Seasonal variability in the sources of particulate organic matter of the Mekong River as discerned by elemental and lignin analyses

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[1] The Mekong River ranks within the top ten rivers of the world in terms of water discharge and sediment load to the ocean, yet its organic matter (OM) composition remains unstudied. This river is experiencing anthropogenically forced changes due to land use and impoundment, and these changes are expected to intensify in the future. Accordingly, we monitored the composition (including vascular-plant signatures) of Mekong River fine particulate organic matter (FPOM) over a one-year period. Autochthonous production comprises a greater proportion of FPOM during the dry season than in the rainy season, as demonstrated by higher percent organic carbon values (7.9 ± 2.4 versus 2.2 ± 0.4%), lower yields of lignin normalized to carbon (0.40 ± 0.05 versus 1.1 ± 0.3 mg (100 mg OC)$^{-1}$), and an increase in N:C ratios toward phytoplankton values during the dry season (from 0.06 to 0.12). Changes in the lignin-phenol composition of FPOM suggest that gymnosperms contribute more toward FPOM composition during the dry season, with angiosperms dominating in the wet season. This is supported by calculations of the lignin phenol vegetation index of riverine FPOM, which increases between the dry to wet seasons (dry: 29.4 ± 15.0 versus wet: 74.6 ± 17.3). These changes likely reflect seasonal differences in the proportion of flow that is coming from the Upper and Lower Basin, corresponding to compositional differences between the vegetation of these regions. Therefore, this work provides a baseline understanding of FPOM variability that can be used to assess how future change will affect this river.

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1. Introduction

[2] The role of rivers as transporters and processors of carbon (C) has recently been recognized to be a fundamental component of the global C cycle [*Cole et al.*, 2007; *Tranvik et al.*, 2009; *Aufdenkampe et al.*, 2011]. Inland waters receive 2.7 Pg C/year [*Battin et al.*, 2009; *Tranvik et al.*, 2009], which is a flux nearly equal to the size of the terrestrial sink for anthropogenic C (2.8 Pg C/yr) [*Canadell et al.*, 2007; *Battin et al.*, 2009]. Understanding the fate of C received by inland waters is of considerable interest, whether it is exported to the ocean, buried within the watershed, or lost to the atmosphere via gas evasion. The particulate organic carbon (POC) carried by rivers is an integral component of each of these fluxes. For example, POC comprises half of the organic carbon (OC) flux discharged by rivers to the ocean [*Degens et al.*, 1991; *Stallard*, 1998]. Deciphering the composition of this material is critical to assessing how much of this material will be permanently buried in marine depositional settings upon reaching saline waters [*Hedges and Keil*, 1995], versus oxidized via respiration while in transit. Understanding particulate organic matter (POM) composition also aids in understanding carbon reallocation on land in response to anthropogenic change. For example, it has recently been suggested that in-land reservoirs exceed OC burial in the ocean by at least 1.5-fold [*Cole et al.*, 2007]. As impoundments trap sediment from the watershed, deciphering the composition of the POM associated with these sediments is key to understanding the reactivity, and ultimate fate of this material. Further, knowledge of compositional attributes may aid in predicting the magnitude of C sequestration in terrestrial settings beyond the lifetime of impoundments (as dams themselves are short-lived) [*Cole et al.*, 2007; *Tranvik et al.*, 2009].

[3] Riverine POM may be derived from a variety of sources, including vascular plants, sedimentary rocks, and phytoplankton [*Hedges et al.*, 1986; *Devol and Hedges*, 2001; *Blair et al.*, 2004; *Coynel et al.*, 2005]. The organic composition of each of these sources differ, as does their lability. While in transit, the composition of these starting materials can be modified by numerous processes, including

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respiration, leaching, sorption, photoxidation, and lateral exchange with the floodplain [*Amon and Benner*, 1996; *Aufdenkampe et al.*, 2001; *Hernes et al.*, 2007; *Eckard et al.*, 2007; *Spencer et al.*, 2010; *Galy et al.*, 2011]. The interplay of these processes can be seasonally and spatially dynamic, leading to substantial variation in the quantity and quality of the organic C exported, particularly in tropical rivers experiencing intense seasonality in precipitation [*Richey et al.*, 1990; *Paolini*, 1995; *Coynel et al.*, 2005]. Such variability may then affect whether carbon is oxidized in transit or preserved in depositional environments. Despite the inherent importance of understanding seasonal variability in POM composition, time series studies documenting changes in the organic matter source of POM are generally lacking. This issue is especially important in the tropics as 64% of the global riverine organic C flux, and 60% of the inland-water gas evasion flux occurs in this region [*Meybeck*, 1982; *Aufdenkampe et al.*, 2011]; yet, the tropics remain poorly studied.

[4] Biomarkers, or compounds indicative of a unique biological source, can be powerful tools to trace the origin, transformation, and preservation of POM [*Goni and Hedges*, 1995; *Tareq et al.*, 2004; *Rezende et al.*, 2010], especially in systems, such as rivers, with a vast heterogeneity of sources. In this study we use time series measurements of biomarker composition to enhance our understanding of these simple, yet, fundamental questions regarding C cycling in fluvial systems: (1) what are the biological sources of POM carried by a large tropical river? and (2) how does the source and degradation-state of POM vary with the hydrological cycle? To answer these questions, we monitored monthly changes in the organic composition of suspended sediment carried by the Mekong River, a globally significant tropical river basin. Specifically, we measured nitrogen:carbon (N:C) ratios and stable isotopes ($\delta^{13}C$) of the fine suspended POM pool. At the molecular-level, we targeted the terrestrial component of fine POM (FPOM) by using lignin-derived phenols, which are biomarkers unique to vascular plants. In large tropical rivers, the carbon contained in fine suspended POM (or fine POC (FPOC)) represents 95% of total riverine particulate carbon [*Richey et al.*, 1990].

[5] Higher-plant derived biomass is an important reservoir of fixed carbon, and lignin represents 40% of the biomass of vascular plants. Consequently, lignin-phenols have been used extensively to understand the fate of terrestrially derived organic matter (OM) in freshwater and marine settings [*Hedges et al.*, 1982, 1986; *Walsh et al.*, 2008; *Rezende et al.*, 2010]. As a basis for determining the vascular-plant contribution to riverine FPOC, we analyzed the composition of the fresh-plant sources from which lignin originates: bark, leaf, and wood tissues from angiosperms found within the Mekong Basin. Considering that higher plants can be degraded to form soil-derived OM, and ultimately converted to carbon dioxide through respiration, comparing the lignin-phenol composition of riverine OM to the original vascular-plant signature provides information about the continental-level processing that occurs as this material travels from source to sink.

[6] The Mekong ranks in the top ten rivers of the world in terms of average sediment load and mean water discharge [*Milliman and Syvitski*, 1992; *Mekong River Commission*, 2005]. Although three dams currently exist on the main stem, the flow regime continues to be dominated by an annual monsoonal flood pulse [*Mekong River Commission*, 2005]. However, this may change in the near future. China is building a series of eight reservoirs along the main stem in the Upper Basin, and in September 2010, Lao People's Democratic Republic petitioned the Mekong River Commission to install eleven dams along the main stem in the Lower Basin [*Grumbine and Xu*, 2011]. In addition to altering the OM flux and composition, these changes will likely impact the Tonle Sap Lake, which is one of the world's most productive freshwater fisheries, as seasonal inputs of sediment and nutrients from the Mekong River have been suggested to be important to the fishery [*Mekong River Commission*, 2005; *Kummu et al.*, 2008]. Despite the potentially large downstream effects of changing the Mekong's sediment load, no study has examined the organic composition of material carried by the Mekong. Our data set is the first attempt to characterize the flow of C through the Mekong prior to major alteration from the building of these dams. Further, an estimate of the flux of particulate organic carbon exported by the Mekong is currently absent from the literature. We consequently present a conservative estimate of the annual flux of suspended sediment, particulate organic carbon, and particulate lignin exported by the Mekong River for the 2006 hydrologic year.

2. Methods

2.1. Study Area

[7] The Mekong River originates in the mountains of the Tibetan Plateau at an elevation of 4,968 m, and flows through China, Myanmar, Lao People's Democratic Republic, Thailand, Cambodia, and Viet Nam during a 4,800 km journey to the South China Sea (Figure 1) [*Hori*, 2000]. The drainage basin (795,000 km^2) has been divided into two regions: the Upper and Lower Basin. In the Upper Basin, the river flows 2,200 km from the headwaters in Tibet to the Lower Basin boundary at the Chinese border. During this journey, it descends 4,000 m in an incised channel, where it is primarily confined to narrow, deep gorges, with few opportunities for sediment deposition [*Mekong River Commission*, 2005; *Gupta*, 2005]. Although its area is only 24% of the watershed, 50% of the sediment exported by the Mekong River comes from the Upper Basin [*Kummu and Varis*, 2007; *Walling*, 2005]. The Lower Basin consists of the portion of the watershed draining the countries downstream of China (Lao People's Democratic Republic, Myanmar, Cambodia, Thailand, and Viet Nam). This 2,600 km stretch of the main stem begins almost entirely mountainous, but then gradually enters a low-relief region, with the flow of the river slowing and the channel widening. Although there are few opportunities for sediment deposition, the main sediment sink on the main stem of the Lower Basin is from Luang Prabang to Nong Khai [*Wang et al.*, 2011], possibly due to local geomorphology in which sediment is stored in the channel against cut banks [*Gupta et al.*, 2002]. It is not until Kratie, Cambodia, that an extensive floodplain develops. The first distributaries form at Phnom Penh, marking the beginning of the Mekong River delta [*Mekong River Commission*, 2005].

[8] The basin is home to over 70 million people and has experienced extensive anthropogenic change. In both the Upper and Lower basins, forests have been replaced with

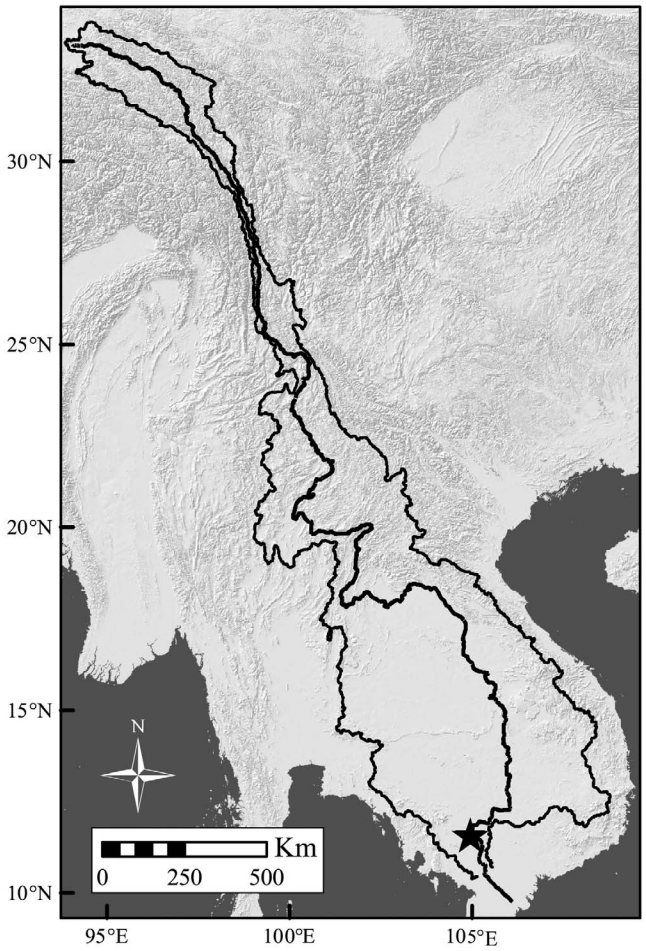

Figure 1. The Mekong River Basin in Southeast Asia. The location at which sampling was conducted for this study is denoted with a star.

agricultural lands and rice production is widespread [*Mekong River Commission*, 2005; *Costa-Cabral et al.*, 2008]. Sixty-four dams are currently in operation within the basin. Most of these are small to medium-sized reservoirs found on tributaries of the Mekong River, although three are located on the main stem in the Upper Basin [*Xue et al.*, 2011]. The impact of the latter impoundments on the sediment load of the river remains unclear due to the sporadic measurements of long-term suspended sediment concentrations prior to dam installation [*Wang et al.*, 2011; *Xue et al.*, 2011]. Regardless, the Mekong River is currently considered to be one of the few large river basins that has not been irreversibly modified by dams [*Kummu and Sarkkula*, 2008].

[9] The climate of the Lower Mekong basin has two monsoon seasons, with the southwest and northeast monsoons dominating during the rainy and dry seasons, respectively. Due to the arrival of the southwest monsoon in May, the Mekong rises from discharges less than 2,500 m^3/s during the low-water period (December through May) to peak flows exceeding 40,000 m^3/s during the rainy season [*Mekong River Commission*, 2005]. During this period, the highest precipitation occurs in the Lower Basin, specifically in southwestern Cambodia and in the Eastern Highlands of Laos (where annual precipitation exceeds 2,500 mm), where rainfall peaks in August. Lowest-year-round precipitation occurs in the Himalayan Highlands of the Upper Basin (500 mm), where precipitation falls largely as snow [*Costa-Cabral et al.*, 2008]. Runoff is dominated by snowmelt from China during the dry season. Much of the annual temporal variability in runoff results from these monsoonal precipitation patterns in addition to basin-wide topography. However, soil moisture levels can explain a significant proportion of the variability, which lags the precipitation peak [*Costa-Cabral et al.*, 2008]. As a result, the Upper Basin contributes over 35% of the Mekong's total flow during the dry season but only 18% over the entire year, indicating that runoff derived from the Upper Basin is disproportionately important during the dry season relative to the rainier periods [*Mekong River Commission*, 2005; *Kummu and Varis*, 2007]. During the latter time, inputs from tributaries from the Lower Basin dominate the hydrograph, with tributaries from Laos being particularly important [*Mekong River Commission*, 2005]. Part of the dry season flow, and the initial rise during the rainy season, is due to the delivery of snowmelt from the mountains [*Kummu and Varis*, 2007; *Wang et al.*, 2011].

[10] In 2006 (the year of this study), the hydrograph was defined by an unusual second peak in discharge that occurred during the first week of October in response to the tropical storm Xangsane. This storm was widespread over the central and southern portion of the Mekong Basin, with some areas receiving as much as 400–500 mm of rain. Without this storm, the volume of water discharged during the flood season in the Lower Basin would have been among the lowest ever recorded over the past 80 years [*Mekong River Commission*, 2007].

2.2. Field Measurements

[11] Water samples were collected on the Mekong River just north of the confluence with the Tonle Sap River (11.595617°N, 104.94299°E). This site was selected because it is not affected by seasonal changes in flow from the Tonle Sap Lake, and it is the most downstream point on the Mekong River that remains uninfluenced by tides and/or salinity [*Mekong River Commission*, 2005]. Samples were collected once to twice a month between 6 January 2006 and 1 November 2006. Water samples were collected at half of the total depth of the river using an in-line submersible pump. This depth was chosen as it has been previously demonstrated to yield a representative sample of the silt and clay-sized particles as compared to sediments obtained from a depth-integrated sampling scheme [*Moody and Meade*, 1994].

[12] Coarse particulate organic carbon (CPOC) was first removed with a 63 μm sieve due to the observation that it comprises a small proportion of the total organic carbon pool of similar large tropical rivers [*Richey et al.*, 1990]. Sieved river water was homogenized with a churn splitter [*Wilde and Radtke*, 2003], then filtered onto a pre-weighed 0.45 μm cellulose acetate filter. The fine suspended sediment (FSS) concentration was determined by the mass difference of the dry filter weighed before and after FSS collection. A known quantity of sieved water was also passed through a precombusted GF/F filter, which was later analyzed for N:C ratios, $\delta^{13}C$, and lignin-derived phenols. All filters were dried for 24 h at 60°C.

[13] A variety of plant end-member types from various aquatic and upland areas around Cambodia were collected, including the Tonle Sap Lake, the Mekong River, the Kulen Mountains (upland primary forest), and agricultural settings outside of the cities of Siem Reap and Phnom Penh, taking care to select vegetation that is representative of that which is growing in the basin. Vegetation was air-dried and coarsely ground prior to analysis. When necessary, bark was first removed from wood samples. Wood, bark, and leaves were analyzed for lignin-derived phenols from 17 angiosperm species.

2.3. Hydrograph Measurements

[14] We used the definitions applied by the Mekong River Commission to determine the onset of the different stages of the hydrograph [*Mekong River Commission*, 2007]. The rising water stage began when discharge rose to over twice that of the annual minimum. The flood season occurred when the daily discharge was greater than the mean annual discharge, and the falling water stage began when a consistent decrease in daily flow of 1% was observed daily for a two-week period. Discharge at Kratie, Cambodia was obtained from records provided by the Mekong River Commission.

2.4. Laboratory Analyses

[15] Carbonates were removed from FSS on GF/F filters by vapor-phase acidification prior to organic C analyses [*Komada et al.*, 2008]. Wetted filters were placed in a glass desiccator with 125 mL of 12 N HCl overnight. Acidified filters were then air-dried for 24 h, and then dried at 60°C overnight. N:C ratios and δ^{13}C were measured using a PDZ Europa ANCA-GSL elemental analyzer interfaced to a PDZ Europa 20–20 isotope ratio mass spectrometer (IRMS) at the Stable Isotope Facility at the University of California-Davis with a precision of 0.2‰. Samples were run with laboratory standards calibrated against NIST standard reference materials. δ^{13}C is expressed relative to PeeDee Belemnite. FPOC concentrations were obtained by multiplying weight % OC by the FSS concentration. The weight % OC and δ^{13}C of the vegetation end-members were analyzed at the University of Washington's IsoLab using a Thermo-Finigan MAT 253 IRMS coupled with a Costech ECS4010 elemental analyzer with a precision that ranged from 0.1 to 0.3‰.

[16] To liberate lignin phenols from sample matrices, samples were subjected to microwave CuO oxidation [*Goni and Montgomery*, 2000]. Prior to microwaving, the sample (either the finely ground plant or the GF/F filter containing the sediment) was placed in an acid-washed Teflon vessel with copper (II) oxide powder, ferrous ammonium sulfide, and 20 mL of 2 M NaOH. For each sample, the size was adjusted so that between 3 and 5 mg of OC was present in each vessel. Nitrogen-purged vessels were microwaved in a MARS 5 Accelerated Reaction System that was held at 144°C for ninety minutes at a pressure that ranged from 45 to 70 PSI. Once the reaction was complete, 50 μL of 1 mM recovery standards (ethyl vanillin and trans-cinnamic acid) were added. The sample was acidified to pH 1, and phenols were extracted using ethyl acetate. Water was removed using sodium sulfate, and the extract was dried under a gentle stream of N_2 gas at 45°C. The sample was then resuspended in pyridine, and lignin-derived phenols were derivitized with trimethylsilyl ethers and esters using BSTFA (N,O-bis(trimethylsilyl) trifluoroacetamide) and TCMS (trimethylchlorosilane). Phenols were quantified by gas chromatography using flame ionization detection (GC-FID). Blank GF/F filters were used to calculate blank-corrected lignin phenol concentrations. Filters were subject to a solvent extraction step prior to being microwaved so that the extracts could be used for subsequent lipid analyses. Comparisons between extracted and non-extracted samples indicated that solvent extraction did not affect lignin recovery.

[17] The effects of over-drying the ethyl acetate extracts on the recovery of phenols were assessed because this can lead to significant loss of adehyldes and ketones [*Keil and Neibauer*, 2009]. The recovery standard, ethyl vanillin, was subject to more loss than trans-cinnamic acid. The following phenols were corrected for loss during drying from post-microwave processing using ethyl vanillin: 4-hydroxybenzaldehyde, vanillin, and acetovanillin. All others were corrected with trans-cinnamic acid.

[18] λ_8 is reported as the sum of the eight phenolic products within the individual vanillyl (V), syringyl (S), and cinnamyl (C) families, normalized by 100 mg of OC (i.e., mg V + S + C/100 mg OC) [*Hedges and Mann*, 1979]. Further, Σ_8 is the sum of these same 8 phenols normalized to 10 g of bulk sediment (i.e., mg V + S + C/10 g sediment). We report C:V and S:V ratios (the ratio of the lignin-phenol derivatives in the cinnamyl and syringyl families to that of the vanillyl family) to differentiate samples among tissue types. We also express the ratio of the acidic functional group (ad) to the aldehyde (al) group within the vanillyl and syringyl families (denoted as (ad:al)v and (ad:al)s) as an indicator of degradation [*Hedges et al.*, 1982; *Ertel and Hedges*, 1984]. Finally, we calculated the Lignin Phenol Vegetation Index (LPVI) for both plant end-members and sediments to further identify the vegetation source of riverine POM [*Tareq et al.*, 2004]:

$$LPVI = [S(S+1)/(V+1)+1]^*[C(C+1)/(V+1)+1] \quad (1)$$

where V, S, and C are expressed in % of the λ_8.

[19] The total flux of FSS, FPOC, and particulate lignin exported by the Mekong River at Phnom Penh was calculated for the 2006 hydrologic year. The FSS flux was calculated by linearly interpolating between the field measurements of FSS to produce daily values, which were then multiplied by daily discharge values, and then summed over one year. To obtain a FPOC flux, percent OC values were taken from field measurements, linearly interpolated between samples to a daily record, and then multiplied by the daily FSS flux, and summed accordingly. Finally, the particulate lignin flux was calculated from daily interpolations of the percent lignin from the FSS flux.

2.5. Statistical Analyses

[20] All statistical analyses were performed in version 15.0 of SPSS. Only comparisons between mean values of environmental parameters were made between the dry and rainy season, as only two samples were collected during the rising-water period. We used the Mann-Whitney U-test to determine significant differences in a variable between two groups, the Kruskal-Wallis test to determine differences among more than two groups. Further, Spearman's rank

Table 1. The Organic Composition of Angiosperm End-Members Found in the Mekong River Basin[a]

Name	Type[b]	Tissue[c]	N:C	$\delta^{13}C$	% OC	S:V	C:V	(Ad:Al)v	(Ad:Al)s	λ_8	Σ_8	LPVI
Aglaia pisifera	A	W		−27.3	38.4	1.48	0.01	0.11	0.15	20.1	773.1	90.0
Alstonia scholaris	A	W		−29.5	43.0	1.89	0.03	0.15	0.18	21.9	943.8	130.2
Antidesma ghaesembilla	A	W		−29.6	40.1	1.82	0.02	0.13	0.16	21.2	852.6	120.0
Artocarpus heterophylus	A	W		−32.4	41.8	3.48	0.02	0.09	0.12	26.5	1107.9	285.8
Baccaurea ramiflora	A	W		−29.1	37.4	0.67	0.04	0.17	0.18	13.3	497.4	31.7
Cassia siamensis	A	W		−31.0	46.2	2.15	0.01	0.14	0.13	21.5	994.9	147.2
Dipterocarpus alatus	A	W		−33.2	42.3	2.02	0.03	0.11	0.34	9.1	385.3	140.0
Feroniella lucida	A	W		−30.8	42.1	1.29	0.01	0.18	0.17	19.2	808.3	73.4
Garcinia lanessani	A	W		−30.2	41.9	0.72	0.02	0.15	0.01	17.9	747.5	32.1
Irvingina malayana	A	W		−30.5	42.0	1.58	0.01	0.13	0.18	17.1	715.8	98.0
Lagerstroemia calyculata	A	W		−29.3	45.4	1.86	0.01	0.13	0.16	18.2	825.6	122.2
Nephelium hypoleucum	A	W		−27.2	43.6	3.02	0.01	0.14	0.18	24.8	1081.1	223.3
Peltophorum dasyrachis	A	W		−30.4	41.1	2.15	0.01	0.12	0.13	19.0	782.4	147.6
Schleicheria oleosa	A	W		−29.6	46.9	1.43	0.01	0.12	0.10	12.7	594.5	86.0
Streblus asper	A	W		−33.1	45.0	1.38	0.03	0.14	0.15	20.6	926.6	84.2
Tamarindus indica	A	W		−30.1	42.5	1.31	0.01	0.18	0.21	18.7	793.9	75.5
Srol wood	A	W		−29.6	43.3	3.04	0.02	0.13	0.12	20.3	877.9	229.2
Wood average (n = 17)				−30.2	42.5	1.92	0.02	0.14	0.16	18.9	806.4	124.5
Standard deviation				1.7	2.5	0.82	0.01	0.02	0.07	4.3	189.4	68.6
Aglaia pisifera	A	B	0.052	−29.5	46.4	0.98	0.86	0.18	0.29	2.2	103.7	939.0
Alstonia scholaris	A	B	0.039	−32.1	41.8	0.72	0.10	0.18	0.25	8.0	332.9	48.8
Antidesma ghaesembilla	A	B		−29.7	48.5	2.13	0.16	0.20	0.22	8.3	402.4	264.0
Artocarpus heterophylus	A	B	0.062	−34.0	46.7	0.85	0.11	0.23	0.33	8.6	409.0	67.0
Baccaurea ramiflora	A	B	0.048	−30.6	40.2	0.46	0.08	0.25	0.33	8.0	323.1	22.8
Cassia siamensis	A	B	0.054	−33.0	46.5	0.96	0.09	0.26	0.29	10.9	506.3	68.7
Dipterocarpus alatus	A	B		−33.4	46.2	1.78	0.10	0.11	0.61	6.7	310.0	159.1
Feroniella lucida	A	B	0.042	−31.7	48.1	0.74	0.58	0.20	0.22	5.7	275.6	386.6
Garcinia lanessani	A	B		−31.4	44.4	0.74	0.14	0.17	0.21	10.0	445.3	63.6
Irvingia malayana	A	B		−30.6	49.1	0.98	0.06	0.24	0.34	9.6	471.4	58.7
Lagerstroemia calyculata	A	B	0.047	−31.0	45.3	0.48	0.13	0.27	0.37	9.7	437.5	34.6
Nephelium hypoleucum	A	B	0.037	−32.1	49.7	0.91	0.09	0.20	0.23	8.7	434.3	63.1
Peltophorum dasyrachis	A	B		−34.5	46.7	0.66	0.11	0.20	0.23	5.6	261.3	147.3
Schleicheria oleosa	A	B	0.034	−30.4	50.7	0.47	0.19	0.17	0.17	4.2	212.3	48.9
Srol bark	A	B	0.043	−30.6	50.2	1.33	0.36	0.21	0.18	4.3	216.6	400.7
Streblus asper	A	B	0.065	−34.5	35.0	1.16	0.15	0.25	0.33	8.5	298.6	122.5
Tamarindus indica	A	B	0.035	−32.2	41.6	0.63	0.07	0.18	0.19	9.2	383.4	33.3
Bark average (n = 17)			0.046	−31.8	45.7	0.94	0.20	0.21	0.28	7.6	342.6	172.3
Standard deviation			0.010	1.6	4.1	0.45	0.21	0.04	0.10	2.4	107.7	230.1
Aglaia pisifera	A	L	0.048	−29.6	47.0	0.15	0.35	0.03	0.15	2.4	110.8	26.6
Alstonia scholaris	A	L	0.046	−29.2	36.5	1.25	0.24	0.09	0.14	2.2	81.5	224.0
Antidesma ghaesembilla	A	L	0.038	−31.8	34.1	0.87	0.16	0.11	0.13	3.2	110.5	92.4
Artocarpus heterophylus	A	L	0.050	−34.0	40.2	1.33	1.14	0.10	0.17	2.7	109.3	1981.3
Baccaurea ramiflora	A	L	0.042	−29.5	36.3	0.73	0.17	0.13	0.17	3.6	130.8	76.9
Borassus flabellifer	A	L	0.035	−32.2	38.0	2.20	0.14	0.13	0.18	14.0	530.4	247.5
Cassia siamensis	A	L	0.045	−31.7	45.9	1.03	0.32	0.17	0.20	6.3	288.4	258.3
Dipterocarpus alatus	A	L	0.039	−33.8	46.2	0.48	0.04	0.13	0.17	5.3	245.5	18.8
Feroniella lucida	A	L	0.054	−31.8	40.6	0.72	0.43	0.15	0.24	6.4	258.1	246.9
Garcinia lanessani	A	L	0.038	−30.8	43.3	1.41	0.16	0.11	0.13	7.0	300.9	166.5
Irvingina malayana	A	L	0.031	−30.0	43.0	0.77	0.13	0.16	0.20	8.0	344.3	65.0
Lagerstroemia calyculata	A	L	0.033	−30.2	46.0	0.99	0.09	0.12	0.16	6.4	294.4	71.3
Nephelium hypoleucum	A	L	0.039	−31.0	41.8	1.14	0.51	0.10	0.12	9.9	415.3	556.8
Peltophorum dasyrachis	A	L	0.031	−32.7	45.7	1.42	0.46	0.12	0.15	5.5	249.8	594.6
Schleicheria oleosa	A	L		−32.4	44.3	0.47	0.14	0.17	0.29	7.7	342.6	34.3
Srol leaves	A	L	0.049	−29.7	38.7	2.07	0.71	0.19	0.21	4.8	184.4	1641.7
Streblus asper	A	L	0.051	−32.9	33.4	1.66	0.48	0.11	0.12	2.1	68.9	744.6
Tamarindus indica	A	L	0.052	−32.6	38.2	0.89	0.47	0.14	0.19	5.0	189.9	366.5
Leaf average (n = 18)			0.043	−31.4	41.1	1.09	0.34	0.13	0.17	5.7	236.4	411.9
Standard deviation			0.008	1.5	4.4	0.54	0.27	0.04	0.04	3.0	125.6	553.7
Tonle Sap	M_g	L	0.064	−29.9	36.3	0.84	1.03	0.18	0.45	10.7	388.4	978.2
Treng	M_g	L	0.049	−11.9	43.4	0.79	1.28	0.16	0.46	10.2	441.8	1166.9
Bamboo	M_g	L	0.065	−27.3	34.7	1.70	1.29	0.16	0.28	15.7	531.8	2623.6
Eichornia crassipes	M_a	L	0.058	−29.7	40.6	0.08	0.98	0.45	0.36	1.0	40.3	65.8
Ipomoea aquatica	M_a	L	0.088	−28.0	36.4	0.19	1.19	0.27	0.31	2.3	82.6	165.1

Table 1. (continued)

Name	Type[b]	Tissue[c]	N:C	$\delta^{13}C$	% OC	S:V	C:V	(Ad:Al)v	(Ad:Al)s	λ_8	Σ_8	LPVI
Macrophyte average (n = 5)			0.065	−25.3	38.3	0.72	1.15	0.24	0.37	8.0	297.0	999.9
Standard deviation			0.015	7.6	3.6	0.65	0.14	0.12	0.08	6.2	221.5	1028.8
Grass average (n = 3)			0.059	−23.0	38.2	1.11	1.20	0.17	0.39	12.2	454.0	1589.6
Standard deviation			0.009	9.7	4.6	0.51	0.15	0.01	0.10	3.1	72.5	900.5
Aquatic plants (n = 2)			0.065	−28.8	38.5	0.14	1.08	0.36	0.33	1.6	61.5	115.5

[a]Attributes of the organic composition of angiosperms are defined as follows: N:C, nitrogen to carbon ratio; $\delta^{13}C$, ^{13}C of the bulk organic matter; % OC, percent organic carbon; S:V, syringyl to vanillyl ratio; C:V, cinnamyl to vanillyl ratio; (Ad:Al)v, acid to aldehyde ratio of vanillyl phenols; (Ad:Al)s, acid to aldehyde ratio of syringyl phenols; λ_8, mg of 8 lignin phenols (100 mg OC)$^{-1}$; Σ_8, mg of 8 lignin phenols (10 gdw)$^{-1}$; LPVI, Lignin Phenol Vegetation Index.
[b]Type denotes the type of angiosperm: A is a tree or shrub, M_g is grass, and M_a is an aquatic plant.
[c]Tissue types W, B, and L refer to wood (W), bark (B), and leaves (L).

correlation procedure was used to determine associations between variables.

3. Results

3.1. Plant End-Members in the Mekong Watershed

[21] The % OC of plant end-members ranged from 34.7 to 50.7 across all vegetation types, but was similar within vegetation types (Table 1). On average, angiosperm barks had the highest % OC values (45.7 ± 4.1%) and grasses had the lowest values (38.2 ± 4.6%). $\delta^{13}C$ values ranged from −11.9‰ to −34.5‰; grasses were the most enriched (averaging −23.0 ± 9.7‰) and angiosperm barks were the most depleted (averaging −31.8 ± 1.6‰).

[22] Angiosperm woods could be statistically differentiated from other angiosperm tissues by the high amount of lignin-derived phenols normalized to both C (λ_8) and gram dry weight (gdw) (for λ_8, $\chi^2 = 34.201$, df = 2, $p < 0.0005$ and for Σ_8, $\chi^2 = 34.238$, df = 2, $p < 0.0005$). Woody tissue λ_8 values averaged 18.9 ± 4.3 mg (100 mg of OC)$^{-1}$, whereas Σ_8 averaged 806.4 ± 189.4 mg (10 gdw)$^{-1}$). Leaves had considerably lower yields, with λ_8 averaging 5.7 ± 3.0 mg (100 mg of OC)$^{-1}$ and Σ_8 averaging 236.4 ± 125.6 mg (10 gdw)$^{-1}$. Bark fell between these values (Table 1).

[23] The average C:V ratio, which differentiates wood from non-woody vegetation [*Hedges and Mann*, 1979], increased among angiosperm tissues taken from trees and shrubs as follows: wood had the lowest C:V ratio (C:V = 0.02 ± 0.01), whereas barks were intermediate (0.20 ± 0.21), and leaves had the highest values (0.34 ± 0.27). Out of all vegetation types studied, macrophytes (grasses and aquatic plants) had the highest C:V values observed (1.20 ± 0.15 and 1.08 for grasses and aquatic plants, respectively) (Table 1). The S:V ratio differentiates angiosperms from gymnosperms, as expected, since gymnosperms do not make syringyl-phenol derivatives [*Hedges and Mann*, 1979]. Angiosperm wood had the highest S:V ratio (1.92 ± 0.82), and bark, leaves, and grasses had S:V ratios near 1 (see Table 1). Aquatic plants can be physically separated from other vegetation types based on the low S:V ratio (0.14) and high C:V ratio (1.08).

[24] The LPVI has recently been proposed to be an additional tool to partition OM among end-member sources [*Tareq et al.*, 2004]. Although the average LPVI value of angiosperm leaves was higher than that of wood and bark (Table 1), the high variability of the leaves (411.9 ± 553.7) and similar values of wood and bark tissues (124.5 ± 68.6 and 172.3 ± 230.1, respectively) indicates that it is not statistically possible to differentiate between these three tissue types using the LPVI ($\chi^2 = 3.464$, df = 2, p = 0.177). Grasses had the highest LPVI value of 1589.6 ± 900.5,

Table 2. Bulk Characteristics of Fine Suspended Sediment

Date	FSS[a] (mg/L)	FPOC[b] (mg/L)	% OC	N:C	$\delta^{13}C$ (‰)
Low-Water Stage					
6 Jan 2006	26.2	0.94	3.6	0.06	−30.3
20 Jan 2006	10.8	0.86	8.0	0.07	−29.9
3 Feb 2006	6.1	0.50	8.2	0.10	−31.3
17 Feb 2006	6.7	0.72	10.7	0.09	−29.2
2 Mar 2006	7.0	0.73	10.5	0.09	−28.8
17 Mar 2006	7.9	0.68	8.6	0.10	−30.4
31 Mar 2006	6.1	0.58	9.5	0.09	−29.4
21 Apr 2006	12.4	0.61	5.0	0.10	−29.3
19 May 2006	10.5	0.72	6.9	0.12	−27.7
Average	10.4	0.71	7.9	0.09	−29.6
Standard deviation	6.3	0.14	2.4	0.02	1.0
Rising-Water Stage					
9 Jun 2006	29.3	1.17	4.0	0.07	−30.4
23 Jun 2006	36.7	1.04	2.8	0.07	−30.5
Average	33.0	1.10	3.4	0.07	−30.5
Standard deviation	5.2	0.09	0.8	0.00	0.1
High-Water Stage					
7 Jul 2006	196.7	3.99	2.0	0.09	−29.5
21 Jul 2006	136.3	3.28	2.4	0.09	−29.3
4 Aug 2006	259.8	6.34	2.4	0.08	−30.1
18 Aug 2006	173.6	3.78	2.2	0.09	−29.0
6 Oct 2006	219.6	4.41	2.0	0.08	−29.7
20 Oct 2006	103.1	2.71	2.6	0.06	−31.1
1 Nov 2006	206.1	3.04	1.5	0.09	−29.8
Average	185.0	3.94	2.2	0.08	−29.8
Standard deviation	52.6	1.21	0.4	0.01	0.7
Overall average	80.8	2.01	5.2	0.09	−29.8
Standard deviation	91.4	1.75	3.3	0.01	0.9

[a]FSS denotes the FSS concentration (measured in units of mg/L).
[b]FPOC is the FPOC concentration (in mg/L). All other variables are defined in Table 1.

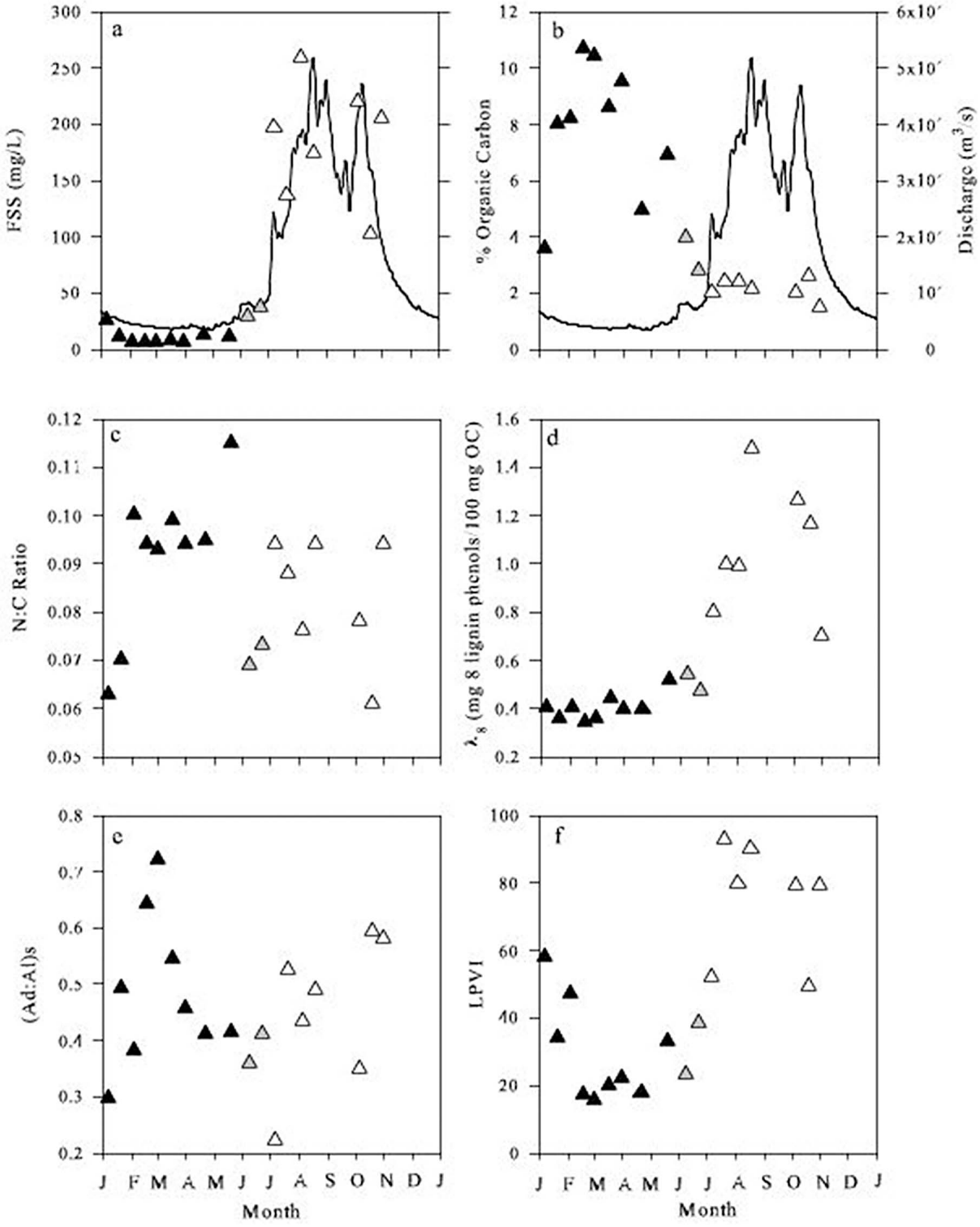

Figure 2. Seasonal variability in suspended sediment attributes found in the Mekong River at Phnom Penh. The hydrograph is plotted concurrently with (a) FSS concentration, (b) % organic carbon of FSS, (c) N:C ratio of FPOM, (d) λ_8 (mg lignin (100 mg OC)$^{-1}$), (e) (Ad:Al)s ratio, and (f) the LVPI of suspended sediment. The low-water, rising-water, and high-water periods are denoted by black triangles, gray triangles, and white triangles, respectively.

Table 3. Riverine Vascular-Plant Biomarker Concentration and Composition of FPOM[a]

Date	Total Lignin (μg/L)	Vanillyl (μg/L)	Syringyl (μg/L)	Cinnamyl (μg/L)	S:V	C:V	(Ad:Al)v	(Ad:Al)s	λ_8	Σ_8	LPVI
Low-Water Stage											
6 Jan 2006	3.82	2.14	1.36	0.32	0.64	0.15	0.34	0.30	0.41	1.46	57.9
20 Jan 2006	3.12	1.96	0.96	0.20	0.51	0.11	0.71	0.49	0.36	2.53	34.0
3 Feb 2006	2.00	1.19	0.60	0.21	0.50	0.17	0.31	0.38	0.40	3.30	47.4
17 Feb 2006	2.47	1.69	0.63	0.14	0.37	0.09	0.43	0.64	0.34	3.66	17.0
2 Mar 2006	2.63	1.81	0.68	0.13	0.38	0.07	0.62	0.72	0.36	3.74	15.9
17 Mar 2006	3.02	2.04	0.77	0.21	0.40	0.10	0.43	0.54	0.44	3.81	19.7
31 Mar 2006	2.28	1.49	0.66	0.13	0.44	0.09	0.52	0.46	0.39	3.74	22.1
21 Apr 2006	2.44	1.66	0.64	0.14	0.39	0.08	0.39	0.41	0.40	1.97	17.6
19 May 2006	3.76	2.28	1.27	0.22	0.56	0.10	0.29	0.42	0.52	3.59	32.8
Average	2.84	1.81	0.84	0.19	0.46	0.11	0.45	0.48	0.40	3.09	29.4
Standard deviation	0.58	0.31	0.26	0.06	0.09	0.04	0.14	0.14	0.05	0.92	15.0
Rising-Water Stage											
9 Jun 2006	6.35	4.13	1.83	0.39	0.44	0.09	0.50	0.36	0.54	2.17	23.4
23 Jun 2006	4.88	2.94	1.57	0.37	0.53	0.13	0.52	0.41	0.47	1.33	38.3
Average	5.62	3.53	1.70	0.38	0.49	0.11	0.51	0.38	0.51	1.75	30.9
Standard deviation	1.03	0.84	0.18	0.02	0.06	0.02	0.01	0.04	0.05	0.59	10.5
High-Water Stage											
7 Jul 2006	31.97	18.35	11.22	2.40	0.62	0.13	0.47	0.22	0.80	1.63	52.2
21 Jul 2006	32.73	16.60	13.19	2.94	0.79	0.18	0.62	0.52	1.00	2.40	93.0
4 Aug 2006	62.66	32.00	25.75	4.90	0.80	0.15	0.48	0.43	0.99	2.41	79.6
18 Aug 2006	55.67	28.37	22.24	5.06	0.78	0.18	0.49	0.49	1.47	3.21	89.9
6 Oct 2006	55.71	28.78	22.32	4.60	0.78	0.16	0.43	0.35	1.26	2.54	79.1
20 Oct 2006	31.65	17.87	11.55	2.23	0.65	0.12	0.77	0.59	1.17	3.07	49.2
1 Nov 2006	21.44	11.21	8.32	1.90	0.74	0.17	0.47	0.58	0.70	1.04	79.4
Average	41.69	21.88	16.37	3.43	0.74	0.16	0.53	0.46	1.06	2.33	74.6
Standard deviation	15.90	7.77	6.86	1.37	0.08	0.02	0.12	0.13	0.27	0.77	17.3
Overall average	18.26	9.81	6.98	1.47	0.57	0.13	0.49	0.46	0.67	2.64	47.1
Standard deviation	21.45	10.95	8.73	1.81	0.16	0.04	0.13	0.13	0.36	0.91	26.9

[a]Total lignin concentration is the sum of the vanillyl, syringyl, and cinnamyl phenol concentrations in (μg/L). All other variables are defined in Table 1.

but the range of LPVI values for angiosperm leaves overlaps with that of the grasses.

3.2. Mekong River Sediment Fluxes, Concentrations, and Organic Matter Compositions

[25] In 2006, the rising-, flood-, falling-, and low-water stages for the Lower Basin were from 13 May to 5 July, 6 July to 1 November, 2–28 November, and 29 November to 30 May, respectively [*Mekong River Commission*, 2007]. FSS concentrations varied by two orders of magnitude throughout 2006, with a minimum of 6.1 mg/L and a maximum of 259.8 mg/L (Table 2 and Figure 2a). They were positively correlated with discharge ($r = 0.850$, $p = <0.0005$, $n = 18$), with higher concentrations observed during the flood season compared to the dry season (185.0 ± 52.6 versus 10.4 ± 6.3) ($u = 0.000$, $p = 0.001$, $n = 16$). The total annual flux of FSS for 2006 was 76.3 MT/yr.

[26] FPOC concentrations ranged from 0.5 to 6.3 mg/L (Table 2) and were also positively correlated with discharge ($r = 0.904$, $p = <0.0005$, $n = 18$) and FSS ($r = 0.941$, $p < 0.0005$, $n = 18$). FPOC was fivefold higher during the flood-period than the low-water period (3.9 ± 1.2 mg/L versus 0.7 ± 0.1) ($u = 0.000$, $p = 0.001$, $n = 16$). The flux of FPOC exported by the Mekong at Phnom Penh was 1.67 MT/yr.

[27] The % OC of FSS also exhibited significant seasonal variability, with values ranging from 1.5 to 10.7% (Table 2 and Figure 2b). It was inversely correlated with discharge ($r = -0.847$, $p = <0.0005$, $n = 18$) and FSS concentrations ($r = -0.943$, $p < 0.0005$, $n = 18$). Values sharply declined from 10.7 to 4.0% as FSS concentrations increased from 7 to 29 mg/L, and then plateaued between 2 and 3% as FSS values increased to 260 mg/L thereafter. Dry-period values were higher than those from the rainy period (7.9 ± 2.4 versus 2.2 ± 0.4%, respectively) ($u = 0.000$, $p = 0.001$, $n = 18$). Maximum values peaked during the first-half of the low-water season (February 17), and declined to high-water values during the remainder of the low-water period.

[28] The N:C ratio of FPOM ranged from 0.06 to 0.11 and averaged 0.09 ± 0.02 (Table 2 and Figure 2c). It increased from 0.06 to 0.11 throughout the low-water season and was negatively correlated with discharge ($r = -0.479$, $p = 0.044$, $n = 18$). However, there was no significant difference between the low-water and high-water N:C ratios ($u = 17.500$, $p = 0.137$, $n = 16$). The $\delta^{13}C$ of FPOC ranged between -27.7 to -31.3‰ with no seasonal trends (Table 2).

3.3. Measurements of Lignin-Derived Phenols Associated With FPOM

[29] Lignin concentrations in the FPOM fraction ranged from 2.00 to 62.7 μg/L (Table 3) and were positively correlated with the hydrograph ($r = 0.909$, $p < 0.0005$, $n = 18$). Average high-water values (41.7 ± 15.9 μg/L) were over

fourteen-fold greater than low-water values (2.84 ± 0.58 μg/L). Vanillyl phenol concentrations were always the highest, followed by concentrations from the syringyl family, and then the cinnamyl family. The total lignin flux exported by the Mekong at Phnom Penh was 0.019 MT/yr.

[30] Σ_8 ranged from 1.04 to 3.81 mg (10 gdw)$^{-1}$ (Table 3), and there was no significant correlation between Σ_8 and discharge (r = −0.416, p = 0.086, n = 18). Total lignin yields (λ_8) ranged from 0.34 to 1.47 mg (100 mg OC)$^{-1}$ (Table 3 and Figure 2d). Values were positively correlated with discharge (r = 0.857, p < 0.0005, n = 18), with the dry season having lower λ_8 values (0.40 ± 0.05 mg (100 mg OC)$^{-1}$ than the flood period 1.1 ± 0.3 (100 mg OC)$^{-1}$ (u = 0.000, p = 0.001, n = 16). When λ_8 was divided into individual phenol families ($\lambda_{vanillyl}$, $\lambda_{syringyl}$, and $\lambda_{cinnamyl}$), the vanillyl family contributed the highest proportion of OC, followed by the syringyl, and then the cinnamyl families (mean λ_v = 0.4 ± 0.2, λ_s = 0.2 ± 0.2 and λ_c = 0.05 ± 0.03 mg C/100 mg OC) (data not shown). The S:V ratio ranged from 0.37 to 0.80 and the C:V ratio ranged from 0.07 to 0.18 (Table 3). Both parameters were significantly correlated with discharge (r = 0.866 and 0.724 for S:V and C:V, respectively) and each other (r = 0.830, p < 0.0005, n = 18). Values of (Ad:Al)v and (Ad:Al)s ranged from 0.29 to 0.77 and 0.22 to 0.72, respectively (Table 3). Neither was correlated with discharge, and no difference between these ratios was observed between seasons. However, the (Ad:Al)s ratio clearly peaked during the middle of the low-water season (Figure 2e). LPVI ranged from 15.9 to 93.0 and was also positively correlated with discharge (r = 0.857, p < 0.0005, n = 18) (Table 3 and Figure 2f), with dry season FPOM samples having lower LPVI values (29.4 ± 15.0) than rainy season samples (74.6 ± 17.3) (u = 2.000, p = 0.002, n = 16).

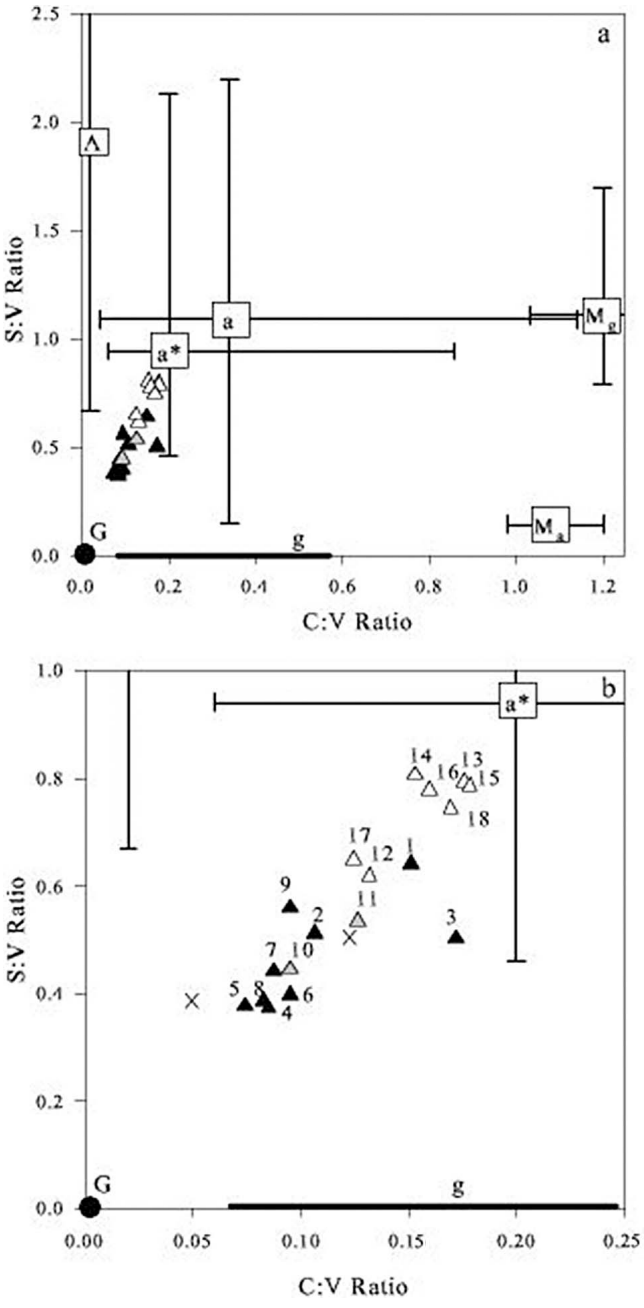

Figure 3. The S:V and C:V ratios of Mekong River FPOM and vegetation end-member tissues specific to the Mekong River Basin. FPOM samples from different seasons are differentiated using the same symbols described in Figure 2. End-member types are identified as follows: angiosperm woods (A), angiosperm leaves (a), angiosperm barks (a*), gymnosperm woods (G), gymnosperm leaves (g), grasses (M_g), and floating aquatic plants (M_a). Error bars indicate the range of values observed for each tissue type. Gymnosperm values were taken from *Hedges and Mann* [1979] and *Opsahl and Benner* [1995] as none were analyzed in this study. (a) FPOM is plotted among all the end-members. (b) Scaled with a higher resolution so that seasonal patterns can be observed. Samples are numbered in the order that they were collected, with the first collection occurring close to the beginning of the low-water period. Two additional samples from the Upper Basin are plotted in Figure 3b, which are represented with a cross.

4. Discussion

[31] The discharge and concentration of fine suspended sediment of the Mekong River varied by almost two orders of magnitude during the 2006 study period, with both parameters being dominated by the annual monsoonal flood pulse. Given this extreme variability, one might expect vast differences in POM composition between seasons. However, intensity in rainfall is not unique to the Mekong Basin, nor is seasonal variability in sediment concentrations. The Amazon River, which is perhaps the most thoroughly studied of the large tropical rivers, experiences little seasonal variability in POM composition relative to the large changes observed between dissolved and particulate organic matter [*Hedges et al.*, 1986; *Quay et al.*, 1992]. Such homogeneity is likely due to the role of the extensive lowland floodplain in both storing and resuspending previously deposited sediment during various stages of the hydrograph [*Hedges et al.*, 1986, 1994; *Dunne et al.*, 1998]. Although a floodplain does develop when the Mekong River reaches Cambodia, the Mekong is largely constrained to its channel during the vast majority of its journey [*Mekong River Commission*, 2005].

Accordingly, distinct compositional differences are observed in Mekong POM composition at both the bulk and the molecular level. We argue below that the variability in riverine POM source signatures over the course of one year is due to seasonal differences in autochthonous production, degradation, and the mobilization of sediment from different regions of the Mekong Basin.

4.1. Variability in Lignin-Derived Phenols in Regional Vascular Plants

[32] As organic matter composition is inherently linked to remineralization and carbon preservation in both marine and terrestrial reservoirs, understanding the biological source of riverine organic matter, and its associated reactivity, will improve our understanding of the global carbon cycle. Riverine organic matter is important to understanding the inland-water gas evasion flux because it is a substrate for heterotrophic respiration, which drives gas evasion [*Richey et al.*, 2002; *Cole et al.*, 2007; *Tranvik et al.*, 2009]. Several studies suggest that suspended particles contain labile OM that support bacterial metabolism and respiratory carbon dioxide fluxes in rivers [*Grossart and Ploug*, 2000; *Mayorga et al.*, 2005; *Ellis et al.*, 2012]. As a starting point to assess riverine OM source, it is first necessary to compare the same compositional signatures found in riverine POM to that found in the potential end-members from which this material is derived.

[33] Accordingly, we used lignin-phenols to deduce whether riverine FPOM is derived from vascular plants, and to assess how the vascular-plant contribution to riverine POM changes seasonally. The range of lignin-phenol compositions for fresh angiosperms found within the Mekong Basin suggests that riverine POM can potentially be partitioned among three angiosperm end-members as they have distinct signatures: (1) woods, (2) leaves and barks, and (3) macrophytes (Table 1 and Figure 3). To the best of our knowledge, ratios for barks have not previously been reported in the tropics, but the values reported here are similar to previous results from a watershed in the south of France [*Cotrim da Cunha et al.*, 2001]. Although there are gymnosperms growing within the basin [*Rundel*, 2007; *Costa-Cabral et al.*, 2008], none were found in the pristine forests visited in this study. Accordingly, the non-overlapping S:V ratios between fresh angiosperms from this study and gymnosperms reported from the literature [*Hedges and Mann*, 1979; *Opsahl and Benner*, 1995] enables riverine POM to be partitioned between these respective sources.

[34] The composition of Mekong angiosperms suggests that the LPVI may be of diminished utility as a tool to partition POM among end-member sources in this watershed. This index linearizes the S:V and C:V ratios to a single value [*Tareq et al.*, 2004]. Its presumed advantage is that each vegetation type and tissue type has non-overlapping boundary lines along this index. However, in this study, the LPVI of angiosperm woods, barks, leaves, and aquatic plants significantly overlap with each other (Figure 4 and Table 1) making differentiation between these sources using only the LPVI tenuous. The low amount of cinnamyl and/or syringyl phenols in some of the angiosperm leaf samples analyzed in this study (relative to those presented by *Tareq et al.* [2004]) could account for the overlap between the LPVI of angiosperm leaves and woods.

4.2. Seasonal Variability in Vascular-Plant Derived Particulate Organic Matter Transported by the Mekong River

[35] As the Mekong River descends from its headwaters to the sea, it drains a landscape with a rich diversity of vascular plant types, and transitions from cool temperate to tropical climates. Along the way, the Mekong passes through numerous vegetation zones with distinct communities. In the Upper Basin, subalpine shrublands and alpine meadows are found at elevations greater than 5,000 m, whereas coniferous forests dominate between 1,000 to 4,000 m in elevation, and evergreen and deciduous broadleaved forests are found at elevations below 1,000 m [*Costa-Cabral et al.*, 2008; *Rundel*, 2007]. Although conifers are found in some montane forests of the Lower Basin, agriculture production and broadleaf forests cover much of the lowlands [*Costa-Cabral et al.*, 2008], with deciduous dipterocarp (angiosperms) forests being the most common forest type of the Lower Basin [*Rundel*, 2007]. Therefore, considering that approximately twice as much flow (40%) comes from the Upper Basin during the dry season relative to the rainy season (20%) [*Mekong River Commission*, 2005], it is possible that the vascular plant composition of riverine FPOM could reflect seasonal variation in the origin of flow and the associated regional vascular-plant-derived OM that is concurrently mobilized.

[36] Our results indicate that vascular plants comprise a significant component of riverine FPOM (λ_8 values range between 0.34 and 1.47 mg (100 mg OC)$^{-1}$), with significant compositional changes observed between seasons. The most striking compositional trend observed using lignin phenols is that Mekong FPOM lies on a mixing line between angiosperm bark/leaf tissues and gymnosperm wood tissues, with clear compositional changes between seasons (Figure 3). Dry season samples had lower S:V and C:V ratios than rainy season samples, suggesting that gymnosperms exert a larger influence on FPOM composition during this period, whereas higher ratios during the rainy season suggest that angiosperms are proportionally more important. However, this trend could potentially be explained without invoking a change in sources between seasons, as degradation can cause the same compositional patterns. Degradation by white-rot fungi preferentially removes precursors of syringyl and cinnamyl phenols relative to vanillyl phenols, so degradation tends to force S:V and C:V ratios toward the origin [*Hedges et al.*, 1988; *Benner et al.*, 1991]. Thus, an alternative possibility is that all of the FPOM originates from angiosperm bark/leafy tissues, and seasonal differences in degradation state, not variability in tissue type, drives this compositional trend.

[37] To assess this possibility, we monitored the acid/aldehyde ratios of lignin phenols. Previous work has demonstrated that this ratio can be used to assess degradation given that ratios in diagenetically altered samples are elevated relative to those found in fresh plants [*Ertel and Hedges*, 1984; *Hedges et al.*, 1988; *Opsahl and Benner*, 1995]. Although the (Ad:Al)s and (Ad:Al)v ratios for FPOM were higher than our plant end-members (Tables 1 and 3), there was no observable difference between (Ad:Al)v and (Ad:Al)s between the dry and rainy season.

[38] As a second test to ensure that degradation is not driving the variability in C:V and S:V ratios between

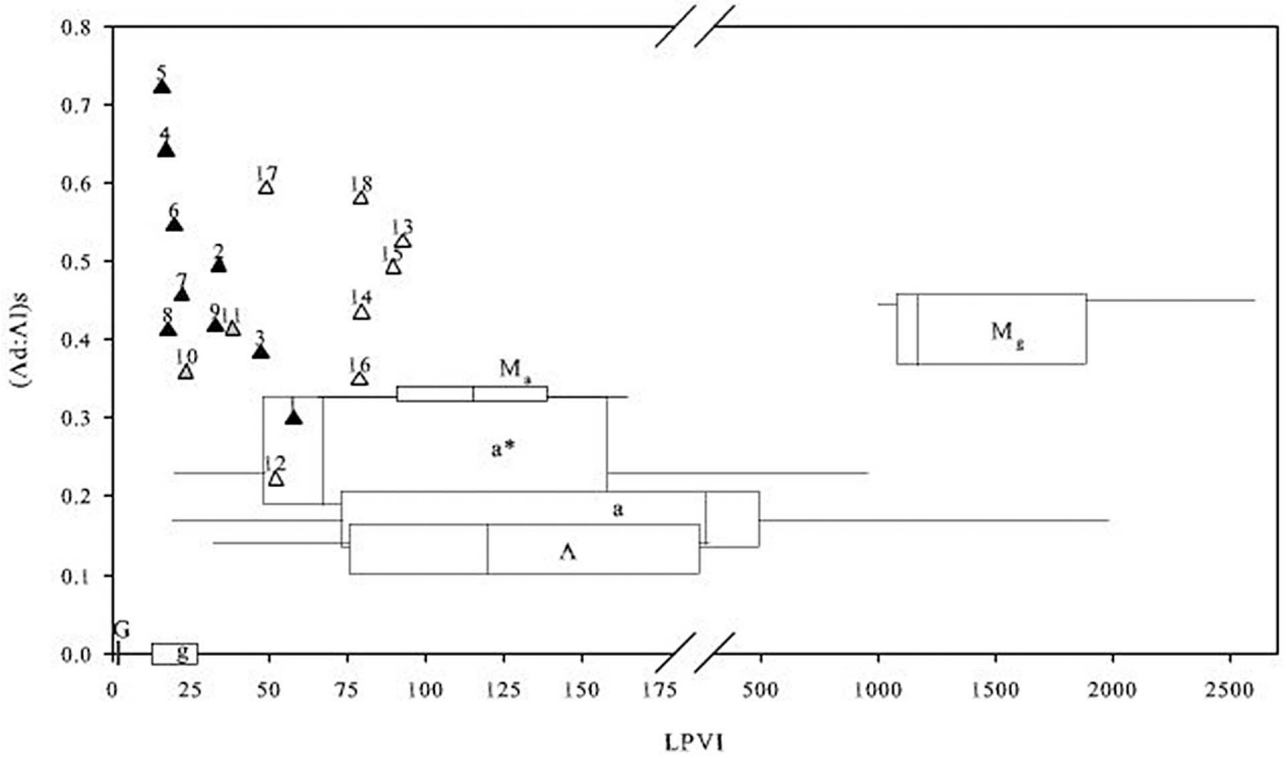

Figure 4. Lignin Phenol Vegetation Index as a function of the acid-to-aldehyde ratio of syringyl phenols of Mekong River FPOM samples. Symbols are as in Figure 2, and vegetation tissue is identified as in Figure 3. The LPVI and (ad:al)s of angiosperm end-members from this study are displayed using box-and-whisker plots. The range of LPVI values for gymnosperm samples are taken from *Tareq et al.* [2004]. Samples are numbered in the order that they were collected (see the Figure 3 caption).

seasons, relationships between the LPVI and Ad:Al ratios were examined. As proposed by *Rezende et al.* [2010], the LPVI should decrease with diagenesis given that cinnamyl and syringyl phenols are preferentially removed relative to vanillyl phenols during degradation [*Hedges et al.*, 1988; *Benner et al.*, 1991]. Accordingly, a negative correlation between Ad:Al ratios and LPVI should be observed if degradation is affecting FPOM signatures. No correlation was found for the whole data set (Figure 4), likely because both (Ad:Al)v and (Ad:Al)s were equally scattered within the dry and rainy season. In contrast, LPVI values were significantly lower during the dry season, with FPOM values lying within the range reported for gymnosperm leaves (12–27) [*Tareq et al.*, 2004], while rainy season FPOM values overlapped with the LPVI of angiosperms from the Mekong basin. Thus, the separation of dry and wet season samples by LPVI (which may be influenced by both diagenesis and OM source), but not (Ad:Al) (primarily affected by degradation), provides further support for changes in vegetation type being an important driver of variability in FPOM composition between seasons.

[39] While great differences in composition were observed between the wet and dry seasons, significant compositional changes were also observed within seasons. During the low-water period, the FPOM derived from angiosperms became more degraded as the dry season initially progressed, with (Ad:Al)s ratios abruptly peaking in the middle of the dry season (Figure 2e and Figure 4). Degradation by white-rot fungi occurs in subaerial environments [*Hedges et al.*, 1988], so it may be expected that degradation would increase as soils dry out. Therefore, the initial decline in LPVI to gymnosperm values (from sample 1 to samples 4–8) that is accompanied by elevated (Ad:Al)s ratios could be explained by the progressive degradation of FPOM derived from an angiosperm source as the low-water season progresses. However, after reaching its peak (sample 5), (Ad:Al)s fell from 0.72 to 0.41, consistent with less degraded material being transported at the end of the dry season. Interestingly, the LPVI experienced little change during this period, continuing to reflect a gymnosperm source. The large change in degradation state without a concurrent change in the LPVI suggests: (1) the LPVI is relatively insensitive to changes in degradation relative to (Ad:Al)s and/or (2) the importance of angiosperms relative to gymnosperms has become significantly reduced as the dry season progresses. Either way, both of these possibilities suggest that the decline in LPVI during the dry season is primarily controlled by the increasing importance of gymnosperms as the dry season progresses, with degradation being of secondary importance.

[40] Compositional variability occurs within the rainy period as well. The freshest angiosperm tissues may be transported during the beginning of the rainy season, likely being derived from undegraded angiosperm bark/leaf sources (Figure 4). As the rainy season progresses, the (Ad:Al)s ratio

of these angiosperms indicate that they generally become more degraded. Therefore, although the LPVI is consistent with the mean for angiosperm-bark end-members, the elevated (Ad:Al)s of these samples suggests that angiosperm leaves and wood may have been the ultimate starting material (Figure 4). The high variability observed in compositional signatures may be expected as many different mechanisms transport FPOM to the river during this period (i.e., overland flow, throughflow, mass wasting, sediment resuspension, etc.).

4.3. Mechanisms for a Seasonal Shift in Vascular Plant Composition

[41] It is likely that vascular-plant compositional changes in riverine FPOM are ultimately caused by seasonal differences in the contribution of various regions of the basin to the flow of the Mekong River. In addition to having proportionally more flow coming from the Upper Basin during the dry season relative to the wet season [*Mekong River Commission*, 2005; *Costa-Cabral et al.*, 2008], research conducted in the Lower Basin (in Laos) suggests that eroded material is simply redistributed among the hillslope initially during the rainy season; it is not until after the catchment is sufficiently saturated that precipitation-induced runoff is able to transport sediment [*Chaplot et al.*, 2005]. Therefore, it is possible that soils from the Lower Basin may become an increasingly important source of sediment as the rainy season progresses. Similarly, with more of the flow originating from the Upper Basin during the dry season, a greater proportion of dry season FPOM could be expected to be derived from Upper Basin sources relative to the rainy season. Therefore, the predominance of gymnosperms observed in dry season samples may reflect the abundance of coniferous forests of the Upper Basin. Similarly, the predominance of angiosperms during the wet-season may reflect the relative importance of these plant types in the Lower Basin [*Rundel*, 2007].

[42] To assess the likelihood that dry-season FPOM is derived from the Upper Basin, we analyzed POM samples taken from two locations from the main stem in the Chinese portion of the Upper Basin during the low-water period (i.e., April 2006). These samples cluster within the variability of the S:V and C:V ratios of our lowland dry-period samples (Figure 3b), and their origin is consistent with gymnosperms. Therefore, they provide further support that the FPOM exported from the Mekong Basin during the dry season has a gymnosperm compositional signature that emanates from the Upper Basin.

[43] Compositional parameters that are linked to sediment coming from different regions of the basin could be a useful tool to assess the impacts of reservoir construction in different regions. For example, China is building eight reservoirs along the main stem in the Upper Basin [*Grumbine and Xu*, 2011], with a likely consequence being that carbon that would otherwise have different downstream fates will likely remain trapped in the Upper Basin. Accordingly, a diminished gymnosperm signal could be expected in POM exported by the Mekong during the dry season. However, 200 other reservoirs are projected to be completed basin-wide over the next several decades [*Xue et al.*, 2011], making predictions as to how that could affect organic matter composition complex.

4.4. Variability in the Vascular Plant Contribution to Particulate Organic Matter

[44] Lignin-phenol concentrations (μg/L) of POM from the Mekong River followed the hydrograph, indicating that more plant-derived material is transported from the watershed during the wet season (Table 3). This is consistent with studies of POM on the Tech River in the south of France [*Cotrim da Cunha et al.*, 2001], and studies of dissolved OM in the Congo River basin [*Spencer et al.*, 2010]. In the Congo basin, higher Σ_8 and λ_8 values of dissolved OM occurred during the flush relative to the post-flush/dry season, which was attributed to the intense leaching of the organic-rich surface horizons and increased runoff rates during the rainy season. Low values during the dry season, in turn, were likely due to increased residence time (enabling more degradation), and the extensive leaching of the source material during the preceding flush period [*Spencer et al.*, 2010]. In this study, Σ_8 values of FPOM were not significantly different between seasons, suggesting that there was no variability in the dilution of the lignin pool by mass with sediment between seasons. However, it is possible that sampling at a higher resolution (i.e., more frequently than twice a month) may have been necessary to see significant differences in Σ_8, especially during the rising-water period when discharge increases rapidly. Regardless, the higher concentration of lignin (μg/L) observed during the rainy season is likely a result of more sediment mobilization caused by increased precipitation.

[45] Normalizing lignin yields by OC content (λ_8) indicates that a greater fraction of Mekong FPOC is derived from vascular plants during the wet season relative to the dry period: λ_8 is twice as high during the rainy season (Figure 2d and Table 3). Dilution of λ_8 by phytoplankton occurs in other river systems, such as the Mississippi [*Bianchi et al.*, 2007]. To assess this possibility, we examined the % OC of FSS. The global riverine average of 1% typically reflects the high mineral content of FSS, whereas higher values may be indicative of algae, which can be up to 50% OC [*Meybeck*, 1982]. Indeed, a strong inverse relationship was observed between suspended sediment concentrations and the % OC on the Mekong (Figure 2b). A likely explanation for this relationship is light inhibition of in situ production during the rainy season [*Ittekot and Laane*, 1991]. Alternatively, although there are generally few opportunities for sediment deposition along the Mekong, this same trend could be caused by an increase in sediment resuspension during the rainy season in the Lower Mekong. However, simultaneous work at this exact location demonstrated that the ratio of gross photosynthesis to community respiration was twice as high during the dry season relative to the rainy season during the period examined in this study (D. E. Lockwood, unpublished data, 2006). Accordingly, increased phytoplankton production during the dry season is likely the primary mechanism that elevates OC contents and dilutes λ_8 values during the dry season.

[46] To further assess the influence of phytoplankton, N:C ratios and the isotopic signature (δ^{13}C) of FPOM were also measured. Unfortunately, δ^{13}C was of limited utility due to the overlap between phytoplankton δ^{13}C values with that of leaves and needles (Figure 5). An increase in N:C ratios was observed throughout the low-water season (Figures 2c and 5),

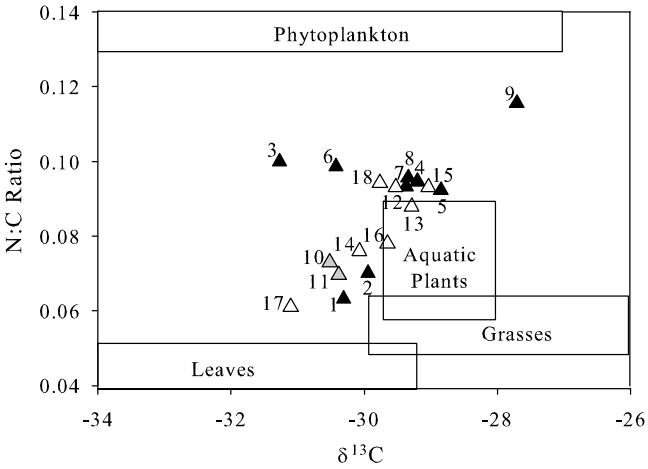

Figure 5. Partitioning of Mekong FPOM among end-member sources using δ^{13}C and N:C ratios. Symbols are described in the caption of Figure 2, and each point is labeled as in Figure 3. Boxes indicate the range of values reported for each end-member. The δ^{13}C and N:C ratios of vascular plants are from Table 1 in this study. The N:C of phytoplankton was calculated from the C:N ratios reported for phytoplankton from the Amazon Basin (5 to 8) [*Devol and Hedges*, 2001]. The δ^{13}C of phytoplankton was estimated from the δ^{13}C of DIC and the fractionation that occurs between DIC and phytoplankton (−21 to −24‰) [*Tan and Strain*, 1983]. Because DIC ranged from −6‰ to −14‰ from the dry to the rainy season (J. E. Richey, unpublished data, 2006), autochthonous OM should be between −27‰ and −38‰. However, during the dry season, it should be on the enriched side of this range. The δ^{13} of grasses extends to −11.9‰, which is beyond the scale of this figure.

suggesting that FPOM transitions from a vascular-plant source with low N:C ratios toward a phytoplankton source with high values as the dry season progresses. However, there was no observable difference between seasons, due to the variability in N:C ratios during both the dry and rainy seasons. High N:C ratios may also be a result of the sorption of nitrogen-rich dissolved OM onto minerals [*Aufdenkampe et al.*, 2001]. Soils from Laos have N:C ratios that range from 0.07 to 0.13 [*de Rouw et al.*, 2010], which is considerably lower than the vascular plant N:C ratios reported here. Accordingly, the rising/high-water FPOM likely reflects mixing between leaf and soil-derived FPOM. Despite no significant difference between the N:C ratios and the δ^{13}C of POM between seasons, the increase in N:C ratios throughout the low-water period combined with the strong seasonal signals in % OC further suggests that phytoplankton are significantly contributing to the FPOM composition of the dry period.

5. Implications and Conclusions

[47] Riverine POM is intricately tied to understanding long-term C burial in marine sediments and elucidating CO_2 production mechanisms, such as respiration, in rivers. Therefore, understanding the provenance of POM signatures and their variability will improve our understanding of these important aspects of the C cycle. In contrast to the Amazon Basin, where little seasonal variability is observed in POM composition relative to the large compositional differences between the coarse and fine fraction [*Hedges et al.*, 1986], distinct seasonal patterns are observed at both the bulk and molecular-level in this large tropical river draining a high-relief catchment. We speculate that such temporal variability occurs because the Mekong is generally constrained to its channel during much of its journey to the South China Sea, with a short transit time from source to sink. Temporal changes in the composition of this material have implications for POM reactivity and preservation.

[48] First, measurements made at the bulk level suggest that FPOM has more of an autochthonous influence during the low-water season relative to the flood season. In the Amazon basin, high rates of water column respiration support high CO_2 gas evasion fluxes [*Richey et al.*, 2002], and recent work examining the variability in basin-wide respiration rates demonstrates that the highest rates are observed in rivers in which the FPOM was derived from autochthonous origins [*Ellis et al.*, 2012]. Thus, given the lability of phytoplankton [*Sun et al.*, 1997], a high proportion of FPOM carried by the Mekong during the dry season would be expected to be consumed while in transit rather than preserved in coastal sediments.

[49] In contrast, during the rainy season, the autochthonous signal carried by Mekong FSS is quickly diminished and the proportion of vascular-plant-derived FPOM increases. However, this does not indicate that this material is of limited bioavailability. Consistent increases in organic C concentrations and dissolved CO_2 with discharge, and simultaneous declines in dissolved oxygen have been observed across a continuum of river sizes in the Amazon Basin [*Richey et al.*, 2009]. These data are consistent with terrestrially derived material supporting water column respiration during the rainy season. Further support for the reactivity of plant-derived FPOM comes from the observation that downstream changes in the δ^{13}C of FPOC carried by the Solimões and Amazon rivers mirrors the expected high-altitude enrichment of 1‰ for every 1,000 m change in elevation for terrestrial plant biomass [*Hedges et al.*, 2000]. This occurs even though the majority of the sediment originates in the Andes. Finally, lignin in soils is highly reactive [*Kiem and Kogel-Knabner*, 2003]. Taken together, a strong case can be made in support of the idea that material derived from higher-plants is bioavailable. Accordingly, it appears likely that although the composition of the material exported by the Mekong differs between seasons, there are labile pools of carbon available to support riverine metabolism throughout the hydrograph.

[50] Gymnosperms contribute to a higher proportion of vascular-plant material during the dry-season, whereas the FPOM transported during the rainy season predominantly reflects angiosperm leaf and bark material. We hypothesize that this variability reflects changes in the proportion of flow coming from the Upper and Lower regions of the Mekong, and speculate that differences in the vascular plant composition of these regions explains the compositional changes observed in riverine FPOM downstream. Thus, we have identified compositional parameters that may be linked to

sediment coming from different regions of the basin, which could potentially be used to evaluate the impact of reservoirs on sediment mobilization within the basin.

[51] We estimate that the total annual flux of FSS, FPOC, and lignin exported by the Mekong River was 76.3 MT/yr, 1.67 MT/yr, and 0.019 MT/yr during the 2006 hydrologic year. This material was then delivered to the Mekong Delta and then on to the South China Sea. An important question is, what are the implications of current and future dams on the composition and magnitude of these fluxes? Three dams are currently in operation on the Mekong main stem in the Upper Basin, and 200 more are projected to be completed within the basin over the next several decades [*Xue et al.*, 2011]. As reservoirs trap material that would have otherwise had another fate, the impacts of these dams will likely have a profound impact on both the quantity and quality of C leaving the catchment via fluvial export. More terrestrially derived C will likely be preserved within the basin rather than being carried downstream and respired, and more autochthonous C will likely be produced due to sediment settling in reservoirs, increasing light penetration [*Downing et al.*, 2008]. These impacts will likely affect Asia's largest freshwater fishery, the Tonle Sap Lake, considering that Mekong sediments are hypothesized to be important in sustaining it [*Mekong River Commission*, 2005; *Kummu et al.*, 2008]. This study provides the beginning of the composition and flux data necessary to assess the impacts of future anthropogenic change within the basin.

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