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Characterization of Wood Mulch and Leachate/Runoff from Three Wood Recycling Facilities

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Abstract

Large-scale open storage of wood mulch is common practice at wood recycling facilities. During rain and snow melt, leachate with soluble compounds and suspended particles is released from mulch stockpiles. The objective of this study was to determine the quality of leachate/runoff from wood recycling facilities to evaluate its potential to contaminate receiving waterbodies. Wood mulch (n = 30) and leachate/runoff (n = 26) samples were collected over 1.5 years from three wood recycling facilities in New Jersey, USA. Differences by site were found ($p < 0.05$) for most of the 21 constituents tested in the solid wood mulch samples. Biochemical oxygen demand (range <20 – 3000 mg/L), chemical oxygen demand (134 - 6000 mg/L) and total suspended solids (69 - 401 mg/L) median concentrations of the leachate/runoff samples were comparable to those of untreated domestic wastewater. Total Kjeldahl N, total P and fecal coliform median values were slightly lower than typical wastewater values. Dose-response studies with leachate/runoff samples using zebrafish (*Danio rerio*) embryos showed that mortality and developmental defects typically did not occur even at the highest concentration tested, indicating low toxicity, although delayed development did occur. Based on this study, leachate/runoff from wood recycling facilities should not be released to surface waters as it is a potential source of organic contamination and low levels of nutrients. A study in which runoff from a controlled drainage area containing wood mulch of known properties is monitored would allow for better assessment of the potential impact of stormwater runoff from wood recycling facilities.

Keywords: wood recycling; wood mulch; leachate; water pollution; stormwater runoff; aquatic toxicity

1. Introduction

The amount of woody waste recycled as wood mulch in the USA is substantial, but not easy to determine; the materials come from several different waste categories and mulch production is not separated out among recycling options. As a component of municipal solid waste (MSW), wood wastes (mainly furniture and pallets) in 2011 were estimated at 14.6 million metric tonnes; of this, 2.2 million tonnes of pallets were recycled as either wood mulch or animal bedding (USEPA, 2013). However, the USEPA (2013) also estimated that 30.6 million tonnes of yard waste were generated as municipal solid waste that year, of which 17.5 million tonnes were processed at composting and wood recycling facilities. Falk and McKeever (2004) earlier estimated that 58% of yard waste was woody material (wood chips, stumps, tree tops, logs, brush). Further, there are other wood wastes, such as land clearing debris, that often ends up being recycled as mulch, as well as construction and demolition waste, which normally does not.

Facilities where wood is processed and stockpiled include wood mulch recycling facilities, composting facilities, log yards, log storage and sorting areas, saw mills, de-barking and bark pressing operations, and pulp and paper mills. The products and residuals stored at these facilities include sawdust, wood chips, logs and bark, thus representing a wide range of particle sizes. Based on Canadian studies, leachate from logs and wood waste has the potential to be acidic, nutrient poor, toxic to aquatic life and have very high oxygen demand (Taylor et al., 1996; Tao et al., 2005; Tao et al., 2007; Taylor and Carmichael, 2003; Woodhouse and Duff, 2004). However, the contaminant levels in these studies varied widely ranging from no toxicity and minimal contaminant loads to high toxicity and loads. This disparity is likely due to the variability of source, size and type of wood being handled as well as the frequency, intensity, and duration of the precipitation. Kaczala et al. (2012) observed higher amounts of organic carbon and chemical oxygen demand (COD) released from pine saw dust compared to oak wood chips and suggested that it was mainly due to the smaller particle size resulting in a higher surface area to volume ratio. Similar results were reported by McLaughlan et al. (2009), while Svensson et al. (2013), concluded that dissolved organic carbon released in leachate was six times higher from pine saw dust than from pine wood chips. However, the type of tree also can be important. Aspen (*Populus tremuloides*) leachate, for example, is known to be particularly toxic (Taylor et al. 1996).

Site characteristics, such as pavement, also play an important role in the quantity and quality of leachate/runoff. Fikart (2002) showed that more runoff was generated at a paved log yard compared to an unpaved one during the same rain event. However, total suspended solids (TSS) levels tend to be higher at unpaved sites since runoff can include soil in addition to wood particles (McDougall, 1996).

The part of the tree stockpiled at a facility also may be an important determinant of leachate/runoff quality. Bark leachate is darker in color, has more dissolved organic matter, and is more toxic compared to stem wood leachate (Field et al., 1988). Svensson et al. (2012) indicated that bark is one component of the tree that could be potentially hazardous in the aquatic environment.

Size of the log yard and volume of wood stored also affect leachate quality. McDougall (2002) reported that for five log yard sites, the runoff from the site with the highest ratio of logs stored to yard area consistently showed the highest median phenolic compound concentrations. McDougall (2002) also noted that increased runoff observed during long, high intensity rain events had increased potential to carry suspended solids and other contaminants from log yards.

Generally, stormwater dilutes the leachate; however, it also can contribute pollutants (e.g., from vehicles).

According to the New Jersey Department of Environmental Protection (NJDEP), 0.63 million tonnes of brush and tree parts and 0.075 million tonnes of stumps were recycled in New Jersey in 2011. Data do not exist, but it is assumed that the majority was processed in 68 Class B recycling facilities (NJDEP, 2015a). Class B recycling facilities with wood recycling activities receive, store, process and transfer source-separated wood materials that are non-chemically treated and unpainted, such as "whole trees, tree trunks, tree parts, tree stumps, brush and leaves provided they are not composted" (NJDEP, 2015b). Wood is typically ground twice (and sometimes colored) before being sold as wood mulch. First, the wood material (often with bark attached) is coarsely ground and stockpiled in the open (Grind 1), then more finely ground (Grind 2). The particle sizes of Grind 1 and 2 materials are highly variable over time and also among sites. Usually, wood material is stockpiled as Grind 1 and ground the second time just before it is sold. However, this practice is not strictly followed and storage times vary yearly and among facilities. In general, grinding and sale of wood mulch is more rapid in spring, while in winter wood often is left on site unground or as Grind 1 material. Since the wood mulch is stockpiled in the open, during rainstorms and snow melt, soluble compounds are leached and particles can be carried off site as runoff.

In July of 1999, two reported incidents of water pollution appeared to result from wood recycling facility operations in New Jersey (NJDEP, 2009). The first involved a fish kill and an outbreak of avian botulism in a lake receiving runoff from a wood recycling facility. The runoff leaving the facility was found to have high biochemical oxygen demand (BOD > 2500 mg/L), chemical oxygen demand (COD > 5400 mg/L), and nutrient loads. In the second incident, *Sphaerotilus* (filamentous bacteria previously called “sewage fungus”) growth was observed in a stream receiving runoff from a wood recycling facility.

Although yard waste composting, a process with some similarities to wood recycling, may produce leachate that requires management (USEPA, 1994, Krogmann and Woyczechowski, 2000), no previously published study on the adverse impact of wood recycling on receiving waters was found in the literature, making it difficult to develop science-based regulations. The work reported here represents the first part of such a study. The focus is specifically on leachate/runoff quality associated with operating wood recycling facilities producing mulch, with the objective of evaluating its potential to contaminate receiving waterbodies and, if necessary, serving as a basis for the development of management practices to reduce potential pollution. Samples of solid wood mulch and leachate/runoff were collected from three Class B wood recycling facilities in New Jersey over a 1.5 year period. The leachate/runoff samples were analyzed for typical water pollutants as well as for “tannin/lignin/phenolic” compounds (TLP), which might be considered indicative of wood wastes. Additionally a zebrafish embryo assay was chosen to examine toxicity, in part because it can detect developmental effects as well as acute mortality. As a result of site layout and operations, leachate that had passed through wood mulch piles and runoff at the study sites that had not passed through piles were mixed in unknown ratios. Therefore, water samples collected at the sites are assumed to have originated from a combination of both sources and are identified as leachate/runoff.

2. Materials and Methods

2.1. Site characteristics

The two main factors used in selecting wood recycling facilities for this study were the sites' compliance with NJDEP regulations and willingness of personnel at a facility to participate and provide assistance. Additionally, sites were sought in which leachate/runoff from the wood recycling activities and other operations were separated, and at which different site characteristics were represented. The three selected NJDEP Class B facilities are located in different geographical regions of New Jersey.

Site B, located in the coastal plain, accepts concrete, brick, and asphalt in addition to wood, but leachate/runoff from those operations discharges into a separate retention pond. However, leachate/runoff from leaf composting located on site flows into the retention pond for the runoff from the wood recycling activities.

Site C, in the piedmont region, grinds leaves and prepares and sells top soil in addition to wood mulch. The site does not have a retention pond. Among the three sites, site C had the highest ratio of volume of wood mulch stored to area of the recycling facility.

At Site D, in the highlands region, leaves were not mixed into wood mulch but were used in top soil preparation. Runoff from wood mulch and leaf stockpiles were directed to the same retention pond, which also received runoff from a steep slope covered with dense vegetation.

2.2. Wood mulch sampling and analysis

A total of 15 representative wood mulch samples were collected in duplicate from among the three sites on seven dates from October 2010 to April 2011. All samples were collected according to the protocols specified by the US Composting Council (2010). After opening with a front end loader, samples were collected from the face of a pile on three equidistant horizontal planes, with the lowest and highest being 1 foot (0.3 m) from the ground and 1 foot from the top of the pile. A total volume of 76 L of sample was collected in the ratio of approximately 3:2:1 (bottom: middle: top). The collected material was well mixed before coning and quartering twice, each time discarding one randomly chosen quarter. Approximately 4 L was placed in a plastic bag and shipped overnight in a cooler to the Agricultural Analytical Services Laboratory (AASL), University Park, PA, USA for analysis. A portion of the remainder was used immediately to determine the moisture content (ASAE, 1992) for comparison with the shipped sample.

2.3. Leachate/runoff sampling

A total of 26 leachate/runoff samples were collected (Table 1) either during or within 24 h of the occurrence of 13 different rainfall events. In general, samples were collected from up to three locations depending on availability at each facility: 1) a puddle immediately next to a Grind 1 and/or Grind 2 pile, 2) runoff flow leaving a pile area (not used for site D), and/or 3) an infiltration pond located on the site (none at site C). Samples were refrigerated and analyzed within the method-specific holding times. Unlike wood mulch samples, leachate/runoff samples could not be differentiated into samples collected from Grind 1 and Grind 2 piles.

2.4. Leachate/runoff analysis

2.4.1. Physical, chemical and microbiological analysis

Leachate/runoff samples were analyzed for pH, biochemical oxygen demand (BOD), chemical oxygen demand (COD), total suspended solids (TSS), settleable solids (Sett.S), total Kjeldahl nitrogen (TKN), total phosphate-phosphorus (TP), tannin/lignin/phenolic (T/L/P) compounds, and fecal coliforms. COD and T/L/P analyses were carried out using a HACH kit (Hach Company, Loveland, CO), followed by spectrophotometric measurements (Spectronic®

20 Genesys™ for COD, Hach DR/850 for T/L/P). All other tests were conducted following Standard Methods for the Examination of Water and Wastewater (Clesceri et al., 1998). Method 5210 was followed for the BOD test and Method 2540 D for TSS. For TSS determination, at least 50 mL of sample was vacuum filtered in most cases and a smaller volume was used only if filtration took more than 10 minutes. Method 4500-N_{org} C without distillation followed by Method 4500-NH_3 D (electrode) for ammonia measurement was used for TKN. For TP, Method 4500-P B (digestion) followed by Method 4500-P E (ascorbic acid colorimetric determination) was employed. Fecal coliform testing, performed using a 5 tube per 10-fold serial dilution multiple tube fermentation procedure was done only on samples from 2011 (n=12). The settleable solids test also was discontinued once values were seen to routinely be below detection (<0.5 mL/L).

2.4.2. Aquatic toxicity dose-response studies

Dose-response studies using 8-24 h old zebrafish (*Danio rerio* strain AB) embryos were conducted based on McCormick et al. (2011) using approved protocols (Cooper, 2009). Dilutions (0.625 – 50% leachate/runoff in dechlorinated, filtered tap water passed through an activated carbon column) were chosen based on a visual estimation of sample strength. Except for the first sample (C-2 from April 5, 2010), dilutions were adjusted to pH 7 using 10 N HCl or NaOH, and 10 mL portions of each were transferred to 20 mL scintillation vials. Five embryos were transferred into each vial, and three vials (15 embryos total) used for each concentration and for controls with dilution water only. After recording the stage of each embryo (based on Kimmel et al., 1995), all vials were incubated at 25ºC. Observations were made daily and any abnormal development or mortality was recorded until the control embryos reached the "protruding mouth" post hatch stage (usually 5 days). Abnormalities looked for included: 1) abnormal circulation, 2) pericardial edema, 3) yolk sac edema, 4) kink tail, 5) abnormal spine curvature, 6) decreased body and retinal pigmentation, and 7) size of head. Percentages reported are based on the survivors at each time period.

The LC_{50} (lethal concentration of leachate/runoff for 50% of exposed embryos by the end of incubation period) was determined by plotting percent mortality (probit scale) vs. log leachate/runoff concentration and manually drawing the best-fit line (Reish and Oshida, 1986). EC_{10} (effective leachate/runoff concentration at which a specific developmental defect was observed in 10% of exposed embryos) was determined similarly. The concentration at which there was a one stage delay in development compared to controls was determined by assigning a numerical value in increasing order to each stage (Kimmel et al., 1995) before the final "protruding mouth stage", which is expected to be achieved by healthy embryos in five days after fertilization. A numerical value was also assigned to controls in which the final protruding mouth stage was not reached. This value was subtracted from the treatment values to determine "delay". The weighted (by percentage of surviving fish) averages of the delays among embryos at each concentration compared to the control were then used to determine (by linear regression) the concentration at which there was a one stage delay in development.

2.5. Statistical analysis

One-way and two-way unbalanced analyses of variance (ANOVA) were performed for wood mulch and leachate/runoff samples using SAS 9.3 (SAS Institute Inc. Cary, NC). A post-hoc Tukey-Kramer test was conducted to determine the means that were significantly different from each other. Coefficients of determination (R^2) obtained from simple linear regressions using Excel 2007 were used to test correlations between values of leachate/runoff parameters.

3. Results and Discussion

3.1. Wood mulch

A two-way unbalanced ANOVA (using site and grind) indicated that among the 20 parameters tested (ammonium omitted because most values were below detection), 17 showed significant differences ($p < 0.05$) in concentrations among the three sites (Table 2; additional results are given in the Supplementary Materials, Table S1). Values of electrical conductivity and sulfur concentrations also differed significantly with grind of the material, and electrical conductivity and lead differed based on the interaction between site and grind.

These differences among sites indicate differences in the material being processed, the processing itself, and/or site soils and topography. Inorganic constituents of wood are highly variable as they are dependent on tree species, soil chemistry and climatic conditions (NCASI, 2005). Concentrations also change with time as the tree undergoes chemical and biological degradation after harvesting (Samis et al. 1999), although in this case, because wood mulch was repeatedly added and removed from these operational piles, samples taken later in the study do not necessarily represent older materials; hence aging of the material (and its effect on leaching, as well) could not be evaluated.

The analysis showed that there was no effect of grind for most of the parameters. Because of the 5% probability of finding a significant difference even when there is only random variation (at 95% significance level), the "significant" differences involving grind observed for 3 (electrical conductivity, sulfur, and lead) out of 20 tests (15%) should not be over-emphasized without confirmation from additional testing.

Table 2 also shows some analyses of wood from the literature. This comparison suggests important differences between the mulch samples and wood. In particular, the mulch has a much lower organic matter content and much higher iron and calcium concentrations, suggesting the likely incorporation of considerable soil (aluminum is also high). This could be from site activities, as well as from bringing in woody material with soil entrained. Decomposition of some wood mulch organic matter occurs during storage, as indicated by strong self-heating, but this does not fully account for the lowered levels. (Note that a 50% loss of wood organic matter would only produce a drop from 98% to ~96% organic material.) The higher levels of nitrogen and phosphorus in the mulch might reflect the presence of leaves incorporated during grinding.

Wood mulch samples were slightly acidic with the average value ranging from 5.8 to 6.8 among the three sites. Total nitrogen was almost entirely in the organic form with ammonium-nitrogen concentrations usually below detection. Samples from Site B had significantly ($p < 0.05$) lower electrical conductivity, N, P, K, Ca, and Mn (Table 2). Site C samples had higher organic matter and C, but lower Fe. Site D had the highest P, Mg, S, Na, Al, Fe, Mn, and Pb, but the lowest organic matter levels. The higher metal concentrations (especially Al and Fe) and lower organic matter observed in Site D samples could be due to greater "contamination" of the mulch with soil. In all cases, concentrations of the 5 regulated metals were well below the standards used in site remediation (Supplementary Table S2).

The moisture content of the samples (wet basis) varied from 31% to 60% with an average value of 47%. The average moisture content was slightly higher for site C compared to the other two sites. As might be expected, there was no significant correlation of the 20 mulch parameters tested with the geometric mean diameters (particle sizes) calculated in the laboratory (Subroy et al., 2014) for the 30 mulch samples.

3.2. Leachate/runoff

3.2.1. Physical, chemical and microbiological analysis

Table 3 summarizes the results of the physical, chemical and microbiological analysis of the leachate/runoff samples, which were highly variable, as well as typical values for sewage and urban stormwater. Standard deviations along with comparisons to some additional materials are provided in the Supplemental Materials (Table S2). In general, BOD, COD, and TSS concentrations were comparable to those of untreated domestic wastewater (raw sewage), although both lower and higher individual values were observed. TKN, TP and fecal coliform values were usually slightly lower than is typical for untreated domestic wastewater. BOD, COD, TSS, TKN, and TP median values were all much higher than for typical urban stormwater runoff, while fecal coliform levels were lower. These findings are similar to those reported by Hedmark and Scholz (2008) in their review of runoff mainly from log yards. It might be expected that grinding and the subsequent microbial activity that occurs in mulch piles would contribute to greater leaching compared to sites with more intact wood, but this is difficult to confirm amidst all of the variability. Likewise the effect of on-site composting of leaves cannot be separated from the wood mulch only effects, although samples right next to piles (especially B-1 and D-1) are likely mainly affected by the mulch.

Figure 1 shows box and whisker plots for the leachate/runoff results. Long whiskers indicate high variability. Potential sources of variability that were not included separately in the statistical analysis include the specific materials handled on site, their age and processing method, storm intensity and duration, and the site geology and hydrology. A log transformation was used to normalize the data and a one-way ANOVA (each site/location considered separately) was run with the Tukey-Kramer test to determine which means differed. The analysis found no significant differences except for pH.

The pH values of the leachate/runoff samples ranged from acidic (4.8) to slightly basic values (7.6) with a median of 6.8. Others (Samis et al., 1999; Tao et al., 2007) have found leachate from wood materials to be acidic. The lower pH values for C-1 samples may also reflect the leaves at the site, and mulch from Site C had the lowest average pH, although the differences were not found to be significant ($p < 0.05$).

BOD ranged from below detection (<20 mg/L) to greater than 2,900 mg/L while COD ranged from 130 to 6,000 mg/L. The high oxygen demand presumably is due to the oxidation of the intermediate products from the decomposition of the wood, which include a mixture of tannins and lignins, volatile fatty acids (C2-C6), and carbohydrates in the form of cellulose and hemicelluloses (Tao et al. 2005; Taylor et al. 1996; Taylor and Carmichael, 2003; Woodhouse and Duff, 2004; Zenaitis et al. 2002). Higher BOD and phenol concentrations, both natural products of decomposition, also are seen with composting of leaves (Richard, 1996). Low BOD/COD ratios indicate a low biodegradation rate for the organic matter in the leachate/runoff and the relatively low proportion of easily biodegradable compounds. Lignins, the major non-carbohydrate constituents of wood, are highly recalcitrant (Komilis et al. 2003). Tannins, lignins and phenolic compound concentrations ranged from 6 to 690 mg/L. Again, some of the high BOD and COD values observed for C-1 samples may arise from the leaf composting operations.

TSS concentrations in the leachate/runoff samples ranged between 70 and 400 mg/L. The TSS appeared to consist of mainly wood and bark particles, although soil also was likely present. Most of the studies done on log storage and handling facilities have reported low TKN and TP concentrations in the leachate (Hedmark and Jonsson, 2008; Jonsson et al., 2004 and 2006). In this study, TKN concentrations ranged from 1 to 70 mg/L and phosphorus concentrations from 0.18 to 8 mg/L. Interestingly, TP values tended to be lowest in leachate/runoff samples from Site

D, even though mulch samples from that site had the highest phosphorus concentrations. Fecal coliform concentrations ranged from below detection (< 450) to greater than 1.6×10^6 most probable number (MPN) per 100 mL.

3.2.2. Aquatic toxicity dose-response studies

Lethal concentration (LC_{50}) values for mortality and effective concentration (EC_{10}) values for developmental defects were calculated for each sample where possible. The results are shown in Table 4.

The relatively high mortality at low sample concentrations observed in the first tested sample, C-2, can likely be attributed to low pH (3.44). Natural waters would typically be better buffered than the dilution water used, so that addition of stormwater runoff would not have such a dramatic effect on pH. For this reason, all later samples were adjusted to pH 7. In the other samples, where pH was adjusted to neutral, mortality and developmental effects, if observed at all, occurred at higher leachate/runoff concentrations. The developmental effects observed were yolk sac edema, abnormal spine curvature, kink tail, and delay in development. Only delay in development regularly occurred at lower concentrations.

Numerous authors (e.g., Masbough et al. 2005, Peters et al. 1976, Tao et al. 2007, Samis et al. 1999) have reported that tannin, lignin, and other phenolic compounds in wood or produced by its decomposition can be directly toxic to aquatic life. However, little acute toxicity other than the one case attributable to low pH was observed in the mulch leachate/runoff samples, and no correlation was found between delayed development and T/L/P (see next section)..

In addition to direct toxic effects, indirect toxicity can occur when high concentrations of organic constituents decrease dissolved oxygen in aquatic environments, or when release of highly colored compounds decreases light penetration and thereby obstructs photosynthesis.

Wood leachate also appears to support abundant growth of the filamentous bacteria *Sphaerotilus,* which can cover and entangle aquatic invertebrates and fish in early life stages (Schuytema and Shankland, 1976).

3.2.3. Correlations among chemical parameters and toxicity of leachate/runoff

Table 5 shows the correlations among different variables for the leachate/runoff samples. Significant negative correlation was observed between stage delay concentrations and COD. The negative slope for this relationship indicates that with increased COD concentration in a sample, the volume causing developmental delay decreased (higher toxicity). However, this correlation likely indicates leachate/runoff concentration rather than direct toxicity of COD. Only further tests aimed at determining the source of toxicity can establish the specific compounds of concern. COD is positively correlated with tannins/lignins/phenolics, BOD, and TKN and negatively correlated with pH. A strong positive correlation was observed between BOD and tannins/lignins/phenolics. BOD is negatively correlated with pH. Despite the visual observation of wood particles in the leachate/runoff, COD was not correlated with TSS.

4. Conclusions

Wood mulch varied significantly between sites in a variety of physical-chemical analyses. However, the effect of these differences on the resulting leachate/runoff from the sites was not obvious, probably due to all of the other varying site and precipitation conditions. There was also evidence that the mulch contained considerable amounts of soil.

The results of this study help in better understanding the concentration ranges that can be expected in wood mulch site leachate/runoff samples. Generally, the organic matter concentrations in these samples were comparable to those in raw sewage, while concentrations of nutrients were slightly lower. Substantial direct toxicity was not observed, but ecological toxicity still needs to be explored. Leachate/runoff from wood mulch stockpiles thus is a potential source of organic matter contamination and low levels of nutrients to receiving waterbodies.

Best management practices must take into account the volume of the material being stockpiled, proximity and size of the receiving water, and local precipitation patterns. Even though nutrient concentrations were not high, considering BOD levels of up to 3,000 mg/L and COD levels of up to 6,000 mg/L, it is recommended that leachate be treated before it is released into the off-site environment. Potential treatment technologies include infiltration ponds and constructed wetlands.

These results also point out the limitations of an observational field project and stress the need for a more controlled study. Considerable variability was found among samples for most parameters tested. Lack of control over and/or detailed knowledge of site geology, hydrology, wood material stored on site and its age and processing, conditions and microbial activity within the piles, and storm intensity, frequency, and duration precludes making a detailed assessment of pollutant loads. Thus while these results provide information on leachate/runoff quality at actual sites, they also point to the need for controlled experiments in order to understand and better predict the potential impacts of stormwater runoff from wood recycling facilities. Based on the work described here, such a study might focus particularly on the factors that affect the concentrations and loads of organic matter potentially released.

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Table 1. Site, location within a site, and sampling dates for leachate/runoff samples.

Site-Location[a]	Sampling Dates 2010	Sampling Dates 2011	n
B-1	9/17, 11/5	5/5, 8/15	4
B-2	10/1, 11/5	4/13, 8/15	4
B-3	10/1, 11/5	4/13, 8/15	4
C-1	6/10, 8/25, 12/1	6/15, 9/7	5
C-2	4/5[b], 6/10, 8/25, 12/1	1/15, 9/7	5
D-1	7/29	7/19	2
D-3	7/29	7/19	2

[a] B, C, and D are the three wood recycling facilities, and 1, 2 and 3 are the sampling locations within a site: 1, "puddle" next to or between piles; 2, runoff leaving the site; and 3, water accumulated in retention pond.

[b] Toxicity testing only.

Table 2. Mean values of wood mulch properties at each sampled site. Within a row, means followed by the same letter are not significantly different based on site (Tukey-Kramer test, $p < 0.05$). Except for pH and C:N ratio, parameters are in mg/kg dry weight unless noted.

Property	Site B	Site C	Site D	Temperate woods[c]	Soft-woods[d]
pH	6.0 a	5.5 a	6.4 a		
C:N ratio	75 a	74 a	50.5 a		690
Electrical Conductivity (mS/cm)[a,b]	0.34 a	0.85 b	0.97 b		
Moisture (% wet weight)	44 a	53 b	43 a		
Organic matter (%)	73 b	91 c	59 a	98.9-99.8	98.8
Carbon (%)	34 a	45 b	32 a		54
Total Nitrogen	4741a	6385 b	6750 b		780
Organic Nitrogen	4726 a	6375 b	6700 b		
Phosphorus	398 a	508 b	847 c	10-260	
Potassium	1986 a	2614 b	3208 b	100-2800	840
Calcium	4840 a	7866 b	8800 b	100-1200	
Magnesium	1237 a	958 a	3175 b	30-370	
Sulfur[a]	388 a	425 a	675 b		220
Sodium	90 a	114 a	257 b	5-940	
Aluminum	3429 a	986 a	5746 b		
Iron	5436 b	1515 a	9571 c	6-100	
Manganese	87 a	143 b	276 c	<10-150	
Copper	64 a	13 a	36 a	4-73	
Zinc	47 a	35 a	67 a	2-38	
Lead[b]	15 a	12 a	25 b		

[a] Differences in grind also significant (Grind 2 means were higher).
[b] Interaction between site and grind also significant.
[c] Range of values from up to 13 North American temperate woods, except for organic matter, which is based on 30 (Pettersen, 1984).
[d] Calculated based on Thy et al. (2006), for mixed white fir and ponderosa pine.

Table 3. Summary statistics for the leachate/runoff samples collected from all three sites combined. Included for comparison are typical values measured in sewage and urban stormwater. (T/L/P is tannin, lignin, and phenolic compounds; FC is fecal coliform most probable number.) (Note: An expanded version of this table showing standard deviations and additional comparison wastewaters is available in Supplemental Materials Table S2.)

Statistic	pH	BOD (mg/L)	COD (mg/L)	Sett. S (mL/L)	TSS (mg/L)	TKN (mg/L)	TP (mg/L)	T/L/P (mg/L)	FC (MPN/100 mL)
No. of samples	26	24	26	14	26	26	26	26	12
Minimum	4.8	<20	134	<0.5	69	1.3	0.18	6	≤450
Maximum	7.6	≥2900	5991	0.9	401	70	8	690	>$1.6x10^6$
Mean	6.74	415 [a]	1247	-	171	12.7	3.2	74	-
Median	6.75	107	566.5	<0.5	145	6.3	2.6	44	<1800
Sewage [b]	-	110-350	250-800	5-20	120-400	20-70	4-12	-	10^3-10^8
Stormwater [c]	-	8	65	-	67	1.3	0.3	-	400-50000

[a] Calculated mean for BOD includes one value below the detection limit that was included as ½ the detection limit and one value above measurement limit for the dilutions used, which was taken as that limit.
[b] Typical domestic wastewater influent values, USA (Tchobanoglous et al., 2003).
[c] Median (range, for fecal coliforms) for mixed urban stormwater runoff (USEPA, 1999).

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Table 4. Lethal concentration to 50% of population (LC_{50}) of zebrafish embryos, effective concentration causing an abnormality in 10% of population (EC_{10}), and concentration causing a one stage delay. Samples as described in Table 1. Concentrations are expressed as % leachate/runoff after dilution.

Sample	Sample Date	LC_{50}[a] (%)	EC_{10}[b] (%)	Conc. (R^2) [c]
B-1	9/17/2010	>20	ND	0.28 (0.51)
	11/5/2010	>30	16 (ASC)	**1.83**[f] (0.92)
	5/11/2011	8.5	ND	**2.7** (0.99)
B-2	10/1/2010	>20	26 (ASC)	**1**[f] (0.7)
	11/5/2010	>30	ND	**7.34** (0.67)
	4/13/2011	19	ND	**2.2** (0.92)
B-3	11/5/2010	> 30	ND	**6.1** (0.69)
	4/13/2011	>20	14 (ASC)	2.85[f] (0.57)
C-1	12/1/2010	>100	ND	**2.97** (0.78)
	6/15/2011	4.5	3 (ASC)	**1.27** (0.99)
C-2	4/5/2010	2.7[d]	ne	**0.8**[d] (0.99)
	6/10/2010	>10	4.9 (YSE), 1.4 (ASC)	**0.88**[f] (0.88)
	12/1/2010	>27	ND	3.23 (0.65)
	6/15/2011	40	29 (ASC)	**6.2** (0.71)
D-1	7/29/10[e]	>20	9 (KT)	2.49 (0.01)
	7/19/2011	9	19 (ASC)	5.16 (0.080)
D-3	7/29/2010	>20	3.3 (YSE)	**0.001** (0.7)
	7/19/2011	33	>20 (ASC), 35 (YSE)	**2.6** (0.96)

[a] In many cases (indicated by “>”), 50% mortality did not occur at the highest concentration tested.

[b] YSE = Yolk sac edema; ASC = Abnormal spine curvature; KT = Kink tail; ND = none detected; ne = not examined. Abnormalities not detected in any sample: circulation, pericardial edema, decreased pigmentation, size of head.

[c] Concentration for 1-stage delay. Values in bold are statistically significant ($p < 0.05$).

[d] Only sample in which pH was not adjusted to neutral.

[e] Normal holding time exceeded.

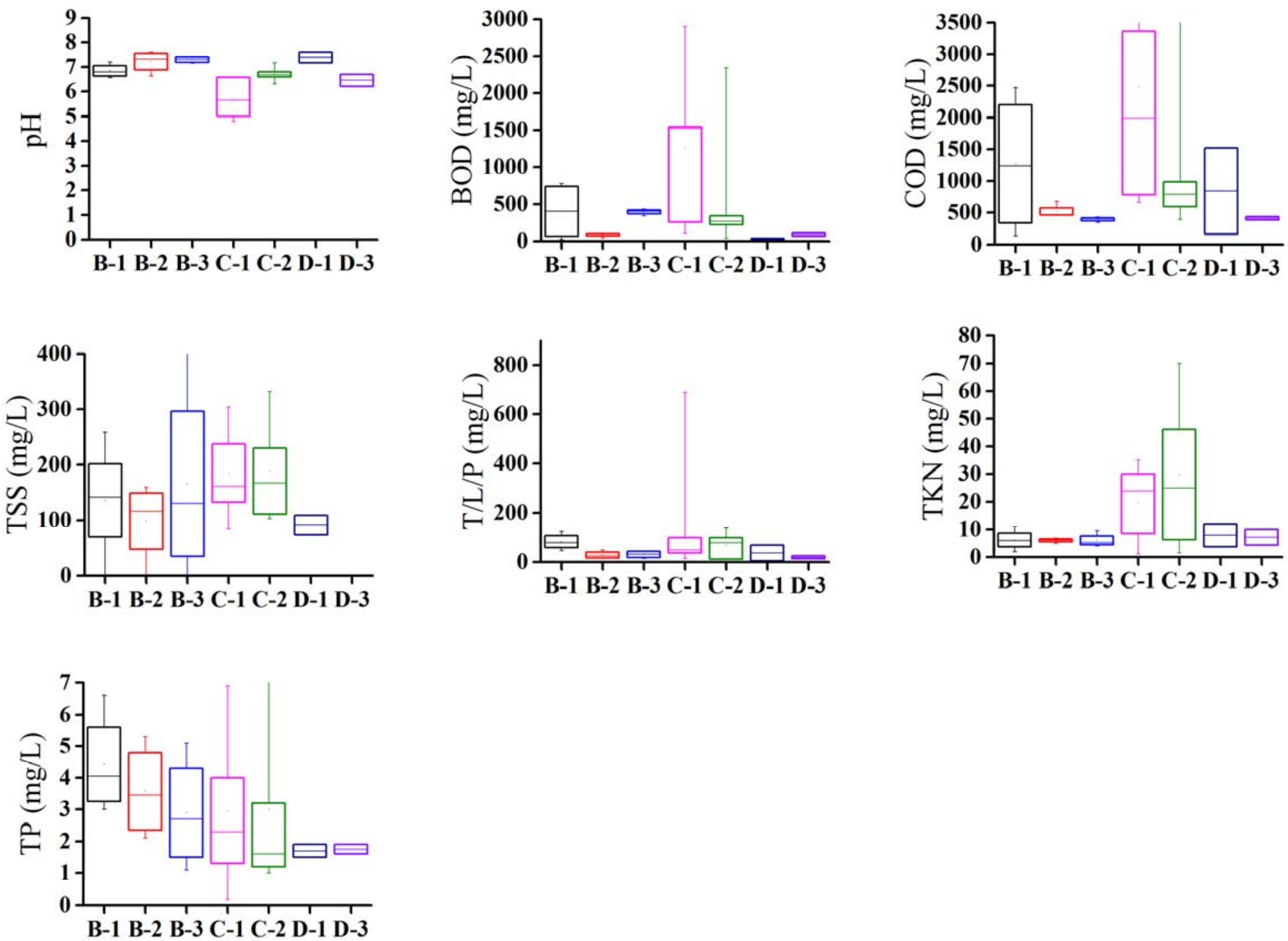
[f] Stage delay concentration calculated based on stages from the 6th day of incubation compared to controls. For other samples, stages from the 5th day were used.

Table 5. The *p*-values from linear correlations among variables for leachate/runoff samples. Bold values indicate significance ($p < 0.05$).

Parameter	1-Stage Delay Conc.	BOD	COD	TSS	T/L/P	TKN	TP
BOD	0.09[a]						
COD	**0.04**[a]	**0.0002**					
TSS	0.33[a]	0.27	0.61				
T/L/P	0.07[a]	**$1.3x10^{-5}$**	**$2.5x10^{-5}$**	0.11			
TKN	0.47[a]	**0.002**	**$8.1x10^{-5}$**	0.8	0.1		
TP	0.76[a]	0.83	0.56	0.8	0.49	0.46	
pH	0.28[b]	**0.0002**[a]	**0.003**[a]	0.38	0.93	**0.04**[a]	0.58

[a] Correlation coefficient is negative.
[b] The pH was adjusted to neutral for toxicity testing (except one sample).

Figure 1. Box and whisker plots for the leachate/runoff results for samples described in Tables 1 and 3. The midline represents the median, the box gives the 25 and 75% values, and the whiskers show the range. Colored dots within boxes are mean values.



Supplementary Material

Table S1. Mean and standard deviation (sd) of wood mulch properties for samples collected at three sites in New Jersey. Except for pH and C:N ratio, parameters are in mg/kg dry weight unless noted.

Parameter	Site B Grind 1 (n= 6)		Site B Grind 2 (n=10)		Site C Grind 1 (n= 4)		Site C Grind 2 (n= 4)		Site D Grind 1 (n= 2)		Site D Grind 2 (n=2)		SRS[a]
	mean	sd	mean	sd	mean	sd	mean	sd	mean	sd	mean	sd	
pH	5.8	0.2	6.2	0.7	6.1	0.5	5	1	6.8	0.1	6	0.4	-
C:N ratio	73	16	77	20	73	21	75	21	56	24	45	16	-
Cond. (mS/cm)	0.3	0.05	0.4	0.1	0.6	0.1	1.1	0.5	0.4	0.2	1.5	0.9	-
Moisture(%)[b]	39	6	48	10	53	5	53	3	46	4	40	1	-
Org.matter(%)	71	11	75	6	93	2	89	2	55	4	63	3	-
C (g/kg)	320	37	366	45	453	44	450	20	311	8	324	103	-
Total N	4483	736	5000	1250	6500	1400	6270	1226	6150	2475	7350	354	-
Org. N	4483	736	4970	1260	6500	1400	6250	1211	6150	2475	7250	495	-
NH_4^+-N	[c]	-	<5	-	<5	-	[d]	-	<5	-	96	76	-
P	405	99	391	90	451	70	565	75	832	213	862	176	-
K	1972	630	2000	500	2437	230	2790	215	2875	766	3542	295	-
Ca	4750	1330	4930	1720	7200	876	8533	1116	7800	1700	9800	1131	-
Mg	1283	620	1190	710	800	163	1117	160	3200	424	3150	70	-
S	367	80	410	70	350	58	500	89	650	212	700	141	-
Na	85	25	97	50	109	32	120	28	226	100	287	106	-
Al	3724	2140	3135	3810	773	314	1200	381	6867	300	4625	147	78000
Fe	5684	2240	5190	2940	1065	554	1965	672	11300	358	7840	1030	-
Mn	102	25	71	30	138	47	149	12	300	48	255	1	11000
Cu	54	56	74	146	11	3	14	4	39	1.4	33	3	3100
Zn	47	17	47	44	30	5	40	5	69	11	65	9	23000
Pb	16	6	14	5	8	3	17	4	30	7	20	3	400

[a] SRS - New Jersey soil remediation residential direct cleanup standards (NJDEP, 2011).
[b] Wet weight basis.
[c] All values below detection limit except for one sample.
[d] All values below detection limit except for two samples.

Table S2. Expanded (from Table 3) summary statistics for the leachate/runoff samples collected from all three sites combined. Included for comparison are typical values measured in sewage and urban stormwater. (T/L/P is tannin, lignin, and phenolic compounds; FC is fecal coliform most probable number.)

Statistic	pH	BOD (mg/L)	COD (mg/L)	TSS (mg/L)	Sett. S (mL/L)	TKN (mg/L)	TP (mg/L)	T/L/P (mg/L)	FC (MPN/100 mL)
Runoff/Leachate									
No. of samples	26	24	26	26	14	26	26	26	12
Minimum	4.8	<20	134	69	<0.5	1.3	0.18	6	≤450
Maximum	7.6	≥2900	5991	401	0.9	70	8	690	$>1.6\times10^{6}$
Mean	6.74	415 [a]	1247	171	-	12.7	3.2	74	-
Median	6.75	107	567	145	<0.5	6.3	2.6	44	<1800
Standard deviation	0.71	745 [a]	1550	86	-	16	2.0	131	-
Wastewater Comparisons									
Sewage [b]									
low strength	-	110	250	120	5	20	4	-	10^{3}-10^{5}
medium strength	-	190	430	210	10	40	7	-	10^{4}-10^{6}
high strength	-	350	800	400	20	70	12	-	10^{5}-10^{8}
Urban stormwater [c]	-	8	65	67	-	1.3	0.3	-	400-50000
Port log yard inf.[d]	-	-	3200	480	-	-	-	-	-
Log yard.[e]	6.7-8.1	0-48	0-14700	7-20100	-	-	-	-	-
Log yard, spruce.[e]	6.7-6.9	-	-	-	-	1.2-2.7	1.2-3.9	-	-
Wood waste.[e]	4.0-4.6	-	2500–6600	-	-	0.1-1.2	0.2-3.1	490–2500[e]	-
Integrated pulp & paper mill influent [f]	6.5	1197	3791	1241	-	-	-	-	-
Papermill woodyard & chipping [f]	7	556	1275	7150	-	-	-	-	-
Yard waste composting leachate [g]	7.75	> 41	56	-	-	- [g]	0.07	0.2 [g]	-

[a] Calculated mean for BOD includes one value below the detection limit that was included as ½ the detection limit and one value above measurement limit for the dilutions used, which was taken as that limit.
[b] Typical domestic wastewater influent values (Tchobanoglous et al., 2003).
[c] Median (range, for fecal coliforms) for mixed urban stormwater runoff (USEPA, 1999).
[d] Influent to a wastewater treatment plant at a port's log yard (Tope 2015).

[e] From review by Hedmark and Scholz (2008); for the spruce logyards leachate, total organic carbon (TOC) concentration = 112-191 mg/L; the number given for woodwaste under T/L/P is for tannin and lignin

[f] From review by Kamali and Khodaparast (2015).

[g] From USEPA (1994), based on work by T.L. Richard and M. Chadsey; mean of 16 samples of leachate from Croton Point, NY, site. BOD included 3 samples > 150 mg/L; value listed as T/L/P was reported as total phenols; TKN not reported, but ammonium-N was 0.44 and nitrite- plus nitrate-N was 0.98 mg/L