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Influences of wood preservation, lumber size, and weather on field leaching of red pine lumber

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HIGHLIGHTS

- Less leachate is generated by rain with small lumber and ACQ-treated lumber.
- Leachate volume is correlated to rainfall depth and temperature before rainfall.
- Leachate quality was similar between two sizes of lumber.
- ACQ treatment increased copper, arsenic, and total dissolved solids in wood leachate.
- Leachate copper content is correlated to weather conditions and rain properties

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ABSTRACT

Alkaline copper quaternary (ACQ) is a widely used wood preservative. This study evaluated leachate volume generation and contaminant leaching from ACQ-treated lumber during rainfall events in comparison to untreated lumber. The influences of wood preservation with ACQ, lumber size, and weather on leachate generation ratio and contaminant concentrations in wood leachate were investigated with four red pine lumber piles exposed to natural weather conditions. The average volumetric ratio of leachate to rainfall was significantly higher for the large-lumber piles (0.62) compared with the small-lumber piles (0.35). Less leachate was generated in the ACQ-treated lumber piles (0.42) than the untreated lumber piles (0.55). Leachate volume could be predicted with rainfall depth, air temperature, and wetted lumber surface area. Lumber size did not make a statistically significant difference in leachate quality except for zinc concentration. The average copper concentrations were 4034 μg/L in the leachate from the ACQ-treated lumber piles and 87 μg/L in the leachate from the untreated lumber piles. Moreover, ACQ treatment significantly increased leaching of arsenic and total dissolved solids. Copper concentration in leachate from ACQ-treated lumber can be predicted with rainfall intensity, the time interval between two consecutive leachate-generating events, rain copper concentration, and rain pH

1. Introduction

Except for naturally durable species such as redwood and cedar, wood in outdoor applications is usually pressure treated with preservatives. Preservatives are forced into the cellular structure of wood to deter wood decay. Alkaline copper quaternary (ACQ) is one of the wood preservatives used worldwide for both commercial and residential wood products. The most common formula of ACQ (type D) contains 66.7% copper oxide and 33.3% quat as didecyldimethylammonium chloride [1]. Its primary biocide, copper (Cu), can leach out of wood in outdoor storages and uses by rainfall [2]. Wood leachate generated intermittently by rainfall at various storage sites of freshly treated lumber and decommissioned wood may percolate to groundwater, be discharged to surface waters, or infiltrate through soil. The leached Cu can be toxic to aquatic organisms and pose human health risks at trace levels [1,3].

Environmental authorities are increasingly concerned with environmental impacts of wood preservation [1]. In New York State, for example, Multi-Sector General Permit for Stormwater Discharges Associated with Industrial Activity covers log storage and handling facilities, general sawmills and planing mills, and wood preserving facilities [4]. This general permit sets the benchmark monitoring cutoff of wood leachate and contaminated stormwater at 0.012 mg/L total recoverable Cu for the timber products sector.

Metal leaching from pressure treated wood has been investigated with laboratory tests, such as continuous leaching with synthetic precipitation or an extraction solution [5–9]. However,

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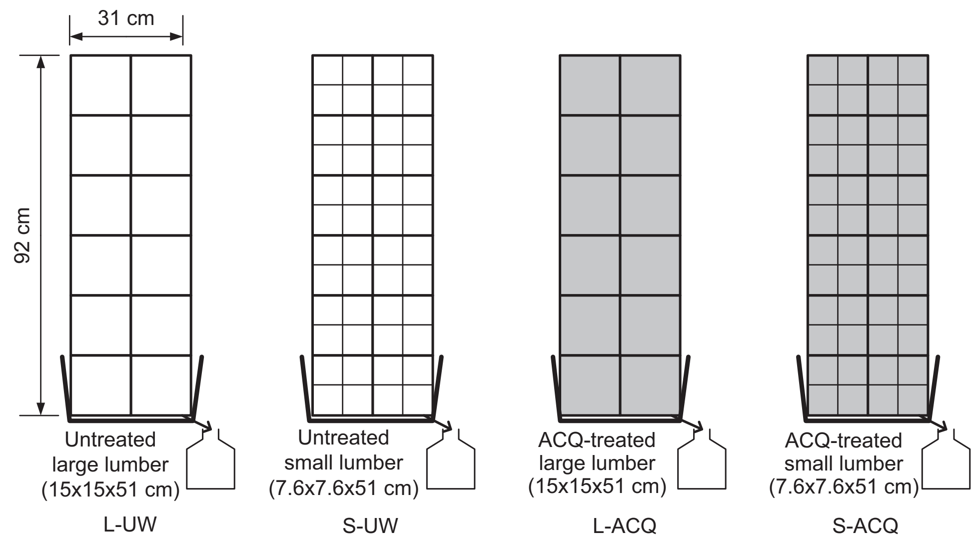

Fig. 1. Sketch of leaching piles with ACQ-treated and untreated red pine at two lumber sizes.

laboratory tests with wood subjected to continuous leaching ignore the influence of weather conditions, water absorption by wood, and weathering on leachate volume generation and metal leaching under field conditions [2,10,11]. Leachate is generated when the volume of rain that falls on a pile of wood is in excess of the volume that the wood can absorb. Laboratory test results do not reflect metal leaching dynamics and intensity in the field and provide no information about the amount of leachate that could actually be generated. Field studies [10,11] have reported influences of rainfall intensity and duration on metal leaching from wood treated with chromated copper arsenate preservatives. Nevertheless, only one recent study [2] investigated metal leaching from ACQ-treated wood under field conditions. Moreover, Hasan et al. [2] only monitored wood leachate weekly, rather than tracking metal leaching across rainfall events. Leachate volume generation and metal leaching from treated wood could vary with weather conditions, wood species, lumber size, and preservative type. No study has yet quantified leachate volume generation and Cu leaching from ACQ-treated wood on a rainfall event basis under field conditions. Investigation into event-based Cu leaching will provide a more realistic evaluation of environmental impacts of Cu leached from ACQ-treated wood.

Trees assimilate various metals, many of which are macro- and micro-nutrients. The inorganic content of wood can be up to 1% of wood dry mass, varying with wood species, soil chemistry and climatic conditions [12]. Metals could leach out of untreated wood as well [13–17]. Moreover, leachate from untreated wood has high concentrations of organic substances [12,18]. Some of the organic substances such as tannins and phenols are toxic to aquatic organisms, though the greatest concern is oxygen demand of the organic substances [12,18]. Pressure treatment deters wood decay, but erosion of wood fiber from lumber surfaces still occurs [7]. The effects of pressure treatment with ACQ on leaching of oxygen-demanding substances and non-biocide metals are unknown. Furthermore, different sizes of lumber have different surface areas exposed to precipitation when stored outdoors, which affects initial water retention and could result in different leachate characteristics [6,11]. To the authors' knowledge, there are no side-by-side comparisons of organic matter and non-biocide metals leaching from untreated and treated wood in the literature.

The objectives of this study were 1) to evaluate the intensity and dynamics of leachate volume generation and contaminant leaching from newly ACQ-treated red pine lumber in comparison to those from untreated lumber during rainfall events; 2) to examine the effects of lumber size on wood leaching; and 3) to develop regression models to predict leachate volume generation and contaminant leaching with meteorological parameters.

2. Materials and methods

2.1. Wood leaching piles

Four wood leaching piles were set up on March 6, 2010 in an open area located in Syracuse, New York, USA. The dimensions and configuration of the piles were selected to simulate wood leaching by rainfall in outdoor storage situations. Each pile was stacked with new red pine lumber (51 cm long) to 92 cm tall in a rectangular polypropylene tank with interior floor dimensions of 32 cm by 53 cm. The lumber pieces in each tank were tied with a plastic string to keep the piles straight and stable. The four piles were differentiated by lumber size (15 cm × 15 cm vs. 7.6 cm × 7.6 cm) and ACQ treatment as shown in Fig. 1. The ACQ-treated red pine lumber was produced for above ground use, which had a preservative retention level of 2.4 kg Cu/m^3 as specified by AWPA [19]. Each pile had a total lumber volume of 0.14 m^3 and a top surface area of 0.16 m^2. Each tank holding a lumber pile had top dimensions of 34 cm by 54 cm. The slightly larger top dimensions relative to the floor dimensions were chosen to minimize direct rainfall to the open space of the tanks and prevent rainfall on the piles from significant loss due to splashes. When leachate was generated during rainfall events it was drained freely by gravity into acid-washed polypropylene bottles. A sheet of 1-cm polystyrene foam was laid on the bottom of each tank to avoid submersion of wood in leachate. A Taylor 1″ analog rain gauge was set up beside the wood piles to record rainfall depth and collect rain samples for individual rainfall events.

Syracuse has a humid continental climate, with cold, snowy winters and relatively cool summers. Mean annual precipitation is 1017 mm. Precipitation is well distributed throughout the year, while snow falls mostly in the period from December to March. Daily average temperatures recorded at the nearby SUNY ESF Weather Station [20] were collected for the study period. The average daily temperature during the study period from March 6 to November 18, 2010 was 15.8 °C, with a maximum temperature of 31.2 °C and a minimum temperature of −7.2 °C.

2.2. Water sampling and laboratory analysis

There were 25 rainfall events that generated leachate between March 6 and November 18, 2010. Rainfall depth and leachate volume were recorded at the end of each rainfall event. Rainfall and leachate samples were collected in 17 of the 25 events for laboratory analysis. No samples were collected in the other 8 events over the study period because of logistical reasons. Samples were collected at the end of individual leachate-generating events except for two additional times of sampling during the second event.

Chemical oxygen demand (COD) was determined colorimetrically with a DR 2800 spectrophotometer (Hach Company, Loveland, CO, USA), following Standard Method 5220-D [21]. Total dissolved solids (TDS) and pH were measured using a HQ40d meter with a CDC401 conductivity probe (Hach Company, Loveland, CO, USA) and a portable pH meter (pH 6 model, Oakton Instruments, Vernon Hills, IL, USA), respectively. Metal concentrations were determined with an Inductively Coupled Plasma-Mass Spectrometer (ICP-MS, ElanDRC-e, Perkin Elmer, USA), following Standard Method 3125 [21]. The subsamples for dissolved metal analysis were filtered through acid-washed membrane filters (pore size 0.45 μm), following Standard Method 3030-B [21]. The subsamples for total metal analysis were acid digested using Standard Method 3030-E [21]. Before digestion and ICP-MS analysis, subsamples were preserved immediately with nitric acid at pH < 2 and stored at 4 °C. The detection limits of the ICP-MS were ≤0.1 μg/L for the metals. Quality control measures for ICP-MS included instrument blanks (deionized water), method blanks (deionized water through digestion), sample spikes, and duplicates.

Concentrations of dissolved Cu, COD and TDS plus pH values in rain and leachate were determined for all the samples. Concentrations of dissolved Al, Ca, Cd, Fe, K, Mg, Mn, Mo, Na, Ni, Pb, and Si were determined only for the second, third, and fourth leachate-generating events to examine potential effects of pressure treatment on leaching of these non-biocide metals. Concentrations of total As, Cr, Cu, and Zn in the unfiltered subsamples were determined only for the first five leachate-generating events to examine the differences between total metal concentrations and dissolved metal concentrations.

2.3. Data analysis

Leachate generation ratio was calculated as leachate volume divided by rainfall volume for individual leachate-generating events. Two-factor analysis of variance (ANOVA) was performed to examine the effects of lumber size and wood treatment with ACQ on leachate quality and generation ratio. One-factor ANOVA was used to test the differences between dissolved and total metal concentrations in wood leachate. One-factor ANOVA was also used to test the differences between rain and wood leachate. Least significant difference (LSD) was calculated to identify the significant differences in one-factor ANOVA [22]. Spearman rank trend test was performed to identify the trends of contaminant leaching over time [22]. Multiple linear regression was performed with a backward elimination approach to analyze how weather conditions affected leachate volume and contaminant concentrations. Histograms were plotted to examine normality of frequency distribution. When normal distribution and homoscedasticity were not met for ANOVA and regression, data transformation (log or square root) was applied to increase normality of frequency distribution. The significance level (p value) was set at 0.05 for all statistical analyses. Coefficient of determination (R^2) was calculated to evaluate the similarity in leached Cu mass between the observations and predictions.

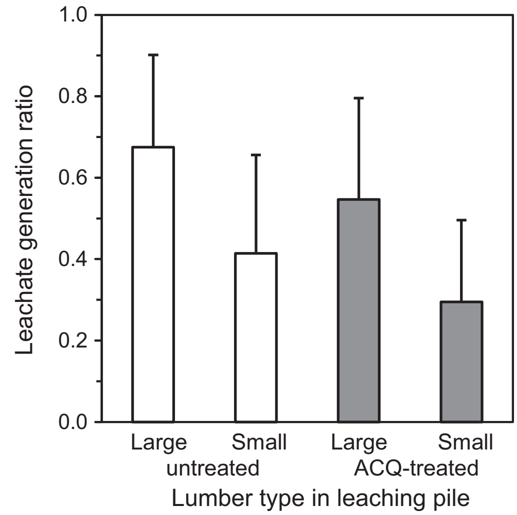

Fig. 2. Variation of average leachate generation ratio (leachate volume: rainfall volume) with lumber size and ACQ treatment. $n = 25$; error bar = standard deviation.

3. Results and Discussion

3.1. Factors influencing leachate volume generation

There were 25 rainfall events that produced leachate during the study period, covering a wide variety of weather conditions. The remaining rainfall events did not produce leachate because the rain water was all absorbed into the lumber. The leachate-generating events had rainfall in the range of 8.5–140.4 mm. The durations of these events ranged from 40 to 2130 min. Rainfall intensity was 0.01–0.46 mm/min. There were 1.1–24.2 dry days between two consecutive leachate-generating events. The average temperature between two consecutive events varied between 3.6 and 24.4 °C.

Two-factor ANOVA showed that both ACQ treatment and lumber size made significant differences in leachate generation ratio ($p = 0.02$ and <0.01, respectively), and lumber size and ACQ-treatment had independent effects on leachate generation ratio ($p = 0.85$). The average leachate generation ratio was higher in the piles with large lumber and untreated lumber (Fig. 2). ACQ-treatment might have increased the equilibrium moisture content of treated wood [23], resulting in a lower leachate generation ratio because a higher equilibrium moisture content of a wood product means a greater capacity to absorb rain water until equilibrium with its surroundings. The difference in leachate generation ratio between the large-lumber and small-lumber piles could be attributed to the different surface areas available to absorb rain.

Fig. 3 shows the rainfall depths and leachate volumes across the leachate-generating events during the study period. A portion of the initial rainfall could be absorbed by wood through the wetted surfaces. Equilibrium moisture content or hygroscopic capacity of lumber is affected by air temperature between two consecutive rainfall events [23,24]. Therefore, leachate volume from each leaching pile was significantly correlated with rainfall depth (total depth of rainfall during an event), air temperature between two consecutive leachate-generating events, and wetted surface area (Table 1). Wetted surface area is related to lumber size and pile stacking pattern. It was observed that the wetted surface areas were 2.61 m^2 for each large-lumber pile and 4.48 m^2 for each small-lumber pile, including the top surface and all vertical surfaces. Eqs. (1) and (2) are the regression models that can be used to predict leachate volume. Regression analysis showed that the time interval between two consecutive leachate-generating events, rainfall intensity, and

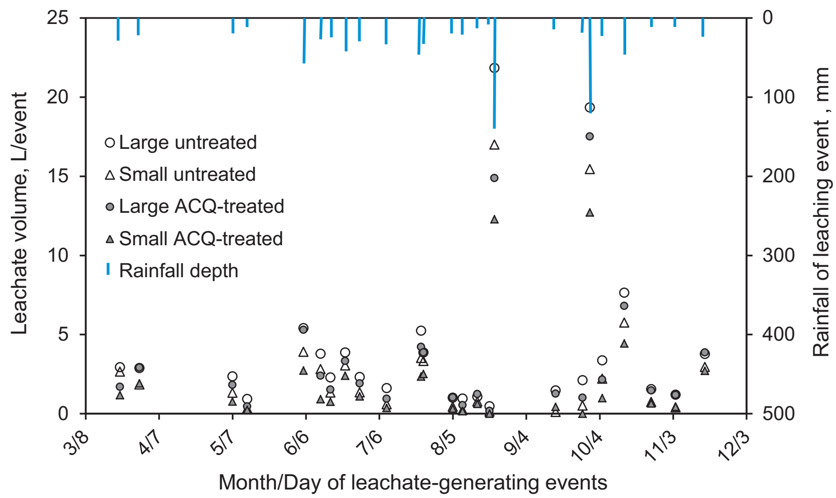

Fig. 3. Rainfall depth and leachate volume generated in red pine lumber piles during individual rainfall events.

event duration had insignificant roles in leachate volume generation.

$$\sqrt{V} = 0.030D - 0.026T - 0.205A + 1.75 \text{ for untreated lumber} \quad (1)$$

$$\sqrt{V} = 0.027D - 0.025T - 0.231A + 1.71 \text{ for ACQ-treated lumber} \quad (2)$$

where V is the leachate volume generated during a rainfall event (L); A is wetted surface area of a lumber pile (m^2); D is rainfall depth in a leachate-generating event (mm); and T is the average air temperature between two consecutive leachate-generating events (°C).

3.2. Effects of ACQ treatment and lumber size on leachate quality

Leachate quality is summarized in Table 2. Two-factor ANOVA analysis showed that there were insignificant differences between the two sizes of lumber in leachate pH and concentrations of COD, TDS, dissolved As, dissolved Cr, and dissolved Cu (p = 0.24–0.99). The only exception was that Zn concentration was significantly higher (p = 0.02) in the leachate from the small-lumber piles than leachate from the large-lumber piles. The insignificant differences in leachate quality between the two sizes of lumber were possibly because the difference in the wetted surface areas was not large enough. The significant effect of lumber size on leachate Zn concentration could be attributed to the higher mobility of Zn in wood compared with Cu and Cr [25], which resulted in more Zn leached from the larger wetted surface area of the small-lumber piles.

Table 1
Properties of multiple linear regression for wood leachate volume generated in untreated and ACQ-treated lumber piles during individual rainfall events. n = 50.

	Eq. (1) for untreated lumber	Eq. (2) for ACQ-treated lumber
Adjusted R^2	0.92	0.88
p-value		
Overall	<0.001	<0.001
Coefficient of rainfall depth (D)	<0.001	<0.001
Coefficient of temperature (T)	<0.001	0.002
Coefficient of wetted surface (A)	<0.001	<0.001
Intercept	<0.001	<0.001

Total metal concentrations were found to be statistically similar to dissolved metal concentrations for As, Cu, Cr, and Zn in both wood leachate and rain (one-factor ANOVA, p = 0.09–0.95), indicating that these four metals were mainly in dissolved forms. Dissolved Cu concentration in the leachate from the ACQ-treated lumber piles was 41–51 times those in the leachate from the untreated lumber piles (Fig. 4). Leachate from untreated lumber had average Cu concentrations slightly higher than rain (Table 2), although the New York State benchmark monitoring cutoff concentration for stormwater discharges associated with timber products sector is even lower, 12 μg/L acid recoverable Cu [4]. Hasan et al. [2] found lower Cu concentrations for rain and untreated wood leachate (7.6 and 4.5 μg/L), but moderate Cu concentration in leachate from ACQ-treated southern yellow pine (637 μg/L). The lower Cu concentration in the leachate of southern yellow pine boards was possibly because of a lower ACQ retention level in its boards (0.88 kg Cu/m^3) and more frequent rainfall events in Miami, Florida than in this study.

Dissolved As concentrations in leachate were significantly affected by ACQ treatment (two-factor ANOVA, p < 0.01). Rain had significantly lower As concentration than the leachate from the ACQ-treated lumber (Table 3). This raises a question whether ACQ treatment increases the leachability of As that was originally assimilated in red pine wood. Similarly, Hasan et al. [2] reported that As concentration in leachate from ACQ-treated southern yellow pine was nearly two times that from untreated pine.

ACQ treatment did not result in a significant difference in leachate concentration for dissolved Cr (two-factor ANOVA, p = 0.39) and Zn (p = 0.10). There were no significant differences in the Cr concentrations between rain and leachate (one-factor ANOVA, p = 0.13). Rain had significantly lower Zn concentration than leachate (one-factor ANOVA, p < 0.01; LSD = 129 μg/L), indicating Zn leached from both untreated and ACQ-treated red pine. This confirms that Zn has a higher mobility than Cr and Cu in wood [25]. The concentrations of the other metals were similar in the leachate from the two sizes of red pine lumber. As shown in Table 3, the concentrations of the other metals in ACQ-treated lumber leachate were similar to or slightly higher than those in untreated lumber leachate, indicating little effect of ACQ treatment on the leaching of other metals.

Although leachate TDS concentration decreased significantly over time with both the ACQ-treated and untreated lumber piles (Spearman rank, p = 0.008–0.010) as shown in Fig. 5a, the TDS concentrations in the leachate from ACQ-treated lumber was significantly higher than those in the leachate from untreated

Table 2
Characteristics of rain and leachate generated by rainfall from red pine lumber piles[a].

	Rain	Untreated lumber		ACQ-treated lumber		Benchmark monitoring cutoff [4]
		Large	Small	Large	Small	
pH	5.2 ± 1.0	5.8 ± 0.4	5.6 ± 0.4	5.9 ± 0.8	5.9 ± 0.4	6–9
COD (mg/L)	131 ± 112	237 ± 117	277 ± 110	291 ± 143	328 ± 161	120
TDS (mg/L)	18.3 ± 17.1	33.5 ± 11.5	35.9 ± 11.6	51.6 ± 27.6	55.9 ± 19.4	n/a
Cu (μg/L)	48 ± 52	90 ± 67	85 ± 51	3704 ± 1927	4363 ± 1542	12
As (μg/L)	0.2 ± 0.1	2.0 ± 0.4	3.1 ± 0.9	11.2 ± 3.9	10.1 ± 3.2	150
Cr (μg/L)	1.1 ± 0.8	2.3 ± 1.1	3.3 ± 2.0	3.3 ± 2.9	3.9 ± 2.9	1800
Zn (μg/L)	34 ± 26	79 ± 47	229 ± 202	188 ± 128	290 ± 98	110

[a] Mean ± standard deviation. $n = 7$ for dissolved arsenic (As), dissolved chromium (Cr), and dissolved zinc (Zn); $n = 17$ for pH, dissolved copper (Cu), chemical oxygen demand (COD), and total dissolved solids (TDS).

lumber (two-factor ANOVA, $p < 0.01$). The TDS concentrations in rain were significantly lower than those in leachate (one-factor ANOVA, $p < 0.01$; LSD = 13.0 mg/L). The TDS increase with ACQ treatment indicated that ACQ treatment not only increased Cu leaching, but leaching of other unidentified constituents as well.

Leachate pH was not significantly affected by ACQ treatment (two-factor ANOVA, $p = 0.16$). Leachate had significantly higher pH values than rain (one-factor ANOVA, $p = 0.01$; LSD = 0.4), indicating that both untreated and treated wood buffered rain pH (Table 2). This was consistent with results reported by Hasan et al. [2] for ACQ-treated southern yellow pine boards.

Leachate COD concentration was not significantly affected by ACQ treatment (two-factor ANOVA, $p = 0.11$). The leachate COD could be partially attributed to the high COD concentration in the rain, although the average rain COD concentration was significantly lower than the leachate COD concentrations (one-factor ANOVA, $p < 0.01$; LSD = 94 mg/L).

The average concentrations of dissolved Cr and As in rain and leachate from the lumber piles (Table 2) were lower than the New York State benchmark monitoring cutoff concentrations [4]. Nevertheless, leachate Zn and COD concentrations were typically higher than the benchmark monitoring cutoff concentrations [4]. The leachate pH values were out of the general permit limits for

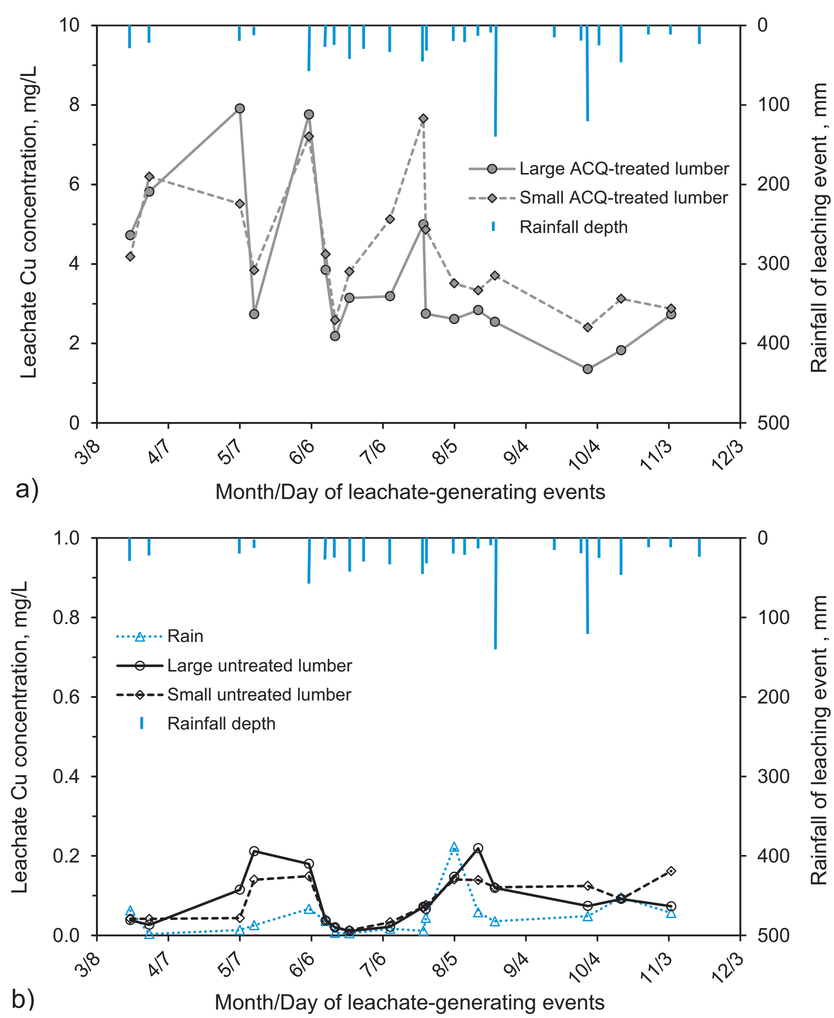

Fig. 4. Dynamics of Cu concentration in leachate from ACQ-treated and untreated red pine lumber piles.

Table 3
Concentrations of dissolved metals in leachate of red pine lumber and runoff from wood handling sites.

	Rain[a]	Red pine lumber[a]		Log yards			Cedar wood waste [17]
		Untreated	ACQ-treated	Reference [14]	Reference [16]	Reference [15]	
Ca (mg/L)	0.52–3.56	5.57–9.82	8.33–16.35	4.44–26.0	85.4		83
K (mg/L)	0.25–2.03	7.21–10.71	6.54–9.88	8–26	178		
Mg (mg/L)	0.10–0.67	1.48–2.82	2.64–5.54	4.9–41.6	167		44
Mn (mg/L)	0.01–0.05	0.52–0.99	0.34–0.73	0.24–1.45	15.4		
Na (mg/L)	0.76–0.78	3.00–5.24	4.29–10.76	72–385	1424	0.8–8.9	
Si (mg/L)	0.11–0.17	0.10–0.27	0.15–0.30	1.28–10.10	7.6	<0.1–1.5	
Al (μg/L)	26–48	43–163	41–74	500–6000	9	1.2–34.5	19 000
As (μg/L)	0.1–0.3	1.4–4.9	6.5–16.0	ND		13–132	
Cd (μg/L)	<0.1–0.3	0.4–0.6	0.4–1.0	ND			
Cr (μg/L)	0.2–2.7	0.9–7.1	0.5–9.2	ND			41
Cu (μg/L)	3–223	11–219	1353–7912	ND-30	79	2.6–44.7	<100
Fe (μg/L)	101–106	64–144	69–116	640–8790	9.2	0.5–27.1	75 000
Mo (μg/L)	0.2–3.2	0.4–9.2	0.5–4.5	ND		25.7–537	
Ni (μg/L)	0.6–8.5	1.1–21.1	1.6–3.1	ND			<100
Pb (μg/L)	0.6–21.0	0.3–3.6	0.9–2.3	ND			<400
Zn (μg/L)	13–74	39–678	79–479	23–186	1180		156–400

[a] This study: $n = 17$ for Cu; $n = 7$ for As, Cr and Zn; and $n = 3$ for the other metals.

stormwater discharges associated with the timber products sector in New York State [4]. Therefore, leachate either from ACQ-treated lumber or untreated lumber should be treated for discharge permit compliance.

3.3. Other factors influencing leachate quality

There are considerable variations in non-biocide metal concentrations of leachate from treated and untreated wood (Table 3). Similar to organic substances in leachate of untreated wood and runoff from wood handling sites [12,18], the variations in metal concentrations across studies could be attributed to differences in wood species, climate, site pavement, wood products, and storage or service duration. Logyard runoff tends to have higher concentrations of Ca, K, Mg, Mn, Na, Si, Al, and Fe (Table 3), which may result from dissolution of soil metals. Concentrations of the other metals in the leachate of red pine lumber in the present study were of a magnitude similar to those in logyard runoff as shown in Table 3. Compared with leachate from wood waste, leachate in the present study had lower metal concentrations except for Cu from ACQ-treated red pine lumber. The difference could be attributed to the larger surface area of wood waste (wood chips, shredded bark, and sawdust) subjected to rainfall percolation [24]. Due to the same reason, red pine lumber leachate in the present study also had lower concentrations of TDS and COD (Table 2) than those in wood waste leachate, 6508 mg/L and 3908–12626 mg/L, respectively [24].

Al, Ca, K, Mg, Mn, and Si are major inorganic components of wood, while heavy metals and other trace elements such as Ba, Ni, and Zn are minor components [12]. Dissolved metal concentrations in the red pine leachate were higher than those in rain for most of the metals, especially some nutrient metals such as Ca, K, Mg, Mn, Na, and Zn (Table 3). It seems there were no considerable differences in nutrient metal concentrations between the ACQ-treated lumber and untreated lumber.

3.4. Correlation of copper leaching with meteorological parameters

Fig. 5a,c show the dynamics of leachate TDS and COD concentrations across the leachate-generating events. There were no significant relationships of leachate concentration with meteorological parameters and rain concentration for COD or TDS. TDS concentration in the leachate from the ACQ-treated lumber was relatively higher in the first leachate-generating event, likely due to wash-out of original surface residues.

There were no significant trends of dissolved Cu concentration in rain and leachate from the untreated lumber over the study period (Spearman rank, $p = 0.12$ and 0.18, respectively) as shown in Fig 4b. Metals bound on the surface of the treated wood leach rapidly in the early stages [26]. Dissolved Cu concentration in the leachate from the ACQ-treated lumber decreased significantly over time (Spearman rank, $p = 0.003$). Similarly, laboratory tests indicated decreasing Cu leaching rates in initial stages of leaching [27,28]. Nevertheless, treated lumber is rarely stored outdoors for longer than a year.

No significant correlations were found with multiple linear regression between dissolved Cu concentration in the leachate from the untreated lumber and meteorological parameters. However, Cu concentration in the leachate from the ACQ-treated lumber was strongly correlated with weather and rain quality as shown by Eq. (3) (Table 4). Leaching of metals from pressure-treated wood involves metal dissolution and transport [10,11,26]: rain water penetrates into wood where fixed and complexed metals are hydrolyzed and/or dissolved into water; the dissolved metals then diffuse to the surface of the wood, precipitate as water evaporates, and become available for wash-out during the next rainfall event. Copper tends to bind weakly to labile wood cellulose [11]. Longer dry days allow more dissolved Cu to diffuse to and precipitate at the lumber surface, where the subsequent leachate-generating events could wash the precipitates out and leach more Cu. A lower pH can increase solubility and mobility of metals [5,8,10], causing a relatively higher metal concentration in wood leachate. A higher Cu concentration in rain could indicate higher concentrations of organic and inorganic ligands in the rain, which lower the labile fraction of Cu and result in lower Cu concentrations in leachate [5]. Metals may also be washed out wood surfaces with fiber and due to UV breakdown of lignin [7,11], which is enhanced by a greater rainfall intensity.

$$\sqrt{C} = 29.7\sqrt{I} + 1.742N - 0.138R_{Cu} - 3.00\,R_{pH} + 61.52 \quad (3)$$

where C is the dissolved Cu concentration in leachate from ACQ-treated red pine lumber (mg/L); I is rainfall intensity of a leachate-generating event (mm/min); N is the number of days between two consecutive leachate-generating events; R_{Cu} is rain Cu concentration (mg/L); and R_{pH} is rain pH value.

The concentrations of Cu in rain varied greatly compared with those in the leachate in the present study (Tables 2 and 3). Both rain

Table 4
Properties of multiple linear regression for concentration of copper in leachate from ACQ-treated lumber. $n = 17$.

	Regression with different independent variables			
	I, N, R_{Cu}, & R_{pH}	I, N, & R_{pH}	I, N, & R_{Cu}	I, & N
R^2	0.80	0.61	0.75	0.55
Adjusted R^2	0.74	0.52	0.69	0.48
p-value				
Overall	<0.001	0.006	<0.001	0.004
Coefficient of rainfall intensity (I)	0.062	0.895	0.074	0.900
Coefficient of dry days (N)	<0.001	0.002	<0.001	0.002
Coefficient of rain Cu (R_{Cu})	0.005		0.006	
Coefficient of rain pH (R_{pH})	0.099	0.181		
Intercept	<0.001	<0.001	<0.001	<0.001

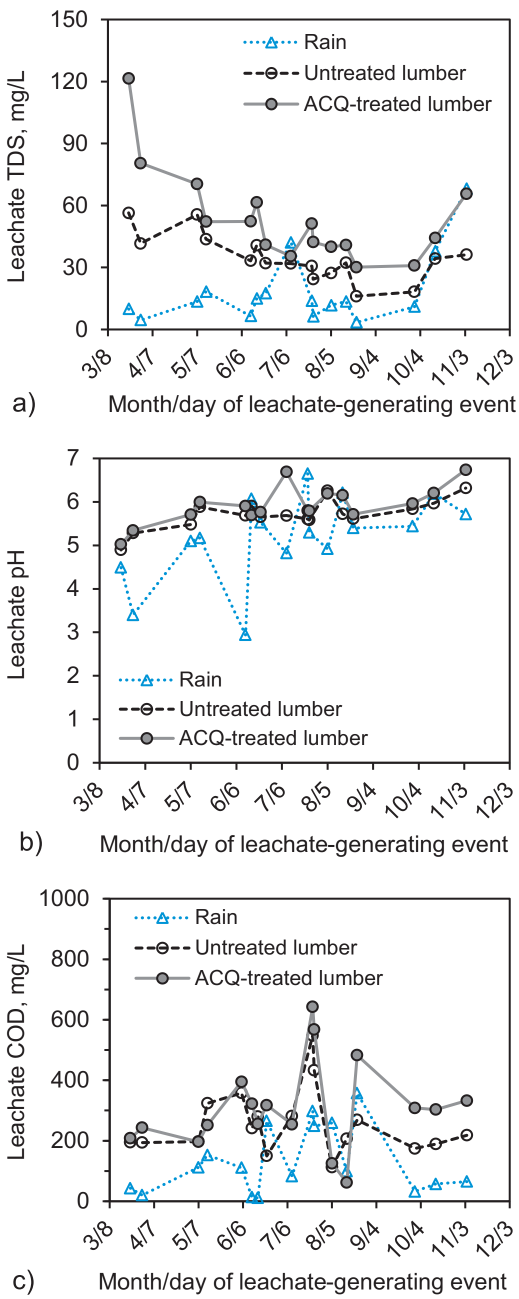

Fig. 5. Dynamics of total dissolved solids (TDS) concentration, pH, and chemical oxygen demand (COD) concentration in leachate from ACQ-treated and untreated red pine lumber piles.

and leachate pH values increased significantly over time as well (Spearman rank, $p = 0.002$–0.010). If rain Cu concentration and pH were excluded to simplify the regression equation, then R^2 would decrease and p-values could increase (Table 4).

Simulating both leachate volume and Cu concentration with the regression models provides basic information for design of leachate treatment processes and preparation of management measures, especially for new timber facilities. Eqs. (2) and (3) together provide a convenient tool to estimate mass of Cu leached from ACQ-treated red pine lumber in individual leachate-generating events. As shown in Fig. 6, the predictions of Cu mass for individual leachate-generating events match closely the mass calculated with

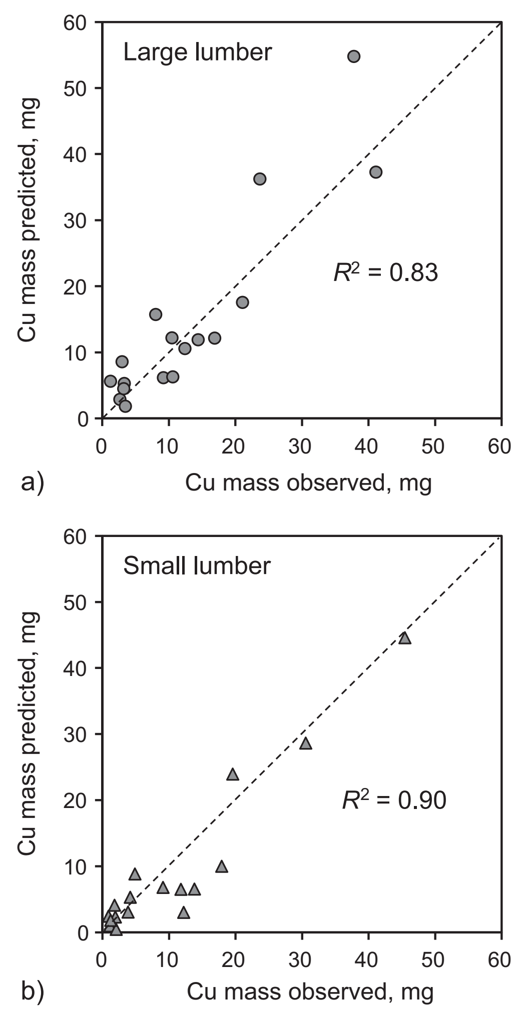

Fig. 6. Observed and predicted Cu mass in leachate from the large and small ACQ-treated lumber piles during individual rainfall events.

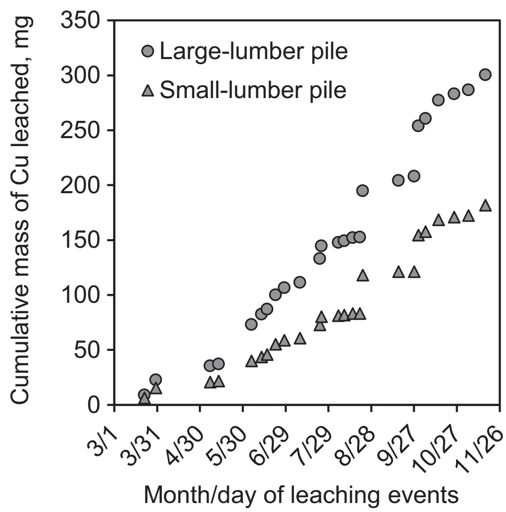

Fig. 7. Cumulative mass of copper leached from two ACQ-treated red pine piles with different lumber sizes over 25 leachate-generating events in 2010.

measurements of leachate volume and Cu concentration. The mass of dissolved Cu leached from the large ACQ-treated lumber pile was often higher than that from the small ACQ-treated lumber pile during individual events because of similar leachate Cu concentrations and higher leachate volume generated from the large-lumber pile. Consequently, large-lumber piles of ACQ-treated red pine would pose greater cumulative effects of Cu on the receiving waters when the wood leachate is discharged without treatment in relative to small-lumber piles as shown in Fig. 7. The copper leaching rate was 2.01 g Cu/m^3 rain for the large ACQ-treated lumber pile and 1.65 g Cu/m^3 rain for the small ACQ-treated lumber pile on average in this study. Based on weekly sampling, Hasan et al. [2] reported a similar Cu leaching rate (1.82 g Cu/m^3 rain) for new ACQ-treated southern yellow pine boards in Miami, Florida.

4. Conclusions

This study investigated leachate volume generation from pressure-treated wood during rainfall events. It evaluated the influences of red pine treatment with ACQ, lumber size, and weather conditions on field leaching of metals, chemical oxygen demand, and total dissolved solids during rainfall events. The following are conclusions from this study:

- The volumetric ratio of wood leachate to rainfall is significantly affected by ACQ treatment and lumber size. Leachate volume was significantly higher with large lumber and lower with ACQ-treated lumber. Leachate volume could be predicted based on rainfall depth from individual leachate-generating events, air temperature between two consecutive events, and wetted lumber surface area.
- Arsenic, copper, chromium, and zinc in the leachate were mostly in dissolved forms. Lumber size did not make a statistically significant difference in leachate concentrations of copper, chromium, arsenic, chemical oxygen demand, and total dissolved solids. ACQ treatment significantly increased leachate arsenic and total dissolved solids concentrations in addition to significant Cu leaching.
- Copper leaching from ACQ-treated lumber is a dynamic process, which can be simulated with rainfall intensity, time interval between two consecutive leachate-generating events, rain copper concentration, and rain pH.
- The average Cu concentration in the leachate of the ACQ-treated lumber was 4034 μg/L, 336 times the New York State benchmark monitoring cutoff concentration for stormwater discharges associated with the timber products sector. Proper treatment is needed for stormwater from ACQ-treated lumber storage sites.

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