

Translocation of soil organic matter following reservoir impoundment in boreal systems: Implications for in situ productivity

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Abstract

To evaluate the effect of reservoir flooding on carbon cycling over time, we studied sedimentary environments in three natural lakes that existed prior to impoundment and that have been incorporated in the larger lentic system of a 70-yr-old boreal reservoir in Quebec. Elemental and biomarker analyses were determined in all core intervals and three soil profiles to characterize source inputs of organic matter to sediments. Following impoundment, a two- to threefold increase in lignin concentrations (5 to 10–25 mg [10 g dry wt]⁻¹) associated with similar decreases in selected terrigenous biomarker ratios (cinnamyl phenols: vanillyl phenols, C:V, 1.0–2.0 to 0.2–0.5; 3,5-dihydroxybenzoic acid: vanillyl phenols, 3,5-Bd:V, 0.2–0.5 to 0.1–0.2; and p-hydroxyl phenols: sum of vanillyl and syringyl phenols, P:[V+S], 0.3–0.7 to 0.1–0.4) illustrate the effect of soil erosion and subsequent translocation of surface soil organic matter (SOM) to sedimentary deposits. Using a mixing model based on mass-normalized biomarker yields in identified surface and mineral soil end-members, we show that although the proportion of mineral-derived SOM predominates in the receiving sedimentary systems during preflooding conditions (97–99% of allochthonous inputs), translocated surface SOM increased in postflooding sediments to comprise up to 5–30% of the total allochthonous organic matter inputs. Using a similar quantitative mixing model based on elemental C and N contents in the autochthonous and the mixed allochthonous end-members, we estimated that concomitant to the redistribution of eroded soil organic matter, reservoir flooding induces a 1.5- to 2-fold increase in the fraction of autochthonous organic matter transferred to sedimentary environments of the reservoir. Results presented here suggest that reservoir flooding induces a change of state in lake systems that may perdure over time scales of decades to centuries.

Large-volume reservoirs, built primarily for hydroelectric power generation, are among the largest man-made water infrastructures and hold at any given time 10–20% of the global mean river runoff (Rosenberg et al. 1997; Vörösmarty and Sahagian 2000). Beyond their recognized disruptions of water flow and impairment of water quality (Rosenberg et al. 1997; Hambright et al. 2004), large-scale reservoirs also result in significant shifts in trophic structure during the transition from a primarily lotic to a lentic system (Kalff 2002). Indeed, the creation of new reservoirs frequently leads to short-term spikes in nutrient levels and anomalously high in situ productivity including sharp increases in bacterial bio-

mass (Schetagne 1994; Paterson et al. 1997; Hall et al. 1999). One process invoked to explain such ecological change is that large-scale erosion and leaching of organic matter and associated nutrients from surface flooded soils (Kennedy and Walker 1990; Paterson et al. 1997) entail a relatively short-term fertilization and a shift in natural productivity and food web structure (Paterson et al. 1997; Kalff 2002). Following this first phase in “trophic upsurge,” a gradual decline or “trophic depression” is predicted to occur and yield a more stable community composition and community (Kalff 2002), although the time of onset of such trophic depression is yet ill defined.

Additionally, the induced increases in carbon cycling within these systems has recently been recognized as a significant contributor of greenhouse gas (GHG) emissions to the atmosphere (Duchemin et al. 2002; Bodaly et al. 2004; Soumis et al. 2004). This latter effect has been the focus of increasing attention from both the research community and the private energy sector because hydroelectric dams have historically been perceived as carbon-free alternatives to power generation and accounted as such in energy production scenarios (Hoffert et al. 1998; Victor 1998). Sustained degradation of flooded soil organic matter (SOM) was initially proposed as a working hypothesis to explain observed supersaturation in CO₂ and CH₄ that led to increased evasion

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rates from reservoirs' water columns (Rudd et al. 1993; Kelly et al. 1997; Rosenberg et al. 1997). However, contrary to this view, carbon stocks in flooded soils do not seem to degrade at the rate necessary to sustain GHG emissions observed over several decades in these systems (Weissenberger et al. 1999; Houel 2003). Furthermore, carbon gas emissions show poor correlations with total carbon stored in flooded soils and peats (Huttunen et al. 2002; Bodaly et al. 2004), suggesting that GHG emissions in recently flooded reservoirs are more related to a discrete carbon stock present in fresh plant or litter biomass, rather than to the decomposition of recalcitrant soil and peat organic carbon (Huttunen et al. 2002; Bodaly et al. 2004). An alternative explanation for long-term evasions of GHG from reservoir systems proposes that following a short-lived pulse in soil organic matter degradation (Houel 2003), emissions are supported by increased turnover of organic matter produced outside a reservoir's boundaries (allochthonous or exogenous) and flowing through the system (Weissenberger et al. 1999; Bodaly et al. 2004). In natural lakes, recent studies on net ecosystem production (NEP) and food web dynamics have shown that inputs of allochthonous carbon support net heterotrophy and can contribute up to half of the carbon sources used by secondary production (Cole et al. 2002; Pace et al. 2004). Although the substantial subsidy of NEP by what is considered recalcitrant organic matter is somewhat of a paradox (c.f. Hedges et al. 1997), more evidence points to high rates of mineralization of this material in aquatic systems (Cole and Caraco 2001; Richey et al. 2002; Pace et al. 2004).

Boreal reservoirs do indeed receive higher proportions of detrital terrigenous organic matter from the surrounding landscape than natural lakes, and during early stages of impoundment this material acts as the predominant carbon source that fuels local food webs (Paterson et al. 1997; Montgomery et al. 2000). Over time, however, the pathway of carbon flow to secondary producers has also been shown to shift from an exclusive allochthonous base to include increased phytoplankton production suggesting dynamic ecosystem responses in these systems (Paterson et al. 1997). Hence, considering that GHG emissions remain high in some reservoirs close to 100 yr after their impoundment (St. Louis et al. 2000; Duchemin et al. 2002), the influence of flooding on ecological shifts and carbon cycling in these systems may thus need to be constrained on time scales of decades to centuries, rather than a few years.

Among the few studies that have addressed the influence of impoundment on flooded soils, most have focused on identifying sources of mercury to reservoir food webs (Louchouart et al. 1993; Montgomery et al. 2000), whereas only a few were directed at balancing GHG emissions with changes in soil carbon stocks (Weissenberger et al. 1999; Houel 2003). To our knowledge, however, none has assessed the fate of eroded soil constituents and their effect on long-term trophic status of flooded systems. In boreal reservoirs, Houel (2003) showed that erosion was the predominant process of carbon losses from flooded soils contributing to a translocation of ~30% of the original organic horizon carbon loads (g C m^{-2}) within the first 10 yr of impoundment. Assuming that some of that carbon is not remineralized in or flushed out of the system, we should then observe a re-

distribution of this material to sedimentary deposits providing a record of past erosional inputs to the aquatic system. Multiproxy studies involving molecular, isotopic, and elemental signatures of sedimentary organic matter have been used successfully in the past to reconstruct shifts in water column productivity and anthropogenic organic matter inputs to lake sediments due to large-scale transformations in their basins such as overload of agricultural fertilizers and increased urbanization (Filley et al. 2001; Routh et al. 2004). However, we are aware of only a few studies that have used soil signatures to reconstruct allochthonous organic matter inputs to aquatic systems with a particular emphasis on tracing disturbances to their watersheds (Louchouart et al. 1999; Farella et al. 2001). Using soil signatures, rather than fresh plant end-members, during source reconstructions carries the considerable advantage of integrating diagenetic changes that may have altered terrigenous organic matter prior to its introduction to the aquatic environment and permits a more meaningful reconstruction of terrigenous organic matter inputs to aquatic systems. For example, the contrastingly different compositions of organic matter in surface versus deep tropical soils have allowed Farella et al. (2001) to document the drastic effect of deforestation and erosion on terrigenous organic matter fluxes to Amazonian river/lake systems from normal surface litter-derived inputs toward that of deep soil horizons. Using a similar approach, we report and quantify the shifts in inputs of allochthonous organic matter to lake sedimentary deposits following reservoir impoundment. Additionally, these results suggest that concomitant to the redistribution of eroded soil organic matter, reservoir flooding induces a significant increase in the proportion of autochthonous organic matter transferred to sedimentary environments of the reservoir.

Study area—Cabonga Reservoir (434 km²), created in 1928, is located in the east-central region of La Verendrye Wildlife Natural Park, approximately 350 km northwest of Montreal, Canada. To evaluate the influence of flooding on carbon cycling over time we selected three natural lakes that existed in the area prior to impoundment and that have been incorporated in the larger lentic system of the reservoir. In each lake, we collected a sediment core in the old sedimentary deposit at stations identified as 1001, 1200, and 999 (Fig. 1). Sta. 1001 is located in a small bay near a native reservation, the only permanent human settlement on the shores of the reservoir. Of the remaining two stations, one was selected to represent the most open zone of the reservoir (Sta. 1200), whereas the other is situated in a narrow channel surrounded by a high abundance of islands (Sta. 999). The depth of the water column at the different stations is comparable and ranges between 10 and 12 m.

Materials and methods

Sampling—The sediment cores were sampled using a Mackereth corer (Mackereth 1958) in September 1998 at Sta. 1001 and in July 1999 at the other two stations. Each core measured over 65 cm, but we analyzed only the first 45–50 cm. Subsampling was performed at every centimeter interval by removing the sediments in contact with the cor-

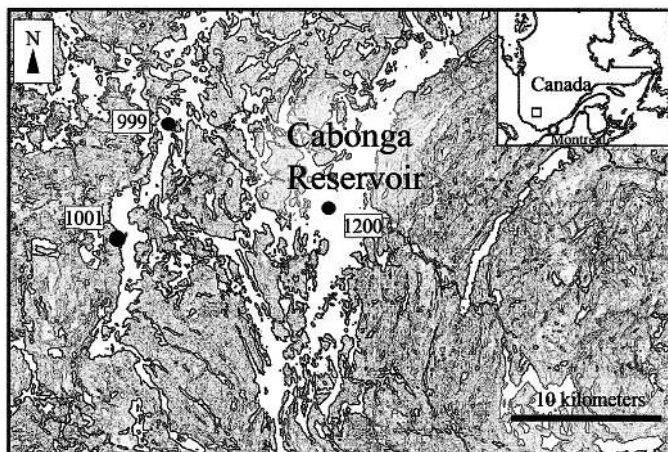


Fig. 1. Location of sampling stations in Cabonga Reservoir. From Landsat 7 image, 18 July 2002.

ing tube to prevent cross contamination and introduction of a stratigraphic artifact through friction along the walls of the tube. Following subsampling, sediments were freeze-dried and homogenized prior to analysis. The bulk density of each sediment interval was calculated using the measured water content of each interval and an assumed dry grain density of 2.5 g cm^{-3} (Berner 1971). Each sedimentary record recovered the pre- and postflooding accumulation period. Preflooding sediments represent conditions when each lake had its own watershed and internal dynamics, whereas postflooding is characterized by sediments accumulated in the interconnected dendritic reservoir and which integrate the perturbations caused by the creation of the Cabonga Reservoir ~ 70 yr prior to sampling. Two soil litters were also sampled near Sta. 1001. Additionally, six characteristic soil profiles (podzols) were collected at one site near Laforge 1 Reservoir using PVC tubes 15 cm in diameter and inserted manually down to the inorganic soil horizons. These soil cores were then extruded and cut at every centimeter with attention paid not to sample the material directly in contact with the tube. Three cores were used for biomarker analyses, whereby subsamples were transferred into cryovials and immediately preserved in liquid nitrogen. The remaining three cores were then subsampled separately into acid-cleaned glass vials for reactive iron and aluminum analyses. Freeze dried soil samples were then ground and homogenized in the laboratory prior to chemical analysis.

Chemical analyses—Sediment accumulation rates in the reservoir were determined by radiometric measurement of ^{210}Pb activity using alpha spectrometric measurement of the activity of the daughter product ^{210}Po (Flynn 1968) and assuming secular equilibrium between the two isotopes. Specifically, aliquots of 0.3 g of sediment were spiked with ^{209}Po yield tracer and digested in Teflon vials using a concentrated 5:4:1 mixture of HNO_3 : HCl : HF . The residue was converted to a chloride salt by repeated evaporation with 12 mol L^{-1} HCl . The salt was then dissolved into a 0.5 mol L^{-1} HCl solution, to which 0.2 g of ascorbic acid were added. Po isotopes were deposited on a spinning Ag disc (Hamilton

and Smith 1986) and the activity measured by α -spectrometry using ORTEC silicon surface barrier detectors coupled to a PC running under Maestro data acquisition software. Blanks were run through the same analytical procedures to test for any contamination from analytical reagents. The supported ^{210}Pb activity for each core was estimated from the series of deepest samples whose measured activity did not deviate by more than 1 standard deviation from the mean of these measurements.

Total carbon and nitrogen were determined by combustion using a Carlo Erba (NA-1500) elemental analyzer. The analytical variability of elemental analyses was better than $\pm 1\%$. Aluminum and iron oxyhydroxydes (Al_{cdB} and Fe_{cdB} , respectively) were extracted from bulk soil samples using the method developed by Lucotte and d'Anglejan (1985) with modifications. The precision of the method is better than $\pm 7\%$ at the 95% confidence level initially cited by Lucotte and d'Anglejan (1985) and averages $\pm 2\%$ (Louchouart et al. 1997b). Molecular analyses of biomarkers were performed according to the CuO oxidation method initially developed by Hedges and Ertel (1982) and modified by Goni and Montgomery (2000). This new method replaces the combustion oven with microwave digestion and uses ethyl acetate instead of ether in the liquid-liquid extraction procedure. Briefly, sediment samples were weighed to include 2–5 mg of organic carbon in the Teflon-lined vessels of an analytical microwave oven (CEM MDS 200). Samples were digested with CuO in presparged 2 mol L^{-1} NaOH and in the absence of O_2 at 150°C for 90 min. Extracted reaction products were converted to trimethylsilyl derivatives and analyzed on a GC/MS system (Varian 3800/Saturn 2000) fitted with a fused capillary column (DB-1, 60 m \times 0.32 mm i.d.; J&W Scientific). Replicate analyses of two “standard” sediment samples ($n = 2$ –11) showed that the analytical variability of the major CuO-oxidation products and related parameters ranged 2–19% and averaged $10\% \pm 7\%$ (Table 1). Similarly, the intercomparison of these sediment analyses between different laboratories shows that the yields and composition parameters produced in this study match those obtained with both the microwave digestion (Goni and Montgomery 2000) and with the “traditional” oven procedure (Louchouart et al. 2000).

Statistical analyses—Statistical tests were applied to differentiate between parameter signatures in the surface versus deep soil horizons and sediment intervals (organic vs. inorganic layers, and post- vs. preflooding accumulation, respectively). In most cases, the normal distribution of the data in the sample sets allowed the use of the Student parametric test (*t*-test), which was performed using a statistical analysis program (Sigma Stats) to evaluate statistically significant differences ($p < 0.05$) between the average values of studied parameters measured in these soil and sedimentary environments. When the assumption of the normal distribution failed, a nonparametric Mann-Whitney test was used.

Results

Soils—All soil profiles discussed (Figs. 2–4) represent averages of elemental concentrations and molecular signatures

Table 1. Comparison and variability of CuO-oxidation parameters measured from two reference sediments: Lake Washington sediment mud (freshwater lake, LWSM) and Sag 05 (estuarine sediments). Oxidation method in this study and Goni and Montgomery (2000) was performed using a microwave oven, whereas values from Louchouart et al. (2000) were produced using conventional oven.

	LWSM		SAG 05	
	This study (n=2)	Goni and Montgomery (n=6)	This study (n=7)	Louchouart et al. (n=11)
$\lambda 8$	2.36 (± 0.05)	2.40 (± 0.01)	2.81 (± 0.31)	3.53 (± 0.17)
3,5-Bd	0.11 (± 0.01)	ND	0.23 (± 0.02)	0.20 (± 0.05)
V	1.51 (± 0.02)	1.47 (± 0.10)	2.11 (± 0.29)	2.73 (± 0.11)
S	0.65 (± 0.03)	0.71 (± 0.10)	0.44 (± 0.03)	0.56 (± 0.05)
P	0.61 (± 0.03)	ND	0.34 (± 0.04)	0.54* (± 0.10)
C	0.20 (± 0.01)	0.21 (± 0.03)	0.26 (± 0.04)	0.23 (± 0.04)
S:V	0.43 (± 0.01)	0.49 (± 0.09)	0.21 (± 0.03)	0.21 (± 0.02)
C:V	0.13 (± 0.00)	0.15 (± 0.03)	0.13 (± 0.02)	0.08 (± 0.01)
3,5-Bd:V	0.07 (± 0.01)	ND	0.11 (± 0.01)	0.08 (± 0.02)
P:(V+S)	0.28 (± 0.01)	ND	0.14 (± 0.02)	ND
(Ad:Al)v	0.53 (± 0.02)	0.47 (± 0.05)	0.48 (± 0.09)	0.38 (± 0.05)
(Ad:Al)s	0.51 (± 0.06)	0.41 (± 0.13)	0.47 (± 0.13)	0.38 (± 0.05)

* Values from Louchouart (1997). ND, Not determined; V, S, C, milligrams of vanillyl, syringyl, and cinnamyl phenols, respectively, normalized to 100 mg of organic carbon; $\lambda 8$, the sum of V, S, and C. 3, 5-Bd, 3, 5-dihydroxybenzoic acid normalized to 100 mg of organic carbon; C:V and S:V, the ratios of cinnamyl and syringyl phenols to vanillyl phenols, respectively; 3, 5-Bd:V, the ratios of 3, 5-dihydroxybenzoic acid over vanillyl phenols; P:(V+S), the ratios of p-hydroxyl phenols to the sum of vanillyl and syringyl phenols; (Ad:Al)v and (Ad:Al)s, the weight ratios of phenolic acid to phenolic aldehyde for the vanillyl and syringyl phenol families, respectively. \pm Values represent 1 standard deviation.

for each interval across the three cores collected. The organic horizon in such soils is thick (>8 cm) and enriched in organic matter as indicated by high and stable carbon and lignin concentrations ($45.9\% \pm 6.6\%$ and 74.8 ± 30.3 mg $[10$ g dry wt] $^{-1}$, respectively), a major and unique structural component of vascular plants (Sarkanen and Ludwig 1971). These concentrations drop quickly and by an order of magnitude in the inorganic horizons ($2.5\% \pm 1.5\%$ and 1.8 ± 1.8 mg $[10$ g dry wt] $^{-1}$, respectively). Similarly, the atomic

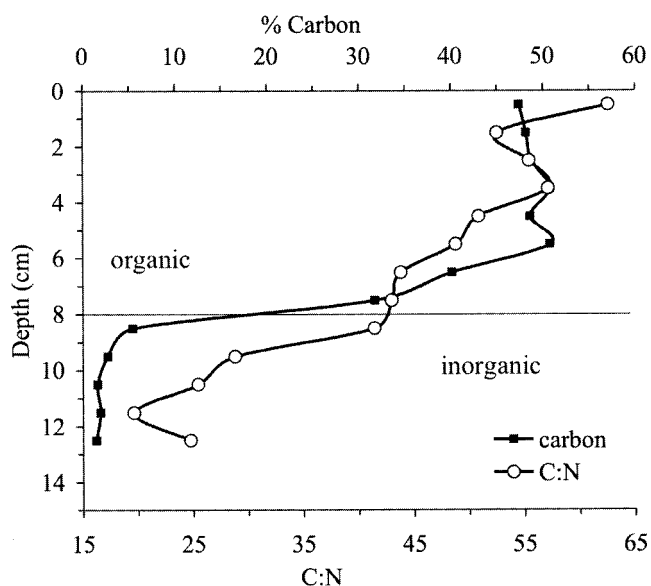


Fig. 2. Organic carbon concentrations and atomic C:N ratios in the soil profiles.

C:N ratio drops substantially from a value of 52 ± 7 in the organic layers to a value of 28 ± 8 in the inorganic horizon (Fig. 2). The difference in the average values of all these parameters (C and lignin content, and atomic elemental C:N ratios) between the organic and inorganic horizons is highly significant ($p < 0.001$). Below the organic horizons, the strong and rapid increase in the storage of reactive iron and aluminum (Fig. 4) points to the increased availability of reactive and sorption sites tied to aluminosilicates and/or oxyhydroxides in the mineral horizons of these soils (i.e., Kaiser and Guggenberger 2000). Various ratios of CuO oxidation byproducts are used extensively to describe both the source and degradation state of terrestrial organic matter in soils and sediments (see discussion following). Among these, the ratio of syringyl to vanillyl phenols (S:V; Fig. 3b) is variable but shows a decreasing trend from a value of 0.22 ± 0.09 in surface organic horizons to a value of 0.12 ± 0.03 in the subsoil. Although this difference is significant ($p = 0.04$), the degree of confidence is close to the set limit of significance and the power of the performed test (0.48) is below the desired power (0.80); as such we should interpret this difference cautiously. The ratio of cinnamyl to vanillyl phenols (C:V) remains stable at around 0.52 ± 0.23 in the surface organic layers and increases sharply and significantly ($p < 0.05$) right beneath the organic-inorganic interface to reach an average value 1.04 ± 0.50 (Fig. 3b). Similarly, additional ratios such as 3,5-dihydroxybenzoic acid:vanillyl phenols (3,5-Bd:V; Fig. 3c), p-hydroxyl phenols:sum of syringyl and vanillyl phenols (P:[V+S]; Fig. 3d), vanillic acid:vanillin ([Ad:Al]v; Fig. 3c), and p-coumaric acid:ferulic acid (Cd:Fd; Fig. 3e) all show a 5- to 10-fold increase from the surface organic layers to the deeper mineral hori-

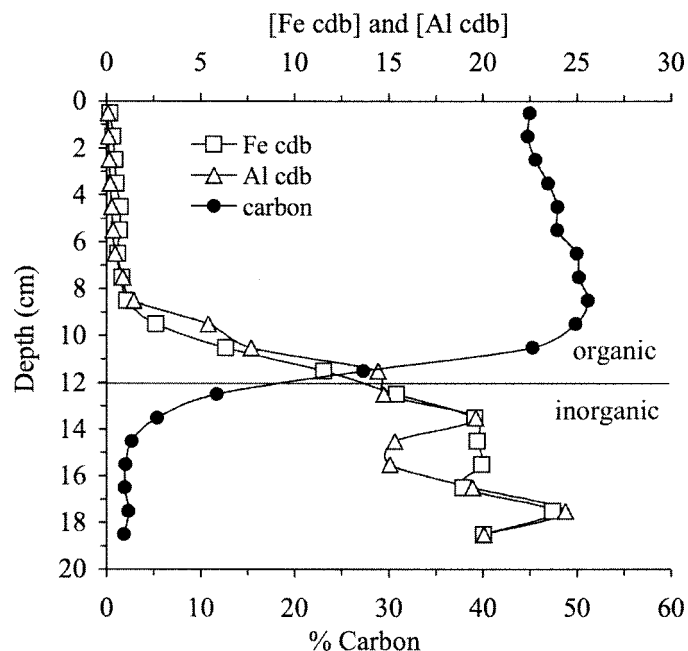
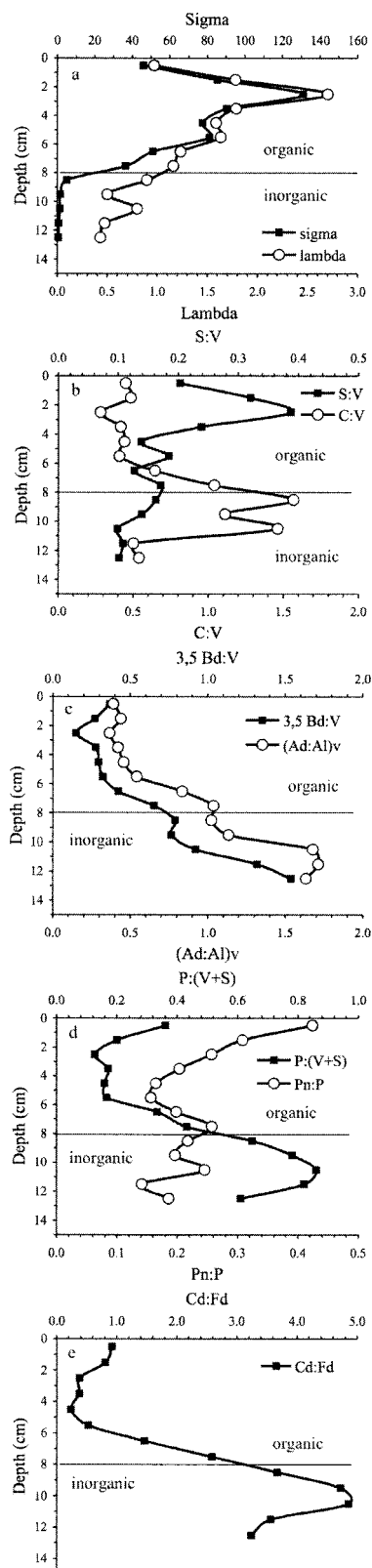


Fig. 4. Organic carbon concentration (%) and Fe-Al oxyhydroxide loads (g m^{-2}) in the soil profiles. The soil cores collected for density mass calculations and metal loads were subsampled adjacent to the ones used for chemical analyses.

zons. All of these parameters show highly significant differences ($p < 0.001$) between their organic and inorganic soil averages. Finally, although the ratio of p-OH acetophenone : p-hydroxyl phenols (Pn:P) decreases slightly along the soil depth profile (0.25 ± 0.09 vs. 0.20 ± 0.04 , respectively), the difference between these averages is not statistically significant.

Sediments: Radiometric measurements—The ^{210}Pb activity in surface sediments varies from ca 15, 32, and 100 dpm g^{-1} in cores 1001, 999, and 1200, respectively (Fig. 5a,c,e). The chronology was established using the downcore unsupported ^{210}Pb ($^{210}\text{Pb}_{\text{ex}}$; 22.3 half-life), a constant initial concentration model (CIC), and dividing nonlinear profiles into linear segments assuming constant sedimentation for each of these segments (Appleby and Oldfield 1990). The high correlation coefficients within each segment ($r^2 = 0.91\text{--}0.98$; Fig. 5b,d,f) suggest minimal mixing of sediments and that accumulation rates were close to constant during each specific period. Approximate ages of sediments were determined using the slope of the relationship between cumulative dry

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Fig. 3. Molecular signatures in the soil profiles. (a) Lignin concentrations (Σ : $\text{mg [10 g dry wt]}^{-1}$) and carbon-normalized yields (λ : $\text{mg [100 mg OC]}^{-1}$). (b) S:V, ratio syringyl to vanillyl phenols; C:V, ratio of cinnamyl to vanillyl phenols. (c) 3,5-Bd:V, ratio of 3,5-dihydroxybenzoic acid over vanillyl phenols; (Ad:Al)v, ratio of vanillic acid to vanillin. (d) P:(V+S), ratio of p-hydroxyl phenols to the sum of vanillyl and syringyl phenols; Pn:P, ratio of p-hydroxyacetophenone to p-hydroxyl phenols. (e) Cd:Fd, ratio of p-coumaric acid over ferulic acid.

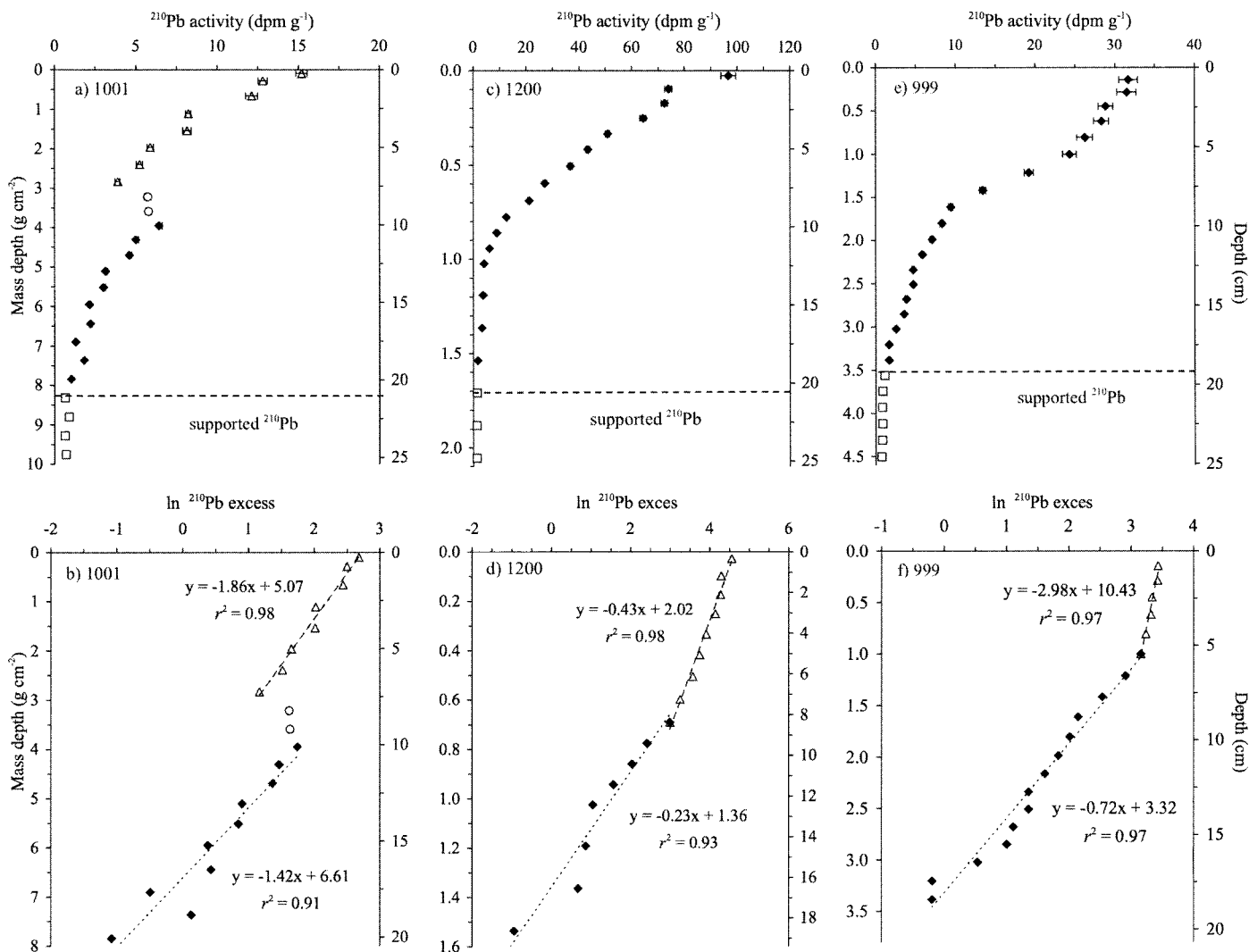


Fig. 5. (a, c, e) Total ^{210}Pb activity; (b, d, f) natural logarithm of the unsupported ^{210}Pb activity ($^{210}\text{Pb}_{\text{ex}}$) versus cumulative dry mass (g cm^{-2}) and depth in sediments of the Cabonga Reservoir.

mass (g cm^{-2}) and the natural logarithm of $^{210}\text{Pb}_{\text{ex}}$, and the ^{210}Pb decay constant (λ). This sediment chronology locates the period of impoundment (1928) below the intervals of 9–10 cm (~ 67 yr) at Sta. 1200 and 12–13 cm (~ 70 yr) at Sta. 999 (Fig. 5d,f). At Sta. 1001, the two points not falling on any segment (8–10 cm; Fig. 5b) probably indicate a period of sediment slumping that accumulated rapidly in this system. Using a time for slumping deposition ranging from 1 to 5 yr does not change the chronology and sets the time of impoundment (~ 70 yr) at 11–12 cm depth. We have thus marked the time of flooding with a horizontal bar on all subsequent sediment profiles. This “interface” is represented for all subsequent parameters on their respective figures to discern between pre- and postflooding conditions in the three sedimentary systems studied.

Sediments: Molecular and elemental markers—Lignin yields specific byproducts upon CuO oxidation that can serve as unambiguous markers of vascular plant material inputs to aquatic systems. In the three lacustrine cores stud-

ied (Fig. 6), we observed a 1.5 to 4-fold increase in mass- and carbon-normalized lignin values ($\Sigma 6$ and $\lambda 6$ values, respectively). The significant increases in $\Sigma 6$ and $\lambda 6$ values ($p < 0.001$) thus seem to record a shift in terrigenous organic matter inputs with increased proportions of lignin-rich constituents in sediments accumulated after impoundment. In core 1001, we also observe a slight increase in lignin oxidation products between 14.5 and 21.5 cm depth prior to the chronological mark of impoundment. We believe this change traces historical changes in land use in the vicinity of this station (old lake), which is located near a native reservation. This may suggest that human settlements on lakeshores had local effects on terrigenous organic matter inputs to the aquatic system (c.f. Farella et al. 2001).

The difference in the mean values of the ratio of syringyl to vanillyl phenols (S:V; Fig. 7) between the two time periods is significant only at Sta. 1001 and 999 ($p < 0.001$). However, these ratios remain relatively low throughout all sediment core depths (0.21–0.31 pre-impoundment to 0.21–0.25 post-impoundment) and oscillate without much discern-

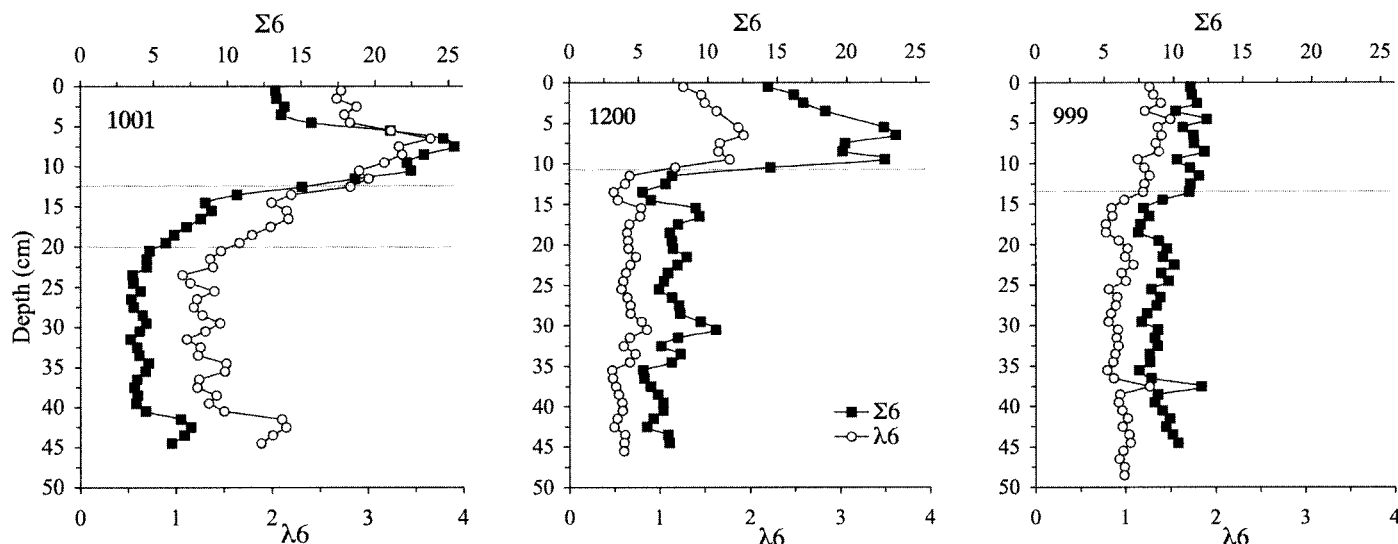


Fig. 6. Mass- and carbon-normalized yields of the sum of lignin-derived vanillyl and syringyl oxidation byproducts ($\Sigma 6$ and $\lambda 6$, respectively) in sediments of the Cabonga Reservoir. $\Sigma 6$, mg (10 g dry wt) $^{-1}$; $\lambda 6$, mg (100 mg OC) $^{-1}$.

ible pattern across the pre- to postflooding interface. These values are typical of mixed deciduous forest SOM comprising combined inputs from both angiosperm and gymnosperm sources but with predominance of the latter (Guggenberger et al. 1994; Prahl et al. 1994; Louchouart et al. 1999). In contrast, a highly significant ($p < 0.001$) decrease in the cinnamyl over vanillyl ratio (C:V; Fig. 7) occurs between sediments accumulating during undisturbed lake conditions (1.0–2.0) and those accumulating after flooding and changes in watershed land use (0.2–0.5). Within all sediment cores, (Ad:Al)v ratios vary between 0.6 and 1.1 (Fig. 8), suggesting a relatively degraded source of vascular plant constituents (Hedges et al. 1988; Goni et al. 1993). We found no significant pattern of change between pre- and postflooding sediments. Contrastingly, 3,5-Bd:V ratios drop substantially and highly significantly ($p < 0.001$) from values ranging from 0.2–0.5 in preflooding sediments to 0.1–0.2 in surface postflooding intervals (Fig. 8), suggesting a shift in the nature of inputs of terrigenous organic matter to these sedimentary systems. Similarly, the ratio of nonmethoxylated to methoxylated phenols in CuO oxidation mixtures (P:[V+S]) drops substantially and highly significantly ($p < 0.001$) from pre- to postflooding sediments (0.3–0.7 to 0.1–0.4, respectively; Fig. 9), pointing again to a shift in the nature of sedimentary organic matter. In contrast, Pn:P ratios remain relatively constant (~ 0.2 ; Fig. 9) throughout all core depths. Although the difference between pre- and post-impoundment averages at stations 1200 and 1001 are significant (0.13 ± 0.01 vs. 0.15 ± 0.01 and 0.17 ± 0.01 vs. 0.19 ± 0.01 , respectively), the variation amounts to only a 10%–15% increase in the ratio, suggesting no substantial change in the proportional delivery of p-hydroxyacetophenone to total p-hydroxyls to sediments across the flooding interface.

In addition to the biomarker parameters previously presented and which are specific to terrigenous organic matter, we also use carbon content and atomic ratios of organic carbon to nitrogen (C:N), the latter being an integrated indicator of organic matter inputs from both the landscape (al-

lochthonous) and in situ productivity (autochthonous; Meyers and Ishiwatari 1993; Routh et al. 2004). The C:N profiles match the behavior of organic carbon concentrations (Fig. 10). Preflooding conditions are characterized by constant C:N ratios (14.8 ± 0.4 , 17.6 ± 1.3 , and 17.3 ± 0.5 for Sta. 1200, 1001, and 999, respectively) and carbon concentrations ($11.5\% \pm 0.4\%$, $3.4\% \pm 0.6\%$, and $9.6\% \pm 0.3\%$ for Sta. 1200, 1001, and 999, respectively). Following impoundment, we observed marked subsurface maxima in carbon concentrations and C:N ratios at Sta. 1001 (7–8% and 25–29, respectively) and 1200 (13% and 18, respectively). At Sta. 999, a significant decline in these two parameters was observed ($8.7\% \pm 0.5\%$ and 16.0 ± 0.8 ; $p < 0.001$) some time after impoundment had occurred.

Discussion

Sediment geochemistry—The range in specific activity for unsupported ^{210}Pb at all three stations (15–95 dpm g^{-1}) was typical of that reported in temperate lakes in northeastern North America (Davis et al. 1984). This variability may reflect differences in the deposition environment and/or sedimentological parameters such as the sources of minerals, grain size, and organic matter content. In reservoirs with dendritic structures, such as those like the Cabonga Reservoir, a spatial difference in hydrodynamic regime (Osidele and Beck 2004) may lead to a strong heterogeneity in sediment accumulation rates between different sites. Sta. 1200 is representative of a typical open channel central basin acting as an integrator of regional sediment inputs, whereas Sta. 1001 and 999 are characteristic of a littoral station within a protected cove and a narrow and peripheral channel area surrounded by multiple islands, respectively (Fig. 1). A potential explanation for the observed differences in surface ^{210}Pb activities across these sites is a change in the proportion of sedimentary size fractions (fine to coarse) typical of sedimentary deposits along hydrodynamic gradients (Thorn-

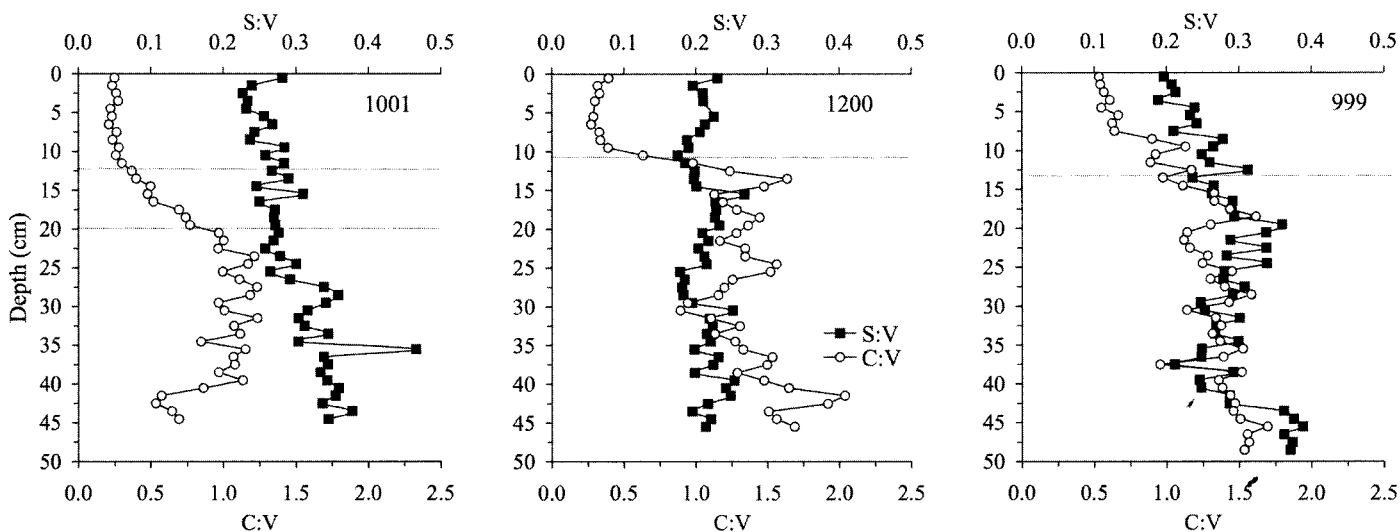


Fig. 7. Vertical profiles of lignin source ratios in sediments of the Cabonga Reservoir. S:V, ratio syringyl to vanillyl phenols; C:V, ratio of cinnamyl to vanillyl phenols.

ton 1990). Because both Sta. 1001 and 999 are more susceptible to local inputs of watershed materials and undergo fewer hydrodynamic sorting processes, accelerated deposition of coarse materials may lead in these systems to a dilution of $^{210}\text{Pb}_{\text{ex}}$ concentrations in surface sediments (Sorente et al. 1999). In contrast, the central reservoir station (1200), as a focal point of fine sediment accumulation for the entire reservoir, is less affected by local inputs from the littoral zones and potential dilution effects and is thus characterized by a higher $^{210}\text{Pb}_{\text{ex}}$ concentration in surface sediments.

Sediment accumulation rates in recent sediments generally increased at all stations with respect to pre-impoundment conditions from 0.02 to 0.09 $\text{g cm}^{-2} \text{yr}^{-1}$ at Sta. 999, from 0.04 to 0.06 $\text{g cm}^{-2} \text{yr}^{-1}$ at Sta. 1001, and 0.007 to 0.013 $\text{g cm}^{-2} \text{yr}^{-1}$ at Sta. 1200. The increased mass accumulation, although consistent with common models of sedimentation

in reservoirs (Thornton 1990; Hambright et al. 2004), shows a 20-yr delay with respect to impoundment in the case of Sta. 1001 and 1200. This lag in sedimentation response may be related to the timing of reservoir filling or other factors (i.e., the geomorphology of the reservoir itself), leading to a delay in reaching a new equilibrium in sedimentation rates responsible for the smooth ^{210}Pb profiles observed in the last 50 yr at these two sites. At Sta. 999, however, the increase in accumulation rate has occurred only in the last 20 yr and is probably not directly related to the flooding itself. A possible cause for such a change may be attributed to local changes in the watershed (fire, logging).

Prior work in boreal reservoirs of the regions has shown that following impoundment, erosional processes are responsible for the translocation of about half of the SOM to the water column (Houel 2003). The fate of this remobilized

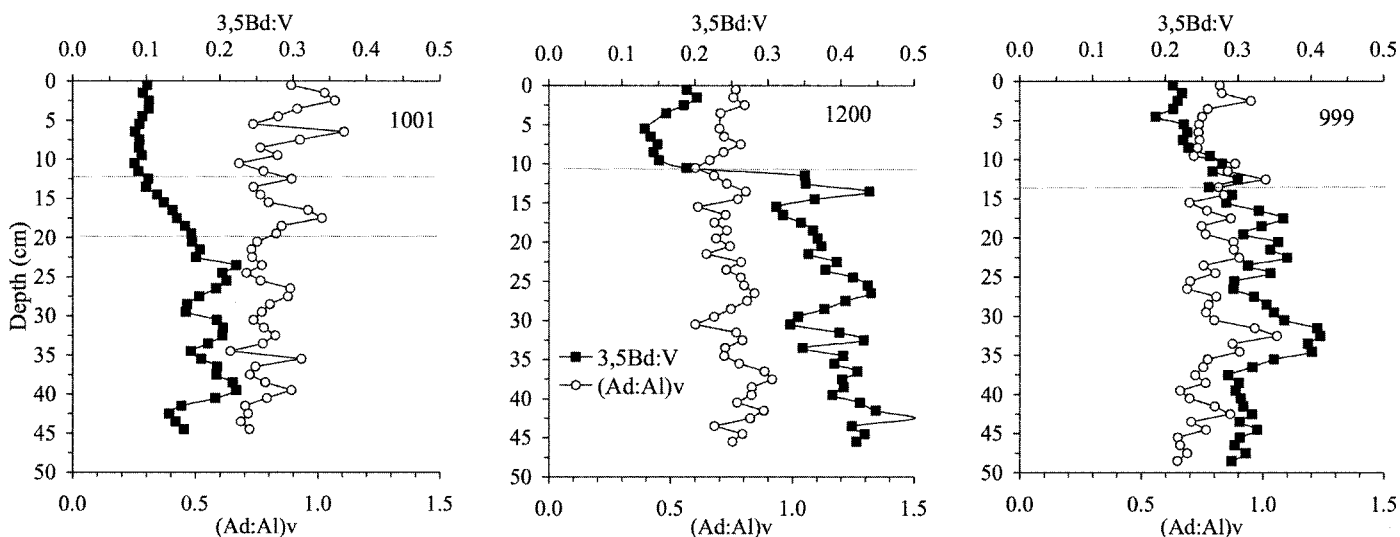


Fig. 8. Vertical profiles of lignin and organic matter degradation state indicators in sediments of the Cabonga Reservoir. 3,5Bd:V, ratio of 3,5-dihydroxybenzoic acid over vanillyl phenols; (Ad:Al)v, ratio of vanillic acid to vanillin.

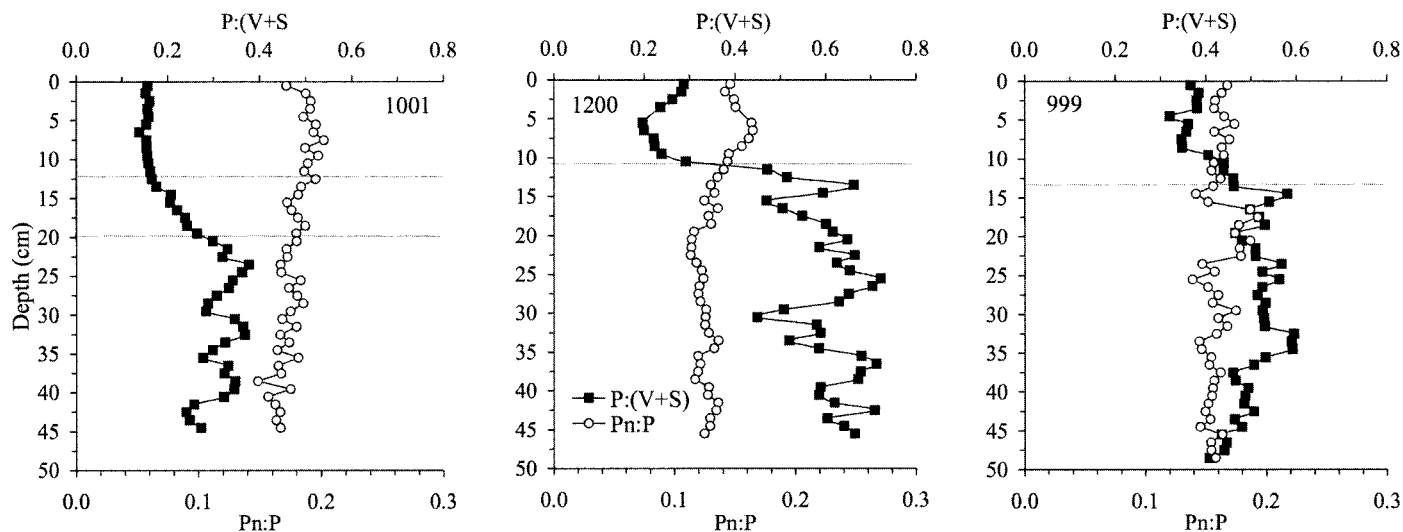


Fig. 9. Vertical profiles of p-hydroxyl phenol ratios in sediments of the Cabonga Reservoir. P:(V+S), ratio of p-hydroxyl phenols to the sum of vanillyl and syringyl phenols; Pn:P, ratio of p-hydroxyacetophenone to p-hydroxyl phenols.

material is not yet known and may include remineralization, redistribution to sedimentary deposits, and export to downstream ecosystems. However, it was our assumption here that if significant amounts of surface SOM were redistributed toward sedimentary deposits within the reservoir, it would translate to measurable shifts in the composition of accumulated organic matter. Hence, under large-scale erosional processes in the basins, organic-rich coarse materials, more typical of soil litter and organic horizons, may predominate in sedimentary materials. In contrast, under undisturbed watershed conditions, the enhanced sorption of hydrophobic soil DOM on fine clays and hydroxide particles (Kaiser and Guggenberger 2000; Hagedorn et al. 2000) may impart a selective signature to fine particulates reaching aquatic systems particularly under hydrodynamic sorting conditions (c.f. Louchouart et al. 1999; Farella et al. 2001; Bianchi et al. 2002).

We used the vertical transport and sorption processes known to occur in podzols, enriched in oxi-hydroxides in the inorganic horizons, as a proxy environment to illustrate the lateral leaching/adsorption processes that occur in lakes under undisturbed conditions. Soils characterized by strong vertical transport processes and having mineral horizons enriched in clay minerals and/or reactive Fe-Al oxy-hydroxides often show contrastingly different molecular compositions along their vertical profiles (Kaiser and Guggenberger 2000; Rumpel et al. 2002). Indeed, the lignin content and signature of SOM sorbed to fine clay minerals and hydroxides in deep soils strongly resemble that of DOM in organic/litter soil solutions but not that of litter organic matter solid phases (Kaiser et al. 2001; Kaiser and Guggenberger 2002). For example, Engelhaupt and Bianchi (2001) have observed that lignin signatures in high molecular weight DOM collected in a southern Louisiana tidal stream are not similar to those

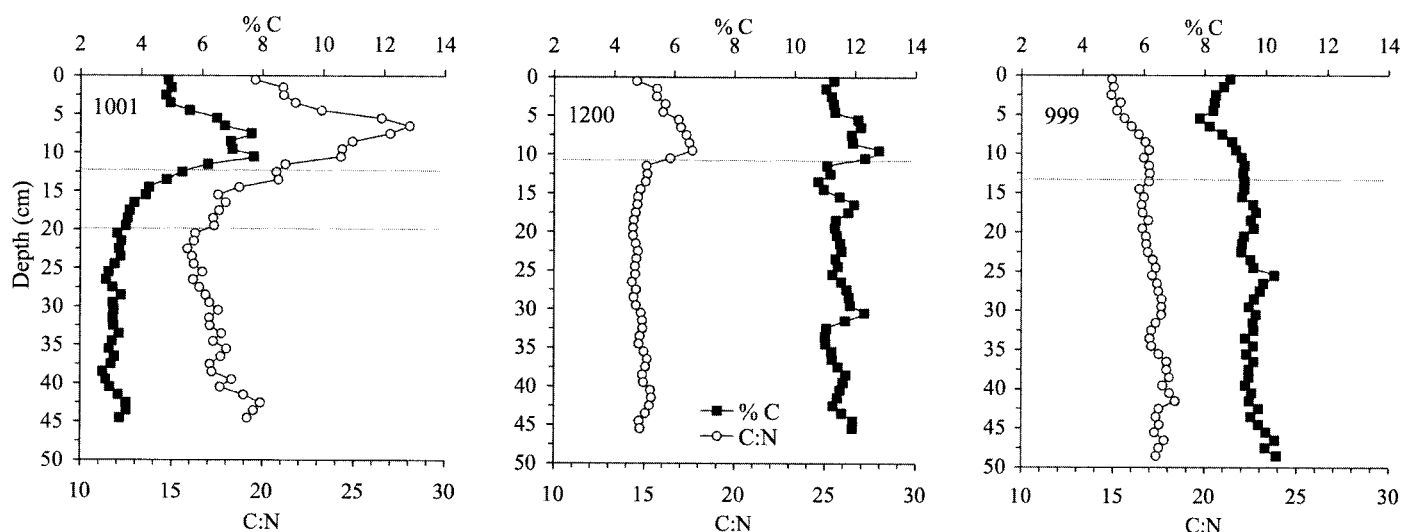


Fig. 10. Organic carbon concentrations and atomic C:N ratios in sediments of the Cabonga Reservoir.

obtained from surface SOM, but rather are characterized by higher C:V and (Ad:Al)v ratios, signatures that are more consistent with those of deep SOM. In addition, Hagedorn et al. (2000) showed that in spite of a strong potential for selective sorption of aromatic DOM on Fe hydroxides in deep soil mineral horizons, large runoff events tend to facilitate rapid flushing of topsoil DOM and export to aquatic ecosystems. In turn, this DOM tends to sorb/coprecipitate on iron-rich particulates and redeposit in these systems (Hagedorn et al. 2000). We thus applied the yields and composition of terrigenous organic matter in surface and deep soils as model end-members of natural versus erosional inputs, respectively, to characterize the potential shifts in sources of SOM inputs to the lake sedimentary deposits prior to and following impoundment.

In the present study, postflooding sediments do show a substantial and significant enrichment in lignin-derived components (Fig. 6) along with a concomitant and significant drop in the C:V ratio (Fig. 7). These changes are consistent with the remobilization and sedimentation, upon flooding, of surface SOM, which is both enriched in lignin and characterized with a lower C:V ratio (Fig. 3). This latter ratio in preflooding sediments is more characteristic of signatures from SOM sorbed on deep fine soil minerals. Although the C:V ratio is known to decrease during the transfer of terrigenous organic matter from the water column to sediment deposits due to a selective sensitivity of cinnamyl moieties to diagenetic alterations (Opsahl and Benner 1995; Klap et al. 1999; Louchouart et al. 1999), in situ alteration of lignin composition is usually minor within sediments (Hedges et al. 1982; Ishiwatari and Uzaki 1987; Louchouart et al. 1997a) and C:V signatures of mature organic matter can still retain a good amount of source information (Louchouart et al. 1999; Hedges et al. 2000). It thus seems that the strong shift in C:V signatures observed here are indicative of substantial changes in the source materials, rather than large-scale diagenetic transformation of sedimentary organic matter.

The Cd:Fd ratios in sediments further support this interpretation, with preflooding values in sediments averaging 12.7 ± 4.5 versus 4.8 ± 3.6 in recent postflooding sediments at all stations. The much higher values observed in sediments versus those of soils suggest that the Cd:Fd ratios can only be used as qualitative markers of selective dissolution/sorption processes. It is obvious here from the observed increase in these ratios in aquatic systems versus soils that other processes, such as a potentially higher susceptibility of Fd than Cd to degradation and/or higher leaching of Cd than Fd, influence these ratios in their transfer to sedimentary systems.

The decreased proportion of 3,5-Bd to vanillyl phenols across the pre- to postflooding interface (Fig. 8) also parallels the substantial shift in this ratio observed between inorganic and organic layers of the proxy soils (Fig. 3c). The phenolic compound 3,5-Bd is a common product of soil degradation processes (Christman and Oglesby 1971; Ugolini et al. 1981) and has been found regularly within aquatic sediments (Prahl et al. 1994; Louchouart et al. 1999; Farella et al. 2001). It has been inferred that the likely precursors of this phenolic product are not altered lignin macropolymers

but tannins and other flavonoids with hydroxy groups present in alternate positions on the aromatic rings (Christman and Oglesby 1971; Goni and Hedges 1995). Because tannin-like materials tend to accumulate within decaying cells (de Leeuw and Largeau 1993), the relative increase of this compound in deep soil minerals may be related to the extent of degradation and humification of fresh vascular plant tissues. This is supported by extremely low 3,5-Bd content and 3,5-Bd:V ratios produced by fresh plant materials upon CuO oxidation (Louchouart 1997). Several studies (Prahl et al. 1994; Louchouart et al. 1999; Farella et al. 2001) have thus used 3,5-Bd:V ratios to identify humified SOM associated with fine clay minerals in aquatic systems.

A similar relationship between sediment and soil profiles is also observed for the ratio of p-hydroxyl phenols to the sum of vanillyl and syringyl phenols (P:[V+S]; Figs. 3d, 9). Although elevated 3,5-Bd:V is a general indicator of humified SOM sorbed on fine particles (Prahl et al. 1994; Louchouart et al. 1999; Farella et al. 2001), an increase in the P:(V+S) ratio marks a specific lignin degradation pathway (brown-rot), leading to demethylation of methoxylated vanillyl and syringyl constituents (Dittmar and Lara 2001). A caveat to these interpretations, however, is that both 3,5-Bd and P compounds are not exclusive biomarkers of vascular plant tissues (Hedges et al. 1988; Goni and Hedges 1995), thus potentially impacting the above-mentioned ratios in sediments receiving substantial inputs of kelps and brown macroalgae (3,5-Bd) and plankton-derived amino acids (P). Although macroalgal sources of 3,5-Bd might be significant in selected marine systems comprising minimal fractions of terrigenous organic matter (c.f. Goni and Hedges 1995), in freshwater systems this interference is unlikely to affect 3,5-Bd:V ratios substantially. In oligotrophic inland aquatic systems such as the reservoirs studied here, p-hydroxyl phenol inputs from autochthonous sources are probably negligible, thus having a limited effect on P:(V+S) signatures as indicated by constant ratios of p-hydroxyacetophenone to total p-hydroxyl phenols (Pn:P) in all sedimentary profiles (0.16 ± 0.02 ; Fig. 9). Indeed, p-hydroxyacetophenone is derived exclusively from lignin constituents (Hedges et al. 1988), and constant Pn:P ratios in sedimentary systems have been used to verify lignin sources for this phenol family (Dittmar and Lara 2001).

The potential shift in the source of organic matter to the reservoir sediments, however, does not affect predictably the signature ratios of certain lignin-derived parameters (S:V and [Ad:Al]v; Figs. 7, 8). Although we observed some slight changes in S:V ratios along the soil profiles, S:V signatures remained low in post-impoundment sediments (0.2–0.3), rather than increased slightly to match signatures observed in surface soil organic layers. Similarly, the relatively constant (Ad:Al)v ratios across the pre- to postflooding interface in sediments (0.77 ± 0.08 and 0.81 ± 0.12 , respectively; Fig. 8) did not provide an appropriate indicator for input variations of surface versus mineral SOM to these systems. One reason for this discrepancy may be that both ratios are particularly sensitive to photochemical degradation processes, which substantially decrease S:V and increase (Ad:Al)v in DOM (Opsahl and Benner 1998; Hernes and Benner 2003). Potential photochemical degradation of erod-

Table 2. Elemental and biomarker mass-normalized concentrations of diverse end-members used in this study. All units are in mg (g dry wt)⁻¹. All abbreviations as in Table 1.

End-member	C	N	Σ6	V	3, 5-Bd	P
litter SOM (<i>n</i> =2)	526±2	8±4	16.8±1.3	13.2±2.3	0.69±0.09	2.25±0
Mineral-derived SOM (<i>n</i> =2)*	30±10	1.1±0.3	0.107±0.005	0.090±0.008	0.074±0.001	0.080†±0.027
Autochthonous (<i>n</i> =187)	421±75	67±32				

* These values represent the average of two separate soil mineral horizons (5 cm) each integrating five depths.

† The corrected mass-normalized yield for P used in the mixing model was 0.107 (see discussion).

ed materials in the water column prior to sedimentation may thus have pushed both ratios in the direction opposite to that of surface soil signatures, resulting in little change in sedimentary signatures across the two periods. Little is known about the effect of photochemical degradation on other CuO-oxidation parameters other than that the effect on C : V ratios seems limited to only a slight increase (Opsahl and Benner 1998). However, if photochemical alteration of eroded SOM is significant in this system, it did not seem to overshadow the large-scale input changes that provided an imprint to biomarker yields (λ and Σ) as well as signatures such as C : V, 3,5-Bd : V, Cd : Fd, and P : (V+S) ratios.

Mixing model: Allochthonous inputs—To further test the hypothesis that terrigenous organic matter signatures in lake sediments vary in relation to a proportional mixture of surface versus deep SOM, we used a quantitative mixing model based on mass-normalized yields of the different CuO-oxidation parameters in two distinct end-members, namely soil litter and deep mineral SOM (c.f. Louchouart et al. 1999). We tested such a model using 3,5-Bd : V and P : (V+S) ratios versus carbon-normalized lignin yields (λ_6) according to the mixing equation

$$\left(\frac{A}{B}\right)_{\text{sed}} = \frac{(A_o \times F_o) + [A_i \times (1 - F_o)]}{(B_o \times F_o) + [B_i \times (1 - F_o)]} \quad (1)$$

where *A* represents the mass-normalized yields for the parameter in the numerator of the ratio (3,5-Bd, P, and Σ_6); *B* represents the mass-normalized yields for the parameter in the denominator (V, [V+S], and OC, respectively); *F_o* is the fraction of organic matter derived from surface soil litter in the sedimentary mixture; and the subscripts represent the mass-normalized yields observed for the different parameters within sediment intervals (sed), deep soil mineral horizons (*i*), and surface organic-rich soil litter (*o*). All mass-normalized yields of the different CuO-derived parameters used in this model are presented in Table 2.

The strong nonlinear relationship between carbon-normalized lignin yields (λ_6) and biomarker ratios (3,5-Bd : V and P : [V+S]) in sediments ($r^2 = 0.91$ – 0.92 ; Fig. 11a,b) are consistent with the mixing behavior predicted above. The apparent match between the 3,5-Bd : V versus λ_6 model (Fig. 11a) and sedimentary data implies that a conservative mixture between the two identified pools is controlling these signatures in sediments of the system of study. In contrast, the mixing model using P : (V+S) versus λ_6 (Fig. 11b) does not fit well the observed sedimentary data (mixing model 1), suggesting that the soil P : (V+S) signatures may not fully represent the true allochthonous endmembers being mixed

in the receiving sedimentary deposits. On the other hand, using the maximum value in the confidence interval of the mass-normalized P yields (mean + 1 SD; from 0.080 to 0.107 mg [g dry wt]⁻¹) for the mineral soil end-member generates a relationship that plots right through the observed sediment values (mixing model 2; Fig. 11b). This slight correction reveals a high sensitivity of the mixing lines to the yields of p-hydroxyl phenols in the mineral soil end-member. The processes that control such a sensitive response may include the influence of soil size fraction inputs to sediments, the proportion of soil-specific end-members in the direct drainage basin, and/or geomorphologic characteristics of the drainage basin emphasizing inputs from specific soil sources.

The use of these two empirical relationships (in the case of P : [V+S], we used the adjusted P content in mineral soils) in combination with λ_6 values measured in all sediments allowed the reconstruction of sedimentary 3,5-Bd : V and P : (V+S) signatures with both a good fit and a slope of one (Fig. 11a,b, inserts). These results confirm conceptual models of lake sediment delivery, which suggest that (1) lake sediments seem to integrate, over a wide range of organic matter concentrations, basin-scale processes such as dissolution/sorption of SOM, sediment transport and hydrodynamic sorting, and erosional inputs of specific soil horizons; and (2) there exists a certain predictive element to lakes' sedimentary signatures of allochthonous SOM.

To evaluate quantitatively the influence of erosional processes on recent inputs of surface SOM to reservoir sediments, we thus used a reorganized version of Eq. 1 that models the sedimentary fraction of surface SOM (*F_o*) as a function of mass-normalized concentrations of 3,5-Bd, V, P, and V+S in the two soil end-members and the 3,5-Bd : V and P : (V+S) ratios measured in sediments. The results of both models (3,5-Bd : V and P : (V+S)) are similar during preflooding conditions (Fig. 12) and indicate that the inputs of allochthonous organic matter to deep sediments are consistently and overwhelmingly comprised (97–99%) of degraded constituents characteristic of SOM sorbed on fine particles. The uniformity in this high proportion of fine-grain SOM at all stations further suggests that prior to impoundment, sedimentary deposits received relatively similar inputs from their undisturbed and respective basins. In contrast, the increased proportion of litter-derived organic matter in recent sediments shows that erosional and translocation processes have led to the redistribution of surface SOM to deep sedimentary deposits shortly after impoundment. Although these inputs of eroded SOM seem to have abated in recent times, they still remain two- to fivefold higher than under preflooding conditions. A particular departure from the pre-impound-

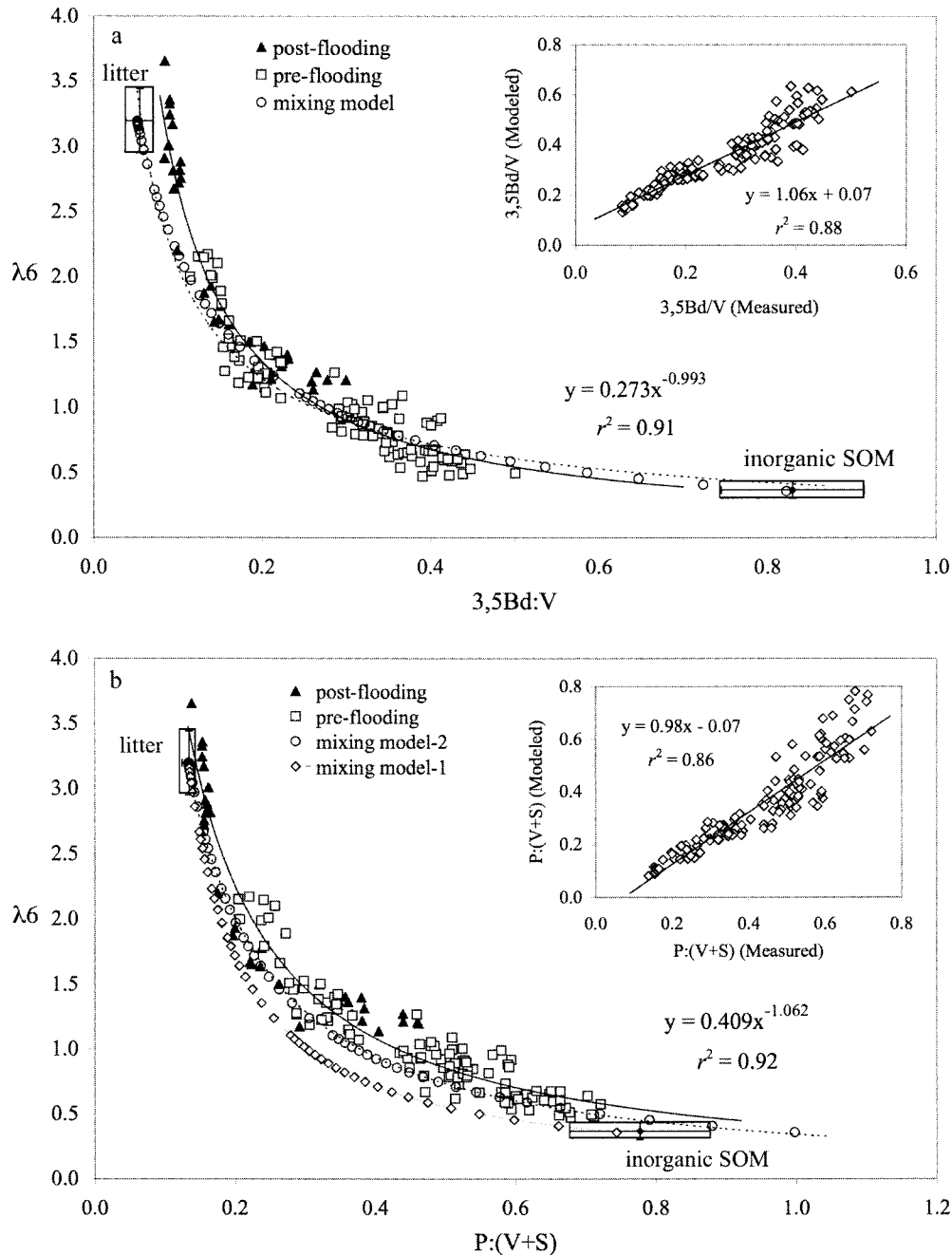
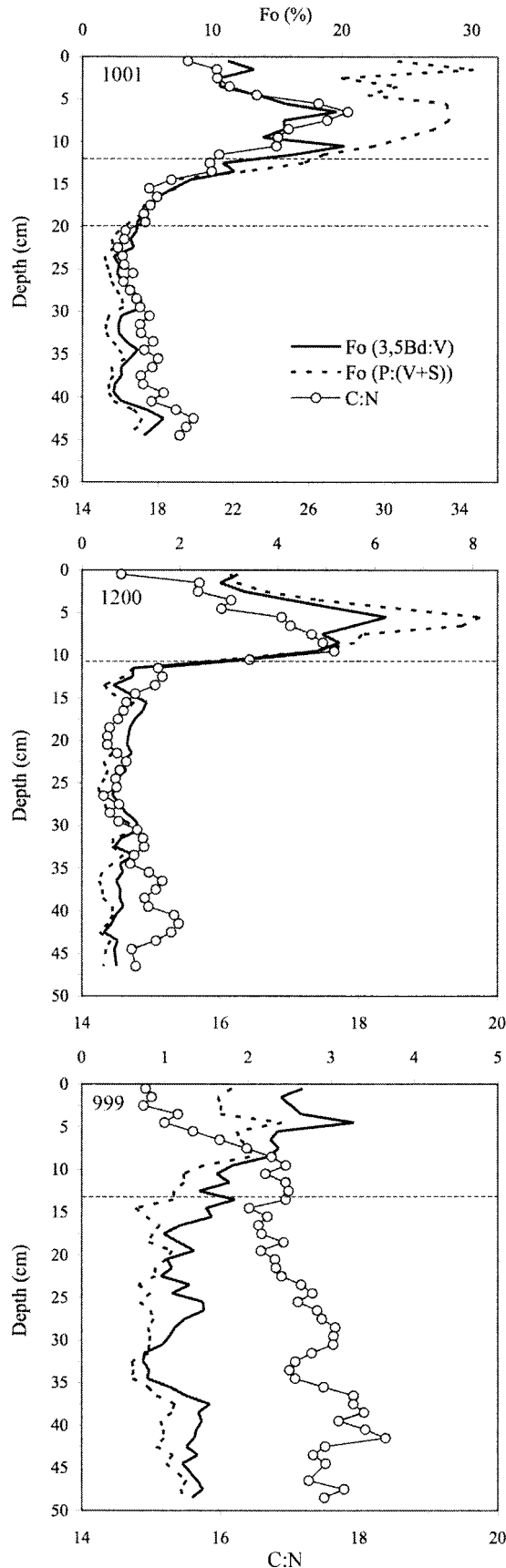


Fig. 11. Measured biomarker signatures in the Cabonga Reservoir sediments and comparison to theoretical values based on a mixing model between surface litter and deep mineral SOM end-members. (a) Mixing lines between $\lambda 6$ and the 3,5-Bd:V ratio. (b) Mixing lines between $\lambda 6$ and the P:(V+S) ratio. "Mixing Model 2" in panel (b) is derived using a P yield of $0.107 \text{ mg (g dry wt)}^{-1}$ instead of $0.080 \text{ mg (g dry wt)}^{-1}$.

ment conditions, however, is the lack of uniformity in the proportion of surface SOM redistribution among all sedimentary deposits. Sta. 1001, the most littoral station within a protected cove, seems more affected by erosional processes (F_o max: 20–30%) than the central lake sediments of Sta. 1200 (F_o max: 6–8%) and the narrow channel area of Sta. 999 (F_o max: 2–3%). In reservoir systems such as Cabonga, a spatial heterogeneity in sedimentary dynamics as well as ecological attributes has been invoked to result from their

large storage capacity and dendritic morphology (Thornton 1990; Osidele and Beck 2004). The spatial heterogeneity in allochthonous organic matter redistribution observed in this study may thus be attributed to the many embayments and narrow channels interspersed by islands (Sta. 1001 and 999; Fig. 1) with limnological characteristics that are different from those of the adjacent main-channel sections of the reservoir (Sta. 1200). Additionally, although the two models tend to provide relatively comparable estimates of F_o at most



depths, in recent sediments of Sta. 1001 the P:(V+S) model consistently generates higher F_o estimates than those obtained from the 3,5-Bd:V model (factor of 1.5–2). We discussed previously the sensitivity of the P:(V+S) parameter to small shifts in constituent yields within any end-member. The discrepancy between the predictions from the two models in recent sediments of Sta. 1001 may thus suggest that the P:(V+S) ratio is more prone to overestimating surface SOM sources when such inputs are large. At Sta. 1001, the strong correspondence existing between surface SOM fractions estimated with the 3,5-Bd:V model and the measured shifts in C:N ratios (Fig. 12) implies that this modeling approach records more accurately the perturbations to sedimentary systems and may be more conservative than P:(V+S) under large erosional inputs.

Mixing model: Autochthonous inputs—The parallel post-flooding increase in the proportion of surface SOM and C:N ratios at Sta. 1001 and 1200 (Fig. 12) are in line with an increased proportion of nitrogen-poor vascular plant macrodebris from litter constituents in recent sediments. In contrast, at Sta. 999 although our estimated value of F_o does increase after impoundment (as well as lignin yields, Fig. 6), a delayed decrease in C:N values suggests that this latter ratio may record a substantial influence of water column productivity on sedimentary elemental signatures (Meyers and Ishiwatari 1993; Herczeg et al. 2001; Routh et al. 2004).

Because lake sediments receive organic matter derived from both water column productivity and watershed inputs, we thus tested our mixing model using the specific C:N signatures in soil versus plankton end-members to quantify the relative contribution of allochthonous versus autochthonous organic matter in sediments. The elemental composition of the composite allochthonous pool was calculated using the proportions of its two sources in sediments (F_o and F_i) estimated with both 3,5-Bd:V and P:(V+S) models, as well as their respective mass-normalized concentrations (Table 2). The elemental composition of the autochthonous end-member was derived from measured values of plankton collected from boreal reservoirs and natural lakes in the region of study (Table 2; Montgomery 2000). The resulting plankton C:N values ranged 6–10 and were consistent with published values for organic matter derived from water column productivity in freshwater lakes (Meyers and Ishiwatari 1993; Meyers and Teranes 2001).

In each core, we represent the estimated proportion of autochthonous organic matter as an “enrichment factor” along the core depth with respect to pre-impoundment conditions (Fig. 13). This enrichment factor was calculated by normalizing the estimated proportion of autochthonous organic matter in every sedimentary interval to the average proportion of autochthonous organic matter accumulated in pre-impoundment sediments. This approach allows us to discuss relative variations in the proportions of autochthonous

Fig. 12. Vertical profiles of the calculated surface SOM in the allochthonous organic fraction (F_o) using both models and measured C:N ratios in sediments of the Cabonga Reservoir.

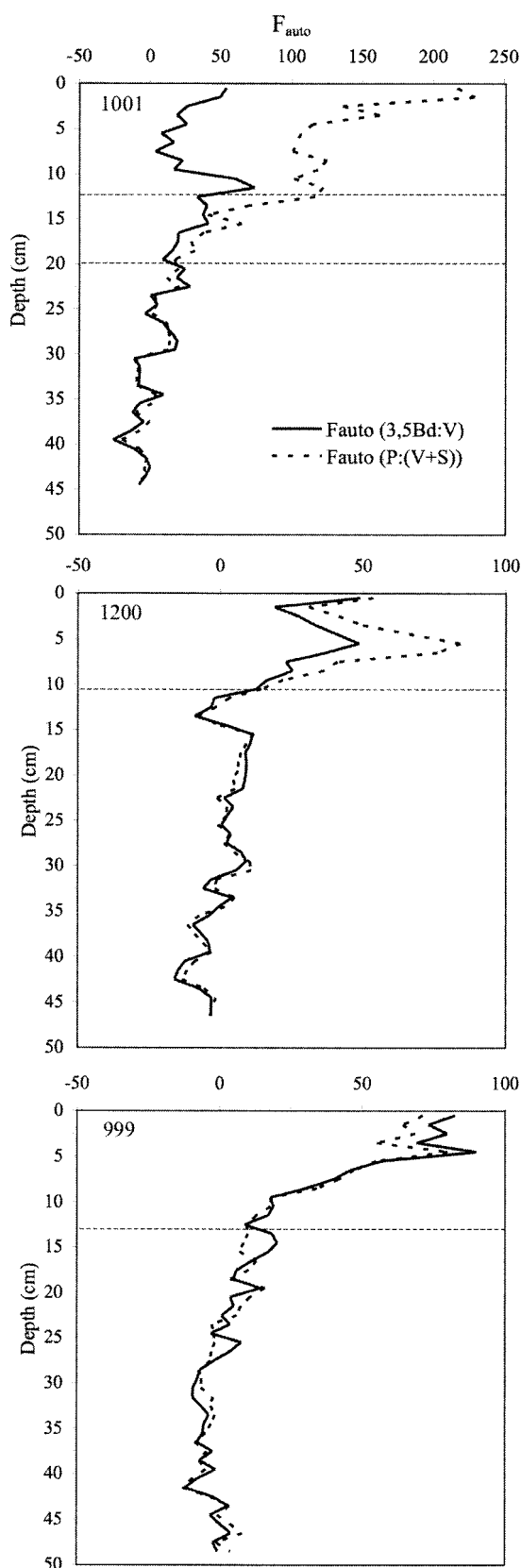


Fig. 13. Vertical profiles of the calculated autochthonous organic fraction (F_{auto}) in sediments of the Cabonga Reservoir using both models (3,5-Bd:V and P:[V+S] ratios).

organic matter in recent sediments due to impoundment, rather than to assign absolute values to these proportions in the lake system.

The low variability of F_{auto} in pre-impoundment sediments of all lakes studied (<10% of average) shows that the proportion of autochthonous organic matter was relatively constant in each individual lake prior to the building of the reservoir. Recent sediments, however, exhibited a marked and synchronous enrichment (1.5–2) in the proportion of autochthonous organic matter in the central lake sediments (Sta. 1200) and the narrow channel (Sta. 999). Although the 3,5-Bd:V model suggests there has also been a slight increase in the proportion of autochthonous organic matter at Sta. 1001, this fraction is estimated to have increased two- to threefold under the P:(V+S) model's predictions. Because of the better match between the F_o values derived from the 3,5-Bd:V model and C:N sedimentary signatures (*see discussion above*), we believe that this latter model is more conservative and less sensitive than P:(V+S) to specific source variations. Hence, in most sediments the values provided by the P:(V+S) model in surface sediments should be regarded as maximum estimates, particularly under strong surface SOM inputs.

Although an increase in autochthonous organic matter proportions seems counterintuitive with respect to previously demonstrated increased proportions of surface SOM inputs (Fig. 12), it also suggests that redistribution of eroded SOM to the water column may have enhanced in situ production. Indeed, to maintain sedimentary C:N values at ~ 28 , ~ 18 , and ~ 15 in recent sediments of Sta. 1001, 1200, and 999, respectively, inputs of nitrogen-rich autochthonous organic matter need to increase to counterbalance the substantial increases in the proportion of surface SOM (F_o), which raises the C:N ratios (ranging 37–60) of the total allochthonous pool reaching the sediments. Moreover, it has been shown that in reservoirs the recycling of allochthonous organic matter enhances aquatic productivity and that the main pathway of carbon flow to secondary producers shifted from an autochthonous to an allochthonous base originating from flooded vegetation (Paterson et al. 1997). In some natural lakes, the substantial increase in runoff of soil nutrients resulting from large-scale perturbations to their watershed (deforestation, Carignan et al. 2000; wetland drainage, Hambright et al. 2004) has also been invoked to explain enhanced levels of primary productivity and concomitant transfer of algal carbon to sediments (Meyers 2003). The study at the Experimental Lake Area in northwestern Ontario (Paterson et al. 1997) revealed that the response of in situ productivity and food web structure to reservoir flooding is rapid and results from pulse inputs of phosphorus, nitrogen, and dissolved carbon to the water column (c.f. Schetagne 1994). Similar dynamic responses in secondary production have been demonstrated in experimental manipulations of dissolved inorganic carbon sources in two natural lakes, suggesting that up to 50% of zooplankton carbon could be derived from terrestrial detrital sources in these systems (Pace et al. 2004).

In the Cabonga Reservoir—even after 70 yr of impoundment—we observe that the proportion of sedimentary autochthonous fractions remains higher than during the pre-

flooding state, particularly in its central section, which should integrate larger scale homogeneous processes, rather than the heterogeneous processes characteristic of littoral zones (c.f. Hambright et al. 2004; Osidele and Beck 2004). Such long-standing change is consistent with the phenomenon of hysteresis observed in lakes receiving pulsed inputs of nutrients (van der Molen et al. 1998; Scheffer et al. 2001; Hambright et al. 2004). In shallow lakes initially exposed to nutrient enrichment, internal nutrient loading and resuspension of sediments are strong stabilizing factors supporting continued eutrophic conditions despite amelioration of external loadings (Kennedy and Walker 1990; van der Molen et al. 1998). This effect is compounded by increased hydraulic residence times in reservoirs (Hambright et al. 2004). Because we observe a continued effect of erosion and remobilization of surface SOM in the reservoir of study (Fig. 12), we hypothesize that such a hysteresis effect may be effective here due to internal loadings of nutrients from translocated soil constituents and thus may explain long-term changes in sedimentary proportions of autochthonous organic matter.

Because we now know that bacterial respiration of terrestrial allochthonous carbon sustains net heterotrophy in many freshwater and estuarine systems (Cole and Caraco 2001; Pace et al. 2004), it is imperative that we investigate the fate of eroded SOM, and its potential support of heterotrophic metabolism, through the construction of quantitative mass balances at the watershed scale. Because benthic degradation of SOM cannot explain long-term GHG emissions from reservoirs (Houel 2003), mass balance calculations including evolution of carbon and nutrient fluxes to sediments over time may help better constrain the source of organic matter responsible for direct and indirect reservoir GHG emissions.

Finally, this study also provides some grounds to reconsider the generally accepted concept that allochthonous organic matter in sedimentary deposits of reservoirs are derived predominantly from the watershed and transported into the reservoir by headwater streams (Thornton 1990). The results presented here suggest that internal dynamics, such as erosion and soil translocation, can supply significant amounts of materials (including SOM) to deep sedimentary deposits in reservoirs. Whether there exists a balance between eroded/remobilized SOM and the fraction that re-accumulates in deep deposits still remains to be tested. Any losses in SOM during transit from eroded flooded soils—through degradation, photooxidation, or simply export by hydrodynamic forcing—may ultimately contribute to enhanced GHG evasion rates either directly from the surface of the reservoirs or indirectly from the spiraling of that material into receiving streams and, most importantly, estuaries (c.f. Rabouille et al. 1998; Cole and Caraco 2001). The mass balance of such redistribution terms from flooded soils thus remains a question to be addressed in assessing the role of reservoirs as potential long-term GHG emitters.

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