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Composition and provenance of terrigenous organic matter transported along submarine canyons in the Gulf of Lion (NW Mediterranean Sea)

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Abstract

Previous projects in the Gulf of Lion have investigated the path of terrigenous material in the Rhone deltaic system, the continental shelf and the nearby canyon heads. This study focuses on the slope region of the Gulf of Lion to further describe particulate exchanges with ocean's interior through submarine canyons. Nine sediment traps were deployed from the heads to the mouths of Lacaze-Duthiers and Cap de Creus submarine canyons and on the southern open slope from October 2005 to October 2006. Sediment trap samples were analyzed by CuO oxidation to investigate spatial and temporal variability in the yields and compositional characteristics of terrigenous biomarkers such as lignin-derived phenols and cutin acids.

Sediment trap data show that the Dense Shelf Water Cascading event that took place in the months of winter 2006 (January, February and March) had a profound impact on particle fluxes in both canyons. This event was responsible for the majority of lignin phenol (55.4%) and cutin acid (42.8%) inputs to submarine canyons, with lignin compositions similar to those measured along the mid- and outer- continental shelf, which is consistent with the resuspension and lateral transfer of unconsolidated shelf sediment to the canyons. The highest lithogenic-normalized lignin-derived phenols contents in sediment trap samples were found during late spring and summer at all stations (i.e., $193.46 \mu\text{g VP g}^{-1}$ lithogenic at deep slope station), when river flow, wave energy and total particle fluxes were relatively low. During this period, lignin compositions were characterized by elevated cinnamyl to vanillyl phenol ratios (>3) at almost all stations, high p-coumaric to ferulic acid ratios (>3) and high yields of cutin acids relative to vanillyl phenols (>1), all trends that are consistent with high pollen inputs. Our results suggest marked differences in the sources and transport processes responsible for terrigenous material export along submarine canyons, mainly consisting of fluvial and shelf sediments during winter and atmospheric dust inputs during spring and summer.

28 **1 Introduction**

29 Understanding the delivery, cycling and fate of terrigenous organic matter (terrigenous
30 OM) to the oceans is of the utmost importance to understand the global marine carbon
31 cycle (Hedges et al., 1997; Sarmiento and Sundquist, 1992; Falkowski et al., 2000).
32 Much of the particulate terrigenous OM delivered to the oceans is composed of the
33 degraded remains of continental primary production, including materials that have
34 experienced significant alteration during pedogenesis (e.g., Goñi et al., 1998; Blair et
35 al., 2010; Hatten et al., 2012). As a consequence, a large fraction of the particulate
36 organic matter (OM) delivered by rivers ($0.15 \cdot 10^{15}$ g C yr⁻¹; Hedges et al., 1997) is
37 thought to be relatively refractory and can potentially accumulate in the marine system
38 (Hedges and Keil, 1995).

39 While the fate of terrigenous OM in deltas and shelves has been studied in some detail
40 in different margins (e.g., Goñi et al., 1997; 1998; Aller et al., 1998, 2004; Louchouart
41 et al. 1997, 1999; Leithold et al., 1999; Bianchi et al., 2002), much less is known about
42 its transport further offshore (Tesi et al., 2010). Studies of sediment and particle
43 dynamics in the Gulf of Lion, Northwestern Mediterranean Sea, have investigated the
44 connections between shelf and the deep basin. More specifically, studies have focused
45 on particle transport along the Lacaze-Duthiers and Cap de Creus submarine canyon
46 system where the most active off-shelf export of Gulf of Lion sediments takes place
47 (Palanques et al., 2006). Findings from previous researches have resulted in a number
48 of publications describing the physical controls on particle export and organic carbon
49 sources and degradation (e.g., Sanchez-Vidal et al., 2008; 2009; Pasqual et al., 2010;
50 2011; Palanques et al., 2012). During the sampling period covered by the present study,
51 the forcing affecting the off-shelf export of particles was a Dense Shelf Water
52 Cascading (DSWC) event that took place in the winter first months of 2006. Strong
53 down-canyon currents during the DSWC event efficiently and quickly eroded and
54 transported material seaward inducing significant increases in particle flux.

55 The objectives of this paper are to analyze temporal and spatial changes in the
56 contribution and composition of terrigenous OM exported offshore and to use these
57 biogeochemical data to ascertain both terrigenous OM provenance and transport

58 processes along- and across- slope. With these goals in mind, we characterized the
59 composition of terrigenous OM in samples collected by sediment traps located along the
60 two canyons by measuring a variety of biomarker compounds (i.e., lignin phenols, LP,
61 and cutin acids, CA). These compounds are uniquely synthesized by vascular land
62 plants and have been used extensively to trace the contribution and source of
63 terrigenous OM in diverse environments (e.g., Gough et al., 1993; Opsahl and Benner,
64 1997; Goñi and Thomas, 2000; Goñi et al., 1998, 2009; Keil et al., 1998; Hernes et al.,
65 2002; Gordon and Goñi, 2003). We use these markers in combination with bulk
66 geochemical characteristics of the trap sediment (e.g., lithogenic content) to gain insight
67 into the sources and pathways of terrigenous OM in the Gulf of Lion system.

68

69 **2 Background**

70 Starting in 1983 with the ECOMARGE program (Monaco et al., 1990), several
71 multidisciplinary experiments, mainly under the umbrella of EC-funded projects like
72 EUROMARGE, EUROSTRATAFORM, HEMES and HERMIONE, have focused on
73 studying the cycle of particulate matter in the Gulf of Lion. Knowledge of the
74 biogeochemistry of the region has progressively increased as a result of these
75 investigations, which have also shed new light on the physical drivers of the system
76 (Monaco et al., 1990; Canals et al., 2006; Durrieu de Madron, 2008). Several studies
77 have investigated inputs of terrestrial material to the Gulf of Lion and lead to significant
78 advances in our understanding of terrigenous OM cycling in the area (Gough et al.,
79 1993; Buscail et al., 1995; Bouloubassi et al., 1997; Tesi et al 2007; Kim et al., 2010).

80 Sediments and terrigenous OM entering the Gulf of Lion are mainly delivered by the
81 Rhone River, which accounts for 90% of the total annual freshwater and sediment flux
82 (Bourrin et al., 2006). Sediments in the Rhone River prodelta are exposed to continuous
83 physical reworking that resuspend and export the finest particles towards the adjacent
84 continental margin in a southwestwards direction following an anticlockwise near-
85 bottom flow (Gough et al., 1993; Buscail et al., 1995; Palanques et al., 2006; Roussiez
86 et al., 2006; Tesi et al 2007, 2010). Along this southwestwards pathway, a number of
87 small coastal rivers contribute additional terrigenous OM that accounts for 10-20 % of

88 the total sediment entering the Gulf of Lion. Most continental inputs to prodeltas occur
89 during fall floods resulting from heavy precipitation events typical of the Mediterranean
90 climatic regime and also during spring-early summer floods caused by snowmelt in the
91 headwaters of river basins (Buscail et al., 1995; UNEP, 2003; Cathalot et al., 2010).

92 Sediment trap studies have documented increases in near-bottom sediment transport
93 through the Gulf of Lion westernmost canyons of Lacaze-Duthiers and Cap de Creus,
94 especially when storm-induced downwelling conditions are coupled with DSWC.
95 Dense shelf water cascading typically occurs during several week-long events from late
96 winter to early spring when the intense evaporation and cooling of shelf waters due to
97 the effect of strong and persistent northern winds produce cold dense water that
98 overflows the shelf and cascades down slope. This phenomenon results in near-bottom
99 currents of dense cooled water that efficiently transfer material and energy from the
100 continental shelf and the upper ocean layers to the deep basin (Canals et al., 2009).
101 Under these conditions, resuspended sediments are transported as bottom turbid layers
102 with the coarser fraction abrading canyon floor sediments (Canals et al., 2006,
103 Palanques et al., 2006; Puig et al., 2008). The downcanyon transport of sediments
104 during the cascading typically occurs as a multi-step process (Palanques et al. 2012).
105 Shelf sediments are accumulated at the upper slope by storms and DSWC. Then, deep
106 DSWC pulses resuspend and transport upper canyon and slope these deposits to the
107 lower canyon and slope. As a result of this lateral advective transport from shallow
108 waters, the deep Northwestern Mediterranean Basin likely is a significant sink for
109 terrigenous OM (Gough et al., 1993; Bouloubassi et al., 1997). The aim of the work
110 presented here is to investigate the transport of terrigenous OM between the continental
111 shelf and the deep basin, therefore filling a gap in our current understanding of carbon
112 fluxes and cycling in advection-dominated marine system.

113

114 **3 Methods**

115 Field strategy involved the deployment of instrumented moorings at upper (300 m),
116 middle (1000 m water depth) and lower (1500 m) canyon reaches and at canyon mouths
117 (1900 m) where Lacaze-Duthiers and Cap de Creus canyons converge (Pasqual et al.,

2010). Moorings were also deployed at 1000 and 1900 m along the southern open slope (Fig. 1). The sampling period extended from mid October 2005 to late October 2006. Each mooring was equipped with a PPS3 Technicap sequential sediment trap (12 collecting cups, 0.125 m² opening and 2.5 height/diameter aspect ratio for the cylindrical part) 30 m above bottom and an Aanderaa current meter (RCM9/11) 5 meters above the bottom (mab). Mooring LDC1000 also included an extra trap and current meter pair at 500 mab (LDC1000-500). Sediment traps were programmed to collect samples in individual cups for 15 day periods whereas current meters collected data at 30-minute intervals. The receiving cups of the traps were filled up before deployment with a buffered 5% (v/v) formaldehyde solution in 0.45 µm filtered seawater. Traps worked according to the sampling plan, with two exceptions due to temporary failure of the rotating motors of CCC300 and LDC1000 traps during six and two months periods, respectively. In early January 2006 the receiving cup from CCC1000 trap overflowed, which probably caused the excess material entering into the following cup when the cup-supporting carousel rotated to the next 15 days interval, thus resulting in two consecutive flux measurements that were under- and over-estimated, respectively. Trap hydrodynamic forcing, swimmer intrusions and solubilisation processes may bias the measurement of particle fluxes (Buesseler et al., 2007 and references therein), so that mass fluxes presented in this study should be regarded as semi-quantitative.

3.1 Sample treatment

Following recovery, trap samples were stored in the dark at 2-4 °C until they were processed in the laboratory according to the procedure described by Heussner et al. (1990). Samples were (i) split into aliquots using a high precision peristaltic pump robot to obtain 10-20 mg sub-samples, (ii) filtered through combusted and pre-weighted glass-fiber filters for carbon and nitrogen analysis and 0.45 µm pore size cellulose membranes for total mass determination and biogenic Si analysis, (iii) rinsed with distilled water and (iv) dried at 40°C during 24 h for dry weight determination. The precision of mass estimates, as measured by the coefficient of variation was 4.1%. Aliquots of non-filtered particulate samples were separated by centrifugation, rinsed with distilled water and freeze-dried for alkaline CuO oxidation.

149 **3.2 Elemental analyses**

150 Total carbon, organic carbon (OC), and total nitrogen (TN) contents were measured on
151 the whole glass-fiber filters samples with an elemental analyzer (EA Flash series 1112
152 and NA 2100) according to the procedure described in Nieuwenhuize et al. (1994).
153 Samples for OC analysis were first decarbonated using repeated additions of 100 μ l
154 25% HCl with 60°C drying steps in between until effervescence ceased. OM content
155 was estimated as twice the total OC content. Inorganic carbon was calculated
156 subtracting the OC fraction to the total carbon. Then, carbonate content was calculated
157 assuming that all inorganic carbon is contained within the calcium carbonate (CaCO_3)
158 fraction, thus using the molecular mass ratio 100/12. Uncertainties were lower than
159 0.1% as determined from replicates of the certified estuarine sediment MESS-1.
160 Biogenic Si was analyzed using a two-step extraction with 0.5 M Na_2CO_3 (2.5 hours
161 each) separated after filtration of the leachate. Si and Al contents in both leachates were
162 determined by Inductive Coupled Plasma Atomic Emission Spectroscopy (ICP-AES).
163 A correction of the Si of the first by the Si/Al relation of the second one was applied to
164 obtain opaline Si concentrations (Fabres et al., 2002). Corrected Si concentrations were
165 transformed to opal after multiplying by a factor of 2.4 (Mortlock and Froelich, 1989).
166 The analytical precision of opal measurements was 4.5%. The lithogenic fraction was
167 calculated assuming $\% \text{ lithogenic} = 100 - (\% \text{ OM} + \% \text{ CaCO}_3 + \% \text{ opal})$.

168 **3.3 CuO oxidation**

169 Sediment trap samples were oxidized with alkaline CuO following Goñi and
170 Montgomery (2000). An amount of freeze dried sample containing 2 – 5 mg of OC was
171 oxidized in pressurized Teflon vessels containing 8% NaOH, CuO and $\text{Fe}(\text{NH}_4)\text{SO}_4$ at
172 150 °C for 90 min under an oxygen-free atmosphere. Known amounts of recovery
173 standards (trans-cinnamic acid and ethylvanillin) were added after the oxidation. After
174 separating the hydrolysate from the solids by centrifugation, the alkaline solution was
175 acidified to pH 1 with concentrated HCl and extracted with ethyl acetate. The organic
176 solvent was subsequently evaporated and the samples redissolved in pyridine. The CuO
177 oxidation products were derivatized with BSTFA + 1% TCMS at 60°C for 10 min prior
178 to chromatographic analysis. The yields of lignin and cutin oxidation products were
179 quantified by gas chromatography-mass spectrometry (GC–MS) using Hewlett Packard

6890 GC linked to a 5973 Mass Detector (Goni et al., 2009). Yields of lignin phenols were calculated using five-point calibration curves derived from periodically injected mixtures of commercial standards with varying concentrations ($r^2=0.999$) (Goni et al., 2009). Yields of cutin acids, for which commercial standards are not available, were quantified using the calibration curve of trans-cinnamic acid. We report the different CuO oxidation products as sums of distinct compound categories, including vanillyl phenols (VP; vanillin, acetovanillone, vanillic acid), syringyl phenols (SP; syringaldehyde, acetosyringone, syringic acid), cinnamyl phenols (CP; *p*-coumaric acid, ferulic acid) and CA (16-hydroxyhexadecanoic acid, hexadecanedioic acid, 18-hydroxyoctadecenoic acid, 8,16-dihydroxyhexadecaonic acid, 9,16-dihydroxyhexadecanoic acid, 10,16-dihydroxyhexadecanoic acid, 7-hydroxyhexadecanedioic acid and 8-hydroxyhexadecanedioic acid). Ratios of these compounds are used to characterize terrigenous OM (Hedges and Mann, 1979; Goñi and Hedges, 1990a, 1990b).

194

195 **4 RESULTS**

196 **4.1 Temporal flux variability**

Total mass fluxes, and OC, opal, calcium carbonate and lithogenic contents and fluxes in samples from sediment trap deployments between October 2005 and October 2006 published by Pasqual et al. (2010, 2011) revealed distinct spatial and temporal trends in flux composition and magnitude along Lacaze-Duthiers and Cap de Creus canyons and in the slope (Fig. 2). Three distinct periods characterized by contrasting physical and biological processes were identified. The period from January to March 2006 was characterized by strong temperature anomalies and increased current speeds (Fig. 3A) consistent with the occurrence of a DSWC event and the flux of massive amounts of material (up to $90 \text{ g m}^{-2} \text{ d}^{-1}$ at the middle Cap de Creus canyon). The general mass flux increase during the DSWC event was not detected at Lacaze-Duthiers upper canyon station (LDC300) possibly because upper canyons acted as bypass areas of material during the period of strong downcanyon currents (Pasqual et al., 2010). The cascading waters signal was detected at all stations (from 300 m to 1900 m water depth inside the

canyons and the open slope) (Pasqual et al., 2010) and the material reaching the sediment traps was characterized by lower OM contents diluted by lithogenic materials (Fig. 2). The following period (post-DSWC period) was defined from April to mid-July 2006. After March total mass flux decreased progressively reaching values similar to pre-DSWC in mid-July 2006. This tendency was probably due to the settling of material from intermediate or bottom nepheloid layers and the addition of pelagic settling particles as a consequence of the surface algal bloom that took place in March (Fig. 3B; Pasqual et al., 2010). Finally, the period from mid-July to October 2006 (late post-DSWC period) was characterized by the advection of shelf material along the upper and middle canyon due to an energetic wave regime (Fig. 3C; Pasqual et al., 2010). In contrast, at the deeper canyon and open slope stations, pulses opal and calcium carbonate materials associated to high concentrations of marine OM (based on chloropigments and amino acids content) were observed during this period and interpreted to be the result of biological processes occurring in the upper water column (Pasqual et al., 2010, 2011). The pre-DSWC period from October to December 2005 displayed characteristics similar to the mid-July to October 2006 period. It is in this setting that we investigate the flux magnitude and composition of terrigenous OM using specific biomarkers (lignin and cutin products) as tracers.

4.2 CuO products

4.2.1 Organic-Carbon normalized yields

OC -normalized yields of lignin-derived vanillyl, syringyl and cinnamyl phenols, and cutin-derived hydroxyl fatty acids in sediment trap samples displayed marked spatial and temporal differences (Fig. 4). Lignin-derived vanillyl phenols (VP) ranged from 0.47 to 6.96 mg g OC⁻¹. Maximum values were detected in the middle Cap de Creus canyon (CCC1000) and upper slope (SOS1000) stations, where VP yields reached 6.96 and 3.81 mg g⁻¹ OC, respectively, during early -January. Peaks in VP yields coincided with mass flux peaks during the DSWC event. At the slope stations values remained relatively high (up to 1.95 mg g OC⁻¹) during January, February and March 2006. The yields of syringyl phenols (SP) were comparable to VP, ranging from 0.69 to 4.76 mg g OC⁻¹. As with VP, maximum SP values coincided with DSWC pulses in CCC1000 and

240 SOS1000 stations (Fig. 2A, 3B). In contrast to VP, the concentration of SP yields at
241 1900 m depth stations (CCC1900 and SOS1900) during the DSWC event increased up
242 to 2 mg g OC⁻¹. Lower canyon, slope and canyon mouth stations displayed maximum
243 SP values in June-July. Yields of cinnamyl phenols (CP) ranged from 0.42 to 6.12 mg g
244 OC⁻¹. In contrast to VP and SP, CP maximum concentrations were found at the lower
245 canyon stations. Unlike the other two lignin phenol classes, CP showed a general
246 increase in yields, reaching 6.12 mg g⁻¹ OC at CCC1500, during July 2006, except in
247 upper canyon stations where the maxima occurred in May. At that time, we also
248 observed elevated yields of cutin acids (CA), which reached 1.54 and 1.18 mg g⁻¹ OC at
249 CCC300 and LDC300 respectively. CA values increased in all sediment traps during
250 August-September 2006 (except in station LDC300), reaching values of 1.99 mg g⁻¹ OC
251 (LDC1000), 2.21 mg g⁻¹ OC (SOS1000) and 2.19 mg g⁻¹ OC (LDC1500).

252 Yields of lignin-derived phenols in sediment trap samples were comparable to values
253 measured in surface sediments from the Gulf of Lion mid-shelf mud belt (0.10-0.60 mg
254 100 mg⁻¹ OC, 0.11-0.46 mg 100 mg⁻¹ OC and 0.03-0.12 mg 100 mg⁻¹ OC for VP, SP
255 and CP, respectively) (Tesi et al., 2007) (Table 1). All combined, lignin-derived
256 products ranged from 2.16-13.07 mg g⁻¹ OC, values virtually identical to concentrations
257 found in the upper Cap de Creus canyon during trap deployments made in 2004-05
258 (Tesi et al., 2010). The yields of lignin-derived phenols in the Gulf of Lion are
259 comparable to those found in surface sediments from the Gulf of Cádiz (Sánchez-García
260 et al., 2009), the Gulf of Mexico (Goñi et al., 1998; Gordon and Goñi, 2003) and higher
261 than yields in sediment traps from Cariaco Basin (with mean values of 0.54, 0.35 and
262 0.23 mg g⁻¹ OC for VP, SP and CP), an upwelling region in the Caribbean Sea where
263 most OM sinking through the water column is of autochthonous marine origin (Goñi et
264 al., 2009).

265 4.2.2 Lithogenic normalized yields

266 Variations in the yields of OC-normalized lignin and cutin products reflect changes in
267 the contribution of marine OM and terrigenous OM composition. To investigate
268 changes in terrigenous contributions independently from variations in marine OM, we
269 calculated the yields of these two terrigenous biomarker classes normalized to the mass
270 of lithogenic material. Since both terrigenous biomarkers and lithogenic (siliclastic)

271 contributions reflect inputs from land, such a ratio allows an evaluation of differences in
272 the nature of allochthonous materials collected by the sediment traps that is independent
273 of variations in contributions from marine materials.

274 Lithogenic-normalized (litho-normalized) yields of VP, SP and CP, as well as CA are
275 presented in Figure 4. As observed for OC-normalized yields, concentrations of lignin-
276 derived phenols showed significant temporal and spatial differences. VP ranged from
277 14.98 to 193.46 $\mu\text{g g}^{-1}$ litho, with maximum values detected in June at CCC300,
278 LDC1500 and SOS1900. Litho-normalized VP values were especially high in
279 CCC1000 and SOS1000 during the DSWC period. SP yields were comparable to VP
280 (15.55 – 244.83 $\mu\text{g g}^{-1}$ litho), displaying a similar overall behavior except for the middle
281 slope station where contributions did not increase during the DSWC event.

282 Unlike VP and SP, the litho-normalized yields of CP were markedly high from May to
283 September throughout the different stations along the two canyons. For instance, CP
284 yields were highest during late May at CCC300 and LDC300 (e.g., 243.73 $\mu\text{g g}^{-1}$ litho
285 in LDC300) whereas they peaked in late-June at CCC1000, LDC1000 and SOS1000
286 (e.g., 143.95 $\mu\text{g g}^{-1}$ litho at CCC1000). Deeper stations showed the highest litho-
287 normalized CP yields in July (CCC1500, CCC1900 and SOS1000 stations) and August
288 (SOS1900). CA ranged from 0.21 to 378.3 $\mu\text{g g}^{-1}$ litho. Maximum values coincided with
289 the phytoplankton bloom period in LDC300, CCC300 (e.g., 80.72 $\mu\text{g g}^{-1}$ litho in
290 LDC300) before a generalized increase occurred during the post-bloom period, in
291 August, with values $>100 \mu\text{g g}^{-1}$ litho at SOS1000, SOS1900 and CCC1900. The only
292 station where such a generalized increase was not recorded is LDC300.

293 4.2.3 Biomarker ratios

294 Different vascular plant tissues display contrasting lignin and cutin compositions that
295 have been used to determine the provenance of terrigenous OM in a variety of
296 environments (e.g., Hedges and Mann, 1979; Goñi and Hedges, 1990, Goñi and
297 Eglinton, 1996; Goñi and Thomas, 2000). To further investigate specific changes in the
298 sources and composition of terrigenous OM we calculated a variety of biomarker ratios
299 (e.g., SP/VP, CP/VP, CA/VP) for our sediment trap samples (Figure 5). Angiosperm
300 (flowering) plants synthesize lignin that contains both SP and VP whereas gymnosperm

301 (conifer) plants do not have the ability to synthesize SP. Therefore, the SP/VP ratio can
302 provide information about the vegetation sources of lignin in sediment samples. In
303 contrast to woody tissues, non-woody tissues of vascular plants, such as leaves, needles,
304 grasses, bark and pollen, yield high amounts of CP resulting in elevated CP/VP ratios.
305 CA are synthesized by vascular plants to make up the cuticular covering of soft tissues
306 (e.g., leaves, needles). Thus, CA/VP ratios also provide information regarding woody
307 vs. non-woody terrigenous OM inputs.

308 SP/VP ratios in our samples ranged from 0.40 to 2.7. During the phytoplankton bloom
309 period time-series, SP/VP peaked at almost all stations, in May and July, with SP/VP
310 values > 2. CP/VP ratios ranged from 0.17 to 5.1. As observed for the SP/VP ratio, the
311 CP/VP ratio exhibited two marked peaks also during the bloom period, in May and in
312 July, reaching values >3 at almost all stations. CA/VP ratios ranged from 0.1 to 2.0,
313 with maximum values also detected during the bloom period, in May, in upper canyon
314 stations where ratios reached 1.7 and 0.95 at LDC300 and CCC300, respectively. At
315 deeper stations and SOS (i.e. LCD1000, LDC1500, CCC1000, CCC1500, CCC1900,
316 SOS1000 and SOS1900), maximum values occurred during the post-bloom period, in
317 August, with values up to 2.0 at CCC1900 and SOS1900.

318

319 **5 DISCUSSION**

320 **5.1 Overall implications for terrigenous OM transport of along** 321 **canyons**

322 5.1.1. Compositional characteristics of mobilized terrigenous OM

323 Our results show that lignin and cutin products are detected at all locations along
324 submarine canyons indicating the presence of land-derived OM throughout the region.
325 The correlation matrix in Table 2 summarizes the relationships among the yields of VP,
326 SP and CP, and CA, the total mass flux, and the OC and lithogenic fractions. TMF are
327 positively correlated with the lithogenic fraction content and negatively with the OC
328 content which reflected the dilution of the OM by siliciclastic material during the
329 cascading event (January-March 2006) (Fig. 2). All four terrigenous biomarker classes

(VP, SP, CP and CA) are negatively correlated with lithogenic and positively correlated with OC indicating that even lignin phenols contribution to the OC are low (0.1-0.8% of OC coming from lignin phenols, Pasqual et al., 2011) they may be indicators of OC inputs. Thus, these correlations suggest that during the cascading events when TMF increase several orders of magnitude, the material is not especially enriched in SP, CP and CA due to the dilution in the lithogenic matrix. VP in contrast do not show significant correlation with TMF may be due to the fact that VP concentration during the DSWC in some stations as middle Cap de Creus and Open Slope stations increases (Fig. 5A) indicating a different behavior of these lignin derived phenols.

In terms of provenance indicators, lignin-derived CP/VP and SP/VP ratios range from 0.39-2.8 for SP/VP and 0.16-5.1 higher than those previously reported from the Gulf of Lion continental shelf by Tesi et al., (2007) when values detected were SP/VP <1.2 and CP/VP <0.5. Increases in CP/VP and SP/VP ratios with increasing depth have also been measured in surface sediments from a variety of margins, including the Northwest Mediterranean and the Northeast Atlantic (Gough et al., 1998)), the Galician margin (Schmidt et al., 2010), the Gulf of Mexico (Goñi et al., 1998), and the Washington coast (Keil et al., 1998). Such spatial changes in biomarker ratios can be caused by either biological or physical alteration of lignin composition during offshore transport. Notably, microbial attack of lignin-containing OM has been shown to decrease rather than increase the SP/VP and CP/VP ratios of the degraded residue (e.g., Hedges et al., 1988; Hedges and Weliky, 1989; Benner et al., 1990; Opsahl and Benner, 1995), thus suggesting that this process is not likely responsible for the observed trends. Instead, the elevated SP/VP and CP/VP ratios in deeper sediment traps are likely caused by hydrodynamic sorting involving the preferential transport of fine, less dense particles enriched in CP and SP relative to VP. Previous studies have shown that fine particles with significant contributions from soil OM have lignin signatures with elevated SP/VP and CP/VP ratios relatively to coarser particles enriched in vascular plant detritus (e.g., Goñi et al., 1997, 1998, 2009; Bianchi et al 2002; Houel et al., 2006). These compositional contrasts among lignin moieties from different size/density classes reflect alteration processes (e.g., degradation, leaching, sorption onto mineral surfaces) of this macromolecule during pedogenesis (see review by Thevenot et al., 2010 and references therein).

362 The diagenetic state of lignin in natural samples can be estimated from the acid to
363 aldehyde ratios of vanillyl (Vd/Vl) and syringyl (Sd/SI) CuO oxidation products (e.g.,
364 Hedges et al., 1988; Goñi et al., 1993; Louchouart et al. 1999; Houel et al., 2006;
365 Sanchez-Garcia et al., 2009). The oxidative alteration of lignin sidechains by
366 microorganisms such as white-rot fungi has been shown to result in elevated acid to
367 aldehyde ratios (> 0.4) in degraded wood samples (Hedges et al., 1988; Goñi et al.,
368 1993; Opsahl and Benner, 1995). Furthermore, high acid to aldehyde ratios have been
369 measured in oxidatively altered geochemical samples, such as those from humic
370 horizons of mineral soils (Ertel and Hedges, 1984). Besides microbial degradation,
371 selective leaching and sorption of lignin molecules on to clay surfaces can result high
372 Ad/Al ratios (Hernes et al., 2007). Thus, higher Ad/Al ratios can be regarded as general
373 indicators of overall lignin alteration. Selective sorting of fine soil particles during
374 transport can also modify Ad/Al ratios of sedimentary particles (Houel et al. 2006;
375 Louchouart et al. 1999; Sanchez-Garcia et al. 2009), as was the case for SP/VP and
376 CP/VP ratios (see above). In our samples, Ad/Al ratios, which ranged from 0.4 to 0.8
377 (Fig. 7B), showed spatial and temporal variability indicative of differences in the
378 contributions of altered lignin. Overall, our results suggest that terrigenous OM
379 deposited along the studied submarine canyons, containing partly degraded lignin from
380 non-woody sources, originates from highly altered soil OM closely associated to clay
381 particles (e.g., Gough et al., 1993; Goñi et al., 1998; Keil et al., 1998; Gordon and Goñi
382 2004).

383 In addition to the spatial trends in lignin phenol signatures that can be ascribed to
384 hydrodynamic sorting, temporal variability in several compositional parameters suggest
385 a change in the source and nature of the terrigenous OM entrained into the canyon
386 systems during specific periods. As we discuss below, several of the lignin/cutin ratios
387 (e.g., pCd/Fd, CA/VP) show marked peaks during summer and later fall that are
388 consistent with significant contributions from pollen, which is characterized by uniquely
389 elevated values for such ratios. We speculate that these characteristic compositions
390 reflect the importance aeolian dust inputs to the area, which can dominate the
391 terrigenous OM fluxes during periods of low river discharge and/or lateral inputs from
392 the shelf (see section 5.3).

393 5.1.2. Trends within the regional (Gulf of Lion) context

394 Results from the Lacaze-Duthiers and Cap de Creus canyon system in combination with
395 results from previous studies on the Rhone River (Gough et al., 1993), its prodelta and
396 mid-shelf mud belt in the Gulf of Lion (Tesi et al., 2007), and the Cap de Creus canyon
397 head (Tesi et al., 2010) provide an opportunity to track the path of land-derived material
398 in the Gulf of Lion from its main source the Rhone River, down to the outer deep
399 continental margin (Table 1). All four studies have detected southwestward decrease in
400 LP concentration from river mouths along the Gulf of Lion sediment dispersal system
401 (Gough et al. 1993; Tesi et al., 2007). The physical reworking of the prodelta
402 superficial sediments causes a sorting of land-derived materials. The finest fraction
403 associated with soil-derived OM is selectively transported along the Gulf of Lion
404 dispersal system (Buscail et al., 1995; Tesi et al., 2007; Salvadó et al., this volume). By
405 the time the soil-derived OM reaches the southwestern end of the Gulf of Lion, LP
406 concentrations are 1/3 lower than those measured at Rhône River mouth and prodelta.
407 As discussed above, the particle sorting processes, combined with degradation of the
408 land-derived compounds and dilution with other sources of organic matter (e.g., marine
409 OM) during the repeated stages of settling and resuspension that characterize sediment
410 transport along and across this margin helps to explain these trends.

411 Lignin concentrations in stations from the upper canyons and lower slope are
412 comparable to the ones in surface sediments of the Gulf of Lion mid-shelf mud belt
413 (Table 1). This finding is not surprising given that most of the sediment and associated
414 OM transported along canyons primarily during DSWC periods originate from the mid-
415 shelf. Therefore, shelf-derived materials funneled to the lower slope along submarine
416 canyons in the Gulf of Lion undergo little dilution or alteration compared to the changes
417 observed between river mouths and the continental shelf. The available data also
418 suggest that the Lacaze-Duthiers and Cap de Creus canyons system and its slope receive
419 sediment inputs that are relatively enriched in lignin compared to other slopes in the
420 Northwestern Mediterranean Sea and the Algero-Balearic deep basin (Gough et al.,
421 1993; Table 1). Moreover, the fact that deep basin samples have the lowest lignin-
422 phenols yields suggest that terrigenous OM is being diluted or degraded by the time it
423 reaches such deep environments. The relatively high values of lignin concentrations

424 found in this study can also be viewed as an indication of the transport efficiency of the
425 Gulf of Lion dispersal system, which is able to mobilize land-derived compounds from
426 the Rhone River mouth to submarine canyons located 150 km away.

427 The advection of land-derived OM into Lacaze-Duthiers and Cap de Creus canyons
428 appears to occur continuously throughout the study period, even though the quantities
429 and compositions change with physical forcings. Similar findings were observed at the
430 Cap de Creus canyon head during a sediment trap deployment from October 2004 to
431 March 2005 (Tesi et al., 2010), and also in Bari Canyon (Tesi et al., 2008) and the
432 Cariaco Basin (Goñi et al., 1998). Inputs of terrigenous materials into all those settings
433 suggest persistent and steady supply of highly altered terrigenous OM to the sea floor
434 under changing physical and geomorphologic regimes.

435 **5.2 Inputs during Dense Shelf Water Cascading (DSWC) event**

436 The DSWC event was the major physical driver of particle fluxes in the studied area
437 representing up to 83.5 % of the total mass settled at CCC1000 station during the
438 monitored period (Pasqual et al., 2010). Major amounts of lignin and cutin products are
439 detected also during the DSWC period (up to 362.61 and 102.49 mg LP m⁻² and up to
440 20.22 and 6.29 mg CA m⁻² at CCC1000 and SOS1000 respectively) (Table 4). On
441 average the period of deep shelf water cascading, which represents ~30% of the time
442 monitored by our sediment trap study, transported the 55% of the LP and 43% of CA
443 products. In some stations, such as CCC1000, DSWC conditions accounted for 86.3%
444 of LP inputs. These estimates reinforce the importance of continental margin cascading
445 events on the transport of terrigenous materials seawards.

446 The composition of terrigenous biomarkers in samples collected during deep water
447 formation periods displayed significant spatial variability between the two canyon
448 systems, suggesting differences in the nature of materials being entrained by this
449 processes. Samples from middle Lacaze-Duthiers canyon were characterized by
450 terrigenous OM with lower LP yields, elevated CP/VP, SP/VP, Vd/Vl and Sd/SI ratios,
451 relative to samples at middle Cap de Creus canyon (Table 3). As explained before,
452 higher Vd/Vl and Sd/SI ratios suggest elevated levels of lignin alteration and higher
453 CP/VP and SP/VP ratios are related to enhanced contributions from fine particles.

454 Therefore, the inter-canyon contrast observed during the DSWC event are consistent
455 with finer, more degraded terrigenous OM being transported through the Lacaze-
456 Duthiers canyon and a less altered terrigenous OM mobilized through the Cap de Creus
457 canyon (Cowie and Hedges, 1992; Goñi et al., 1998).

458 Overall, the differences in the nature of the terrigenous OM advected through Lacaze-
459 Duthiers canyon relative to that flushed through the Cap de Creus canyon during DSWC
460 conditions suggest different source-areas of particulate materials for the two systems
461 during high energetic events. Such interpretation is consistent with suspended sediment
462 concentration and current measurements at the heads of both canyons (Palanques et al.,
463 2012). The compositional differences in terrigenous OM between the Lacaze-Duthiers
464 and Cap de Creus canyons are comparable to those observed between the terrigenous
465 OM signatures of sediments in the outer shelf and inner shelf (Table 1). Thus, we infer
466 that during DSWC events the Lacaze-Duthiers canyon system receives materials
467 resuspended from seabed on the outer shelf. The terrigenous OM in this region of the
468 shelf is highly altered and the compositions obtained in the sediment trap samples
469 reflect this origin. In contrast, during DSWC conditions, resuspended sediments from
470 the inner shelf containing fresher terrigenous OM are funneled through the Cap de
471 Creus canyon. Such explanation is reasonable if one considers the closeness of the Cap
472 de Creus canyon head to the coastline (4 km), the contrast in current direction near the
473 heads of both canyons during dense water events and the processes that focus coastal
474 turbid waters at the southern wall of the Cap de Creus canyon (Canals et al., 2006;
475 Lastras et al., 2007; Puig et al., 2008; Palanques et al., 2012).

476 **5.3 Atmospheric inputs of terrigenous OM**

477 In addition to lateral inputs from the shelf, our analyses revealed unique compositional
478 characteristics that strongly indicate both canyons receive terrigenous OM from a
479 separate source with distinct forcings – atmospheric inputs. For example, as the April
480 to mid-July 2006 bloom period ended, sediment trap samples exhibited unique lignin
481 and cutin biomarker ratios that are consistent with a distinct provenance. From early
482 May to July we measured unusually high cinnamyl to vanillyl phenol ratios (CP/VP
483 from 2.75 up to 5) at most stations (Fig. 6B). Notably, the composition of the cinnamyl
484 phenols during this period were characterized by very high p-coumaric acid contents,

485 that resulted in highly elevated with p-coumaric:ferulic acid ratios ($pCd/Fd > 3$; Fig.
486 6D). In addition, the peaks in CP/VP generally coincided with elevated cutin acid to
487 vanillyl phenol ratios ($CA/VP > 1.2$; Fig. 6C). Moderately elevated CP/VP and CA/VP
488 ratios (e.g., 0.2 to 1.1, 0.5 to 2.5, respectively) have been utilized to distinguish
489 contributions of non-woody plant tissues, such as gymnosperm needles and angiosperm
490 leaves, which are characterized by moderately elevated ratios (Goñi and Hedges, 1990b,
491 1992; Goñi and Thomas, 2000), from woody tissues that contain only trace amounts of
492 CP and CA. However, the only vascular plant material analyzed to date yielding such
493 high CP/VP and pCd/Fd ratios (> 2.5 and > 1.5 , respectively) upon CuO oxidations are
494 pollen samples from angiosperm and gymnosperm plants (Keil et al., 1998; Hu et al.,
495 1999). Indeed, pollen grains are known to highly resistant to microbial degradation and
496 can contribute significantly to allochthonous OM present in coastal sediments (Keil et
497 al., 1998). We infer from the unusual compositions that pollen inputs to the particle
498 flux along the canyons may be significant during specific time intervals (i.e. late spring
499 to early summer).

500 High CP/VP, CA/VP and pCd/Fd ratios were measured from June to August 2006,
501 when river discharges were minimal, there was no deep water cascading and overall
502 mass fluxes were particularly low (Fig. 2A), all indicating negligible advection of
503 allochthonous materials from land and the continental shelf (Pasqual et al.; 2011).
504 During this time sediment trap samples exhibit moderate increases in LP and CA
505 contents, which in previous publications were attributed to the slow settling of fine
506 particles originally mobilized during the preceding DSWC event (Tesi et al., 2010;
507 Pasqual et al., 2011). It was also hypothesized that the time lag between the cascading
508 event and the deposition of these particles reflected the low settling velocity of the finest
509 sediment fraction mobilized from the shelf. However, the unique terrigenous biomarker
510 signatures (i.e., markedly high CP/VP, CA/VP and pCd/Fd ratios) were detected almost
511 synchronously at all stations (Fig. 6). We infer that these compositions indicate
512 enhanced inputs of terrigenous OM associated with atmospheric sources, including
513 pollen (i.e. high CP/VP, pCd/Fd) and dust, which can be enriched in cuticular (i.e. high
514 CA:VP) compounds (Huang et al., 2000).

515 The importance of atmospheric dust inputs, representing the main source of terrigenous
516 particles to offshore locations and of aerial transport of pollen from land have been
517 highlighted in several studies (Gagosian and Peltzer, 1986; Franzen et al., 1994; Keil et
518 al., 1998, Hu et al., 1999; Izquierdo et al., 2010; Shakya et al., 2011). Measurements of
519 pollen concentrations in the air above the Western Mediterranean indicate the summer
520 and early fall are periods characterized by high pollen input (i.e., Meiffren, 1998; Recio
521 et al., 2006). Estimates of atmospheric particle fluxes to the Western Mediterranean Sea
522 average $27.4 \text{ mg m}^{-2} \text{ d}^{-1}$ (Guerzoni et al., 1997) and these materials have a measurable
523 amounts (1 to 3 wt.%) of organic matter (Tomadin et al., 1984). Given these
524 observations and the unique compositions of the materials recovered in sediment traps
525 from June to August, we conclude that aeolian inputs make up a considerable fraction of
526 the terrigenous OM inputs during this period of otherwise low allochthonous fluxes.
527

528 **6 Conclusions**

529 Our results unveil different sources and transport processes involved in terrigenous OM
530 transport along the submarine canyons and open slope of the Gulf of Lion. The flux and
531 composition of terrigenous biomarkers (i.e., lignin phenols and cutin acids) is strongly
532 affected by winter-early spring DSWC, which is responsible for the majority of their
533 transport even though it only represents one fourth of the period studied. The fresher
534 character of the terrigenous OM advected during DSWC along the Cap de Creus
535 canyon, relative to the Lacaze-Duthiers canyon is consistent with a more direct linkage
536 of the former with the continental shelf during high energetic conditions typical of
537 DSWC events. The temporal resolution of sediment trap sampling allows the
538 characterization of terrigenous OM during low energy conditions (late spring and
539 summer) when biomarker signature strongly point towards pollen and dust associated
540 with atmospheric inputs as major contributors to the overall fluxes of allochthonous
541 materials. Overall, our results show that there is a continuous and steady supply of
542 highly altered terrigenous OM to the continental slope under contrasting hydrographic
543 regimes, with Lacaze-Duthiers and Cap de Creus canyons appearing as the most active
544 and efficient systems funneling terrigenous OM to the deep Northwestern
545 Mediterranean margin and basin.

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References

- Aller, R.C., 1998. Mobile deltaic and continental shelf muds as suboxic, fluidized bed reactors. *Marine Chemistry* 61, 143-155.
- Aller, R.C., Blair, N.E., 2004. Early diagenetic remineralization of sedimentary organic C in the Gulf of Papua deltaic complex (Papua New Guinea): net loss of terrestrial C and diagenetic fractionation of C isotopes. *Geochimica et Cosmochimica Acta* 68, 1815-1825.
- Benner, R., Weliky, K., Hedges, J. I., 1990. Early diagenesis of mangrove leaves in a tropical estuary: Molecular-level analyses of neutral sugars and lignin-derived phenols. *Geochimica et Cosmochimica Acta* 54, 1991-2001.
- Bianchi, T.S., Mitra, S., McKee, B.A., 2002. Sources of terrestrially-derived organic carbon in lower Mississippi River and Louisiana shelf sediments: implications for differential sedimentation and transport at the coastal margin. *Marine Chemistry* 77, 211-223.
- Blair, N.E., Leithold, E.L., Brackley, H., Trustrum, N., Page, M., Childress, L., 2010. Terrestrial sources and export of particulate organic carbon in the Waipaoa sedimentary system: Problems, progress and processes. *Marine Geology* 270(1-4), 108-118.
- Bouloubassi, I., Lipiatou, E., Saliot, A., Tolosa, I., Bayona, J.M., Albaigés, J., 1997. Carbon sources and cycle in the western Mediterranean—the use of molecular markers to determine the origin of organic matter. *Deep-Sea Research Part II* 44, 781-799.
- Bourrin, F., Durrieu de Madron, X., 2006. Contribution of the study of coastal rivers and associated prodeltas to sediment supply in North-western Mediterranean Sea (Gulf of Lions). *Vie Milieu* 56(4), 307-314.
- Buesseler, K.O., Antia Avan, N., Chen, M., Fowler, S.W., Gardner, W.D., Gustafsson, O., Harada, K., Michaels, A.F., Rutgers van der Loeff, M., Sarin, M., Steinberg, D., Trull, T., 2007. An assessment of the use of sediment traps for estimating upper ocean particle fluxes. *Marine Research* 65, 345-416.

- Buscail, R., Pocklington, R., Germain, C., 1995. Seasonal variability of the organic matter in a sedimentary coastal environment: sources degradation and accumulation (continental shelf of the Gulf of Lion - northwestern Mediterranean Sea). *Continental Shelf Research* 15(7), 843-869.
- Canals, M., Puig, P., Durrieu de Madron, X., Heussner, S., Palanques, A., Fabres, J., 2006. Flushing submarine canyons. *Nature* 444, 354-357.
- Canals, M., Danovaro, R., Heussner, S., Lykousis, V., Puig, P., Trincardi, F., Calafat, A.M., Durrieu de Madron, X., Palanques, A., Sánchez-Vidal, A., 2009. Cascades in Mediterranean Submarine Grand Canyons. *Oceanography* 22 (1), 26-43.
- Cathalot, C., Rabouille, C., Pastor, L., Deflandre, B., Viollier, E., Buscail, R., Grémare, A., Treignier, C., Pruski, A., 2010. Temporal variability of carbon recycling in coastal sediments influenced by rivers: assessing the impact of flood inputs in the Rhône River prodelta. *Biogeosciences* 7, 1187-1205.
- Cowie, G.L., Hedges, J.I. 1992. The role of anoxia in organic matter preservation in coastal sediments: relative stabilities of the major biochemicals under oxic and anoxic depositional conditions. *Organic Geochemistry* 19(1-3), 229-234.
- Durrieu de Madron, X., Wiberg, P.L., Puig, P., 2008. Sediment dynamics in the Gulf of Lions: The impact of extreme events. *Continental Shelf Research* 28, 1867-1876.
- Ertel, J.R., Hedges, J.I., 1984. The lignin component of humic substances: Distribution among soil and sedimentary humic, fulvic, and base-insoluble fractions. *Geochimica et Cosmochimica Acta* 48(10), 2065-2074.
- Fabres, J., Calafat, A., Sanchez-Vidal, A., Canals, M. Heussner, S., 2002. Composition and spatio-temporal variability of particle fluxes in the Western Alboran Gyre, Mediterranean Sea. *Journal of Marine Systems* 33-34, 431-456.
- Falkowski, P., Scholes, R.J., Boyle, E., Canadell, J., Canfield, D., Elser, J., Gruber, N., Hibbard, K., Höglberg, P., Linder, S., Mackenzie, F.T., Moore III, B., Pedersen, T., Rosenthal, Y., Seitzinger, S., Smetacek, V., Steffen, W., 2000. The Global Carbon Cycle: A Test of Our Knowledge of Earth as a System. *Science* 290, 291-296.

- Franzen, L.G., Hjelmroos M., Kallberg, P., Brorstrom-Lunden, E., Juntto S., Savolainen A.L., 1994. The 'yellow snow' episode of northern Fennoscandia, March 1991 -a case study of long-distance transport of soil, pollen and stable organic compounds. *Atmospheric Environment* 28(22), 3587-3604.
- Gagosian, R., Peltzer, E., 1986. The importance of atmospheric input of terrestrial organic material to deep sea sediments. *Advances in Organic Geochemistry* 10, 661-669.
- Goñi, M.A., Hedges, J.I., 1990a. Cutin-derived CuO reaction-products from purified cuticles and tree leaves. *Geochimica et Cosmochimica Acta* 54(11), 3065-3072.
- Goñi, M.A., Hedges, J.I., 1990b. Potential applications of cutin-derived CuO reaction products for discriminating vascular plant sources in natural environments. *Geochimica et Cosmochimica Acta* 54, 3073-3081.
- Goñi, M.A., Eglinton, T.I., 1996. Stable carbon isotopic analyses of lignin-derived CuO oxidation products by isotope ratio monitoring-gas chromatography-mass spectrometry (irm-GC-MS). *Organic Geochemistry* 24, 601-615.
- Goñi, M.A., Montgomery, S., 2000. Alkaline CuO oxidation with a microwave digestion system: lignin analyses of geochemical samples. *Analytical Chemistry* 72 (14), 3116-3121.
- Goñi, M.A., Thomas, K.A., 2000. Sources and transformations of organic matter in surface soils and sediments from a tidal estuary (north inlet South Carolina, USA). *Estuaries* 23, 548-564.
- Goñi, M.A., Nelson, B., Blanchette, R.A., Hedges, J.I., 1993. Fungal degradation of wood lignins—geochemical perspectives from CuO-derived phenolic dimers and monomers. *Geochimica et Cosmochimica Acta* 57, 3985-4002.
- Goñi, M.A., Rittenberg, K.C., Eglinton, T.I., 1997. Sources and contribution of terrigenous organic carbon to surface sediments in the gulf of Mexico. *Nature* 389, 275-278.

- Goñi, M.A., Ruttenberg, K.C., Eglinton, T.I., 1998. A reassessment of the sources and importance of land-derived organic matter in surface sediments from the Gulf of Mexico. *Geochimica et Cosmochimica Acta* 62(18), 3055-3075.
- Goni, M.A., Aceves, H., Benitez-Nelson, B., Tappa, E., Thunell, R., Black, D.E., Muller-Karger, F., Astor, Y., Varela, R., 2009. Oceanographic and climatologic controls on the compositions and fluxes of biogenic materials in the water column and sediments of the Cariaco Basin over the Late Holocene. *Deep Sea Research I* 56(4), 614-640.
- Gordon, E.S., Goñi, M.A., 2003. Sources and distribution of terrigenous organic matter delivered by the Atchafalaya River to sediments in the northern Gulf of Mexico. *Geochimica et Cosmochimica Acta* 67(13), 2359-2375.
- Gordon, E.S., Goñi, M.A., 2004. Controls on the distribution and accumulation of terrigenous organic matter in sediments from the Mississippi and Atchafalaya River Margin. *Marine Chemistry* 92, 331-352.
- Gough, M.A., Fauzi, R., Mantoura, C., Preston, M., 1993. Terrestrial plant biopolymers in marine sediments. *Geochimica et Cosmochimica Acta* 57(5), 945-964.
- Guerzoni, S., Molinaroli, E., Chester, R., 1997. Saharan dust inputs to the western Mediterranean Sea: depositional patterns geochemistry and sedimentological implications. *Deep-Sea Research II* 44, 631-654.
- Hatten, J.A., Goñi, M.A., Wheatcroft, R.A., 2012. Chemical characteristics of particulate organic matter from a small mountainous river in the Oregon Coast Range, USA. *Biogeochemistry* 107, 43-66.
- Hedges, J.I., 1992. Global biogeochemical cycles: progress and problems. *Marine Chemistry* 39, 67-93.
- Hedges, J.I., Mann, D.C., 1979. The characterization of plant tissues by their lignin oxidation products. *Geochimica et Cosmochimica Acta* 43(11), 1803-1807.
- Hedges, J.I., Weliky, K., 1989. Diagenesis of conifer needles in a coastal marine environment. *Geochimica et Cosmochimica Acta* 53, 2659-2673.

Hedges, J.I., Keil, R.G., 1995. Sedimentary organic matter preservation: an assessment and speculative synthesis. *Marine Chemistry* 49, 127-136.

Hernes, J.P., Benner, R., 2002. Transport and diagenesis of dissolved and particulate terrigenous organic matter in the North Pacific Ocean. *Deep-Sea Research I* 49, 2119-2132.

Hedges, J.I., Blanchette, R.A., Weliky, K., Devol, A.H., 1988. Effects of fungal degradation on the CuO oxidation products of lignin: A controlled laboratory study. *Geochimica et Cosmochimica Acta* 52, 2717-2726.

Hedges, J.I., Keil, R.G., Benner, R., 1997. What happens to terrestrial organic matter in the ocean?. *Organic Geochemistry* 27, 195-212.

Hernes, P. J., Robinson, A. C., Aufdenkampe, A. K. 2007. Fractionation of lignin during leaching and sorption and implications for organic matter “freshness”, *Geophysical Research Letters* 34, L17401.

Heussner, S., Ratti, C., Carbonne, J., 1990. The PPS 3 time-series sediment trap and the trap sample processing techniques used during the ECOMARGE experiment. *Continental Shelf Research* 10, 943-958.

Houel, S., Louchouart, P., Lucotte, M., Canuel, R., Ghaleb, B., 2006. Translocation of soil organic matter following reservoir impoundment in boreal systems: Implications for in situ productivity. *Limnology and Oceanography* 51(3), 1497-1513.

Hu, F.S., Hedges, J.I., Gordon, E.S., Brubaker, L.B., 1999. Lignin biomarkers and pollen in postglacial sediments of an Alaskan lake. *Geochimica et Cosmochimica Acta* 63(9) 1421-1430.

Huang, Y.S., Dupont, L., Sarthein, M., Hayes, J.M. and Eglinton, G., 2000. Mapping of C(4) plant input from North West Africa into North East Atlantic sediments. *Geochimica et Cosmochimica Acta* 64(20) 3505-3513.

- Ishiwatari, R., Uzaki, M., 1987. Diagenetic changes of lignin compounds in more than 0.6 million-year-old lacustrine sediment (Lake Biwa Japan). *Geochimica et Cosmochimica Acta* 51, 321-328.
- Izquierdo R. Belmonte J. Avila A. Alarcón M. Cuevas E. Alonso-Pérez S., 2011. Source areas and long-range transport of pollen from continental land to Tenerife (Canary Islands). *International Journal of Biometeorology* 55(1), 67-85.
- Keil, R.G., Tsamakis, E., Giddings, J.C., Hedges, J.I., 1998. Biochemical distributions (amino acids, neutral sugars, and lignin phenols) among size-classes of modern marine sediments from the Washington coast. *Geochimica et Cosmochimica Acta* 62, 1347-1364.
- Kim, J., Zarzycka, B., Buscail, R., Peterse, F., Bonnín, J., Ludwig, W., Schouten, S., Sinninghe Damste J.S., 2010. Contribution of river-borne soil organic carbon to the Gulf of Lions (NW Mediterranean). *Limnology and Oceanography* 55, 507-518.
- Lastras, G., Canals, M., Urgeles, R., Amblas, D., Ivanov, M., Droz, L., Dennielou, B., Fabr s, J., Schoolmeester, T., Akhmetzhanov, A., Orange, D., Garc a-Garc a, A., 2007. A walk down the Cap de Creus canyon, northwestern Mediterranean Sea: recent processes inferred from morphology and sediment bedforms. *Marine Geology* 246 (2–4), 176–192.
- Leithold, E.L., Hope, R.S., 1999. Deposition and modification of a flood layer on the northern California shelf: lessons from and about the fate of terrestrial particulate organic carbon. *Marine Geology* 154, 183-195.
- Louchouart, P., Lucotte, M., Canuel, R., Gagn , J.P., Richard, L.F., 1997. Sources and early diagenesis of lignin and bulk organic matter in the sediments of the Lower St. Lawrence Estuary and the Saguenay Fjord. *Marine Chemistry* 58(1-2), 3-26.
- Louchouart, P., Lucotte, M., Farella, N., 1999. Historical and geographical variations of sources and transport of terrigenous organic matter within a large-scale coastal environment. *Organic Geochemistry* 30(7), 675-699.

- Meiffren, I., 1988. Airborne Pollen of Toulouse, Southern France. *Grana* 27(3), 183-201.
- Monaco, A., Courp, T., Heussner, S., Carbonne, J., Fowler, S.W., 1990. Deniaux B. Seasonality and composition of particulate fluxes during ECOMARGE-I, western Gulf of Lions. *Continental Shelf Research* 10, 959-987.
- Mortlock, R. A. and Froelich, P. N., 1989. A simple method for the rapid determination of biogenic opal in pelagic marine sediments, *Deep-Sea Reserach* 36, 1415-1426.
- Nieuwenhuize, J., Maas, Y.E.M., Middelburg, J.J., 1994. Rapid analysis of organic carbon and nitrogen in particulate materials. *Marine Chemistry* 45, 217-224.
- Opsahl, S., Benner, R., 1995. Early diagenesis of vascular plant tissues: Lignin and cutin decomposition and biogeochemical implications. *Geochimica et Cosmochimica Acta* 59, 4889-4904.
- Opsahl, S., Benner, R., 1997. Distribution and cycling of terrigenous dissolved organic matter in the ocean. *Nature* 386, 480-482.
- Palanques, A., Durrieu de Madron, X., Puig, P., Fabres, J., Guillén, J., Calafat, A., Canals, M., Heussner, S., Bonnin, J., 2006. Suspended sediment fluxes and transport processes in the Gulf of Lions submarine canyons. The role of storms and dense water cascading. *Marine Geology* 234, 43-61.
- Palanques, A., Puig, A., X. Durrieu de Madron, X., Sanchez-Vidal, A., Pasqual, C., Martín, J., Calafat, A., Heussner, S., Canals, M. 2012. Sediment transport to the deep canyons and open-slope of the western Gulf of Lions during the 2006 intense cascading and open-sea convection period. *Progress in Oceanography*. DOI: 10.1016/j.bbr.2011.03.031.
- Pasqual, C., Sanchez-Vidal, A., Zúñiga, D., Calafat, A., Canals, M., Durrieu de Madron, X., Puig, P., Heussner, S., Palanques, A., Delsaut, N., 2010. Flux and composition of settling particles across the continental margin of the Gulf of Lion: the role of dense shelf water cascading. *Biogeosciences* 7, 217-231.

- Pasqual, C., Lee, C., Goñi, M., Tesi, T., Sanchez-Vidal, A., Calafat, A., Canals, M., Heussner, S., 2011. Use of organic biomarkers to trace the transport of marine and terrigenous organic matter through the southwestern canyons of the Gulf of Lion. *Marine Chemistry* 126(1-4), 1-12.
- Puig, P., Palanques, A., Orange, D.L., Lastras, G., Canals, M., 2008. Dense shelf water cascades and sedimentary furrow formation in the Cap de Creus Canyon, northwestern Mediterranean Sea. *Continental Shelf Research* 28(15), 2017-2030.
- Recio, M., Trigo, M.M., toro, F.J., Docampo, S., García-González, J.J., Cabezudo, B., 2006. A three-year aeropalynological study in Estepona (Southern Spain). *Annals of Agricultural and Environmental Medicine* 13, 201-207.
- Roussiez, V., Ludwig, W., Monaco, A., Probst, J.L., Bouloubassi, I., Buscail, R., Saragoni, G., 2006. Sources and sinks of sediment-bound contaminants in the Gulf of Lions (NW Mediterranean Sea): A multi-tracer approach. *Continental Shelf Research* 26(16), 1843-1857.
- Salvadó, J.A., Grimalt, J.O., López, J.F., Durrieu de Madron, X., Pasqual, C., Canals, M., 2012. Distribution of organochlorine compounds in superficial sediments from the Gulf of Lion, northwestern Mediterranean Sea. *Progress in Oceanography* (this volume).
- Sánchez-García, L., de Andrés, J.R., Martín-Rubí, J.A., Louchouart, P., 2009. Diagenetic state and source characterization of marine sediments from the inner continental shelf of the Gulf of Cádiz (SW Spain), constrained by terrigenous biomarkers. *Organic Geochemistry* 40(2), 184-194.
- Sanchez-Vidal, A., Pasqual, C., Kerhervé, P., Calafat, A., Heussner, S., Palanques, A., Durrieu de Madron, X., Canals, M., Puig, P., 2008. Impact of dense shelf water cascading on the transfer of organic matter to the deep western Mediterranean basin. *Geophysical Research Letters* 35, L05605.
- Sanchez-Vidal, A., Pasqual, C., Kerhervé, P., Heussner, S., Calafat, A., Palanques, A., Durrieu de Madron, X., Canals, M., Puig, P., 2009. Across margin export of organic

matter by cascading events traced by stable isotopes northwestern Mediterranean Sea. *Limnology and Oceanography* 54(5), 1488-1500.

Sarmiento, J.L., Sundquist, E.T., 1992. Revised budget for the oceanic uptake of antropogenic carbon dioxide. *Nature*, 356, 589-593.

Schmidt, F., Hinrichs, K., Elvert, M., 2010. Sources, transport, and partitioning of organic matter at a highly dynamic continental margin. *Marine Chemistry* 118, 37-55.

Shakya, K.M., Louchouart, P., Griffin, R.J., 2011. Lignin-Derived Phenols in Houston Aerosols: Implications for Natural Background Sources. *Environmental Science Technology* 45, 8268-8275.

Tesi, T., Miserocchi, S., Goñi, M.A., Langone, L., 2007. Source transport and fate of terrestrial organic carbon on the western Mediterranean Sea, Gulf of Lions, France. *Marine Chemistry* 105, 101-117.

Tesi, T., Puig, P., Palanques, A., Goñi, M.A., 2010. Lateral advection of organic matter in cascading-dominated submarine canyons. *Progress in Oceanography* 84(3-4), 185-203.

Tomadin, L., Lenaz, R., Landuzzi, V., Mazzucotelli, A., Vannucci, R., 1984. Wind-blown dusts over the Central Mediterranean. *Oceanologica acta* 7, 13-23.

Thevenot, M., Dignac, MF., Rumpel, C., 2010. Fate of lignins in soils: A review. *Soil Biology and Biochemistry*, 42(8), 1200-1211.

UNEP (United Nations Environment Programme), 2003. Riverine transport of water, sediments and pollutants to the Mediterranean Sea. MAP Tech. Rep. No. 141, Athens, 111 p.

TABLE CAPTIONS

Table 1. Lignin derived phenol concentrations in marine sediment and trap samples from different experiments.

Table 2. Correlation matrix for the total mass flux and organic biomarkers data (mg g^{-1} sediment). Cursive indicate significant correlation at the 0.01 level (2-tailed).

Table 3. Mean value and standard deviation of CP/VP, SP/VP, Sd/Sl and Vd/Vl ratios from middle Lacaze-Duthiers and Cap de Creus canyons during the DSWC event.

Table 4. LP and CA cumulative masses, fluxes and %, at the studied mooring stations separated according to environmental conditions: pre-DSWC (October - December 05), DSWC (January - March 06), post-DSWC (April - mid-July 06) and late post-DSWC (mid-July - October 06).

FIGURE CAPTIONS

Fig. 1. Bathymetry map of the Gulf of Lion and North Catalan margin in the Western Mediterranean Sea and location of the sediment trap mooring stations (white dots) along the three studied transects: Lacaze-Duthiers Canyon (LDC), Cap de Creus Canyon (CCC) and its Southern Open Slope (SOS). The path of the mesoscale Northern Current is also indicated (NC, grey arrow). LFC: La Fonera Canyon. BC: Blanes Canyon.

Fig. 2. Time-series data in settling particles of the studied mooring stations. A) Total mass fluxes ($\text{g m}^{-2} \text{d}^{-1}$). B) OC concentrations (%). C) Lithogenic concentrations (%). The background patterns mark the different ambient conditions: pre-DSWC (squared), DSWC (grey), post-DSWC (white) and late post-DSWC (striped). Numbers along the horizontal scale correspond to year months (October 2005 to October 2006). Numbers to the right of each station three letters code correspond to water depth.

Fig. 3.A) Current speed (cm s^{-1}) and temperature ($^{\circ}\text{C}$) recorded at 5 mab at each station location. B) Mean chlorophyll *a* (chl *a*) concentration (mg m^{-3}) recorded at coastal

(black) station and open ocean (blue). C) Significant wave Heights $H_{1/3}$ (m) recorded at the Sète wave buoy offshore station.

Fig. 4. Organic carbon (OC)-normalized time-series data in settling particles of the studied mooring stations in mg g^{-1} OC. A) Vanillyl phenols (VP). B) Syringyl phenols (SP). C) Cinnamyl phenols (CP). D) Cutins. The background patterns mark the different ambient conditions: pre-DSWC (squared), DSWC (grey), post-DSWC (white) and late post-DSWC (striped). Numbers along the horizontal scale correspond to year months (October 2005 to October 2006). Numbers to the right of each station three letters code correspond to water depth.

Fig. 5. Lithogenic (litho)-normalized time-series data in settling particles of the studied mooring stations in $\mu\text{g g}^{-1}$ litho. A) Vanillyl phenols (VP). B) Syringyl phenols (SP). C) Cinnamyl phenols (CP). D) Cutins. The background patterns mark the different ambient conditions: pre-DSWC (squared), DSWC (grey), post-DSWC (white) and late post-DSWC (striped). Numbers along the horizontal scale correspond to year months (October 2005 to October 2006). Numbers to the right of each station three letters code correspond to water depth.

Fig. 6. Time-series data of various phenol ratios in settling particles of the studied mooring stations. A) Syringyl phenols to vanillyl phenols ratio (SP/VP). B) Cinnamyl phenols to vanillyl phenols ratio (CP/VP). C) Cutins to lignin-derived phenols ratio (CA/VP). The background patterns mark the different ambient conditions: pre-DSWC (squared), DSWC (grey), post-DSWC (white) and late post-DSWC (striped). Numbers along the horizontal scale correspond to year months (October 2005 to October 2006). Numbers to the right of each station three letters code correspond to water depth.

Fig. 7. A) Mean values of syringyl phenols to vanillyl phenols ratio (SP/VP) and cinnamyl phenols to vanillyl phenols ratio (CP/VP). B) Mean values of vanillic acid to vanillin ratio (Vd/Vl) and syringic acid to syringaldehyde ratio (Sd/SI). Confidence bars represent 1 Standard Deviation.

TABLE 1

Reference	Region	Location	Lignin-derived phenols	
			mg 100 mg OC ⁻¹	µg g ⁻¹ DW
Gough et al., 1993	Gulf of Lion	Rhone River and delta	1.29 - 2.08	
		Gulf of Lion shelf	0.14 - 2.95	
		NW Mediterranean slope	0.16 - 0.88	
		NW Mediterranean basin	0.031	
Tesi et al., 2007	Gulf of Lion	Rhone prodelta	2.30 - 4.07	
		Mid-shelf mud belt	0.25 - 1.16	
Tesi et al., 2010 (sediment traps)	Gulf of Lion	Inner shelf	1.77 - 2.62	
		Mid shelf	1.16 - 3.07	
		Outer shelf - canyon head	0.14 - 0.79	
		Upper slope close to CCC	0.74 - 1.54	
		Upper slope close to LDC	0.23 - 0.28	
		CCC (before DSWC)	0.39 - 0.71	
		CCC (during DSWC)	0.25 - 1.30	
		CCC (after DSWC)	0.28 - 0.52	
This study (sediment traps)	Gulf of Lion	Southwestern margin	0.21 – 1.307	31 - 296
Tesi et al., 2008	Adriatic Sea	Shelf-canyon	0.32 – 0.95	
		Bari canyon (sediment traps)	0.17 - .040	
Sánchez-García	Gulf of Cádiz	Guadiana River	3.15 ± 0.56	
		Tinto-Odiel River	2.18 ± 1.18	
		Inner shelf	1.57 ± 0.82	
Gough et al., 1993	North East Atlantic	Celtic shelf	0.012	
		Hatton-Rockall basin	0.055 - 0.120	
		Porcupine abyssal Plain	0.006 - 0.011	
Schmidt et al., 2010	NW Iberian margin	Mid-shelf mud belt		43.05 - 189.26
		Inner-mid shelf		6.89 - 153. 54
		Outer shelf		5.70 - 104.13
		Continental slope		6.51 - 24.85
Gordon et al., 2002	Gulf of Mexico	River SS	2.23 - 3.22	
		Attchafalaya delta and bay	3.45 - 4.06	
		Shoal stations	0.56 - 0.57	
		Inshore stations	1.36 - 2.73	
		Offshore stations	0.96 - 1.26	
Goñi et al., 2008	Gulf of Mexico	Shelf	0.50 - 1.37	
		Slope	0.37 - 0.81	
Goñi et al., 2009 (sediment traps)	Cariaco Basin	Upwelling season	0.100 ± 0.013	
		Stratified season	0.123 ± 0.021	
		Rainy season	0.130 ± 0.020	
		Dry season	0.090 ± 0.010	

TABLE 2

	TMF	Litho (%)	OC (%)	VP	SP	CP	CA
TMF	1,00	0,39	-0,31	0,14	-0,14	-0,32	-0,22
Litho (%)		1,00	-0,77	-0,27	-0,53	-0,63	-0,67
OC (%)			1,00	0,38	0,62	0,68	0,85
VP				1,00	0,73	0,37	0,52
SP					1,00	0,81	0,60
CP						1,00	0,60
CA							1,00

TABLE 3

Station		CP/VP	SP/VP	Sd/SI	Vd/VI
CCC1000	N	6	6	6	6
	Mean	0.44	0.72	0.40	0.46
	SD	0.34	0.11	0.05	0.05
LDC1000	N	3	3	3	3
	Mean	1.83	1.65	0.51	0.52
	SD	0.66	0.61	0.12	0.05

TABLE 4

Station	Period	Sampling days	Days %	LP mg m ⁻²	LP mg m ⁻² d ⁻¹	LP %	CA mg m ⁻²	CA mg m ⁻² d ⁻¹	CA %
LDC300	pre-DSWC	53	17	41.33	0.78	29.0	1.08	0.02	13.7
	DSWC	75	25	7.05	0.09	5.0	0.37	0.00	4.7
	post-DSWC	107	35	33.77	0.32	23.7	3.32	0.03	42.1
	Late post-DSWC	69	23	60.20	0.87	42.3	3.11	0.05	39.4
	Total	304	100	142.34	0.47	100.0	7.88	0.03	100.0
LDC1000	pre-DSWC	60	21	9.57	0.16	8.2	1.00	0.02	9.7
	DSWC	45	16	47.25	1.05	40.6	2.71	0.06	26.1
	post-DSWC	92	33	34.78	0.38	29.9	2.80	0.03	27.0
	Late post-DSWC	84	30	24.74	0.29	21.3	3.85	0.05	37.2
	Total	281	100	116.33	0.41	100.0	10.35	0.04	100.0
LDC1500	DSWC	90	38	21.02	0.23	56.1	2.12	0.02	48.1
	post-DSWC	91	39	12.16	0.13	32.5	1.29	0.01	29.2
	Late post-DSWC	54	23	4.27	0.08	11.4	1.00	0.02	22.7
	Total	235	100	37.45	0.16	100.0	4.40	0.02	100.0
CCC300	post-DSWC	92	57	53.76	0.58	44.7	7.49	0.08	55.4
	Late post-DSWC	69	43	66.39	0.96	55.3	6.04	0.09	44.6
	Total	161	100	120.15	0.75	100.0	13.54	0.08	100.0
CCC1000	pre-DSWC	60	18	9.58	0.16	2.3	1.58	0.03	5.7
	DSWC	90	28	362.61	4.03	86.3	20.22	0.22	72.7
	post-DSWC	107	33	30.51	0.29	7.3	3.79	0.04	13.6
	Late post-DSWC	69	21	17.26	0.25	4.1	2.20	0.03	7.9
	Total	326	100	419.95	1.29	100.0	27.80	0.09	100.0
CCC1500	DSWC	90	54	17.62	0.20	62.5	2.75	0.03	63.0
	post-DSWC	76	46	10.58	0.14	37.5	1.62	0.02	37.0
	Total	166	100	28.21	0.17	100.0	4.37	0.03	100.0
CCC1900	DSWC	75	41	7.87	0.10	42.9	0.57	0.01	35.4
	post-DSWC	92	51	9.46	0.10	51.6	0.57	0.01	35.3
	Late post-DSWC	15	8	1.02	0.07	5.6	0.47	0.03	29.4
	Total	182	100	18.34	0.10	100.0	1.61	0.01	100.0
SOS1000	DSWC	90	43	102.49	1.14	88.2	6.29	0.07	76.8
	post-DSWC	61	29	10.00	0.16	8.6	0.96	0.02	11.8
	Late post-DSWC	60	28	3.74	0.06	3.2	0.94	0.02	11.4
	Total	211	100	116.23	0.55	100.0	8.19	0.04	100.0
SOS1900	DSWC	75	38	10.97	0.15	49.6	0.69	0.01	31.8
	post-DSWC	91	46	8.62	0.09	39.0	0.78	0.01	36.1
	Late post-DSWC	31	16	2.51	0.08	11.4	0.69	0.02	32.0
	Total	197	100	22.10	0.11	100.0	2.17	0.01	100.0
Total	pre-DSWC	233	10	65.84	0.28	5.9	4.50	0.02	5.0
	DSWC	720	30	613.95	0.85	55.4	38.72	0.05	42.8
	post-DSWC	916	38	235.26	0.26	21.2	26.48	0.03	29.2
	Late post-DSWC	520	22	192.36	0.37	17.4	20.86	0.04	23.0
	Total	2389	100	1107.40	0.46	100.0	90.57	0.04	100.0

Fig. 1

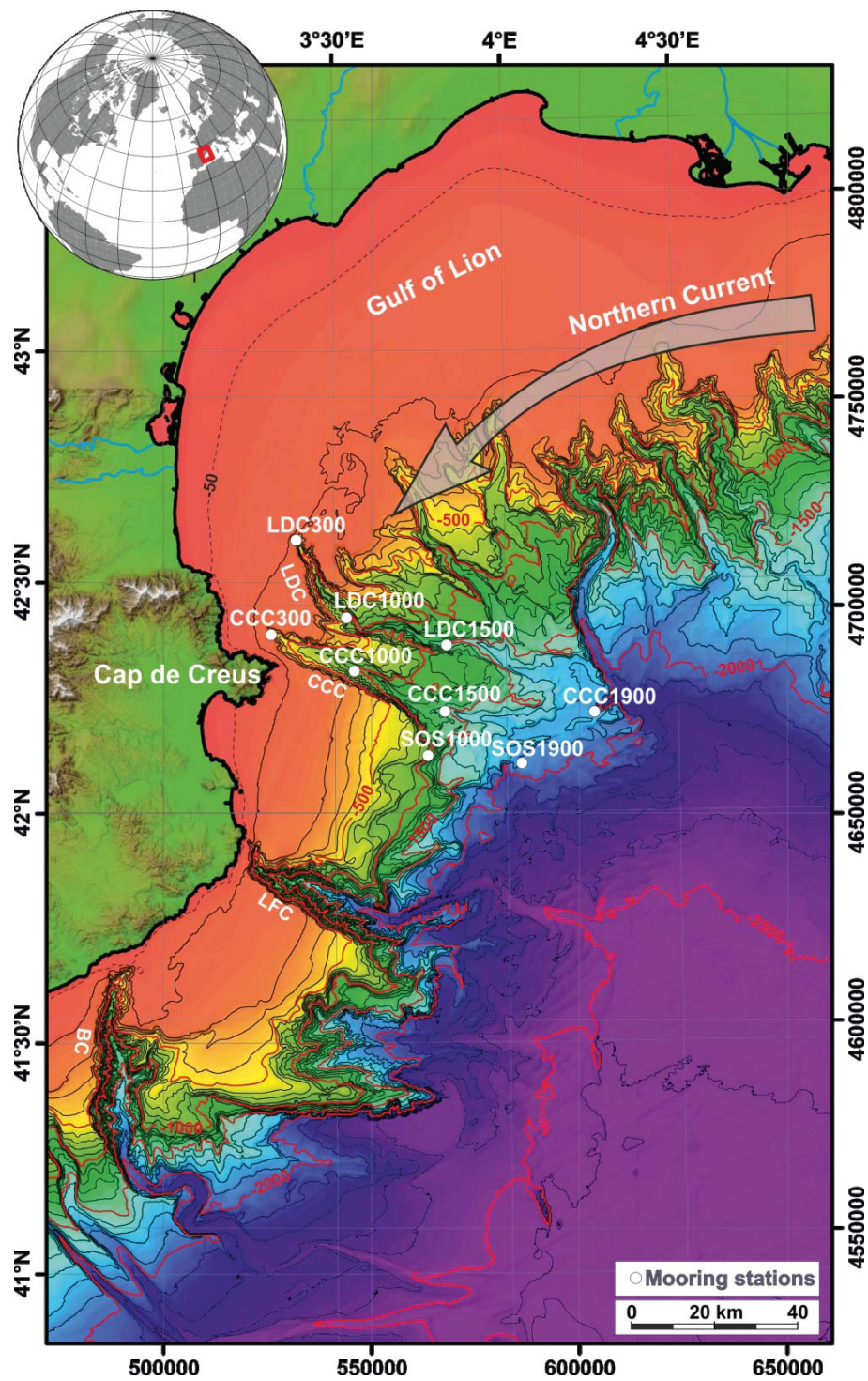


Fig. 2

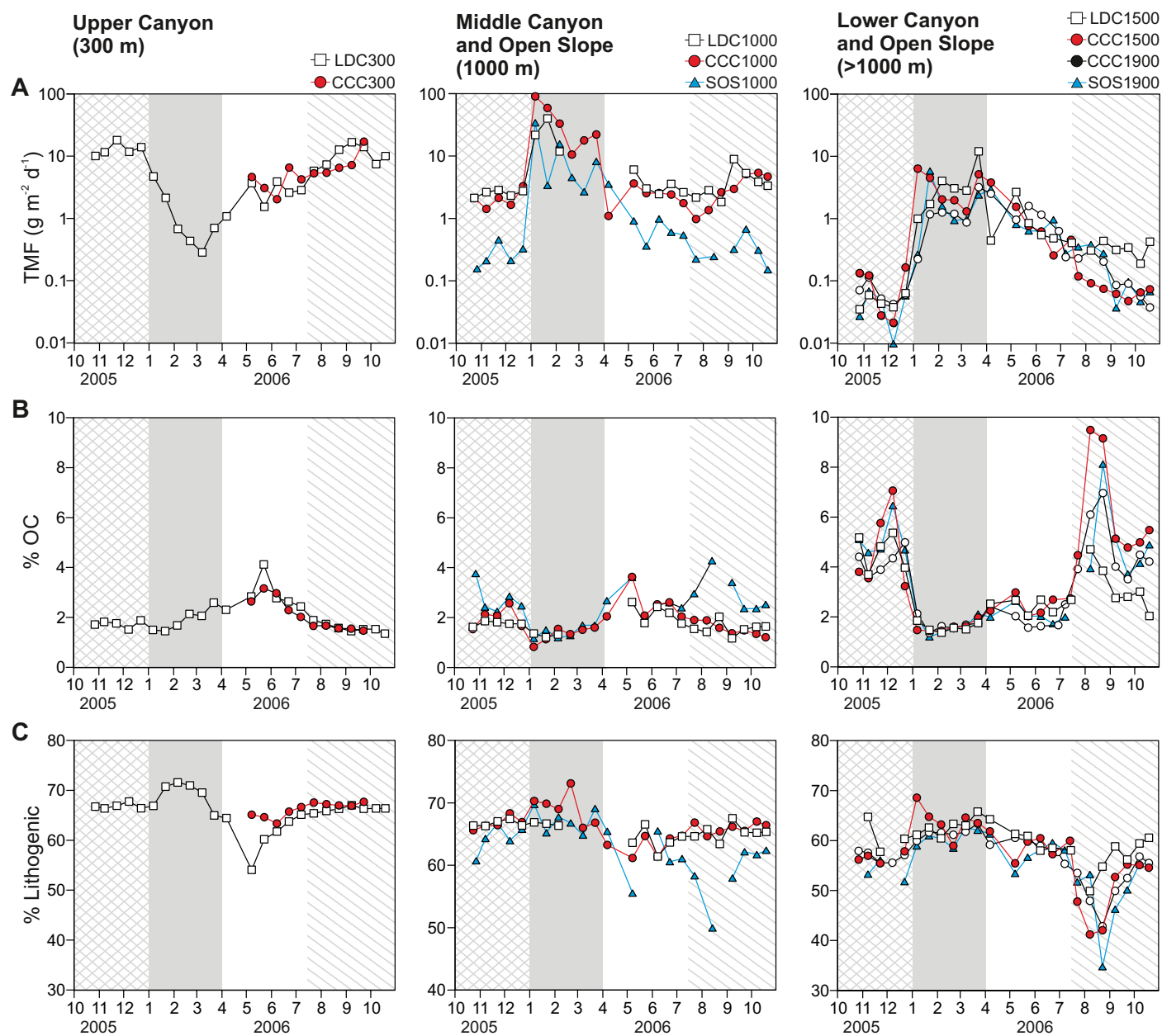


Fig. 3

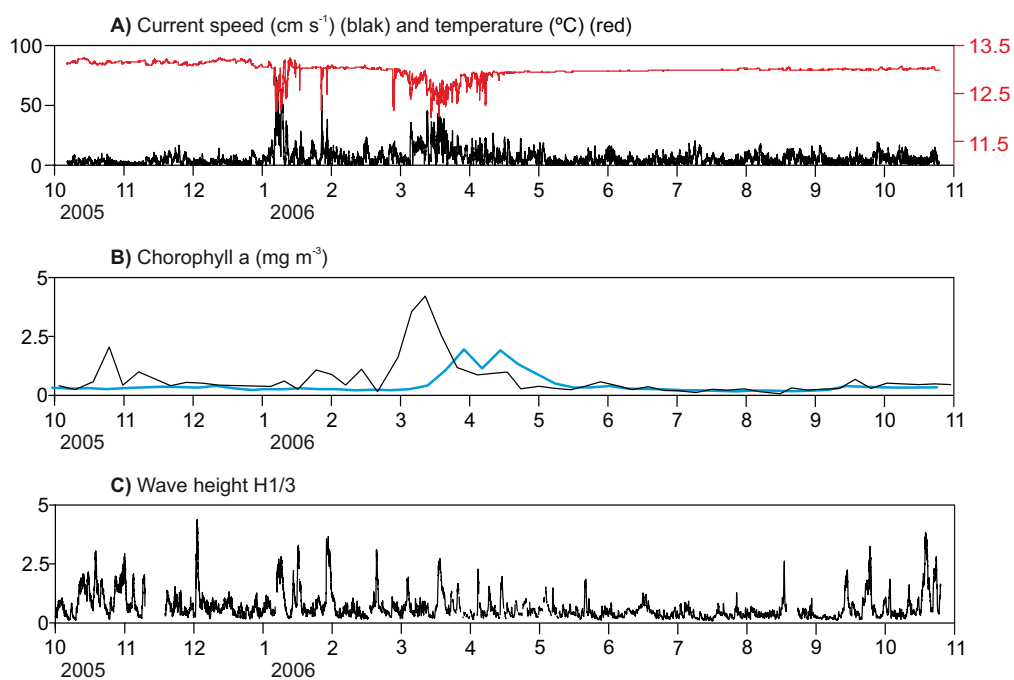


Fig. 4

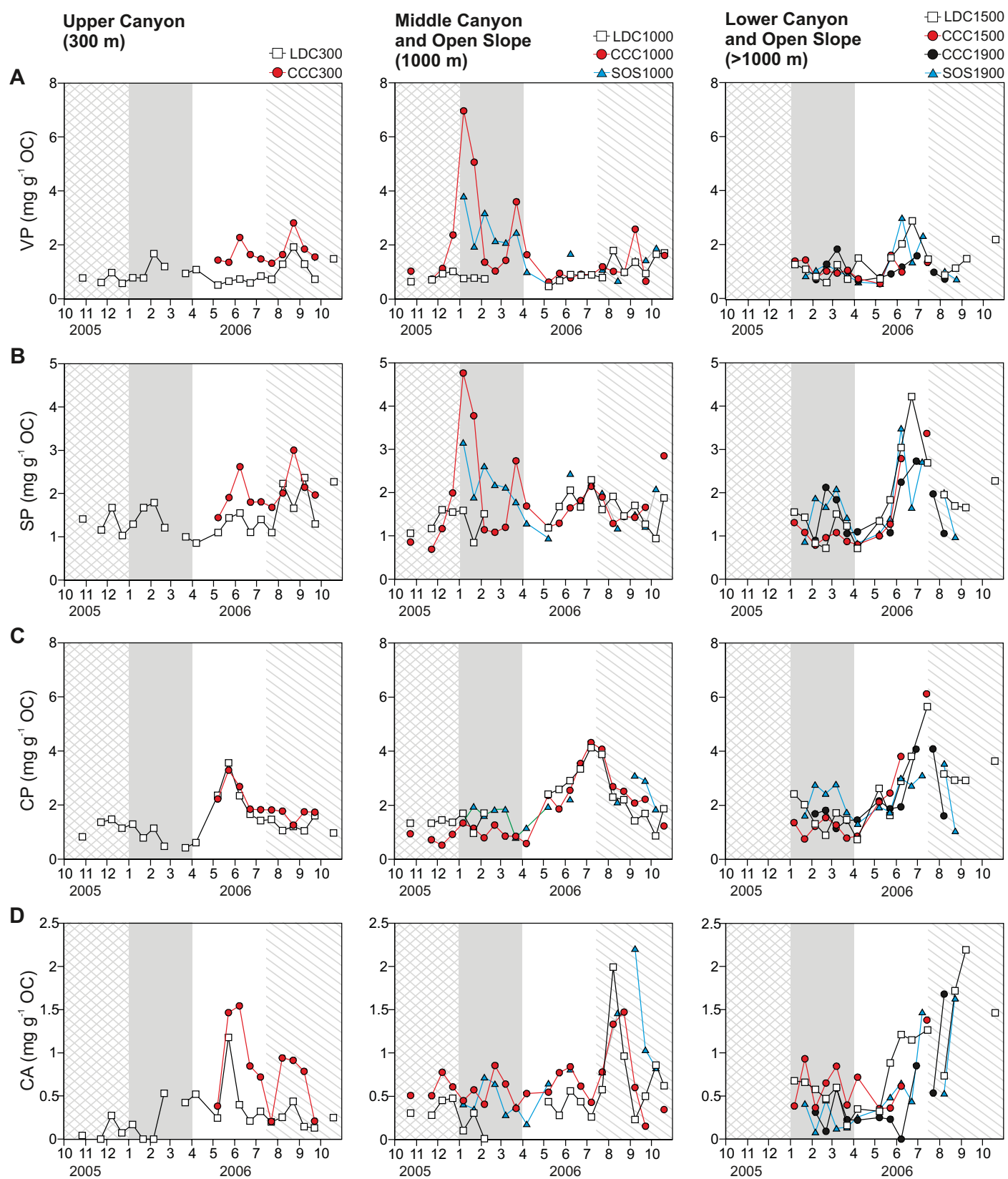


Fig. 5

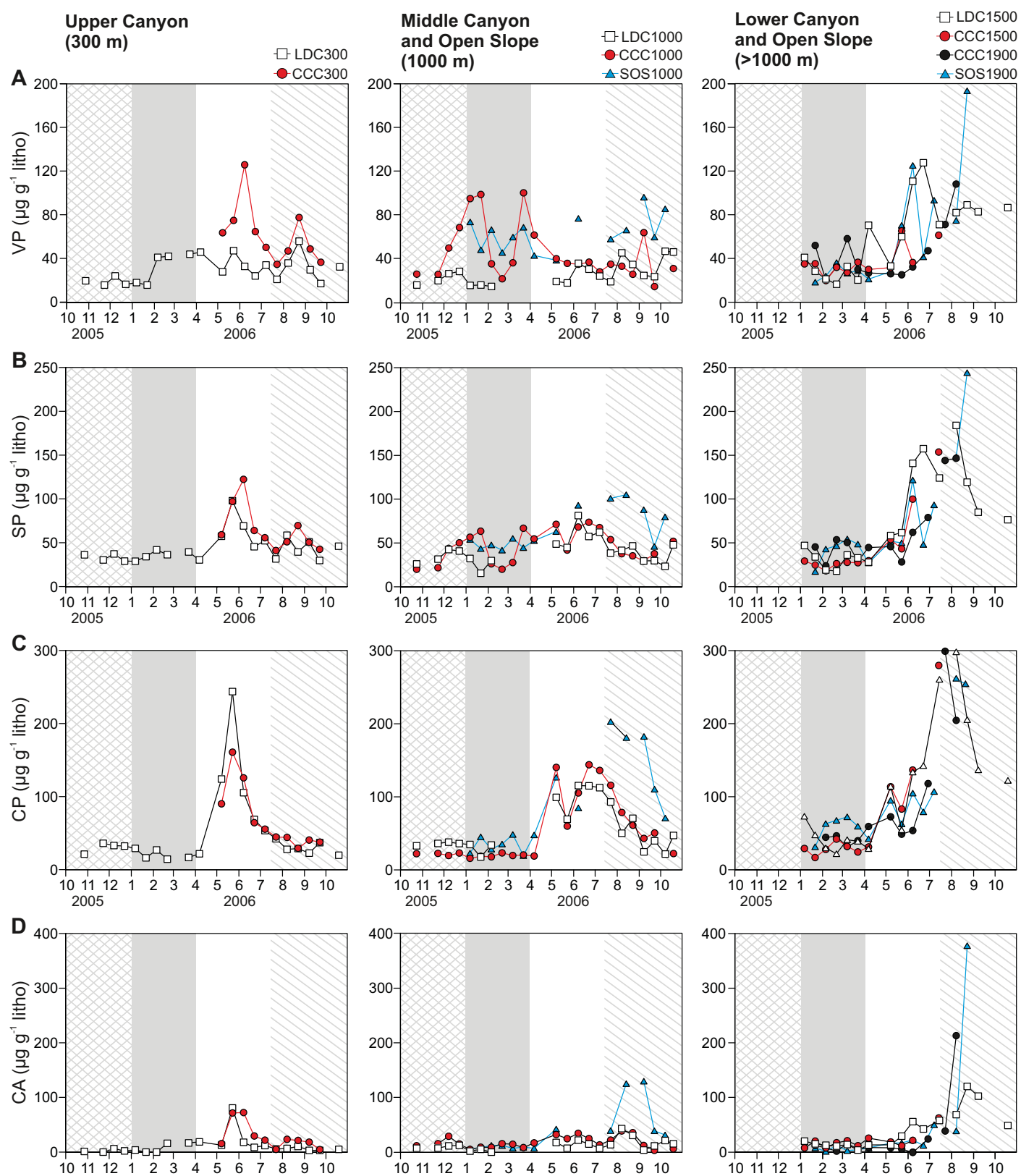


Fig. 6

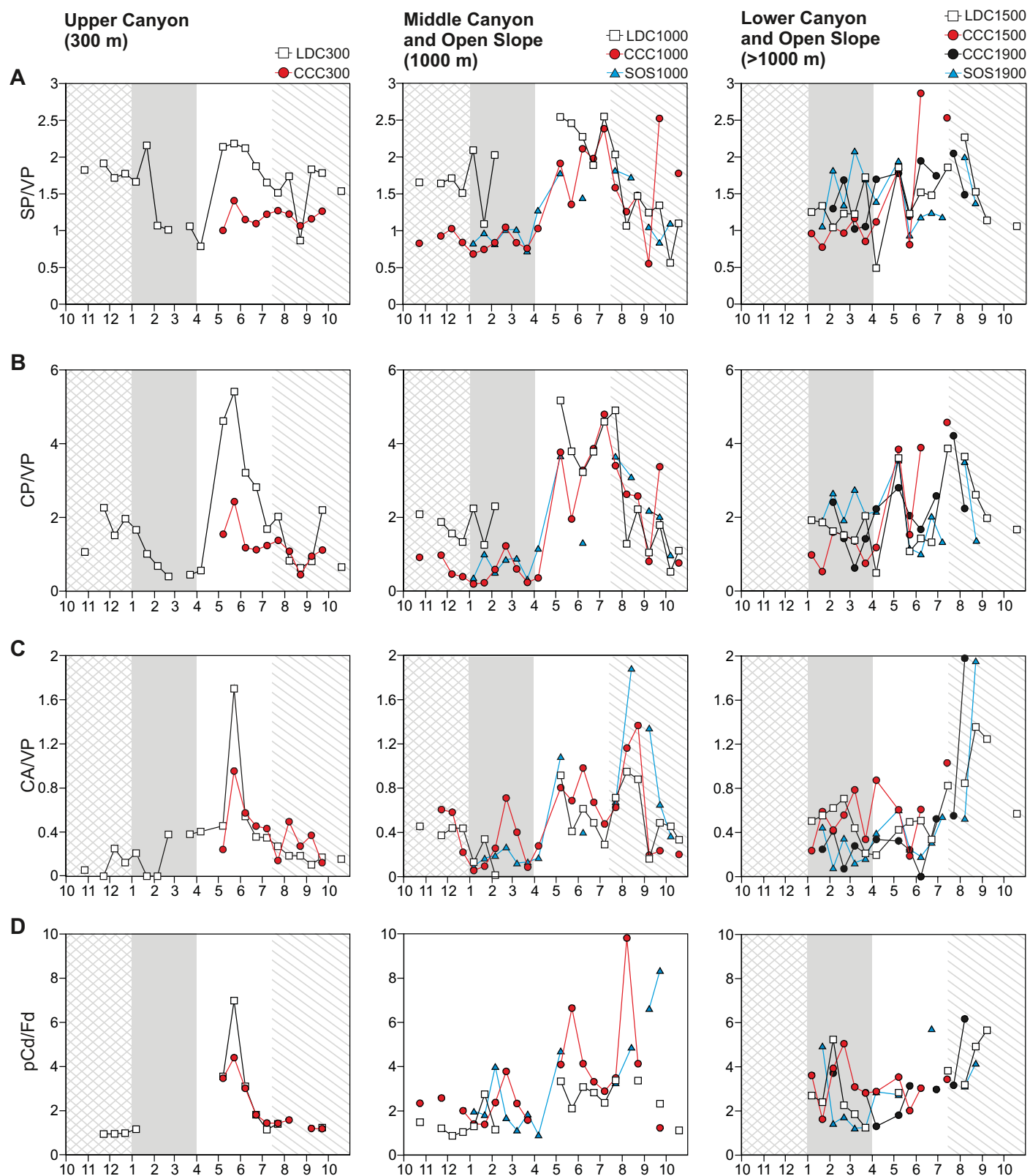


Fig. 7

