

DEPLETION OF WOOD PRESERVATIVES – SOME ISSUES

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Abstract

This paper focuses on factors that affect leaching losses of wood preservatives in service and the potential for modeling these losses. Many factors affect the leaching of preservatives from wood, including the type and retention of preservative, the conditions of exposure, treating parameters, size and geometry of the structure or component, installation and maintenance practices. Test methods that incorporate diffusion modeling in the analysis have the potential to correct for variables such as preservative type, conditions of exposure, dimensions and wood orientation and provide a scientific basis for predicting leaching under a wide range of conditions.

Introduction

One of the most important attributes of a wood preservative is its stability and persistence when exposed to natural in-service conditions. Depending on the preservative type and formulation, depletion of preservative in service may result from a number of phenomena including leaching, volatilization, bleeding and physical or biological breakdown. Any chemical added to wood has measurable water solubility and the significance of leaching losses in terms of loss of efficacy and potential for health and environmental impacts must be determined by long term testing and risk assessment under the specific application considered. In fact, leaching from contact with moisture is the predominant depletion mechanism and the one most often considered. Leaching evaluation is required as part of the process for adoption of preservatives in preservative standards to provide evidence of long term efficacy of a treated product. Simplified laboratory tests such as AWWA E11 and EN 84 were developed primarily with this objective in mind.

Leaching performance data are now mandated by most regulatory agencies as part of the risk assessment process to determine potential environmental impacts in use. There is increasing recognition that the above tests are not representative of leaching rates in service and there is interest in developing short term laboratory tests that would emulate the environmental loss rates to be applied to risk assessments during the registration or re-registration of wood preservatives. For example, the EU under the Biocidal Products Directive (BPD) requires the evaluation of leaching as a means of predicting potential environmental impacts. The OECD is currently trying to develop standard procedures that would be acceptable to all member states, including the EU (BPD), USA (EPA) and Canada (PMRA). Also, attempts have been made to predict preservative loss rates using empirical or other models to allow results from small scale tests to be extrapolated to real world conditions.

This paper focuses on factors that affect leaching losses of wood preservatives in service and the potential for modeling these losses. A more complete discussion of factors affecting leaching is

available in review articles that discuss the broad spectrum of variables (e.g. Cooper 1994, Lebow 1996, Hingston et al. 2000, Lebow et al 2000).

It should be noted that *some leaching may be necessary or beneficial to the efficacy of the treatments*. Toxic components must be available to denature fungal enzymes (extra-cellular), to be absorbed into fungal tissue or be released from particles ingested by insects to have a toxic effect on the organism. Very low solubility compounds such as copper oxalate have low efficacy against fungi, and some copper-tolerant fungi can render some copper compounds less effective by producing oxalates. Mobility of small amounts of preservative components into checks that develop through the treated zone interfere with spore germination inside checks, contributing to the efficacy of the treatment (Choi et al. 2001, 2002).

Measuring leaching

Most leaching studies are based on test methodologies designed to predict long-term efficacy against decay organisms by estimating the % loss from small specimens after aggressive leaching exposure. This information is not useful for predicting potential impacts of preservative leaching on the environment or human health risk. Leaching studies need to generate information on component loss rates expressed as flux or emission rates (e.g.: $\mu\text{g}/\text{cm}^2/\text{hour}$ or day). These values, in combination with the structure dimensions and the volume of the receiving water, soil or sediment sink allow estimation of the “Predicted Environmental Concentration” (PEC), which can be compared with the “Predicted No Effect Concentration” (PNEC) for risk assessment (Derubaix et al. 2000). The relationship between these two approaches is shown in Figure 1 for leaching from AWP E11 blocks.

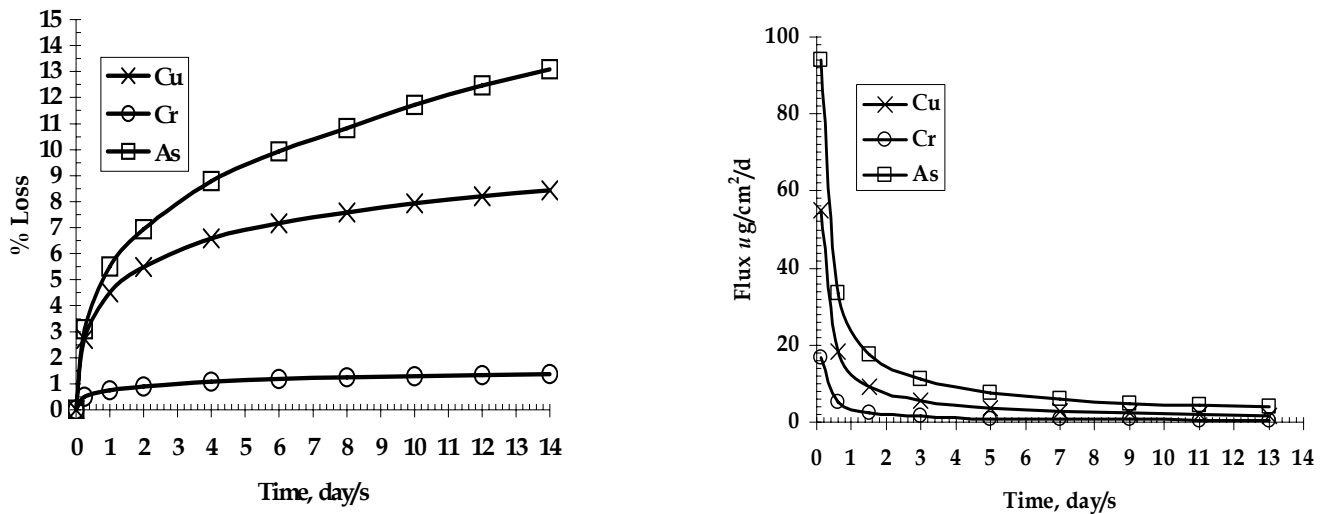


Figure 1.- Leaching of arsenic from CCA-C components from treated SYP ($5.5 \text{ kg}/\text{m}^3$) AWP E11 blocks (Taylor 2001) – Comparison of % loss and leaching rate

The rate of leaching declines with time and the highest potential for health and environmental exposures from direct contact with leachate is during the initial exposure to leaching conditions. Thus, risk assessments, for example for aquatic exposure, focus on this initial flux rate. The “equilibrium” flux rate typifies long-term or chronic exposure effects. The total leached amount (area under the flux rate curve) is indicative of the total possible loading on soil and other compartments of the local environment. The declining leaching rate is often attributed to initial high losses of poorly fixed or unfixed surface components, and this may be a contributing factor, but it also typifies the inherent nature of the diffusion process as discussed later.

Organic wood preservatives

Wood preservatives that are dissolved in oil or other organic solvents or emulsified in water for treatment may be depleted from wood by a combination of mechanisms including leaching, bleeding, evaporation and microbial degradation. The relative importance of the different mechanisms depends on the preservative and solvent properties as well as the in-service application and time in service. Studies on factors affecting the leaching of organic wood preservatives are less common than for inorganic preservatives but there are a number of known or anticipated effects.

Creosote consists of a large number of PAH’s and other components with variable vapour pressures, viscosities and water solubilities so individual components are depleted at different rates and by different mechanisms. For example, creosoted ties that were evaluated for relative losses by evaporation, bleeding and leaching (Chakraborty 2001) showed the relative losses tabulated below. Evaporation was highest for the low MW components, bleeding was more significant for newer ties and leaching was a significant component of losses, especially in older ties.

Component	New Ties			Old Ties		
	Evap.	Leach	Bleed	Evap.	Leach	Bleed
Naphthalene	85	12	3	-	-	-
Acenaphthalene	8	85	7	3	97	0
Phenanthrene	10	78	12	0.2	99.5	0.3
Pyrene	15	80	5	1	98	1

Other organic preservatives are affected by other variables. For example, pentachlorophenol is highly dissociated and water soluble under high pH conditions so it cannot be used under alkaline conditions (including in sea water). Triazoles, such as tebuconazole have very low water solubility and leaching will be limited where water exposure is limited. For example in some standard leaching tests tebuconazole will reach its limit of solubility in the water bath relatively quickly and leaching estimates will be lower than for conditions where the volume of

water contacting it is not limited. Organic co-biocides such as quaternary ammonium compounds that are ion exchanged to wood are likely to be vulnerable to leaching by solutions with high cation content such as sea water and some soils.

Leaching of inorganic (copper based) wood preservatives

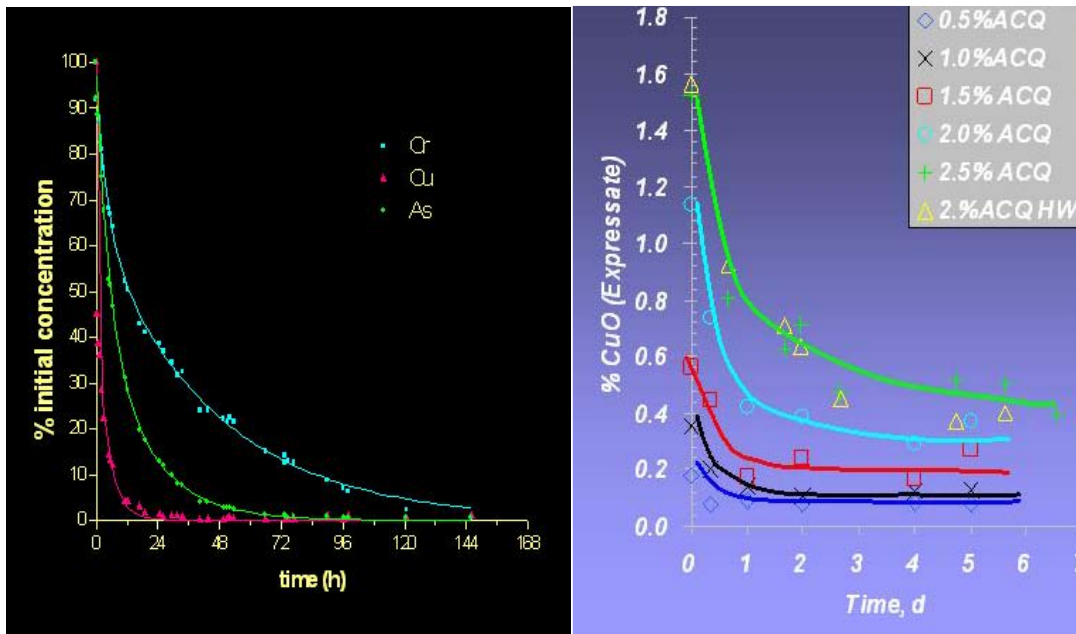
Leaching rates of copper based preservatives are affected by many variables, including:

1. Treatment type, fixation
2. Preservative retention
3. Wood species/type
4. Exposure conditions (soil, water, above ground, wetting duration, product orientation, configuration of structure, UV)
5. Weather conditions (rainfall and temperature)
6. Characteristics of leach water: pH, ionic strength, T etc.)
7. Wood grain orientation and dimensions and checking in service
8. Effects of coatings or water repellents

1. Effect of preservative type and fixation.

Different preservative components and preservative types will respond differently to different variables. Leaching will be higher in wood that has not completed its fixation reactions, and adequate time must be allowed for these reactions to proceed as far as possible.

CCA fixation reactions proceed to a higher level compared to copper in copper amine preservative systems (e.g. Fig. 2a). As a result, initial leaching rates are higher for the copper amine system than for CCA components.



(a)

(b)

Figure 2: Comparison of rate and extent of reaction of CCA components (a) (Radivojevic 2006) and copper component of ACQ (b) at 50°C (Ung and Cooper 2005)

2. Preservative retention effects

Depending on the preservative system, retention can affect both relative leaching rates (e.g. % losses) and absolute leaching (e.g., $\mu\text{g}/\text{cm}^2/\text{day}$). For CCA, the leaching amount, expressed as a percentage of total, declines with increased retention for all three components. However emission or flux rate of copper increases with increasing concentration while that of chromium and arsenic may drop with increasing retention depending on the retention. At very low retentions, arsenic (Figure 3) and to some extent chromium leaching increases drastically. We attribute this to reaction of wood extractives with chromium in such a way that the chromium is reduced, but remains somewhat soluble and does not complex arsenic into normal chromium arsenate fixation products (Radivojevic 2006). The retention effect is very visible in species like red maple, beech and oak, which are characterized by high arsenic leaching at normal retentions (Fig. 5) (Stevanovic-Janesic et al 2001, Radivojevic 2006). This effect is clearly linked to content of sugars and other extractives. In this case there is a strong relationship with chromium fixation rate with rapidly fixing samples having higher arsenic leaching.

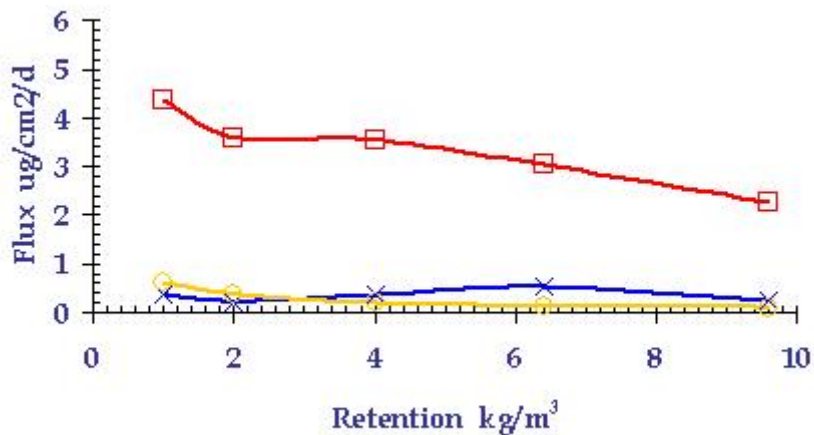


Figure 3: Comparison of flux rates of CCA components at different wood retentions of total CCA – red line arsenic, blue line copper and yellow line chromium (adapted from Jin and Preston 1993)

In the case of CCA, wood contains sufficient reducing groups to reduce the hexavalent chromium component of the treating solution, even at the highest preservative loadings used for marine applications. This reduction is accompanied by precipitation of stable chromium arsenates and oxidation of wood functional groups which simultaneously increase wood's capacity to bind copper by ion exchange. As a result, fixation of all components is high even at high retentions.

In contrast, copper amine based wood preservatives, rely on a smaller number of anionic sites to bind copper and, in the case of ACQ, the quaternary ammonium component. There are insufficient sites to bind all of the copper at high retentions so the degree of copper fixation is low at very high retentions (Fig. 2b). This results in high copper leaching rates under these

conditions (e.g., Figure 4).

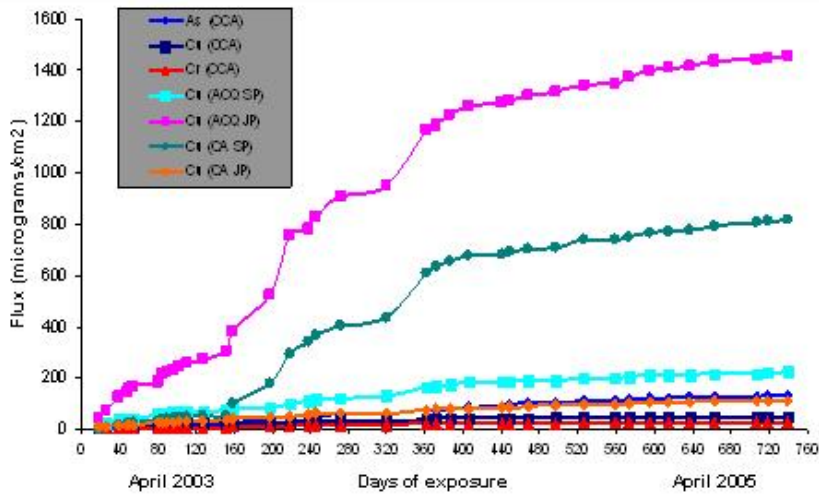


Figure 4: Effect of preservative and retention on leaching of inorganic components. Note ACQ jack pine and CA southern pine were treated to above normal ground contact specified retentions (Stefanovic 2006).

3. Wood species effects

As noted above, the extractive content of wood can have a large effect on the arsenic leaching properties of different species (Figure 5). It would be expected that leaching emissions would be higher from wood species with higher preservative contents (retentions). However, we noted for CCA treatment of well treated southern pine, moderately treated jack pine and poorly treated spruce that during the first year of natural weathering, the emission or flux rates were similar for the three species resulting in much higher % losses from the poorly treated species (Figure 6 – Taylor and Cooper 2005). Most softwood species have similar copper stabilization properties when treated with ACQ, although complexing extractives such as in Douglas-fir may enhance copper stabilization and result in lower copper leaching properties (Ruddick 1996, Ung and Cooper 2005).

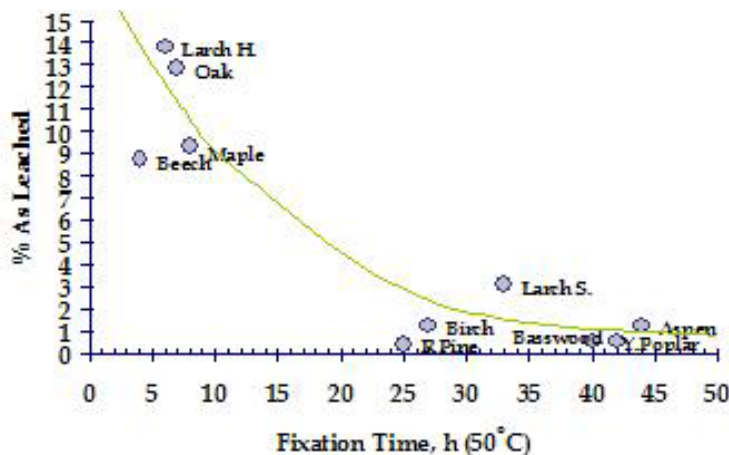


Figure 5: Effect of wood species on arsenic leaching from fixed treated wood (adapted from Stevanovic-Janesic et al. 2001)

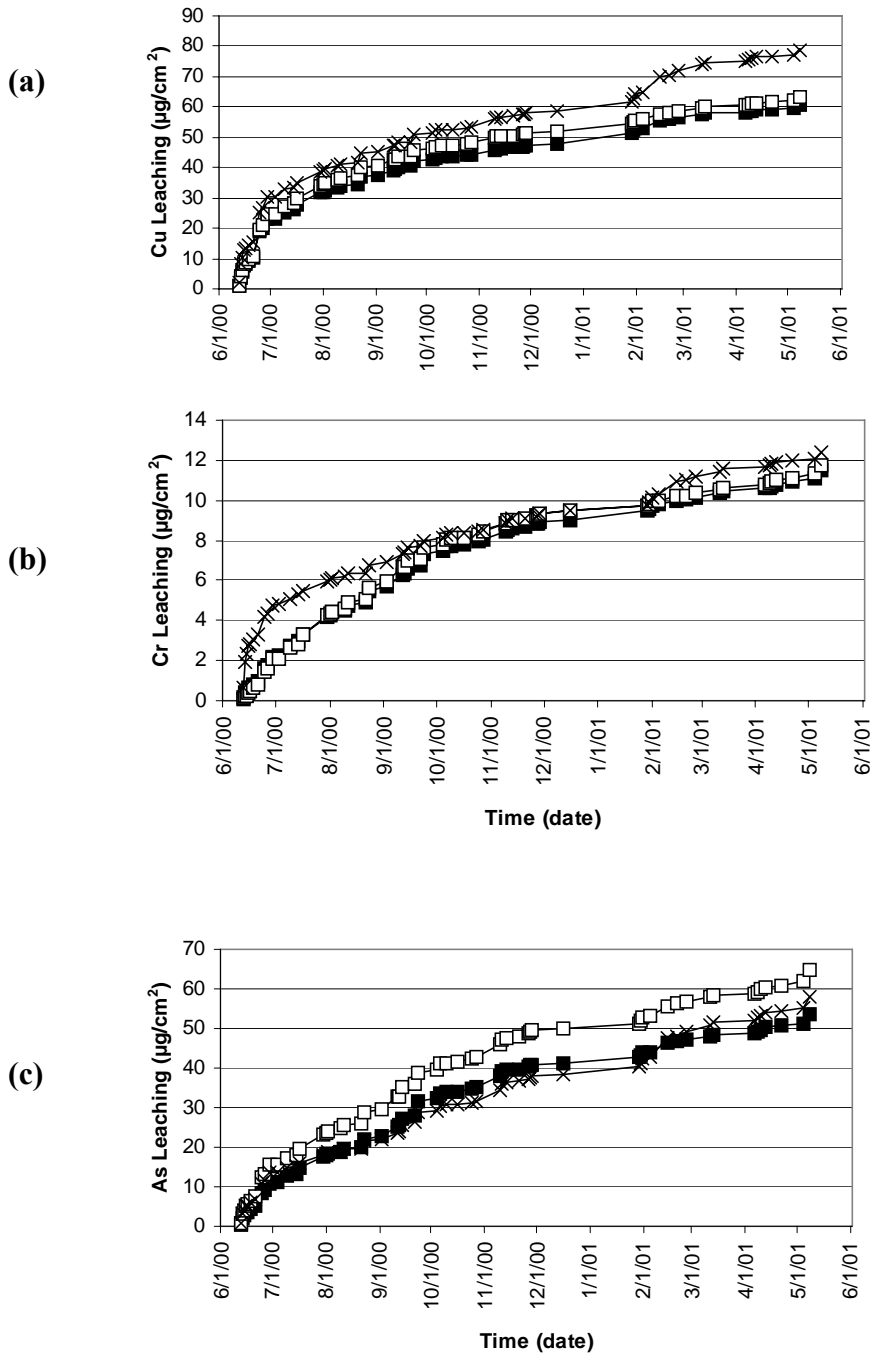


Figure 6: Leaching of CCA components from CCA treated black spruce (□□□), southern pine (x x x) and jack pine (■ ■ ■). (a) copper; (b) chromium; (c) arsenic (Taylor and Cooper 2003)

4. Exposure conditions (soil, water, above ground, wetting duration, product orientation, configuration of structure, UV)

Leaching amounts depend on the design and exposure conditions of a structure. Generally leaching increases according to the following hierarchy of leaching exposures:

- 1. Protected from rain (patios, gazebos)**
- 2. Partial rain exposure (fence boards, siding, deck sub-structure)**
- 3. Exposed to rain (decking, roofing, rails, steps)**
- 4. In soil contact (posts, poles, retaining walls)**
- 5. Fresh water contact (piling, cribs)**
- 6. Marine water exposure (piling)**
- 7. Extreme conditions (cooling tower slats, silos, chemical tanks)**

Leaching caused by precipitation varies depending on the type of structure. For example, vertically exposed wood (such as fence boards and cladding) is subjected mainly to driving rain from only one direction at a time and is less exposed than horizontally applied decking. Wood used in covered structures, such as gazebos, has even less exposure to precipitation. Other structures such as poles have a relatively small surface area in relation to the wood volume and relative leaching is low. In horizontal structures such as decks, the highest soil concentrations of CCA constituents are usually found along the base of the vertical poles and in drip lines of decking.

In situations where wood is continuously submerged in water, leaching with time will respond to changes in ambient conditions and loss of available components. Above ground exposure is much more complex and difficult to predict.

One implication of this complex above ground mechanism is that it is not possible to greatly accelerate the leaching process on full size samples simply by increasing the intensity of rainfall, since the rate is controlled by dissolving of the material between rain exposures and by the rate of component diffusion.

5. Weather conditions (rainfall and temperature)

Leaching in above ground applications depend on rainfall characteristics. As noted above, leaching is more related to duration of rain events than to intensity of rainfall. All leaching processes are somewhat diffusion controlled and diffusion rate increases with temperature and leaching will be higher in the summer than in colder seasons for the same type of water exposure. These factors will necessarily vary greatly depending on the geographic location and climatic variation from year to year. The effect of temperature on leaching of borate in a laboratory test is shown in Figure 7. The change in leaching rate (normalized to rainfall) with the season is shown in Figure 8 for CCA treated samples exposed during one year.

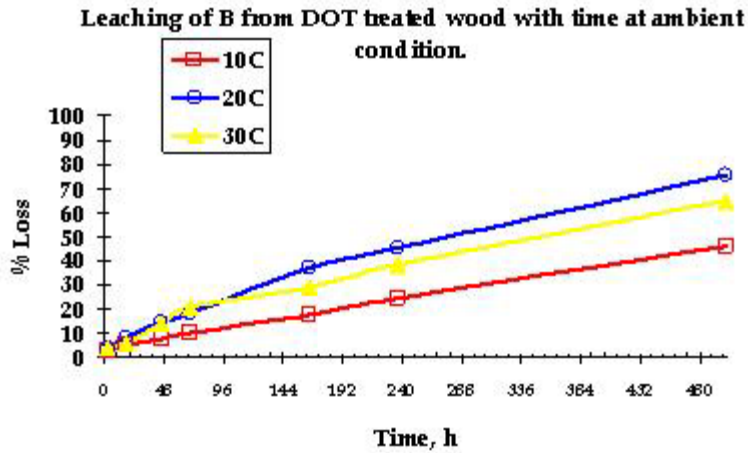


Figure 7: Effect of water temperature on leaching of borate from treated red pine (AWPA E11 samples)

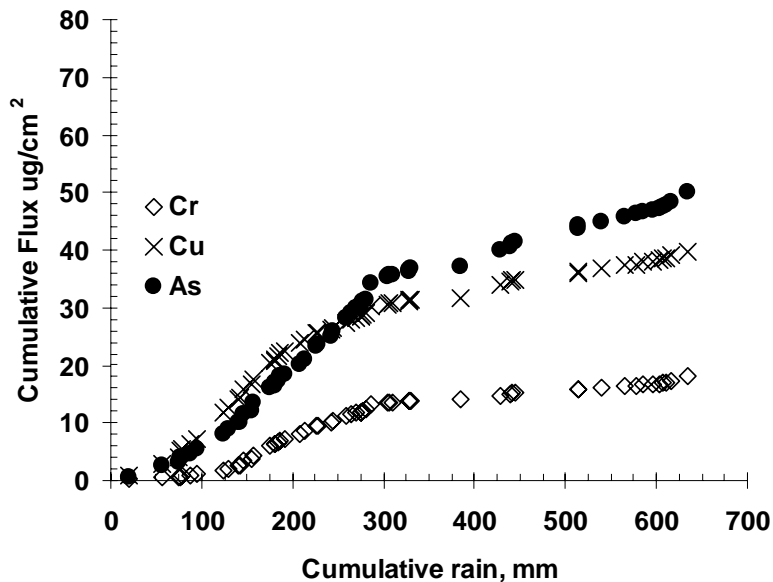


Figure 8: Leaching flux rates of CCA components from samples exposed to natural weathering (June to May). Note change in slope of flux rate for the winter period (Taylor and Cooper 2003)

6. Characteristics of leach water: pH, ionic strength, T etc.)

The potential for acidic precipitation, acid surface water or other acidic conditions to cause abnormal leaching has been considered by many. Persistent exposure to low pH conditions increases leaching of copper from CCA treated wood (Murphy and Dickinson 1990, Kim and Kim 1993, van Eetvelde et al. 1995). Both wood and soil can buffer acid rain effects above and

below ground reducing potential effects of acidic precipitation (Murphy and Dickinson 1990). Radivojevic (2006) showed that leaching of inorganic components of CCA and ACQ was only affected by pH at extreme levels < 3 or > 9 (Figure 9) although she showed that some alkaline solutions at pH 8 or higher oxidize trivalent chromium to soluble hexavalent chromium.

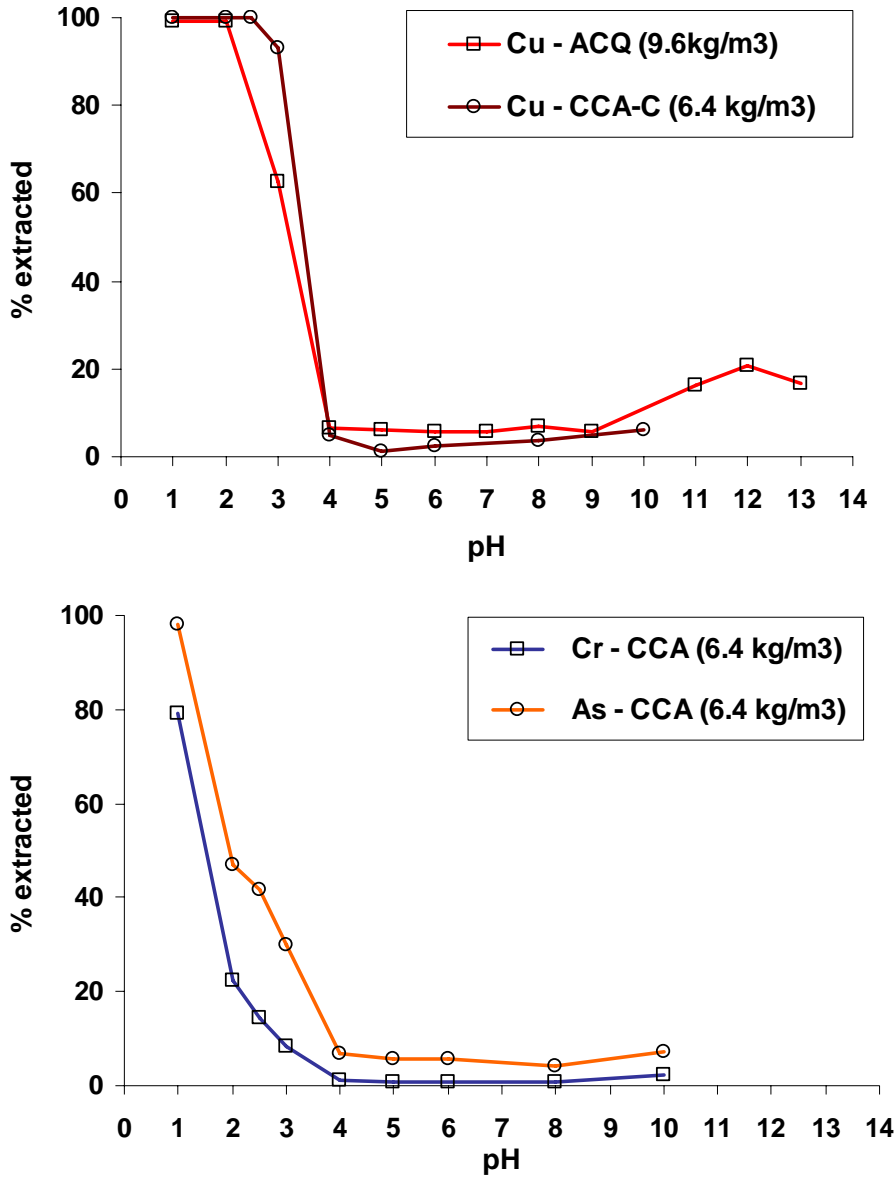


Figure 9: Effects of water pH on leaching of CCA and ACQ components (Radivojevic 2006)

Evans (1987) showed that organic acids in silage (formic, lactic, acetic etc.) extracted a significant amount of all CCA components from treated wood used to construct the silo walls. Similarly, the organic acids produced by the decomposition of organic matter in compost bins

leaches more CCA (mainly copper) than other soil contact (Cooper 1992) as does poorly drained organic acid rich boggy sites (Cooper et al. 2001). Hingston (2002) showed lower copper leaching in marine applications at salinities of 5 or 25 parts per thousand (ppt) with increased copper leaching at 10, 15 and 35 ppt).

For treated wood in soil contact, leaching potential depends on the moisture levels and physical and chemical properties of the soil (Archer and Jin 1994, Wang et al. 1998, Crawford et al. 2002). Fine textured soils (clay/silt) have high water retention ensuring that soil water is in contact with the treated wood, allowing continuous leaching at the surface under moderately wet conditions. Wang et al (1998) found that the highest copper and arsenic leaching was in a poorly drained high organic soil with high cation exchange capacity. Least leaching was from a poorly drained clay soil. Leaching was greater from wet soil than from water for Cu but not for As. Soil exposure may be more aggressive when soils have a strong affinity for the CCA components such as high humic acid soils, high organic and high cation exchange capacity (CEC) soils.

7. Wood grain orientation and dimensions and checking in service

Leaching amounts expressed as % loss for a given leach cycle are higher from wood samples with higher relative amounts of end grain surface. Wood absorbs water more readily through the end grain and rates of diffusion of CCA components are faster along the grain than across the grain. The combination of these factors results in much greater leaching from exposed end grain than from side grain of wood. Furthermore, leaching is faster in the radial direction than the tangential direction (e.g., Figure 10). Thus the grain orientation of a board may have a large effect on its leaching properties.

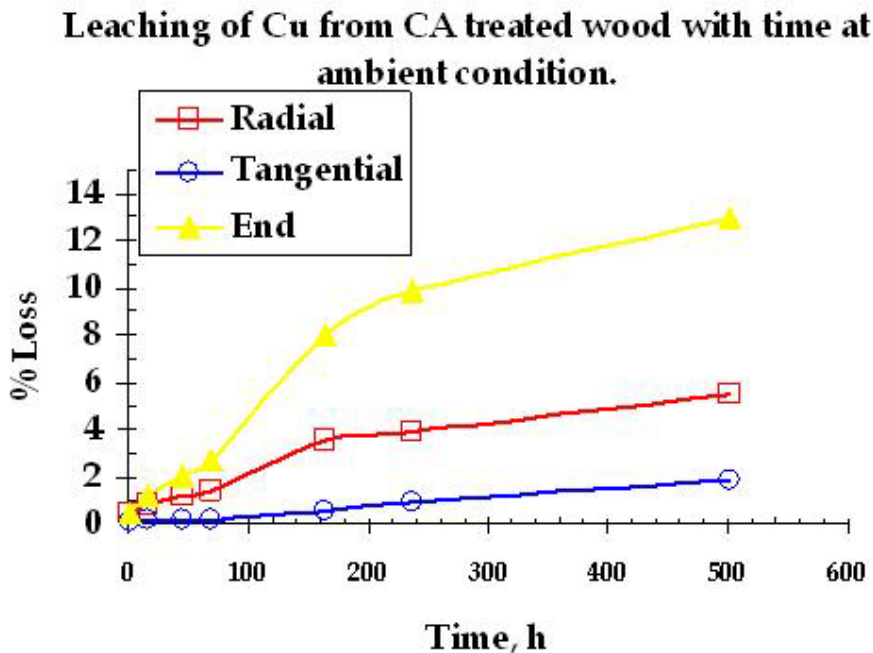


Figure 10: Effect of orientation on leaching of copper from copper azole treated wood

Structures made of many short pieces of wood or wood machined to increase the relative amounts of end grain (such as turned products) will have higher leaching than longer pieces with less relative end grain area.

Leaching rates are significantly higher from small dimension materials and leaching evaluations made on small specimens result in high leaching estimates. The magnitude of this effect can be seen for copper leaching comparisons from a number of laboratory and field tests with CCA-C treated SYP (Figure 11).

Leaching rates can increase dramatically as a result of wood splitting in service (Taylor and Cooper 2003). The open checks increase the surface area being leached and allow faster access to preservative deeper in the wood.

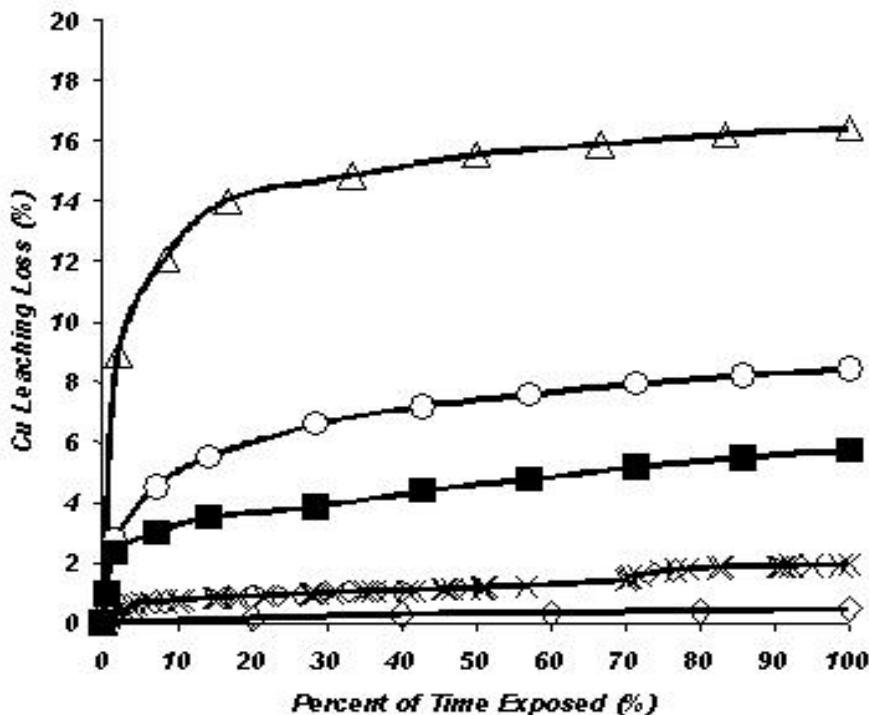


Figure 11: Comparison of leaching of: Δ sawdust 2 weeks; \circ AWPA E-11 2 weeks; \blacksquare EN 84 2 weeks; \times natural weathering (Toronto) 2" X 6" decking one year; and \diamond laboratory water spray 2"X6" lumber 60h.

8. Effects of coatings and water repellents on leaching

Several studies (e.g. Cooper and Ung 1997, Stilwell 1998, Cui and Walcheski 2000) have confirmed that application of a stain, paint or water repellent to treated wood decreases the component leaching significantly. Additional results are shown for penetrating stains in the paper by Nejad and Cooper in these proceedings.

Diffusion modeling of preservative leaching processes

Leaching of preservatives from wood in contact with water is governed to a large extent by diffusion processes and loss of preservative components can be modeled effectively by a “membrane” diffusion model (Waldron et al 2005a,b) shown in its simplest form in the equation below. It is possible to extend this to three dimensions as long as the relative diffusion rates are known for the different orientations (Waldron 2005). For leaching losses in one direction, the mass loss of preservative at time t (M_t) relative to the amount available for leaching (M_∞) is related to the Diffusion coefficient D , the elapsed time of water exposure t and the dimension of the sample in the diffusion direction (l).

$$\frac{M_t}{M_\infty} = \sqrt{4 \frac{Dt}{\pi l^2}}$$

This model takes specimen dimensions and different leaching rates in different grain directions into account and can be extended to apply to three dimensional leaching. It allows prediction of losses during the period that the wood is in contact with water (e.g. continuously for submerged products, or while subjected to rainfall for above ground applications). Application of the diffusion equation allows estimation of the leaching from any configuration of wood as long as information on the amounts available for leaching amounts and diffusion coefficients in the different directions are available. These can be determined for a given wood species and treatment, through a few simple experiments (Waldron et al. 2005b) and the effect of water temperature can also be incorporated in the estimated diffusion coefficients.

An example of the application of such a model (in three dimensions) to estimate the leaching rates from 2X6 pine treated with different preservatives and continuously exposed to a water shower is demonstrated in Figure 12.

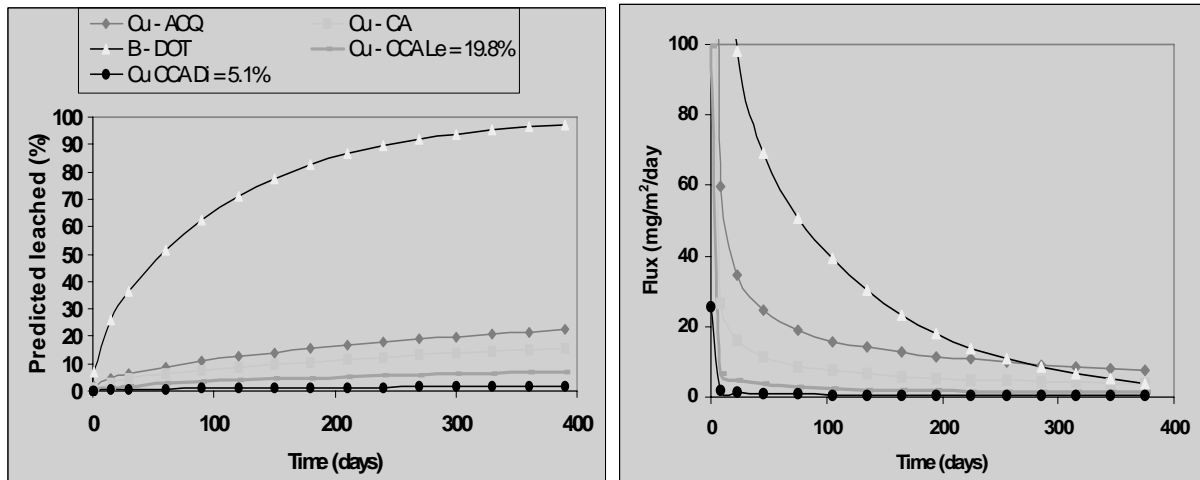


Figure 12:- Modeling of leaching by a three dimensional diffusion model (Waldron 2005)

Summary

Many factors affect the leaching of preservatives from wood, including the type and retention of preservative, the conditions of exposure, treating parameters, size and geometry of the structure or component, installation and maintenance practices. Test methods that incorporate diffusion modeling in the analysis have the potential to correct for variables such as preservative type, conditions of exposure, dimensions and wood orientation.

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