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1	Modern sedimentation and geochemical imprints in sediments from the NW Madagascar margin
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13

14 Abstract

The NW Madagascar continental margin receives high loads of terrigenous particulate organic matter 15 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 alongslope 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 δ^{13} 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 δ^{13} C signature of organic 46 matter through its mineralisation (M^carthur et al., 1992), the isotopic composition of several 47 elements in foraminifera tests through the dissolution/reprecipitation of carbonates phases (Edgar et al., 2015), or the magnetic properties of sediment through the dissolution and precipitation of iron 48 phases (Larrasoana et al., 2009). In environments where diagenetic processes are highly diverse like 49 50 in cold seeps areas or deltaic environments, it can be intricate to quantify their relative impact on paleosignatures. Thus, studies aiming at reconstructing paleosignals or determining the source of
 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 continental slope (Acoustic anomalies detected in the water column - Dupré et al., 2019). The 55 sediment dynamics and geochemical signatures on the continental slope under the influence of the 56 57 rivers input have been monitored using one mooring with a sediment trap, temperature and current velocity measurements, and interface sediment cores. The objectives of this work were to 1) give a 58 59 first insight on the modern sedimentary patterns along this slope and 2) decipher the relative impact of the sedimentation versus methane escape on geochemical signatures (Ti/Ca, Ti/Al, δ^{13} C of organic 60 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 basin). This basin partly covers onshore and offshore areas (Tari et al., 2004). In this paper, we only 65 66 discuss data from the offshore Majunga basin. The Betsiboka River is the largest river in Madagascar with a length of 605 km and a watershed of 49,000 km² (Randrianarijaona, 1983). Spectacular gullies, 67 68 named lavakas by the Malagasy people, are widespread in Madagascar and are especially dense in the highlands of central Madagascar around the Betsiboka watershed (Cox et al., 2010). The river 69 supplies the coastal area with sediment eroded from ferruginous and ferralitic soils of the Majunga 70 basin and of the lavakas (Ramifehiarivo et al., 2017). Fluvial discharges varies from 35 m³ s⁻¹ during a 71 very dry season to 4,500 m³ s⁻¹ during wet periods, with extreme values during flood reaching > 72 73 25,000 m³ s⁻¹ (Berthois and Crosnier, 1966; Chaperon et al., 1993). From measurements in surface waters, solid transport was estimated in the 1960s at about 1660 t yr⁻¹, which corresponds to 1.1 mm 74

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

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

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

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

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

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

123 3.2 Samples treatment

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

Interface sediments were collected with a multicorer equipped with eight Plexiglas tubes (96 mm 131 132 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 depth and then every 2 cm downcore. Each layer was immediately frozen (-80 °C) on board in a pre-135 136 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 freezedried weight and assuming a sediment bulk density of 2.7 g cm⁻³ (standard value for mud - (Hamilton, 138 139 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. One aliquot (2-4 mL) was stored in a 10 mL glass vial with 15 µl of HgCl₂, and sealed for further 141 142 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 using a pre-cut syringe, directly transferred into a 20 mL glass vial with 5 mL 1M NaOH, sealed and
stored head-down at 4°C before methane measurement at the laboratory.

146 3.3 Analyses

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

151 Crystalline oxi-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 δ^{13} C 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 carbonate (LSVEC). An acetalinide standard (Costech) was inserted every ten samples to correct for 161 162 the drift, with a measured value of $-33.50 \pm 0.02\%$ (6 replicates). Precision was typically within 0.03 163 ‰ for a triplicate.

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

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

The bulk sediment semi-quantitative geochemical composition was measured with an Avaatech X-ray fluorescence (XRF) core scanner on half sections. Measurements were carried out every one millimeter applying a basic voltage of 10 and 30 kV to estimate elemental relative concentrations (aluminum (AI), sulfur (S), calcium (Ca) and titanium (Ti)). Results are given in counts per second (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. TiO₂, CaO, Al₂O3, Fe₂O₃). Half a gram of dried 180 sediment was fused at 1050°C with 9 g of fusion flux (90 % lithium tetraborate (LiB₄O₇)/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 reference materials Project" (CCRMP - FER-1, FER-2, WMG-1), the "Geological Survey of Japan" (GSJ -184 185 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 third run in case of divergence. The relative error on major elements (up to 1%) is below 5%. 187 188 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 of 7.8 cm s⁻¹ (Figure 2). Daily means ranged between 4.3 and 15.5 cm s⁻¹. Main current directions 198 199 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 °C with an averaged value of 7.8 °C (Figure 2A). A 201 slight decrease in temperature from 8.0 to 7.7 °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 mg m⁻² 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

cumulated TMF remains correct. TMF varied between 51 and 750 mg m⁻² d⁻¹ during the dry season. 215 During the wet season, the daily averaged particulate mass export was multiplied by 2.8 compared to 216 217 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 higher values during the dry season. δ^{13} C of organic carbon varied from -21.6 to -20.6 ‰ (average -220 21.0 ‰) (Figure 4). ²¹⁰Pb_{xs} activities present also a limited variability, between 1794 and 2256 mBq g⁻ 221 222 ¹, 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 μ mol g⁻¹ during the wet season, with a marked increase of concentration during the wet season. Fe_{dith} ranged between 488 and 527 µmol g⁻¹ with no clear pattern during the 224 225 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

²¹⁰Pb_{xs} profiles were determined on 3 cores at site 2 (S2-PE1; S2-PE2; S2-G - Figure 6). ²¹⁰Pb_{xs} activities 241 are rather comparable in the top sediment, ranging between 548 and 845 mBq g⁻¹, and present an 242 exponential decrease with depth to reach negligible level at about 10 cm. The limited variability in 243 244 sedimentation rates among the different cores at site 2, regardless of their position, results in very 245 similar ²¹⁰Pb_{xs} profiles and thus similar SAR and MAR. At the edge of a pockmark (S2-PE1 and S2-PE2), the SAR is 0.052 \pm 0.004 cm yr⁻¹ and 0.039 \pm 0.005 cm yr⁻¹ while the MAR is 249 g m⁻² yr⁻¹ and 230 g m⁻² 246 247 2 yr⁻¹, respectively. On a gas escape at S2-G, the SAR is 0.059 ± 0.006 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 three sites, indicating a rather homogenous particle deposition on the area. The profile of ²¹⁰Pb_{xs} is 249 250 completely different at site 1 (Figure 5): first, the top ²¹⁰Pb_{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 ²¹⁰Pb_{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 16 cm, the SAR is 0.249 \pm 0.054 cm yr⁻¹ (Figure 5), which corresponds to a mean MAR of 1381 g m⁻² yr⁻¹ 254 255 ¹ (Fontanier et al., 2018). Deeper in the sediment (> 16 cm; before 1950s), the SAR is much lower (0.057 ± 0.007) cm yr⁻¹ (Figure 5), which corresponds to a MAR of 320 g m⁻² yr⁻¹. In addition to the 256 257 main pattern, ²¹⁰Pb_{xs} activities present low activities at 2-3, 13-14 and 19 cm (Figure 5), that could be the result of a temporary input of low ²¹⁰Pb_{xs} particles. 258

259 4.2.3 Geochemical data

Sedimentary OC concentrations ranged between 0.8% and 2.4% (Figures 5 and 6), IC concentrations between 0.3% and 5.7%, and total S between 0.07% and 3.7%. S1-P recorded the highest values of OC and S, and the lowest values of IC. In all cores, Mn_{dith} ranged between 0.5 and 17.9 µmol g⁻¹ and Fe_{dith} between 95.7 and 710.8 µmol g⁻¹.

Ca values measured by semi-quantitative usually ranged between 1.10⁵ and 2.10⁵ Cps, with lower values at the top of cores from sites S2-P2, S2-G and S1-P. S values measured by semi-quantitative 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 in average around 0.04-0.06, with high variations at the top of S2-P2, S2-G and S1-P. Ti/Al ranged between 2 and 5 in all sites.

- A ternary plot between OC, S and Fe_{dith} separate 2 distinct groups of sites (Figure 7): the first group with a higher relative proportion of total S and lower Fe_{dith} contribution (S2-G, S2-P1, S2-P2 and S1-P), the second group with inverse proportions of S and Fe_{dith} (S2-PE1, S2-PE2, S2-OUT and S1-ST).
- δ^{13} C of organic carbon varied between -30.0‰ and -23.5‰ in S2-G (Figure 6) and between -27.3‰ and -21.1‰ in S1-P (Figure 5), and was more stable in other cores between -23.8‰ and -19.9‰ (Figure 6). Ratios measured in trapped particles were -21.0 ± 0.3 ‰. Overall, ratios in trapped particles and seabed sediment are in the same range than values from the Betsiboka estuary measured in intertidal sediments samples (-19.8 ‰ ± 1.6‰), or in particles from the marine part of the estuary (between -23.6 and -16.9 ‰) (Ralison et al., 2008), except for sites S1-P, S2-G and S2-P2 (Figure 8).
- 279 Methane was measured on all cores but only detected on three, with concentrations up to 18.3 μ M 280 at S2-P2, 14.1 μ M at S2-G, and < 1 μ M at S1-P.

281 5 DISCUSSION

- 282 5.1 Modern sedimentation along the Madagascar margin
- 283 5.1.1 Seasonal sediment transfers

During most of the wet season, some sediment is transported offshore. The high continental fluxes favor the downslope transfer of sediments, that reach site 1, and to a lesser extent site 2. However, 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 sediment transport towards the continental slope, as observed on a satellite image of the 16th March 288 289 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 ²¹⁰Pb produced by the decay of ²²⁶Ra in the water column, this concentration decreasing by 296 297 radioactive decay once sedimented.

298 5.1.2 Long-term sedimentation

Past sedimentation rates were similar until the 1950s at both sites, around 0.04-0.06 cm yr⁻¹ (Figures 299 300 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 particles collected with the sediment trap at S1-P, as the Chedza storm was sampled. Their 310 311 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 zone and intensified in the Mozambique channel before quickly crossing the Island from West to 313 314 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 in not one of the extreme events 316 responsible for anomalies in Ti/Ca ratios, can we elaborate on its contribution over the annual sedimentation? Particulate fluxes were around 2000 mg m⁻² d⁻¹ during the 3 months preceding the 317 318 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 g m⁻² yr⁻¹. If we compare this value to the MAR calculated from the ²¹⁰Pb_{xs} profile in S1-P of 1381 g m⁻² yr⁻¹ after the 320 321 1950s, the latter is slighly higher, still in the same order of magnitude. Now if we integrate the entire sampling period for S1-ST, this accumulation rate lowers down to around 285 g m⁻² yr⁻¹, very close to 322 the 320 g m⁻² yr⁻¹ estimated from the $^{210}Pb_{xs}$ below 16 cm at S1-P (Figure 5). The tropical storm 323 324 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

After the 1950s, the sedimentation rate at site 1 drastically increased (Figure 5), contrary to those of site 2 that remained the same than before the 1950s (Figure 6). One event, characterized by a peak in Ti/Ca ratio, was recorded at S2-P2 and probably preserved because of a lower bioturbation activity at this site (Figure 6). Several layers at S1-P (up to 6 as described in figure 5) could represent different depositional conditions. Four of these events (number 1, 3, 4, 5 – Figure 5) present a sharp increase in Ti/Ca ratios (Fontanier et al., 2018), explained by very low Ca contents. These layers, most likely 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 canyon in Africa (Shepard and Emery, 1973), Monterey Canyons in America (Smith et al., 2007; Maier 347 348 et al., 2018), Cap-Breton canyon (Gaudin et al., 2006; Guiastrennec-Faugas et al., 2020) and the Var 349 canyon in Europe (Khripounoff 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,

and this type of event could promote their erosion and lead to a very intense and fast transportation
of terrestrial material. The dilution with marine particles could then be less efficient, explaining
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 well as the manifestation of gas or fluid expulsion supported by acoustic anomalies detected in the 368 369 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 ΣH_2S 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 δ^{13} 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?

Sedimentation is a well-known controlling factor for organic matter mineralization and preservation, the latter being enhanced when sediment accumulation rates increase (Blair and Aller, 2012). As sedimentation rates increase, organic material inputs also increase and oxygen is rapidly consumed within a few millimeters of sediment (Glud, 2008). Anoxic processes, and mainly sulfate reduction, then become the major process affecting organic matter mineralization (Henrichs and Reeburgh, 387 1987; Pastor et al., 2011). Then, sedimentation is a controlling factor for sulfide production and iron 388 oxides inputs, and ultimately pyritization. In deep sea sediments, where sedimentation rates are very 389 low and organic carbon highly degraded, sulfate reduction rates never reach high enough levels to 390 convert all the reactive iron into pyrite, resulting in the preservation of iron oxides (Raiswell and 391 Canfield, 2012). In coastal areas, with shallower water depth and fresher organic carbon compounds, 392 even at very low sedimentation rates and low sulfate reduction rates, sulfide production rates are 393 still higher than the inputs of reactive iron (Canfield, 1989), represented here by the fraction of iron 394 extracted with dithionite (Fe_{dith} = amorphous FeIII oxides, crystalline FeIII oxides, AVS - (Kostka and 395 Luther III, 1994)) and thus all the iron is pyritized. In the Majunga basin, at both sites 1 and 2, 396 medium sedimentation rates < 0.07 cm yr⁻¹ and organic carbon available in relatively high 397 concentrations (> 1%) should cause the complete pyritization of the reactive iron. Typical values for 398 reactive iron contents in marine sediment are between 0.6 to 1.5% (Raiswell and Canfield, 2012), but 399 here the red ferriginous/ferralitic soils from Madagascar provide the coastal area with iron rich 400 minerals, leading to reactive iron contents close to 2-3% in surface sediments (0-5 cm), and still 401 >0.5% deeper in the sediment (20-30 cm) at both sites. These high values promote the local 402 precipitation of pyrite in this area. Figure 7 shows a clear differentiation between cores. Sites inside 403 pockmarks (S1-P, S2-P1 and S2-P2) and the site where gaseous methane in the water column have 404 been detected (S2-G) are distinct from other sites, with large quantities of pyrite produced and 405 concomitant Fedith consumption (Figure 7). This difference cannot be due to a difference in inputs, as 406 cores taken at site 2 are within close proximity to each other. Even if the present methane degassing 407 activity is not homogenous in these areas, these geochemical indicators provide a clear first overview 408 of the impact of fluids on overall past and on-going geochemical processes.

409 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 oxi-hydroxides (up to 710 μmol 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 ΣH_2S 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 measured porosity (0.78-0.92) and a dry density of 2.7 g cm⁻³ (standard value for mud - (Hamilton, 419 420 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 approximately 65 years from ²¹⁰Pb data (at the time of sampling in 2015). This would lead to an 423 integrated rate on S1-P of sulfate reduction of around 7.5 mmolS m⁻² d⁻¹. This would be barely visible 424 425 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 integrated consumption of carbon of between 7.5 and 15 mmolC m⁻² d⁻¹. This would mean that if 427 428 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 δ^{13} C of bulk organic carbon: an indicator of present methane outflux?

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

part of the estuary (Ralison et al., 2008). The trapped particulate matter at site 1 and most sediments 437 fall into this range (Figure 8). Only sediment from S1-P, S2-G and S2-P2 presents more negative 438 values (Figure 8 and supplementary material). At site 2, all cored sites are located within a diameter 439 440 of 5 kilometers and are therefore likely to receive similar settling particles with similar δ^{13} C ratios. 441 The difference in δ^{13} 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 ¹³C, is needed, such as methane for 444 instance that typically exhibits negative δ^{13} 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 δ^{13} 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 δ^{13} C of originally deposited OC, and the δ^{13} C of methane coming from below (and thus a minimum value for associated 448 449 methanotrophs (feeding on this carbon source) and derived organic products), was used to estimate 450 the potential contribution of these methanotrophs on the δ^{13} C of OC at depth. δ^{13} C of methane was 451 measured in deeper sediment cores during the campaign PAMELA-MOZ4 (data not presented here) and a value of -82 % can be used as the first end-member. The second end-member is the δ^{13} C of 452 453 OC from trapped particles of -21‰. Then, for the most negative values recorded for δ^{13} C of OC of -454 30‰ in S2-G, the maximum contribution of these products into the organic matter pool can be estimated around 15% (and thus less for heavier isotopic signatures). Then, δ^{13} C signatures of OC 455 456 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 not considered to be impacted by methane escape using the δ^{13} C signatures of OC. ANMEs have 459 been detected on S1-P, S2-P2 and S2-G but not on S2-P1 (Odobel, 2017), which, associated with the 460

461 clear difference of facies between those cores (Figure 6), underlines the high heterogeneity in this462 system and the complexity of choosing the right proxies to understand past and present processes.

463 6 CONCLUSIONS

This study provides a clear first insight into the sedimentation patterns along the continental slope 464 465 off the Betsiboka and Mahavavy Rivers on the NW coast of Madagascar. During the wet season, particles from the two rivers are transported offshore and provide the continental slope with 466 terrestrial material, leading to a mass accumulation rate ranging from 230-249 g m⁻² yr⁻¹ off the 467 Betsiboka River, to 320 g m⁻² yr⁻¹ off the Mahavavy River, calculated from ²¹⁰Pb^{xs} in surficial sediment 468 cores. The sediment trap off the Mahavavy River on the slope falls well within this range with 285 g 469 m⁻² yr⁻¹ during 2014-2015. Yet since the 1950s, a higher mass accumulation rate of 1381 g m⁻² yr⁻¹ is 470 471 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 produced. This influence is also supported in the δ^{13} C of bulk organic carbon, with a significant 478 479 contribution of methanotrophs to the organic carbon pool.

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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 (S ediment T rap)	15°31.160	45°42.943	781	MLP1	S1-ST	26/10/14 – 15/11/15	781
	inside the P ockmark	15 31.148	45 42.931	789	MOZ1-MTB6/7	S1-P	07/10/2014	789
	inside the P ockmark	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
Site 2		15 21.685	45 57.378	754	MOZ4-MTB2	S2-PE2	14/11/2015	754
	OUT side the pockmarks area	15 22.047	45 59.210	529	MOZ1-MTB1/2	S2-OUT	04/10/2014	529
	On observed G as escape	15 22.230	45 57.110	762	MOZ4-MTB3/4	\$2-G	14/11/2015	762

FIGURE CAPTIONS

- Figure 1: (A) The NW Madagascar margin and the Mahavavy and Betsiboka watersheds. The two main sites (S1, S2) are
 indicated by a star. (B) Detailed bathymetry of the sampled area and location of the sampling sites (C) Satellite image
 (MODIS/Aqua corrected reflectance) of 16 March 2015, obtained from the NASA Worldview application, showing how river
 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.
- Figure 2: (A) Temperature and speed time series at 30 m above the seafloor at S1. The light blue curves represent the raw
 data and the dark blue lines averaged values every 12 h. (B) Rose diagram of current direction and speed at 30 m above the
 seafloor at S1.
- Figure 3: Spectra of kinetic energy based current velocity (A) and temperature (B) at S1 (blue line) and 98% confidence level
 (red lines). M2 is the semi-diurnal tide (12.42 h) and M4 is the quatridiurnal tidal constituent (6.21 h).
- Figure 4: Geochemical data from suspended particulate matter in Site 1 collected with a sediment trap positioned 10 m
 above the sediment surface. TMF stands for total mass flux. The pink shaded bar represents the interval in which the tropical
 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.
- The dashed line represents the limit around 1950s. The orange and gray shaded bars represents the main visual sediment
 layer at the top of each core
- Figure 7: Ternary plot of organic carbon (OC), sulfur content (S) and crystalline iron oxi-hydroxides (Fe_{dith}) for multitube cores
 (MTB) and particles from the sediment trap
- **658** Figure 8: Summary of δ^{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,
- 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
- 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









Oct. Nov. Dec. Jan. Feb. Mar. Apr. May June Jul. Aug. Sept. Oct. Nov. Dec









