

Siliciclastic sediment volumes and rates of the North Pyrenean Retro-Foreland Basin

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| 1 | SILICICLASTIC SEDIMENT VOLUMES AND RATES OF THE NORTH PYRENEAN RETRO- |
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| 2 | FORELAND BASIN |
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36 growth and decay of a mountain belt during orogenic and post-orogenic phases have been 37 largely studied thanks to thermochronological and structural studies. The sedimentary 38 response of the orogenic phases in the preserved sediments of the surrounding basins is well 39 known in terms of sedimentary filling patterns and architecture, but much less better 40 quantified.

Here, we performed a measurement of the siliciclastic sediment volumes of the Pyrenean retro-foreland basin – the Aquitaine Basin and the Bay of Biscay during Cenozoic times – for a better understanding of the erosion and the sediment transfer and deposition during the convergence (syn-orogenic) to post-convergence (post-orogenic) periods of the Pyrenees Mountain. The measurement of the compacted siliciclastic sediment is based on sediment 46 thickness (isopach) maps of known lithologies, derived from the interpretation of 40 000 kms47 of seismic profiles.

Thanks to the siliciclastic sediment volumes quantification and a well-known retro-foreland
basin tectono-sedimentary evolution, we bring quantitative results as:

50 (1) The amount of preserved sediments is of 51500 ± 16800 km³ for the Cenozoic.

- 51 (2) The siliciclastic sediment rate curve during Cenozoic shows two major increase around
- 52 26.0 Ma and 2.5 Ma. The 26.0 Ma increase is clearly related to the erosion of the 53 Pyrenees of tectonic origin. The major 2.5 Ma one would be mainly related to a climatic
- 54 forcing.
- (3) The mass balance between the Aquitaine Platform and the deepest domains change
 through time in favour of the deep domain. This might be explained by the ratio
 between subsidence that created accommodation space and the sediments feed by
- 58 the mountain belt and stored in the Aquitaine Platform.
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60 Keywords: accumulation rates, retro-foreland, Pyrenees, Aquitaine Basin, Bay of Biscay,
61 Cenozoic

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74 1. Introduction

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The growth and decay of mountain belts reliefs result from feedbacks between 76 77 convergence rates, surface processes (Beaumont et al., 1992; Willett, 1999) and isostatic 78 response (Molnar and Lyon-Caen, 1988; Watts, 2001). The topography of a mountain belt is 79 first built by continental crust thickening during convergence, generating loading on the 80 lithosphere and its downward deflection at the origin of the surrounding foreland basins and 81 their vertical motion (Beaumont, 1981; Allen and Allen, 2013). Topographic evolution of the 82 mountain belt is also influenced by climate which can modify the erosion by surface processes 83 (e.g. Whipple, 1999). Finally, the base level variations in the foreland basins may play a 84 dominant role, on piedmont dynamics, in preserving reliefs (base level rise with emplacement 85 of endorheism, Babault et al., 2005) or enhancing its incision (base level fall by major capture 86 for instance, see Burbank and Anderson, 2009)

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The estimation of denudation, i.e. the amount of eroded rocks, through time and its controls by lithospheric deformation and/or climate (precipitation) are of primary importance for understanding (1) the topographic growth and decay of mountain belts, but also (2) the volumes and lithologies of the sediments produced and transferred to the surrounding basins. The quantification of the denudation of a mountain belt is mainly based on low temperature chronology (e.g. fission track and (U-Th)/He on apatite and zircons – ZFT, AFT, ZHe, AHe) that provides a cooling history classically converted into exhumation with using thermal numerical
models (Braun, 2003 for instance, Ketcham, 2005; Gallagher et al., 2009). The sediment
production (volumes, petrography and grain-size) was performed on several foreland basins
(Molnar and England, 1990; Einsele, 2000; Najman and Garzanti, 2000; Goodbred and Kuelh,
2000; Schlunegger et al., 2001; Garzanti, 2019). In the Pyrenean domain, the P. Allens's group
has carried out numerous studies on these thematic, focus on the southern side (e.g. Michael
et al., 2013, 2014 a,b; Armitage et al., 2015).

101 The quantification of the siliciclastic sediment volume (sink) in foreland basins was first 102 performed at mountain-scale in the Alps (England, 1981, Kuhlemann, 2000; Kuhlemann et al., 103 2002), but most of them concerns the latest deposition stages found in the ultimate sink: the 104 passive margin laterally connected to the foreland basins. This is the case for Asian mountains 105 (Métivier et al., 1999; Clift et al., 2006) or Rocky Mountains and the margins of the Gulf of 106 Mexico (Galloway and Williams, 1991; Galloway et al., 2011). They all show an increase of the 107 sedimentation rates up to today, a critical point discussed by P. Molnar (Molnar and England, 108 1990; Molnar, 2004) that might be related to global climate cooling (e.g. Herman and 109 Champagnac, 2016) and/or large erosion/recycling of the foreland deposits (Schlunnegger and 110 Mosar, 2011; Leroux et al., 2017), linked frontal accretion, climate variations and drainage 111 network reorganization. Thus, the volumes distribution (storage, transfer, recycling) between 112 the foreland and the final sink at different stages of mountain belt evolution have not been fully explored since the synthesis of Hinderer, 2012. 113

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We propose in this study to measure the siliciclastic sediment volumes in a retroforeland basin where limited frontal accretion allows a more complete preservation of syntectonic deposits – the Aquitaine Basin and the Bay of Biscay during Cenozoic times for a better understanding of the erosion, the sediment transfer and deposition during the convergence to post-convergence periods of the Pyrenees Mountains and discuss at the first order the tectonic and climatic control factors. The measurement of the compacted siliciclastic sediment volumes (for a solid rocks equivalence) are based on sediment thickness (isopach) maps of known lithologies, built from an extensive dataset of seismic lines interpreted using the principles of seismic stratigraphy and dated on wells (Ortiz et al., 2020).

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125 2. Geological setting

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127 The North Pyrenean retro-foreland and its lateral deep area of ultimate sediment 128 deposition – the deep Bay of Biscay Basin – is segmented into three geographical-geological 129 units (Fig. 1): (1) upstream, bounded westward by the Pamplona transfer zone, the Aquitaine 130 Platform (i.e. the modern onshore Aquitaine Basin and its shelf), (2) the Landes Plateau, 131 bounded westward by the Santander "soft" transfer zone, is a step (1000-1800 m deep) 132 bounded by slopes and canyons (Capbreton, Cap-Ferret) and (3) the deep Bay of Biscay (4000-133 4500 m deep).

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135 **2.1. Tectonic, relief and exhumation evolution of the Pyrenees**

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137 The Pyrenees Mountains are a collisional belt bringing Eurasian lithosphere over the 138 Iberian lithosphere (e.g. Roure et al., 1989; Muñoz, 1992). Both Pyrenees and Basque-139 Cantabrian Mountains result from the inversion and compression of the Albian hyperextended 140 Eurasian lithosphere (e.g. Fabriès et al., 1991, 1998; Lagabrielle et Bodinier, 2008; Jammes et 141 al., 2009; Lagabrielle et al., 2010; Masini et al., 2014; Clerc et al., 2012, 2016; Ducoux et al., 2019; Saspiturry et al., 2019; Ducoux et al., 2021, Tugend et al., 2014). The relative motion of
Eurasia and Africa plates controlled the orogeny: the compression started at the time of
convergence of the two plates, i.e. at the end of the Santonian (83.6 Ma, Schettino and Turco,
2011) and ended around the Oligocene-Miocene boundary (chron 6c, Roest and Srivastava,
1991, Macchiavelli et al., 2017).

147 Thermochronological data also document the early mountain building with a cooling phase at around 70 Ma (Whitchurch et al., 2011; Beamud et al., 2011; Mouthereau et al., 148 149 2014; Ternois et al., 2019; Waldner et al., 2021). During Cenozoic, several studies resolve the 150 increase of exhumation during late Eocene-Oligocene times (Fitzgerald et al., 1999; Sinclair et 151 al., 2005; Gibson et al., 2007; Jolivet et al., 2007; Gunnel et al., 2009; Metcalf et al., 2009; 152 Whitchurch et al., 2011; Fillon and van der Beek, 2012; Bosch et al., 2016) following an earlier 153 uplift of the eastern range during Middle Eocene times (Maurel et al., 2008; Beamud et al., 154 2011; Mouthereau et al., 2014; Vacherat et al., 2014; 2016; Ternois et al., 2019; Whitchurch 155 et al., 2011; Waldner et al., 2021). Finally, a late Miocene exhumation phase is recorded in the 156 Western Pyrenean Range by several authors (Jolivet et al., 2007; Bosch et al., 2016; Fillon et 157 al., 2021)

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2.2. Stratigraphic and tectonic evolution of the Aquitaine Basin and Bay of Biscay deep Basin
Several studies have been carried out on the tectono-stratigraphic evolution of the
Aquitaine Basin since the late Cretaceous (Brunet, 1984; Desegaulx and Brunet, 1990; Brunet,
1994; Sztrákos et al., 1998; Serrano et al., 2001; Ford et al., 2016; Sztrákos and Steurbaut,
2017; Angrand et al., 2018). They identified two subsiding phases (Latest Santonian - Danian
and Thanetian – Oligocene) separated by the so called "Palaeocene quiet period"
characterized by a low to near zero subsidence phase (Ford et al., 2016). The first subsidence

phase has been largely influenced by the post rift thermal subsidence because of the short time between the end of the rifting and the beginning of the convergence (around 10 Ma, Angrand et al., 2018). Serrano et al., (2001) differentiate two distinct phases in the Western Aquitaine Basin (1) from Palaeocene to Middle Eocene - compressional basin due to lithospheric buckling (mixed system – East-West oriented turbidites and carbonates platform) and (2) from Middle Eocene to Oligocene – foreland stage (progressive continentalization on the eastern part and deltaic progradation toward the West).

173 Recently, through an extensive bio- and seismic stratigraphy study of both basins, Ortiz 174 et al. (2020) identified a set of several discontinuities of tectonic origins (fig. 2). The orogenic 175 period is subdivided into two steps (fig. 2): (1) the dominant lithosphere flexure with the 176 formation of foredeeps up to the uppermost Lutetian and (2) the propagation of the 177 deformation from the orogenic wedge through the basin mainly along salt-controlled thrusts 178 and anticlines locating subbasins. From the basin record point of view, the transition between 179 orogenic and post-orogenic periods is Chattian in age, from 27.1 to 25.2 Ma. This time interval 180 corresponds to the end of thrusting/inversions and a modification of the sediment 181 preservation pattern. The post-orogenic period is also subdivided into two intervals (fig. 2): 182 (1) a sharp decrease of the subsidence rates over the Aquitaine Platform, mainly filled by 183 continental sediments during early Miocene times (25.2-16.4 Ma) and (2) an uplift of the 184 Aquitaine Platform from 16.4 Ma to today characterized by a low-preservation domain up to 185 10.6 Ma. The main uplift phase occurred at 10.6 Ma and caused a generalized by-pass on the 186 Aquitaine Platform (Ortiz et al., 2020).

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188 **3. Measurement of siliciclastic sediment volumes and rates**

190 The measurement of siliciclastic sediment (sink) volumes is based on four sediment 191 thickness (isopach of preserved sediments) maps: Palaeocene-Eocene (66-33.9 Ma - fig. 3), 192 Oligocene (33.9-23.03 Ma - fig. 4), Miocene (23.03-5.3 Ma - fig. 5) and Pliocene-Pleistocene 193 (5.3-0 Ma - fig. 6). These isopach maps are the products of a seismic stratigraphic analysis of 194 an extensive seismic line dataset (TotalEnergies, BRGM, BSS, fig. 1). This quite low time-195 resolution is due to the absence of wells in the deep Bay of Biscay and the difficulty to extend 196 timelines from the Aquitaine Platform where several dated wells are available (see Ortiz et al., 197 2020). For each interval the main lithologies proportion is known in the Aquitaine platform 198 and in the Landes Plateau (see Ortiz et al., 2020) and for the distal part of the Bay of Biscay, 199 we used the studies available in this domain about the Cenozoic sedimentary record (Cremer, 200 1983; Iglesias, 2009; Brocheray et al., 2014).

201 The sediment volumes measured from the isopach maps were later decompacted and 202 the amount of sediments produced in the basin (here mainly carbonates) was removed to get 203 solid rock volumes. We used the protocol and code published by Guillocheau et al. (2012) that 204 consider uncertainties or ranges for (1) time-depth conversion parameters, (2) absolute ages 205 of the different horizons, (3) carbonate content and (4) surface porosity and e-folding depth 206 for porosity decrease. During Cenozoic times the deep-sea Bay of Biscay fan fed through the 207 Capbreton and Cap-Ferret Canyon by the sediments coming from the Aquitaine Basin has 208 progressively migrated westward (Cremer, 1983; Iglesias, 2009; Brocheray et al., 2014), our 209 calculation area is evolving accordingly (dashed line on figs, 3, 4, 5, 6). In order to compare the 210 volumes, they will be presented through the evolution of the ratio between Aquitaine Basin 211 volume (V_{On}) and Bay of Biscay volume (V_{Off}) as follow: $R_{OnOff} = V_{On}/V_{Off}$.

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4. Isopach maps: results and interpretation

215 4.1. Paleocene-Eocene (66-33.9 Ma)

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The *Paleocene-Eocene* (66-33.9 Ma - Fig. 3) time interval is characterized by sediment preservation along the North Pyrenean Thrust on the Aquitaine Platform, with three subsiding domains - 10-50 km large - filled by 3500 m of sediments accumulated in 32 Ma. Two less subsiding domains were located north of the previous ones and bounded by two ridges with very low accumulation. Few sediments were preserved along the Landes Plateau. In the deep Bay of Biscay, two depocenters occurred south of the South Armorican Margin and north of the Biscay Wedge Front.

224 Interpretation: The three subsiding domains correspond to a segmented foredeep 225 (Ortiz et al., 2020) controlled by the inherited structures from the Early Cretaceous rifts history 226 (Angrand et al., 2018). These domains are mainly filled during the first orogenic phase (up to 227 uppermost Lutetian) and the small depocenter in the northern part of the ridges correspond 228 mainly to the second orogenic phase, when the deformation is propagated basinward. The 229 two deep depocenters of the Bay of Biscay are disconnected from the Aquitaine Platform and 230 bounded by the inverted extensional structures of the Gascogne Dome Bulge (Thinon, 1999; 231 Thinon et al., 2001). The first depocenter located in the Armorican subbasin, was fed by 232 sediments coming from the Loire River (fig. 1) or from the Armorican Massif (Guillocheau et al., 2003). 233

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235 4.2. Oligocene (33.9-23.03 Ma)

237 The Oligocene (33.9-23.03 Ma - fig. 4) was a period of more widely distributed 238 sediments accumulation along the three domains. In the Aquitaine Platform depocenters are 239 located (i) on both sides of salt-related controlled thrusts forming anticlines or ridges 240 (Maubourguet-Antin, Audignon) or (ii) along N140° faults (e.g. Téthieu Fault, location on 241 figures 1 and 4). In the south Landes Plateau, north of the Basque-Cantabrian Mountains, a 60 242 km-large depocenters occurred. In the deep Bay of Biscay, a NW-SE trending depocenter is 243 located south-west of the inverted structures of the Gascogne Dome Bulge and north of the 244 Biscay Wedge Front.

245 Interpretation: Oligocene is a period during which (1) segmented foredeep was no 246 longer subsident and (2) compressive deformation propagated from the orogenic wedge to 247 the retro-foreland basin along salt decollement levels localizing little subbasins (Serrano, 248 2001; Ortiz et al., 2020). The depocenters of the south Landes Plateau may be due to the 249 loading effect of the inverted and thrusted Basque Cantabrian Margin starting at 37 Ma and 250 ending at 28.5 Ma (Gomez et al., 2002). The depocenter of the deep Bay of Biscay is 251 disconnected from the Aquitaine Platform and moderately fed by the erosional product 252 coming from the inversion and deformation of the Cantabrian Margin and the uplift of the 253 Cantabrian Mountains from Late Eocene to Oligocene times (Gallastegui et al., 2002; Pedreira 254 et al., 2015; Fillon et al., 2016).

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256 4.3. Miocene (23.03-5.3 Ma)

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The *Miocene* (23.03 to 5.3 Ma – fig. 5) recorded a major change in the sediment distribution. The Aquitaine platform was henceforth a place of low sediment preservation (less than 200 m for 17.5 Ma). Most of the sediments were transferred and stored along the Landes Plateau and the deep Bay of Biscay. Two major sediment accumulations were active in the Landes Plateau, (1) to the north, a progradational wedge (Ortiz et al., 2020) located at the transition between the southeastern part of the South Armorican shelf and the Landes High and (2) to the south, an E-W trending depocenter located north of the Capbreton canyon. The preserved sediments along the deep Bay of Biscay are organized as an E-W trending body more than 100 km-large located on the Biscay Wedge Front. It is upstream in connection with the two depocenters of the Landes Plateau.

268 Interpretation: The Aquitaine Platform was no longer a significant subsiding domain. 269 Most of the sediments bypassed to deepest areas: (1) through deltaic progradational wedges 270 on the Landes Plateau, (2) through canyons and at the end of the system (3) as deep-sea fans. 271 The depocenters located north to the Capbreton canyon correspond to the northern levee of 272 the canyon already active at this period (Cremer, 1983; Iglesias, 2009, Ortiz et al., 2020). 273 Progradationnal deltaic wedge accumulate at the head of the Cap-Ferret canyon (western 274 extension of the north Landes Plateau depocenter – Cremer, 1983) and downstream (East of 275 the deep Bay of Biscay depocenter), an erosion domain where the canyon is active. These two 276 canyons domains fed downward, the Cap-Ferret deep-sea fan (Cremer, 1983; Iglesias, 2009) 277 in the deep Bay of Biscay.

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279 **4.4. Plio-Pleistocene (5.3-0 Ma)**

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The *Pliocene-Pleistocene* (5.3-0 Ma – fig. 6) time-interval shows an accentuation of the sediment distribution pattern established during Miocene. (1) No to very few sediments were preserved on the Aquitaine Platform. (2) Two depocenters occurred on the Landes Plateau: (i) a N-S trending one north to the Landes High (progradationnal wedges – Ortiz et al., 2020) and (ii) a N-S trending one south of the Landes High. (3) The depocenters of the deep Bay of Biscay
are located (i) at the base of slope of the South Armorican shelf and (ii) along the Biscay Wedge
Front in connection with the northern depocenter of the Landes Plateau.

Interpretation: The Aquitaine Plateau was upstream uplifted and incised by rivers (Mouchené et al., 2017; Ortiz et al., 2020). All the products of erosion are transferred to the deepest domains with by-pass to very low sedimentation along the modern Landes Forest (Dubreuilh et al., 1995). The prograding sedimentary wedge is now located at the border of the Aquitaine shelf and the two canyons (Capbreton and Cap-Ferret) alternated periods of erosion and deposition (Cremer, 1983; Iglesias, 2009).

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295 5. Measurement of the siliciclastic sediment budget: results (fig. 7)

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297 In the deep-sea plain, siliciclastic volume measurements were only performed in the 298 south deep Bay of Biscay Basin, south of the Gascogne Dome Bulge, the sediment located 299 northward (Armorican Subbasin) being fed from the Loire River (fig. 1) and the Armorican 300 Massif (see 4.1). In addition, during Cenozoic times the deep-sea Bay of Biscay fan fed through 301 the Capbreton and Cap-Ferret Canyon by the sediments coming from the Aquitaine Basin is 302 progressively migrating westward (Cremer, 1983; Iglesias, 2009; Brocheray et al., 2014). This 303 is the reason why, our calculation area is evolving according to the deep-sea fan location (Figs, 304 3, 4, 5, 6).

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The total amount of siliciclastic sediments (Tab. 1, Fig. 7) deposited from the Aquitaine Platform to the south deep Bay of Biscay during Cenozoic times is 51 500 km³ with an uncertainty of 16 800 km³, i.e. 33%.

310 The four-time intervals here defined, have not the same duration and therefore 311 siliciclastic sedimentation rates had to be used for a comparison between time intervals instead of volumes (Tab. 1, fig. 7). At first order the sedimentation rates are increasing from 312 313 535 km³/Myr (between 66 and 33.9 Ma) to 3150 km³/Myr (from 5.3 to 0 Ma). Oligocene and 314 Pliocene-Pleistocene seems to be significant instants of siliciclastic sediment rates increase. 315 316 The siliciclastic sediment budget comparison (Tab. 1, fig. 7) between the onshore 317 (Aquitaine Basin) and offshore (Aquitaine shelf, Landes Plateau, south Bay of Biscay) shows a shift, trough Cenozoic times, from onshore to offshore preferential accumulation since 318 319 Oligocene-Miocene boundary. The sediment ratio between onshore and offshore (R_{OnOff}) is (i) 320 25.3 from 66 to 33.9 Ma, (ii) 7.5 from 33.9 to 23.03 Ma, (iii) 0.18 from 23.03 to 5.3 Ma and (iv) 321 0.01 from 5.3 to 0 Ma. 322 323 6. Discussion on the siliciclastic sediment budget 324 325 Volume comparison with South Pyrenean Foreland and others orogenic adjacent basins - The 326 amount of siliciclastic sediments preserved in northern foreland is 51 500 ± 16 800 km³ for 327 Cenozoic times. A comparison with the sediment volumes of other orogenic systems 328 supposes to get a sediment budget for both sides of the Pyrenees, i.e. including the South 329 Pyrenean Foreland basin. Several studies have been conducted for quantifying the preserved 330 sediment volumes in the South Pyrenean sink (i.e. Ebro and Valencia Basins)(Nelson, 1990;

331 Garcia-Castellanos et al., 2003; Babault et al., 2006; Filleaudeau, 2011; Arche et al., 2010;

332 Watts and Torné, 1992). The Ebro Basin has been under endoreic conditions since 36.2 Ma

333 (Costa et al., 2010) and then opened towards the Mediterranean Sea between 12 Ma and 334 7.5 Ma (Garcia-Castellanos et al., 2003; Fillon et al., 2012; Garcia-Castellanos and 335 Larrasoana, 2015). The total amount of preserved sediments in the Ebro and Valencia Basin 336 is 260 300 km³ since 66 Myr. For a better sediment budget on both sides of the Pyrenees, 337 only the sediments coming from the mountain belt should be considered, i.e. the total 338 Cenozoic preserved sediments volumes in the Ebro basin and the Neogene for the Valencia Basin. This corresponds to a range between 78 000 km³ (Filleaudeau, 2011) and 110 000 km³ 339 340 ± 5000 km³ (Garcia-Castellanos et al., 2003) in the Ebro Basin and 92 000 km³ in the Valencia 341 Basin (Garcia-Castellanos et al., 2003). This gives a total of preserved sediments between 342 170 000 km³ and 202 000 km³. At the first order, this result shows a factor of four between 343 the northern retroforeland (our study) and the South Pyrenean proforeland.

344 The first-order estimation of total volume preserved in the Pyrenean adjacent basins 345 (i.e. Aquitaine Basin, Bay of Biscay, Ebro Basin, Valencia Basin) is between 220 000 km³ and 346 250 000 km³. The total volume of siliciclastic sediments produced by the Alps since Oligocene 347 times is of 890 000 km³ (220 000 km³ from the Eastern Alps and 670 000 km³ from the western 348 ones- Kuhlemann et al., 2002) i.e. four times more than the Pyrenees. However, for larger 349 orogens the order of magnitude changes with 3.35 10⁶ km³ of sediment produced by the Rocky 350 Mountains on its continental part and stored in the Gulf of Mexico (Galloway et al., 2011) and 351 the 4.95 10⁶ km³ by the western side of the Himalaya and accumulated along of the Indus 352 Delta (Clift et al., 2006).

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Volume comparison with Pyrenean thermochronological data – We present here some
 volumes estimation for the Cenozoic period based on published themrochronology data in the
 Pyrenees.

- Too few data (AFT and AHe) provide Palaeocene to early Eocene ages to obtain a
 consistent exhumation trend across the entire Pyrenean range.

359 - During the late Eocene/lower Oligocene (from 37 to 30 Ma), orogenic exhumation (> 360 0.5 km/Myr) is recorded in Central and Eastern Pyrenean Massif (Maladetta, Nogueres, 361 Canigou) according to studies from Fitzgerald et al., (1999), Gibson et al., (2007), Fillon and 362 Van der Beek, (2012) and Gunnel et al., (2009). These authors modeled an exhumation rate of 363 1.5 to 4 km/Myr. Considering an average of 2 km/Myr integrated over the whole area 364 (inducing a strong assumption of a continuous constant exhumation) during the period of 35-365 30 Ma, the eroded volume should be of 50 000 km³. This value is very high compared to what 366 has been measured in the Aquitaine Basin during the same period (16 500 km³ from base 367 Cenozoic to base Oligocene). This volume seems therefore to supply mainly the Ebro basin 368 with a significant increase observed at the base of Oligocene (i.e. 33.9 Ma) (Filleaudeau, 2011).

369 - Since middle Oligocene times, a moderate exhumation (between 0.1 and 0.5 km/Myr) 370 is proposed by several authors in Western, Central and Eastern massifs of the Pyrenean range 371 (DeFelipe et al., 2019; Bosch et al., 2016; Fillon et al., 2021; Fitzgerald et al., 1999; Gibson et 372 al., 2007). An average exhumation rate of 0.25 km/Myr across the entire range provides an 373 estimation of 17 500 km³ for the Middle to Late Oligocene period. This volume is, like during 374 Late Eocene/Lower Oligocene, one order of magnitude higher than what we measure in the 375 Aquitaine Basin. This volume supply mainly the Ebro basin where the Oligocene volumes are 376 very large (around 45 000 km³ in Filleaudeau, 2011).

Finally, the important increase of exhumation rate recorded in the Western Pyrenees
(see Fillon et al., (2021) for the data synthesis and modelling) infers a volume estimation of
2 000 km³ for the late Miocene period. This quantification is low compared to the volumes of
preserved sediments.

We draw attention to the fact that these estimation are produces from an average of 382 the results of modelling data done in 1D and extrapolated on a surface. These volumes based 383 on thermochronological data are probably overestimated.

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386 6.1. Age estimation of siliciclastic sediment flux increase from interpreted seismic data 387

388 As we mentioned earlier, the low stratigraphic resolution (Palaeocene-Eocene, 389 Oligocene, Miocene and Pliocene-Pleistocene) is due to difficulties to propagate the high 390 resolution age model (established in shallow marine setting in the Aquitaine Platform and 391 Landes Plateau – Ortiz et al., 2020) across the whole seismic survey in the deep Bay of Biscay 392 Basin and continental Aquitaine series. In order to be more precise about the age of the 393 siliciclastic rate increase during the two key periods (Oligocene and Plio-Pleistocene) and their 394 possible links to climatic or geodynamic event, we performed a 2D quantification on the cross 395 section published in Ortiz et al., (2020). This quantification aims at comparing on three 396 different locations (proximal Aquitaine platform – deep Bay of Biscay Basin – progradational 397 wedges in between) the percentage of 2D surface (comprising sediment) normalized on their 398 duration between each high-resolution timeline.

399 The figure 8 present the results and shows a refined estimate of increase at 26.0 ± 2.0 400 Ma for Oligocene period and 2.5 ± 0.5 Ma for Pliocene-Pleistocene period.

401

402 6.2. The 26.0 ± 2.0 Ma increase

The measured Late Oligocene (26.0 \pm 2.0 Ma) siliciclastic rate increase could be explained by both a tectonic factor – the accretion of Pyrenees basal units – and a climatic factor – the icehouse-greenhouse transition and the late Oligocene warming (Zachos et al., 2001).

408 Tectonic - A Late Eocene to Oligocene/Miocene paroxysm of denudation (37-29 Ma 409 central Pyrenees and 37-20 Ma western Axial Zone) is measured by the numerous 410 thermochronological studies (see references in section 2.1) and by the dated active tectonics 411 mainly in the Southern Pyrenees (Montsec and Sierras exteriores thrust systems associated to Orri and Rialp thrust sheets, see Munoz, 1992, Teixell, 1998, Vergés et al., 2002 and 412 413 Mouthereau et al., 2014), or in the Jaca Basin cover thrust and associated Gavarnie and more 414 frontal thrusts (Labaume et al., 2016). During late Oligocene time, the retro-foreland is 415 affected by several truncation/erosional surfaces from 27.1 to 25.2 Ma (Ortiz et al., 2020). 416 During this event, some syn-orogenic deposits are recycled and can be transferred to more 417 distal sink. The observed increasing volumes and increasing export of sediment to the Bay of 418 Biscay is thus contemporaneous with a (long) denudation paroxysm, but also with a more 419 temporally resolved drop of preservation in the foreland which becomes less subsident and 420 might be locally eroded and recycled. The observed modification of source to sink pattern 421 might record combination of two processes, denudation increasing phase and cessation of subsidence in the foreland could explain the increase export toward the final sink and 422 423 sedimentation rate increase observed in the western Aquitaine Platform part (fig. 4).

424 *Climate* - Few palaeoclimatological data dealing with the palaeoprecipitation record 425 are available in the Aquitaine Basin. A palaeobotanical synthesis (Dupéron-Laudoueneix and 426 Pons, 1985) indicate a cooling expressed by a decrease of the tropical taxon from the Eocene 427 to the Oligocene that corresponds to the Eocene-Oligocene global cooling event (Zachos et
428 al., 2011). This event takes place at 33.9 Ma, thus well before the observed 26 Ma event.

429 The 26.0 \pm 2.0 Ma increase event is contemporary with the Late Oligocene Warming, 430 that correspond to a sustained *ca*. 6°C global warming (Zachos et al., 2001). Warming events 431 (such as PETM) can cause sediment pulses (Foreman et al., 2012). Schlunegger and Norton, 432 (2015) have tested this increase hypothesis in the Alpine Foreland Basin. They focus on the 433 Napf megafan sedimentary evolution in the Alpine foothills during this period of sudden 434 warming. As in our study the period is also a time of increasing exhumation phase in the Alpine 435 mountain (Schmid et al., 1996). They didn't observe remarkable shift in the water discharge, 436 and they ruled out the hypothesis of sediment rate increase due to the Late Oligocene 437 warming but linked it to tectonic process.

In our case, the Late Oligocene (26.0 ± 2.0 Ma) siliciclastic rate increase is linked to the
high erosion rates and lower preservation in the Aquitaine Platform controlled by the orogenic
tectonic processes. This increase is also registered in the Ebro basin (Filleaudeau, 2011).

441

442 6.3. The 2.5 ± 0.5 Ma increase and the long-term volume increase

443

The change in the rate of siliciclastic sediment supply during Plio-Pleistocene (at 2.5 ± 0.5 Ma) is significant (fig. 8) and sharply accentuates the overall increasing rate trend through Cenozoic times. This confirms for this area, the Hay's curve (Hay et al., 1989, 1990; Wold et Hay, 1990) showing a world-scale increase of the siliciclastic fluxes up to today, interpreted by Molnar (Molnar and England, 1990; Molnar, 2004) as the consequence of the global Earth cooling during Plio-Pleistocene times. Herman et al., (2013) further suggest a Plio-Pleistocene (at around 6 Ma) increase in erosion rates and with an acceleration at around 2 Ma that they 451 attribute to the global cooling Pliocene and Pleistocene episodes that enhanced erosion in452 mountainous glaciated area.

453 Onshore, the most sensible low-temperature thermochronological data, ((U-TH)/He 454 and Fission tracks on apatites) do not show any Pliocene increase of denudation either in the 455 Pyrenees Mountains (e.g. Bosch et al., 2016) or in the French Massif central (T. François, 456 personal communications). The youngest cooling episode is recorded at around 10 Ma (Late 457 Miocene) in the western axial zone and no basin erosion > 1.5 km could be retrieved in the 458 Aquitaine Basin for the last 20 Myrs (Fillon et al., 2021), except in easternmost Aquitaine Basin 459 (Corbières area) Al Reda et al., (2021) althought this denudation event is temporally poorly 460 resolved between 20 and 5 Ma, this event is attributed by the authors to early Miocene and 461 opening of the Gulf o Lion.

462 With these observations, several hypothesis have yet to be tested: (1) a climate effect 463 on the intensity and pattern of erosion, with the effect of glaciations and change of the pattern 464 of erosion, from widely distributed before 5.3 Ma to localized in valleys after with a greater 465 climate effect from 2 Ma, which could be associated to (2) an "underestimated" significant re-466 erosion of previously deposited sediments in the Aquitaine Basin ("cannibalization") and (3) 467 lateral supply of sediments from other sources in the deep-sea domain under the action of 468 deep-sea currents (Le Danois Contourite Depositional System due to Mediterranean Outflow 469 Water, Van Rooij et al., 2010). Moreover, Pliocene-Pleistocene evolution of the Aquitaine 470 Platform records a two steps evolution, the emplacement of megafans with the most striking 471 one i.e. Lannemezan megafan starting during Messinian up to Uppermost Pliocene (Ortiz et 472 al., 2020). This megafan mainly source by the Neste River is dissected during Late Quaternary 473 (at or before 300 ka, Mouchené et al., 2017). This indicates a shift from a sedimentation phase

474 to an incision phase, that takes place between 2.6 Ma and 300 ka. This supports the hypothesis
475 of recycling by erosion of previous deposits sediments in the Aquitaine Platform.

Although glaciations is well documented during Late Quaternary in the Pyrenees (Calvet et al., 2011; Delmas et al., 2008), their importance in terms of sedimentary budget is still not quantified. Identified glacial remnants are restricted to the upper part of the valleys and mostly confined in the mountains belts (Delmas et al., 2011). The preservation over large areas of preglacial flat surfaces in the high chain (Bosch et al., 2016) claim for erosion localized mainly in glacial valleys. This supports the hypothesis of a more localized erosion in the mountain valleys.

483

484

485 **6.4. Sediment budget evolution between onshore and offshore**

486

The change of the sediment ratio between onshore and offshore (R_{OnOff}) from 25.3 (66 to 33.9 Ma) to 0.01 (5.3 to 0 Ma) has to be related to the general evolution of the two basins as summarized in item 2.2. This drastic change of ratio in favour of the "ultimate sink" is in line with the synthesis of Hinderer, 2012.

The first interval (66.0-33.9 Ma) spans a long period of time and several steps of the
 orogeny. A first phase (66-56 Ma) of relatively quiescent with very low relief in the
 mountain belt and siliciclastic input, a second phase (56-33.9 Ma) that correspond to
 progressive mountain building with increasing relief and erosion. At around 40 Ma,
 deformation propagate in the foredeep and segmented the foredeep. During this entire
 period, accommodation space created by the flexure was large enough to trap siliciclastic

497 sediments coming from the incipient mountain belt. This may explain the general trend498 between 66 and 33.9 Ma.

The second period (33.9-23.03 Ma) also spans different periods, paroxysm of denudation
 in the Mountain belt, while in the Aquitaine Platform, two periods are deciphered, a first
 period of accumulation and deformation (up to 25.2 Ma) and a second with a decrease of
 subsidence and increase of export to the distal sink (since 25.2 Ma). As a consequence,
 since 25.2 Ma, siliciclastic supply is too important compare to the accommodation space
 creation and the sediments are transferred to the progradational wedges in the Aquitaine
 platform. This explains the trend during Oligocene and particularly since 25.2.

During the post-orogenic period, first the Aquitaine platform subsidence drastically
 decreased (25.2-10.6 Ma) and second this domain was partly uplifted (10.6-0 Ma). This
 favored the sediment export to the deep-sea domain, first poorly balanced (R_{OnOff} at 0.18

- map 23.03-5.3 Ma) and then unbalanced (R_{OnOff} at 0.01 - map 5.3-0 Ma).

510

511 **7. Conclusion**

512

513 Our objective was to measure the siliciclastic sediment volumes and rates of a retro-514 foreland basin and its lateral deep equivalent, during the end tectonic and topographic 515 evolution of the Pyrenees mountain belt.

516

520

517 (1) The amount of sediments is of 51 500 \pm 16 800 km³ for the Cenozoic.

518 (2) The siliciclastic sediment rate curve during Cenozoic shows two major increase around
519 26.0 and 2.5 Ma. This last one is a major increase. This is in agreement with the world-

scale trend of Hay et al. (1989), expected of climatic origin by Molnar and England (1990).

| 521 | The 26.0 Ma increase is clearly related to the exhumation of the Pyrenees of tectonic |
|-----|---|
| 522 | origin. The discussion on the major 2.5 Ma episode is still open. |

(3) The mass balance between the Aquitaine Platform and the deepest domains (Landes Plateau, Bay of Biscay) change through time in favour to the deep domain. This might be explained by the ratio between subsidence created accommodation space and the produced siliciclastic sediments by the mountain belt on the platform. This confirms and quantifies the stratigraphic model of sink preservation proposed by Ortiz et al. (2020). This also suggests the sediment accumulation in the deep part of the margin is only controlled

- 529 by the accommodation/sedimentation balance on the platform.
- 530

531 Data Availability Statement

- 532 Research data are not shared
- 533

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882 **Table 1 caption:** Siliciclastic sediment volumes and rates values, with the uncertainties

| Time interval | (Ma) | Volume (× 1 km ³) | σ | Rate (× 1 km ³ / Myr) | σ |
|------------------------|--------------|-------------------------------|-----------|----------------------------------|---------|
| Pliocene - Pleistocene | 5.33 - 0 | 228.6 | ± 10.8 | 43.1 | ± 2.0 |
| Aquitanian - Messinian | 23.03 - 5.33 | 1 370.3 | ± 385.2 | 77.2 | ± 21.7 |
| Rupelian - Chattian | 33.9 - 23.03 | 7 894.7 | ± 1 949.9 | 726.2 | ± 179.3 |
| Danian - Priabonian | 66 - 33.9 | 16 545.6 | ± 5 563.1 | 515.4 | ± 173.3 |

Accumulated solid volumes, rates and associated variance for the Aquitaine Basin

Accumulated solid volumes, rates and associated variance for the southern Bay of Biscay

| Time interval | (Ma) | Volume (× 1 km ³) | σ | Rate (× 1 km ³ / Myr) | σ |
|------------------------|--------------|-------------------------------|-----------|----------------------------------|-----------|
| Pliocene - Pleistocene | 5.33 - 0 | 16 445.0 | ± 5 929.1 | 3 084.2 | ± 1 111.9 |
| Aquitanian - Messinian | 23.03 - 5.33 | 7 301.3 | ± 2 231.5 | 411.8 | ± 125.9 |
| Rupelian - Chattian | 33.9 - 23.03 | 1 043.7 | ± 351.8 | 96.0 | ± 32.4 |
| Danian - Priabonian | 66 - 33.9 | 652.7 | ± 352.0 | 20.6 | ± 11.1 |

Accumulated solid volumes, rates and associated variance for the whole basins

| Time interval | (Ma) | Volume (× 1 km ³) | σ | Rate (× 1 km ³ / Myr) | σ |
|------------------------|--------------|-------------------------------|-----------|----------------------------------|-----------|
| Pliocene - Pleistocene | 5.33 - 0 | 16 673.6 | ± 5 940.0 | 3 145.9 | ± 1 120.8 |
| Aquitanian - Messinian | 23.03 - 5.33 | 8 671.7 | ± 2 616.8 | 489.1 | ± 147.6 |
| Rupelian - Chattian | 33.9 - 23.03 | 8 938.4 | ± 2 301.7 | 822.3 | ± 211.7 |
| Danian - Priabonian | 66 - 33.9 | 17 198.3 | ± 5 915.2 | 535.8 | ± 184.3 |

Table 2 caption: Percentage of carbonates for the studied time interval 884

| Time interval | (Ma) | Carbonate fraction (%) | σ |
|------------------------|--------------|------------------------|-------|
| Pliocene - Pleistocene | 5.33 - 0 | 7.5 | ± 7.5 |
| Messinian | 7.24 - 5.33 | 14 | ± 10 |
| Aquitanian - Tortonian | 23.03 - 7.24 | 18 | ± 10 |
| Rupelian - Chattian | 33.9 - 23.03 | 44 | ± 15 |
| Danian - Priabonian | 66 - 33.9 | 60 | ± 20 |

| Sarbonate fraction for the «Façade Atlantique» Basin | | | | | |
|--|---------------|------------------------|------|--|--|
| īme interval | (Ma) | Carbonate fraction (%) | σ | | |
| Pliocene - Pleistocene | 5.33 - 0 | 5 | ± 5 | | |
| Tortonian - Messinian | 11.63 - 5.33 | 10 | ± 10 | | |
| anghian - Serravallian | 15.97 - 11.63 | 20 | ± 20 | | |
| Burdigalian | 20.44 - 15.97 | 50 | ± 20 | | |
| Aquitanian | 23.03 - 20.44 | 60 | ± 30 | | |
| Rupelian - Chattian | 33.9 - 23.03 | 40 | ± 20 | | |
| Priabonian | 37.8 - 33.9 | 50 | ± 20 | | |
| Danian - Bartonian | 66 - 37.8 | 40 | ± 20 | | |

Carbonate fraction for the Arzacq Basin

| Time interval | (Ma) | Carbonate fraction (%) | σ |
|-------------------------|---------------|------------------------|------|
| Pliocene - Pleistocene | 5.33 - 0 | 5 | ± 5 |
| Tortonian - Messinian | 11.63 - 5.33 | 10 | ± 10 |
| Langhian - Serravallian | 15.97 - 11.63 | 20 | ± 20 |
| Burdigalian | 20.44 - 15.97 | 50 | ± 20 |
| Aquitanian | 23.03 - 20.44 | 60 | ± 30 |
| Rupelian - Chattian | 33.9 - 23.03 | 20 | ± 10 |
| Priabonian | 37.8 - 33.9 | 20 | ± 10 |
| Danian - Bartonian | 66 - 37.8 | 40 | ± 20 |

Carbonate fraction for the Carcassonne Basin

| Time interval | (Ma) | Carbonate fraction (%) | σ |
|-------------------------|---------------|------------------------|------|
| Pliocene - Pleistocene | 5.33 - 0 | 5 | ± 5 |
| Tortonian - Messinian | 11.63 - 5.33 | 10 | ± 10 |
| Langhian - Serravallian | 15.97 - 11.63 | 20 | ± 20 |
| Burdigalian | 20.44 - 15.97 | 50 | ± 20 |
| Aquitanian | 23.03 - 20.44 | 60 | ± 30 |
| Rupelian - Chattian | 33.9 - 23.03 | 25 | ± 20 |
| Priabonian | 37.8 - 33.9 | 20 | ± 20 |
| Danian - Bartonian | 66 - 37.8 | 40 | ± 20 |

Carbonate fraction for the Tarbes Basin

| Time interval | (Ma) | Carbonate fraction (%) | σ |
|-------------------------|---------------|------------------------|------|
| Pliocene - Pleistocene | 5.33 - 0 | 5 | ± 5 |
| Tortonian - Messinian | 11.63 - 5.33 | 10 | ± 10 |
| Langhian - Serravallian | 15.97 - 11.63 | 20 | ± 20 |
| Burdigalian | 20.44 - 15.97 | 50 | ± 20 |
| Aguitanian | 23.03 - 20.44 | 60 | ± 30 |
| Rupelian - Chattian | 33.9 - 23.03 | 20 | ± 10 |
| Priabonian | 37.8 - 33.9 | 15 | ± 10 |
| Danian - Bartonian | 66 - 37.8 | 40 | ± 20 |

Carbonate fraction for the Castres Basin (1.4.

| lime interval | (ivia) | Carbonate fraction (%) | 0 | |
|-------------------------|---------------|------------------------|------|--|
| Pliocene - Pleistocene | 5.33 - 0 | 5 | ± 5 | |
| Tortonian - Messinian | 11.63 - 5.33 | 10 | ± 10 | |
| Langhian - Serravallian | 15.97 - 11.63 | 20 | ± 20 | |
| Burdigalian | 20.44 - 15.97 | 50 | ± 20 | |
| Aquitanian | 23.03 - 20.44 | 60 | ± 30 | |
| Rupelian - Chattian | 33.9 - 23.03 | 40 | ± 20 | |
| Priabonian | 37.8 - 33.9 | 40 | ± 20 | |
| Danian - Bartonian | 66 - 37.8 | 40 | + 20 | |

| Carbonate fraction for the Aquitaine platform | | | | | | | |
|---|---------------|------------------------|------|--|--|--|--|
| Time interval | (Ma) | Carbonate fraction (%) | σ | | | | |
| Pliocene - Pleistocene | 5.33 - 0 | 5 | ± 5 | | | | |
| Tortonian - Messinian | 11.63 - 5.33 | 10 | ± 10 | | | | |
| Langhian - Serravallian | 15.97 - 11.63 | 20 | ± 20 | | | | |
| Burdigalian | 20.44 - 15.97 | 50 | ± 20 | | | | |
| Aguitanian | 23.03 - 20.44 | 60 | ± 30 | | | | |
| Rupelian - Chattian | 33.9 - 23.03 | 20 | ± 10 | | | | |
| Priabonian | 37.8 - 33.9 | 15 | ± 10 | | | | |
| Danian - Bartonian | 66 - 37.8 | 40 | ± 20 | | | | |

Carbonate fraction for the Tartas Mirande Basin

| Time interval | (Ma) | Carbonate fraction (%) | σ | |
|-------------------------|---------------|------------------------|------|--|
| Pliocene - Pleistocene | 5.33 - 0 | 5 | ± 5 | |
| Tortonian - Messinian | 11.63 - 5.33 | 10 | ± 10 | |
| Langhian - Serravallian | 15.97 - 11.63 | 20 | ± 20 | |
| Burdigalian | 20.44 - 15.97 | 50 | ± 20 | |
| Aguitanian | 23.03 - 20.44 | 60 | ± 30 | |
| Rupelian - Chattian | 33.9 - 23.03 | 30 | ± 20 | |
| Priabonian | 37.8 - 33.9 | 50 | ± 10 | |
| Danian - Bartonian | 66 - 37.8 | 40 | ± 20 | |

Table 2 885



| 888 | Figure 1: A: Main physiographic and structural features and available dataset of seismic |
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| 889 | reflection lines in the Aquitaine Basin, Landes Plateau and Biscay Bay deep-basin. B: Location |
| 890 | of the studied area in Europe. |
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- **Figure 2:** Synthetic chart of the main events (deformation, topography, sediment routing) of
- 915 the Aquitaine Basin to deep Biscay Bay Basin sedimentary system

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938 Figure 4

| 939 | Figure 3: A: Palaeocene-Eocene preserved sediment thickness (isopach, meters) map. The |
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| 940 | volume quantification western limit is according to the deep sea fan location (Cremer, 1983; |
| 941 | Iglesias, 2009). B: interpreted cross-section from the proximal Aquitaine Basin part to the deep |
| 942 | Bay of Biscay Basin, the considered period (isopach map) is highlighted by colour in the cross- |
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| 966 | Figure 4: A: Oligocene preserved sediment thickness (isopach, meters) map. The volume |
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| 967 | quantification western limit is according to the deep sea fan location (Cremer, 1983; Iglesias, |
| 968 | 2009). B: interpreted cross-section from the proximal Aquitaine Basin part to the deep Bay of |
| 969 | Biscay Basin, the considered period (isopach map) is highlighted by colour in the cross-section. |
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| Figure 5: A: Miocene preserved sediment thickness (isopach, meters) map. The volume |
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| quantification western limit is according to the deep sea fan location (Cremer, 1983; Iglesias, |
| 2009). B: interpreted cross-section from the proximal Aquitaine Basin part to the deep Bay of |
| Biscay Basin, the considered period (isopach map) is highlighted by colour in the cross-section. |
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| 1021 | Figure 6: A: Pliocene-Pleistocene preserved sediment thickness (isopach, meters) map. The |
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| 1022 | volume quantification western limit is according to the deep sea fan location (Cremer, 1983; |
| 1023 | Iglesias, 2009, Brocheray et al., 2014). B: interpreted cross-section from the proximal |
| 1024 | Aquitaine Basin part to the deep Bay of Biscay Basin, the considered period (isopach map) is |
| 1025 | highlighted by colour in the cross-section. |
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| 1049 | Figure 7: Siliciclas | stic sedimentation | volumes | and | rates | of the | e Aquitaine | Basin | and | Bay | of |
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| 1050 | Biscay. | | | | | | | | | | |
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Figure 8: In the cross sections published in Ortiz et al., (2020), some parts of the interpretation 1075 1076 are of higher resolution than the resolution of the calculations in this paper (i.e. Palaeocene-1077 Eocene, Oligocene, Miocene, Pliocene-Pleistocene). For example, the figure 5 in Ortiz et al., 1078 (2020) shows several high resolutions surfaces (thanks to orbitostratigraphy) at 32.2, 32.0, 1079 31.3, 29.4, 27.1, 26.4, 25.2, 24.5, 24.4 Ma for the Oligocene period. This resolution on different 1080 parts of the cross section allows us to make a precise quantification shown in the figure. This 1081 figure is a compilation of the percentage of 2D area normalized to their duration thanks to 1082 high resolution timeline on different cross section published in Ortiz et al., (2020) 1083