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► To cite this version:

Sabine Schmidt, L. Pastor, C. Brandily, E. Miramontes, M. Péron, et al.. Modern sedimentation and geochemical imprints in sediments from the NW Madagascar margin. *Marine Geology*, 2020, 426, pp.106184. 10.1016/j.margeo.2020.106184 . hal-03431785

HAL Id: hal-03431785

<https://hal.science/hal-03431785>

Submitted on 20 May 2022

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1 Modern sedimentation and geochemical imprints in sediments from the NW Madagascar margin

2 ¹Pastor L., ¹Brandily C., ²Schmidt S., ³Miramontes E., ¹Péron M., ¹Appere D., ⁴Chéron S., ⁴Boissier A.,
3 ⁵Jouet G.

4 (1) IFREMER, Centre de Bretagne, REM/EEP, Laboratoire Environnement Profond, F-29280
5 Plouzané, France

6 (2) UMR5805 EPOC, CNRS, OASU, Université de Bordeaux, 33615 Pessac, France

7 (3) MARUM – Center for Marine and Environmental Sciences, University of Bremen,
8 Klagenfurter Straße 2-4, Bremen, 28359, Germany

9 (4) IFREMER, Centre de Bretagne, REM/GM, Laboratoire Cycles Géochimiques et ressources, F-
10 29280 Plouzané, France

11 (5) IFREMER, Centre de Bretagne, REM/GM, Laboratoire Géodynamique et enregistrement
12 Sédimentaire, F-29280 Plouzané, France

13

14 **Abstract**

15 The NW Madagascar continental margin receives high loads of terrigenous particulate organic matter
16 during the wet season and especially linked to extreme events, originating from two major rivers, the
17 Betsiboka and the Mahavavy Rivers. This particulate matter contains a high content of iron minerals
18 from the weathering of red ferruginous/ferralitic soils of Madagascar. The presence of pockmarks,
19 i.e. gas or fluid expulsion features on the continental slope, testifies to past/present methane
20 migration through the sedimentary column, associated with early diagenetic processes. This study
21 globally aims at deciphering the interactions between episodic sedimentation and geochemical
22 processes influenced by fluids upward migration, using a sediment trap mooring and interface
23 sediment cores at two sites on the continental slope. The present-day sedimentation along this
24 margin undergoes two patterns. During the wet season, high continental fluxes generally result in an
25 increase in particle fluxes on the slope. Longshore currents may deflect river plumes along slope

26 resulting in some periods of low particle flux on the slope during the wet season. During the dry
27 season, the particles collected in the water column are probably originated from sediment
28 remobilization from the shelf and slope. The observed progradation of the Mahavavy River delta and
29 prodelta between 1984 and 2016 argues for an increase in temporary connections of the river with
30 the canyon head during extreme events, inducing pulsed sedimentation offshore. This pulsed
31 sedimentation could be responsible for enhancing pyritisation on surface sediments, due to higher
32 inputs of terrigenous organic matter and iron oxides. Finally, methane upward migration also
33 influences the pyritisation process through anaerobic oxidation of methane using sulfate as an
34 electron acceptor and methanotrophs associated with this reaction significantly impact the $\delta^{13}\text{C}$ of
35 organic carbon towards more negative values.

36 1 INTRODUCTION

37 Rivers represent the largest transportation pathway of organic particulate matter to coastal areas,
38 and subsequently to continental shelves, slopes and deep ocean basins (Bauer et al., 2013).
39 Continental margins, which encompass the continental shelf, slope and rise, are thus impacted by
40 high inputs of terrigenous organic material, 55– 80 % of this flux being remineralized in the upper
41 sedimentary column along these margins (Burdige, 2005). The remainder could undergo long term
42 biogeochemical reactions like oil and gas formation at depth that could eventually migrate through
43 the sedimentary column, or be finally buried. Geochemical signatures in margin sediments are thus
44 influenced by several factors, including sedimentation intensity and nature, episodic events and
45 diagenetic processes. Diagenetic processes can modify, for instance, the $\delta^{13}\text{C}$ signature of organic
46 matter through its mineralisation (McArthur et al., 1992), the isotopic composition of several
47 elements in foraminifera tests through the dissolution/reprecipitation of carbonates phases (Edgar et
48 al., 2015), or the magnetic properties of sediment through the dissolution and precipitation of iron
49 phases (Larrasoana et al., 2009). In environments where diagenetic processes are highly diverse like
50 in cold seeps areas or deltaic environments, it can be intricate to quantify their relative impact on

51 paleosignatures. Thus, studies aiming at reconstructing paleosignals or determining the source of
52 sediments have to take early diagenesis into account.

53 The NW continental margin of Madagascar receives large amount of sediment from two main rivers,
54 the Betsiboka and the Mahavavy Rivers. There are also evidences of local methane escapes on the
55 continental slope (Acoustic anomalies detected in the water column - Dupré *et al.*, 2019). The
56 sediment dynamics and geochemical signatures on the continental slope under the influence of the
57 rivers input have been monitored using one mooring with a sediment trap, temperature and current
58 velocity measurements, and interface sediment cores. The objectives of this work were to 1) give a
59 first insight on the modern sedimentary patterns along this slope and 2) decipher the relative impact
60 of the sedimentation versus methane escape on geochemical signatures (Ti/Ca, Ti/Al, $\delta^{13}\text{C}$ of organic
61 matter, sedimentary sulfur and iron) of sediment, in order to distinguish between changes in inputs
62 versus changes in diagenetic processes.

63 2 REGIONAL SETTING

64 The Majunga basin is one of the two major sedimentary basins of Madagascar (with the Morondava
65 basin). This basin partly covers onshore and offshore areas (Tari *et al.*, 2004). In this paper, we only
66 discuss data from the offshore Majunga basin. The Betsiboka River is the largest river in Madagascar
67 with a length of 605 km and a watershed of 49,000 km² (Randrianarijaona, 1983). Spectacular gullies,
68 named lavakas by the Malagasy people, are widespread in Madagascar and are especially dense in
69 the highlands of central Madagascar around the Betsiboka watershed (Cox *et al.*, 2010). The river
70 supplies the coastal area with sediment eroded from ferruginous and ferralitic soils of the Majunga
71 basin and of the lavakas (Ramifehiarivo *et al.*, 2017). Fluvial discharges varies from 35 m³ s⁻¹ during a
72 very dry season to 4,500 m³ s⁻¹ during wet periods, with extreme values during flood reaching >
73 25,000 m³ s⁻¹ (Berthois and Crosnier, 1966; Chaperon *et al.*, 1993). From measurements in surface
74 waters, solid transport was estimated in the 1960s at about 1660 t yr⁻¹, which corresponds to 1.1 mm

75 of soil eroded per year for the whole watershed (Chaperon et al., 1993). During flood events, the
76 plume of fine suspended particles reaches up to 17 km offshore towards the north and about 13 km
77 in the east-west direction alongshore (Berthois and Crosnier, 1966). Otherwise, particles remain
78 confined within the estuary (coarse material) and within a few kilometers north of the outlet on the
79 shelf (fine particles).

80 About 50 km westward, the Mahavavy River is 410 km long and has a watershed of 18,500 km². Its
81 connection to the ocean forms a long delta spread along approximately 20 km of coastline. Discharge
82 was measured randomly at Marovato between 1950 and 1973 with a range of 26-4,300 m³ s⁻¹
83 (Chaperon *et al.* 1993). There is currently no estimate for solid transport.

84 There are no recent hydrogeological data for these two rivers. Both river outlets are located on the
85 northern tip of Madagascar that has been uplifting at a rate of 0.02-0.08 mm yr⁻¹ over the last 0.1–
86 0.8 Ma (Battistini, 1972; Roberts et al., 2012). Accretion and mangrove colonization on new-formed
87 mudflats have been evidenced (Salomon, 2009). Continuous deforestation on the island since 1600
88 AD coupled with a growing population relying on agriculture (Harper et al., 2007; Klein, 2002)
89 contributes to the very high erosion rates, between 250 and 400 t ha⁻¹ yr⁻¹, in the highlands, among
90 the highest rates measured worldwide (Randrianarijaona, 1983).

91 The circulation in the Comoros Basin and the NW margin of Madagascar is characterized by intense
92 mesoscale eddies that result in a high variability in the direction and intensity of oceanic currents
93 (Collins et al., 2014; 2016; Pripp et al., 2014). Anticyclonic eddies are mainly generated west of Cape
94 Amber (northernmost tip of Madagascar) and are associated with the North East Madagascar
95 Current (NEMC), while cyclonic eddies are mainly formed along the northwestern margin of
96 Madagascar due to strong baroclinic instability (Collins et al., 2014). The NW Madagascar margin is
97 very often affected by cyclonic eddies that generate currents flowing SW along the slope that can

98 deflect the river plumes and affect sediment transfer from the continent to the deep sea (Figure 1C;
99 Collins et al., 2016; Pripp et al., 2014).

100 3 MATERIEL AND METHODS

101 3.1 Sampling strategy

102 The North western Madagascar margin in the Comoros Basin was investigated during two cruises:
103 the PAMELA-MOZ1 cruise in 2014 (Olu, 2014) aboard the RV L'Atalante and the PAMELA-MOZ4
104 cruise in 2015 (Jouet and Deville, 2015) aboard the RV Pourquoi pas?. The two main sampling sites
105 are located on the upper continental slope, offshore the outer shelf carbonate reefs where water
106 depths increase from a few meters to a few hundred meters. This area is characterized by a number
107 of pockmarks (i.e. sedimentary depressions formed during gas or fluid expulsion) located between
108 400 and 750 m depth (Figure 1).

109 Site S1 is located 30 km off the Mahavavy River at a water depth of 789 m (Figure 1 - Table 1). At this
110 site, a mooring (MLP1), equipped with a current-meter and a sediment trap, was deployed in
111 October 2014 and retrieved in November 2015. It was positioned at a water depth of 781 m (Figure
112 1 – Table1). It was equipped with a RCM 11 current meter (Aanderra) positioned 30 m above the
113 bottom (mab) and coupled to a sediment trap (PPS 4/3, 24 sampling bottles), with a sampling surface
114 of 1m², positioned 10 mab. Additionally, two multicorers were deployed within a pockmark
115 (diameter = 600 m) in 2014 (S1-P - Table 1 - Figure 1). Site S2 is located ~30 km north-eastwards of
116 site 1, 50 km off the Betsiboka River estuary, at a water depth ranging from 529 to 762 m (Figure 1 -
117 Table 1). At this site, one pockmark was sampled through multicores taken inside (diameter = 220 m,
118 S2-P1 in 2014 and S2-P2 in 2015 - Figure 1) and outside the same pockmark around its edge (S2-PE1
119 in 2014 and S2-PE2 in 2015 - Figure 1). Two additional multicores were collected, one outside the
120 pockmarks area in 2014 (S2-OUT - Figure 1) and one in 2015 on a slope where gas seeping was

121 evidenced using water column acoustic data (Dupré et al., 2019) (S2-G - Figure 1). Salinity was
122 measured in the water column using a SEACAT SBE19 CTD.

123 3.2 Samples treatment

124 Before deployment of the mooring, sampling bottles were filled with filtered seawater and sodium
125 borate-buffered formalin to a final concentration of 3%. Settling particles were collected from 26
126 October 2014 to 15 November 2015 with a constant sampling interval of 17 days. Samples were kept
127 at 4°C before treatment. In the laboratory, the content of each bottle was visually examined to
128 retrieve swimmers, identified as organisms larger than 1 mm, and rinsed with Milli-Q water to
129 remove salts and formalin. Particles were freeze-dried, weighed to determine mass flux, and crushed
130 for further geochemical analysis.

131 Interface sediments were collected with a multicorer equipped with eight Plexiglas tubes (96 mm
132 internal diameter). Twenty to forty centimetres of undisturbed sediment cores were retrieved with
133 their overlying water. Photographs were taken of the half section of one core. One core was sliced
134 horizontally every 0.5 cm from the sediment-water interface to 2 cm depth, every 1 cm to 10 cm
135 depth and then every 2 cm downcore. Each layer was immediately frozen (−80 °C) on board in a pre-
136 weighed vial. Back at the laboratory, sediment was weighed, freeze-dried, weighed again and then
137 homogeneously crushed into a fine powder. Porosity was calculated considering the wet and freeze-
138 dried weight and assuming a sediment bulk density of 2.7 g cm⁻³ (standard value for mud - (Hamilton,
139 1976)). It was corrected for salt content using measured bottom water salinity. A second core was
140 used to sample porewater using rhizons® (Rhizosphere Research Products) usually every 1 or 2 cm.
141 One aliquot (2-4 mL) was stored in a 10 mL glass vial with 15 µl of HgCl₂, and sealed for further
142 methane measurements. As rhizons are not adequate to measure gas concentration, one core S2-G
143 was sub-sampled during its extrusion; 3 cc of sediment were immediately collected in each layer

144 using a pre-cut syringe, directly transferred into a 20 mL glass vial with 5 mL 1M NaOH, sealed and
145 stored head-down at 4°C before methane measurement at the laboratory.

146 3.3 Analyses

147 Total sulfur and carbon were determined using a Leco CNS-2000 auto-analyzer. The organic carbon
148 content (OC) was measured with a Leco CS-125 elemental analyzer after removing carbonates
149 overnight at a temperature of 60°C with 30% HNO₃. Inorganic carbon (IC) was calculated from the
150 difference between total and organic carbon.

151 Crystalline oxo-hydroxides of iron and manganese were determined by dithionite extractions (Mn_{dith}
152 and Fe_{dith}) (Kostka and Luther III, 1994) coupled to ICP-AES determination using a Horiba Jobin Yvon®
153 Ultima 2 spectrometer. A certified sediment (MESS-4, NRC-CNRC) was used to check the repeatability
154 of the extractions through time. The global variability of MESS-4 was around 5% (standard error).

155 δ¹³C of the sedimentary organic matter was determined using Combustion Module-Cavity Ring
156 Down Spectroscopy (CM-CRDS - Picarro) (Balslev-Clausen et al., 2013). Around 250 mg of samples
157 were first acidified with successive additions of HCl 1N at 40°C until complete carbonate dissolution
158 and rinsed twice with milli-Q water to retrieve acid. After freeze-drying, around 10 mg were then
159 introduced into 5 × 9 mm tin capsules for analysis. Calibration was performed using International
160 Atomic Energy Agency (IAEA) reference material: calcite (NBS-18), sucrose (CH-6) and lithium
161 carbonate (LSVEC). An acetanilide standard (Costech) was inserted every ten samples to correct for
162 the drift, with a measured value of -33.50 ± 0.02‰ (6 replicates). Precision was typically within 0.03
163 ‰ for a triplicate.

164 ²¹⁰Pb and ²²⁶Ra were measured on dried particles and sediments using a high-efficiency, low-
165 background well-type germanium detector (Canberra) (Schmidt et al., 2009). The standard used for
166 the calibration of the gamma spectrometer is IAEA RGU-1. Excess ²¹⁰Pb in settling particles and in
167 sediments, ²¹⁰Pb_{xs}, was calculated by subtracting the activity supported by its parent isotope, ²²⁶Ra,

168 from the total measured ^{210}Pb activity. Errors on $^{210}\text{Pb}_{\text{xs}}$ were calculated by propagating the errors on
169 ^{210}Pb and ^{226}Ra measurements. Sediment accumulation rate (SAR, expressed in cm yr^{-1}) was
170 calculated from the decrease of $^{210}\text{Pb}_{\text{xs}}$ with depth using the CFCS (constant flux and constant
171 sediment model (Schmidt et al, 2009)). The mass accumulation rate (MAR, expressed in $\text{g m}^{-2} \text{cm}^{-1}$) is
172 obtained by plotting the regression of $^{210}\text{Pb}_{\text{xs}}$ against cumulative mass.

173 The bulk sediment semi-quantitative geochemical composition was measured with an Avaatech X-ray
174 fluorescence (XRF) core scanner on half sections. Measurements were carried out every one
175 millimeter applying a basic voltage of 10 and 30 kV to estimate elemental relative concentrations
176 (aluminum (Al), sulfur (S), calcium (Ca) and titanium (Ti)). Results are given in counts per second
177 (Cps).

178 Quantitative X-ray Fluorescence (XRF) analyses were carried out on selected samples from S1-ST in
179 order to obtain the elemental composition (e.g. TiO_2 , CaO , Al_2O_3 , Fe_2O_3). Half a gram of dried
180 sediment was fused at 1050°C with 9 g of fusion flux (90 % lithium tetraborate (LiB_4O_7)/10 % of
181 lithium fluoride (LiF)) and 500 μL of lithium bromide solution (LiBr) at 250 g L^{-1} . The obtained fused
182 beads were analyzed on a sequential X-ray wavelength dispersive spectrometer (WD-XRF) with a Rh
183 x-ray tube (S8 Tiger from Bruker). A set of certified geostandards from the “Canadian Certified
184 reference materials Project” (CCRMP - FER-1, FER-2, WMG-1), the “Geological Survey of Japan” (GSJ -
185 JB-3, JCFA-1, JLS-1, JSD-2) and the “Centre de Recherches Pétrochimiques et Géochimiques”
186 in France (CRPG - BE-N, IF-G) was used for the calibration. All samples were analyzed twice, with a
187 third run in case of divergence. The relative error on major elements (up to 1%) is below 5%.
188 Variations in Ti/Ca and Ti/Al ratios calculated from semi-quantitative XRF throughout the cores are
189 used to point depositional events, while molar ratios calculated from quantitative XRF on S1-ST are
190 used to check variations during the sampling time.

191 Methane concentrations were measured by headspace gas chromatography with a PR2100 gas
192 chromatograph equipped with a flame ionization detector (GC/FID Perichrom, France) connected to
193 a headspace injector (dani HSS 86.50)(Sarradin and Caprais, 1996).

194 4 RESULTS

195 All geochemical data are provided in supplementary material.

196 4.1 Currents and particulate fluxes in site 1

197 Current velocities measured 30 m above the seafloor reached up to 42 cm s^{-1} with an averaged value
198 of 7.8 cm s^{-1} (Figure 2). Daily means ranged between 4.3 and 15.5 cm s^{-1} . Main current directions
199 were N-NNE and S-SSW (Figure 2B). These changes in current direction are related to tides (Figure
200 3A). Temperature ranged between 6.5 and $9.4 \text{ }^\circ\text{C}$ with an averaged value of $7.8 \text{ }^\circ\text{C}$ (Figure 2A). A
201 slight decrease in temperature from 8.0 to $7.7 \text{ }^\circ\text{C}$ on average is also visible from mid-February to
202 September (Figure 2A). A tropical storm (Chedza) occurred on 16 January 2015, but no significant
203 changes in current nor temperature were observed associated with this event (Figure 2).
204 Temperature shows high-frequency oscillations related to tides (Figure 3B).

205 The cumulated rainfall recorded in Majunga, at the Betsiboka outlet, indicates that the wet season
206 spread from November to March/April, with a daily maximum of 59.8 mm the January 11th
207 (www.worldweatheronline.com/majunga-weather-history/mahajanga/mg.aspx). During the 13 days
208 preceding the tropical storm Chedza, total rain of 361 mm was recorded in Majunga. The total
209 sediment mass flux (TMF) recorded in the sediment trap varied between 0 (from February 22nd to
210 March 11th) and $2837 \text{ mg m}^{-2} \text{ d}^{-1}$ (from January 2nd-19th), this maximum being recorded around the
211 Chedza storm (Figure 4). From January 2nd to February 5th, we suspect that particles have
212 overflowed the sampling bottles, which may lead to an underestimation of the bi-weekly TMF during
213 this period, as part of the particles may have been stored in the next one or two bottle(s). Then, even
214 if the absolute bi-weekly TMF from January 2nd to February 22nd should be taken with caution, the

215 cumulated TMF remains correct. TMF varied between 51 and 750 mg m⁻² d⁻¹ during the dry season.
216 During the wet season, the daily averaged particulate mass export was multiplied by 2.8 compared to
217 the dry season (353 versus 124 g m⁻² d⁻¹). Organic and inorganic carbon contents ranged between
218 1.9% and 3.0%, and 1.8% and 2.8%, with averaged values of 2.3% and 2.2%, respectively, without
219 drastic evolution (Figure 4). Total sulfur (Figure 4) varied from 0.16% to 0.51% (average 0.37%), with
220 higher values during the dry season. δ¹³C of organic carbon varied from -21.6 to -20.6 ‰ (average -
221 21.0 ‰) (Figure 4). ²¹⁰Pb_{xs} activities present also a limited variability, between 1794 and 2256 mBq g⁻¹,
222 the lowest levels correspond to the lowest mass fluxes. Mn_{dith} ranged between 3.5 μmol g⁻¹ during
223 the dry season up to 12.9 μmol g⁻¹ during the wet season, with a marked increase of concentration
224 during the wet season. Fe_{dith} ranged between 488 and 527 μmol g⁻¹ with no clear pattern during the
225 year. TiO₂, CaO, Al₂O₃ and Fe₂O₃ ranged between 0.62-0.71%, 8.1-12.1%, 17.0-18.4%, 6.4-7.1%,
226 respectively, through the sampling period. Ti/Ca and Ti/Al ratios ranged between 0.040-0.054 and
227 0.023-0.026, respectively, with no significant variations.

228 4.2 Sediments characteristics

229 4.2.1 Visual description of the cores

230 The image of S1-P highlights marked layers of sediments of different colors (0-16 cm), from orange at
231 the top, followed by a well-defined black layer, a brownish/dark orange layer, some diffuse black
232 sediment and dark grey sediment at depth (Figure 5). S2-P1 and S2-P2 were taken at the same
233 surface GPS point during MOZ1 (2014) and MOZ4 (2015) cruises, respectively, and look very different
234 (Figure 6). S2-P1 exhibits an orange layer at the top (0-10 cm) with traces of bioturbation, followed
235 by light greenish sediment. S2-P2 is much darker with a black layer at the top (0-6 cm). S2-PE1 and
236 S2-PE2 were taken at the same surface GPS point during MOZ1 (2014) and MOZ4 (2015) cruises,
237 respectively, and both exhibit an orange layer at the top (thicker in S2-PE2) and a light greenish
238 sediment underneath (Figure 6). S2-G is dark gray all along the core. Finally, S2-OUT has a thick
239 orange layer at the top (0-20 cm) followed by light greenish sediment (Figure 6).

240 4.2.2 Sediment and mass accumulation rates

241 $^{210}\text{Pb}_{\text{xs}}$ profiles were determined on 3 cores at site 2 (S2-PE1; S2-PE2; S2-G - Figure 6). $^{210}\text{Pb}_{\text{xs}}$ activities
242 are rather comparable in the top sediment, ranging between 548 and 845 mBq g^{-1} , and present an
243 exponential decrease with depth to reach negligible level at about 10 cm. The limited variability in
244 sedimentation rates among the different cores at site 2, regardless of their position, results in very
245 similar $^{210}\text{Pb}_{\text{xs}}$ profiles and thus similar SAR and MAR. At the edge of a pockmark (S2-PE1 and S2-PE2),
246 the SAR is $0.052 \pm 0.004 \text{ cm yr}^{-1}$ and $0.039 \pm 0.005 \text{ cm yr}^{-1}$ while the MAR is $249 \text{ g m}^{-2} \text{ yr}^{-1}$ and 230 g m^{-2}
247 yr^{-1} , respectively. On a gas escape at S2-G, the SAR is $0.059 \pm 0.006 \text{ cm yr}^{-1}$ and the MAR is 243 g m^{-2}
248 yr^{-1} . In fact, the mass accumulation rate, which integrates the compaction effect, is equivalent at the
249 three sites, indicating a rather homogenous particle deposition on the area. The profile of $^{210}\text{Pb}_{\text{xs}}$ is
250 completely different at site 1 (Figure 5): first, the top $^{210}\text{Pb}_{\text{xs}}$ activity is much higher compared to S2
251 cores (1372 mBq g^{-1}), and is closer to the activities recorded in the sediment trap. In addition, the
252 $^{210}\text{Pb}_{\text{xs}}$ profile in core S1-P presents a decrease according to two slopes, which is interpreted as a
253 change in sedimentation intensity. In the most recently deposited sediments, from the top to about
254 16 cm, the SAR is $0.249 \pm 0.054 \text{ cm yr}^{-1}$ (Figure 5), which corresponds to a mean MAR of $1381 \text{ g m}^{-2} \text{ yr}^{-1}$
255 (Fontanier et al., 2018). Deeper in the sediment ($> 16 \text{ cm}$; before 1950s), the SAR is much lower
256 ($0.057 \pm 0.007 \text{ cm yr}^{-1}$) (Figure 5), which corresponds to a MAR of $320 \text{ g m}^{-2} \text{ yr}^{-1}$. In addition to the
257 main pattern, $^{210}\text{Pb}_{\text{xs}}$ activities present low activities at 2-3, 13-14 and 19 cm (Figure 5), that could be
258 the result of a temporary input of low $^{210}\text{Pb}_{\text{xs}}$ particles.

259 4.2.3 Geochemical data

260 Sedimentary OC concentrations ranged between 0.8% and 2.4% (Figures 5 and 6), IC concentrations
261 between 0.3% and 5.7%, and total S between 0.07% and 3.7%. S1-P recorded the highest values of
262 OC and S, and the lowest values of IC. In all cores, Mn_{dith} ranged between 0.5 and $17.9 \mu\text{mol g}^{-1}$ and
263 Fe_{dith} between 95.7 and $710.8 \mu\text{mol g}^{-1}$.

264 Ca values measured by semi-quantitative usually ranged between 1.10^5 and 2.10^5 Cps, with lower
265 values at the top of cores from sites S2-P2, S2-G and S1-P. S values measured by semi-quantitative
266 XRF are usually < 2000 Cps, with higher values in S2-P2, S2-G and S1-P (up to 7500 Cps^{s-1}). Ti/Ca are
267 in average around 0.04-0.06, with high variations at the top of S2-P2, S2-G and S1-P. Ti/Al ranged
268 between 2 and 5 in all sites.

269 A ternary plot between OC, S and Fe_{dith} separate 2 distinct groups of sites (Figure 7): the first group
270 with a higher relative proportion of total S and lower Fe_{dith} contribution (S2-G, S2-P1, S2-P2 and S1-
271 P), the second group with inverse proportions of S and Fe_{dith} (S2-PE1, S2-PE2, S2-OUT and S1-ST).

272 $\delta^{13}\text{C}$ of organic carbon varied between -30.0‰ and -23.5‰ in S2-G (Figure 6) and between -27.3‰
273 and -21.1‰ in S1-P (Figure 5), and was more stable in other cores between -23.8‰ and -19.9‰
274 (Figure 6). Ratios measured in trapped particles were $-21.0 \pm 0.3 \text{‰}$. Overall, ratios in trapped
275 particles and seabed sediment are in the same range than values from the Betsiboka estuary
276 measured in intertidal sediments samples ($-19.8 \text{‰} \pm 1.6\text{‰}$), or in particles from the marine part of
277 the estuary (between -23.6 and -16.9‰) (Ralison et al., 2008), except for sites S1-P, S2-G and S2-P2
278 (Figure 8).

279 Methane was measured on all cores but only detected on three, with concentrations up to $18.3 \mu\text{M}$
280 at S2-P2, $14.1 \mu\text{M}$ at S2-G, and $< 1\mu\text{M}$ at S1-P.

281 5 DISCUSSION

282 5.1 Modern sedimentation along the Madagascar margin

283 5.1.1 Seasonal sediment transfers

284 During most of the wet season, some sediment is transported offshore. The high continental fluxes
285 favor the downslope transfer of sediments, that reach site 1, and to a lesser extent site 2. However,
286 from February to May 2015, sediment fluxes at site 1 were very low despite the relative high rainfall

287 (Figure 4). Longshore currents sometimes deflect the river plumes southwards and may reduce the
288 sediment transport towards the continental slope, as observed on a satellite image of the 16th March
289 2015 (Figure 1C), which corresponds to the period of low sediment fluxes in the slope (Figure 4). The
290 particles recovered during the dry season at S1-P could be due to specific resuspension events,
291 affecting sediment accumulated on the slope. The geochemical composition remains globally similar
292 in the collected particles through the sampling period, except for Mn_{dith} and S, which could evidence
293 that some geochemical processes already altered their concentration before the resuspension
294 happened (manganese oxides reduction and pyrite formation). This functioning scheme could be
295 supported by lower ²¹⁰Pb_{xs} during the dry season (Supplementary material), as particles scavenge the
296 ²¹⁰Pb produced by the decay of ²²⁶Ra in the water column, this concentration decreasing by
297 radioactive decay once sedimented.

298 5.1.2 Long-term sedimentation

299 Past sedimentation rates were similar until the 1950s at both sites, around 0.04-0.06 cm yr⁻¹ (Figures
300 5 and 6), suggesting a moderate offshore sedimentation originated from coastal rivers. A few
301 sedimentation events (anomalies in Ti/Ca ratios) are recorded prior to 1950s at site 2, especially in
302 S2-G, located on a steep slope of a sediment wave. This specific localization favors the preservation
303 of sediment and prevent their erosion, with a physical protection against the currents in a slight
304 depression. Then, sediment cores from site 2, located in different morphologies (pockmarks,
305 slopes...), are subject to the same inputs but do not prevent bottom current erosion. Ti/Ca ratio is
306 typically used as a proxy for comparing marine and terrigenous sediment input, due to the higher
307 content of calcium in marine sediment and higher content of titanium in terrigenous sediment (van
308 der Lubbe et al., 2014). The question is: do these Ti/Ca anomalies recorded in the sedimentary phase
309 came from episodic events? To start answering this question, one might look at the composition of
310 particles collected with the sediment trap at S1-P, as the Chedza storm was sampled. Their
311 composition do not change through time, and anomalies in the Ti/Ca ratio were not recorded with

312 constant values around 0.047. The tropical storm Chedza formed from the intertropical convergence
313 zone and intensified in the Mozambique channel before quickly crossing the Island from West to
314 East. During this storm, heavy rains occurred in central Madagascar and induced intense flooding of
315 the Betsiboka and Mahavavy Rivers. If the Chedza storm is not one of the extreme events
316 responsible for anomalies in Ti/Ca ratios, can we elaborate on its contribution over the annual
317 sedimentation? Particulate fluxes were around $2000 \text{ mg m}^{-2} \text{ d}^{-1}$ during the 3 months preceding the
318 storm, and increased by 1/3 during the event (Figure 4). The mass accumulation rate in January 2015
319 (cumulated on 2 sampling periods including the tropical storm Chedza) was $900 \text{ g m}^{-2} \text{ yr}^{-1}$. If we
320 compare this value to the MAR calculated from the $^{210}\text{Pb}_{\text{xs}}$ profile in S1-P of $1381 \text{ g m}^{-2} \text{ yr}^{-1}$ after the
321 1950s, the latter is slightly higher, still in the same order of magnitude. Now if we integrate the entire
322 sampling period for S1-ST, this accumulation rate lowers down to around $285 \text{ g m}^{-2} \text{ yr}^{-1}$, very close to
323 the $320 \text{ g m}^{-2} \text{ yr}^{-1}$ estimated from the $^{210}\text{Pb}_{\text{xs}}$ below 16 cm at S1-P (Figure 5). The tropical storm
324 Chedza can then be considered as an event integrated in the base sedimentation of this area, and is
325 not extreme enough to provide the area with several centimeters of new sediment. The season
326 2014-2015 was a “normal” season with around 50 days of storm or cyclone activity, when
327 exceptional seasons (1970-1971 or 1993-1994) can reach more than 90 days of activities
328 (<http://www.cycloneoi.com/pages/cyclone-ocean-indien/donnees-remarquables.html>).

329 5.1.3 Episodic deposition at S1-P

330 After the 1950s, the sedimentation rate at site 1 drastically increased (Figure 5), contrary to those of
331 site 2 that remained the same than before the 1950s (Figure 6). One event, characterized by a peak
332 in Ti/Ca ratio, was recorded at S2-P2 and probably preserved because of a lower bioturbation activity
333 at this site (Figure 6). Several layers at S1-P (up to 6 as described in figure 5) could represent different
334 depositional conditions. Four of these events (number 1, 3, 4, 5 – Figure 5) present a sharp increase
335 in Ti/Ca ratios (Fontanier et al., 2018), explained by very low Ca contents. These layers, most likely
336 from terrestrial origin, are not very clearly defined in the S and Ti/Al profiles. An increase in OC is

337 generally observed during these deposition events, even if the lower resolution could explain some
338 discrepancies. As there is only evidences for these repetitive episodic events at site 1, the simpler
339 hypothesis would be a better connectivity between sediment sources (i.e Mahavavy outlet) and the
340 slope depositional area. Between 1984 and 2016, images retrieved from Google earth show a
341 difference in the coastline boundaries (Figure 9). The Mahavavy River delta and prodelta moved
342 toward the ocean about 1 km and 2 km, respectively (Figure 9). Our hypothesis is that this modern
343 delta and prodelta progradation favors temporary connections of the river through an incised valley
344 located around 11 km off the delta, to the canyon head (Figure 9 - (SHOM, 2013)). Only a few canyon
345 systems around the world are currently able to collect the sediments and favor focused downslope
346 turbidity flows, in such sea-level highstand mode of the present interglacial (ie. the Congo/Zaire
347 canyon in Africa (Shepard and Emery, 1973), Monterey Canyons in America (Smith et al., 2007; Maier
348 et al., 2018), Cap-Breton canyon (Gaudin et al., 2006; Guiastrennec-Faugas et al., 2020) and the Var
349 canyon in Europe (Khripunoff et al, 2012)). On these narrow shelves, the canyon heads are close
350 enough to the river outlets to be supplied with sediment by longshore drift which can directly
351 cascade down submarine canyon heads by cooling of shelf water masses (Canals et al., 2006). It has
352 also been evidenced that cyclones or stormy conditions can trigger very dense sediment flows, and
353 can be responsible for significant deposits even within a disconnected canyon (Hale et al., 2012; Pope
354 et al., 2017; Guiastrennec-Faugas et al., 2020). As the distance between the Outlet and the preserved
355 incised valley is now reduced, the sediment loads, when intense enough, do not have time to be
356 deflected by the littoral currents, and are thus mostly focused in the shelf incision and ultimately sink
357 in the canyon head. The axial incision of the Mahavavy canyon (Figure 1) evidences modern turbidite
358 flows that could also spread into the surrounding interfluves to reach site 1. Most of the cyclones in
359 this area hit the north-eastern coast and cross the island from east to west (Fitchett and Grab, 2014;
360 Mavume, 2009). During this type of cyclones, torrential rainfall and substantial floods are generated.
361 The high density of lavakas in central Madagascar (Cox et al., 2010) are drained by both watersheds,

362 and this type of event could promote their erosion and lead to a very intense and fast transportation
363 of terrestrial material. The dilution with marine particles could then be less efficient, explaining
364 higher Ti/Ca ratios in some local deposits.

365 5.2 Geochemical evidences of confined past-present methane escapes

366 Sites 1 and 2 are characterized by the presence of pockmarks (Figure 1), dissolved methane
367 measured in the upper 30 cm of the sedimentary column in some cores (supplementary material), as
368 well as the manifestation of gas or fluid expulsion supported by acoustic anomalies detected in the
369 water column (Dupré et al., 2019). Usually, when methane migrates through the sedimentary
370 column, it is partially or totally oxidized by a consortium of anaerobic methanotrophic (ANME) and
371 sulfate-reducing bacteria with sulfate as the terminal electron acceptor (Boetius et al., 2000; Orphan
372 et al., 2001). This reaction, so-called anaerobic oxidation of methane (AOM) produces $\Sigma\text{H}_2\text{S}$ and
373 dissolved inorganic carbon (DIC), which could precipitate to form sulfur minerals (FeS, FeS₂...) and
374 carbonates. The presence of large amount of pyrite can be related to layers in the sediment where
375 AOM takes place (Berner, 1984; Kasten et al., 1998). Sulfate reduction can also occur through organic
376 matter mineralization, especially in the first centimeters of sediment. Nevertheless, in the next
377 discussion section, we tested two proxies to discriminate the sampling cores regarding to
378 past/present methane outflux: 1) the total sulfur (mainly pyrite as revealed by additional XRD
379 measurements) together with reactive iron for past activities, in addition to 2) the $\delta^{13}\text{C}$ of organic
380 matter that reflects more recent activities through the microbial biomass.

381 5.2.1 S/Fe contents: indicators of the past?

382 Sedimentation is a well-known controlling factor for organic matter mineralization and preservation,
383 the latter being enhanced when sediment accumulation rates increase (Blair and Aller, 2012). As
384 sedimentation rates increase, organic material inputs also increase and oxygen is rapidly consumed
385 within a few millimeters of sediment (Glud, 2008). Anoxic processes, and mainly sulfate reduction,
386 then become the major process affecting organic matter mineralization (Henrichs and Reeburgh,

1987; Pastor et al., 2011). Then, sedimentation is a controlling factor for sulfide production and iron oxides inputs, and ultimately pyritization. In deep sea sediments, where sedimentation rates are very low and organic carbon highly degraded, sulfate reduction rates never reach high enough levels to convert all the reactive iron into pyrite, resulting in the preservation of iron oxides (Raiswell and Canfield, 2012). In coastal areas, with shallower water depth and fresher organic carbon compounds, even at very low sedimentation rates and low sulfate reduction rates, sulfide production rates are still higher than the inputs of reactive iron (Canfield, 1989), represented here by the fraction of iron extracted with dithionite (Fe_{dith} = amorphous Fe(II) oxides, crystalline Fe(II) oxides, AVS - (Kostka and Luther III, 1994)) and thus all the iron is pyritized. In the Majunga basin, at both sites 1 and 2, medium sedimentation rates $< 0.07 \text{ cm yr}^{-1}$ and organic carbon available in relatively high concentrations ($> 1\%$) should cause the complete pyritization of the reactive iron. Typical values for reactive iron contents in marine sediment are between 0.6 to 1.5% (Raiswell and Canfield, 2012), but here the red ferruginous/ferralitic soils from Madagascar provide the coastal area with iron rich minerals, leading to reactive iron contents close to 2-3% in surface sediments (0-5 cm), and still $>0.5\%$ deeper in the sediment (20-30 cm) at both sites. These high values promote the local precipitation of pyrite in this area. Figure 7 shows a clear differentiation between cores. Sites inside pockmarks (S1-P, S2-P1 and S2-P2) and the site where gaseous methane in the water column have been detected (S2-G) are distinct from other sites, with large quantities of pyrite produced and concomitant Fe_{dith} consumption (Figure 7). This difference cannot be due to a difference in inputs, as cores taken at site 2 are within close proximity to each other. Even if the present methane degassing activity is not homogenous in these areas, these geochemical indicators provide a clear first overview of the impact of fluids on overall past and on-going geochemical processes.

5.2.2 The influence of episodic extreme deposition events on pyrite formation

As explained in the previous section, a rapid increase in accumulation rates, through episodic events for instance, carrying more organic carbon, and more iron oxo-hydroxides (up to $710 \mu\text{mol g}^{-1}$) may be

412 responsible for an enhancement in organic matter mineralization and pyritization. The large
413 accumulation of total sulfur (mainly authigenic pyrite) in site 1 within the first 16 cm (concomitant to
414 the deposition of sediment enriched in organic carbon (Figure 5)) can only result from the
415 precipitation of the $\Sigma\text{H}_2\text{S}$ produced *in situ* during sulfate reduction. The large amount of iron in
416 ferruginous soil exported during these events supplied the area with excess labile iron that favored
417 pyrite formation. A sulfur budget was estimated from total sulfur measured in the sediment core,
418 minored by a background input estimated at 0.4% from the sediment trap results (Figure 5). The
419 measured porosity (0.78-0.92) and a dry density of 2.7 g cm^{-3} (standard value for mud - (Hamilton,
420 1976)) were used to calculate the amount of pyrite formed throughout the entire sediment core in
421 S1-P, which would represent 3.5% of the total sulfate pool (28 mM in seawater). If we only consider
422 the sulfur peak formed between 0 and 16 cm depth, this would correspond to a period of
423 approximately 65 years from ^{210}Pb data (at the time of sampling in 2015). This would lead to an
424 integrated rate on S1-P of sulfate reduction of around $7.5 \text{ mmolS m}^{-2} \text{ d}^{-1}$. This would be barely visible
425 in the sulfate profile. During organoclastic sulfate reduction 2 mole of carbon are degraded per 1
426 mole of sulfate, and only 1 mole of carbon during AOM per 1 mole of sulfate, this would lead to an
427 integrated consumption of carbon of between 7.5 and $15 \text{ mmolC m}^{-2} \text{ d}^{-1}$. This would mean that if
428 AOM was 100% responsible for the pyrite build-up, methane concentrations could have reached
429 significant values (mM) in the top layer (0-30 cm) and thus high fluxes. Conversely, if organoclastic
430 sulfate reduction was the main process leading to pyrite build-up, then the deposited layers rich in
431 OM would have enhanced sulfate reduction locally. In the end, the labile iron supplied by the events
432 certainly enhanced the rate of pyrite formation, independently of the sulfate reduction process
433 involved.

434 5.2.3 $\delta^{13}\text{C}$ of bulk organic carbon: an indicator of present methane outflux?

435 At the Betsiboka River outlet, $\delta^{13}\text{C}$ signatures of particulate organic carbon were $-19.8 \text{ ‰} \pm 1.6 \text{ ‰}$ in
436 intertidal sediments samples, and ranged between -23.6 and -16.9 ‰ in particles from the marine

437 part of the estuary (Ralison et al., 2008). The trapped particulate matter at site 1 and most sediments
438 fall into this range (Figure 8). Only sediment from S1-P, S2-G and S2-P2 presents more negative
439 values (Figure 8 and supplementary material). At site 2, all cored sites are located within a diameter
440 of 5 kilometers and are therefore likely to receive similar settling particles with similar $\delta^{13}\text{C}$ ratios.
441 The difference in $\delta^{13}\text{C}$ ratios in some cores is then not due to a change in the organic matter source,
442 but most certainly to diagenetic processes within the sedimentary column. To decrease these ratios,
443 up to -30‰ in S2-G, another source of carbon, very depleted in ^{13}C , is needed, such as methane for
444 instance that typically exhibits negative $\delta^{13}\text{C}$ ratios (Whiticar, 1999). Studies have shown the
445 contribution of products from methanotrophs into specific organic compounds and bulk OC, leading
446 to very negative $\delta^{13}\text{C}$ ratios (Freeman et al., 1990; Hinrichs et al., 1999; Hinrichs et al., 2003; Hinrichs
447 et al., 2000). A simple mixing model using two end-members, namely the $\delta^{13}\text{C}$ of originally deposited
448 OC, and the $\delta^{13}\text{C}$ of methane coming from below (and thus a minimum value for associated
449 methanotrophs (feeding on this carbon source) and derived organic products), was used to estimate
450 the potential contribution of these methanotrophs on the $\delta^{13}\text{C}$ of OC at depth. $\delta^{13}\text{C}$ of methane was
451 measured in deeper sediment cores during the campaign PAMELA-MOZ4 (data not presented here)
452 and a value of -82 ‰ can be used as the first end-member. The second end-member is the $\delta^{13}\text{C}$ of
453 OC from trapped particles of -21‰. Then, for the most negative values recorded for $\delta^{13}\text{C}$ of OC of -
454 30‰ in S2-G, the maximum contribution of these products into the organic matter pool can be
455 estimated around 15% (and thus less for heavier isotopic signatures). Then, $\delta^{13}\text{C}$ signatures of OC
456 probably reflect the present abundance of methanotrophs and their by-products in sites with current
457 high active methane escape (S2-G), past high methane escape (S1-P) and current low flux of methane
458 (S2-P2). Compared to the two distinct groups evidenced in Figure 7 using the S/Fe indicators, S2-P1 is
459 not considered to be impacted by methane escape using the $\delta^{13}\text{C}$ signatures of OC. ANMEs have
460 been detected on S1-P, S2-P2 and S2-G but not on S2-P1 (Odobel, 2017), which, associated with the

461 clear difference of facies between those cores (Figure 6), underlines the high heterogeneity in this
462 system and the complexity of choosing the right proxies to understand past and present processes.

463 6 CONCLUSIONS

464 This study provides a clear first insight into the sedimentation patterns along the continental slope
465 off the Betsiboka and Mahavavy Rivers on the NW coast of Madagascar. During the wet season,
466 particles from the two rivers are transported offshore and provide the continental slope with
467 terrestrial material, leading to a mass accumulation rate ranging from 230-249 g m⁻² yr⁻¹ off the
468 Betsiboka River, to 320 g m⁻² yr⁻¹ off the Mahavavy River, calculated from ²¹⁰Pb^{xs} in surficial sediment
469 cores. The sediment trap off the Mahavavy River on the slope falls well within this range with 285 g
470 m⁻² yr⁻¹ during 2014-2015. Yet since the 1950s, a higher mass accumulation rate of 1381 g m⁻² yr⁻¹ is
471 recorded in sediment cores off the Mahavavy River. This increase is probably due to temporal
472 reconnection of the river with the head of the canyon during extreme events, as supported by a
473 significant progradation of the delta and prodelta of around 1 and 2 km respectively, and a well
474 incised canyon reflecting recent turbidite activity. We suspect that these pulsed deposits, rich in
475 organic carbon and iron oxides, enhance some geochemical processes and ultimately the pyritization
476 process. More globally, the amount of pyrite on the continental slope seems to be driven by
477 methane upward migration, locally enhancing sulfate reduction and thus the amount of free sulfide
478 produced. This influence is also supported in the δ¹³C of bulk organic carbon, with a significant
479 contribution of methanotrophs to the organic carbon pool.

480 **Acknowledgements**

481 We would like to thank the captain and crew members of the R/V L'Atalante and RV Pourquoi pas?,
482 the chiefs scientist (Karine Olu, Stéphanie Dupré, Gwenael Jouet and Eric Deville) and all scientists
483 who took part in PAMELA-MOZ1 and PAMELA-MOZ4 cruises. Thanks also go to Céline Liorzou at the
484 PSO/IUEM (France) for technical support with the ICP-AES, Philippe Noël and Jean-Pierre Brulport

485 from Ifremer for technical help with mooring and sampling, Alexis Khripounoff from Ifremer for
486 current data, Bernard Dennielou, Angélique Roubi and Jérémie Gouriou from Ifremer for semi-
487 quantitative XRF, Alison Chalm from Ifremer for english editing and Julie Tourolle from Ifremer for
488 help with the SIG. We would like to thank the two anonymous reviewers for their constructive
489 comments that helped improve this manuscript. This study was completed as part of the PAMELA
490 project (“Passive Margin Exploration Laboratories”) funded by TOTAL and Ifremer. The authors
491 acknowledge the use of imagery from the NASA Worldview application
492 (<https://worldview.earthdata.nasa.gov>), part of the NASA Earth Observing System Data and
493 Information System (EOSDIS).

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632 Table 1: Position of sampling. MLP stands for mooring and MTB for sediment core.

Site	Position	Latitude (S)	Longitude (E)	Depth (m)	Cruise label	Designation	Date (dd/mm/yy)	Depth (m)
Site 1	above the pockmark (Sediment Trap)	15°31.160	45°42.943	781	MLP1	S1-ST	26/10/14 – 15/11/15	781
	inside the Pockmark	15 31.148	45 42.931	789	MOZ1-MTB6/7	S1-P	07/10/2014	789
Site 2	inside the Pockmark	15 21.815	45 57.648	740	MOZ1-MTB8/9	S2-P1	09/10/2014	740
		15 21.812	45 57.628	735	MOZ4-MTB1	S2-P2	13/11/2015	735
	at the Pockmark Edge	15 21.695	45 57.388	757	MOZ1-MTB3	S2-PE1	06/10/2014	757
		15 21.685	45 57.378	754	MOZ4-MTB2	S2-PE2	14/11/2015	754
	OUTside the pockmarks area	15 22.047	45 59.210	529	MOZ1-MTB1/2	S2-OUT	04/10/2014	529
	On observed Gas escape	15 22.230	45 57.110	762	MOZ4-MTB3/4	S2-G	14/11/2015	762

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FIGURE CAPTIONS

637 *Figure 1: (A) The NW Madagascar margin and the Mahavavy and Betsiboka watersheds. The two main sites (S1, S2) are*
 638 *indicated by a star. (B) Detailed bathymetry of the sampled area and location of the sampling sites (C) Satellite image*
 639 *(MODIS/Aqua corrected reflectance) of 16 March 2015, obtained from the NASA Worldview application, showing how river*
 640 *plumes are deflected towards the north by alongshore currents and sediment in suspension is carried from the northern part*
 641 *towards the south by longshore currents.*

642 *Figure 2: (A) Temperature and speed time series at 30 m above the seafloor at S1. The light blue curves represent the raw*
 643 *data and the dark blue lines averaged values every 12 h. (B) Rose diagram of current direction and speed at 30 m above the*
 644 *seafloor at S1.*

645 *Figure 3: Spectra of kinetic energy based current velocity (A) and temperature (B) at S1 (blue line) and 98% confidence level*
 646 *(red lines). M2 is the semi-diurnal tide (12.42 h) and M4 is the quadridiurnal tidal constituent (6.21 h).*

647 *Figure 4: Geochemical data from suspended particulate matter in Site 1 collected with a sediment trap positioned 10 m*
 648 *above the sediment surface. TMF stands for total mass flux. The pink shaded bar represents the interval in which the tropical*
 649 *storm Chedza happened.*

650 *Figure 5 Geochemical data and photograph of sediment core from S1-P. SR: Sediment accumulation Rates. OC: Organic*
 651 *Carbon content. The dashed line represents the limit around 1950s. The top part of the core has been divided in 6 layers*
 652 *which could represent different sedimentation events*

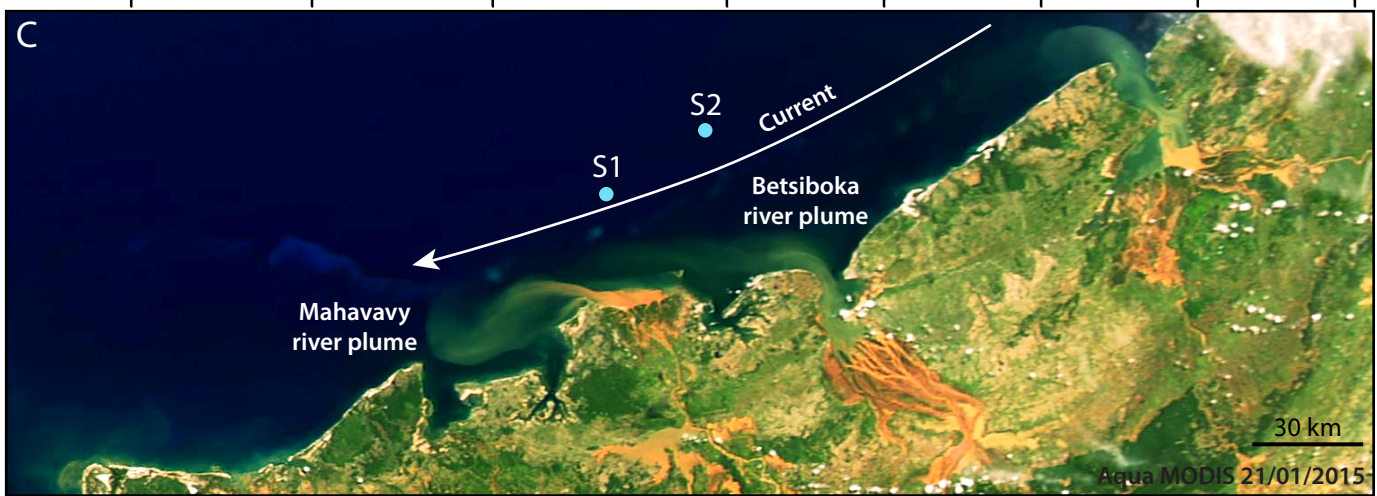
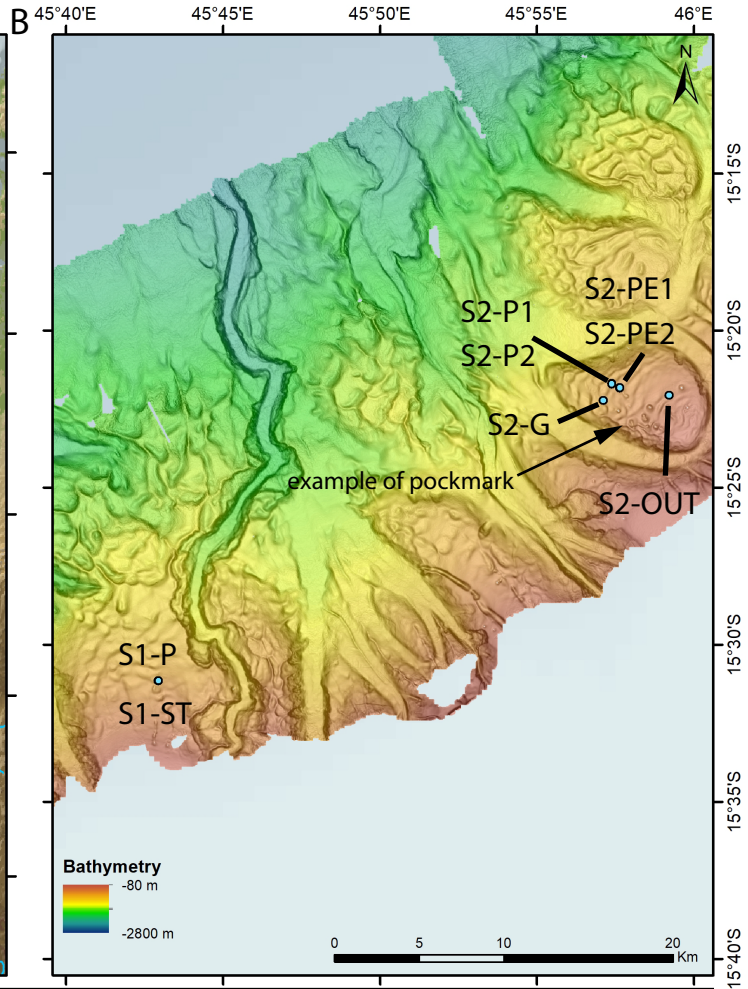
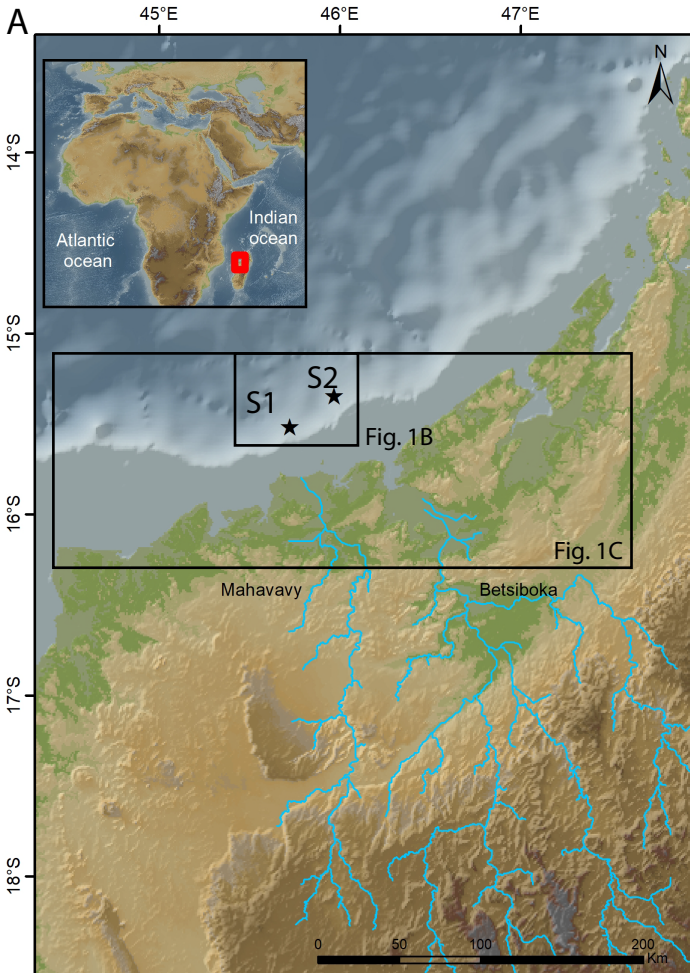
653 *Figure 6: Geochemical data and photographs of sediment cores from Site 2. SR stands for the Sediment accumulation Rates.*
 654 *The dashed line represents the limit around 1950s. The orange and gray shaded bars represents the main visual sediment*
 655 *layer at the top of each core*

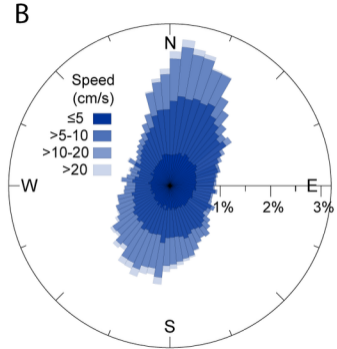
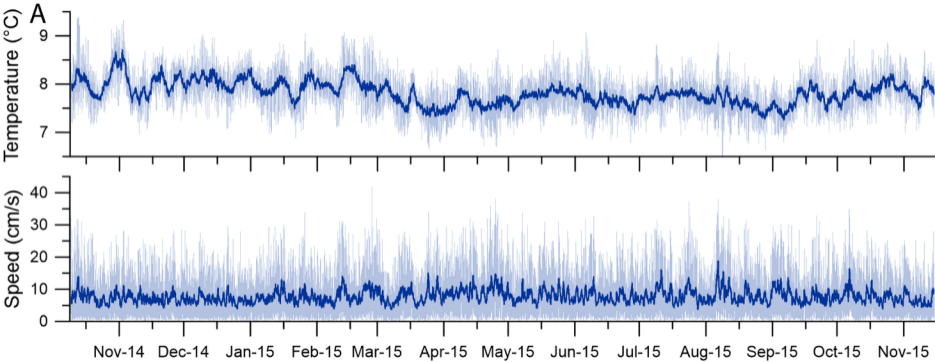
656 *Figure 7: Ternary plot of organic carbon (OC), sulfur content (S) and crystalline iron oxi-hydroxides (Fe_{dith}) for multitube cores*
 657 *(MTB) and particles from the sediment trap*

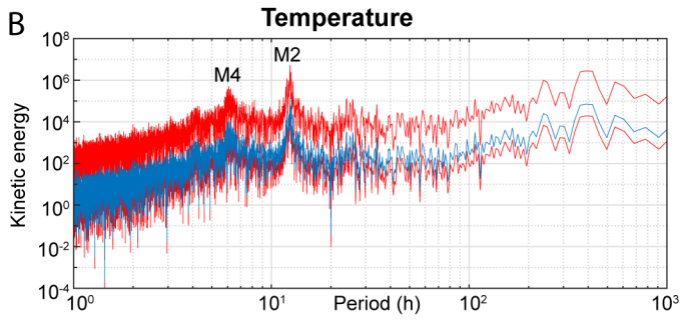
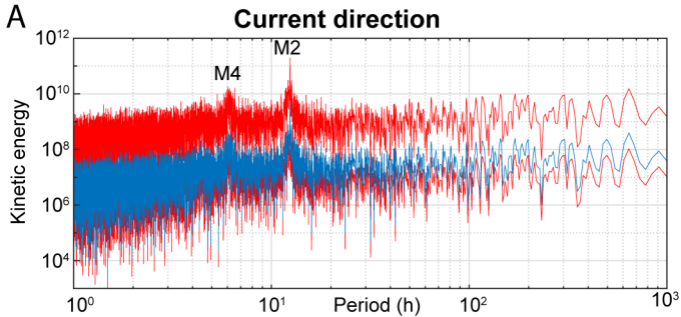
658 *Figure 8: Summary of $\delta^{13}C$ values on particulate organic matter from the moored sediment trap and the sediment cores.*
 659 *Data are shown as boxplots; lines represents the median and 75% interquartile ranges, error bars show the 90% intervals,*
 660 *and extremes are shown as black circles. For comparison, the typical range of $\delta^{13}C$ values for C3 and C4 derived organic*
 661 *matter are indicated as between dotted lines, and the range of $\delta^{13}C$ values of particulate organic carbon and intertidal*
 662 *sediments (75% interquartile ranges) (from (Ralison et al., 2008) as a hatched area*

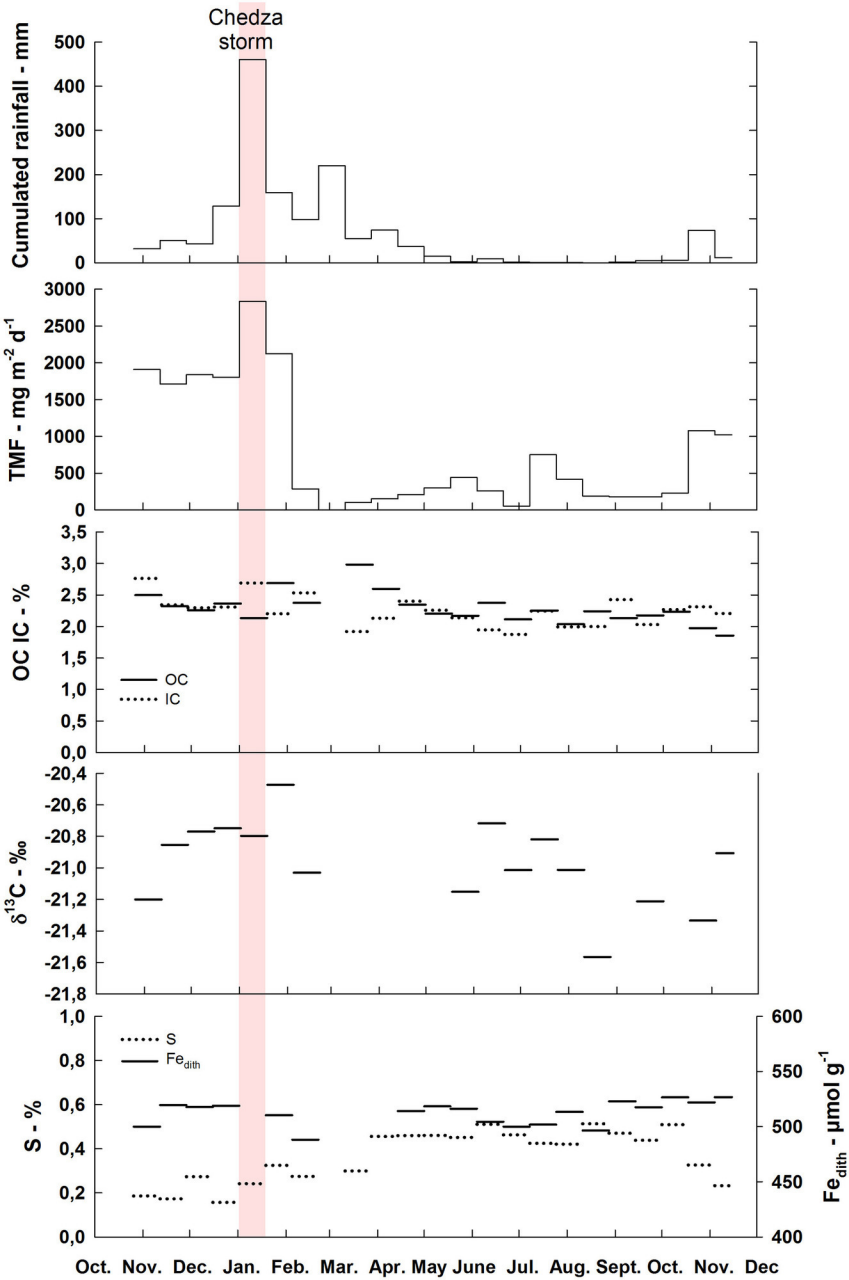
663 *Figure 9: Satellite image of the Mahavavy delta and prodelta in 2016. White dashed lines represent the location of the delta*
664 *and prodelta in 1984. The red dashed line stands for particles export through the incised valley to the head of the canyon*
665 *during extreme events. Images from google maps*

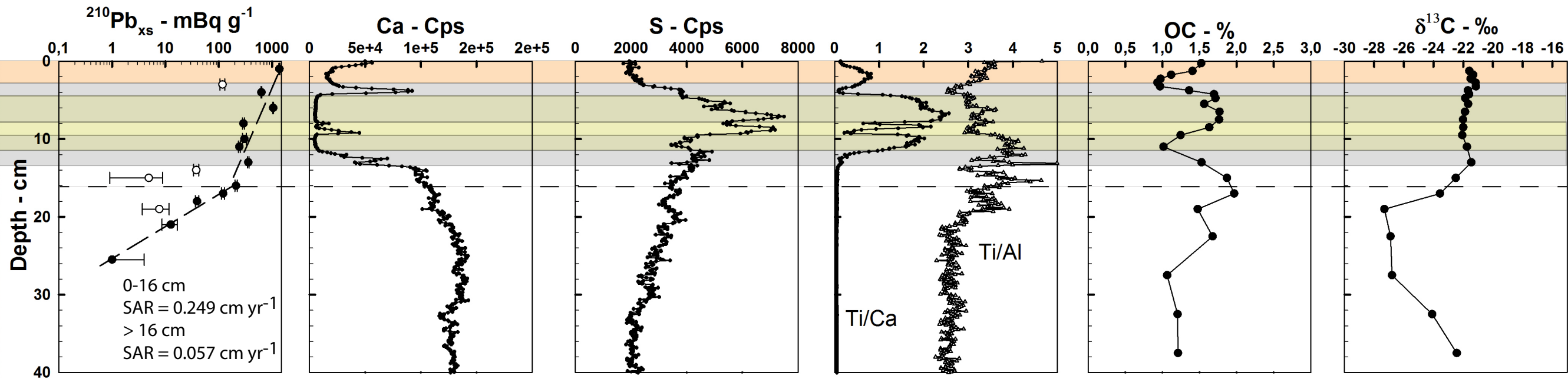
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S1-P

- S1-ST
- S2-G
- S2-PE2
- S1-P
- S2-OUT
- S2-P1
- S2-PE1
- S2-P2

