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Treatment of Log Yard Runoff in an Aerobic Trickling Filter

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Contaminated stormwater runoff from log yards is generated when precipitation comes into contact with wood, woody debris and equipment at outdoor wood sorting, processing and storage facilities. Nine runoff samples collected at a sawmill had biochemical oxygen demand (BOD), chemical oxygen demand (COD), tannins and lignin (T+L), and total suspended solids (TSS) levels ranging from 25 to 745 mg/L, 125 to 4610 mg/L, 10 to 1505 mg/L, and 65 to 2205 mg/L, respectively. Six samples were acutely toxic (EC50 <100%) based on the Microtox assay.

The samples were effectively treated using a laboratory-scale, attached microbial growth reactor. Treatment for 24 hours at 34°C resulted in substantial reductions in BOD (94–100%), COD (86–93%) and T+L (91–97%). Near complete removal of acute toxicity and colour were also observed. Twenty-four-hour treatment at lower temperatures, 24 and 5°C, reduced BOD concentrations by 97 and 76%, COD by 91 and 64%, and T+L by 95 and 67%, respectively.

Key words: **log yard, runoff, trickling filter, biofilm, biological treatment, wood extractives**

Introduction

Raw logs are often stored and processed at outdoor log yards, where they are exposed to precipitation. Precipitation falling on a log yard also comes into contact with machinery and buildings, as well as large quantities of bark and woody debris. The stormwater runoff generated at a log yard is frequently deeply coloured and may contain suspended solids, foam, oil and/or grease. Runoff can also be generated from water sprinkled to prevent fire, used to clean equipment, or carried over as raw logs are pulled from a river or the ocean (Samis et al. 1999).

Log yard stormwater runoff is often toxic and can exceed regulated water quality parameters for industrial discharge (Bailey *et al.* 1999a,b; Borga et al. 1996; McDougall 1996; Zenaitis and Duff 2002). The strength of log yard runoff varies considerably; biochemical oxygen demand (BOD), chemical oxygen demand (COD), and total suspended solids (TSS) levels range from 6 to 4950 mg/L, 11 to 14,723 mg/L, and up to 20,077 mg/L, respectively (McDougall 1996; deHoop et al. 1998). Runoff toxicity varies from non-toxic to highly toxic (McDougall 1996; Bailey et al. 1999a). In a study of British Columbia (B.C.) sawmills, Bailey *et al.* (1999a,b) found that 72 to 96% of runoff samples were toxic to rainbow trout. Approximately 85% of B.C. wood processors recently surveyed (Orban et al. 2002) produced a visible runoff and most applied only primary treatment (debris removal, oil/water separation) or no treatment to the runoff stream.

While many potential treatment options have been proposed for log yard runoff (Samis et al. 1999), few have been tested. Ozonation has been shown to effectively decrease acute toxicity, but had little effect on COD or BOD (Zenaitis and Duff 2002). Borga *et al.* (1996) showed that recycled water at a log yard supported biological activity, and that the biological activity resulted in improved water quality. Batch biological treatment, using seed from a pulp mill activated sludge treatment system, reduced BOD, COD and tannin and lignin (T+L) concentrations by 99, 80 and 90%, respectively (Zenaitis et al. 2002). Acute (Microtox) toxicity decreased during the treatment, from an initial EC50 of 1.83% to a value of 50.4% after 48 hours of treatment. Pre- or post-biological treatment ozonation produced modest improvements in final effluent quality. Frankowski (2000) treated cedar log fuel leachate in both laboratory- and pilot-scale constructed wetlands. Substantial reductions in BOD, COD and T+L concentrations (63 to 94%), as well as toxicity removal were achieved. However, contaminant and toxicity removal decreased to 20 to 49% in pilot-scale trials.

Aerobic attached-growth biological systems offer many potential advantages over conventional biological treatment (Gavrilescu and Macoveanu 2000; Lazarova and Manem 1996), and are often more economical (Parker 1999). The objectives of this study were to characterize runoff from a Vancouver Island log yard, and investigate the effectiveness of aerobic attached growth biological treatment at reducing runoff BOD, COD, T+L and toxicity.

Materials and Experimental Methods

Runoff Sample Collection and Characterization

Stormwater runoff for this project was collected from the paved log yard at a sawmill on Vancouver Island,

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B.C., which is adjacent to a river. Outdoor processing of logs generates bark and wood debris which covers the surface of the yard. Rainfall becomes contaminated through contact with the debris, then runs off the site into the river. A runoff treatment system is planned.

Sample #1 (Table 1) was collected from standing water after a light rain event. Samples #2 to 9 were collected during rain events, from the runoff flowing into the Courtenay River. The samples were delivered to the laboratory within two to four days of collection.

To minimize degradation during storage, the pH of each sample was lowered to less than 2 with sulfuric acid. The samples were stored in the dark, at 5°C, in high-density polyethylene containers.

Attached-Growth Batch Reactor Design

The attached-growth batch reactor (Fig. 1) consisted of a 2-L mixing vessel and a 1-L trickling filter vessel, both made of Pyrex glass. A bed of 48 plastic biofilm support media pieces (Hydroxyl Systems Inc., Victoria, B.C.) filled the bottom half of the trickling filter vessel. A VWR Scientific model 1137 water bath heated the jacketed trickling filter vessel to 34°C unless otherwise specified.

Liquid cycled continuously between the mixing and trickling filter vessels during the batch treatment of each sample. A Masterflex speed-controlled peristaltic pump continuously pumped liquid through Masterflex PharMed tubing from the mixing vessel up to a stainless steel, atomizing nozzle (1/4 LNN, Spraying Systems Co., Wheaton, Illinois) suspended about 10 cm above the support media bed. The liquid passed through the nozzle at a flowrate of approximately 0.21 L/min, gently sprayed over the support media, and drained from the bottom of the trickling filter vessel through Masterflex Tygon tubing into the mixing vessel, completing the liquid cycle.

Biofilm Growth Phase

To colonize the support media, a mixture of runoff, methanol, nutrients and microbial seed was continuously circulated through the system for 41 days prior to the onset of treatment trials. Runoff (sample #7, centrifuged at 1360 × g for 10 min) was augmented with methanol to provide the biofilm with an easily degradable carbon source while acclimating it to the runoff (Annachhatre and Bhamidimarri 1992; Burgess et al. 1999). Methanol, nutrients and potassium bicarbonate (to provide alkalinity) (solution A, Table 2) were added to the centrate. The microbial seed, return activated sludge (RAS) from Pope and Talbot's Harmac Kraft Pulp Operations in Cedar, B.C., was suspended in the solution for a final concentration of 2240 mg/L in the reactor. The final pH of the mixture was adjusted to 6.5 before pouring the liquor into the mixing vessel.

The biofilm colonized the media and grew for 41 days (biofilm growth phase) before the first runoff treatment trial was conducted. During this phase, 1 to 1.5 L of solution B (Table 2) was continuously circulated through the system, wetting the biofilm. A methanol and nutrient stock solution was pumped into the mixing vessel every five hours, to yield the concentrations described. Runoff centrate was also added to the system every two to four days by hand or with a third peristaltic pump. Chrontrol XT timing devices regulated the automated additions. The mixing vessel pH was constantly monitored with a Cole-Parmer digital bench-top pH meter (model 05669-20) and a 30 cm-long, Orion gel-filled combination pH electrode (model 912600). The addition of alkalinity to the system was sufficient to maintain the liquid in the reactor at a pH level between 6 and 8. The biofilm grew in abundance and frequently clogged the bottom of the trickling filter, the tubing and the nozzle. Excess suspended growth was cleared every four to five days and discarded.

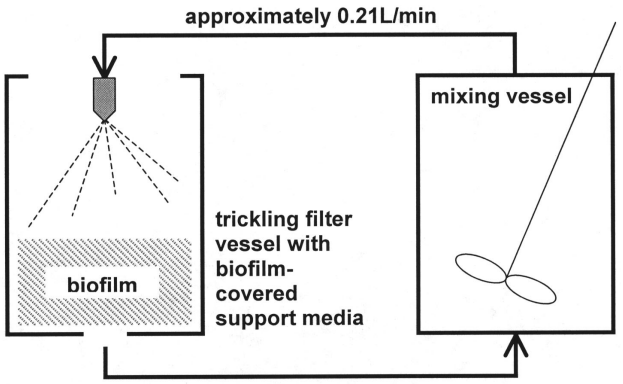

Fig. 1. Aerobic trickling filter schematic. Liquid flows from the mixing vessel to the trickling filter vessel at a flow rate of about 0.21 L/min, is sprayed over the biofilm-covered support media, and cycles back into the mixing vessel.

TABLE 1. Dates and mill furnish for runoff sample collection

Sample number	*Sample date*	*Mill furnish at time of sample*
1	May 4, 2001	Hemlock
2	Oct. 10, 2001	Douglas Fir
3	Oct. 10, 2001	Douglas Fir
4	Oct. 10, 2001	Douglas Fir
5	Oct. 10, 2001	Douglas Fir
6	Nov. 21, 2001	Douglas Fir
7	Nov. 21, 2001	Douglas Fir
8	Nov. 21, 2001	Douglas Fir
9	Apr. 16, 2002	Douglas Fir

Runoff Treatment Trial Phase

Runoff treatment trials were conducted over a period of 120 days. Only the higher strength samples, #2, 3, 4, 7

TABLE 2. Solutions A through E, which were used during the biofilm growth and runoff treatment phases

	Concentration (mg/L)				
Constituent	*A*	*B*	*C*	*D*	*E*
Methanol	214	180–270	0	214	0
$(NH_4)_2SO_4$	120	80–120	40	120	81
NaH_2PO_4	19	10–20	17	19	34
$KHCO_3$	130	90–130	65	130	130
Runoff (% v/v)	50	0–100	66	0	100

and 8, were chosen for treatment in the trickling filter. Before treatment, the runoff samples were adjusted to their original pH (Table 3) and solids were removed by centrifugation at 1360 × g for 10 min. The reactor was cleared of excess suspended growth before each treatment trial. In between trials, the biofilm was maintained with substrate and nutrients as previously described.

Each runoff treatment trial consisted of: 1) overnight biofilm acclimation to the runoff sample and temperature under investigation (solution C, 16 to 22 h); 2) methanol removal to roughly gauge the activity of the biofilm at the time of the trial (solution D, 80 to 90 min); and 3) runoff treatment (solution E, 24 h). The tricking filter vessel temperature, flow rate through the nozzle, and runoff sample used were kept constant through the three steps. All of the liquid was removed from the reactor after each step.

During the second step, which generated a methanol degradation rate for the biofilm at the time of each runoff treatment trial, samples were taken from the mixing vessel at 0, 10, 20, 40 and 80 or 90 min. The methanol samples were centrifuged at 8385 × g for 10 min in a Sanyo MSE MicroCentaur centrifuge and stored in glass vials in the fridge or freezer until analysis.

Prior to the third step, nutrients were added to 1.2 L of runoff centrate, according to solution E (Table 2), to achieve a BOD:N:P ratio equal to 100:5:1 (Annachhatre and Bhamidimarri 1992). The pH of the solution was then adjusted to 6.5, before pouring the solution into the mixing vessel. No additional runoff or nutrients were added to the reactor during the trial. The runoff solution was cycled through the reactor system for about five minutes before the first (time = 0 h) sample was taken. Samples of 25 to 30 mL were taken from the mixing vessel at 0, 1, 2, 4, 8, 12 and 24 h. The system was monitored to ensure that clogging from excess biofilm growth did not occur and that the nozzle flow rate remained relatively constant throughout the trial.

Runoff samples taken from the mixing vessel during the treatment trials were immediately centrifuged at 887 × g for 10 min. The centrates were transferred to clean FisherBrand 50-mL centrifuge tubes for storage at 5°C until analysis. All seven samples from a treatment trial were analyzed together immediately following the trial. The samples were analyzed for BOD, COD, T+L and Microtox toxicity.

Treatment at Sub-optimal Temperatures

The first sub-optimal temperature runoff treatment trial was conducted by switching off the water bath and allowing the reactor to operate at an ambient temperature of approximately 24°C. The reactor system was transferred to a cold-storage room, where it operated at

TABLE 3. Characterization of runoff samples from the Vancouver Island sawmill for the present study

Sample number	*pH*	*BOD (±) (mg/L)*		*COD (±) (mg/L)*		*T+L (±) (mg/L)*		*Toxicity EC50 (95% C.I.) (% v/v)*
		Total	*Soluble*	*Total*	*Soluble*	*Total*	*Soluble*	*Soluble*
1	7.00	335 (35)	325 (35)	1400 (15)	1360 (45)	—	435 (5)	13 (11 to 15)
2	5.36	485 (30)	395 (20)	3190 (75)	1710 (40)	770 (25)	600 (10)	7.11 (2.9 to 18)
3	5.23	—	—	4610 (55)	1780 (20)	815 (20)	470 (5)	—
4	4.85	745 (15)	660 (15)	4485 (90)	2995 (75)	1505 (30)	1210 (10)	4.35 (2.2 to 8.5)
5	6.29	30 (10)	0 (5)	125 (15)	50 (5)	15 (0)	10 (0)	>100
6	6.40	50 (5)	35 (15)	1180 (50)	260 (30)	100 (5)	65 (5)	91 (1.0 to 8367)
7	5.09	685 (45)	625 (45)	4010 (105)	2370 (25)	785 (5)	685 (5)	4.9 (2.6 to 9.2)
8	4.96	575 (10)	515 (25)	3590 (175)	2100 (10)	845 (15)	655 (10)	5.1 (2.8 to 9.4)
9	5.64	25 (5)	25 (5)	135 (10)	40 (30)	10 (0)	5 (0)	>100

a temperature of 5°C, for the second sub-optimal temperature trial. The second sub-optimal temperature trial was conducted after biofilm-covered support media were removed from the trickling filter for biomass quantification experiments (not described here). For this reason the trickling filter vessel contained only 28 biofilm-covered support media during the 5°C trial (previous trials were conducted with 48 media pieces).

Control Trial without Biomass

After all of the experimental work with the attached-growth reactor had been completed, the reactor was completely cleared of biomass. The vessels were cleaned with detergent and the tubing replaced. New support media was added to the trickling filter vessel. A runoff treatment trial without biomass was then conducted at 34°C, using sample #2.

Analyses

BOD, COD, TSS and T+L were assayed in triplicate according to procedures 5210B, 5220D (closed reflux), 2540D and 5550B of Standard Methods (American Public Health Association 1992). Acute toxicity was assessed using the Microtox basic testing protocol (Microbics Corp. 1992). Since, after dilution, the runoff samples were only slightly coloured, colour correction was not done for the Microtox assay.

Methanol concentrations were determined in triplicate using a Varian CP-3800 gas chromatograph equipped with a Supelco (Supelcowax-10 24080-U) capillary column (fused silica, 30 m x 0.32 i.d. x 0.25 µm film thickness). Helium, hydrogen and air flow rates were 25, 30 and 300 mL/min, respectively. For all determinations 1-butanol was used as the internal standard. The detection method consisted of initially holding the column oven temperature at 45ºC for 2 min then ramping at 20ºC/min up to 65ºC and holding for 2.75 min. The flame ionization detector and injector were both maintained at 250ºC.

Results and Discussion

Characterization of Runoff Samples

The nine runoff samples varied in strength and physical appearance (Table 3). The samples ranged from light beige-grey in colour and slightly turbid, with some small black solids to dark brown to black in colour and more turbid, with larger wood pieces and a distinct woody odour. In four previous samplings at the same site, BOD, COD, T+L and Microtox EC50 values ranged from 300 to 1900 mg/L, 2380 to 8760 mg/L, 510 to 1550 mg/L and 1.86 to 16.1 % v/v, respectively (Zenaitis et al. 2002; Zenaitis and Duff 2002). The parameter values from the current work were typically in the low end of these ranges.

Biofilm Development and Performance Over Time

A brown biofilm began to colonize the support media within 4 days, and was allowed to develop for 41 days before runoff treatment trials commenced. The biofilm may still have been developing throughout the runoff treatment phase, as the start-up period for biofilms can often take up to several months (Annachhatre and Bhamidimarri 1992). Biofilm activity was gauged by the results of the methanol removal trials. The average methanol degradation rate (at 34ºC) was 1.59 ± 0.50 mg/L min.

Runoff Treatment

Batch treatment of runoff resulted in rapid and complete toxicity removal, as well as substantial decreases in soluble BOD, COD and T+L concentrations (Fig. 2). Effluent pH increased from 6.5 to about 7.7 over the course of the 24 h of treatment. Substantial colour reductions in runoff samples were also observed during the treatment trials. Deeply coloured orange or purple runoff became almost colourless by the end of each trial (Fig. 3). Coloured runoff components may have been degraded by or sorbed to the biomass. The results from all of the treatment trials are similar to those presented and a summary of the results for all trials is given in Table 4. The first-order rate constant for biodegradation of each of the measured parameters was calculated (Fig. 4). There was no relationship between the soluble start of treatment BOD, COD or T+L concentrations with the first-order rate constants.

Table 4 also includes data from runoff treatment trials described in the literature. The attached-growth

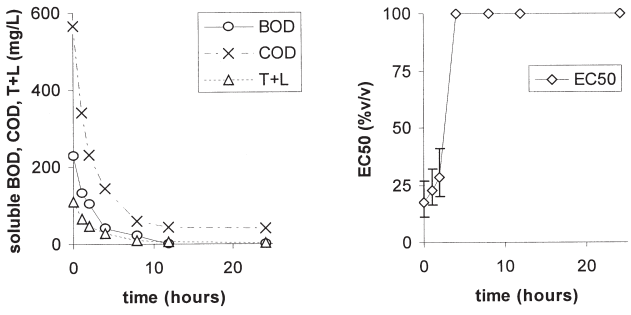

Fig. 2. Degradation of runoff sample #2 (at time of sampling, total BOD = 485 mg/L, COD = 3190 mg/L, T+L = 770 mg/L, soluble EC50 = 7.1%) during trickling filter treatment. Standard deviations are less than the size of the data points for BOD, COD and T+L. Ninety-five percent confidence intervals are shown for the EC50 data. EC50 values were greater than 100% after 4 h.

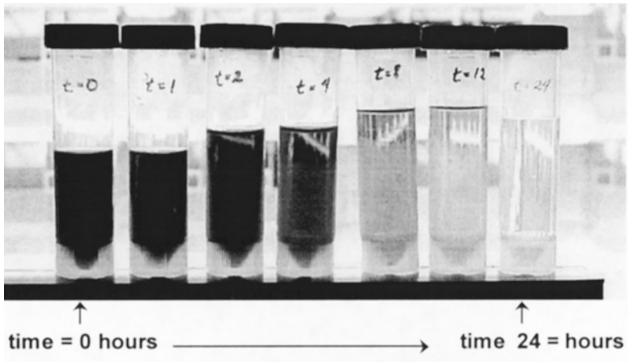

Fig. 3. A picture of all seven samples taken after the treatment of runoff sample #3 (at time of sampling, total COD = 4610 mg/L, T+L = 815 mg/L).

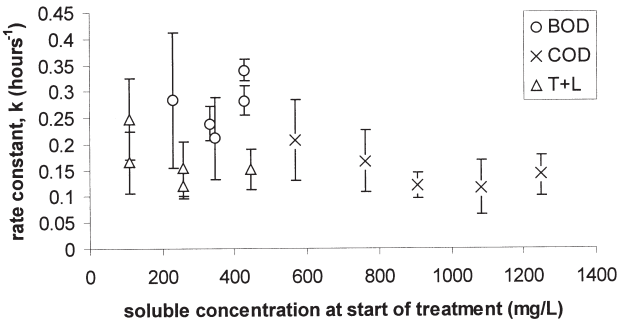

Fig. 4. First-order degradation rate constant as a function of initial runoff strength for all samples treated (error bars represent 95% confidence intervals). For each point (x,y), x is the BOD, COD or T+L concentration at the start of the degradation trial, and y is the rate constant for the corresponding BOD, COD or T+L removal rate.

reductions of BOD, COD and T+L concentrations are similar to results from 48-h runoff treatment trials in an aerated suspended growth batch reactor (Zenaitis et al. 2002). The removal of T+L by the attached-growth reactor was also similar to (but much slower than) that observed in ozonation trials (Zenaitis and Duff 2002). However, the attached-growth reactor removed an average of 72% more BOD and 52% more COD from runoff than did ozonation. Zenaitis and Duff (2002) note that ozonation of the runoff reduced toxicity by 86.2 ± 2.6% and the dehydroabietic acid (a wood extractive) concentration by 100%.

Treatment of Runoff at Sub-optimal Temperatures

Because precipitation events occur in British Columbia primarily during the winter months, it was desirable to characterize the performance of the fixed film bioreactor at sub-optimal temperatures. The soluble BOD, COD and T+L concentrations were almost completely removed in 24 h of treatment at 24°C (Table 5). There was no significant difference in the rate of removal of BOD, COD and T+L between trials conducted at 24 and 34°C (Fig. 5). However, it took longer (8 h) to render the effluent non-toxic at 24°C, as compared to 4 h at 34°C.

The biofilm was less active at 5°C (Fig. 5). This may have been due to the decreased amount of biomass in the reactor during the 5°C trial, however previous methanol removal rates were not affected by biomass amount (work not presented here). Significant removal of runoff components was achieved at 5°C: 76% of BOD, 64% of COD and 67% of T+L was removed in 24 h at this temperature. Runoff toxicity was not completely eliminated during 24 h of treatment at 5°C; the EC50 after 24 h was 69% (95% confidence interval: 21 to 230%).

TABLE 4. Extent of degradation and final concentration achieved for runoff samples treated in the trickling filter at 34°C and compared to literature values

	Soluble BOD (±)		*Soluble COD (±)*		*Soluble T+L (±)*	
Sample	*Reduction (%)*	*24 h (mg/L)*	*Reduction (%)*	*24 h (mg/L)*	*Reduction (%)*	*24 h (mg/L)*
#2 without biomass	11 (9.9)	246 (20.4)	5 (0.9)	573 (4.4)	32 (1.1)	77 (0.8)
#2	100 (1.7)	0 (3.8)	93 (1.3)	40 (5.8)	97 (0.3)	4 (0.3)
#3	94 (6.4)	22 (22)	91 (0.7)	71 (4.5)	96 (0.5)	7 (0.8)
#4	100 (1.9)	0 (8)	92 (1.1)	106 (13)	96 (0.2)	19 (1.0)
#7	98 (1.7)	7 (7)	86 (0.4)	147 (4.4)	95 (0.8)	14 (2.1)
#8	97 (1.1)	9 (4)	88 (0.4)	113 (3)	91 (0.2)	24 (0.5)
Average	98 (1.4)	8 (5.0)	90 (0.4)	95 (3.2)	95 (0.2)	14 (0.5)
Suspended[a]	99[c]	32[c]	80[c]	1046[c]	90[c]	132[c]
Ozonation[b]	25	n.a.	35	n.a.	90	n.a.

[a]Zenaitis et al. (2002).
[b]Zenaitis and Duff (2002), final values not available.
[c]48 h of treatment in a suspended biomass reactor.

TABLE 5. Extent of degradation and final concentration achieved for runoff samples treated in the trickling filter at 5°C (sample #2) and 24°C (sample #3), and compared with previous results at 34°C

	Soluble BOD (±)		*Soluble COD (±)*		*Soluble T+L (±)*	
Temperature (°C)	*Reduction (%)*	*24 h (mg/L)*	*Reduction (%)*	*24 h (mg/L)*	*Reduction (%)*	*24 h (mg/L)*
Sample #3						
24	97 (0.4)	12 (1.4)	91 (0.3)	75 (2.2)	95 (0.2)	8 (0.2)
34	94 (6.4)	22 (22)	91 (0.7)	71 (4.5)	96 (0.5)	7 (0.8)
Sample #2						
5	76 (5.6)	59 (12.9)	64 (3.3)	201 (15)	67 (0.2)	40 (0.2)
34	100 (1.7)	0 (3.8)	93 (1.3)	40 (5.8)	97 (0.3)	4 (0.3)

Conclusions

Five log yard runoff samples were treated using an aerobic attached-growth, cycling batch bioreactor. In 24 h of treatment, BOD concentrations were reduced by 94 to 100%, COD by 86 to 93% and T+L by 91 to 97%. Complete toxicity removal and substantial colour reductions were also observed. Significant runoff degradation was achieved over 24 h of treatment at 24 and 5°C, with reductions in BOD concentrations of 97 and 76%, COD of 91 and 64%, and T+L of 95 and 67%, respectively. Toxicity was completely removed after 8 h of treatment at 24°C, and substantially reduced after 24 h of treatment at 5°C.

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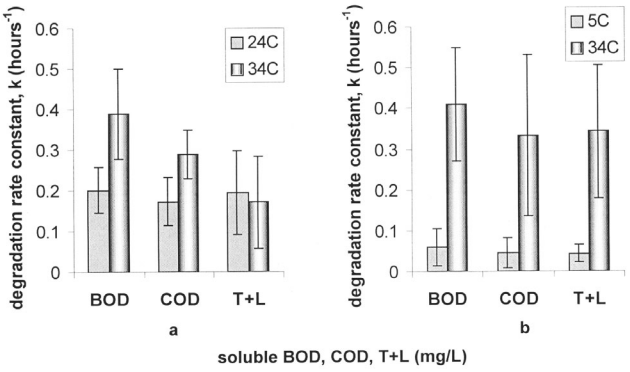

Fig. 5. First-order rate constants for the degradation of runoff samples #3 (a) and #2 (b) at different temperatures (error bars represent 95% confidence intervals).

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