



# Holocene hydrological changes of the Rhone River (NW Mediterranean) as recorded in the marine mud belt

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#### 8 Abstract

9 Expanded marine Holocene archives are relatively scarce in the Mediterranean Sea because most of the 10 sediments were trapped in catchment areas during this period. Mud belts are most suitable targets to access 11 expanded Holocene records. These sedimentary bodies represent excellent archives for the study of sea-land 12 interactions and notably the impact of the hydrological activity on sediment accumulation. We retrieved a 7.2 m-13 long sediment core from the Rhone mud belt in the Gulf of Lions in an area where the average accumulation rate 14 is of ca. 0.70 m/1000 years. This core thus provides a continuous and high-resolution record of the last 10 ka cal 15 BP. A multi-proxy dataset (XRF-core scan, <sup>14</sup>C dates, grain size and organic matter analysis) combined with 16 seismic stratigraphic analysis was used to document decadal to centennial changes of the Rhone hydrological 17 activity. Our results show that 1) the Early Holocene was characterized by high sediment delivery likely 18 indicative of local intense (but short duration) rainfall events , 2) important sediment delivery around 7 ka cal BP 19 roughly presumably related to increased river flux, 3) a progressive increase of continental/marine input during 20 the Mid-Holocene despite increased distance from river outlets due to sea-level rise possibly related to higher 21 atmospheric humidity caused by the southward migration of the storm tracks in the North Atlantic, 4) multi-22 decadal to centennial humid events in the Late Holocene. Some of these events correspond to the cold periods 23 identified in the North Atlantic (Little Ice Age, LIA; Dark Age) and also coincide with time intervals of major 24 floods a in the Northern Alps. Other humid events are also observed during relatively warm periods (Roman 25 Humid Period and Medieval Climate Anomaly).

#### 26 1. Introduction

27 The Holocene climate is characterized by centennial-scale climate changes that punctuated the final deglacial warming after the Younger Dryas (Ishihama et al., 2014; Ishihama et al.; 28 29 Renssen et al., 2009; Rogerson et al., 2011; 2004; Wanner et al., 2008). Wanner et al. (2014) 30 provided an extensive review of Holocene climate variability mainly based on chronologically well-constrained continental temperature time series that emphasize the 31 superimposition of the insolation-driven climate changes with those induced by other external 32 forcings such as solar activity, volcanism and greenhouse gases (CH<sub>4</sub>, CO<sub>2</sub> and NO<sub>2</sub>). Based 33 on existing data, Holocene climate can be divided into four periods: 34





- a) the early Holocene (between 11.7 and 8.2 ka cal BP, Walker et al., 2012) characterized by a
  progressive warming inducing ice-cap melting and outbreaks of freshwater from North
  America glacial lakes leading to a regional cooling in the Northern Hemisphere, *i.e.* the 8.2 ka
  cal BP cold event (Barber et al., 1999);
- b) the warm middle Holocene (between 8.2 and 4.2 ka cal BP, Walker et al., 2012) that
  coincides approximately with the Holocene Thermal Maximum (HTM) and is punctuated by
  several cold relapses (CR) (Wanner et al., 2011). Events at 6.4, 5.3 and 4.2 ka cal BP are the
  most significant in terms of temperature change (Wanner et al., 2011). The 4.2-ka event
  corresponds to enhanced dryness in the Southern Mediterranean, Asia and North America,
  that may have presumably played a role in the collapse of various civilizations (Cronin et al.,
  2013; Magny et al., 2013).
- c) the cold late Holocene (from 4.2 ka cal BP to the mid 19<sup>th</sup> century, Walker et al., 2012) that
  includes the 2.8 ka cal BP cold event possibly responsible for the collapse of the Late Bronze
  Age civilization (Do Carmo and Sanguinetti, 1999; Weiss, 1982) and the Migration Period
  cooling around 1.4 ka cal BP (Wanner et al., 2014). The late Holocene cooling trend
  culminated during the Little Ice Age (LIA) between the 13<sup>th</sup> and 19<sup>th</sup> century (Frezza and
  Carboni, 2009);
- d) the warm Industrial Era from 1850 AD onwards (Cronin et al., 2002; Rogerson et al., 2011;
  Wanner et al., 2011).
- In contrast to these cool events, the Medieval Climate Anomaly (MCA, 800-1300 AD) is often described as a warm period characterized by intense dryness in some regions of the Northern Hemisphere, such as for example Europe and the Mediterranean region (Cronin and Raymo, 1997) although not synchronous worldwide (PAGES 2k Consortium, 2013).
- The causes of Holocene climate variability are not yet fully understood despite recent 58 59 advances achieved through the study of climate archives from all around the word from both 60 marine and continental settings. To what extent these well-known climate events are global 61 rather than regional and what are the driving mechanisms at play are still open questions. 62 Numerical modelling allows examining in more details and on a broader geographical scale causes of rapid climate changes and the role of natural or anthropogenic forcings by better 63 integrating data from marine, land and ice archives (Fontanier et al., 2005; Fontanier et al., 64 2006; Müller and Suess, 1979; Perez-Sanz et al., 2014; Puig et al., 2004; Schmiedl et al., 65 66 2004; Van der Zwaan et al., 1999). Nonetheless, there are significant discrepancies between proxy reconstructions and numerical simulations that suggest the need to generate better 67 chronologically constrained high-resolution proxy records from continental and marine 68





69 archives and develop new approaches (Anchukaitis and Tierney, 2013; Evans et al., 2013). Of particular interest are the locations that allow developing paleo-hydrological and paleo-70 71 environmental investigations at the land / sea interface to better link atmospheric circulation 72 controlling the precipitation pattern over the continent and changes in the thermohaline circulation. Sediment drifts fed by water streams connected to the deep sea such as the Var 73 (Bonneau et al., 2014) or mid-shelf mud belts are interesting locations to recover sedimentary 74 75 archives where both continental and marine proxies can be analyzed. Mid-shelf mud belts, in 76 particular, are depot centers fed by streams that result from various processes including 77 diffusion under the influence of storms, advection by currents and transport by gravity flows 78 (Hill et al., 2007). They often form elongated sediment bodies, between 10-30 m and 60-100 m water depth, roughly parallel to the coastline. Such sediment bodies can reach several 79 10<sup>th</sup> of meters thickness when they are associated to large streams, and form infralittoral 80 prograding prisms (sometimes called subaqueous deltas) as for instance along the Italian 81 82 Adriatic coast (Cattaneo et al., 2003). Somehow, they are shallow-water equivalents to contourites, but they generally display higher accumulation rates making them ideal targets 83 84 for paleo-environmental reconstructions.

In this study, we present a continuous record of the Holocene climate obtained from a 7.2 m-85 86 long sediment core retrieved from the Rhone mud belt in the Gulf of Lions. Owing to the high sedimentation rate of this environmental setting, we could generate sedimentological data at 87 88 decadal scale resolution for sediment grain size and semi-quantitative chemical composition obtained by mean of continuous X-ray fluorescence. Organic matter parameters and the 89 overall seismic architecture of the mud-belt were also used to reconstruct the terrigenous flux 90 and the degree of alteration of land-derived material for investigating the relationship between 91 92 detritic fluxes and the paleohydrology of peri-Mediterranean rivers. Based on the comparison of available data, we explored the linkages between rapid climate changes and continental 93 paleo-hydrology with a focus on the Rhone river flood activity. 94

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#### 96 2. Environmental and climatic framework

#### 97 2.1. The Gulf of Lions geological and oceanographic settings

98 The Gulf of Lions (GoL) is a passive and prograding continental margin with a relatively 99 constant subsidence and a high sediment supply (Berné and Gorini, 2005). Located in the 100 north-west sector of the Mediterranean Sea, the GoL is bounded to the West and to the East





101 by the Pyrenean and Alpine orogenic belts, and comprises a crescent-shaped continental shelf with maximum width of 72 km near the mouth of Rhone (Berné et al., 2004). The general 102 103 oceanic circulation is dominated by the geostrophic Liguro-Provencal or Northern Current (Millot, 1990), which is the northern branch of the general cyclonic circulation in the western 104 Mediterranean basin. This current flows southwestward along the continental slope and 105 temporally intrudes on the continental shelf during northwesterly winds events (Millot, 1990; 106 Petrenko, 2003). Surface water circulation in the GoL shelf is wind-dependent (Millot, 1990). 107 108 Different wind patterns affect the circulation and transport of suspended particles on the shelf 109 and produce distinctive wave regimes. The continental cold and dry winds known as the Mistral and Tramontane, blowing from the N and NW through the passages between the 110 Pyrenees, Massif Central and the Alps, are associated with a short fetch that generate small 111 waves on the inner shelf. During winter, these winds induce strong cooling and mixing of the 112 113 shelf-waters triggering dense water formation (Estournel et al., 2003) and locally generating 114 upwelling (Millot, 1990). Episodic and brief E-SE (Marin or Maritime regime) winds are associated with long fetch and large swells. This wind regime induces a rise in sea level along 115 116 the shore and intense cyclonic circulation on the shelf (Ulses et al., 2008) producing alongshore currents and down-welling (Monaco et al., 1990). Transport of humid marine air 117 masses over the coastal relief induces abundant precipitations often accompanied by river 118 119 flooding.

The main source of sediment in the GoL is the Rhône River (Pont et al., 2002) and to a lesser 120 extent, small rivers of the Languedoc-Roussillon region (Hérault, Orb, Aude, Agly, Têt, Tech) 121 122 (Figure 1). The latter experience episodic discharges (*flash floods* in spring and fall) that are 123 difficult to quantify. The terrigenous sediment supply originating from the Rhone River represents 80% of the total sediment deposited on the shelf (Aloisi et al., 1977). The Rhone 124 River drains a largely mountainous catchment area of 97 800 km<sup>2</sup> incising a geologically 125 heterogeneous substrate, consisting of siliciclastic and carbonate sedimentary rocks in valley 126 infills and a crystalline (plutonic and metamorphic from the Alpine domain) bedrock. The 127 mean annual water discharge measured at Beaucaire gauging station, downstream the last 128 confluence is 1,701 m<sup>3</sup> s<sup>-1</sup> (mean for 1961-1996); the solid discharge varies between 2 to 20 129  $10^{6}$  tons yr<sup>-1</sup> (Eyrolle et al., 2012; Pont et al., 2002). 130

Most of the sediment delivered by the Rhone is trapped on the inner shelf mainly in prodeltas (Fanget et al., 2013; Ulses et al., 2008) but redistribution processes operating along the shelf create mid-shelf depocenters of fine sediments. The sediment accumulation rate varies from





134 20 to 50 cm yr<sup>-1</sup> at the present Roustan mouth of the Rhone River and strongly decreases with the distance from the river. Sediment is exported seaward by several turbid layers: the surface 135 136 nepheloid layer, related to river plume; an intermediate nepheloid layer that forms during periods of water-column stratification; and a persistent bottom nepheloid layer whose 137 influence decreases from the river mouth to the outer shelf (Calmet and Fernandez, 1990; 138 Naudin et al., 1997). The surficial plume is typically a few meter-thick close to the mouth but 139 rapidly thins seaward to few centimeters (Millot, 1990); it is deflected southwestward by the 140 141 surface water circulation on the GoL shelf. The predominance of the Rhone River in the sediment supply and the continental shelf circulation allow the identification of several zones 142 in the GoL (Durrieu De Madron et al., 2000): i) the deltaic and prodeltaic sediment units 143 where most of the sediments are trapped, ii) the mid-shelf mud belt between 20 and 50-90 m 144 depth resulting from sediment transport under the influence of the main cyclonic westward 145 146 circulation and iii) the outer shelf where fine-grained sedimentation is presently very low and 147 where relict fine sands are episodically reworked during extreme meteorological events (Bassetti et al., 2006). 148

#### 149 2.2. Holocene paleohydrology in the western Mediterranean

The Mediterranean hydrology is controlled by the seasonality of precipitation as well as the 150 catchment geology, vegetation type and geomorphology of the region. In northwestern 151 Mediterranean the most important fluvial discharges occur in spring and autumn, while 152 153 minimum flow is observed in summer (Thornes et al., 2009). On Holocene time scale, the Mediterranean fluvial hydrology is characterized by the alternation of wet and dry episodes 154 related to changes in atmospheric circulation leading to a North-South hydrological contrast 155 in the Mediterranean region with climate reversal occurring at about 40°N (Magny et al., 156 157 2013). Complex climate regimes result from external forcings (orbital, solar activity, volcanism) as well as from internal modes of atmospheric variability such as the North 158 Atlantic Oscillation, East Atlantic, East-Atlantic-West Russian or Scandinavian modes (Josey 159 et al., 2011; Magny et al., 2013). 160

In the NW Mediterranean, the Holocene fluvial hydrology has been reconstructed using major hydrological events (extreme floods and lake levels) recorded in lake and fluvial sediments (Arnaud et al., 2012; Benito et al., 2015; Magny et al., 2013; Wirth et al., 2013). Overall, the early Holocene climate was generally dry except for short pulses of higher fluvial activity reported in the Durance and southern Alps rivers (Arnaud-Fassetta et al., 2010). A marked cooling trend is observed with a major change around 7,500 a cal BP (Fletcher and Sánchez





167 Goñi, 2008) corresponding to humid conditions in the Iberian peninsula (Benito et al., 2015). The mid-Holocene (from ca. 7,000 to 5,000 a cal BP) also records low torrential activity but 168 169 increasing flood frequency between 6,000 and 4,500 a cal BP in Spain, Tunisia and southern France (Arnaud-Fassetta, 2004; Benito et al., 2003; Faust et al., 2004) that evolves in the late 170 Holocene to a general increase of fluvial activity, at least in the Rhone basin catchment and 171 north Alps domain (Wirth et al., 2013). In addition, anthropogenic activities (agriculture and 172 deforestation) over the last 5,000 years have modified the erosional rate in the catchment area, 173 resulting in increased/decreased sediment delivery to the sea depending on the 174 175 deforestation/forestation phases related to the agricultural development (Arnaud-Fassetta et al., 2000; van der Leeuw, 2005). 176

#### 177 2.3. Deglacial and Holocene history of the Rhone Delta

During the last ca. 20 ka, the morphology of the Rhone delta strongly evolved in response to 178 sea-level and climate changes. At the end of the Last Glacial Maximum, the Rhone reached 179 the shelf edge and directly fed the Petit Rhone Canyon (Figure 1) (Lombo Tombo et al., 180 2015). The disconnection between the river and the canyon head is dated at 19 ka cal BP in 181 182 response to the acceleration of sea-level rise (*ibid.*). The retreat path of the estuary mouth on the submerged shelf has be tracked through the mapping and dating of paleo-deltas (Berné et 183 184 al., 2007; Fanget et al., 2014; Gensous and Tesson, 2003; Jouet, 2007; Lombo Tombo et al., 2015) and, onshore, through the study of ancestral beach ridges (Arnaud-Fassetta, 1998; 185 L'Homer et al., 1981; Vella and Provansal, 2000). During the Younger Dryas, an "Early 186 Rhone Deltaic Complex" (ERDC) formed at depths comprised between -50 and -40 m below 187 present sea level (Berné et al., 2007). The estuary then shifted to the NW as sea-level rose 188 during the Early Holocene (Fanget et al., 2014). The period of maximum flooding in the delta 189 (the turnaround between coastal retrogradation and coastal progradation) is dated at ca. 8,500 190 -7,500 a cal BP (Arnaud-Fassetta, 1998). Around this time, the mouth of the Rhone was 191 situated about 15 km North of its present position. Between this period and the Roman Age 192 193 (approximately 20 BC-390 AD in Western Europe), the position of the Rhone outlet(s) are not precisely known and many distributaries, with their associated deltaic lobes, have been 194 identified. However, there is a general consensus on the eastward migration of the delta from 195 the St Ferreol Distributary that occupied the position of the modern Petit Rhone between ca. 196 6,000 and 2,500 a cal BP, and the modern Grand Rhone, built at the end of the 19th century. 197 198 To the West of the Rhone, a mud belt/subaqueous delta, about 150 km in length, up to 20 m





- 199 thick, is observed (Figure 1). So far, little attention has been paid to this sediment body, and
- 200 neither seismic data nor detailed core analysis were available.

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#### 202 3. Material and methods

203 The gravity core KSGC-31 was retrieved from the Rhone mud belt (43°0'23''N; 3°17'56''E, water depth 60 m) during the GM02-Carnac cruise in 2002 on the R/V "Le Suroît". Seismic 204 data were acquired in 2015 aboard R/V Néréis during the Madho1 cruise, using an SIG<sup>TM</sup> 205 sparker. The shooting rate was 1s. Data were loaded on a Kingdom<sup>TM</sup> workstation. An 206 average seismic velocity of 1,550 ms<sup>-1</sup> (based on measurements of sonic velocity with a 207 Geotek<sup>TM</sup> core logger) was used to position the core data on seismic profiles. The uncertainty 208 in the position of time lines on the seismic profile at the core position is on the order of  $\pm$ 209 0.5 m, taking into account the resolution of the seismic source, the errors in positioning and 210 211 sound velocity calculation. Due to the shallow water depth, core deformation by cable stretching is considered as negligible. 212

Grain size analyses were carried out by mean of a Malvern<sup>TM</sup> Mastersize 3000 laser 213 diffraction particle size analyzer using a HydroEV dispersing module, which measures 214 particle grain-sizes between 0.04 and 3,000 µm. Samples were dispersed in a solution of 215 (NaPO<sub>3</sub>)<sub>6</sub> (1.5 gr/l of distilled water) for 1 hour in order to better disaggregate the sediment. 216 Before each measurement, the sample was stirred on a rotating mixer during 20 minutes. 217 Grain-size parameters were measured all along the core every cm. Three size ranges were 218 used to classify the grains: clay (<8 µm, as recommended by Konert and Vandenberghe 219 (1997), coarse silt (>8  $\mu$ m and <63  $\mu$ m), sand (>63  $\mu$ m and < 250  $\mu$ m). The D50, representing 220 221 the maximum diameter of 50% of the sediment sample was calculated.

Core KSGC31 was analyzed using an Avaatech XRF Core Scanner at IFREMER (Brest, 222 223 France). This non-destructive method provides semi-quantitative analyses of major and minor elements by scanning split sediment cores (Richter et al., 2006). Measurements were 224 performed every 1 cm with a counting time of 20 sec and a 10kV and 30kV acceleration 225 226 intensity. Resulting element abundances are expressed as element-to-element ratio. Three 227 ratios are used in this work: 1) Ca/Ti ratio, to account for two end-members in the sediment composition. The Ca is supposedly mostly derived from biogenic carbonates, while Ti is 228 commonly used for tracking terrigenous sediments, even if usually found in small amounts. 229 Nonetheless, it is worthwhile reminding that calcite of detritic origin might be important in 230





231 the fluvial Rhône waters but this kind of CaCO<sub>3</sub> is mainly accumulated in the sand fraction and quickly decreases seaward of the river mouth (Chamley, 1971). A small fraction of 232 233 detritic calcite can be preserved in the clay fraction but represents a minor component; 2) Zr/Rb reflects changes in grain size, with higher values in the relatively coarse grained 234 sediments. Zr is enriched in heavy minerals and commonly associated with the relatively 235 coarse-grained fraction of fine-grained sediments (highest Zr values are found in sandstones), 236 whereas Rb is associated with fine-grained fraction, including clay minerals and micas 237 (Dypvik and Harris, 2001); 3) K/Ti values can be related to illite content. Illite is formed by 238 weathering of K-feldspars under subaerial conditions and most of the K leached from the 239 rocks is adsorbed by the clay minerals and organic material before it reaches the ocean 240 (Weaver, 1967). In the case of the GoL, the Rhône waters deliver mainly illite and chlorite to 241 the Mediterranean Sea whereas rivers flowing from Massif Central, Corbières and Pyrénées 242 mainly carry illite and montmorillonite (Chamley, 1971). Thus, illite (K) is thought to be 243 abundant in fluvial waters ending in the GoL, and thus K relative abundances can be used as a 244 proxy for sediment continental provenance. Because illite might be depleted in K upon 245 246 pedogenetic processes, the K/Ti ratio can be considered as an indirect proxy for the intensity of chemical weathering (Arnaud et al., 2012). The XRF raw data were smoothed using a 5-247 point moving average to remove background noise. 248

In addition semi-quantitative bulk geochemical parameter such as total carbon (TC), organic 249 carbon (OC) and total nitrogen (TN) were determined from freeze-dried homogenized and 250 precisely weighed sub-samples of sediment using the Elementar Vario MAX CN automatic 251 elemental analyzer. Prior to the OC analyses, samples were acidified with 2M HCl overnight 252 at 50°C in order to remove carbonates (Cauwet et al., 1990). The precision of TC, OC and TN 253 254 measurements was 5 and 10%. The calcium carbonate content of the sediments was calculated from TC-OC using the molecular mass ratio (CaCO<sub>3</sub>: C= 100:12). Results are expressed as 255 the weight percent of dry sediment (% d.w.). The atomic C:N ratio (C:N<sub>a</sub>) was calculated and 256 used as a qualitative descriptor of organic matter (OM). Moloney and Field (1991a) proposed 257  $C:N_a = 6$  for OM of marine origin because of the high protein content of organisms such as 258 phytoplankton and zooplankton. Higher plant-derived OM of terrestrial origin have higher 259 C:N<sub>a</sub> ratios (>20) than marine organisms because of a high percentage of non-protein 260 constituents (Meyers and Ishiwatari, 1993). In marine sediment, C:Na ratios are usually higher 261 262 than phytoplankton. C:Na ratios comprised between 6 and 10 are indicative of degraded 263 organic detritus resulting from the breakdown of the more labile nitrogenous compounds and





values of C:N<sub>a</sub> ratio > 13 indicate a significant contribution of terrestrial organic matter (Goñi
et al., 2003).

- The age model is based on 21 radiocarbon dates (Table 2) obtained by Accelerator Mass 266 Spectrometry (AMS) at the Laboratoire de Mesure du Carbone 14, Saclay (France). The two 267 uppermost dates were performed at Beta Analytic Radiocarbon Dating Laboratory and 268 indicate post-bomb values (AD 1950). The  $^{14}\!C$  dates were converted into  $1\sigma$  calendar years 269 using Calib7.1 (Stuiver and Reimer, 1993) and the MARINE 13 calibration dataset including 270 the global marine reservoir age (400 years) (Charmasson et al., 1998). We used a local marine 271 272 reservoir age correction of  $\Delta R = 23 \pm 71$  years (http://calib.gub.ac.uk/marine/regioncalc.php). The age model was obtained by polynomial interpolation between <sup>14</sup>C dates excluding the 273 minor reversal at 18.5 cm ( $350 \pm 78$  yrs) and the two post-bomb dates. Timing and 274 uncertainty of CR1 to CR6 is estimated using the Bayesian approach of OxCal 4.2 (Ramsey 275 and Lee, 2013) (Table 3). We used the same age model as in Jalali et al. (this volume). Age 276 inversions are not used in the estimation of the sedimentation rate (SR) (Table 2). 277
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#### 279 4. Results

#### 280 4.1. Age model, sedimentological core description

Based on the 21 <sup>14</sup>C dates, the average SR has been estimated to ~0.70 m/1,000 years. The absolute chronology allows identifying three stratigraphic intervals corresponding to the formal subdivision of the Holocene epoch proposed by (Walker et al., 2012). The well-known cold events (Cold Relapses, CRs) are defined on the basis of this chronology (Figure 3, Table 1).

The core is predominantly composed of silt (60-70%) and clay. The clay content is highly 286 variable but no more than 50% between 10,000 and 4,000 a cal BP, and between 50 and 60% 287 288 in its upper 350 cm corresponding to the last 4,000 years (Figure 3). Small-size shell debris are randomly mixed with the clayey silt but become more abundant between 400 and 500 cm 289 depth. Abundant and well-preserved Turritella sp shells certainly not reworked are found 290 291 between 680 and 640 cm. The sand fraction is generally very low (0.5-5%) except for the lowermost 30 cm (50%, Figure 3). At visual inspection, the thin sandy base (between 703 and 292 293 690 cm) contains very abundant shell debris. Weak bioturbation is visible on the X-ray images as well as the occurrence of sparse articulated shells. 294

Core KSGC31 was retrieved at the seaward edge of the Rhone mud belt. The seismic profileat the position of the core displays the architecture of this mud belt that drapes Pliocene rocks





297 and continental deposits of the Last Glacial Maximum (Figure 2). The bottom of the core corresponds to the ravinement surface (RS in Figure 2) that formed by wave erosion at the 298 299 time of marine flooding during the deglacial period. This heterolitic material includes fluvial and coastal sands and gravels mixed with marine shells in a muddy matrix. At the position of 300 core KSGC31, it is postdated by the overlying muds immediately above (ca. 10,000 a cal BP). 301 The period of "turn around" between coastal retrogradation and coastal progradation is well 302 marked on the seismic profile by a downlap surface dated at ca. 7.5 ka cal BP at the position 303 304 of the core. It corresponds to the Maximum Flooding Surface in the sense of Posamantier and 305 Allen (1999). Two other distinct seismic surfaces (higher amplitude, slightly erosional) can be recognized in the upper part of the wedge (Figure 2), they are dated at ca. 4.2 and 2.5 ka 306 307 cal BP from the core.

#### 308 4.2. Elemental and geochemical distribution

Ca/Ti, K/Ti and Zr/Rb ratios were generated and cross-analyzed with grain-size (clay content
 -D50 computed curve) and C:Na to assess changes in geochemical composition.

In the Early Holocene, the Ca/Ti ratio is fairly constant and relatively high. The carbonate 311 content is high (>45% CaCO<sub>3</sub>, Figure 4b), whereas C:N<sub>a</sub> values are highly fluctuating 312 between values of 20 (~ 10 ka) and lower values of 13 towards the mid-Holocene (Figure 4a). 313 Zr/Rb ratios gradually decrease while K/Ti show a relatively stable behavior. Between 7,000 314 and 9,000 a cal BP, K/Ti and Zr/Rb indicate lower values, yet with a peak in the mid-interval, 315 around 8,200-8,300 a cal BP (Figure 4d,e). All over the period, clay content is comprised 316 approximately between 24 and 52% (Figure 5c), D50 is generally >10 µm and variable 317 (Figure 4f). A significant drop of Ca/Ti and D50 are observed in the 7,000-6,400 a cal BP 318 interval (Figure 4c, f). Similar trends are observed for the K/Ti and Zr/Rb, but the most abrupt 319 320 drop occurs between 6,500 and 6,400 a cal BP. No significant changes are detected in the main lithology (mostly clayey, Figure 3). C:Na ratios decrease (<13) due to a better 321 322 preservation of nitrogen in clay deposits.

After 6.4 ka cal BP, Ca/Ti a constant decreasing trend until 4,200 a cal BP. On the other hand, C:N<sub>a</sub> between 6,400 and 4,200 a cal BP reveals two prominent peaks (>15) culminating at 5,700 and 4,800 a cal BP (Figure 4a) that roughly correspond to low K/Ti and Zr/Rb values (Figure 4d, e), higher clay (Figure 5c) and lower carbonate sediment contents (Figure 4b). The most pronounced changes in the elemental ratio are observed after 4,200 a cal BP (Figures 4 and 5). Millennial-scale oscillations are discernible in the Ca/Ti record (Figure 4c) and coherent with changes in K/Ti and Zr/Rb ratios (Figure 4d, e) and, to some extent, with





330 the D50 values (Figure 4f). Six main episodes of high terrigenous inputs (lowest Ca/Ti) are clearly expressed in the XRF data at ~ 3,500  $\pm$  170, ~ 2,840  $\pm$  172, ~ 2,200  $\pm$  145, ~ 1,500  $\pm$ 331 332 124, ~ 1,010  $\pm$  75 and ~ 720  $\pm$  72 a cal BP (Figure 4, Table 4). Considering the age uncertainty, only some of those events might coincide with CRs (CR6, CR5, CR4, Figure 4). 333 334 The peaks in the clay content correspond to low Ca/Ti ratios of variable amplitude. The clay content of the 2,840 and 2,200 a cal BP events are among the highest (~35%, Figure 5e). 335 336 From 4,200 a cal BP to present, the C:N<sub>a</sub> values decrease gradually. Between  $\sim$ 4,200 and 3,200 a cal BP, some values exceed 13. Thereafter, the  $C:N_a$  values range between 9 and 10 337 (Figure 4a). The Late Holocene is also characterized by decreasing carbonate content with a 338 339 drastic drop around 2,000 a cal BP (Figure 4b). The SR is also higher than during the Mid-Holocene lying between 0.5 and 1 mm/year. 340

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#### 342 5. Discussion

343 Early Holocene (11.7-8.2 ka cal BP)

Based on seismic evidences, the KSGC31 basal deposits correspond to the early setting of the 344 345 mud belt unconformably lying on older fluvial sediments (Figure 2). The mud belt initiation phase is marked by coarse (silty-sand) grained sediments (coastal and deltaic) with abundant 346 347 shell debris transported when the relative sea-level was -30/40 m lower than today. This interval is marked by highest SR values and high terrestrial supply as also indicated by the 348 349 high C:N<sub>a</sub> ratio (>13) (Figure 4a) (Buscail and Germain, 1997; Buscail et al., 1990; Gordon and Goñi, 2003; Kim et al., 2006). Of note, the C:Na ratios ~ 20 indicative of even larger 350 351 enrichment in organic material originating from soils or plant debris in the coarse deposit at the very bottom of the core (700 cm) (Hedges and Oades, 1997{Meyers, 1993 #2518; Meyers 352 353 and Ishiwatari, 1993). A layer of high Turritella abundances is identified in the fine-grained sediments just above the sandy interval (680-640 cm, i.e. 8,500 -8,000 a cal BP) (Figure 3). 354 Then Turritella shells disappear gradually towards the top of the core, suggesting an upward 355 deepening environment. The high Turritella level could indicate a change in Northern 356 357 Hemisphere climate and can be hypothetically related to the "Turritella Layer" described by 358 Naughton et al. (2007) on the NW Atlantic shelf, therefore suggesting a regional change between 8,700 and 8,400 a cal BP, possibly in relation with the southward migration of the 359 360 Boreal biogeographical zone. The Maximum Flooding Surface (MSF) is dated around 7,500 a cal BP (Figure 2). This age may vary at different locations because it depends upon the ratio 361 362 between sediment delivery and accommodation space, but it matches well the age of delta initiations observed worldwide by Stanley and Warne (1994). 363





The increase of K/Ti between 9,000 and 7,000 a cal BP might reflect the gradual decrease of the contribution of weathered material from the river catchment areas, which can thus be interpreted as a signal of weaker pedogenetic processes and lower soil erosion due to dry climate in European Alps (Figure 4d) (Arnaud et al., 2012).

The period between ~ 12,000 and 7,000 a cal BP is marked by a continuous retreat of Arctic 368 continental ice-sheets until the complete disappearance of the Fennoscandian and Laurentide 369 ice cap (Tornqvist and Hijma, 2012; Ullman et al., 2015). Ice sheet melting is seen in the 370 371 general sea-level rise and also manifested by short-lived water releases into the ocean when sufficient enough to perturb the North Atlantic Ocean circulation and climate over Europe (for 372 example the 8,200 cal BP event, here CR0). It is worthwhile to note that around CR0, the 373 K/Ti ratio shows a peak within an interval of low values between approximately 7,900 and 374 8,300 a cal BP. This peak would identify an increase of continental supply and low chemical 375 weathering corresponding to cold (weak soil formation) and wet (high physical erosion) 376 377 conditions over mid-latitude Europe in response to the 8,200 a cal BP cooling (Arnaud et al., 2012; Magny et al., 2003) also attested by higher lake levels in Western Europe (Figure 5d, f) 378 379 concurrent with CR0 (Magny et al., 2013). Note that no clear temperature drop in the alkenone-derived SST record generated in the core has been detected (Jalali et al., this 380 381 volume).

382

383 Mid-Holocene (8.2-4.2 ka cal BP)

Values of Ca/Ti and C:Na ratios indicate a progressive increase of terrestrial inputs (Figure
4ac) during the Mid-Holocene, despite increasing distance of KSGC31 site from river outlets
due to sea-level rise and the progressive shift of the Rhone delta to the East (Fanget et al.,
2014).

The Ca/Ti ratio values drop drastically between 7,000 and 6,400 a cal BP. In Eastern 388 Mediterranean enhanced sediment delivery is documented around 7,000 a cal BP (Castañeda 389 390 et al., 2016; Skonieczny et al., 2015) during the Saharan Humid Period (SHP). In Western 391 Mediterranean, torrential discharges are observed in most rivers of Southern France and thought to be responsible for a regional episode of river incision between 7,200 and 6,800 a 392 cal BP (Benito et al., 2015). These events coincides with the deposition of large amount of 393 fine-grained sediments at the site of KSGC31 (Figure 4). We thus hypothesize high fluvial 394 395 activity between 7,000 and 6,400 a cal BP, coinciding with fine-grained deposits, the coarser fraction being trapped in the deltaic system (Fanget et al., 2014). Possible connections 396 between runoff from the South Mediterranean as a consequence of a strengthened North 397





African monsoon and increased precipitations over the Mediterranean basin itself have been
recently investigated (Bosmans et al., 2015; Kutzbach et al., 2013).

400 Our observations raise the question if an increase in the discharge of NW Mediterranean 401 rivers might be synchronous with the SHP. Some climate archives like speleothems and lake 402 records show evidence for a substantial increase in precipitation both in the Western and 403 Eastern Mediterranean (Zanchetta et al., 2007; Zhornyak et al., 2011). However, in the case of 404 Rhone River, we cannot rule out the possibility that alpine glacier meltwater could have at 405 least in part contributed to increase the Rhone River discharge.

The Mid-Holocene is described as a period of relatively mild and high atmospheric moisture 406 balance (Cheddadi et al., 1998) that favored the maximum expansion of the mesophytic forest 407 leading to a maximum land cover over Europe. Nonetheless, two main short-lived climate 408 anomalies are reported at 6,600 - 5,700 a cal BP (CR1, Tables 2 and 3) and 5,300 - 5,000 a cal 409 BP (CR2, Tables 2 and 3) over the North Atlantic (Wanner et al., 2011) and in Europe 410 411 (Magny and Haas, 2004; Roberts et al., 2011a; Roberts et al., 2011b), at the time of global cooling. CR1 is associated with drying climate in eastern Europe and Asia and has been 412 413 related to the weakening of the Asian monsoon and the decrease of summer insolation (Gasse et al., 1991), while CR2 coincides with weaker solar activity as indicated by maximum 414 atmospheric <sup>14</sup>C around 5,600–5,200 a cal BP (Stuiver et al., 2006), lower tree lines (Magny 415 and Haas, 2004) and colder sea-surface temperatures (Jalali et al., this volume). 416

According to our data, during CR1 and CR2, there was no significant change in terrigenous 417 inputs (Ca/Ti ratio, Figure 4c), mean SR was generally low (<1 mm/yr on average, Table 2) 418 and chemical weathering was weak as suggested by high K/Ti values (Figure 4d). The C:Na 419 ratios indicate better preservation of nitrogen organic compounds preferentially adsorbed in 420 the clay fraction (Figure 4a). The reduction of the vegetation cover in the river catchment, 421 combined with lower (1-1.5°C) temperatures in the European Alps (Haas et al., 1998), may 422 explain the low chemical degradation state of the illite minerals. A drop in sea surface 423 424 temperature is also recorded by alkenones in the core (Jalali et al., this volume), confirming 425 the global climate impact of the cold relapses in the Mediterranean area (Figure 4i).

426

#### 427 Late Holocene (4.2-0 ka cal BP)

Multi-decadal to century-scale wet episodes are evidenced from ~ 4,200 a cal BP, that marks
the Mid-Late Holocene transition (Figure 3). In the KSGC31 core, wetter intervals are
expressed by highly fluctuating Ca/Ti, Zr/Rb and K/Ti ratios, C:N<sub>a</sub> and grain size values.





431 From present to 3.5 ka cal BP, the C:Na ratio shows an increase from 9 to 11 (Figure 4) testifying active diagenetic processes, due to preferential degradation of nitrogen relative to 432 433 carbon during burial. Some values > 13 are still observed between 4,200 and 3,500 a cal BP, indicating enhanced terrestrial inputs. Episodes of enhanced terrigenous inputs are detected by 434 low Ca/Ti ratios that also coincide with low Zr/Rb and low D50 values, indicating general 435 smaller-size terrigenous grains as also suggested by high clay content (Figure 5c). Thus, 436 intensified hydrological activity associated with high terrestrial inputs would have prevailed 437 during the Late Holocene, as also suggested by higher SR (Table 2). The enhanced terrestrial 438 inputs are inferred from XRF ratios discussed here, but also by the biomarker data. Indeed, 439 Jalali et al. (this volume) also highlighted enhanced flood activity during the Late Holocene 440 based on TERR-alkane concentrations, which are among the highest of the entire Holocene 441 442 record.

A similar signature of continental runoff in marine sediments (low Ca/Ti ratio) during the past 443 ~ 6,500 years has been reported in the central Mediterranean and related to climatically driven 444 wet periods (Moloney and Field, 1991b). Indeed, some of these events (~2840 a, ~ 1500 and 445 446 ~720 a cal BP) encompass cold events in the North Atlantic (LIA-CR6; Dark Age-CR5; CR4, Figure 4) and coincide with periods of increasing flood frequency in the Northern Alps as 447 reconstructed by Wirth et al. (2013) (Figure 5d). However, other humid episodes occur during 448 warmer periods such as the MCA at ~ 3,500; ~ 2,200 and ~1,000 a cal BP (Figure 4) 449 suggesting different causes for enhanced precipitations. 450

451 The North Atlantic Oscillation (NAO) exerts a strong influence on the precipitation pattern in Europe and the NW Mediterranean region. Today, precipitation in the western Mediterranean 452 region and southern France is lower during positive NAO. Rainfall increases under negative 453 NAO due to the southern shift of the Atlantic storm tracks leading to enhanced cyclogenesis 454 455 in the Mediterranean Sea (Trigo et al., 2000). The position of the ITCZ is also important in the precipitation pattern of the Mediterranean region and its southernmost position is the 456 457 probable cause for extremely dry conditions between 2,500 and 2,000 a cal BP 458 (Schimmelpfennig et al., 2012). The reconstructed NAO index (Olsen et al., 2012) indicates a predominance of positive state between 5,000-4,500 and 2,000-550 a cal BP, in agreement 459 with a) an increased frequency of floods in Northern Alps (Wirth et al., 2013), b) higher lake 460 levels at Accesa (Central Italy), Ledro (Northern Italy) and in Central-Western Europe 461 462 (Magny et al., 2013), all together suggesting more humid conditions in west-central Europe (Figure 5). However, between 2,500 and 2,000 a cal BP, very negative NAO and 463





southernmost position of the ITCZ (Haug et al., 2001) would have led to several centuries ofextreme dryness.

466 Late Holocene human settlements along the Rhone valley and South France may have had an impact on the origin and amounts of eroded sediments in the river catchment areas. When 467 examining the chemical signature of KSGC-31 sediments, low K/Ti ratios co-eval with wet 468 events (Figure 4d) reflecting soil weathering due to terrain degradation, that could be 469 interpreted as the result of widespread deforestation for agropastoral activities in this area 470 471 since the end of the Neolithic. The most extensive erosion episodes in the Rhone valley correspond to 1) the end of Neolithic (~ 4,000 BC; 6,000 a cal BP) after the first phase of 472 human expansion linked to the development of the agriculture 2) the end of the Bronze Age 473 (~ 2,000 BC; 4,000 a cal BP) and 3) the Roman Period when a rapid transformation of 474 landscape is operated by deforestation and their replacement by intensively cultivated 475 476 agricultural land (van der Leeuw, 2005).

477 Disentangling human impact from climate control on environmental changes in the Late Holocene is not an easy task, and requires the study of other river catchment basins to confirm 478 479 the regional character of these observations. However, assuming that climate variability is the major factor influencing soil pedogenesis, we can hypothesize that the elemental composition 480 of marine sediments reflects erosion and transport because of a good correspondence with 481 482 temperature variability in the Mediterranean Sea (Jalali et al., this volume), and because, on a regional scale, both marine and continental climate proxies indicate co-eval signals. Despite 483 the fact that the characteristics in amplitude and duration of these climate intervals slightly 484 differ geographically, there seems to be a general agreement on their origin and the role of 485 486 solar forcing and large-scale atmospheric circulation. However, amplification of soil degradation following human occupation waves should be further explored through accurate 487 correlation between archeological data and paleoenvironmental proxies in order to better 488 489 evaluate the importance of land use on sedimentary signals.

490

#### 491 **6. Conclusions**

This work represents the first attempt to detect and decipher the linkages between rapid
climate changes and continental paleo-hydrology in the NW Mediterranean shallow marine
setting during the Holocene.

Based on the combination of sedimentological and geochemical proxies we coulddemonstrate that between 11 and 4 ka cal BP, terrigenous input broadly increased and reached





497 a maximum around 7,000 a cal BP probably as a result of a more humid phase. From ca. 4,000 a cal BP to present, the sediment flux proxies indicate enhanced variability in the 499 amount of land-derived material delivered to the Mediterranean by the Rhone River input. We 500 suggest that this late Holocene variability is due to changes in large-scale atmospheric 501 circulation and rainfall patterns in Western Europe including the variability of continental 502 glaciers. Anthropogenic impact such as deforestation resulting in higher sediment flux into 503 the Gulf of Lions are also likely and should be better taken into account in the future.

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#### 812 Tables

#### 813 Table 1: Chronology of Holocene cold relapses (CR) based on existing literature.

Event	Time slice	References
	(ky)	
CR0	8.2	(Barber et al., 1999)
CR1	6.4-6.2	(Wanner et al., 2011)
CR2	5.3-5.0	(Magny and Haas, 2004; Roberts et al., 2011a)
CR3	4.2-3.9	(Walker et al., 2012)
CR4	2.8-3.1	(Chambers et al., 2007; Swindles et al., 2007)
CR5	1.45-1.65	(Wanner et al., 2011)
CR6	0.65-0.45	(Wanner et al., 2011)

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Denth	Material	Laboratory	Radiocarbon age	Calibrated	±1σ	Sedimentation
(cm)		Luboratory	±1σ error (vr BP)	Age (cal BP)	- 10	
()				go (	error	Rate (SR)
						(mm/year)
5.5	Bittium sp.	Beta Analytics	420 <u>+</u> 30	24ª	60	-
11.5	<i>T. U</i> :		420.20	2.43	(0)	
11.5	Tellina sp.	Beta Analytics	430±30	34"	60	-
18.5	Pecten sp.	Beta Analytics	$720 \pm 40$	350 <sup>b</sup>	78	-
25.5	Venus sp.	LMC14	640 ± 30	234	99	1,34
41	Pecten sp.	LMC14	700 ± 30	339	79	1,48
52	Indet. bivalve	LMC14	960 ± 30	551	59	0,52
71	Arca tetragona	LMC14	$1340 \pm 30$	851	80	0,63
110.5	Venus sp.	LMC14	1465 ± 30	992	85	2,80
1965	No	LMC14	2225 + 40	1905	00	0.02
180.5	<i>Nucuta</i> sp.	LMC14	$2235 \pm 40$	1805	99	0,95
251	Juvenile bivalve	LMC14	$2940\pm30$	2674	100	
	shells (ind.)					0,74
330.5	Venus cosina	LMC14	$3870\pm30$	3796	106	0,71
370.5	Nuculana sp.	LMC14	4170 ± 30	4223	113	0,94
390.5	<i>Turritella</i> sp.	LMC14	4500 ± 30	4676	106	0,44
460	Venus sp.	LMC14	$5530 \pm 45$	5873	106	0,58
481	Ostrea sp	LMC14	5955 ± 35	6348	78	0,44
501.5	<i>Turritella</i> sp.	LMC14	$6380 \pm 50$	6826	107	0,43
552	Shells (mixed)	LMC14	$7215\pm30$	7653	75	0,61
583	<i>Turritella</i> sp.	LMC14	7860 ± 60	8288	92	0,49
652	<i>Turritella</i> sp.	LMC14	8310 ± 35	8843	121	1,24
700.5	<i>Turritella</i> sp.	LMC14	9215 ± 30	10006	123	0,42
701	Turritalla sp	LMC14	0100 ± 50	0068	145	
/01	<i>Turrueuu</i> sp.	LIVIC 14	9190 ± 50	7700	145	-

### 817 Table 2: <sup>14</sup>C dates performed on core KSGC31





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Cold relapses	Central Age year BP $\pm 1\sigma$ uncertainty	Age interval year BP	Duration year	Amplitude °C
CR1	$6175 \pm 133$	6600 - 5750	850	$1.3\pm0.3$
CR2	$5195 \pm 196$	5350 - 5040	310	$1.3 \pm 0.3$
CR3	$4130\pm126$	4340 - 3920	420	$2.4\pm0.3$
CR4	$2355\pm142$	2530 - 2180	350	$1.4 \pm 0.3$
CR5	1365 ± 119	1770 - 960	810	$2\pm0.3$
CR6	$320\pm75$	490 - 150	340	$1.1\pm0.3$

819 Table 3: Timing of Holocene cold relapses (CRs; Jalali et al, this volume).

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Table 4: Time uncertainty  $(1\sigma)$  of "wet spells" identified on the Ca/Ti and K/Ti ratios

823 (Figure 4)

		Maximum			
Abbreviation	Start	(yr cal	±1 σ	End (yr cal	
in the text	(yr cal BP)	BP)	uncertainty	BP)	Proxy
0.72 ka	645	720	72	800	Ca/Ti
1.01 ka	1000	1015	75	1070	Ca/Ti
1.5 ka	1400	1500	124	1640	Ca/Ti
2.2 ka	2080	2200	145	2300	Ca/Ti
2.84 ka	2700	2840	172	2900	Ca/Ti
3.5 ka	3350	3500	170	3615	Ca/Ti
4.8 ka	4670	4800	150	4960	K/Ti
5.7 ka	5530	5700	162	5770	K/Ti

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Figure 1: Bathymetric map of the Gulf of Lions and position of core KSGC31. The
approximate extent of the Rhone mud belt is represented in green; the arrow represents the
direction of dominant transport of suspended sediments . Bathymetric map based on Berné et
al. (2007). Contour lines every 5 m on the shelf. The dotted line corresponds to the retreat
path of the Rhone during the Deglacial (based on Gensous and Tesson, 2003; Berné et al.,
2007; Jouet, 2007; Fanget et al., 2014, Lombo Tombo et al., 2015). ERDC: Early Rhone
Deltaic Complex.







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835 Figure 2: Seismic profile across the Rhone Mud Belt at the position of core KSGC31 836 (position in Figure 1). SB: Sequence Boundary- surface formed by continental erosion during the Last Glacial Maximum (LGM). RS: Ravinement Surface formed by wave erosion during 837 838 sea-level rise. It corresponds to the coarse interval at the base of the core. MFS : Maximum Flooding Surface (MFS). It corresponds to the phase of transition between coastal 839 retrogradation and coastal progradation. It is dated here at ca. 7.5 ka cal BP (i.e. the period of 840 global sea-level stabilization). S1 and S2 are seismic surfaces used as time lines (on the basis 841 of the age model in Figure 3 (respectively  $4.2 \pm 0.5$  ka cal BP and  $2.5 \pm 0.5$  ka cal BP). 842 843 Horizontal bars every meter along the core.







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Figure 3: Correlation age-depth in core KSGC31. The Holocene time is divided into Early,
Middle and late Holocene according Walker et al. (2012). General core lithology is shown
through distribution of three grain-size classes: <8μm, 8-63 μm, 63 μm.</li>

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852 Figure 4: KSGC31 geochemical and sedimentological proxies: (a) C:Na atomic ratio is used 853 as qualitative descriptor of organic matter nature. Values of  $C:N_a$  ratio > 13 indicate significant amount of terrestrial organic matter, according to Goñi et al. (2003); (b) CaCO<sub>3</sub> 854 content (%) calculated from TC-OC using the molecular mass ratio (CaCO<sub>3</sub>: C= 100:12); (c) 855 856 Ca/Ti ratio is used for estimating the degree of detritism, since Ti is commonly found in terrigenous sediments; (d) K/Ti ratio can be related to illite content, formed by weathering of 857 858 K-feldspars. Illite might be depleted in K upon pedogenetic processes, the K/Ti ratio can be considered as an indirect proxy for the intensity of chemical weathering (Arnaud et al., 2012); 859 860 (e) Zr/Rb is known to reflect changes in grain size. Zr is commonly associated with the relatively coarse-grained fraction of fine-grained sediments, whereas Rb is associated with 861 fine-grained fraction; (f) D50 represents the maximum diameter of 50% of the sediment grain 862 size. These plots are correlated to reconstruction of NAO (h) from a lake in Greenland (Olsen 863 864 et al., 2012) and reconstructed SST (C°) from alkenones (j) in core KSGC31 (Jalali et al., this 865 volume).





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Figure 5: Ca/Ti ratio (a) and percentage of fine-grained (<8µm) sediment (c) compared to: b)</li>
Periods of intense fluvial activity based on hydromorphological and paleohydrological
changes in the Rhone delta (Arnaud-Fassetta et al., 2010); d) Holocene flood frequency (%) in
Southern and Northern Alps estimated on the basis on lake flood records by Wirth et al.

872 (2013); e); f); g) lake level fluctuations in Central and Eastern Europe during the Holocene

873 (Magny et al., 2013)