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Leaching of dissolved organic carbon, dissolved organic nitrogen, and other solutes from coarse woody debris and litter in a mixed forest in New York State

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Abstract. Coarse woody debris (CWD) may play a role in nutrient cycling in temperate forests through the leaching of solutes, including dissolved organic carbon (DOC) and dissolved organic nitrogen (DON), to the underlying soil. These fluxes need to be considered in element budget calculations, and have the potential to influence microbial activity, soil development, and other processes in the underlying soil, but studies on leaching from CWD are rare. In this study, we collected throughfall, litter leachate, and CWD leachate *in situ* at a young mixed lowland forest in NY State, USA over one year. We measured the concentrations of DOC, DON, NH_4^+, NO_3^-, dissolved organic sulfur, SO_4^{2-}, Cl^-, Al, Ca, K, Mg, Na, and P, estimated the flux of these solutes in throughfall, and measured the cover of CWD to gain some insight into possible fluxes from CWD. Concentrations of DOC were much higher in CWD leachate than in throughfall or litter leachate (15 vs. 0.7 and 1.6 mM, respectively), and greater than reported values for other leachates from within forested ecosystems. Other solutes showed a similar pattern, with inorganic N being an exception. Our results suggest that microsite scale fluxes of DOC from CWD may be An high relative to throughfall and litter leaching fluxes, but since CWD covered a relatively small fraction (2%) of the forest floor in our study, ecosystem scale fluxes from CWD may be negligible for this site. Soil directly beneath CWD may be influenced by CWD leaching, in terms of soil organic matter, microbial activity, and N availability. Concentrations of some metals showed correlations to DOC concentrations, highlighting the possibility of complexation by DOM. Several solute concentrations in throughfall, including DOC, showed positive correlations to mean air temperature, and fewer showed positive correlations in litter leachate, while negative correlations were observed to precipitation, suggesting both biological and hydrologic control of solute concentrations.

Introduction

Dissolved organic matter (DOM) is involved in numerous processes in forest ecosystems and their drainage waters. The production, movement, and fate of dissolved organic carbon (DOC) and dissolved organic nitrogen (DON) are important components of carbon (C) and nitrogen (N) cycles in forest

ecosystems (Yavitt and Fahey 1986, Michalzik et al. 2001). Dissolved organic compounds contribute to the formation of soil organic matter (Dawson et al. 1978; McDowell and Wood 1984), facilitate metal and organic pollutant mobilization (McCarthy and Zachara 1989; Christensen and Christensen 1999), may play an important role in stream chemistry (Moore and Jackson 1989; Driscoll and Fuller 1994), and react to produce toxic chlorinated organic compounds in drinking water (Hoadley and Gould 1976; Kalmez and Kalmez 1981). Considering the current interest in C and N cycling due to changes in local and global cycles, there is considerable motivation to more completely understand these processes.

Numerous studies have documented the leaching of high concentrations of DOC (mean values up to 7 mM), and associated high fluxes (up to 4 mol m^{-2} y^{-1}) from the litter layer or forest floor in deciduous and coniferous temperate forests across the globe (McDowell et al. 1998, and see reviews by Michalzik et al. 2001 and Neff and Asner 2001). Concentrations and fluxes of DON have been documented in fewer cases, but range from 30 to 240 μM and 20 to 90 mmol m^{-2} y^{-1}, respectively (McDowell et al. 1998; Solinger et al. 2001, and see review by Michalzik et al. 2001).

Coarse woody debris (CWD) may constitute a much larger fraction of the total aboveground detritus in temperate forests than litter (Lang and Forman 1978; Harmon and Cromack 1987; Stewart and Burrows 1994), and thus may contribute a substantial flux of DOC to the underlying soil. Additionally, because of a highly 'clumped' distribution of woody detritus, it may create 'hot spots' (McClain et al. 2003) with very high solute concentrations and fluxes on a microsite scale. Whereas elements with no prominent gaseous phase must leave CWD and enter the soil at approximately the rate at which they fall in CWD on an ecosystem level in systems near steady state, this is not the case for C and N: respiration can cause substantial loss of C, while N_2 fixation can add N to CWD (e.g. Jurgensen et al. 1984; Mattson et al. 1987).

Despite the potential significance of leaching from CWD, few studies have measured the concentrations or estimated the fluxes of solutes in leachate from decaying CWD (Yavitt and Fahey 1985; Mattson et al. 1987; Spears et al. 2003). Simulation models and budget calculations of C movement in forests have assumed this flux to be zero (Raich and Nadelhoffer 1989; Currie and Aber 1997). Ignoring a potentially substantial movement of C could lead to flawed understanding of C cycling in temperate forests, and errors in budget calculations and simulation results.

Understanding the importance of the leaching of other elements (e.g., P and S), as well as inorganic forms of N, from CWD is important for understanding the role of CWD in the cycles of these elements. Many studies on the chemistry of CWD decay have suggested that some elements (especially N) are temporarily immobilized in CWD (Grier 1978; Brown et al. 1996; Laiho and Prescott 1999), while other studies have demonstrated decreases in the concentrations or total mass of N and other elements beginning early on in decomposition (Means et al. 1992; Creed et al. 2004; Ganjegunte et al. 2004). Some of these differences

may be attributed to the use of chronosequences, which have the potential to underestimate nutrient loss, since fragmentation represents a loss of elements that is often not considered (Harmon et al. 1994). However, methods to correct for this loss have been applied (Krakina et al. 1999; Creed et al. 2004), and even observations of CWD over time have resulted in inconsistent responses between locations and tree species (Brown et al. 1996; Laiho and Prescott 1999; Ganjegunte et al. 2004). Additionally, even with a net accumulation of N, CWD could still be a net source of biologically available N to the soil if N_2 fixation, which has been observed in wood in various settings (e.g., Cornaby and Waide 1973; Jurgensen et al. 1984), produces more fixed N than is accounted for by accumulation. Lastly, the demonstration of net changes within CWD provides no information about the pathways of nutrient loss. Considering the shortcomings of these approaches, it is important to study specific pathways of nutrient loss from decaying CWD, including leaching.

In a lowland setting, periodic flooding has the potential to complicate the role of CWD in nutrient cycling, since floodwater may act as a source or sink of solutes for CWD. In the absence of microorganisms, solutes would be expected to move via advective flow, as water moves down a hydraulic potential gradient, or via diffusive flow, as solutes move down a concentration gradient, analogous to solute behavior in ground water (Fetter 2001). Decay organisms, however, have the potential to move elements against a concentration gradient (Aumen et al. 1985; Diez et al. 2002). This process could alter solute concentrations and fluxes in both CWD and, to a lesser extent, stream water.

The primary difficulty in estimating leaching rates from CWD is in estimating the hydrologic flux from CWD. As with soil horizons, it is not valid to assume that the volume of leachate collected in a lysimeter is equal to the hydrologic flux, although water volumes collected in zero-tension lysimeters have sometimes been used as an estimate of water flux under natural conditions, especially for organic horizons (e.g., Qualls et al. 1991; Kaiser et al. 2001; Solinger et al. 2001), and also for CWD (Mattson et al. 1987; Spears et al. 2003). Hydrologic fluxes through soil horizons can be estimated using watershed budget or simulation approaches (e.g. Mitchell et al. 2001b; Qualls et al. 2002), and validated models exist for the latter (though shortcomings clearly exist) (Aber et al. 1995; Federer 1995). Unfortunately, simulation models are not available to simulate water flow through CWD, and watershed budget approaches do not reflect the process.

The objectives of this study were to determine the effects of CWD on solution chemistry of water flowing through it, and assess the significance of CWD as a source of DOC, DON, and other solutes relative to litter and throughfall at both microsite and ecosystem scales in a temperate forest. We collected throughfall, litter leachate, and CWD leachate over a 1-year period at a mixed forest in NY State, USA, and compared solute concentrations among these three sources. Leachates were continuously collected *in situ* using zero-tension lysimeters. We estimated solute fluxes in throughfall, and measured the cover of CWD to examine possible solute fluxes from CWD.

Methods

Study site and plot layout

The study site was a young, mixed lowland forest in southeastern New York State at the Institute of Ecosystem Studies in Millbrook, NY, and was used as a pasture for dairy cows until 1972 (R. Winchcombe, Institute of Ecosystem Studies, personal communication) (Figure 1). The pasture included trees and shrubs, and thus the age of some trees is likely greater than 30 y. The topography of this site is relatively flat, with several braids of the same stream flowing through the lowest portions and much of the area in a floodplain and annually flooded. Elevation ranges from 91 to 94 m above sea level, and the area studied is approximately 7.8 ha. Soils are generally mineral (Fluvaquents-Udifluvents complex [USDA NRCS 2002]), but have high organic matter (ca. 5% organic C), and range from wet (i.e., flooded in the spring) to generally dry (e.g., 20% water-filled pore space in the summer), due to microtopographic relief. Some areas of organic soil are also present within the site. The litter layer at the study site is relatively thin (ca. 10 mm) and is the only organic horizon over most of the site, resulting in a very shallow forest floor, probably due to flooding. Annual precipitation averages 1070 mm (extremes: 797, 1424 mm) with the most precipitation falling in May (120 mm monthly mean) and annual temperature ranges from − 2.7 °C in January (monthly mean) (extreme low: − 32.6 °C) to 21.8 °C in July (monthly mean) (extreme high: 39.2 °C) (data from 1988 to 2002; Kelly 1999, 2002). Tree species present include red maple (*Acer rubrum* L.), sugar maple (*Acer saccharum* Marshall), American sycamore (*Platanus occidentalis* L.), shagbark hickory (*Carya ovata* (Miller) K. Koch.), white pine (*Pinus strobus* L.), yellow birch (*Betula alleghaniensis* Britton), black cherry (*Prunus serotina* Ehrh.), and American elm (*Ulmus americana* L.).

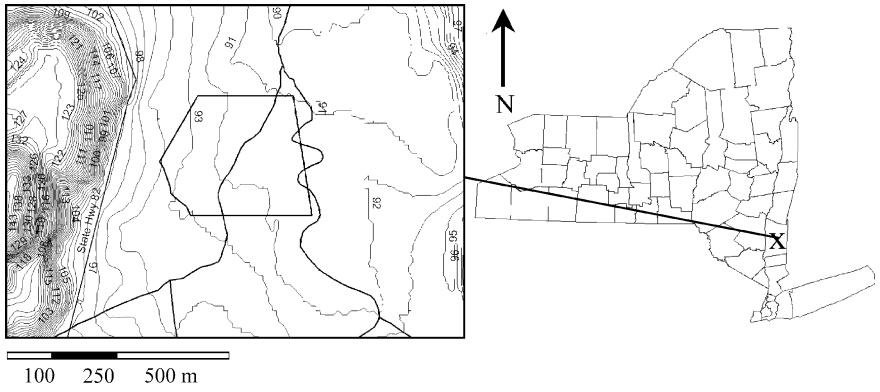

Figure 1. A map of the study site. The 'X' marks the position of the study site within NY State, USA (41° 50′ N, 73° 45′ W). In the close-up at the left, the irregular shape is the approximate boundary of the study site, which has a total area of 7.8 ha. The contour interval is 1.0 m.

Understory species include poison ivy (*Toxicodendron radicans* (L.) Kuntze.), multiflora rose (*Rosa multiflora* Thunb.), barberry (*Berberis* sp.), American hornbeam (*Carpinus caroliniana* Walter), and spicebush (*Lindera benzoin* (L.) Blume). Herbaceous species include skunk cabbage (*Symplocarpus foetidus* (L.) Nutt.), sensitive fern (*Onoclea sensibilis* L.), lady fern (*Athyrium filix-femina* (L.) Roth.), crested wood-fern (*Dryopteris cristata* (L.)), various graminoids (species in the families Poaceae and Cyperaceae), and cattail (*Typha* sp.) in some wet areas beneath an open canopy.

At the site, a census of all downed woody debris in contact with the ground and greater than 20 cm in diameter was taken by walking a series of transects through the entire area (ca. 12 m spacing), and each piece was characterized by decay class, following Lang and Forman (1978), with some modifications. Specifically, a 0.5-cm diameter pointed metal shaft was pushed into each log as far as possible at several points. Logs for which penetration was less than 2.5 cm were assigned to decay class 1, those with penetration between 2.5 and 5 cm were assigned to decay class 2, and those with penetration greater than 5 cm and that readily fell apart were assigned to class 3. Treating each decay class as a stratum, a random sample of two logs was selected from each class, for a total of six logs. The vicinity of each log was treated as an experimental block, and adjacent to each log two square 16-m^2 plots were established for throughfall and litter leachate collection, approximately 1 m from the log's edge.

Determination of solute concentrations in leachate and throughfall

In each litter leachate plot, a zero-tension lysimeter was placed in the center just below the litter layer (the O_i horizon [Brady and Weil 1999]). An identical lysimeter was installed below a central 1-m section of the log at each block, just below, and in contact with, the log. Lysimeters consisted of PVC plastic troughs, approximately 300 × 80 mm, containing a perforated PVC insert and filled with washed sand, and drained into a 4-l plastic bottle through a flexible PVC tube. The surface of each lysimeter was covered with a layer of glass wool to minimize the disruption of sand. Throughfall was sampled in the center of the litter-free plots using a 200-mm diameter funnel at a height of 30 cm that drained into a 4-l bottle. A plug of glass wool at the bottom of the funnel minimized particulate matter contamination of the throughfall when necessary, and a loop in the tubing acted as a trap to minimize evaporation. Installation of collectors took place on May 21, 2002, and the last set of samples was collected on May 18, 2003. Solution collection bottles were emptied twice for most months, though less frequently at times, with collection following storm events where possible. Accumulated volume was measured in the field and a sample was collected in a plastic bottle (rinsed with sample when possible). Samples were brought to the laboratory on ice, and held at 4 °C, where they were stored until analysis or freezing (see below). Dissolved organic carbon

analysis and freezing of subsamples (see below) were completed within 2 weeks of sampling. However, early samples (from the first five intervals) were stored at 4 °C for as long as 2 months before freezing subsamples.

Sample pH was measured using a Corning 245 pH meter (Corning Inc., Corning, NY, USA), and data were analyzed as H^+ concentrations. Subsamples were filtered (0.45 μm) and acidified with 650 μl H_2SO_4 per 40 ml of sample, for DOC analysis. Filtered acidified samples were analyzed for DOC using a Tekmar Phoenix 8000 UV persulfate TOC analyzer (Tekmar-Dorhmann, Cleveland, OH, USA), set for samples between 0.1 and 20 ppm DOC. Samples for which higher DOC concentrations were expected were diluted using deionized distilled water (DDW) prior to filtration. Subsamples were frozen for longer-term storage (i.e., several months), and these were used to determine N, anion, and cation concentrations. Total dissolved N and NH_4^+ were measured using a Technicon Autoanalyzer, with persulfate oxidation carried out prior to analysis for total dissolved N (TDN); NO_3^-, SO_4^{2-}, and Cl^- were measured using a Dionex Ion Chromatograph (Dionex Corporation, Sunnyvale, CA, USA); and DON concentrations were calculated by difference. An Inductively Coupled Plasma Emission Spectrometer (Perkin-Elmer Optima 3300 DV, Perkin Elmer Corp., Wellesly, MA, USA) was used to determine the concentrations of Al, Ca, K, Mg, Na, P, and S. Dissolved organic S (DOS) concentrations were calculated as the difference between total dissolved S (TDS) and SO_4^{2-}.

The annual flux of solutes in throughfall was estimated by multiplying the concentration of each solute from a given collector by the throughfall volume collected in that interval, and these values were summed for the entire year. The mean of these six flux estimates were taken as the estimated flux. Where concentration data were not collected, values were linearly interpolated. Throughfall volume was not determined over three sampling intervals, due to freezing conditions, and instead, it was estimated as 90% of precipitation volume, which was the relationship observed in the preceding and following sampling intervals. Precipitation data were collected at the Institute of Ecosystem Studies weather station (Institute of Ecosystems Studies Environmental Monitoring Program 2002; Kelly 2002). Because of the influences that flooding may have on solute fluxes (i.e. generating horizontal flows of solutes into and out of CWD and the litter layer, and increasing vertical movement of solutes), as well as the lack of a method for estimating hydrologic fluxes from CWD, solute fluxes from CWD and the litter layer were not estimated. However, the cover of CWD was used to gain insight into potential solute fluxes.

Sampling of coarse woody debris

Coarse woody debris was sampled at the site to estimate the cover, volume, and biomass in each decay class. Nine point locations were produced within the study site by randomly generating pairs of Cartesian coordinates. At each

point, CWD with a maximum diameter greater than 10 cm was sampled in a square 400-m^2 plot, and smaller CWD (down to a maximum diameter of 2.5 cm) was sampled in a square 25-m^2 plot. The diameter of each piece was measured at each end using calipers, and the total length was measured. The decay class was determined by measuring the penetration as described above, in six locations. Area was calculated from these data as the projected area, using the formula for the area of a trapezoid. The volume of CWD was estimated using the formula for the frustum of a cone, and the mean density measured for each decay class was multiplied by volume to estimate total biomass. Total C content in CWD was estimated using mean C contents for each decay class. Carbon contents were taken as 43% of total organic matter content (calculated from Lang and Formann 1978), which was determined for each of the six logs from which leachate was collected by combusting samples at 550 °C in a muffle furnace. Total N was estimated using measured total N concentrations for each of the six logs from which leachate was collected. Total N concentrations were measured using a macro-Kjedahl method, using 2 g of wood (Bickelhaupt et al. 1983).

Statistical analyses

The spatial and temporal variances of the solute concentrations were much greater in CWD leachate than in throughfall or litter leachate, and the difference between sources was not normally distributed over space, leading to nonadditivity of factors. Because these qualities deviate from the assumptions of analysis of variance (ANOVA) (Zar 1999), nonparametric analyses were used to detect differences between sources. Friedman's test was utilized to test for an overall effect of source (i.e. throughfall, litter leachate, or CWD leachate) on solute concentrations, and individual sources were compared to each other using the Wilcoxon paired-sample test (Zar 1999). The program used to carry out Friedman's and the Wilcoxon paired-sample tests was written in FORTRAN90 code and is available from the corresponding author upon request. Mean concentrations from each plot over the year were utilized in the analyses. The inclusion of all data in the analyses despite the fact that not all collectors within a block produced a sample at each sampling event potentially introduces bias in the statistical comparisons. However, all data were included to provide the best estimates of mean concentrations, and an equitable distribution of samples over time. Statistical analyses were repeated with a reduced data set that did not contain data collected from blocks on a date when not all collectors produced a sample, to determine the possibility of bias. Correlations between DOC and cation concentrations were quantified separately for each plot using the Spearman rank correlation coefficient (r_s) (Zar 1999). To assess the statistical significance of the observed correlations at the site level, the Wilcoxon paired-sample test was applied to the correlation coefficients as a one-sample test, to determine if r_s differed from zero (Zar

1999). An identical analysis was applied for DON and DOS correlation to DOC. Correlation analyses and the one-sample application of the Wilcoxon paired-sample test were carried out using SAS v. 9.0 (SAS Institute, Cary, NC, USA). To explore relationships between solute concentrations and weather, we used Kendall's robust line-fit method (Sokal and Rohlf 1995) to calculate a regression slope between solute concentrations and mean precipitation rate (mm d^{-1}) or mean air temperature (°C) for each plot (weather data from Kelly [2002]). Mean air temperature was taken as the mean of temperature maxima and minima. The program used to carry out Kendall's robust line-fit method was written in FORTRAN90 code and is available from the corresponding author by request. Statistical significance of the regressions at the site level was assessed by applying the Wilcoxon paired-sample test to the slope estimates as a one-sample test as was done for the r_s estimates. The Spearman rank correlation coefficient was also used to quantify correlation between CWD N concentrations and N species concentrations in leachate. All statistical results were evaluated at $\alpha = 0.05$.

Results

Hydrologic fluxes

Total precipitation flux for the 1-year study period (May 22, 2002-May 18, 2003) was 1162 mm, similar to the annual average of 1070 mm. Total throughfall (including the estimated throughfall for three periods) was 973 mm. On two sampling dates (December 23, 2002 and March 23, 2003) there was evidence of flooding at three experimental blocks, specifically, the water table was above the soil surface in some locations, and the standardized volume collected in some lysimeters was as much as 5-fold that collected in lysimeters at other blocks. Concentration data from these three blocks were excluded from all analyses for those two dates. The mean water volume collected in CWD and litter lysimeters represent annual fluxes of 262 and 644 mm, respectively, excluding flooded samples.

Concentrations of elements in throughfall and leachate

In general, measured solutes showed much higher concentrations in CWD leachate than in litter leachate or in throughfall. Additionally, litter leachate had higher concentrations than throughfall for most solutes. All differences described below are statistically significant unless otherwise stated.

The mean concentration of DOC in leachate from CWD (ca. 15 mM) was an order of magnitude greater than the mean concentration in litter leachate (ca. 1.6 mM), which was twice the mean concentration in throughfall (ca. 0.7 mM) (Table 1, Figure 2a). Leachate from some logs occasionally showed extremely

Table 1. Mean solute concentrations, statistical results, and throughfall fluxes from May 21, 2002–May 18, 2003[a].

Solute	Concentrations (μM) or C:N ratio (g g^{-1})						Statistical results		Flux (mmol m^{-2} y^{-1})
	CWD 1	CWD 2	CWD 3	Mean CWD	TF	Litter	Friedman's χ^2	p	TF flux
DOC	12,500	7120	25,600	15,100	693	1600	12.0	0.001	489
TDN	274	209	168	217	92	116	7.0	0.05	63
NH_4^+	7.5	4.0	7.5	6.3	32	5.5	10.3	0.002	21
NO_3^-	132	80	24	79	38	76	0.3	>0.5	30
DON	134	97	141	124	21	30	10.3	0.002	12
H^+	1.8	2.6	4.1	2.8	2.7	2.3	3.0	0.3	3.8
Al	20	9.2	50	26	1.0	3.8	10.3	0.002	0.84
Ca	741	843	1353	979	51	255	12.0	0.001	48
K	335	95	770	400	68	116	9.3	0.01	48
Mg	168	244	359	257	26	76	12.0	0.001	21
Na	102	493	477	357	18	140	10.3	0.002	18
P	3.9	19	31	18	3.7	3.5	4.0	0.2	2.2
TDS	67	143	144	118	38	60	12.0	0.001	28
SO_4^{2-}	32	117	62	70	33	52	3.0	0.3	25
DOS	29	25	77	44	4.6	7.7	12.0	0.001	3.6
Cl^-	46	474	463	327	41	118	10.3	0.002	32
DOC: DON	93	74	156	105	28	46	7.0	0.05	NA

[a]TF = throughfall, and CWD 1, 2, and 3 refer to the decay classes (see Methods). The sample size was two logs for each CWD decay class and 6 plots for throughfall and litter.The mean for each plot is based on 9–17 measurements. For Friedman's test, a = three blocks and b = six replicates for each solute (Zar 1999). The Friedman's χ^2 and p values presented refer to the null hypothesis that CWD leachate, litter leachate, and throughfall concentrations do not differ. NA = not applicable.

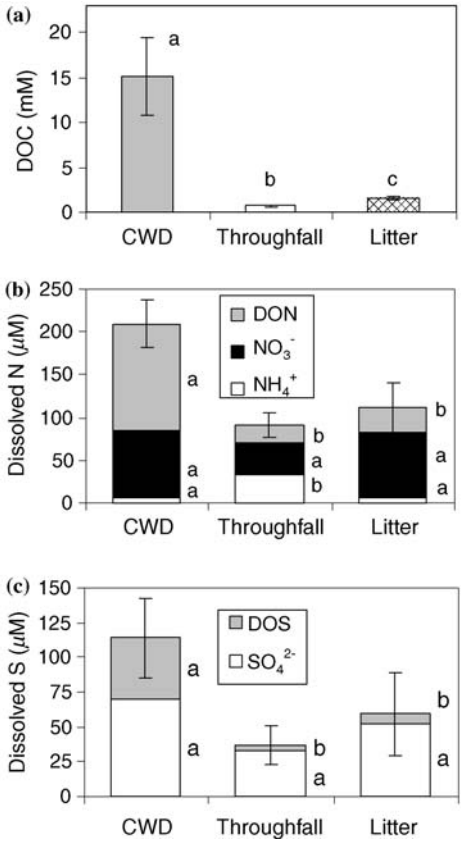

Figure 2. Mean concentrations in throughfall and CWD and litter leachate of DOC (a), dissolved N (b), and dissolved S (c). Concentrations of a solute that do not share a letter are significantly different by the Wilcoxon paired-sample test. The bars represent the standard error ($n = 6$ plots; the mean of each plot is based on 9–17 samples).

high concentrations of DOC (maximum of 77 mM from a log in decay class 3 in August 2002). Dissolved organic N was more concentrated in CWD leachate than in litter leachate or throughfall, NH_4^+ was less concentrated in CWD and litter leachate than in throughfall, and NO_3^- showed no statistically significant differences (Table 1, Figure 2b). Dissolved organic N contributed the greatest fraction of TDN to CWD leachate (ca. 60%), while DON made a smaller contribution to litter leachate and throughfall TDN.

TDS and DOS showed higher concentrations in CWD leachate than in litter leachate and throughfall (Table 1, Figure 2c). Sulfate showed no statistically significant differences (Table 1, Figure 2c). Concentrations of all cations except H^+ (and NH_4^+) were significantly greater in CWD leachate than in throughfall (Table 1, Figure 3a). Additionally, CWD leachate showed a greater concentration of Ca, Mg, and K than litter leachate. The most

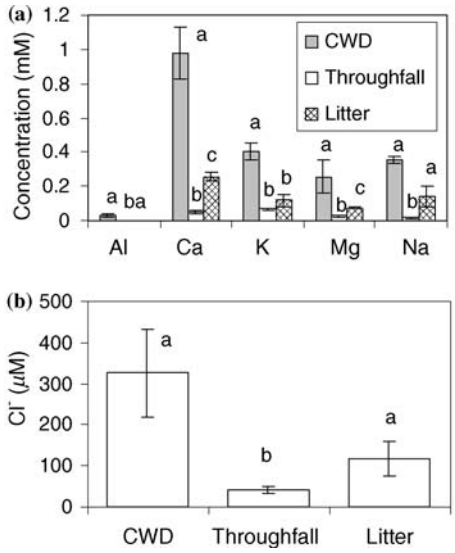

Figure 3. Mean concentrations in throughfall and CWD and litter leachate of cations (a), and Cl^- (b). Concentrations of a solute that do not share a letter are significantly different by the Wilcoxon paired-sample test. The bars represent the standard error (*n* is as with Figure 2).

concentrated cation in CWD leachate was Ca, which reached a mean of ca. 1 mM. Total dissolved P was not significantly different between the three sources, though the mean concentration in CWD leachate was much greater than that in litter leachate and throughfall (and $p < 0.10$ for all comparisons except litter:throughfall) (Table 1). The mean concentration of Cl^- was greater in CWD than in throughfall but otherwise showed no significant differences (Table 1, Figure 3b). Differences in H^+ concentrations were not statistically significant, and all means fell between pH 5 and 6 (Table 1). The mean DOC:DON ratio in CWD was greater than in throughfall (Table 1).

The concentrations of most solutes showed an increase from CWD decay classes 1 and 2 to decay class 3 (Table 1). However, several solutes had greater concentrations in leachate from decay class 1 than decay class 2, and both TDN and NO_3^- concentrations decreased as the degree of decay increased. The sample size used in the study (two logs of each decay class) was too small to adequately assess the importance of these differences.

Statistical conclusions using the reduced data set were identical to those listed above, excepting that K did not show a significant difference between CWD leachate and throughfall ($p = 0.08$) and CWD and litter leachate ($p = 0.08$), and DOC:DON ratio did not show a significant difference between CWD leachate and throughfall ($p = 0.08$).

Concentrations of most solutes were highly variable over time in CWD leachate, and litter leachate and throughfall generally showed lower variability (e.g. Figure 4a). For CWD, the mean concentration of DOC was lowest in

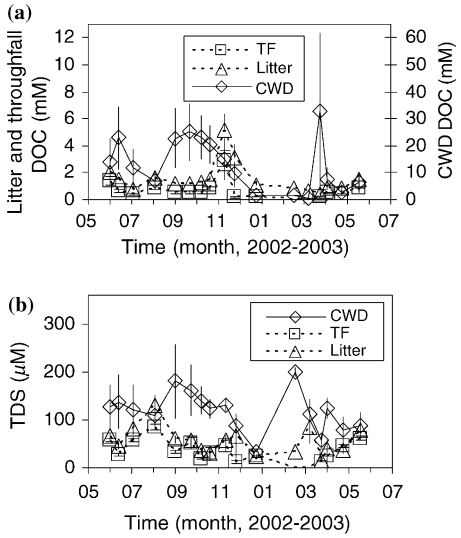

Figure 4. Temporal patterns in DOC (a), and TDS (b) concentrations, for throughfall and leachates. Each point represents the mean of, at most, six plots. Bars represent standard error. The *x* coordinate of each point represents the time at which the water sample was collected (i.e. accumulation occurred from the previous point). Note that for part (a), the scale for CWD leachate is shown on the right. TF = throughfall.

winter, with a > 40-fold difference between the extremes (Figure 4a). Other solutes showed less variability over time, e.g. TDS (Figure 4b).

The estimated throughfall flux for all solutes except DOC was < 70 mmol m^{-2} y^{-1} (Table 1). The lowest estimated fluxes were observed for Al, P, DOS, and H^+.

Solute relationships

All cations except Na showed statistically significant positive correlations to DOC at a site level for throughfall, litter leachate, or CWD leachate (Table 2). CWD leachate showed significant correlations for Al, Ca, K, and Mg, while correlations in litter leachate were only significant for Al, K, and H^+ (Table 2). In throughfall, Al, Ca, K, and Mg showed significant positive correlations to DOC (Table 2). Aluminum concentrations showed the strongest plot level correlations to DOC concentrations (Figure 5a). Dissolved organic N showed a significant positive correlation to DOC only in throughfall, while DOS showed a positive correlation in both throughfall and litter leachate. However, positive correlations between DON and DOC were significant for three CWD plots (Figure 5b).

Several solutes showed significant correlations to mean air temperature and precipitation rate (Table 2). In throughfall, DON, dissolved inorganic N

Table 2. Regression between solute concentrations in throughfall and leachates and weather, and correlation between solute and DOC concentrations[a].

	Slope, concentration vs. precipitation (μM d mm^{-1} or g g^{-1} d mm^{-1})									
	DOC	DON	DOS	TDN	DIN	TDS	SO_4^{2-}	Cl^-	P	DOC:DON
CWD	668 (0.09)	0.58 (0.56)	0.23 (0.31)	−3.8 (0.69)	0.0 (1.0)	−0.97 (0.56)	−0.98 (0.56)	−0.12 (0.84)	0.06 (0.69)	6.6 (0.03)
TF	−21 (0.44)	−1.9 (0.03)	−0.16 (0.56)	−8.8 (0.06)	−7.3 (0.03)	−5.3 (0.03)	−5.3 (0.03)	−1.5 (0.03)	−0.13 (1.0)	0.47 (0.03)
Litter	31 (0.56)	−1.4 (0.09)	−0.33 (0.06)	−5.9 (0.03)	−2.0 (0.22)	−6.3 (0.03)	−6.3 (0.03)	−1.2 (1.00)	−0.33 (0.06)	1.9 (0.03)
	Slope, concentration vs. temperature (μM °C^{-1} or g g^{-1} °C^{-1})									
	DOC	DON	DOS	TDN	DIN	TDS	SO_4^{2-}	Cl^-	P	DOC:DON
CWD	39 (1.00)	0.26 (1.00)	0.02 (0.84)	4.7 (0.16)	1.3 (0.13)	−0.17 (0.84)	−1.3 (0.16)	−21 (0.03)	−0.080 (0.84)	−0.99 (0.22)
TF	17 (0.03)	0.50 (0.03)	0.17 (0.06)	3.3 (0.03)	3.1 (0.03)	1.6 (0.03)	1.2 (0.03)	−0.70 (0.16)	0.10 (0.03)	−0.10 (0.06)
Litter	15 (0.56)	0.96 (0.03)	−0.014 (1.00)	5.2 (0.03)	2.4 (0.06)	1.4 (0.44)	0.91 (0.44)	−2.5 (0.03)	0.15 (0.03)	−0.78 (0.06)
	Spearman rank correlation coefficient (r_s), concentration vs. DOC									
	H^+	Al	Ca	K	Mg	Na	DON	DOS		
CWD	0.36 (0.56)	0.82 (0.03)	0.80 (0.03)	0.76 (0.03)	0.84 (0.03)	−0.29 (0.44)	0.58 (0.09)	0.73 (0.06)		
TF	−0.18 (0.06)	0.43 (0.03)	0.44 (0.03)	0.71 (0.03)	0.74 (0.03)	−0.12 (0.06)	0.40 (0.03)	0.71 (0.03)		
Litter	0.42 (0.03)	0.65 (0.03)	0.53 (0.16)	0.58 (0.03)	0.49 (0.06)	0.02 (1.00)	0.47 (0.09)	0.64 (0.03)		

[a]Values are the median of the six individual estimates calculated for each plot (n = 9– 17 samples), with p values from the Wilcoxon paired-sample test (n = 6 plots) in parentheses. Regression utilized mean precipitation rate (mm d^{-1}) and mean air temperature as independent variables.TF = throughfall.

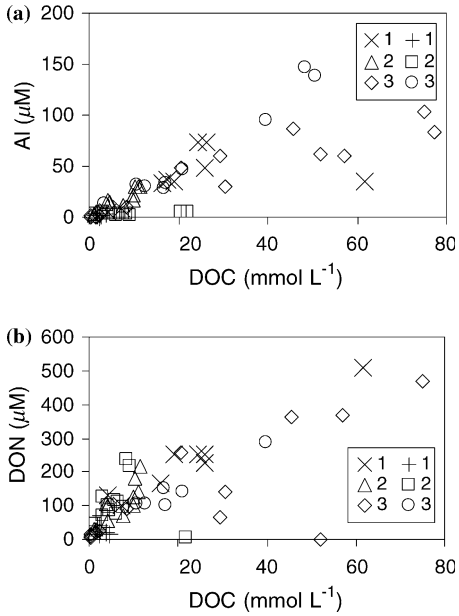

Figure 5. Concentration of Al vs. DOC (a) and DON vs. DOC (b) for the CWD plots only. Different symbols represent individual plots, identified by decay class. For Al vs. DOC, four plots showed significant correlation ($r_s > 0.72$, $p < 0.03$). For DON vs. DOC, three plots (one of each decay class) showed significant correlation ($r_s > 0.68$, $p < 0.01$).

(DIN), TDS, SO_4^{2-}, and Cl^- showed a negative correlation to precipitation, while only TDN, TDS, and SO_4^{2-} showed a negative correlation in litter leachate, and none were significant in CWD leachate. However, DOC:DON ratios were positively correlated to precipitation in all three sources. Mean air temperature was a less effective predictor of solute concentrations. In throughfall, DOC, DON, TDN, DIN, TDS, SO_4^{2-}, and P all showed a positive correlation to temperature. The only solute correlated with temperature in CWD leachate was Cl^-, and the response was negative, as it was in litter leachate. In litter leachate, DON, TDN, and P all showed a positive relationship to temperature.

Concentrations of dissolved N species showed no clear relationship to the concentration of N in wood ($|r_s| < 0.54$, $p > 0.26$) (Figure 6). Similarly, DOC:DON ratios showed no relationship to CWD N concentration ($r_s = -0.26$, $p = 0.62$).

Coarse woody debris characteristics

The estimated volume of CWD at the study site was 26 m^3 ha^{-1} (Table 3), with most of the volume in decay class 3. The projected area of CWD was

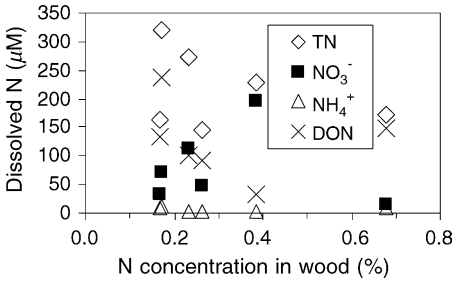

Figure 6. Mean coarse woody debris leachate N concentrations vs. N concentration of wood. Different points represent different plots.

Table 3. Mean area, volume, biomass, and C and N content of CWD[a].

Variable	Class 1	Class 2	Class 3	Total	Standard error
Area (m^2 ha^{-1})	110	38	88	240	54
Volume (m^3 ha^{-1})	7.8	4.9	13	26	6.7
Biomass (Mg ha^{-1})	2.2	1.3	2.8	6.3	ND
C(Mg ha^{-1})	0.93	0.54	1.2	2.7	ND
N(kg ha^{-1})	6.1	3.1	12	21	ND
C:N (g g^{-1})	150	170	100	130	ND

[a]The sample size = nine plots for all area and volume stimates.Biomass, N, and C estimates for each decay class were made using the mean of two measurements. C:N ratios were calculated using the mean C and N contents. ND = not determined.

240 m^2 ha^{-2}, and decay class 1 contributed the greatest area. Biomass of CWD was estimated as 6.3 Mg ha^{-1} with decay class 3 making the largest contribution. The estimated C pool of CWD was 2.7 Mg ha^{-1} (2.2×10^5 mol ha^{-1}), and that of N was 21 kg ha^{-1} (1.5×10^3 mol ha^{-1}), for a C:N ratio of 130 g g^{-1} (150 mol mol^{-1}).

Discussion

Effects of detritus on solution chemistry

The results of our study clearly demonstrate the effect that litter and CWD have on the chemistry of throughfall percolating through them; namely to increase the concentration of most solutes. Additionally, concentrations of most solutes were higher in CWD leachate than in litter leachate, showing that CWD has a much greater effect on solution chemistry than litter. These detrital sources may increase solute concentrations through both evaporation and consequent concentration of throughfall, or leaching of material from within the detritus. Of these, only leaching represents the net transport of elements from detritus to soil layers. The relative change in concentration from

throughfall to CWD leachate differed among solutes, and the magnitude of the concentration increase from throughfall to CWD leachate is large, suggest that leaching is the most important processes altering solute concentrations. Accurate estimates of leaching fluxes would clarify this issue.

Concentrations of most solutes in litter leachate were only slightly greater than those in throughfall. In throughfall, concentrations of most solutes were toward or below the lower end of the range of values reported for other forested ecosystems (Yavitt and Fahey 1986; Moore and Jackson 1989; Qualls et al. 1991; Lovett et al. 2000; Solinger et al. 2001). This small difference between throughfall and litter leachate suggests that the thin litter layer (ca. 10 mm) resulted in a relatively minor effect of litter on solute concentrations at our study site. The change in DOC concentration from throughfall to litter leachate was low for DOC in comparison to other studies at sites where the forest floor layer was more prominent (Yavitt and Fahey 1986; Qualls et al. 1991; Solinger et al. 2001).

Disturbance effects of lysimeter installation have been observed to cause elevated solute concentrations in soil solutions (Shepard et al. 1990; Mitchell et al. 2001a). While it is possible that the lysimeter installations in our study also caused an artificial elevation of solute concentrations in CWD leachate, this seems unlikely for two reasons. First, installation consisted of only cutting out a central 1-m section, rolling this over, installing the lysimeter, and rolling the section back, and thus the structure of the wood itself was minimally disturbed. Second, were disturbance the cause of high concentrations, concentrations would be expected to decline over time, while in fact most solutes did not show a clear decline from initial levels. However, for some solutes, concentrations were generally higher in 2001 than in 2002.

Solute concentrations: Comparison to other studies

The concentrations of DOC that we measured in CWD leachate were higher than previously reported values for both litter and CWD leachate. Mattson et al. (1987) measured concentrations of DOC up to 15 mM, and a mean of 6.3 mM in log leachate in North Carolina, USA. Yavitt and Fahey (1985) reported maximum concentrations of ca. 17 mM and 360 μM of DOC and TDN, respectively, and means of 5.2 mM and 93 μM from CWD in a lodgepole pine forest in WY, USA. Yavitt and Fahey (1985) found that almost all the TDN leached from well-decomposed lodgepole pine logs consisted of DON ($>98\%$). Spears et al. (2003) reported concentrations lower than our values in log leachate from a range of decay classes at Andrews Forest, OR, USA. Mean DOC concentrations from Spears et al. (2003) were ca. 9 mM (data taken from Figure 1a and the text), and the mean concentration of TDN was 81 μM.

Differences between solute concentrations in CWD leachate in our study and previous studies may be due to differences in C and N concentrations across decay classes, as well as differences due to tree species or climate. Mattson et al.

(1987) collected leachate from recently dead wood, and the concentrations of DOC from decay class 1 in our study are comparable to their numbers, though N concentrations differ. Yavitt and Fahey (1985) sampled leachate from well-decayed logs, yet the DOC and dissolved N concentrations were lower than those reported here. Yavitt and Fahey's (1985) study site in WY, USA (Medicine Bow National Forest) is generally colder, and receives less total precipitation (600 mm), with most of this as snow, which melts rapidly in May and June. Therefore, it seems likely that CWD at this site in WY maintains lower temperature and moisture contents than our site in NY, and this likely influences overall decomposition rates, as well as the importance of gaseous losses vs. leaching. Lastly, both Yavitt and Fahey (1985) and Spears et al. (2003) utilized coniferous species in their study, while deciduous species were used in our study. Differences in the composition of wood and the dominant decay fungi between these two groups may influence the concentrations of solutes in leachate. Coniferous species tend to be decomposed by brown-rot fungi, which do not break down lignin, while white-rot fungi, which break down lignin along with cellulose and hemicelluloses, are more important for deciduous species (Levi 1965; Atlas and Bartha 1998).

Concentrations of DOC and DON in litter leachate range from 0.9 to 7 mM and 30–240 μM, respectively, for temperate forests (McDowell et al. 1998; Fitzhugh et al. 2001; Solinger et al. 2001; and see reviews by Michalzik et al. 2001, and Neff and Asner 2001). The concentrations from our study in CWD leachate were much higher than these values for DOC, but within the range observed for DON. The concentrations of DOC and DON in litter leachate that we measured are within the range reported for other ecosystems, but relatively low in comparison to most reported values, probably due to its thinness. The concentrations of cations, P, and TDS in CWD leachate reported in our study are generally higher than those reported for soil solutions and stream water in other forested ecosystems in the northeastern US (Yavitt and Fahey 1986; Driscoll et al. 1988; Thorne et al. 1988; Likens and Bormann 1995; Qualls et al. 1991; Likens et al. 1994; Zhang and Mitchell 1995; Likens et al. 1998; Johnson et al. 2000; Fitzhugh et al. 2001; Likens et al. 2002). This difference demonstrates that CWD has a relatively substantial impact on solute concentrations in water flowing through it, relative to other solute sources within forested ecosystems.

Solute fluxes from CWD

As explained above, an accurate method of determining hydrologic fluxes from CWD does not exist, and therefore solute fluxes from CWD were not estimated in our study. However, the cover of CWD and mean concentrations of solutes in leachate can be used to can give some insight into the potential solute fluxes from CWD. Because CWD is not an evenly distributed layer, two types of fluxes from it can be specified: a microsite flux, directly beneath a log, and an

ecosystem flux, representing the contribution of all CWD within an ecosystem. Where flooding does not contribute to hydrologic fluxes (which is probably not the case at this site), these fluxes are constrained by throughfall (though diffusive flow could make a contribution). Based on mean concentrations (Table 1), microsite fluxes of DOC (the most concentrated solute) from well-decayed wood may be as high as ca. 37-fold (25600 μM/693 μM) the flux in throughfall. Using the throughfall flux from this site, this gives a possible microsite flux of ca. 18 mol m^{-2} y^{-1}. While very high in comparison to DOC fluxes in throughfall and from litter (<4 mol m^{-2} y^{-1}, see Introduction), such a value is smaller than other fluxes of C in forested ecosystems, e.g. soil respiration rates, which may exceed 100 mol C m^{-2} y^{-1} (Raich and Nadelhoffer 1989; Bowden et al. 1993; Valentini et al. 2000). Solute fluxes from CWD are probably reduced well below this value by throughfall loss via evapotranspiration and runoff. Fluxes of other solutes from CWD would be smaller than DOC fluxes and likely less important in comparison to litter and throughfall fluxes, since DOC showed the highest concentrations of all the solutes, and also the highest relative concentrations in comparison to throughfall and litter leachate (e.g. the mean DOC concentration in CWD leachate is ca. 10-fold that in litter leachate, while the next highest solute, DOS, is <7-fold the mean litter leachate concentration). Ecosystem scale solute fluxes from CWD appear to be relatively small, since CWD covered only a small proportion of the forest floor in our study. At this site, ecosystem scale fluxes would be ca. 1/40th (0.024 ha ha^{-1}, Table 3) of microsite scale fluxes. This suggests that ecosystem scale solute fluxes are less than throughfall fluxes, and thus much lower than fluxes from the litter layer. In those ecosystems where the mass and cover of CWD are greater, the role of CWD as a source of leachate is likely to be greater. CWD mass at the study site (6.3 Mg ha^{-1}, Table 3) is substantially lower than typical values for temperate forests, which range from close to this value to more than 200 Mg ha^{-1} (Harmon and Cromack 1987; Means et al. 1992; McGee et al. 1999). Yavitt and Fahey (1985) and Mattson et al. (1987) estimated the ecosystem flux of DOC from CWD as 1 mol m^{-2} yr^{-1} for a lodgepole pine forest in WY, USA, and 1.2 mol m^{-2} yr^{-1} for a clear-cut mixed forest in NC, USA, respectively. These high values reflect the larger amounts of CWD present at these sites.

Dissolved organic matter

The solute with the highest concentration in CWD leachate was DOC, and the concentrations we measured were greater than those reported for other leachates in forested ecosystems (McDowell et al. 1998; Solinger et al. 2001, and see reviews by Michalzik et al. 2001 and Neff and Asner 2001). This observation suggests that decaying CWD and the underlying soil may be the sites of maximum DOC concentration within forested ecosystems. Previous studies and reviews of DOC dynamics in temperate forests have generally

ignored any contributions from CWD (see previous references and Currie and Aber 1997). The high concentration of DOC in CWD leachate would be expected to play a role in soil processes.

DOC collected from forest floors has been shown to generally consist of $<40\%$ labile C (Qualls and Haines 1992; Boyer and Groffman 1996; Kalbitz et al. 2000). Though measurements have not been made on DOC leached from CWD, this suggests it may serve as both a source of labile C for soil microbes, as well as play a role in soil development and other processes. Leaching of DOC from CWD would be expected to contribute to the pool of soil organic matter (SOM) below it, and some fieldwork has shown a higher concentration of humic acids beneath CWD than in soil without CWD in British Columbia (Klinka et al. 1995). Though the ecosystem scale flux of DOM from decaying wood may be small (see above), contributions over a large time scale may be significant, and the microsite contribution to SOM may be substantial, both depending on the recalcitrance of DOC from CWD.

Complexation of metal cations by DOM is an important process in the movement and availability of both nutrient and toxic cations (McCarthy and Zachara 1989; Sparks 1995). The positive correlations between DOC and cations concentrations suggest that complexation is occurring in leachate and throughfall. Thus, soluble organic matter leached from CWD could be important in controlling cation transport and availability at least on a microsite scale.

Correlations between concentrations of DOC and DON have previously been observed at an across-site scale, as well as within individual watersheds (Campbell et al. 2000; Michalzik et al. 2001; Qualls et al. 2002). The only CWD plots to consistently show positive correlations between DOC and DON over time in our study were of decay classes 2 and 3. This suggests that DOC and DON release are often controlled by different factors.

Additions of both labile and recalcitrant C to forest soils in both laboratory and field studies have shown effects on N cycling rates and availability (Groffman 1999; Magill and Aber 2000; Fierer et al. 2001). Thus, the leaching of DOC from CWD may be expected to alter C and N cycling. DOC may lead to decreased N availability both by the adsorption of potential microbial substrates, and through immobilization by increasing microbial uptake of N (Magill and Aber 2000; Fierer et al. 2001). Given the high concentration of DOC in CWD leachate, these mechanisms may contribute to reduced N availability beneath CWD.

High concentrations of DOC directly beneath CWD may affect the proportion of DOC that is lost from the soil solution, thus leading to a disproportionate contribution to ecosystem export. As the soil solution moves through soil horizons, DOC is lost due to both sorption onto soil particles and microbial degradation (Boyer and Groffman 1996; Yano et al. 2000; Qualls et al. 2002). If the affinity of soil particles for DOC (i.e., the partition coefficient) decreases as the concentration of sorbed DOC increases (i.e., a Langmuir adsorption isotherm [Sparks 1995]), higher concentrations of DOC in soil

solutions would lead to proportionally less adsorption on soil. Though some studies on DOC adsorption have demonstrated a linear response of DOC sorption to increasing solution concentration, notably at much lower DOC concentrations than those in CWD leachate (Nodvin et al. 1986; Kaiser et al. 1996; Kaiser et al. 2000), others have shown a Langmuir-type response (van Hees et al. 2003; Lilienfein et al. 2004). Microbial degradation of DOC may also be proportionally lower in soil with higher concentrations of DOC, due to a shift in limitation from C to N or some other element.

A common consequence of forestry practices is a reduction in the quantity of CWD present (Maser et al. 1988; Jurgensen et al. 1997; McGee et al. 1999). Though it is difficult to relate our results to management impacts without accurate determination of solute fluxes, it seems likely that any forestry practice that reduces CWD quantity also reduces the leaching of DOM from CWD, as well as the processes that are associated with this flux.

Controls on solute concentrations

The DOC:DON ratios, or the C:N ratios of DOM, for CWD leachates were lower than the C:N ratios of the logs from which they came for decay classes 1 and 2, and one log in decay class 3, demonstrating a relative enrichment in N in most cases. This observation could be explained by differential destruction of CWD constituents, loss of C via respiration, or gross input of N via N_2 fixation. Determining the mechanisms responsible for these patterns requires mass balances on C and N. The one outlier from this pattern, a log in decay class 3, had the highest N content (0.68%), but still the second highest DOC:DON ratio (14 g g^{-1}), suggesting that this enrichment was not controlled simply by the N content of CWD, but was likely related to the availability of N within woody tissue. This is supported by the lack of a relationship between the DOC:DON ratio and CWD N content.

The large variation in CWD leachate DOC concentrations over the year and the poor correlation to precipitation rate suggests the possibility of biological control of DOC concentrations. The general pattern of CWD DOC concentrations over the year, with the lowest levels in winter, is compatible with biological control, though CWD DOC concentrations did not show a significant correlation to mean air temperature. The variable effect that air temperature may have on biological activity within CWD, through altering e.g. log temperature and available water, as well as the shortcomings of using mean temperatures and concentrations, could explain the lack of a consistent response. Precipitation would also be expected to have variable effects on solute concentrations from CWD, since water may promote and decrease biological activity, affect oxygen status, and dilute dissolved constituents. The ratio of DOC:DON did show a positive correlation to precipitation rate for throughfall, litter leachate, and CWD leachate, suggesting a hydrologic control on the C:N ratio of DOM. However, a mechanism through which increased precipi-

tation leads to an increase in DOC leaching relative to DON leaching is not forthcoming. In addition to the possible effects of temperature and precipitation, DOM concentrations in litter would be expected to respond to changes in the quantity of litter present. The increase in the quantity of litter due to leaf fall likely explains the spike in DOC in litter leachate from the November 23, 2002 samples. The observed positive correlation of DON in litter leachate to air temperature suggests that temperature is controlling DON leaching via activity of decomposer organisms.

In general, correlations between inorganic (or, in the case of P, undifferentiated) solutes and temperature were common in throughfall, suggesting biological controls on solute leaching from the canopy. Similar correlation has been observed elsewhere (Solinger et al. 2001). Less correlation, as well as higher variability in general, for the leachates reflect a more complicated control of mineralization and leaching from litter and CWD. The observed negative correlations of SO_4^{2-}, NO_3^-, and Cl^- to precipitation suggest a simple dilution effect, and hydrologic control of concentrations.

Nitrogen dynamics

Inorganic forms of dissolved N, NH_4^+ and NO_3^-, did not show a clear increase from throughfall to leachate. Though NH_4^+ concentrations should be interpreted cautiously due to sample storage at 4 °C (Klingaman and Nelson 1976; Yorks and McHale 2000), the concentration of NH_4^+ showed a statistically significant decline from throughfall to litter leachate. The decrease in NH_4^+ could be due to nitrification in the litter layer, but the lack of an increase in inorganic N in general suggests that it is conserved to some degree by decomposers in decaying detritus. This observation is what would be expected given the generally high C:N ratio of litter and CWD (i.e. 20–90 for leaf litter and 90–1000 for woody tissue) (Daubenmire and Prusso 1963; Cowling and Merrill 1966; Lang and Forman 1978; Yavitt and Fahey 1986), and is compatible with the observation that N concentration, and often the total mass of N, may initially increase in decaying litter and wood (Grier 1978; Melillo et al. 1982; McClaugherty et al. 1985; Means et al. 1992). Factors that control the loss of N from CWD are clearly more complicated than just the N concentration of wood.

Despite the lack of an increase in inorganic N concentrations in CWD and litter leachate relative to throughfall, TDN concentrations were much greater in CWD leachate than in throughfall. The increase in TDN in CWD leachate was due to the high concentrations of DON. The impact that N leaching has on soil microbiology needs to be assessed in light of the difference between the flux of N from CWD and from litter, as well as the availability of N in CWD leachate. Little work has been done on the biodegradability of DON (Ohta et al. 1986; Qualls and Haines 1992; Scherer et al. 1992), and it is not clear whether DON is less or more labile than DOC Kalbitz et al. 2000). However, DON would be expected to be less available than inorganic N.

Conclusions

The high concentrations of DOC and, to a lesser extent, other solutes, in CWD leachate from this site demonstrate that CWD is a source of DOC and other solutes in this lowland forest. While contributions of CWD to ecosystem scale fluxes are probably small at our study site due to a small mass of CWD, microsite scale fluxes of DOC have the potential to be very high, with numerous potential impacts. Leaching from CWD should be more closely studied for its contribution to nutrient cycling within forested ecosystems, especially where CWD is a larger component of the forest floor.

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