

Modeling of wood preservative leaching in service

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Abstract:

To evaluate the potential environmental and health implications of leaching of wood preservatives in service under different conditions, there is a need for a predictive model that provides estimates of the rate and extent of leaching over a wide range of exposure conditions. In this paper, we test the hypothesis that preservative component emissions can be modeled on the basis of three easily measured parameters: total leachable component based on intensive leaching of fine ground material; amount of dissolved or dissociated component in water saturated wood; and preservative component diffusion coefficients in the longitudinal and transverse directions. These variables are estimated for ACQ, copper azole, CCA and borate wood preservatives and the leaching rates of lumber samples are compared with the rate predicted by the model. This model approach should permit the estimation of leaching rates under a wide variety of product geometries and exposure conditions.

Keywords: Leaching, preservatives, diffusion, dissociation, risk assessment

INTRODUCTION

In the risk assessment of new and currently in use wood preservatives, it is important to have representative estimates of the preservative component leaching rates under different exposure scenarios and to understand how these leaching rates change with time and other variables. This information, in combination with data on dilution and fate of substances in the environment, permits the estimation of predicted environmental concentrations (PEC) at any time in service [1]. These PEC values can be compared to predicted no effect concentrations (PNEC) to assess their potential environmental impacts. There are a number of initiatives through the OECD, EU, AWPA and other organizations and associations [2,3,4] to devise appropriate leaching procedures that can predict both expected long term efficacy of the treatment and potential impacts of leachates on human health and the environment. Willeitner and Peek [6] discussed the difficulty of devising a practical test that is cheap, fast and reproducible while giving a realistic indication of short term and long term impacts. Exposure of the environment to wood preservative contaminants and the resulting risk is best characterised by the rate of preservative emission from the treated wood, expressed as amount released per unit surface area per unit time or flux (typically $\mu\text{g}/\text{cm}^2/\text{hour}$). It has been shown that flux decreases with time in service and that the most acute environmental and health exposure to

leached substances occurs in the period soon after installation [7] while long term accumulations in soil and sediments result from the lifetime exposure to ever-declining emissions. Previous studies [7,8,9,2] have modeled this declining flux rate, usually with exponential equations. These models can be applied to risk assessment scenarios to predict releases at any given time; long term accumulations in water columns, soils and sediments can be estimated by determining the area under the curve by integration or by use of a “growth model” as suggested by Homan and van Oosten [1].

Based on our comparisons of laboratory and field leaching studies, we believe that leaching consists of a number of concurrent and consecutive phenomena, each of which is affected by a number of wood product, preservative and exposure conditions:

- Wetting of wood and penetration of free water into the wood void space (more relevant for intermittent rain exposure than continuous water soaking conditions). This factor depends on wood permeability, surface wetting properties, orientation to the weather, type of exposure etc.
- Dissolving/dissociation of precipitated or reacted preservative components and diffusion out of the cell walls into free water in the cell lumens to the limit of their solubility under the test conditions (depends on preservative, temperature, MC, etc.). This creates the concentration gradient that drives the diffusion process.
- Diffusion of preservative components to the wood surface. The diffusion rate will depend on a number of factors such as wood permeability, moisture content, temperature, dimensions, direction of movement in wood, and especially the nature of the diffusing species and the concentration gradient established; an understanding of the effects of solute ion size, charge, solubility and other chemical properties will be helpful in predicting how new preservatives will perform.
- Drying of wood after rain exposure and possibly, wicking of dissolved components to the wood surface – not relevant for continuous water soaking conditions (depends on ambient T, RH, air flow conditions).

To model leaching emissions, we propose that there are advantages to using diffusion theory rather than empirical models. Leaching of wood components from two opposing surfaces can be described by one-dimensional Fickian diffusion.

$$\frac{\partial C}{\partial t} = D \frac{\partial^2 C}{\partial x^2} \quad (1)$$

For leaching of a preservative component from wood, C is the concentration of preservative in wood at any time t that is available for leaching. D = diffusion coefficient (cm^2/s); l = sample length in the direction of movement of leached component if leached from only one side, or half the sample length if the two opposing faces are leached; x = diffusion direction. The above equation is solved for boundary conditions: (i) Uniform initial analyte concentration in the wood at the start of leaching, ie, the initial concentration C(x,0) is a constant Co, i.e. all of the available solute is dissolved in the cell lumen water and no additional preservative dissociates. (ii) The component concentration is 0 at the wood surface/water interface i.e., the water is changed frequently; (iii) Continuous leaching in one direction only.

In this case, the mass of material M leached as a function of time, relative to the ultimate amount available (M_{inf}) is given by Equation 2. For preservative systems with low solubility reaction products, M_{inf} is the amount of preservative that is available for leaching rather than the amount of preservative in the wood.

$$M/M_{inf} = \sum_{n=0}^{\infty} \frac{8}{(2n+1)^2 \pi^2} e^{-\frac{D(2n+1)^2 \pi^2 t}{4l^2}} \quad (2)$$

Equation 2 estimates cumulative loss (M/M_{inf}) at time t (s), as a function of specimen dimension in the diffusion direction ($2l$ cm for diffusion from two opposing surfaces) and diffusion coefficient (D). M/M_{inf} can be related to the % of total leached which is a common expression of cumulative leaching if the amount available for leaching is known and expressed as a % of total of preservative retained in the wood. Estimates can be converted to flux estimates ($\mu\text{g}/\text{cm}^2/\text{time}$) at any elapsed time (t) by computing M (in μg) at two consecutive time intervals and dividing the change in amount leached over the interval by the exposed sample area and the time interval.

Application of an appropriate diffusion model to the loss of preservative from wood by leaching offers the following advantages:

1. Diffusion models are science based and should describe the physical process more consistently than empirical models. Extrapolation of results based on diffusion theory should be more reliable than extrapolation of empirical models; furthermore, they provide a theoretical basis to assist with the design of appropriate test methods.
2. The diffusion model corrects for effects of specimen dimensions. It should be possible to use small scale short term laboratory tests to predict long term emission characteristics.
3. Diffusion coefficients can be determined for variables of interest such as wood direction (longitudinal vs. transverse), temperature, wood species, preservative component and formulation.
4. Diffusion equations can be solved for virtually any product geometry (pole, board, block) and can be solved in three dimensions to describe practical leaching scenarios.

We hypothesise that the leaching process for a given product under continuous moisture exposure conditions can be described adequately by three fairly simple-to-measure factors:

1. Percentage of preservative components available for leaching;
2. Equilibrium dissociation of preservative components in wood;
3. Diffusion coefficients for movement of preservative components out of wood

MATERIALS AND METHODS

To evaluate the equilibrium dissociation of preservative components, solid treated wood samples (25 mm cubes) at a moisture content of approximately 10 % were cut from southern yellow pine (*Pinus* sp.) 38mm X 140 mm boards commercially treated with chromated copper arsenate (CCA-C), amine copper quaternary (ACQ-C), copper azole (CA-B) and disodium octaborate tetrahydrate (DOT). For more refractory species such as spruce, jack pine and red pine, thinner wafers (20mm X 50mm X 5mm thick) were treated with different concentration ACQ-D preservative solutions. The inorganic preservative component retention of ground samples was determined (kg/m^3) by digesting a 1 g sample in H_2O_2 /Nitric Acid (AWPA 1993) and analyzed by inductively coupled plasma (ICP) spectroscopy for copper (CCA, ACQ, CA), chromium (CCA) arsenic (CCA) and borate (ACQ and DOT). After preservative stabilization, the wafers were vacuum/pressure impregnated with water. The mass uptake of water was determined and assumed to represent the amount of water in the wood containing dissolved components. The rates of change in amounts of preservative components in the free water in the cell lumens was determined by squeezing the blocks in a press to express solution from matched samples at different times. The expressed solution was analysed for the above inorganic components by ICP and the mass of dissolved component (D_i) expressed as a percentage of total content of the wood:

$$D_i = \frac{\text{Water uptake (g)} \times \text{concentration in expressate } (\mu\text{g/g})}{\text{Retention in wood } (\mu\text{g/g}) \times \text{sample mass (g)}} \times 100 \% \quad (3)$$

The percentage of preservative components readily available for leaching was estimated from the leaching performance of finely ground wood. Cross-section samples of the same wood samples as above were ground to pass a 1 mm screen. Five grams of the ground samples were leached in 50 mL water at 21°C and the water exchanged regularly over a 12-week period. Cumulative percent leaching was determined by analysing the leachate by ICP.

Under ideal leaching conditions, when the wood is sufficiently saturated with water and the preservative components are dissolved in the water to their limit of solubility, the diffusion of the components out of wood can be characterized using the non-steady state diffusion model shown in Equation 2.

The maximum amount of preservative available for leaching (M_{inf}) should be the total leachable amount (Le) determined as above but the quantity of dissociated preservative at equilibrium (Di) establishes the concentration gradient that controls the diffusion process. Unless these quantities are identical, use of Le will over-estimate the rate of leaching while use of Di will under-estimate the cumulative amount of leaching at any time. Alternatively, M_{inf} can be estimated from the model fitting exercise. In this study, diffusion values are estimated using the amount of dissociated preservative as the first estimate of M_{inf} . As discussed later this is appropriate for some preservative systems but the model will have to be refined for others.

To ensure uniform initial preservative component concentration at the start of leaching, the blocks were vacuum treated with water and stored for six days to allow full dissociation of the component(s) before starting leaching. Continuous leaching in one direction only was achieved by sealing all except one face with silicone sealant. Samples were placed individually in 300 ml water and the water replaced periodically. The leachate samples were analyzed for preservative content by ICP and the % of total preservative leached plotted vs. leaching time. For some preservatives, leach water was held at different temperatures (10°C, 20°C, 30°C) to compare T effect on leaching rate. The leaching curves at 20°C were fit with the diffusion model (Equation 2) to estimate D values. For this paper, calculations were performed using a FORTRAN program to determine best fit diffusion coefficients by minimizing the sum of square of differences between experimental and calculated leachate quantities. Since Equation 2 converges rapidly, the calculation could also be done using a spreadsheet to sum the first 8-10 terms of the equation.

RESULTS AND DISCUSSION

The dissolving or dissociation process required some time to reach equilibrium and there were differences in rate of equilibration and extent of component solution in the cell lumen space depending on the preservative system, preservative retention, and ambient temperature. The equilibrium dissociation of copper is shown for a number of wood species and retentions for ACQ treated wood at room temperature in Table 1. For the species compared here, there does not appear to be a species effect as all species have similar amounts of available copper dissolved in the water in the cell lumens and similar % total preservative dissociated for a given preservative retention. However, the amount of dissolved copper available for diffusion increases with preservative retention, although the proportion of total copper available relative to the total in wood decreases with increasing retention. For most preservative components, there is an increase in the amount of dissolved or dissociated preservative with increasing temperature (Table 2). This is especially evident for CCA components and borates. Solubility of copper in the copper amine systems seems to be less affected by ambient temperature.

The rates of dissociation and leaching of fine sawdust properties of typical samples of treated wood are shown in Figure 1 (CCA), Figure 2 (ACQ) and Figure 3 (CA-B). The total amounts available for leaching and the equilibrium dissociation amounts for the preservatives and for different preservative retentions are shown in Table 3.

Table 1 Effect of species and preservative retention on dissociation of copper from ACQ-D treated wood

Species	Concentration ACQ (%)	Retention CuO (kg/m ³)	% CuO concentration in lumens	% copper dissociated	% copper available for leaching
Southern pine	-	3.14	0.085	22	-
Jack pine	0.4	1.32	0.042	22	26
Jack pine	0.8	2.87	0.062	15	14
Jack pine	1.3	4.63	0.087	13	12
Jack pine	1.7	6.27	0.122	14	8
Trembling aspen	0.4	1.59	0.053	23	-
Trembling aspen	0.8	2.95	0.083	16	-
Trembling aspen	1.3	5.6	0.101	12	-
Trembling aspen	1.7	7.62	0.129	13	-
Red pine	0.4	1.26	0.039	22	19
Rd pine	0.8	2.64	0.058	15	16
Red pine	1.3	4.13	0.084	14	16
Red pine	1.7	5.81	0.126	16	20
Spruce	0.4	1.36	0.038	22	22
Spruce	0.8	2.92	0.060	16	13
Spruce	1.3	4.78	0.099	16	19
Spruce	1.7	6.55	0.148	17	15

Table 2 Effect of ambient temperature on preservative component dissociation in southern pine

Preservative	Retention (kg/m ³)	Temperature (°C)	% component dissociated			
			Cr	Cu	As	B
CCA-C	4.3	10	1.5	3.0	3.5	-
CCA-C	4.3	20	3.0	6.5	7.5	-
CCA-C	4.3	30	4.0	6.0	9.0	--
DOT	2.28 B ₂ O ₃	10	-	-	-	75
DOT	2.28 B ₂ O ₃	20	-	-	-	100
DOT	2.28 B ₂ O ₃	30	-	-	-	100
ACQ	3.15 (CuO)	10	-	20	-	-
ACQ	3.15 (CuO)	20	-	22	-	-
ACQ	3.15 (CuO)	30	-	16	-	-
CA-B	1.94 (CuO)	10	-	8	-	-
CA-B	1.94 (CuO)	20	-	10	-	-
CA-B	1.94 (CuO)	30	-	12	-	-

For all preservatives, the finely ground treated wood exposed to water lost a proportion of each component rapidly, to an asymptotically approached maximum indicating that the remaining component was highly leach-resistant (DOT was an exception since virtually all borate could be extracted). In practice, it is unlikely that leaching in service will progress beyond this asymptote and its value provides an upper limit to the amount potentially lost by leaching. Even with this finely ground material it was evident that some components such as Cu in ACQ and CA treated wood and borates leached much more quickly than other components.

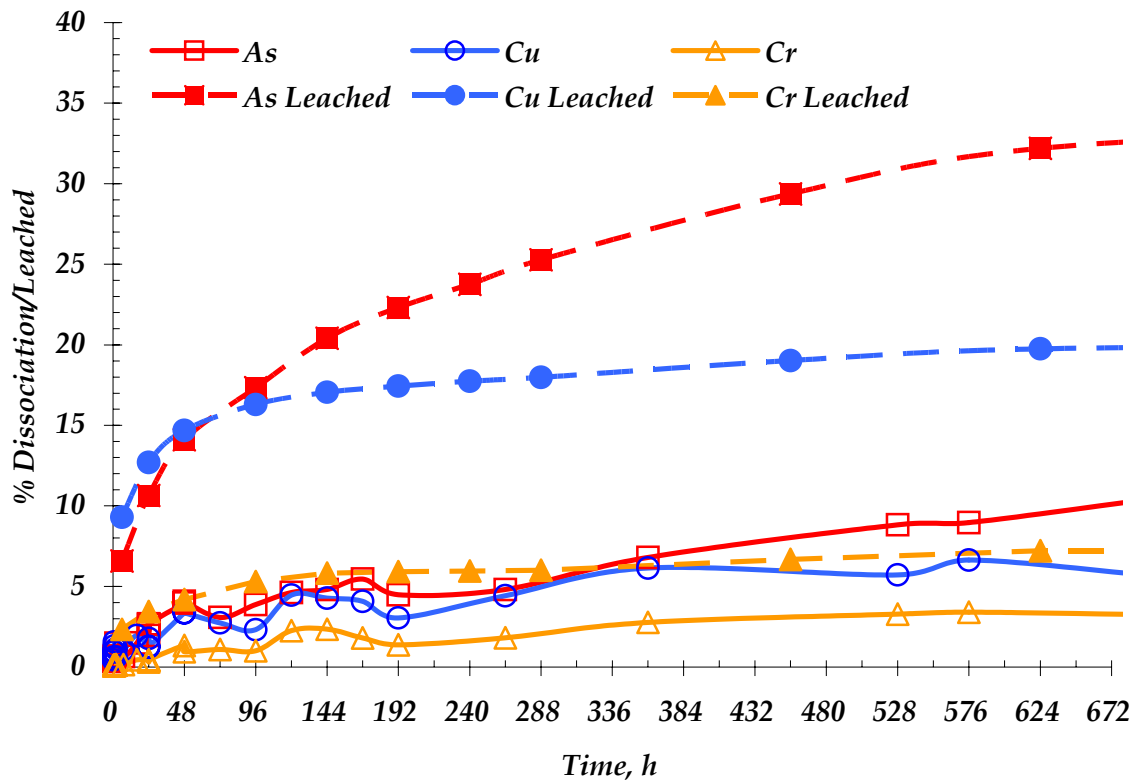


Figure 1 Comparison of rates and amounts dissociated (solid lines) with amounts leached from sawdust (broken lines) for CCA-C components in southern pine

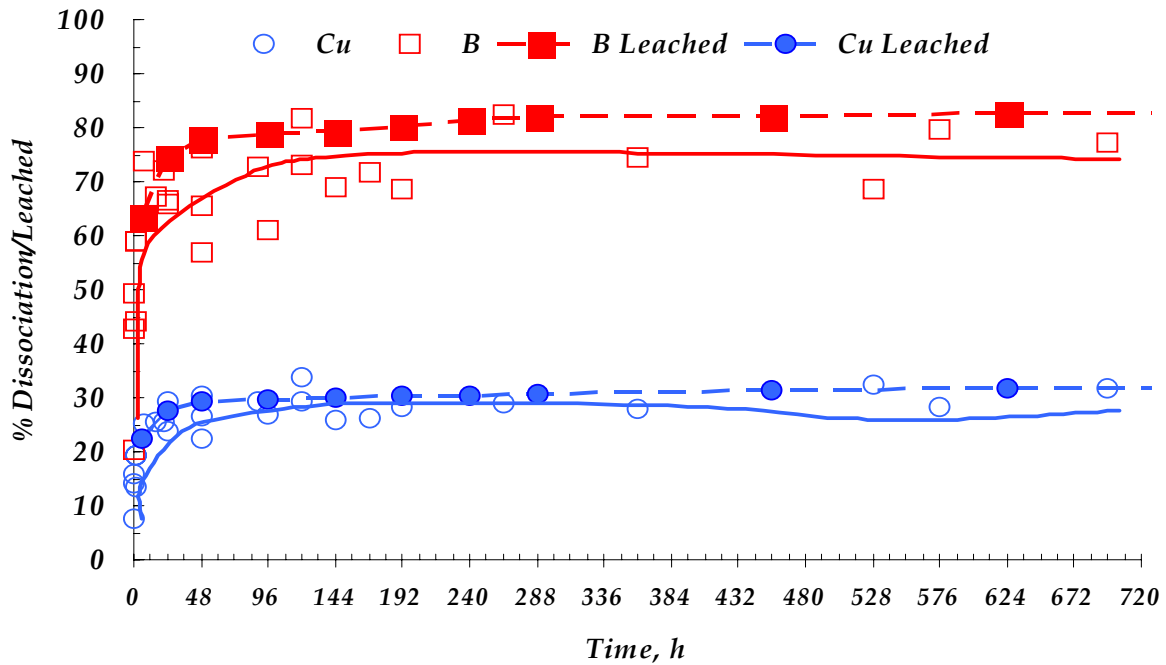


Figure 2 Comparison of rates and amounts dissociated (solid lines) with amounts leached from sawdust (broken lines) for Cu and B components of ACQ-C in southern pine

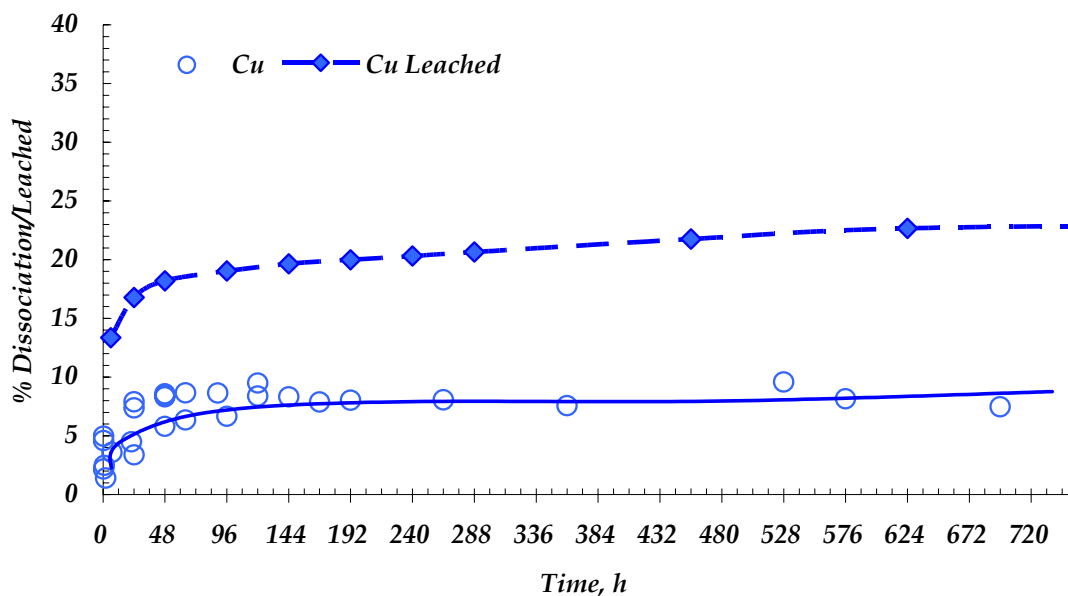


Figure 3 Comparison of rates and amounts dissociated (solid line) with amounts leached from sawdust (broken line) for Cu component of CA-B in southern pine

The rates of dissociation were also different for different preservatives. For example, the more soluble copper in ACQ and borate in ACQ and DOT formulations reached equilibrium sooner than the copper in CA and the CCA components. The arsenic component of CCA was especially slow to equilibrate. If the treated wood undergoes wetting and drying processes, this could significantly affect the leaching rate due to the limited time available for leaching. For CCA (Figure 1) it is evident that the amount dissociated was limited by solubility and it represented only a fraction of the preservative component available for leaching. In this case, the concentration gradient for diffusion will be low resulting in a reduced leaching rate, but there will be a “reservoir” of available preservative that will replace leached components. The net effect should be characteristic slow leaching of CCA components over a very long period of time. As confirmed by numerous studies, the chromium was the least soluble component and these results show that the ultimate amount of chromium available for leaching is also relatively low (7.4 % of total at this retention). The copper component reaches a leaching equilibrium rapidly suggesting that at the retention evaluated here, it will initially leach faster than arsenic, but the rate of leaching will decline faster than arsenic. The arsenic component appears to dissolve and leach slowly but for a long time. The availability of arsenic to leaching is highly dependent on retention in wood [10] and species [11] so these properties will depend greatly on these and other variables.

For ACQ, at the retention evaluated in Figure 2, both the copper and borate components had amounts available for leaching almost identical to the amounts dissolved in the free void space at equilibrium. This means that most of the readily soluble components in the treated wood can dissolve in free water saturating the wood. This creates a steeper concentration gradient during leaching. This was also more or less confirmed for a range of species and ACQ retentions (Table 1, last two columns). This high copper solubility compared to copper in CCA is likely due to free amine dissolved in the cell lumen water.

Table 3: Summary of leaching, dissociation and diffusion parameters for southern pine treated commercially with a number of wood preservatives

Preservative	Retention (kg/m ³)	Total leached from sawdust (%)	Total Dissociated (%)	D _L (10 ⁻⁶ cm ² /s)	D _{Rad.} (10 ⁻⁶ cm ² /s)	D _{Tan} (10 ⁻⁶ cm ² /s)	Quality of fit		
							L	R	T
CCA-As	1.44 As ₂ O ₅	39.5	5.5	1.52	0.20	0.05	P	P	P
CCA-Cu	0.86 CuO	20.4	4.3	0.34	0.05	-	P	G	-
CCA-Cr	2.01 CrO ₃	7.4	2.1	0.77	0.13	-	P	G	-
ACQ	1.82 CuO	34.9	28.1	1.9	0.15	-	M	M	-
CuA	1.21 CuO	26.2	9.3	0.65	0.032	0.002	G	G	P
DOT	2.28 B ₂ O ₃	~100	91.0	0.15	0.05	0.03	M	G	G

M - Moderate; P - Poor; G - Good

At the CA-B retention evaluated (Figure 3), the copper behaved more like the copper in CCA than the copper in ACQ. A much larger amount was available for leaching than was dissolved in the free void space at equilibrium. This is surprising since it is also a copper amine system. The difference is attributed to the lower copper amine concentration compared to the ACQ system, and in fact when higher CA retentions were evaluated, we observed a higher % dissociated (results not shown).

For the DOT treatment virtually all of the borate was available both for leaching and dissolved in the free water in the wood void space. However, the fraction of total B dissolved in the wood cell lumens will be restricted at lower temperatures as seen in Table 2 and at high DOT retentions. This creates a very steep concentration gradient to drive diffusion of boron out of wood during leaching.

It appears that the dissociation characteristics have several important implications:

- Rate of dissolving may be a significant factor in intermittent water exposure conditions where wood dries between exposures;
- In some cases, equilibrium dissociated concentrations in the cell lumens is an indicator of total available preservative component for leaching;
- The high concentration in the cell lumens and reduced concentration at the surface during a leaching event provides the concentration gradient driving the diffusion of components to the wood surface where they are leached away.

The rate of unidirectional leaching is known to depend on both temperature [12,13] and diffusion direction [14]. Preliminary results shown in Table 4 for 20 mm thick, DOT and CA-B samples sealed on all but one face and leached for specific times confirm this. Leaching is much greater along the grain than transversely and in southern pine radial leaching is much greater than in the tangential direction.

Examples of the rate of leaching of 20 mm samples in different directions and the best analytical solution for the diffusion coefficient D assuming M_{inf} is the equilibrium dissociated amount are shown in Figures 4-8.

The diffusion model fits leaching data very well for leaching of Cu and Cr from CCA treated wood in the radial direction (Figures 4 and 5). At the end of the leaching test, the amount of component leached is still much lower than the amount dissociated in the wood so the amount dissociated is a good short term estimate of M_{inf} .

Table 4 Effect of water temperature on leaching of copper from CA-B and borate from DOT – southern pine

Preservative	Water T (°C)	Leaching time (d)	% Component leached		
			Long.	Rad.	Tan.
CA-B (Cu)	10	9	6.2	2.5	0.6
CA-B (Cu)	20	9	10.2	4.2	0.8
CA-B (Cu)	30	9	10.5	4.4	1.1
DOT (B)	10	3	22	12	9
DOT (B)	20	3	26	20	13
DOT (B)	30	3	41	28	19

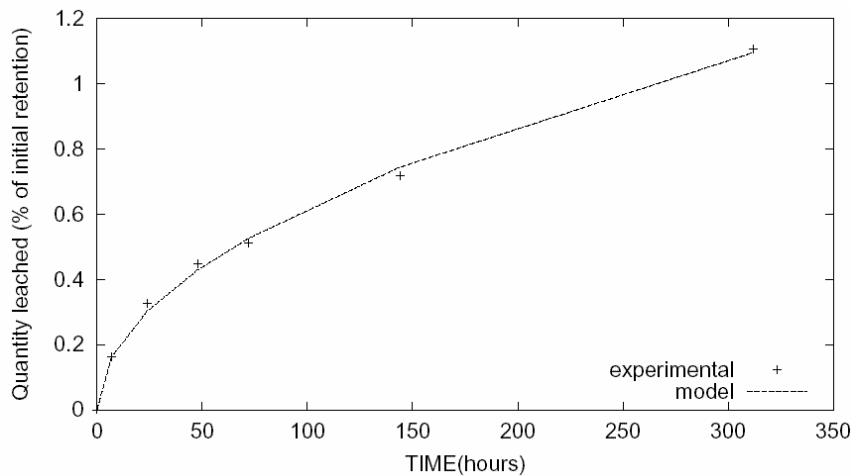


Figure 4 Comparison of leaching values with best fit of diffusion model for copper leaching from CCA treated southern pine – Radial direction

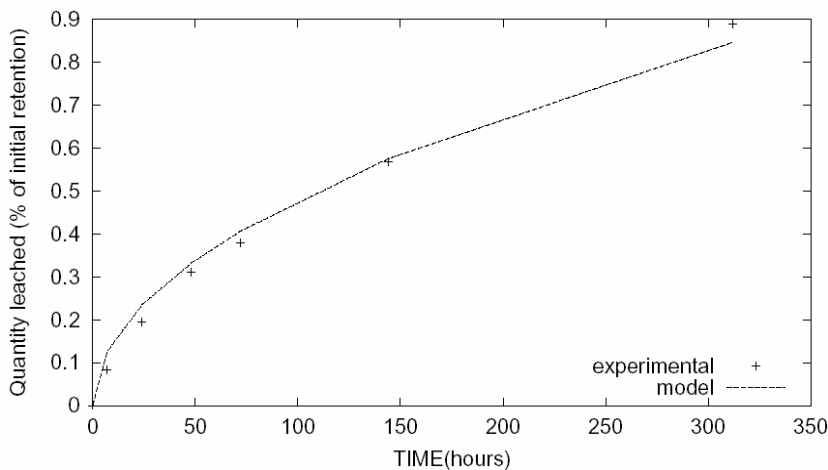


Figure 5 Comparison of leaching values with best fit of diffusion model for chromium leaching from CCA treated southern pine – Radial direction

However, where more vigorous leaching occurs as for arsenic leaching along the grain in CCA treated samples (Figure 6) the model predicts that the leaching rate will level off soon, but this does not occur because of the large arsenic reservoir that can dissolve and leach as the initial dissociated

arsenic is depleted. It will be important to refine the diffusion model in cases where not all preservative available for leaching is dissolved in the wood during the leaching process.

For borate treated samples (Figures 7 and 8) the diffusion model fits experimental values relatively well for both longitudinal and transverse diffusion because the amounts dissociated are approximately equal to the amounts available for leaching (100 % of retention).

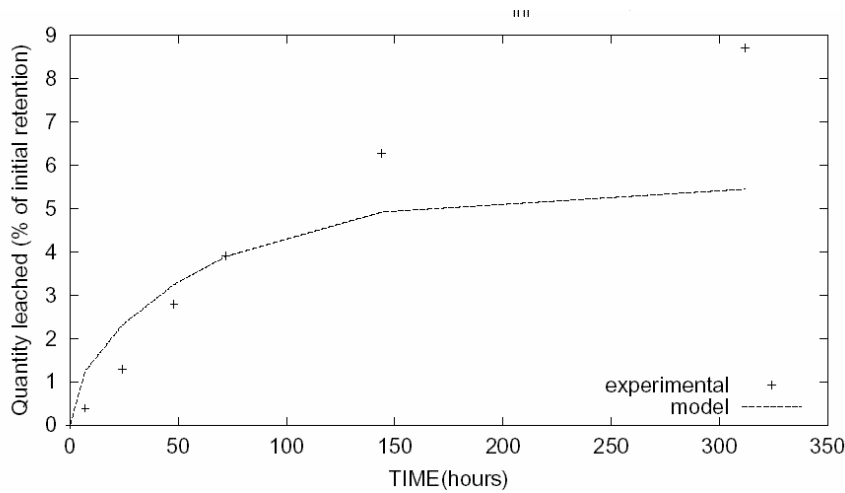


Figure 6 Comparison of leaching values with best fit of diffusion model for arsenic leaching from CCA treated southern pine – Longitudinal direction

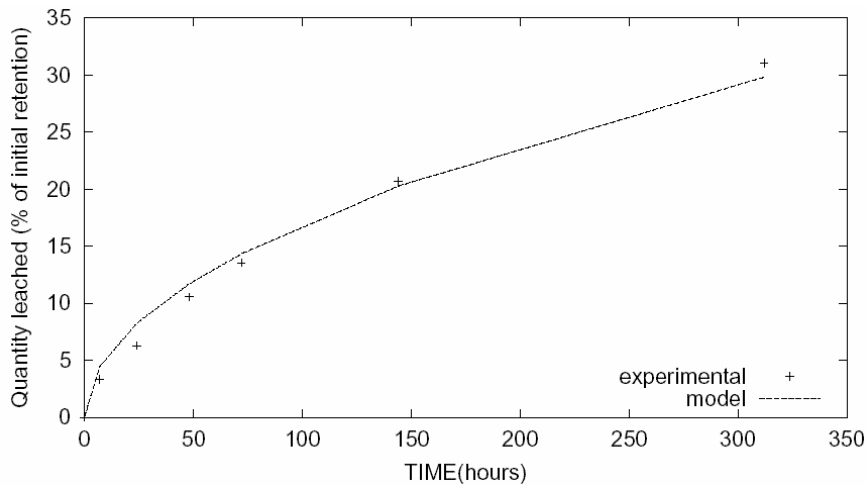


Figure 7 Comparison of leaching values with best fit of diffusion model for borate leaching from DOT treated southern pine – Radial direction

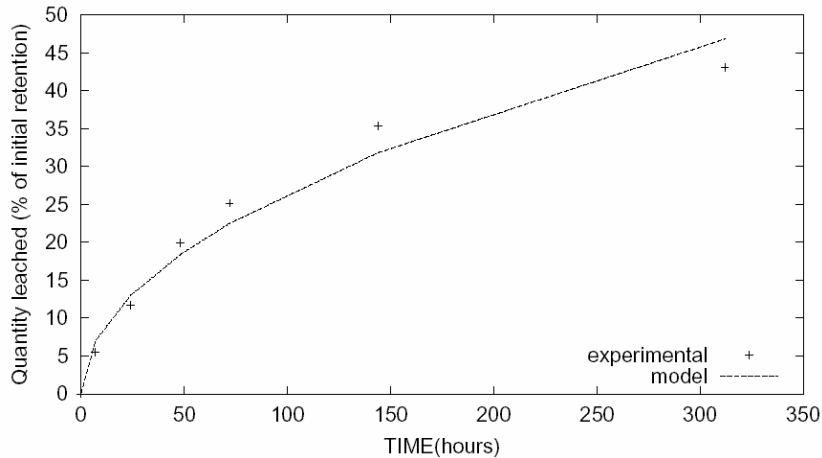


Figure 8 Comparison of leaching values with best fit of diffusion model for borate leaching from DOT treated southern pine – Longitudinal direction

These results show large differences in the inherent diffusion characteristics of the different preservative components and systems. D values were typically greater parallel to grain than across the grain and higher in the radial direction than the tangential direction (Table 3). For borate samples, D values were in the range observed by Ra et al. [14] for boric acid diffusion into southern pine wood.

APPLICATION

Based on the above parameters, it should be possible to estimate the rates and amounts of preservatives that leach from wood under a wide range of conditions. For example, the above parameters were applied to estimate copper and borate leaching from the top surface of southern pine decking (38mm) treated with CCA, ACQ and DOT and *continuously* exposed to rainfall for a 1 year period (Figure 9). For this comparison, dissociated concentration of copper was used in the estimate. It is assumed that all leaching is from the top face of the board. The model predicts that virtually all of the DOT will be lost after 1 year and that about 2/3 of the dissociated copper will be lost from ACQ and CCA treated boards. While these estimates need to be validated, the copper losses from CCA will likely be underestimated since further dissociation of additional leachable copper above the initial dissociation level is not considered. Future work will focus on including the continuing dissociation of these components as leaching proceeds in the diffusion model.

Some Special Implications and Limitations of this Approach:

1. If the above principles are accepted, it is clear that prediction of emissions should be based on time of rain and other water exposure rather than intensity of exposure. This is validated by studies that show that more leaching occurs per unit rainfall by slow steady rainfall than by an intense down-pour [15] and that the amount of leaching occurring over a fixed leaching time is independent of intensity of rainfall [16]. This is unfortunate, since it is more common to record meteorological information as mm rainfall rather than duration of actual precipitation.
2. The models assume that wood is fully saturated with water (dissociation is complete) and the surfaces are in continuous contact with water. Refinements to the model are needed to account for wetting and drying during natural exposure and the effects of moisture content changes on rates of solution of components and wicking of solution to the surface;

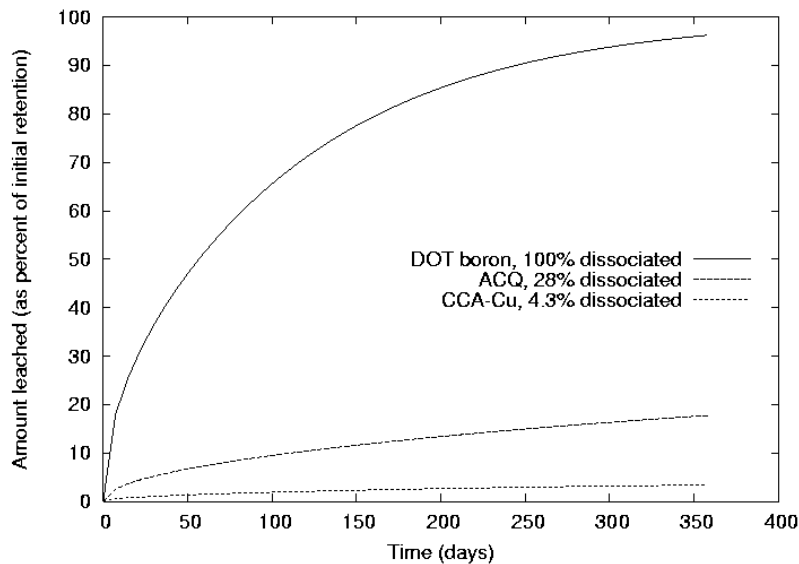


Figure 9: Predicted leaching rate from top surface of 38mm deck board continuously exposed to rainfall

3. The procedure should also be applicable to leaching of organic wood preservatives but it does not predict losses from volatilization or bleeding and these factors would have to be considered in a model for organic substances;
4. No approach, including this one can predict the effects of natural exposure conditions that may affect leaching in an unusual way such as UV exposure [17], exposure to organic and other acids and other aggressive leaching media.
5. At this time, the effect of low solubility of some components such as CCA and CA components relative to their ultimate availability is not taken into account in the simple diffusion models presented here and the long term leaching losses of these components will be under-estimated if the dissociated amount is used in the analysis and the rate of leaching will be over-estimated if the total availability is used.

CONCLUSIONS

An approach is shown to estimate leaching impacts from inorganic preservative treated wood based on laboratory estimation of the amount of preservative component available for leaching, the equilibrium dissociation of preservative into free water in wood and diffusion coefficients in different wood directions. Combining this information with a simple diffusion test allows the estimation of potential risk from leaching over a wide range of product dimensions and specified conditions.

The results predict that ACQ-C will have higher initial leaching rates compared to CCA and CA-B but the latter species will continue to leach copper at a measurable rate for a much longer time.

We plan to continue to investigate the applicability of this approach with particular emphasis on:

- Investigating the rate of replenishment of leached material by continued dissociation and integrating this into a diffusion model;
- evaluating effects of rates of wetting and drying of wood between water exposures and their impacts on leaching;
- Developing models for three dimensional diffusion in rectangular and pole shaped samples.

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