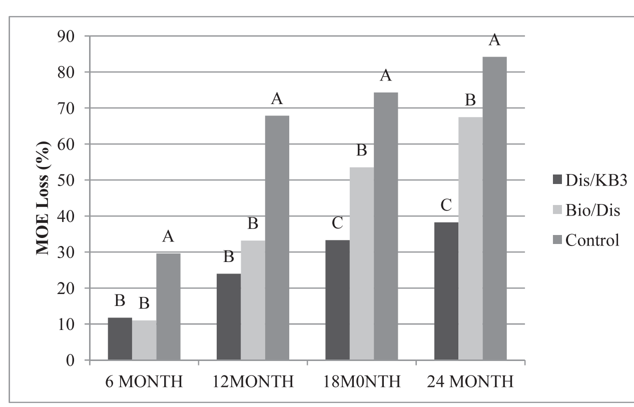

Figure 2.—Modulus of elasticity (MOE) losses for untreated control, diesel/KB3 alone, and diesel/biodiesel alone treatments at four exposure periods. Note: Different letters indicate significant differences within treatments for a given time period.

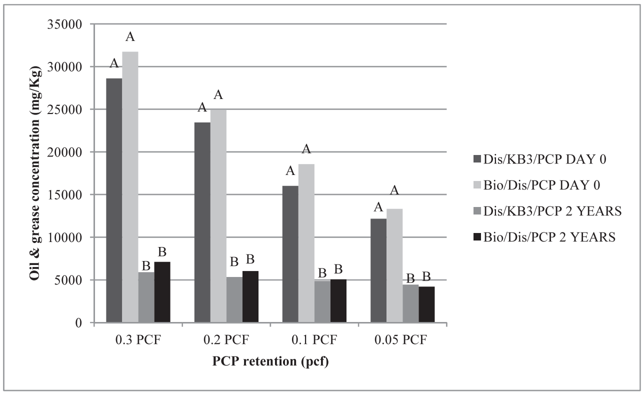

Figure 3.—Oil and grease concentration for treatments containing pentachlorophenol (PCP) on day 0 and at 2 years. Note: Different letters indicate significant differences within a given treatment.

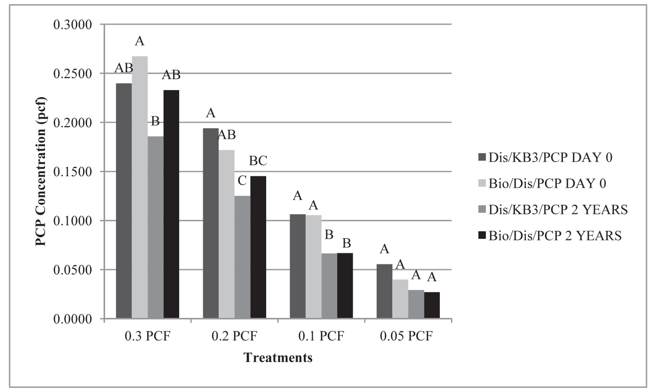

Figure 5.—Pentachlorophenol (PCP) retention in wooden sticks for treatments containing PCP on day 0 and at 2 years. Note: Different letters indicate significant differences within a given treatment.

Because biodiesel/diesel and diesel/KB3 were not diluted at the time of treatment as the treatments containing PCP were, they showed higher initial concentrations of oil and grease. There were no significant differences between treatments with the same PCP retention values in oil and grease after 2 years (Fig. 3).

The percent reduction of oil and grease concentration showed that there were no significant differences in oil and grease concentration for treatments containing PCP (Fig. 4). There were also no significant differences observed in oil and grease reduction in treatments containing biodiesel/diesel alone and diesel/KB3 alone after 2 years (Fig. 4).

PCP concentration

Changes in PCP concentration are shown in Figures 5 and 6. There were no significant differences between day 0 samples and the final samples taken after 2 years in PCP retention for wooden sticks treated with biodiesel/diesel/PCP except for samples treated with PCP at 0.1 per cubic foot (pcf) and diesel/KB3/PCP with PCP at 0.2 pcf (Fig. 5).

There were no significant differences in PCP reduction between treatments with the same PCP retention values for biodiesel/diesel and diesel/KB3 carriers (Fig. 6). These results support results of Morgan (2010), which showed that PCP depletion rates had better fixation/reaction, or more water repellency in treatments using biodiesel as cosolvent for PCP compared with petroleum-based carriers. The reason for this behavior could be better penetration of PCP in biodiesel/diesel systems.

Microorganism populations

In general the number of microorganisms (bacteria and fungi) decreased during this study, but there were still sufficient numbers of microorganisms present in the soil to cause degradation of the wood (Tables 3 through 5). There were no significant differences in populations between treatments with the same PCP retention values for the biodiesel/diesel carrier compared with the diesel/KB3 carrier. As expected, control treatments had the highest number of fungal colonies by the end of this experiment (Table 5). The observed decrease in the number of microorganisms could possibly be attributed to unfavorable effects of diesel and PCP from the wood.

Toxicity

At the end of this study (2 y) there were significant differences in relative toxicity between treatments (Fig. 7). Although there were no significant differences between treatments with the same PCP retention values, there was a trend toward lower toxicity in treatments containing

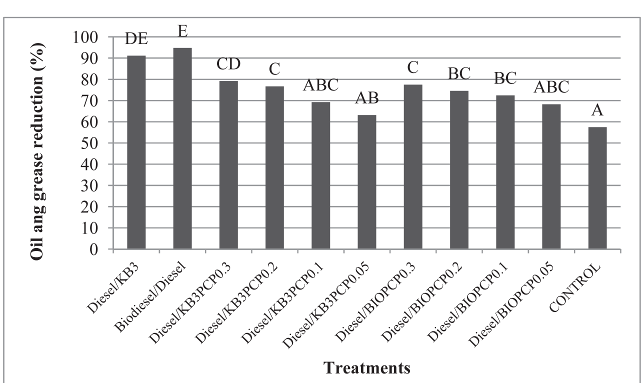

Figure 4.—Percent oil and grease reduction rate in wooden sticks during 2 years. Note: Different letters indicate significant differences between treatments.

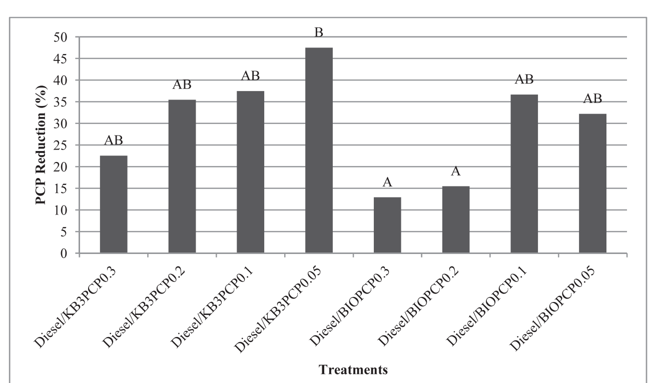

Figure 6.—Percent pentachlorophenol (PCP) reduction rate in wooden sticks during 2 years of exposure. Note: Different letters indicate significant differences between treatments.

Table 3.—Pentachlorophenol (PCP)–acclimated bacteria populations in treatments over time.

Treatments	PCP-resistant bacteria (CFU/mL)				
	Initial	6 mo	12 mo	18 mo	24 mo
Diesel/KB3/PCP 0.05	8.40E+07	7.20E+06	1.22E+05	2.72E+03	2.48E+03
Diesel/KB3/PCP 0.1	7.92E+08	6.40E+06	6.08E+04	2.96E+03	1.40E+03
Diesel/KB3/PCP 0.2	3.74E+08	8.00E+06	1.14E+05	8.00E+02	1.76E+03
Diesel/KB3/PCP 0.3	8.32E+08	9.60E+06	8.24E+04	2.40E+03	1.04E+03
Biodiesel/diesel/PCP 0.05	5.84E+08	6.40E+06	2.62E+05	3.28E+03	1.92E+03
Biodiesel/diesel/PCP 0.1	2.40E+08	9.60E+06	4.76E+05	2.56E+03	1.36E+03
Biodiesel/diesel/PCP 0.2	5.98E+08	7.20E+06	1.76E+04	1.28E+03	1.76E+03
Biodiesel/diesel/PCP 0.3	5.00E+08	5.60E+06	3.28E+04	1.52E+03	3.60E+02

Table 4.—Bacteria populations in treatments over time.

Treatments	PCP-resistant bacteria (CFU/mL)				
	Initial	6 mo	12 mo	18 mo	24 mo
Diesel/KB3	4.34E+07	9.60E+06	1.96E+05	1.90E+05	1.44E+03
Biodiesel/diesel	1.36E+07	8.80E+06	6.04E+05	1.00E+05	1.96E+03
Diesel/KB3/PCP 0.05[a]	1.72E+07	6.40E+06	1.26E+06	9.60E+04	6.40E+02
Diesel/KB3/PCP 0.1	5.36E+07	6.40E+06	5.64E+05	1.70E+05	2.40E+03
Diesel/KB3/PCP 0.2	1.12E+07	8.00E+06	2.68E+06	8.80E+04	1.52E+03
Diesel/KB3/PCP 0.3	2.96E+07	9.60E+06	1.56E+06	9.60E+04	1.12E+03
Biodiesel/diesel/PCP 0.05	2.00E+07	8.80E+06	8.00E+05	1.44E+05	4.40E+03
Biodiesel/diesel/PCP 0.1	1.76E+07	7.20E+06	4.12E+06	1.20E+05	5.32E+03
Biodiesel/diesel/PCP 0.2	2.32E+07	7.20E+06	3.04E+05	1.52E+05	5.20E+02
Biodiesel/diesel/PCP 0.3	2.48E+07	4.80E+06	2.72E+05	1.44E+05	1.60E+03
Control	3.28E+07	8.00E+06	1.92E+05	8.80E+04	1.24E+03

[a] PCP = pentachlorophenol.

Table 5.—Fungal populations in treatments over time.

Treatments	PCP-resistant bacteria (CFU/mL)				
	Initial	6 mo	12 mo	18 mo	24 mo
Diesel/KB3	3.46E+06	5.60E+03	1.36E+04	3.02E+04	5.44E+03
Biodiesel/diesel	1.74E+07	6.40E+03	1.58E+04	2.49E+04	4.64E+03
Diesel/KB3/PCP 0.05[a]	8.98E+06	9.60E+03	1.52E+04	3.76E+04	4.80E+03
Diesel/KB3/PCP 0.1	1.46E+06	1.04E+04	1.68E+04	2.54E+04	6.96E+03
Diesel/KB3/PCP 0.2	1.94E+06	9.60E+03	8.40E+03	3.58E+04	6.88E+03
Diesel/KB3/PCP 0.3	2.24E+06	2.64E+04	1.12E+04	2.49E+04	5.12E+03
Biodiesel/diesel/PCP 0.05	2.84E+06	4.00E+03	1.34E+04	2.76E+04	4.80E+03
Biodiesel/diesel/PCP 0.1	4.32E+06	6.40E+03	1.76E+04	2.73E+04	6.00E+02
Biodiesel/diesel/PCP 0.2	2.30E+06	1.20E+04	5.60E+03	2.22E+04	6.16E+03
Biodiesel/diesel/PCP 0.3	2.86E+06	8.80E+03	5.60E+03	2.66E+04	4.80E+03
Control	2.40E+06	1.28E+04	1.04E+04	1.82E+04	3.76E+04

[a] PCP = pentachlorophenol.

biodiesel/diesel than in treatments containing diesel/KB3 (Fig. 7). The reason for this could be the higher intrinsic toxicity of the diesel/KB3 carrier. The results also show that relative toxicity decreased over time in samples containing biodiesel/diesel, diesel/KB3, and controls (Fig. 8). At the end of 2 years, diesel/KB3 contained significantly higher toxicity compared with the control soil.

Toxicity characteristic leaching potential

The results of TCLP are shown in Figures 9 and 10. There was significantly reduced leaching of PCP between day 0 and 2 years except for diesel/KB3/PCP (0.1 pcf) and biodiesel/diesel/PCP (0.3 pcf; Fig. 9). Higher leaching of PCP in day 0 samples is mainly due to surface PCP leaching from the newly treated wood. The wood sticks were treated with PCP in different carriers only a few days before the start of this study; therefore, it is reasonable to assume there was higher PCP content not fixed into the available wood surface versus the 2-year-old samples. In general, a reduction in PCP leaching was proportional to the PCP concentration; the higher the concentration, the higher the PCP leaching (Fig. 9). A comparison of treatments at the end of the study found no statistical differences when comparing diesel/KB3 with biodiesel/diesel at the same concentration of PCP (Fig. 10). TCLP values for biodiesel/diesel and diesel/KB3 were far below the US EPA permissible limit for treated wood products, which is 100 mg/kg. This finding is generally in agreement with Morgan

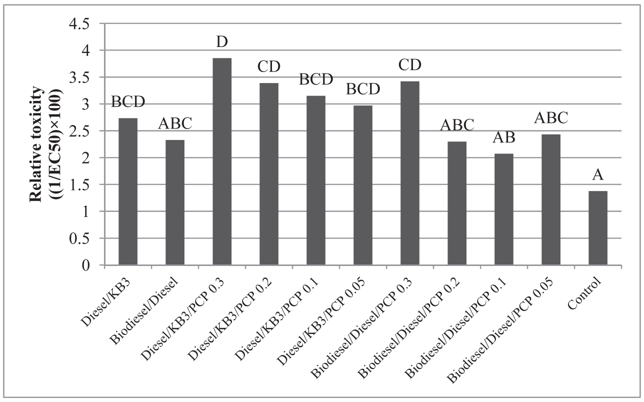

Figure 7.—Toxicity differences between treatments after 2 years of exposure. Note: Different letters indicate significant differences between treatments.

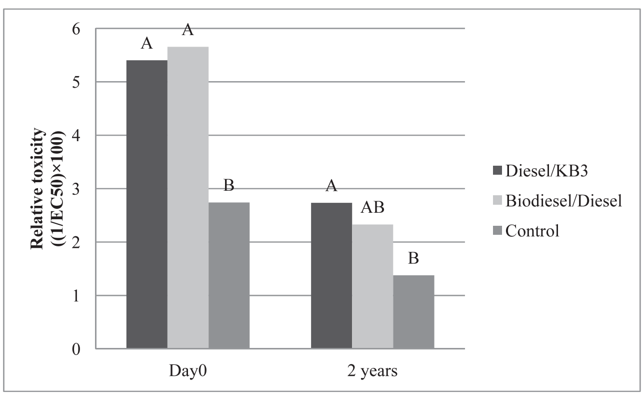

Figure 8.—Toxicity differences between treatments on day 0 and at 2 years. Note: Different letters indicate significant differences within a given time.

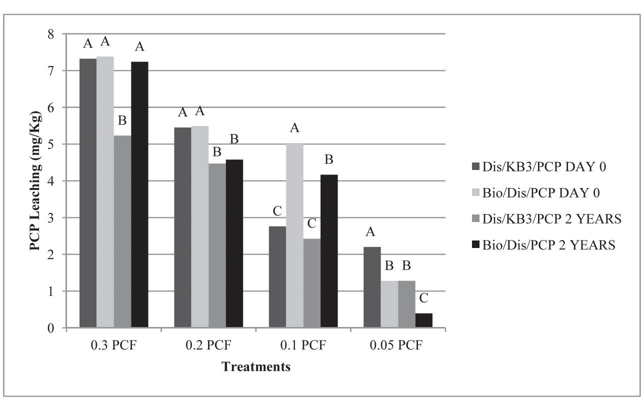

Figure 9.—Pentachlorophenol (PCP) concentrations from leaching (toxicity characteristics leaching procedure) for treatments on day 0 and at 2 years. Note: Different letters indicate significant differences within a given treatment.

(2010), indicating that the PCP leaching values were the same for biodiesel/diesel systems compared with conventional petroleum-based carrier systems.

Conclusions

Although samples treated with the PCP-free biodiesel carrier experienced greater MOE losses than the PCP-free,

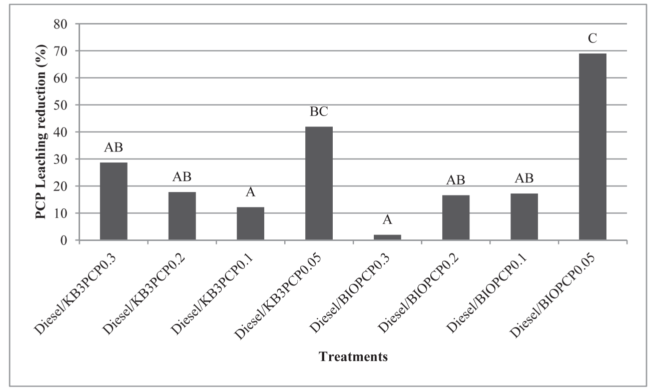

Figure 10.—Percent differences in pentachlorophenol (PCP) leaching among treatments after 2 years of exposure. Note: Different letters indicate significant differences between treatments.

petroleum diesel carrier–treated samples, there was no statistically significant difference in the average MOE loss for wood samples treated with PCP in either of these carriers. Overall this study suggests that PCP formulated in a biodiesel/diesel carrier is not as effective as the conventional diesel/KB3 formulation against wood decay fungi. However, owing to production of different biodiesels from various sources that could affect the efficacy of PCP-treated wood, additional long-term field studies are needed before the potential impact of biodiesel formulations on the long-term performance of PCP-treated wood can be fully assessed.

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