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# Organic matter compositions and loadings in soils and sediments along the Fly River, Papua New Guinea

Miguel A. Goñi [1†], Eric Moore [2*], Andrew Kurtz [2§], Evan Portier [3], Yvan Alleau [1**], David Merrell [1‡]

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[1] College of Earth, Ocean & Atmospheric Sciences, Oregon State University, Corvallis, OR 97331-5504, USA

[2] Department of Earth and Environment, Boston University, 685 Commonwealth Ave., Boston, MA 02215, USA

[3] Ecosystem Sciences, Department of Environmental Science, Policy and Management, University of California Berkeley, California 94720

***Contact Information:***

[†] Corresponding Author: Tel. (541) 737-0578; Fax (541) 737-2064; Email: mgoni@coas.oregonstate.edu

* Present Address: Department of Natural Resources, Southcentral Region Office, 550 West 7th Avenue, Suite 900C, Anchorage, AK 99501, USA. Tel. (907) 269-8548; Email: eric.moore@alaska.gov

[§] Tel. (617) 358-2570; Fax (617) 353-3290; Email: kurtz@bu.edu

[3] Email: efportier@gmail.com

[**] Email: yalleau@coas.oregonstate.edu

[‡] Present Address: Civil and Environmental Engineering, Brigham Young University, Provo, UT 84602, USA. Email: merrell1978@yahoo.com

## ABSTRACT

The compositions and loadings of organic matter in soils and sediments from a diverse range of environments along the Fly River system were determined to investigate carbon transport and sequestration in this region. Soil horizons from highland sites representative of upland sources have organic carbon contents (%OC) that range from 0.3 to 25 wt%, carbon:nitrogen ratios(OC/N) that range from 7 to 25 mol/mol, highly negative stable carbon isotopic compositions ($\delta^{13}C_{org}$ < -26 ‰) and variable concentrations of lignin phenols (1 < LP < 5 mg/100 mg OC). These compositions reflect inputs from local vegetation, with contributions from bedrock carbon in the deeper mineral horizons. Soils developed on the levees of active floodplains receive inputs of allochthonous materials by overbank deposition as well as autochthonous inputs from local vegetation. In the forested upper floodplain reaches, %OC contents are lower than upland soils (0.8 to 1.5 wt%) as are OC/N ratios (9 to 15 mol/mol) while $\delta^{13}C_{org}$ (-25 to -28 ‰) and LP (2 to 6 mg/100 mg OC) values are comparable to upland soils. These results indicate that organic matter present in these active floodplain soils reflect local (primarily $C_3$) vegetation inputs mixed with allocthonous organic matter derived from eroded bedrock. In the lower reaches of the floodplain, which are dominated by swamp grass vegetation, isotopic compositions were less negative ($\delta^{13}C_{org}$ > -25 ‰) and non-woody vegetation biomarkers (cinnamyl phenols and cutin acids) more abundant relative to upper floodplain sites. Soils developed on relict Pleistocene floodplain terraces, which are typically not flooded and receive little sediment from the river, were characterized by low %OC contents (< 0.6 wt%), low OC/N ratios (< 9 mol/mol), more positive $\delta^{13}C_{org}$ signatures (> -21 ‰) and low LP concentrations (~ 3 mg/100 mg OC). These relict floodplain soils contain modern carbon that reflects primarily local ($C_3$ or $C_4$) vegetation sources. Total suspended solids collected along

the river varied widely in overall concentrations (1 > TSS > 9,000 mg/L), %OC contents (0.1 to 60 wt%), OC/N ratios (7 to 17 mol/mol) and $\delta^{13}C_{org}$ signatures (-26 to -32 ‰). These compositions reflect a mixture of $C_3$ vascular plants and freshwater algae. However, little of this algal production appears to be preserved in floodplain soils. A comparison of organic carbon loadings of active floodplain soils (0.2 and 0.5 mg $C/m^2$) with previous studies of actively depositing sediments in the adjacent delta-clinoform system (0.4 to 0.7 mg $C/m^2$) indicates that Fly River floodplain sediments are less effective at sequestering organic carbon than deltaic sediments. Furthermore, relict Pleistocene floodplain sites with low or negligible modern sediment accumulation rates display significantly lower loadings (0.1 to 0.2 mg $C/m^2$). This deficit in organic carbon likely reflects mineralization of sedimentary organic carbon during long term oxidative weathering, further reducing floodplain carbon storage.

## 1. Introduction

The erosion, transport and storage of sediment along the land-ocean continuum fundamentally affect the production, cycling and burial of organic matter at a global scale (e.g., Aufdenkampe et al., 2011; Blair and Aller, 2012; Hilton et al., 2012). Fluvial systems transport materials from upland portions of watersheds to lowland regions (floodplains and deltas) and marine depocenters (continental shelves and slopes), where further alteration and long-term burial can occur. Floodplains in particular are highly dynamic systems (e.g., Tockner et al., 2010; Moreira-Turcq et al., 2013) that can act as both sources and sinks of carbon (e.g., Zehetner et al., 2009), but also as sites of active organic matter transformation (e.g., Hoffmann et al., 2009; Zocatelli et al., 2013). Allochthonous organic matter deposited during high water conditions and autochthonous production within floodplains both can contribute to the net sequestration of carbon in these systems (e.g., Cabezas and Comin, 2010; Hoffmann et al., 2013). Biologically mediated mineralization can result in the efficient recycling of carbon in floodplain soils (e.g., Kaye et al., 2003; Valett et al., 2005) whereas physical processes such as floodplain drainage, channel meandering and bank erosion can export nutrients and organic matter further down river (e.g., Aspetsberger et al., 2002; Samaritani et al., 2011).

The overarching objective of this study is to gain additional insight into the role of floodplains in the cycling of sediment and major elements such as carbon and nitrogen. Our focus is the Fly River, a large tropical fluvial system selected as one of the sites in the "Source to Sink" (S2S) initiative of the MARGINS program. The goal of the S2S program was to investigate material fluxes from source regions to ultimate sinks in order to better understand the fidelity of the stratigraphic record. In the context of this initiative, we determined the compositions and loadings of organic matter in soils from different regions of the Fly River floodplain to investigate how variable rates of channel migration and over-bank deposition affect the distributions of organic carbon and nitrogen.

With this objective in mind, we collected soils from highland locations with contrasting bedrock and weathering profiles and used those data to provide information on organic matter compositions from upland sources. Suspended particles collected from the main stem and major tributaries of the Fly River were also analyzed to provide information on the composition of organic matter transported along the system. To understand the transformations and sequestration potential along different depositional settings of the river floodplain, we collected soil profiles along the proximal levees of different reaches of the Fly River system. These includes floodplain sites from the upper and upper-middle Fly, which are characterized by high rates of lateral migration and overbank deposition and are covered by dense riparian forests. We also collected and analyzed soil profiles from proximal levees along the lower reaches of the Fly and Strickland Rivers, which displayed lower rates of migration and deposition and are characterized by swamp-grass vegetation. As a contrast to these active floodplain soils, we collected and analyzed soil profiles from Plesitocene-era terraces that represent relict floodplains. Because of their higher elevation compared to the active (Holocene-era) floodplain, these relict floodplain sites are rarely flooded and receive little new sediment input. Thus, they provide us with the opportunity to investigate long-term storage of organic matter under subaerial conditions in the absence of active sedimentation. Finally, we compared these data to compositions of sediments from the subaqueous delta and associated clinoform (i.e., the marine sink) to evaluate carbon sequestration across the land-ocean continuum.

## 2. Materials and Methods

### *2.1. Study area*

The Fly River system, which is located in the southern portion of the island of New Guinea along the eastern region of Papua New Guinea (Figure 1), is composed of three major tributaries (Ok Tedi, Fly and Strickland). Combined, these three rivers drain a total area of 75,000 km$^2$ from the Southern Highlands to the Gulf of Papua. Tectonically-active, steep mountainous terrain (peak elevation of 4,000 m) underlain by siliciclastic and carbonate sedimentary rocks and extremely high rainfall rates (> 10 m per year) characterize the upland regions, which make up ~30% of the watershed. These characteristics lead to elevated rates of erosion, high runoff and large sediment yields with marked contrasts between the Strickland (70-80 x $10^6$ tonnes/y) and Ok Tedi/Fly river systems (7-10 x $10^6$ tonnes/y; Pickup et al., 1981). Natural sediment loads have been increased by anthropogenic discharges at both the Ok Tedi mine and at the Porgera mine located at the headwaters of the Ok Tedi and Strickland rivers, respectively (Day et al., 2008 and references therein).

The steep mountainous terrain gives way to the piedmont alluvial plain, a broad lowland region of gentle topography (e.g. Fig. 1 insert), lower precipitation (~ 2 m/y) and lush vegetation. The piedmont plain, which formed deposition of sediment eroded from the highlands during the upper Pliocene and Pleistocene, was incised during periods of low sea level resulting the Ok Tedi, Fly and Strickland river valleys (Blake and Ollier, 1971; Pickup and Marshall, 2009). Following the most recent low sea-level stand, these incised valleys have been filling with fluvial sediment over the Holocene constructing the present-day floodplains (e.g., Dietrich et al., 1999; Lauer et al., 2008). Throughout the lowland regions, the Ok Tedi/Fly and Strickland rivers display meandering channels with active floodplains and well-developed levees connected to off-channel water bodies, including scroll complexes and blocked-valley lakes. The floodplains along the Ok Tedi and Upper Fly and upper portion (between Binge and Agu rivers) of the

Middle Fly below D'Albertis Junction (Fig. 1) are characterized by dense forests. Forests transition into predominantly swamp grasses in the floodplains below the Agu River and past the confluence with the Strickland River.

Our study is focused on the middle portion of the Fly River system (Fig. 1), which includes a well developed floodplain that widens from 4 km at Kiunga to 14 km at Everill Junction. Floodplain migration rates are highest along the Upper Fly and upper portion of the Middle Fly (1-2 m/y) whereas the lower portions of the Middle Fly and Lower Fly display essentially no channel migration within the past 50 years (Dietrich et al., 1999). The central meander belt occasionally cuts into remnants of Pleistocene terraces characterized by highly weathered soils developed on relict floodplain sediments (e.g., Pickup and Marshall, 2009). Sediment is delivered to a narrow, 1-km corridor of the proximal floodplain by overbank deposition through a network of channels that characterize this stretch of the Fly River (Day et al., 2008). The gradual downstream decrease in sediment accumulation can be seen in post-mine deposition rates measured at near-bank locations, which vary from ~3.6 cm/y in the forest reach, to ~2.4 cm/y in the transition zone, to ~1.6 cm/y in the swamp grass reach (Day et al., 2008).

The Fly River discharges its materials into the northwest Gulf of Papua through its delta, which contains multiple islands with lush vegetation and three major distributary channels (Fig. 1). Fluvial discharge combined with tidal and wave energy transport of Fly River sediment across the delta (e.g., Ogston et al., 2008) and its offshore deposition results in the formation of a subaqueous clinoform that dominates the coastal bathymetry of the region (e.g., Wolanski and Alongi, 1995; Harris et al., 2004; Walsh et al., 2004). The delta-clinoform complex extends up to 50 km offshore and is characterized by distinct sediment environments (e.g., Crockett et al., 2008; 2009; Walsh et al., 2004). Briefly, the inner topset (< 10 m water depth) is a zone of abundant sediment supply and moderate rates of accumulation (~1cm/y) whereas the highly energetic outer topset (water depths of ~10 to 20 m) is a zone of negligible sediment accumulation and is considered a sediment bypass region. The highest sediment accumulation rates (> 3 cm/y) are found in the foreset region of the clinoform (20-50 m water depth), with lower rates at its leading edge (bottom set). Previous studies have investigated the distribution of organic matter in sediments from the Fly River delta-clinoform system (e.g., Goñi et al., 2006; 2008) and provide us with an opportunity to contrasts these compositions to those measured in soils and sediments associated with the Fly River floodplain.

### *2.2. Sampling*

The primary objective of this study was to collect and analyze soil samples from a range of locations along the Fly River system (Fig. 1). Because of the remoteness and inaccessibility of this part of Papua New Guinea, we relied on the logistical assistance of the Ok Tedi Mining Ltd's (OTML) Environmental Department. Highland soils were sampled from soil pits dug to bedrock or from cut banks adjacent to tributary streams at sites accessible from roads near Ok Tedi mine and from the road between Tabubil and Kiunga. Floodplain sampling was conducted during an OTML environmental monitoring cruise on January 2007 aboard the R/V Tahua Chief. Soil profiles developed on floodplain levees were sampled from exposed cut banks from Kiunga on the Upper Fly River to just below Obo on the Lower Fly. At these locations, we excavated into the side of the levee to expose fresh surfaces and collected samples from visually or texturally distinct horizons. We also sampled recently-deposited (days or weeks old), unvegetated alluvium from locations along the bank of the Ok Tedi, which were directly affected by mine and dredge operations, and from the floodplain surface of the Lower Fly, which was more representative of natural sedimentation. All floodplain samples were from levee locations adjacent to the river channel as we did not have the time and/or logistical support to access more distal locations of the floodplain (e.g., Swanson et al., 2008; Aalto et al., 2008). Nevertheless, our sampling covered a large geographical area along over 500 km of river and 2,000 m of altitude that expanded forested and swamp grass reaches of the floodplain with contrasting accumulation rates (Fig. 1).

In addition to sampling soils, we also collected water samples to recover suspended sediments from a variety of locations along the Fly River system, including major tributaries and several off-channel water bodies (Fig. 1). Most of the water samples were hand-dipped from the center of the channel and collected into acid-washed bottles. At six locations along the major reaches of the Fly River, we also collected depth-integrated samples at multiple locations across the channel using a USGS-style isokinetic depth-integrated sampler. All water samples collected in this manner were mixed and sub-sampled using a USGS-style ‘churn splitter’. Splits for suspended sediment characterization were filtered onto pre-combusted, pre-weighed 0.45 mm glass fiber filters, oven-dried at 50 $^{o}$C and stored in petri-dishes until analysis.

### *2.3. Soil characterization*

Table 1 summarizes the location, description and mode of sampling for the different soils obtained for this study. Soils were classified by order based on field observations and soil chemistry and for consistency with Bleeker’s (1983) monograph on the soils of Papua New Guinea. Extent of chemical weathering was evaluated based on trends in inorganic carbon content (see below), and supplemented by distributions of major elements and refractory trace elements (Moore, 2011). We sampled soils developing on the major lithologies of the upland source region (Davies et al., 2005), uplifted Cenozoic carbonates (Rendolls in the classification of Bleeker, 1983), Mesozoic siliciclastic sediments (Inceptisols), and on the Tertiary siliciclastic rocks that outcrop in the transitional zone between the highlands and lowland floodplain (Ultisols).

Floodplain soils are distinct from upland soils because they are developed not on bedrock but on alluvial materials ultimately derived from upland sources. We distinguish between those developing on the active floodplain and soils developed on intensely-weathered relict Pleistocene terraces. Active floodplain soils were sampled from forested reaches along the Upper Fly near Kiunga and upper-Middle Fly just below the confluence with the Ok Tedi River near D'Albertis Junction. Active floodplain samples were also collected along grass reaches of the lower-Middle Fly and Lower Strickland above Obo, and the Lower Fly below Everill Junction. Within the active floodplain soils, we used the appearance of elevated copper concentrations (Cu > 400 μg/g) as an chronostratigraphic marker of the onset of elevated sedimentation rates due to mining ca. 1985 (e.g., Day et al., 2009; see Electronic Annex I for details). Intensely weathered relict Pleistocene floodplain soils were sampled from terraces at several locations along the Middle and Lower Fly.

### *2.4. Analyses*

Soil samples were oven-dried, passed through a 2-mm sieve to remove large debris, and homogenized. We used a mortar and pestle to grind soil samples for organic chemical analyses while unground samples were used for mineral surface area analyses. In the case of suspended sediments, the diameter of the sample filter area was measured with a caliper and an exact proportion of that area sub-sampled for elemental (carbon and nitrogen) and stable isotopic ($\delta^{13}C$) analysis using a hole-puncher. Replicate (2 to 4) cuttings from each filter were used to obtain representative sub-samples for analyses. The area ratio of total vs. sub-sampled filter was used to calculate concentrations of measured constituents (e.g., Moskalski et al., 2013).

Bulk and biomarker organic analyses were performed using established techniques that have been applied previously to soil and sediment samples from the region (e.g., Alin et al., 2008; Goñi et al., 2008). Analytical details for each technique are provided in the cited references. Briefly, weight percent total carbon (%TC), organic carbon (%OC) and nitrogen (%N) contents of ground soil samples were measured by high-temperature combustion (Goñi et al., 2006). In the case of %OC and %N contents, samples were exposed to concentrated HCl fumes and 10% aqueous HCl prior to analysis to remove inorganic carbon. For filter samples, blank corrections were made for carbon and nitrogen contents of glass fiber matrix. Inorganic carbon (%IC) contents were determined as the difference between %TC and %OC. Replicate analyses of selected soil and suspended sediment samples yielded analytical variability of less than 4% of the measured values. Stable isotopic compositions of organic carbon ($\delta^{13}C_{org}$) in soils and sediment were measured by high temperature combustion followed by isotope ratio mass spectrometry after removal of carbonates by repeated washings with 10% HCl (e.g., Goñi et al., 2006). Analytical reproducibility for this measurement was better than 0.3 ‰. Radiocarbon contents of selected decarbonated soils were performed by accelerator mass spectrometry (Vogel et al., 1987), reported as fraction modern and in $\Delta^{14}C_{org}$ format, and used to calculate radiocarbon ages for the OC in soils according to convention (e.g., Stuiver and Polach, 1977). Mineral surface area was determined using a 5-point BET measurement after removing soil organic matter by combustion (e.g., Goñi et al., 2008). Replicate analyses of selected samples revealed analytical variability of this measurement of less than 5% of measured value.

Organic biomarkers derived from vascular plants were measured in soils using alkaline CuO oxidation coupled with gas chromatography-mass spectrometry (Goñi and Montgomery, 2000). Yields of individual compounds were measured using selective ion monitoring and multilevel weekly calibrations (e.g., Goñi et al., 2008; 2009). In this study we present data for lignin-derived vanillyl phenols (VP), syringyl phenols (SP), and cinnamyl phenols (CP), as well as for cutin-derived hydroxyl acids (CA) of soil samples. CuO analyses of filter samples were not performed because of sample size constraints. The structures, provenance and ultimate sources of these CuO reaction products have been discussed previously (e.g., Goñi and Hedges, 1990; Crow et al., 2009; Goñi et al., 2009). Briefly, vanillyl and syringyl phenols are major structural components of lignins in all angiosperm vascular plants characteristic of Papua New Guinea whereas cinnamyl phenols and cutin acids are yielded primarily by non-woody vascular plant tissues (e.g., grasses, leaves). Replicate analyses demonstrated analytical reproducibility better than 10% for individual compound types.

## 3. **Results**

### *3.1. Surface area, carbon, and nitrogen distributions in soils along Fly River*

Measurements of mineral surface area (SA) and weight percent contents of organic carbon (%OC), nitrogen (%N) and inorganic carbon (%IC) were carried out for all soil samples and the results presented in Electronic Annex II. In Table 2, we show averages for different soil types and regions throughout the Fly River system. Because several highland soil profiles (FRS01, FRS03 and FRS06) had well-developed O-horizons characterized by markedly elevated %OC and %N contents (12-25 wt% and 0.8-1.6 wt%, respectively) we group these O-horizon soils separately from the underlying mineral soil horizons (Table 2). Furthermore we break the active floodplain soils into different groups based on location and the impact of mine-related sediment (see Electronic Annex I): forested reaches of the Upper Fly (no mine impact) and upper Middle Fly (mine-impacted), transitional forest-grass reaches of the Middle Fly (mine-impacted) and swamp-grass reaches from the Lower Fly (no mine impact) and the Strickland River (no mine impact). Also included in Table 2 are the compositions of recently deposited alluvium (mine impacted) and relict floodplain soils (no mine impact).

An evaluation of the data in Electronic Annex II and Table 2 revealed several significant contrasts among highland mineral soils in terms of surface area and inorganic carbon concentrations, but little difference in organic matter content. Highly weathered soils (Ultisols) developed on Tertiary sedimentary rocks at the base of the highlands displayed the highest SA values (89 ± 52 $m^2/g$) and relatively low %IC contents (0.2 ± 0.2 wt%). Consistent with their carbonate bedrock source, highland Rendoll mineral soils were characterized by extremely high %IC contents (5 ± 0.6 wt%) and relatively low SA values (12 ± 5 $m^2/g$). Assuming all the inorganic carbon is in the form of $CaCO_3$, these %IC contents translate to overall calcite contents of 25 to 50 wt%. Mineral soils from highland Inceptisols (siliciclastic bedrock) displayed moderate SA values (19 ± 10 $m^2/g$) and were characterized by very low %IC contents (0.1 ± 0.1 wt%). In contrast to SA and %IC compositions, there was significant overlap in the %OC contents of highland mineral soils (Electronic Annex II) resulting in overall averages (Table 2) that were not significantly different among Rendolls (1.0 ± 0.6 wt%), Inceptisols (0.8 ± 1.2 wt%) and Ultisols (1.8 ± 1.0 wt%). The same applies to %N contents which displayed overlapping ranges among these three categories (Table 2).

Within the floodplain, we measured significant differences between modern alluvium, active floodplain soils, and relict floodplain soils. Samples of recently deposited alluvium were characterized by relatively low SA values (11.9 $m^2/g$) and moderate %IC contents (0.8 wt%). The %OC (0.9 wt%) and %N (0.09 wt%) contents of alluvium samples also were similar to those from upland mineral soils. Active floodplain soils were characterized by moderately elevated SA values (20 to 30 $m^2/g$ ) and %OC and %N contents (~1 wt% and ~0.1 wt%, respectively) that were similar to those from both upland mineral soils and recent alluvium. Although there were no clear trends in these parameters among the different regions (Fig. 3), it is interesting to note that the mine-impacted, surface (top 1 m) horizons in the floodplain from the upper Middle-Fly (FRS19) displayed considerably lower SA values than those in deeper horizons, which were not impacted by mine activities. Inorganic carbon contents of active floodplain soils ranged from 0.2 to 0.4 wt% and were notably lower than those from alluvium samples (Table 2). %IC contents of active floodplain soils from the Upper and Middle Fly (0.34 ± 0.10 wt%) were significantly higher (t-test, $p<0.05$) than those from the Lower Strickland and Lower Fly (0.13 ± 0.02 wt%). Relict floodplain soils were characterized by very low %OC and %N contents (~0.5 wt% and 0.06 wt%), were almost devoid of inorganic carbon (%IC ~ 0.05 wt%) and displayed significantly higher SA (43 ± 16 $m^2/g$) than most active floodplain soils.

It is informative to examine the profiles of SA, %OC, %N and %IC at several sites as these data illustrate the effects of organic matter inputs as well as distinct depositional and weathering histories (Fig. 2). For example, whereas SA values were rather constant as a function of depth at most of the sites (Fig. 2a), there were marked depth-related decreases in %OC contents at both highland and active floodplain sites (Fig. 2b). In the case of FRS02, an upland Inceptisol profile, the stark contrasts in %OC with depth was likely due to the low organic matter content (%OC < 0.25 wt%) of the underlying siliciclastic bedrock. At FRS11, a relict floodplain site, %OC levels reached extremely low values (< 0.1 wt%) at depth. In contrast, the profile at FRS19, an active floodplain site in the upper-Middle Fly, showed near constant values of %OC (and %N) for the top 150 cm of the soil, with even higher values in deeper horizons, including a maximum (%OC ~ 3 wt%) at ~350 cm below the surface (Fig. 2b, 2c).

### 3.2. *Elemental and isotopic ratios of soil organic matter*

The elemental and stable isotopic ratios of bulk organic matter in the soil samples analyzed show distinct compositions among and within sites (Table 2; Electronic Annex II). The averages in Table 2 show that IC/OC ratios were highest (> 4) in samples from mineral horizons in highland Rendolls and in recently deposited alluvium. These trends are consistent with elevated contributions from carbonate bedrock and relatively low degree of weathering. Surface horizons of several sites from active floodplains were characterized by moderately elevated IC/OC ratios (ca. 0.5 to 1.0), with the highest average values measured in soils from the forested regions of the Upper and Middle Fly (Table 2). At the upper-Middle Fly site (FRS19); surface horizons impacted by elevated inputs of mine-derived sediment (Cu > 400 μg/g; Fig. 2) displayed significantly higher IC/OC ratios than those from deeper horizons not impacted by mine sediments. Although this pattern could be related to changes in sediment source, it is more likely to reflect contrasts in weathering history. Low IC/OC ratios (< 0.5) were found in most all deep soil horizons from active floodplain sites whereas deep relict floodplain soil samples were completely devoid of carbonate (IC/OC = 0).

A statistically significant correlation between the %OC and %N contents of soil samples (Figure 3) displayed a positive %N intercept that suggests contributions from inorganic N sources could be important at some locations (e.g., highland soils, relict floodplains). With this caveat in mind, we calculated the molar OC/N ratios for the analyzed soils (Electronic Annex II) and summarized whole-soil compositions in Table 2. Overall, highland soils displayed generally elevated OC/N values (> 15), especially the O-horizons with values as high as 25. The major exceptions were the deeper mineral horizons from siliciclastic-bedrock soils, which were characterized by very low OC/N ratios (3 to 7) and appeared to contain significant amounts of inorganic N (Electronic Annex II). Whereas alluvium samples displayed widely variable OC/N values (17 ± 8), soils from active floodplain sites in the forested regions of the Upper Fly and upper-Middle Fly were characterized by higher OC/N values (> 12) than their counterparts along the lower-Middle Fly, Lower Strickland and Lower Fly (9 to 10). Notably, OC/N ratios from mine-impacted horizons in the upper Middle-Fly site were somewhat lower than those from deeper (non-mine impacted) horizons; however, these differences were not statistically significant (Table 2). Low OC/N averages (8 ± 5) characterized the soils from relict floodplain sites (Table 2), with the very low values (~4) in deeper horizons. In contrast, surface horizons of relict floodplain displayed OC/N values that were not that different from those of active floodplain soils (Electronic Annex II).

The stable isotopic compositions of organic carbon also displayed marked variability (Electronic Annex II), with several clear trends among whole-soil averages (Table 2). For example, within highland soils, the $\delta^{13}C_{org}$ signatures of O-horizons were generally more negative (< -28 ‰) relative to the compositions of underlying mineral soils (> -28 ‰). Compared to highland soils, floodplain soils were characterized by $\delta^{13}C_{org}$ compositions that were enriched in $^{13}C$, with samples from the Lower Strickland and Lower Fly displaying the least negative $\delta^{13}C_{org}$ signatures (> -24 ‰) among active floodplain soils. Relict floodplain soils were characterized by the most positive $\delta^{13}C_{org}$ compositions (-22 ± 3 ‰) of all the samples analyzed in this study (Electronic Annex II).

Depth profiles of these bulk elemental and isotopic ratios revealed variable distributions among sites (Figure 4). Among active floodplain sites, IC/OC values were generally highest near the surface and decreased with depth. However, profiles from the Middle Fly and Lower Strickland displayed subsurface maxima in IC/OC values indicating otherwise (Fig. 4a). Most soil profiles displayed OC/N ratios that were relatively high (> 10) in surface horizons and then decreased to lower values with depth (Fig. 4b). Profiles of $\delta^{13}C_{org}$ compositions of organic matter in active floodplains did not display consistent trends (Fig. 4c). For example, in some cases (FRS10, FRS18) $\delta^{13}C_{org}$ signatures became more negative with depth, whereas in others (FRS16, FRS19) no trend with soil depth was apparent. In the case of the relict floodplain site (FRS11), $\delta^{13}C_{org}$ compositions were highly enriched in $^{13}C$ (-13 to -20 ‰) in the top 100 cm of soil and became more negative (< -23 ‰) in deeper horizons (Fig. 4c). Contrasts in the sources and degradative histories of organic matter (and carbonate minerals) at different sites and in different horizons likely contribute to these observations.

To gain additional insight into the provenance and composition of organic matter in the soils from the Fly River region, we carried out radiocarbon analyses of organic matter in surface horizons from selected sites (Table 3). These analyses revealed that whereas the organic matter in surface horizons from both the highland soil site and three relict floodplain sites analyzed was modern and had $\Delta^{14}C$ values > 0 ‰, the surface horizons from active floodplain sites contained aged organic matter with bulk ages that ranged from 400 to over 3,000 years before present (ybp) and $\Delta^{14}C$ values between -56 to -343 ‰ (Table 3). The sandy alluvium sampled from the bank of the Ok Tedi river in the uplands (FRS04) yielded the most negative $\Delta^{14}C$ compositions (-696 ‰) consistent with an age for its bulk organic matter greater than 9,500 ybp. Notably several of these samples were characterized by high Cu contents (Electronic Annex II), consistent with significant contribution of mine-derived sediment and recent (within last 30 years) deposition.

### *3.3. Biomarker distributions in soils*

In order to further characterize the composition of organic matter in soils from the Fly River basin, we measured the yields of a variety of CuO products, including lignin-derived vanillyl, syringyl, and cinnamyl phenols, as well as cutin-derived acids. The yields of 17 individual reaction products belonging to these different categories in selected soil horizons are tabulated in Electronic Annex III and whole-soil averages shown in Table 4. The concentrations of lignin-derived vanillyl and syringyl phenols, which are major components of lignin in angiosperm woody tissues, were highest (> 2 and > 1.5 mg/100 mg OC) in highland soils and in active floodplain soils from the Upper and Middle Fly regions (Table 4). In contrast, active floodplain soils from the Lower Strickland and the Lower Fly displayed lower vanillyl and syringyl phenols yields (< 1.5 mg/100 mg OC.) as did the surface horizons from relict floodplains and samples from alluvium. The lowest yields of lignin-derived vanillyl and syringyl products were measured in the mineral horizons in highland Rendolls (< 0.5 mg/100 mg OC) and in the deep, pre-mine horizons in the upper-Middle Fly floodplain (< 1.2 mg/100 mg OC). The concentrations of cinnamyl phenols, which are derived from non-woody tissues of vascular plants (e.g. leaves, grasses), were significantly lower (0.1 to 1.0 mg/100 mg OC) than those of vanillyl and syringyl phenols (Table 4). Unlike their woody-derived counterparts, the distribution of cinnamyl phenols lacked the marked contrasts between the forested regions of the study area (highland soils and upper reaches of the floodplain) and the swamp grass regions (lower reaches of the Fly and Strickland floodplains).

Cutin acids, which are also derived from non-woody tissues of vascular plants, displayed distribution among soils that were quite distinct from those exhibited by lignin phenols (Table 4). For example, unlike lignin phenols, the highest concentrations of cutin acids (> 0.5 mg/100 mg OC) were found in surface and subsurface horizons from active floodplain soils in the Upper and Middle Fly, and in the surface horizons from relict floodplain sites (Electronic Annex III; Table 4). Low concentrations of cutin acids (<0.2 mg/100 mg OC) characterized most highland soils (with exception of subsurface weathered soils), alluvium samples and active floodplain soils from Lower Strickland and Lower Fly. Differences in vegetation (forest vs. swamp grass) and preservation efficiencies under distinct depositional regimes likely contribute to the patterns observed among the vascular plant biomarkers in these samples.

The biomarker profiles in four selected floodplain soils illustrate additional differences among the reaches of the active floodplain and highlight variability among horizons from the same location (Figure 5). Floodplain sites from the upper-Middle Fly and Middle Fly displayed elevated yields of both vanillyl and syringyl phenols (> 2 mg/100 mg OC) in the upper portion of the soil profiles. Steep decreases with soil depth in these two biomarker classes (to < 1 mg/100 mg OC) were observed in floodplain sites from the Lower Strickland and Lower Fly (Fig.5 a,b). In the case of FRS19, there was a sharp drop in vanillyl and syringyl yields below 150 cm, which coincides with the penetration depth of mine-derived Cu. Cinnamyl phenol profiles did not display as large of a contrast between sites from the upper and lower regions of the floodplain (Fig. 5c). Cutin acids, displayed marked contrasts between upper-Middle Fly and Middle Fly sites and those from the Lower Strickland and Lower Fly. Unlike vanillyl and syringyl phenols, cutin acids showed significant decreases with depth over the top 100 cm of the profiles in FRS10 and FRS19 (Fig. 8d).

### *3.4. River suspended sediment concentrations and organic compositions*

Water samples were collected along the main stem of the Upper, Middle and Lower Fly River, the Ok Tedi, the Lower Strickland and at several highland and lowland tributaries as well as within several off-channel water bodies (Table 5). Along the Upper and Middle Fly, sampling occurred after a moderate peak in discharge (~1500 $m^3 s^{-1}$ ) on January 5, which decreased steadily to ~200 $m^3 s^{-1}$ by January 17. At Obo (Lower Fly), discharge was considerably higher (due to inputs from Strickland River) and only decreased slightly from ~2200 to ~2000 $m^3 s^{-1}$ during the same period. Therefore, falling hydrographic conditions were the norm during our sampling and discharges from the floodplain into the main channel were observed throughout.

Total suspended sediment concentrations (TSS, mg $L^{-1}$) along the Upper Fly were relatively low (10-50 mg $L^{-1}$) and did not display any clear pattern with distance from the mouth or collection date (Table 5). Higher TSS values (50 to 345 mg $L^{-1}$) were measured in samples from the mine-influenced Ok Tedi River. One sample collected downstream of the Ok Tedi mine's river dredging operation exceeded 9000 mg $L^{-1}$. Two smaller highland tributaries of the Upper Fly also displayed elevated TSS values (67 and 462 mg $L^{-1}$). TSS concentrations along the Middle Fly ranged from 23 to ~389 mg $L^{-1}$, and were consistently higher than the values measured along the Upper Fly. Inputs from the Ok Tedi and other highland tributaries contributed to the generally elevated TSS contents along this stretch of the Fly River. With the exception of one sample (FRW47), lowland tributaries along the Middle Fly displayed low TSS concentrations (3-18 mg $L^{-1}$). In contrast, the samples from the Lower Strickland were characterized by much higher TSS concentrations (>1400 mg $L^{-1}$), which contributed to the elevated TSS contents (177 to 889 mg $L^{-1}$) measured along the Lower Fly below the confluence with the Strickland. The three off-channel water bodies sampled along the floodplain displayed low TSS concentrations, especially those from the Middle and Lower Fly (1 to 3 mg $L^{-1}$).

Comparison of data for the six locations where surface and depth-integrated samples were collected concurrently show that the latter consistently displayed higher TSS concentrations (Table 5), consistent with higher suspended particle concentrations in the deeper portion of the river channel. Compositionally, some of the depth-integrated samples displayed lower particulate organic carbon (%POC) and particulate nitrogen (%PN) contents relative to their surface-only counterparts, although that was not the case for all the samples. The depth integrated samples displayed OC/N ratios that were generally slightly higher than those of their surface counterparts, suggesting elevated contributions from nitrogen-poor detritus. The $\delta^{13}C_{org}$ signatures were comparable for the two types of samples (Table 5). Overall, these data show that although surface samples consistently underestimated suspended sediment loads along this stretch of the Fly River, the elemental and isotopic compositions were for the most part comparable among the two sample types. In this study, we are interested in determining the provenance, transformation and ultimate fate of organic matter in the floodplains, which receive inputs from the river via overbank-flow. In that respect, the surface samples may be more representative of materials ultimately deposited on floodplains during high flow conditions than their depth-integrated counterparts.

The %POC and %PN contents of suspended particles ranged from~ 0.2 to over 60 wt% and from ~0.02 to 7.5 wt%, respectively (Table 5), and were negatively correlated with TSS (Fig. 6a, %PN plot not shown). The highest %POC contents were found in samples from lowland tributaries and off-channel water bodies, whereas samples from the Ok Tedi River were characterized by the lowest %POC values. Molar OC/N ratios of suspended particles from the Fly River system ranged from 6 to 18 and failed to display any statistically significant relationship with TSS concentrations (Fig. 6b). Low OC/N values (< 8 mol:mol) were measured in both high and low TSS samples, whereas samples from the Upper Fly and upper reaches of the Middle Fly (forested reaches) exhibited the highest OC/N ratios.

The $\delta^{13}C_{org}$ signatures of suspended particles ranged from -32 to -26 ‰ (Table 5) and in contrast to OC/N ratios displayed a significant correlation with TSS (Fig. 6c). The most $^{13}C$-depleted compositions were found in low-TSS samples from lowland tributaries, off-channel water bodies and portions of the Middle Fly. These highly negative $\delta^{13}C_{org}$ values were measured in samples with low OC/N ratios and coincided with periods when low turbidity waters from lowland tributaries and off-channel water bodies were draining into the main stem of the Fly. In contrast, the most $^{13}$C-enriched signatures (> -28 ‰) were measured in high-TSS samples from the Ok Tedi, Strickland and Lower Fly, which were characterized by relatively low OC/N ratios. The observed compositional contrasts indicate marked differences in the nature and provenance of organic matter in river suspended particles collected as part of this study.

## 4. Discussion

To our knowledge, the data presented here represent some of the first measurements on the distribution of organic matter in soils from Papua New Guinea. A previous study by Bird and co-workers (1994) investigated the carbon contents and $\delta^{13}C_{org}$ compositions of organic matter in surface soils from the central and southern highlands of Papua New Guinea. However, most of those samples were from O-horizons with organic carbon contents > 10 wt% and do not provide a comparable data set to our upland mineral soils and floodplain profiles. The studies by Alin and co-workers (2008), who analyzed organic matter from selected horizons in proximal and distal locations along the floodplain of the Strickland River, and those by Goñi and co-workers (2006; 2008), who determined organic matter distributions in offshore sediments along the Fly River delta-clinoform system, provide us with the opportunity to evaluate organic matter composition and sequestration across the land-ocean continuum in this region. Additional studies are currently underway to further characterize organic matter distributions in regions Strickland River watershed, including areas of its floodplain and highlands.

### *4.1. Organic matter provenance in soils from the Fly River drainage basin*

The elemental, isotopic and biomarkers compositions of soil organic matter from the Fly River system indicate variable contributions from different sources. We did not conduct a survey of specific organic matter end-members within the study area (e.g., aquatic macrophytes, terrestrial vegetation). However, we have soil/bedrock samples from the highland source region as well as suspended particulate samples from different tributaries, which combined with end member compositions from previous studies help us interpret the trends presented above and assess the provenance of materials from different locations.

*4.1.1. Stable and radiocarbon compositions* - The stable and radiocarbon compositions ($\delta^{13}C_{org}$ and $\Delta^{14}C_{org}$) of organic matter in the surface soil horizons from different environments within the Fly River drainage basin allow us to constrain the ages (and thus provenance) of different pools of carbon in this system. Comparable $\delta^{13}C_{org}$ and $\Delta^{14}C_{org}$ data are available for surface soils from proximal and distal floodplain locations along the Strickland River system (Alin et al., 2008) and from surface (0-2 cm) marine sediments collected from distinct offshore environments within the Fly River delta-clinoform system (Goñi et al., 2008; see Fig. 1 for locations).

A plot of $\delta^{13}C_{org}$ versus $\Delta^{14}C_{org}$ compositions reveal that the organic matter in surface soils from both the highlands and relict floodplains is composed of modern carbon ($f_{mod} > 1$) that reflects local vegetation sources (Fig. 7). We base this latter assertion on the fact that whereas the highland soil sites display $\delta^{13}C_{org}$ signatures consistent with primarily forest $C_3$ vegetation, relict floodplain sites within the swamp grass reach display $^{13}C$-enriched signatures consistent with contribution from $C_4$ grasses (Bird et al., 1994). In contrast, most samples from the upper reaches of active floodplains along the Fly and the Strickland Rivers plot along a mixing line between $C_3$ plants and the compositions expected from bedrock-derived kerogen (Lewan, 1986).

One notable exception is the organic matter from site FRS16, which is located in the floodplain from the Lower Fly and displays a signature considerably enriched in $^{13}C$ that indicates contributions from $C_4$ plants. Given the regional vegetation contrasts (forests vs. swamp grasses) discussed previously and the radiocarbon data, we infer that the surface horizons from active floodplain soils contain primarily modern carbon ($0.7 > f_{mod} > 0.9$) derived from local vegetation sources, which is mixed with variable amounts of older carbon highly depleted in $^{14}C$.

Although we do not have the data and samples to specifically determine the provenance of the 'aged' organic carbon diluting modern organic matter derived from vegetation, we speculate that kerogen derived from eroded bedrock is a major constituent of this carbon pool. Support for this explanation is the observation that the one sample of freshly-deposited, upland sandy alluvium from the Ok Tedi River (FRS04) plots closest to the kerogen end member. This sample also displays relatively low lignin and cutin acid yields (Electronic Annex III), consistent with the lack of contributions from vegetation sources given its recent deposition and lack of soil development. At this point we cannot discount the presence of pre-aged, soil organic matter exported from upland sites. However, the modern radiocarbon ages in the O-horizons analyzed as well as the high weathering rates and efficient mineralization of organic matter in tropical soils (e.g., Zocatelli et al., 2013), suggest the magnitude of this input is small. Notably, floodplain sites with elevated rates of overbank deposition (e.g., FRS19) display soil profiles with considerable amounts of vegetation-derived organic matter at depth (e.g., Fig. 5). Mobilization of these deeper floodplain deposits during channel migration likely represents a source of pre-aged (non-bedrock derived) organic matter to down-river environments, including marine depocenters within the delta and clinoform (e.g., Goñi et al., 2008).

Organic matter samples from the surface (0-2 cm) marine sediments from delta-clinoform locations plot along the same mixing line between $C_3$ modern vegetation and fossil kerogen delineated by the active floodplain samples (Fig. 7), albeit their compositions indicate lower abundances of modern carbon ($0.3 > f_{mod} > 0.7$). One possible explanation for these compositions is the preferential loss of modern vegetation-derived organic carbon prior to deposition in the clinoform (e.g., Ward et al., 2013; Aller et al., 2008). However, given the elevated yields of terrestrial biomarkers (i.e., lignin phenols) in delta-clinoform sediments (Goñi et al., 2006; 2008; see discussion below), we conclude that the input of materials eroded from deeper soil horizons in active floodplains, which contain pre-aged vegetation-derived carbon along with fossil kerogen, is a more likely explanation for the observed differences in radiocarbon ages.

*4.1.2. Organic matter compositional signatures* – Several compositional parameters provide additional insight into the provenance of organic matter in floodplain soils, which can be further constrained by examining organic compositions in highland soils and suspended particles. For example, a plot of $\delta^{13}C_{org}$ versus molar N/OC ratios (Fig. 8) shows suspended particle compositions that vary along a mixing line delineated by $C_3$ vascular plants and freshwater algae end members. Note that in these plots we use N/OC (rather than OC/N) to evaluate trends in the relative contributions from different carbon (as opposed to nitrogen) sources (e.g., Perdue and Kprivnjak, 2007). Low-TSS samples display compositions that indicate a predominant freshwater algal source and are consistent with high levels of productivity in off-channel water bodies during periods of low turbidity (e.g., Moreira-Turcq et al., 2013). However, low N contents and moderately negative $\delta^{13}C_{org}$ signatures in most surface soils suggest predominant contributions from $C_3$ vascular plants and a low preservation of algal production in floodplain soils (e.g., Zocatelli et al., 2013). The main exception to this trend are the surface horizons from relict floodplain sites, which contain organic matter with low N/OC ratios typical of vascular plant sources but more positive $\delta^{13}C_{org}$ compositions. These characteristics suggest $C_4$ vascular plants are important contributors to the organic matter in surface soils at these sites. Several samples from the lower reaches (swamp grass region) of the floodplain (e.g., Middle Fly, Lower Strickland, Lower Fly) also plot along the mixing line between the $C_3$ and $C_4$ plant end members (Fig. 8), albeit their more negative $^{13}C$ signatures are consistent with smaller $C_4$ contributions.

Assuming all carbon in surface horizons of relict floodplains is modern (e.g., Fig. 7), simple 2-end member mixing calculations indicate $C_4$ contributions account for 50 to 100% of the organic carbon in these sites. In active floodplains, this type of estimate is more difficult to make because of inputs from bedrock-derived sources (Fig. 7), for which we do not have well constrained $\delta^{13}C$ or N/OC signatures. However, if we assume the regional bedrock has moderately negative $\delta^{13}C$ compositions (e.g., -24 ‰; Lewan, 1986) and elevated N/OC ratios (>0.15; Electronic Annex II), we can explain our measurements as a mixture of organic matter from $C_3$ vegetation sources (80 to 100 %) and bedrock sources (0 to 20%). Hence, these results suggest that with the exception of one location in the Lower Fly (FRS16), contributions from $C_4$ vegetation represent a small fraction of the organic matter accumulating in most active floodplain soils. Such findings suggest the establishment of riparian forests along active floodplains may be important for the effective sequestration of vegetation-derived carbon in these environments (e.g., Cabezas and Comin, 2010; Zocatelli et al., 2013).

Many samples from deeper soils in both active and relict floodplains exhibit enriched N-contents that can be interpreted as inputs from freshwater algae or microbial biomass, both of which could be important in periodically flooded soils (e.g., Aspertsberger et al., 2002).

However, several samples, especially some of the deeper horizons from relict floodplains, display N/OC ratios (> 0.18 mol/mol) that are much higher than expected from the typical algal and microbial sources. It is likely that sorption of inorganic nitrogen species (e.g., ammonium, nitrate) onto the mineral surfaces of these highly weathered, high-SA soils with presumably high cation-exchange capacity are responsible for these trends (e.g., Kaye et al., 2003; Krull and Skjemstad, 2003). Extensive diagenetic alterations (i.e., degradation of biogenic organic matter, preferential preservation of petrogenic carbon) during the weathering process also could explain the overall low organic matter contents exhibited by many of the deep soil horizons from relict floodplain sites (e.g., Electronic Annex II; FRS 11 in Fig. 2).

The distribution of biomarker-based parameters among soils from the various highland and floodplain settings (Table 4) indicates that the sources and degradative state of vegetation-derived organic matter are comparable throughout most of the sites in this study. Hence, for example, elevated SP/VP ratios (0.6 to 1.1) and moderate CP/VP ratios (0.1 to 0.4) characterize all the soil types, and suggest the sources of lignin in these samples include mixtures of woody and non-woody tissues (leaves, grasses) from angiosperm plants, consistent with regional vegetation. All samples display elevated acid/aldehyde ratios (0.5-0.8) indicative of extensive lignin decay consistent with the humid tropical climate that favors effective organic matter recycling. Some parameters exhibit distinct spatial differences, which suggest there are contrasts in the sources of vegetation-derived carbon to soils. For example, elevated CP/VP ratios (> 0.2) characterize soils from floodplains along the lower reaches of the Fly River that lie within the swamp grass region and suggest higher contributions from non-woody lignin sources (i.e. grasses) in these sites relative to those along forested reaches of the Upper Fly. Elevated CA/VP ratios characterize the surface horizons of soils from relict floodplain sites, which are consistent with contributions from non-woody ($C_4$) grasses at these locations.

Figure 9 compares the compositions of soil organic matter from different regions within the Fly River drainage basin to organic compositions from proximal and distal floodplain sites along the Strickland River (Alin et al., 2008) and from sediments from the Fly River delta-clinoform complex (Goñi et al., 2006; 2008). For this comparison, we computed the organic carbon-weighted average for the entire soil and/or sediment profile at each site and combined data from different sites to calculate environment-specific compositions. Our results show that the compositions of organic matter in active floodplain soils from the Fly River are generally comparable to those of organic matter sequestered in the Strickland floodplain and in delta-clinoform sediments. In the case of the Strickland samples, contrasts in OC/N ratios and LP concentrations between proximal sites with higher sediment accumulation rates and distal sites with lower rates of sediment accumulation (Aalto et al., 2008) are consistent with the contrasts between the Upper/ Middle Fly floodplain soils and their counterparts from the Lower Fly and relict floodplains, which also are characterized by lower sediment accumulation rates. Differences in the extent of decay of lignin-bearing organic matter and inorganic-N sorption discussed above are likely responsible for these trends.

The overall similarities in OC/N and $\delta^{13}C$ signatures between floodplain soils and marine sediments are consistent with previous studies that showed terrigenous organic matter makes up the bulk of the carbon preserved in the topset region of the Fly River delta-clinoform system (e.g., Goñi et al., 2008; Aller et al., 2008). The higher contributions of lignin phenols to the overall organic matter in inner topset sediments (Fig. 9c) suggest a more efficient preservation of these materials under the subaqueous settings of the delta-clinoform complex. Highland soils, which were dominated by contributions from O-horizons, and relict floodplains showed the most negative and most positive $\delta^{13}C_{org}$ values, respectively, of all the sites analyzed in the study. Contributions from fresh, relatively undegraded $C_3$ plant material in highland soils and contributions from $C_4$ vascular plants growing on the Pleistocene terraces explain these distinct compositions. Based on the comparison with marine sediments, we conclude that the organic matter from these sites contributes little to the materials sequestered in the delta-clinoform system.

## 4.2. *Organic matter preservation in soils from the Fly River drainage basin*

The overall content of organic matter in soils and sediments is a function of several factors, including the magnitude and composition of organic inputs, exposure to efficient oxidants such as oxygen, and the availability of mineral surfaces that can stabilize organic materials against decay (e.g., Hedges and Oades, 1997). In soils from the upland and lowland floodplain regions investigated in this study, all three factors likely are at play. For example, as discussed above and as seen in other fluvial systems (e.g., Gomez et al., 2004; Leithold et al., 2006; Hilton et al., 2011; Zocatelli et al., 2013), the entrainment of ancient organic matter associated with carbonate and/or siliciclastic bedrock often results in inputs of highly recalcitrant organic matter that has a different fate than the more labile, organic materials derived from local vegetation. Differences in reactivity of organic matter from different types of vegetation (e.g., forests vs. grasslands, above- vs. below-ground inputs) also may influence the distribution of organic matter in floodplain soils (e.g., Hibbard et al., 2001; Vargas and Allen, 2008: Cabezas et al., 2009). Contrasts in sediment accumulation rates and inundation frequency among different regions of the floodplain likely affect the redox conditions in the soils and hence the net exposure to oxygen (e.g., Valett et al., 2005). Given these considerations, we expect higher overall preservation of organic matter in soils from the upper (forested) reaches of the Fly River floodplain, which have higher sediment accumulation rates (~3.6 cm/y) than the floodplain sites in the swamp-grass reaches (~1.6 cm/y; Day et al., 2008), where we expect lower preservation.

Because mineral surfaces help stabilize organic substances in soils and sediments (e.g., Hedges and Oades, 1997; de Carvalho et al., 2003; Wagai et al., 2008), it is useful to evaluate the relationship between mineral surface area and organic carbon content when comparing samples from contrasting texture (e.g., Mayer 1994; Goñi et al., 2008; Wagai et al., 2009). Figure 10 illustrates the relationship between these two parameters for the mineral soil samples analyzed as part of this study. Overall, the majority of the soils from both highland sites and active floodplain sites display organic carbon loadings that vary between 0.25 and 0.5 mg C/m$^2$ of mineral soil. Most of the samples with carbon loadings above this level are from surface horizons, which typically have higher carbon contents due to inputs of organic detritus from surface litter. Note that we have excluded the O-horizons in this plot as these latter samples contain predominantly organic litter with little mineral-associated materials. As expected, many of the soil samples from floodplain sites with low accumulation rates in the Middle and Lower Fly display low carbon loadings (0.1 to 0.25 mg C/m$^2$), with even lower values (< 0.1 mg C/m$^2$) measured in soils from the deeper horizons of relict floodplains soils.

To better assess the relationship between organic matter loadings and the extent of weathering, we compared organic carbon- and inorganic carbon ratios-surface area ratios (OC/SA and IC/SA, respectively) in soils along the Upper, Mid and Lower Fly (Fig. 11). Our assumption is that while the active floodplains along this region of the Fly River receive comparable loads of carbonate-bearing sediment derived from eroded bedrock sources in the highlands, contrasts in weathering rates associated with differences in accumulation rates result in different IC/SA ratios.  Active floodplain soils exhibit a range of OC/SA values that are statistically correlated with IC/SA ratios (Figure 11), which we interpret as proxy for carbonate preservation and overall extent of chemical weathering (e.g., Bouchez et al., 2012; Luker et al., 2012).  Our results show that soils with the lowest degree of chemical weathering, as characterized by IC/SA ratios > 0.2 mg C/m$^2$, display the highest carbon loadings (OC/SA > 0.5 mg C/m$^2$).  Most of these samples are either recently deposited alluvium or active floodplain sites from the Upper and Middle Fly, where we expect higher sedimentation rates to reduce both the exposure of organic matter to efficient oxidants such as oxygen as well as the dissolution of carbonate minerals by soil $CO_2$ (e.g., Egli et al., 2008; Ferguson et al., 2011).  Most of the other active floodplain samples exhibit low OC/SA and IC/SA ratios consistent with higher degrees of organic matter oxidation and carbonate dissolution (Fig. 11).  The lowest organic carbon loadings are exhibited by the deeper horizons of relict floodplain soils, which because of their age and lack of active sedimentation have undergone the highest degree of chemical weathering and contain little to no inorganic carbon (Table 3).

To directly evaluate how organic matter preservation in different regions of the Fly River watershed compares to preservation in the marine environment, we calculated average OC/SA values for soils and sediment cores collected along the Fly River continuum (Figure 12). Organic carbon loadings from floodplain soils in the Upper and Middle Fly are on average comparable to those measured in sediment cores from the outer topset region of the Fly River's delta-clinoform system but lower than those from inner topset locations.  Accumulation rates in the outer topset are small to negligible (e.g., Crockett et al., 2008) and sediments are episodically exposed to oxygen by physical mixing, facilitating the oxidation of organic matter and resulting in less efficient carbon preservation (e.g., Aller et al., 2008; Goñi et al., 2008)).  In contrast, sites from the inner topset are characterized by moderately high rates of sediment accumulation, which result in lower rates of organic matter degradation. The significantly higher OC/SA ratios ($p < 0.05$) exhibited by these sediments are consistent with a more efficient carbon preservation at these locations than in floodplain soils. Active floodplain sites from the Lower Fly and Lower Strickland display significantly ($p < 0.05$) lower whole-soil OC/SA ratios than delta-clinoform sediments, suggesting that higher mineralization losses of organic matter in floodplain environments coincide with lower rates of sedimentation. Relict Pleistocene terraces with no active sedimentation are characterized by the lowest whole-soil carbon loadings (OC/SA < 0.2 mg C/m$^2$) that reflect the extensive weathering and organic mineralization in these environments.

## 5. Summary and Future Work

Our results show that loss of soil carbon via both carbonate dissolution and organic matter oxidation can be very intense under the tropical conditions that characterize both the upland and floodplain soils of Papua New Guinea. As in other depositional environments, organic matter preservation in floodplain soils appears to be controlled to a first-order by the balance between sediment accumulation and exposure to effective oxidants. Hence, the ability of floodplain soils to stabilize and accumulate measurable amounts of organic matter appears to be dependent on periodic deposition of fresh sediment and regular inundation. Forested reaches of the Fly River floodplain, which are characterized by elevated accumulation rates, appear to sequester significantly more carbon than their swamp grass counterparts with lower accumulation rates. Deeper soils from relict floodplain sites without active sedimentation contain little organic matter and are extensively weathered. The sources of organic matter present in floodplain soils include materials derived from local (autochthonous) vegetation and aged materials derived from allochthonous sources (e.g., bedrock and older soils eroded from uplands). In general, active floodplain soils along the Fly River contain organic matter with comparable composition to that present in sediments from its delta-clinoform complex, albeit the latter appears somewhat enriched in the aged-carbon pool. We speculate this is due to entrainment of organic matter that has undergone temporary storage within the floodplain prior to its export to sea.

Comparisons of organic matter loadings in floodplain soils and in delta-clinoform sediments suggest the subaqueous environments of actively accumulating regions of the inner topset display higher preservation efficiencies than subaerial environments within active floodplains. Because of changes in river level, exposure to effective oxidants is likely much higher in subaerial floodplains than in actively accumulating clinoform sediments, where particle resuspension is less likely and pore-water oxygen is consumed rapidly (e.g., Ogston et al., 2008; Goñi et al., 2008; Aller et al., 2008). Although our measurements are constrained to proximal levees, we speculate that distal floodplains of the Fly River are likely to be sites of even less efficient carbon sequestration because of their lower accumulation rates (Day et al., 2008; Aalto et al., 2008). One exception is likely to be the off-channel water bodies, which remain saturated throughout most of the year and thus may represent sites with low oxygen exposure and more efficient organic matter preservation (e.g., Moreira-Turcq et al., 2004). The sediment and organic matter dynamics of these systems need further investigation.

Regarding the overall role of floodplains as net sources, sinks and/or transformers of carbon, our work on the Fly River system only begins to address this issue. Studies in other systems (e.g., Amazon, Ganges) indicate that active mineral weathering is prevalent in lowland floodplains of tropical rivers, contributing to significant releases of major elements (e.g., Ca, Mg, Na, K) and a net sink of atmospheric $CO_2$. (e.g., Bouchez et al., 2012; Lupker et al., 2012) Our study shows that regions of the Fly River floodplain with low rates of sediment accumulation are regions of efficient organic matter mineralization and that, although top soils appear to stabilize organic materials from local vegetation, their net storage capacity for organic carbon is quite low. Thus, soils developing in subaerial levee deposits of tropical floodplains are unlikely to represent efficient long-term sinks for organic carbon. However, because channel migration leads to the mobilization of materials from active floodplains, organic matter in surface floodplain soils can be transported to offshore depositional centers. Hence, it is likely that geomorphically active floodplains along tropical rivers may act to both facilitate stabilization of organic materials and to transport these constituents to marine depocenters where long-term storage is favored. To further investigate the role of floodplains in the overall carbon cycle of this region of Papua New Guinea, studies of the much more active Strickland River system are underway. We expect the results of these studies will contribute needed insight into the part of the system responsible for most of the sediment transport. Estimation of accumulation and sequestration rates for the whole system need to wait until that study is completed.

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# Figure Captions

**Figure 1.** Map of the study area illustrating the location of all samples discussed in this study. The gray insert shows the location of soil and water sampling sites relative to altitude and distance from river mouth. The sample codes and numbers identify soil types according to Table 1 and include: RE, rendoll; IN, inceptisol; UL, ultisol; A, alluvium; AF, active floodplain soils; RF, relict floodplain soils. The locations of previously analyzed sediment samples from the Fly River delta-clinoform complex are shown as open diamonds. These include samples from the inner topset (IT), outer topset (OT), foreset (FT) and bottom set (BT) regions. Yellow and red stars identify the location of kasten cores from the inner and outer topset, respectively.

**Figure 2.** Profiles of bulk soil properties from selected sites along the Fly River system. a) SA, mineral surface area; b) %OC, weight percent organic carbon content; c) %IC, weight percent inorganic carbon content; d) %N, weight percent nitrogen content. Site numbers and categories are defined in Tables 1 and 2.

**Figure 3.** Plot of weight percent organic carbon vs. weight percent nitrogen contents of analyzed soil and suspended sediment samples. The line and equation shows the statistical correlation between %OC and %N contents for soil samples and illustrates the statistically significant positive N intercept. Sample categories are as in Tables 1, 2 and 5.

**Figure 4**. Profiles of compositional parameters of bulk organic matter from selected soil sites. a) IC/OC, molar inorganic carbon to organic carbon ratios; b) OC/N, molar organic carbon to nitrogen ratios; c) $\delta^{13}C_{org}$, stable isotopic signature of organic carbon. Site numbers and categories as in Tables 1 and 2.

**Figure5.** Profiles of organic carbon-normalized yields of different biomarker groups from selected soil sites. a) VP, vanillyl phenols; b) SP, syringyl phenols; c) CP, cinnamyl phenols; d) CA, cutin acids. Site numbers and categories as in Tables 1 and 2.

**Figure 6.** Relationships between total suspended sediment concentrations and a) particulate organic carbon content (%POC); b) molar organic carbon to nitrogen ratios (OC/N); c) stable isotopic signature of organic carbon ($\delta^{13}C_{org}$) for water samples collected along the Fly River system. Sample categories are as in Table 5.

**Figure 7.** Plot of the stable carbon vs radiocarbon composition of organic matter in surface soils from the Fly River watershed (highland soils, alluvium, floodplains) and in offshore sediments from different locations along Fly River delta-clinoform system. End member compositional ranges are included for $C_3$ and $C_4$ vascular plants (C3 and C4), freshwater algae and microbial sources (FW&MC) and kerogen from sedimentary bedrock (KER) and are from Goñi et al., 2003; Lewan, 1986. Strickland floodplain data are from Alin et al., 2008 and clinoform data from Goñi et al., 2008.

**Figure 8.** Plot of the stable isotopic composition ($\delta^{13}C_{org}$) vs. nitrogen.organic carbon ratios (N/OC) of organic matter in soils and suspended sediments from the Fly River system. Sample categories are as in Tables 1, 2 and 5.

**Figure 9.** Comparison of organic matter compositional parameters, including a) organic carbon-nitrogen ratios (OC/N), b) stable carbon isotopic compositions ($\delta^{13}C_{org}$), and c) lignin phenol concentrations among soil sites analyzed in this study and previous data from Strickland River floodplain soils and delta-clinoform sediments. The data plotted represent organic carbon-weighted averages for whole soil profiles (O-horizons included) calculated from individual sites (this study), which were then grouped according to location and geomorphic characteristics, and whole kasten cores from different regions of the delta-clinoform complex (Fig. 1). The total number of samples in each category is shown below. Whole-soil and sediment estimates are shown for highland soils (HS, n=16), alluvium (A, n=4), Upper and upper-Middle Fly floodplain soils in the forested reach (UF, n=15), lower-Middle Fly floodplain soils along the transition reach (MF, n=10); Lower Fly and Lower Strickland floodplain soils along the swamp-grass reach (LF, n=22); relict floodplain soils from Pleistocene Terraces (RF, n=30) (see Table 1 and text for further description). The Prox. (n=7) and Dist. (n=7) designations differentiate soils from proximal and distal locations within the Strickland River floodplain (data from Alin et al., 2008). IT (n=95) and OT (n=80) refer to cores located in the inner topset and outer topset of the Fly River delta-clinoform system, respectively (data from Goñi et al., 2008).

**Figure 10.** Relationship between mineral surface area and organic carbon content for analyzed soils. Different levels of organic matter loadings (0.1 to 1.0 mg C per $m^2$ of sediment) are indicated in the graph for reference. Sample categories are as in Tables 1, 2.

**Figure 11.** Relationship between inorganic carbon/surface areal ratios (IC/SA) and organic carbon/surface area ratios (OC/SA) for soil samples from active regions of the Fly River floodplain. A linear regression and confidence intervals are included. Sample categories are as in Tables 1, 2.

**Figure 12.** Comparison of organic matter loadings (OC/SA) among mineral soils and sediments from different regions within the Fly River watershed (highland and floodplain soils; this study) and offshore sediments from the Fly River delta-clinoform system (data from Goñi et al., 2008. The data plotted are unweighted averages of all the data from mineral soils (O-horizons excluded) and offshore sediments. Data were grouped into the same site categories as in Figure 9 and the number of samples used to compute average for each category are indicated below: highland soils (HS, n=10), Upper and upper-Middle Fly floodplain soils (UF, n=15); lower-Middle Fly floodplain soils (MF, n=10), Lower Fly and Lower Strickland floodplain soils (LF, n=21), relict floodplain soils (RF, n=29) and inner topset (IT, n=54) and outer topset (OT, n=39) sediments from the Fly River delta-clinoform complex.

# Electronic Annexes

(supplied in attached Word and Excel files)