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Climate variability and relationship with ocean fertility
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     during the Aptian Stage
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     Abstract
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17 Several studies have been conducted to reconstruct temperature variations across the Aptian 18 Stage, particularly during the Early Aptian Oceanic Anoxic Event (OAE)1a. There is a 19 general consensus that a major warming characterized the OAE 1a, although some studies 20 have provided evidence for transient 'cold snaps' or cooler intervals during the event. The climatic conditions for the middle-late Aptian are less constrained, and a complete record 21 22 through the Aptian is not available. Here we present a reconstruction of surface-water 23 palaeotemperature and fertility based on calcareous nannofossil records from the Cismon and 24 Piobbico cores (Tethys) and DSDP Site 463 (Pacific Ocean). The data, integrated with 25 oxygen-isotope and TEX₈₆ records, provide a detailed picture of climatic and ocean fertility 26 changes during the Aptian Stage, which are discussed in relation to the direct/indirect role of 27 volcanism. Warm temperatures characterized the pre-OAE 1a interval followed by a 28 maximum warming (of $\sim 2-3$ °C) during the early phase of anoxia under intense volcanic activity of the Ontong Java Plateau (OJP). A short-lived (~35ky) cooling episode interrupted 29 the major warming, following a rapid increase of weathering rates. Nannofossils indicate that 30 31 eutrophic conditions were reached when temperatures were at their highest and OJP 32 volcanism most intense, thus suggesting that continental runoff, together with increased input of hydrothermal metals, increased nutrient supply to the oceans. The latter part of OAE 1a 33 34 was characterized by cooling events, probably promoted by CO₂ sequestration during burial 35 of organic matter. In this phase, high productivity was probably maintained by N₂-fixing 36 cyanobacteria while nannofossil taxa indicating high fertility were rare. The end of anoxia 37 coincided with the cessation of volcanism and a pronounced cooling. The mid-Aptian was 38 characterized by high surface-water fertility and progressively decreasing temperatures, 39 probably resulting from intense continental weathering drawing down pCO_2 . The lowest 40 temperatures, combined with low fertility, were reached in the middle-late Aptian across the 41 interval characterized by blooming of Nannoconus truittii. The data presented suggest that OJP activity played a direct role in inducing global warming during the early Aptian, whereas 42 43 other mechanisms (weathering, deposition of organic matter) acted as feedback processes, favouring temporary cooler interludes. 44

45

46 **1** Introduction

The Aptian (~121 to ~113 Ma, Malinverno et al., 2012) has been characterized by climatic 47 changes and profound environmental perturbations including the Oceanic Anoxic Event 1a 48 49 (OAE 1a: ~120 Ma), representing a global phenomenon of organic-matter burial in oxygen-50 depleted oceans. The disturbance in the carbon cycle related to OAE 1a is recorded in 51 sedimentary successions worldwide, presenting a negative carbon-isotope anomaly at the 52 onset of OAE 1a, followed by a positive excursion that extends into the late Aptian (e.g. Weissert, 1989; Weissert and Lini, 1991; Jenkyns, 1995; Menegatti et al., 1998; Bralower et 53 54 al., 1999; Erba et al., 1999; Luciani et al., 2001; Bellanca et al., 2002; Price, 2003; van Breugel et al., 2007; Ando et al., 2008; Méhay et al., 2009; Malkoč et al., 2010; Mahanipour 55 et al., 2011; Millán et al., 2011; Stein et al., 2011; Bottini et al., 2012; Hu et al., 2012). 56 Volcanism, associated with the emplacement of the Ontong Java Plateau (OJP), is thought to 57 be the main triggering mechanism for global anoxia as well as for imposed greenhouse 58 59 conditions and ocean acidification during OAE 1a (e.g. Larson, 1991; Erba, 1994; Bralower et al., 1994; Larson and Erba, 1999; Jones and Jenkyns, 2001; Leckie et al., 2002; Jenkyns,
2003; Méhay et al., 2009; Tejada et al., 2009; Erba et al., 2010; Bottini et al., 2012)

62 Several studies are suggestive of significant temperature increase during OAE 1a, as recorded 63 by different temperature proxies (i.e. oxygen isotopes, TEX₈₆, calcareous nannofossils, palynomorphs) in the Tethys (e.g. Menegatti et al., 1998; Hochuli et al., 1999; Luciani et al., 64 65 2001; Bellanca et al., 2002; Jenkyns, 2003; Millán et al., 2009; Erba et al., 2010; Jenkyns, 2010; Keller et al., 2011; Stein et al., 2011; Bottini et al., 2012; Hu et al., 2012; Husinec et al., 66 67 2012), Vocontian Basin (e.g. Moullade et al., 1998; Kuhnt et al., 2011), Lower Saxony Basin (Mutterlose et al., 2010; Bottini and Mutterlose, 2012; Pauly et al., 2013), North Sea 68 69 (Mutterlose and Bottini, 2013), eastern European Russian Platform (Zakharov et al., 2013), 70 Pacific (e.g. Jenkyns, 1995; Price, 2003; Schouten et al., 2003; Ando et al., 2008; Bottini et 71 al., 2012), and Atlantic Oceans (e.g. Tremolada et al., 2006). Some works have provided 72 evidence for a climatic variability during OAE 1a, identifying short-lived cooling events (e.g. 73 Dumitrescu et al., 2006; Keller et al. 2011; Kuhnt et al., 2011; Jenkyns et al., 2012; Lorenzen 74 et al., 2013). At the end of OAE 1a, a temperature decline is registered in the Tethys (e.g. Weissert and Lini, 1991; Menegatti et al., 1998; Hochuli et al., 1999; Luciani et al., 2001; 75 Bellanca et al., 2002; Millán et al., 2009), Vocontian Basin (e.g. Herrle et al., 2010; Kuhnt et 76 al., 2011), Boreal Realm (e.g. Rückheim et al., 2006; Malkoč et al., 2010; Bottini and 77 78 Mutterlose, 2012; Pauly et al., 2013; Mutterlose and Bottini, 2013), and Pacific Oceans (e.g. 79 Jenkyns, 1995; Jenkyns and Wilson, 1999; Price, 2003; Dumitrescu et al., 2006; Ando et al., 80 2008).

For the late Aptian, cooler conditions have been reconstructed based on migration of boreal 81 species southwards (e.g. Herrle and Mutterlose, 2003; Mutterlose et al., 200 poxygen-82 83 isotope records (e.g. Weissert and Lini, 1991; Jenkyns, 1995; Hu et al., 2012; Price, 2012; 84 Maurer et al., 2012), putative ice-rafted debris in high latitudes (Kemper 1987; Frakes and Francis 1988; De Lurio and Frakes 1999; Price, 1999) and, for sea-bottom temperatures, the 85 presence of glendonites (marine low-temperature hydrated polymorphs of calcium carbonate), 86 (Kemper 1987). Recently McAnena et al. (2013) have documented, on the basis of TEX_{86} 87 data, a ~2 Myr long interval of relatively cool conditions (~ 28-29 °C) in the late Aptian in the 88 Proto-North Atlantic followed by a warming (up to ~31 °C), linked to OAE 1b. 89

Although the amount of information about temperature variations across the Aptian isconsiderable, a complete picture of climatic changes is not available. In most cases, the

92 records are poorly correlated between the different basins and/or cover limited time intervals within the ~ 12 My-long Aptian Stage (Malinverno et al., 2012). In this work, we focus 93 primarily on surface-water temperatures through the Aptian reconstructed on the basis of 94 95 calcareous nannofossils from three well sites: Cismon (Italian Southern Alps), Piobbico 96 (Umbria-Marche Basin, central Italy) and DSDP Site 463 (Mid-Pacific Mountains). The existing stratigraphic framework for the three sites and available cyclochronology for the 97 98 Cismon core (Malinverno et al., 2010), allow high-resolution dating of climatic fluctuations. 99 Calcareous nannoplankton live in the (upper) photic zone and are a good proxy of present and 100 past surface-water conditions, being sensitive to temperature, fertility, salinity and pCO_2 101 (Mutterlose et al., 2005). Extant calcareous nannoplankton occur from coastal areas to the 102 open ocean, although with different abundance and diversity and, together with diatoms, 103 dinoflagellates and bacteria constitute marine phytoplanktonic communities. The Mesozoic 104 geological record confirms the wide geographical/latitudinal distribution of calcareous nannofossils (coccoliths and nannoliths) that are commonly used to trace palaeoecological 105 106 conditions. Within nannofossil assemblages, nannoconids are inferred to have been restricted 107 to the deep photic zone at the base of the mixed layer on top of the thermocline coinciding 108 with a deep nutricline (Erba, 1994). In the studied intervals, nannoconids are relatively scarce, 109 and micrite mostly consists of coccoliths, thus essentially recording the uppermost water 110 masses.

In this work, stable carbon and oxygen isotopes on bulk rock have been measured to 111 112 reconstruct changes in surface-water temperature, taking into account potential diagenetic 113 modification. The preservation of nannofossils provides information on the early diagenetic 114 history of pelagic carbonates, (Erba, 1992b; Herrle et al., 2003; Tiraboschi et al., 2009). Although oxygen-isotope ratios contain a mixing of a primary signal and later diagenetic 115 phases (Marshall, 1992), hampering the use of palaeotemperature values, the δ^{18} O bulk data 116 can be used to derive trends toward warmer/cooler conditions. New oxygen-isotope data for 117 118 DSDP Site 463 and Piobbico have been generated, and are directly correlated with calcareous nannofossil variations as well as with new TEX_{86} data from the Cismon core. 119

120 The aims of our work are to: a) trace climatic variations during the Aptian Stage; b) 121 reconstruct, in high resolution, the climate variability through OAE 1a; c) identify 122 synchroneity and diachroneity of temperature variations in different oceanic basins; c) trace 123 the direct/indirect role of volcanism, weathering rates and pCO_2 on climate changes 124 connected with OAE 1a and its aftermath.

125 We also characterize the evolution of surface-water fertility during the Aptian Stage. Previous 126 studies (e.g. Coccioni et al., 1992; Bralower et al. 1993; Erba, 1994, 2004; Premoli Silva et 127 al., 1999; Leckie et al., 2002; Mutterlose et al., 2005; Tremolada et al., 2006; Bottini and 128 Mutterlose, 2012) mainly focused on the OAE 1a interval, documenting an increase in 129 surface-water fertility accompanied by high primary productivity, but a record throughout the 130 entire Aptian is missing. We therefore highlight fluctuations in fertility during and after OAE 131 1a, identifying potential relationships with climatic changes on both the short- and the long-132 term as well as oceanic nutrification.

133

134 2 Material and methods

135 2.1 Studied sites

We have investigated the Upper Barremian–Aptian interval at three sites in the Tethys andPacific Oceans (Figure 1):

138 The **Cismon core**, drilled in the Southern Alps, north-eastern Italy (46°02'N; 11°45'E; 398 m 139 altitude) is represented by a total stratigraphic thickness of 131.8 m with 100% recovery. The 140 site was located on the southern margin of the Mesozoic Tethys, on the eastward deepening 141 slope between the Trento Plateau (a pelagic submarine high) and the Belluno Basin (Erba and 142 Tremolada, 2004). The Cismon sequence was deposited at an estimated palaeo-depth of 1000-143 1500 m during the Early Cretaceous (Weissert and Lini, 1991; Erba and Larson 1998; 144 Bernoulli and Jenkyns, 2009). In the uppermost part of the cored section (at 7.80 m) there is a major hiatus corresponding to the late Aptian and the early-middle Albian. The Selli Level 145 (sedimentary expression of OAE 1a) is represented by a ~5 m-thick interval, between 23.67 146 and 18.64 stratigraphic metre depths (Erba and Larson, 1998; Erba et al., 1999). 147 148 Lithologically, the Selli Level is characterized by marlstones alternating with black shales and 149 discrete radiolarian-rich beds (Coccioni et al., 1987; Erba et al., 1999). The interval studied 150 extends from 35 m to 10 m.

The **Piobbico core** was drilled at "Le Brecce" (43°35′ 3.78′′N; 12°29′ 10.09′′E), located 3 km west of the town of Piobbico (Marche, Italy), at Km 33 of the Apecchiese State Road No. 153 257, on the left hydrographic side of the Biscubio stream. Coring penetrated the entire Marne 154 a Fucoidi Formation, including the upper transition to the Scaglia Bianca and the lower 155 transition to the Maiolica. The total length of the core is 84 m with 98.8% recovery; after 156 adjusting for dip direction, the stratigraphic thickness equals 77 m. The lithostratigraphy and 157 calcareous plankton biostratigraphy of the core were described by Erba (1988, 1992a) and 158 Tornaghi et al. (1989). The Selli Level, consisting of black shales and radiolarian-rich beds, 159 extends from 75.94 to 73.47 m. The interval studied covers the interval from 77 m to 40 m.

DSDP Site 463 was drilled at a water depth of 2525 m on the ancient structural high of the western Mid-Pacific Mountains (21°21.01′N, 174°40.07′E) during DSDP Leg 62. During the Early Cretaceous, Site 463 was located at a palaeo-latitude of ~20°S, with a palaeo-depth between a few hundred metres (Mélières et al., 1978) and ~1 km (Roth, 1981). The Selli Level equivalent is located between ~626 and 615 mbsf, corresponding to ~12 m of tuffaceous limestones containing a number of discrete organic-rich horizons (Thiede et al., 1981; Erba, 1994). The interval studied covers from 650 to 515 mbsf.

167 **2.2 Calcareous nannofossils** \bigcirc

168 Calcareous nannofossils were investigated under polarizing light microscope at 1250X 169 magnification in smear slides and thin-sections. Smear slides were prepared using standard techniques, without centrifuging or cleaning in order to retain the original sedimentary 170 171 composition. A small quantity of rock was powdered in a mortar with bi-distillate water and mounted on a glass slide with Norland Optical Adhesive. A total of 285 smear slides for the 172 173 Cismon core, 179 smear slides for the Piobbico core and 281 smear slides for DSDP Site 463 174 were investigated. At least 300 nannofossil specimens were counted in each sample and percentages of single taxa were calculated relative to the total nannoflora. 175

176 Thin-sections were polished to an average thickness of 7μ m for optimal view of nannofossils; 177 a total of 85 thin-sections for the Cismon Core (which integrates the data from Erba and 178 Tremolada, 2004), 242 for DSDP Site 463, and 179 for Piobbico core were investigated. 179 Absolute abundances were obtained by counting all nannofossil specimens in 1 mm² of the 180 thin-section.

181 **2.3 Statistical analysis** \bigcirc

182 The software "Statsoft Statistica 6" was used for multivariate "factor analysis" (FA) (R-183 mode) varimax rotation with principal component extraction to determine the relationships 184 between samples and variables, and to identify palaeoceanographic and palaeoecological 185 affinities among selected nannofossil taxa. Factor loadings represent relationships among 186 individual taxa within the main factors, whereas factor scores denote the relationships within the sampled cases (lithological samples). The same software was used for the "principal 187 components and classification analysis" (PCCA) and δ^{18} O values. The species used in FA and 188 189 PCCA statistical analyses (Watznaueria barnesiae, Biscutum constans, Discorhabdus rotatorius, Zeugrhabdotus erec Rhagodiscus asper, Zeugrhabdotus diplogrammus, 190 191 Staurolithites stradneri, Repagulum parvidentatum, Eprolithus floralis, Nannoconus sp. and 192 Cretarhabdus surirellus) have been selected on the basis of their palaeoecological 193 significance (e.g. Roth and Krumbach, 1986; Erba, 1992b; Herrle et al., 2003; Mutterlose et 194 al., 2005; Tiraboschi et al., 2009).

195 **2.4 Oxygen-isotope analysis**

196 New oxygen stable-isotope analyses were performed at Oxford University on bulk carbonate 197 fraction of 57 samples from DSDP Site 463 and of 373 samples from Piobbico. Bulk-rock samples for isotopic analysis were first powdered, cleaned with 10% H₂O₂ followed by 198 199 acetone, and then dried at 60°C. Powders were then reacted with purified orthophosphoric acid at 90°C and analyzed online using a VG Isocarb device and Prism Mass Spectrometer. 200 Long-term reproducibility, as determined from repeat measurements of the in-house standard 201 (Carrara marble), resulted in analytical uncertainties of $\delta^{18}O = -1.86 \pm 0.1$. The values are 202 203 reported in the conventional delta notation with respect to the Vienna Pee Dee Belemnite (V-PDB) standard. For DSDP Site 463 data are drawn from Price (2003), Ando et al. (2008) and 204 this work, and for the Cismon core δ^{18} O come from Méhay et al. (2009) and Erba et al. 205 206 (2010).

207 2.5 TEX₈₆

Sediments from the Cismon core were extracted as described by van Breugel et al. (2007). The polar fractions of the extracts, containing the GDGTs, were dried under a stream of nitrogen (N_2 ,) redissolved by sonication (5 min) in 200 µl hexane/propanol (99:1; vol:vol), 211 and filtered through 0.45 µm polytetrafluoroethylene (PTFE) filters. GDGTs were analyzed 212 by high-pressure liquid chromatography-mass spectrometry (HPLC/MS) following the 213 method described by Schouten et al. (2007). Samples were analyzed on an Agilent 1100 series 214 LC/MSD SL. A Prevail Cyano column (150 mm × 2.1 mm, 3 mm) was used with 215 hexane:propanol (99:1; vol:vol) as an eluent. After the first 5 min, the eluent increased by a linear gradient up to 1.8% isopropanol (vol) over the next 45 min at a flow rate of 0.2 mL 216 min⁻¹. Identification and quantification of the GDGTs isomers was achieved by integrating the 217 218 peak areas of relevant peaks in m/z 1300, 1298, 1296, 1292, 1050, 1036 and 1022 selected ion 219 monitoring scans. The TEX₈₆ ratio was calculated following Schouten et al. (2002):

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 $TEX_{86} = ([GDGT 2] + [GDGT 3] + [crenarchaeol regioisomer]) / ([GDGT 1] + [GDGT 2] + [GDGT 3] + [crenarchaeol regioisomer])$ (1)

223

where numbers correspond to isoprenoid GDGTs from marine Thaumarchaeota with 1, 2 or 3 cyclopentane moieties, and the crenarchaeol regioisomer has the antiparallel configuration of crenarchaeol (Sinninghe Damsté et al., 2002).

The TEX₈₆ values were converted to SST using the most recent core-top calibration as proposed by Kim et al. (2010) for oceans with SST > 15° C:

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230
$$SST = 38.6 + 68.4 \times \log (TEX_{86})$$
 (2)

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The Branched and Isoprenoid Tetraether (BIT) index is based on the relative abundance of non-isoprenoidal GDGTs derived from soil bacteria versus a structurally related isoprenoid GDGT, 'crenarchaeol' with four cyclopentane moieties and one cyclohexane moiety, produced by marine Thaumarchaeota. The BIT index, which thus represents a measure for soil versus marine organic matter input in marine sediments, was calculated according to Hopmans et al. (2004):

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BIT = ([GDGT-I] + [GDGT-II] + [GDGT-III]) / ([Crenarchaeol] + [GDGT-I] + [GDGT-II] +
 [GDGT-III])

242 **3** Stratigraphic framework

In this work, the stratigraphic framework for the three cores investigated is based on carbonisotope stratigraphy calibrated with calcareous nannofossil and foraminiferal biostratigraphy.
For the Cismon core and DSDP Site 463, magnetic chron CM0 has been used to define the
base of the Aptian; this level was not reached with the Piobbico core.

In addition to the two well-known, high-amplitude δ^{13} C Aptian excursions, several minor 247 fluctuations are identified in the carbon-isotope record from the Tethys, Pacific and Atlantic 248 249 Oceans, which allow codification of major and minor perturbations. Menegatti et al. (1998) 250 focused on the late Barremian-early Aptian interval and identified segments C1-C8. 251 Subsequently, Bralower et al. (1999) extended the codification of Menegatti et al. (1998) through the rest of the Aptian Stage (C1-C11). Herrle et al. (2004) introduced new codes for 252 the Aptian starting from Ap6, coinciding with C5 and C6 of Menegatti et al. (1998), to Al2. 253 254 McAnena et al. (2013) used the Ap9-Al3 segments previously identified by Herrle et al. 255 (2004).

256 The studied sections cover the latest Barremian to earliest Albian time interval. We revised 257 the Herrle et al. (2004) carbon-isotope segments by extending their codes down to Ap1 and 258 renaming the earliest Albian fluctuations Al1 to Al3. Segments Ap1-Ap7 coincide with previously identified segments C1-C7 (Menegatti et al., 1998). The δ^{13} C curve for the rest of 259 260 the Aptian shows several fluctuations, allowing a higher resolution subdivision into segments Ap8–Ap18. In this paper, we applied a double coding for Ap1/C1 through Ap7/C7 segments 261 262 and used the new Ap8-Ap18 and Al1-Al3 codes for the late Aptian-earliest Albian time interval. At Cismon, we identify segments Ap1–Ap8, at Piobbico Ap2–Al3, and at DSDP Site 263 264 463 Ap1-Ap15. Segments Ap8-Ap15 are less well defined at DSDP Site 463 due to 265 incomplete core recovery.

In addition to nannofossil zones NC6-NC8 (Bralower et al., 1995), we used the "nannoconid decline" and the "nannoconid crisis" (e.g. Erba et al., 2010) as further bio-horizons. Moreover, in the late Aptian the "*Nannoconus truittii* acme" (Mutterlose, 1989; Erba, 1994; Herrle and Mutterlose, 2003), defines a globally recognized interval where *N. truittii* dominates the assemblages with undances from 5 up to 40 % of the total nannofloras. As far as lithostratigraphy is concerned, the Selli Level or its equivalents are identified at all three sites. A lithological revision of the Piobbico core also allowed the identification of the Kilian Level Equivalent, corresponding to the prominent black shale at the bottom of lithological Unit 12 within core 44 (Erba, 1988). The Kilian level Equivalent in the Piobbico core is characterized by very fine laminations, without bioturbation, and has a thickness of 33 cm (from 45.13 to 44.80 m). Following Petrizzo et al. (2012), the Kilian Level marks the Aptian/Albian boundary.

278 Regarding the Piobbico core, we identify the presence of a hiatus that eliminates part of the 279 basal Selli Level. In particular, on the basis of the correlation between the lithology and 280 carbon-isotope record from the Piobbico core with the equivalent records from the Cismon core and DSDP Site 463 (Figure 2), as well as from other sedimentary basins, we note that: 1) 281 282 the δ^{13} C values from 75.94 to 74.80 m, ranging between 2 and 3 ‰, probably correspond to segments Ap4/C4 and Ap5/C5 rather than to the negative excursion Ap3/C3 where values of 283 Cismon and DSDP Site 463 sediments are below 1‰; 2) δ^{18} O values from 75.94 to 74.80 m 284 285 fall between -3 and -1‰ and never reach the highly negative values (-4‰) characteristic of those in the Ap3/C3 segment of the other two sites, but rather conform to the range of values 286 287 detected in segments Ap4/C4 and Ap5/C5; 3) the Selli Level in the Cismon core is characterized by three lithological sub-units (Erba et al., 1999), the lowermost being 288 represented by laminated black shales corresponding to segment Ap3/C3, the second 289 290 characterized by prevailing light grey marlstones corresponding to segments Ap4/C4 and 291 Ap5/C5, and the uppermost one characterized by laminated black shales corresponding to 292 segment Ap6/C6. The total organic carbon content (TOC) in the Cismon core shows highest 293 values corresponding to segments Ap4/C, the base of segment Ap5/C5, as well as segment 294 Ap6/C6. Similar high values are detected at DSDP Site 463 in coeval stratigraphic positions. 295 At Piobbico, only two lithological sub-units are recognized (Erba, 1988) following the 296 definition of Coccioni et al. (1987, 1989). The lower part, namely the "green interval", is 297 dominated by light green claystones, while the upper "black interval" is characterized by 298 laminated black shales. It is therefore possible that the lowermost black shale interval 299 normally found in the Selli Level equivalents is missing at Piobbico and only the other two 300 lithostratigraphic intervals, corresponding to Ap4/C4-Ap5/C5 and Ap6/C6, respectively, are 301 represented.

302

303 4 Results

304 **4.1 Calcareous nannofossil abundances** $\mathcal{O}\mathcal{O}$

305 Figures 3-5 illustrate the distribution of the high-fertility (D. rotatorius, B. constans, Z. 306 erectus) and low-fertility (W. barnesiae) nannofossil taxa (following Roth and Krumbach, 1986; Premoli Silva et al., 1989a, 1989b; Watkins, 1989; Coccioni et al., 1992; Erba et al., 307 308 1992b; Williams and Bralower, 1995; Bellanca et al., 1996; Herrle, 2002, 2003; Herrle et al., 2003; Bornemann et al., 2005; Mutterlose et al., 2005; Tremolada et al., 2006; Tiraboschi et 309 al., 2009), as well as of the warm-temperature (R. asper, Z. diplogrammus) and cool-310 311 temperature (S. stradneri, E. floralis, R. parvidentatum) taxa (following Roth and Krumbach, 1986; Wise, 1988; Erba, 1992b; Erba et al., 1992; Mutterlose, 1992; Herrle and Mutterlose, 312

313 2003; Herrle et al., 2003; Tiraboschi et al., 200

314 A description of the major trends of these taxa is given for the three sections investigated:

315 In the **Cismon core** (Fig. 3), *W. barnesiae* is the dominant species with mean abundance of 316 66.8%. *Rhagodiscus asper* ranges from 0 to 20% of the total assemblage (mean: 3%) showing 317 the highest peaks in the lowermost part of the Selli Level (segments Ap3/C3 and Ap4/C4 of 318 the carbon-isotope curve). Zeugrhabdotus diplogrammus ranges from 0 to 2% (mean: 0.1%). Staurolithites stradneri ranges from 0 to 4% (mean: 0.2%), and shows peaks in the uppermost 319 320 part of the Selli Level (segment Ap6/C6). Eprolithus floralis ranges from 0 to 6% (mean: 321 0.2%) and shows peaks just above the top of the Selli Level (Ap7/C7). Biscutum constans 322 ranges from 0 to 2% (mean: 0.2%), D. rotatorius from 0 to 3.5% (mean: 0.4%), and Z. erectus 323 from 0 to 4.1% (mean: 0.15%). These three species are more abundant in the lower part of the 324 Selli Level (segments Ap3/C3, Ap4/C4 and part of Ap5/C5). Nannoconids show a decline in abundance starting prior to magnetic chron CM0 (where they show high abundances up to 325 40% in smear slides; $1*10^4$ specimens/mm² in thin-section) and reaching a minimum 326 327 corresponding with segment Ap3/C3 of the carbon-isotope curve where they are virtually 328 absent.

In the **Piobbico core** (Fig. 4), the interval from 75.29 to 73.92 m, within the Selli Level, is barren of calcareous nannofossils. In the rest of studied interval, *Watznaueria barnesiae* is the dominant species with a mean abundance of 62%. *Rhagodiscus asper* ranges from 0 to 7.7% (mean: 2.4%). *Zeugrhabdotus diplogrammus* fluctuates between 0 and 1.2% (mean: 1%).

333 Eprolithus floralis ranges from 0 to 3.5% (mean: 0.5%), R. parvidentatum from 0 to 0.3%

334 (mean: 0.04%), and S. stradneri from 0 to 3% (mean: 0.6%); these taxa are more abundant 335 above the Selli Level, showing the highest values in corresponding with segments Ap11, Ap13-Ap15 of the carbon-isotope curve. *Biscutum constans* ranges from 0 to 4.1% (mean: 336 337 0.1%), D. rotatorius from 0 to 20% (mean: 2.7%), and Z. erectus from 0 to 6.5% (mean: 338 0.9%). The latter three species are more abundant corresponding with segments Ap9-Ap11 of the carbon-isotope curve. Nannoconids are absent to rare throughout most of the studied 339 340 interval except for an interval between 60.37 and 55.61 m, where they show rather high abundances (up to 40 % relative abundance in smear slides; $4*10^3$ specimens/mm² absolute 341 abundance in thin-section). This particular interval is dominated by N. truittii and coincides 342 343 with the "N. truittii acme".

344 At DSDP Site 463 (Fig. 5), the intervals from 624.24 to 623.96 mbsf and from 623.16 to 622.57 mbsf, within the Selli Level, are barren of calcareous nannofossils. Watznaueria 345 346 barnesiae is the dominant species with a mean abundance of 58%. Rhagodiscus asper ranges 347 from 0 to 32% (mean: 6.5%), having the highest values in the lower part of the section below 348 the Selli Level Equivalent. Zeugrhabdotus diplogrammus ranges from 0 to 2.7% (mean: 1%). 349 Eprolithus floralis ranges from 0 to 8.4% (mean: 0.9%) and S. stradneri from 0 to 11.15% 350 (mean: 2.0%); both taxa are particularly abundant in levels corresponding to segments Ap12 351 and part of Ap13. *Biscutum constans* ranges from 0 to 6.7% (mean: 0.6%), *D. rotatorius* from 352 0 to 30% (mean: 2.4%), and Z. erectus from 0 to 16% (mean: 1.2%). These three species are 353 more abundant within the lower part of the Selli Level Equivalent (segments Ap3-Ap4 of the carbon-isotope curve). A peak is also detected around segment Ap8. Nannoconids are 354 355 abundant below the Selli Level Equivalent and between 568.27 and 540.73 mbsf (segments 356 Ap12 and part of Ap13) showing abundances up to 40 % (relative abundance in smear slides) and $4*10^3$ specimens/mm² (absolute abundance in thin-section), dominated by *Nannoconus* 357 358 truittii, which marks the Nannoconus truittii acme interval.

359

For each studied site, two significant factors were extracted from the FA (R-mode) varimax
rotation analysis:

In the **Cismon core** (Fig. 6A, Tab. 1 of Supplementary material), Factor 1 (F1) and Factor 2 (F2) represent 32% of the total variance. F1 (19% of the total variance) shows the highest positive loadings for *W. barnesiae* and the highest negative loadings for *D. rotatorius, B. constans, Z. erectus.* F2 (13% of the total variance) shows positive loadings for *W. barnesiae*, S. stradneri, E. floralis, and negative loadings for R. asper, C. surirellus and Z. *diplogrammus*. F1 is interpreted to correspond with surface-water fertility and F2 to surfacewater temperature, respectively.

In the **Piobbico core** (Fig. 6B, Tab. 2 of Supplementary material). F1 and F2 represent 36% of the total variance. F1 (25% of the total variance) shows high positive loadings for *Z. erectus, D. rotatorius, B. constans, R. irregularis, C. surirellus, R. asper* and high negative loadings for *Nannoconus* sp. and *W. barnesiae*. F2 (11% of the total variance) shows the highest positive loadings for *W. barnesiae, S. stradneri, E. floralis* and the highest negative loadings for *Nannoconus* sp. F1 is interpreted to correspond with surface-water fertility and F2 with surface-water temperature, respectively.

At **DSDP Site 463** (Fig. 6C, Tab. 3 of Supplementary material), F1 and F2 represent 36% of the total variance. F1 (17% of the total variance) shows the highest positive loadings for *D. rotatorius, Z. diplogrammus, B. constans, R. irregularis* and the highest negative loadings for *Nannoconus* sp. F2 (19% of the total variance) shows the highest positive loadings for *Nannoconus* sp., *E. floralis, S. stradneri* and the highest negative loadings for *W. barnesiae, R. asper, B. constans.* F1 is interpreted to correspond with surface-water fertility and F2 with surface-water temperature, respectively.

Nannoconids show apparently a different affinity at Piobbico compared to the Cismon and DSDP Site 463 records, being associated with taxa indicator of warm waters and high nutrients, instead of the cold-water species *S. stradneri* and *E. floralis*. However, this discrepancy can be explained with the record of Piobbico starting around the "nannoconid crisis", thus excluding the latest Barremian–earliest Aptian interval dominated by nannoconids. It is well possible that the results of the FA are in this case not reliable or should

- 389 be considered with caution.
- 390

391 The results of the PCCA analysis are summarized as follow:

In the **Cismon core** (Fig. 6D, Tab. 4 of Supplementary material), the first component (22% of the total variance) shows the highest positive loadings for *D. rotatorius, B. constans, Z. erectus* and the highest negative loadings for *W. barnesiae* and *Nannoconus* sp. The second component (15% of the total variance) shows the highest positive loadings for *S. stradneri, W. barnesiae* and the δ^{18} O (associated variable), and the highest negative loadings for *R. asper* and *C. surirellus*. The 1 axis is interpreted to correspond with surface-water fertility, the 2
axis to surface-water temperature.

399 In the **Piobbico core** (Fig. 6E, Tab. 5 of Supplementary material), the first component (28% 400 of the total variance) shows the highest positive loadings for D. rotatorius, B. constans, Z. 401 erectus and the highest negative loadings for Nannoconus sp. The second component (11% of 402 the total variance) shows the highest positive loadings for E. floralis, S. stradneri, W. *barnesiae* and the highest negative loadings for *D. rotatorius*. The associated variable δ^{18} O 403 404 has loadings close to zero. The 1 axis is interpreted to correspond with surface-water fertility. 405 while the interpretation for the 2 axis is not straightforward, but might correspond with 406 surface-water temperature.

407 The dataset collected in this work for Piobbico core has been integrated with the dataset from 408 Tiraboschi et al. (2009) covering the Albian. The results of the PCCA analysis performed on 409 the integrated dataset are presented in Figure 6F. The first component (32% of the total 410 variance) shows the highest positive loadings for *D. rotatorius*, *B. constans*, *Z.diplogrammus*, 411 R. asper, C. surirellus, R. irregularis, Z. erectus and the highest negative loadings for 412 W.barnesiae and Nannoconus sp.. The second component (13% of the total variance) shows the highest positive loadings for *E. floralis*, *S. stradneri* and *R. parvidentatum* and the highest 413 negative loadings for *R. asper*, *Z. diplogrammus and B. constans*. Also the associated variable 414 δ^{18} O exhibits positive loadings. The 1 axis is interpreted to correspond with surface-water 415 416 fertility, while the 2 axis corresponds with surface-water temperature.

417

At **DSDP Site 463** (Fig. 6G, Tab. 6 of Supplementary material), the first component (24% of the total variance) shows the highest negative loadings for *R. asper, Z. erectus*, and lower negative loadings for *S. stradneri*, *E. floralis*. The second component (16% of the total variance) shows the highest positive loadings for *D. rotatorius*, *R. irregularis*, *B. constans*, *Z. erectus*, and negative loadings for *W. barnesiae*, and *Nannoconus* sp., δ^{18} O (associated variable) has loadings close to zero. The 1 axis is interpreted to correspond with surface-water temperature, the 2 axis with surface-water fertility.

425 **4.2 Nannofossil Temperature and Nutrient Indices**

426 On the basis of reconstructed nannofossil affinities to temperature and nutrient content of 427 surface waters, some authors (e.g. Herrle et al., 2003; Tiraboschi et al., 2009) have proposed 428 two indices: the Temperature Index (TI) and the Nutrient Index (NI). According to these 429 palaeoecological reconstructions, and the results of our FA and PCCA analysis, we modified 430 the formulae of Herrle et al. (2003) by excluding taxa that are sparse and rare in the studied 431 sections. The formulae of the indices used here are (4) and (5):

433
$$TI = (Ss + Ef + Rp) / (Ss + Ef + Rp + Ra + Zd) \times 100$$
 (4)

434

435
$$NI = (Bc + Dr + Ze) / (Bc + Dr + Ze + Wb) \times 100$$
 (5)

436

437 Where: Ss = S.stradneri; Ef = E.floralis; Rp = R.parvidentatum; Ra = R.asper; Zd =

438 Z.diplogrammus; Bc = B.constans; Dr = D.rotatorius; Ze = Z.erectus; Wb = W.barnesiae.

439

The nannofossil TI, calibrated against carbon-isotope stratigraphy, has revealed systematic and synchronous changes in the Cismon core, Piobbico core and at DSDP Site 463. A complete nannofossil record through OAE 1a is available only for the Cismon core, because the Selli Level of the Piobbico core is incomplete and many samples are barren of nannofossils, while at DSDP Site 463 the top of the Selli Level Equivalent is probably not recovered and some samples are barren. In the three investigated sites, the TI and NI show the following fluctuations:

447

448 At **Cismon** (Fig. 3) the TI shows high-frequency fluctuations superimposed on a longer term \bigcirc 449 trend. The warmest temperatures were reached in the early phase of OAE 1a (corresponding 450 to segment Ap3/C3 of the carbon-isotope curve). Cooling interludes are registered within the 451 Selli Level, especially across segments Ap4/C4 to Ap5/C5. The interval represented by the 452 uppermost part of the Selli Level (segment Ap6/C6), suggests that a pronounced cooling 453 episode was followed by another cold snap after deposition of sediments just above the Selli Level. In the overlying interval (Ap7/C7), the TI shows relatively high-amplitude 454 455 fluctuations. The NI indicates that the highest surface-water fertility was recorded in the lower 456 part of the Selli Level (segments Ap3/C3 to base of Ap5/C5). The rest of the Selli Level 457 shows low NI. Fertility started to increase in the Ap7/C7 interval.

458 At **Piobbico** (Fig. 4), the TI shows the warmest temperatures in the lowermost part of the 459 recovered Selli Level corresponding to base of Ap5/C5. All samples in the Ap5/C5-Ap6/C6 460 interval are barren of calcareous nannofossils and therefore the TI cannot be used for relative 461 palaeotemperature fluctuations. Corresponding to segments Ap7/C7–Ap9, a general cooling 462 is detected (Ap8), interrupted by a brief warming. Then, from Ap9, a warming continued through most of Ap11. The rest of the late Aptian was characterized by a prolonged cooling 463 464 episode (from top of Ap11 to top of Ap15) followed, at the end of the Aptian, by a warming 465 trend showing two temperature peaks coinciding with the 113 Level and the Kilian Level at 466 the Aptian/Albian boundary. The earliest Albian (Al1-Al3) shows a brief relative cooling 467 immediately after the Kilian temperature spike, followed by a general warming.

468 The NI exhibits relatively high values in the interval immediately preceding the Selli Level 469 and in its lowermost portions, corresponding to the base of Ap5/C5. All samples in the 470 Ap5/C5–Ap6/C6 interval are barren of calcareous nannofossils and therefore the NI cannot be 471 used for illustrating palaeofertility fluctuations. Above the Selli Level, a long interval of 472 increased fertility (Ap7-Ap11) shows maximum values in the Ap9-Ap10 interval. The 473 Nannoconus truittii acme is characterized by low surface-water fertility, followed by a 474 relative increase of the NI up to Ap15. The Aptian/Albian boundary interval is marked by a 475 decrease of the NI interrupted by a relative increase through the Kilian Level. The lowermost 476 Albian (Al2-Al3) exhibits a trend to increased fertility extending through the Albian 477 (Tiraboschi et al., 2009).

478 At DSDP Site 463 (Fig. 5), the TI indicates warm temperatures just before and at the onset of 479 OAE 1a. The warmest temperatures are reached at the level of segment Ap3/C3. Relative 480 cooling interludes are registered within the Selli Level Equivalent, in segments Ap4/C4 and 481 Ap5/C5. During the late Aptian, a long cooling (Ap7-Ap14) is registered with the coolest 482 temperatures recorded from the top of Ap12 to the base of Ap13. The NI indicates relatively 483 high values in the interval preceding the Selli Level Equivalent. Two maxima are recorded in the Ap3/C3 and at the base of Ap5/C5, respectively. Low NI is detected in the rest of the Selli 484 485 Level Equivalent. An increase of the NI starts in Ap7 and continues up to the base of Ap12, 486 with a maximum corresponding to Ap8. A decrease in then recorded during the Nannoconus 487 truittii acme interval, followed by a relative increase.

488 **4.3 Oxygen isotope fluctuations**

The three oxygen-isotope records are somewhat scattered, and probably reflect a contribution from diagenetic cement. However, δ^{18} O trends are reproduced at the three studied sites independently of lithology, and nannofossil preservation is persistently moderate; we conclude, therefore, that the oxygen-isotope records contain a primary palaeotemperature signal only marginally modified by lithification. The main trends (Figs. 3-5) are summarized as follows:

495 Along segments Ap1/C1 and Ap2/C2 the isotopic ratios are relatively stable, being ~ -2‰ at 496 DSDP Site 463, -1.5‰ at Piobbico and -1‰ at Cismon. At the end of segment Ap2/C2 values 497 start to decrease, reaching -4‰ in correspondence with the negative carbon-isotope excursion (segment Ap3/C3). At Cismon, the decreasing trend is interrupted by a short-lived (\sim 35 ky) 498 interval of higher values (-1.5%). At segment Ap4/C4, the δ^{18} O values start increasing and in 499 500 the middle part of the Selli Level (segment Ap5/C5) they fluctuate: between -1 and -2‰ at 501 Cismon, between -1 and -3 ‰ at Piobbico, and between -1 and -4‰ at DSDP Site 463. Corresponding with segment Ap6/C6, δ^{18} O values are relatively stable between -1 and -2 %. 502 Starting from segment Ap7/C7, oxygen isotopes illustrate progressively increasing ratios 503 504 reaching ~-1‰ around the Nannoconus truittii acme interval and then decrease to a minimum of ~-3‰ close to the Aptian/Albian boundary. With respect to coeval sediments in the Tethys 505 506 and Pacific Ocean, we notice that the oxygen-isotope values of the Cismon are greater by 1‰.

507 4.4 TEX₈₆

508 A total of 32 samples from the Cismon core have been analyzed for TEX₈₆ of which 17 509 contained detectable amounts of GDGTs (Tab. 7). TEX₈₆ data for a number of sediments were 510 excluded as they contained relatively mature organic matter, i.e. the hopane 22S/(22S+22R) 511 ratio was >0.2 (van Breugel et al., 2007) at which level TEX₈₆ values will become biased 512 towards lower temperatures (Schouten et al., 2004). Nearly all sediments have BIT values < 513 0.3, suggesting relatively low input of soil-derived GDGTs, and thus no bias of the TEX_{86} 514 (Weijers et al., 2006). The values obtained for the OAE 1a interval (Figs. 2, 8) comprise one sample having a TEX₈₆ value of 0.57 indicative of ~22°C sea-surface temperature (SST) and 515 corresponding to the most negative δ^{13} C values (segment Ap3/C3). The rest of segment 516 517 Ap3/C3 is characterized by values from 0.67 to 0.61 (SST= \sim 24 to 27°C). Segments Ap4/C4 518 and Ap5/C5 are characterized by relatively stable values lying between 0.66 and 0.64 (SST = 519 ~25–26°C). Corresponding to segment Ap6/C6, one sample gives a TEX₈₆ value of 0.58 (SST 520 = ~22.5°C) and the following one of 0.64 (SST = ~25.5°C).

521

522 **5 Discussion**

523 **5.1** Long- and short-term temperature fluctuations during the Aptian

Long-term temperature variations are comparable with the results of previous studies based on various temperature proxies (i.e. calcareous nannofossils, palynomorph, oxygen isotopes, and TEX₈₆). The high-resolution sampling and the stratigraphic calibration of the studied sections, enabled better constraints on long-term temperature changes and detected short-term variability improving the characterization of climate changes during the Aptian.

529 Long-term temperature fluctuations. A warming pulse (Fig. 7), starting at the time of the 530 "nannoconid crisis", characterized the onset of OAE 1a. The highest temperatures are recorded in the core of the negative carbon-isotope interval (segment Ap3/C3), as also 531 532 documented in other sections in the Tethys (e.g. Menegatti et al., 1998; Hochuli et al., 1999; 533 Luciani et al., 2001; Bellanca et al., 2002; Jenkyns, 2003; Millán et al., 2009; Erba et al., 534 2010; Jenkyns, 2010; Keller et al., 2011; Stein et al., 2011; Bottini et al., 2012; Hu et al., 535 2012; Husinec et al., 2012), Vocontian Basin (e.g. Moullade et al., 1998; Kuhnt et al., 2011), 536 Boreal Realm (Mutterlose et al., 2010; Bottini and Mutterlose, 2012; Pauly et al., 2013; 537 Mutterlose and Bottini, 2013), eastern European Russian Platform (Zakharov et al., 2013), 538 and Pacific Ocean (e.g. Jenkyns, 1995; Price, 2003; Schouten et al., 2003; Ando et al., 2008; 539 Bottini et al., 2012). Warm conditions persisted through OAE 1a, although fluctuations are 540 detected, as discussed below. A major cooling, coeval with segment Ap7/C7, marks the end of global anoxia; it is followed by a warm phase preceding a major long-lasting cooling 541 542 episode starting during segment Ap8 and extending through most of the late Aptian. 543 Minimum temperatures were reached soon after the N. truittii acme, confirming the cooling 544 (of ~4°C down to ~28°C) indicated by TEX₈₆ reconstructed from the Proto-North Atlantic 545 (McAnena et al., 2013). Further evidence of significant cooling during the late Aptian derives 546 from the occurrence of the Boreal (cold water) species R. parvidentatum at low latitudes as documented here for the Piobbico core and DSDP Site 463 (Figs. 4, 5, 7), and in the 547 548 Vocontian Basin, North Sea and Proto-North Atlantic Ocean (Herrle and Mutterlose 2003, 549 Rückheim et al., 2006; Herrle et al., 2010; McAnena et al., 2013). Close to the Aptian/Albian

- boundary, temperatures show a relative increase, with warm peaks at the 113 level and Kilianequivalent.
- **Climate variability during OAE 1a** De integration of nannofossil TI and oxygen-isotope 552 553 data allows the identification of a sequence of synchronous temperature variations labelled A 554 to M (Fig. 8) at the three studied sites. After a warming pulse at the onset of OAE 1a (Interval A), a brief (~35 ky) cooling interlude interrupted warm conditions corresponding to the 555 interval of minimum δ^{13} C values (Interval B). It was followed by a maximum warming in the 556 557 core of segment Ap3/C3 (Interval C). A cooling episode (Interval D) coincides with segment 558 Ap4/C4 and base Ap5/C5. Intermediate climatic conditions, including one minor cooling episode (Interval E), a warm interlude (Interval F) and another minor cooling (Interval G), 559 characterize segment Ap5/C5. Warmer temperatures (Interval H) preceded a more prominent 560 cooling (Interval I) correlating with the latest part of OAE 1a and corresponding with segment 561 Ap6/C6.The end of anoxia was marked by a short-lived warming (Interval L) and a further 562 563 major cooling (Interval M) coinciding with the onset of segment Ap7/C7. A cool snap across 564 segment Ap4/C4 interrupting the main warming has also been detected in the Vocontian Basin (Kuhnt et al., 2011; Lorenzen et al., 2013), Tethys (Menegatti et al., 1998; Luciani et 565 566 al., 2001; Stein et al., 2011), and Turkey (Hu et al., 2012).
- 567 The correlation of oxygen-isotope and calcareous nannofossil datasets with SST estimates from TEX₈₆ is difficult since the TEX₈₆ data available for OAE 1a have a much lower 568 569 resolution and provide relatively scattered records. The new TEX₈₆ data for the Cismon core 570 are suggestive of SSTs ranging between 22°C and 27°C. The lowermost data point, which 571 corresponds to Interval B, indicates an SST of ~22°C which is the coolest value for the 572 studied interval and well matches with cooler conditions reconstructed from other data. The 573 SST values for the following three data points are rather puzzling: two indicate temperatures 574 of ~23-25 °C and fall in Interval C - the warmest of OAE 1a - while the third data point 575 shows almost 27°C although it falls in Interval D, interpreted to correspond to a time of relative coolin \mathcal{O} he rest of the samples, encompassing Intervals E to H, and representing 576 577 minor temperature fluctuations, fall between 25°C and 27°C. We identify one more 578 discrepancy in the relatively low estimated SST (22.5 °C) for one sample falling in Interval H, 579 suggested by TI and oxygen isotopes to be a relatively warm interlude.
- 580 Another problem of the TEX₈₆ data of the Cismon core is related to the SSTs, which are \sim 5°C 581 to 8°C lower compared with the TEX₈₆ records from other sites. TEX₈₆ data from DSDP Site

582 463 cover the Ap4/C4 to Ap6/C6 interval and range between 31°C and 34°C (recalibrated 583 from Schouten et al., 2003 using Eq. 2). At Shatsky Rise, temperatures fall between 30°C and 584 35°C (recalibrated values from Dumitrescu et al., 2006 using Eq. 2). Here, two cooling 585 interludes are detected: the first cooling of 4°C down to 30°C was during segment Ap4/C4 586 and corresponds to the cooling of our Interval D. The second cooling of 5°C down to 30°C 587 seems to corresponds to segment Ap6/C6 and possibly reflects the cooling of Interval I. In the 588 Lower Saxony Basin, SSTs indicate a distinctive warming during OAE 1a (segment C3-C6), 589 with TEX₈₆ temperature estimates of 31–34°C. The TEX₈₆ data for the interval following 590 OAE 1a (the C7 segment) reveal stable SSTs around 30°C (Mutterlose et al., 2010).

591 We notice that although the temperature variability ($\Delta = \sim 4-5^{\circ}$ C) is similar in all sites, at Cismon the absolute temperatures are generally 5°C to 8 °C lower than at DSDP Site 463, 592 593 Shatsky Rise and Lower Saxony Basin. For Cismon, also the highest (coolest temperature) δ^{18} O values are ~1% greater than those registered at DSDP Site 463 and ~0.5% greater than 594 595 those at Piobbico. Generally cooler temperatures for Cismon could be explained by different 596 latitudinal settings, the Cismon site being at $\sim 30^{\circ}$ N, the Shatsky Rise at an almost equatorial 597 position and the DSDP Site 463 at $\sim 20^{\circ}$ S. However, this seems not to apply to the Boreal 598 section (39°N) characterized by the highest (~35°C) SST. Another possible explanation for this discrepancy may be that the TEX_{86} values from the Cismon core are already affected. 599 600 the higher level of thermal maturity (i.e. hopane 22S/(22S+22R)) ratios of 0.1–0.2). It has been 601 documented that destruction of GDGTs during thermal maturation processes results in lower 602 TEX₈₆ values due to the fact that GDGTs with cyclopentane moieties are thermally less stable 603 (Schouten et al., 2004). Finally, it has been shown in several modern settings that TEX_{86} , although calibrated against sea-surface temperature, may sometimes reflect changes in 604 605 subsurface water temperatures as well (e.g. Huguet et al., 2007; Lopes dos Santos et al., 606 2010), possibly because the source organisms, Thaumarchaeota, also reside in the deeper 607 thermocline where nutrients such as ammonia might be available.

608 5.2 Long- and short term changes in surface water fertility

609 The nannofossil NI exhibits similarities between the three studied sites (simplified in Fig. 7, 610 where nannofossil data are calibrated against the δ^{13} C curve, adopting the timescale of 611 Malinverno et al., 2012). The earliest Aptian (segments Ap1/C1 and Ap2/C2) is characterized 612 by low to intermediate NI values suggestive of oligotrophic conditions. The onset of OAE 1a was marked by increasing fertility, which reached a maximum in the core interval of the negative carbon-isotope excursion (segment Ap3/C3). A decrease in surface-water fertility characterized the rest of the Selli Level (segments Ap4/C4–Ap6/C6). A shift to meso- to eutrophic conditions is detected from segment Ap8 to the beginning of the *N. truittii* acme interval, corresponding to minimal fertility conditions. The latest Aptian is then characterized by intermediate NI values, continuing into the earliest Albian.

619

The early Aptian has been generally seen as a time of warm and humid climate, mainly responsible for accelerated continental weathering, and consequent important nutrient fluxes to the ocean sustaining high productivity (e.g. Leckie et al., 2002; Erba, 2004; Föllmi 2012). It has also been proposed that higher fertility in the global ocean was triggered directly by submarine igneous events that introduced enormous quantities of biolimiting metals within hydrothermal plumes (e.g. Larson and Erba, 1999; Leckie et al., 2002; Erba, 2004).

626 Peaks in the NI are detected at the levels of the "nannoconid decline" (~1 Ma before OAE 1a) 627 and the "nannoconid crisis". This relationship is in agreement with the interpretation of 628 nannoconids as oligotrophic taxa, which suffered during episodes of increased surface-water 629 fertility. The results of the FA and PCCA analysis also support this affinity for nannoconids. Their virtual absence during the early phase of OAE 1a has been interpreted as the result of 630 631 widespread meso- to eutrophic conditions (e.g. Coccioni et al., 1992; Bralower et al., 1994; Erba, 1994, 2004; Premoli Silva et al., 1999) combined with excess CO₂ in the ocean-632 633 atmosphere system (Erba and Tremolada, 2004; Erba et al., 2010).

As far as the OAE 1a interval is concerned, the fluctuations in surface-water fertility reconstructed in our work are in agreement with other studies on calcareous nannofossils from the Tethys, Boreal Realm and Atlantic Ocean. Furthermore, other proxies, for example palynomorphs (Hochuli et al., 1999) and phosphorus (e.g. Föllmi et al., 2006; Föllmi and Gainon, 2008; Stein et al., 2012), support this interpretation.

639 5.3 Climate and environmental changes and their relation to igneous-tectonic 640 events during the Aptian

641 Our data confirm a relationship between major volcanic episodes and climate change, with
642 associated (or subsequent) perturbations in ocean chemistry, structure and fertility.
643 Specifically, the construction of the OJP LIP, documented in the Os-isotopic record (Tejada et

644 al., 2009; Bottini et al., 2012), Pb isotopes (Kuroda et al., 2011), and biomarkers (Méhay et al. 2009), suggestive of a stepwise accumulation of volcanogenic CO_2 in the atmosphere (Fig. 645 646 correlates in time with OAE 1a and was marked by global warming at the onset of the mid-647 Cretaceous greenhouse (e.g. Larson and Erba, 1999; Jenkyns, 2003). A short-lived event of 648 possible methane hydrate dissociation probably promoted a ~100 kyr-long interval of 649 accelerated continental weathering, temporarily reducing the CO₂ concentrations and inducing 650 a subsequent cooling interlude (~35 ky). The next interval, marked by a maximum warming, 651 coincided with the beginning of the most intense volcanic phase of OJP (Bottini et al., 2012). 652 This correspondence is suggestive for a (super)greenhouse climate triggered by excess 653 volcanogenic CO_2 . The rest of OAE 1a was accompanied by climate variability including 654 cooling interludes. Termination of widespread anoxia-dysoxia coincided with the end of the 655 main emplacement of the OJP (Bottini et al., 2012) and a major cooling.

656 Large-scale igneous-tectonic events took place also during the late Aptian, but their causal 657 impact on climate changes are less obvious, since palaeotemperatures were generally cooler. 658 We notice that submarine volcanism (construction of OJP, Manihiki Plateau, Hikurangi Plateau, the early phase of Kerguelen Plateau) correlates with global warming, but subaerial 659 660 addition to magmatic fluxes of different orders of magnitude (lower for Kerguelen, see 661 662 Eldholm and Coffin, 2000), subaerial volcanism probably injected ashes and gases into the 663 atmosphere inducing short-term cooling associated with individual degassing phases. 664 Feedbacks related to atmospheric CO₂ drawdown via accelerated weathering were probably most significant, as also suggested by Ca isotopes (Blättler et al., 2012). On the basis of 665 666 pedogenic calcretes from South Korea, Hong and Lee (2012) documented a decrease in CO₂ concentrations from ~1000 to ~500 ppmV for an interval in the late Aptian corresponding, as 667 discussed above, to relatively cooler temperatures. However, these data present a large 668 uncertainty in the age assignment. Recently published data by Li et al. (2014 com similar 669 continental facies in south-east China spanning the same age, are suggestive of higher values. 670 671 Specifically, here we present (Fig. 7) a revised plot of Li et al. (2014) data based on the 672 Malinverno et al. (2012) time scale, which indicate progressively increasing CO₂ concentrations from ~1000 up to ~2000 ppmV across the N. truittii acme interval despite a 673 674 general cooling trend. In correspondence of the lowest temperatures reached in the latest Aptian, CO₂ estimates decrease to ~1600 ppmV. The following early Albian warming trend 675 676 was instead paralleled by increasing CO₂ concentrations up to 2600 ppm.

677 The long-lasting cool conditions of the late Aptian have been recently quantified using TEX_{86} data (McAnena et al., 2013), which indicate a total decrease of palaeotemperatures of ~ 4°C 678 (from $32^{\circ}C$ to $28^{\circ}C$) in the equatorial Proto-Atlantic Ocean followed by a warming (~4°C) 679 680 linked to the earliest Albian OAE 1b. The trends of our nannofossil TI curve are similar to these TEX₈₆-reconstructed SST changes (Fig. 7), so we adopt the values of McAnena et al. 681 (2013) to estimate climate variations through the Aptian using the nannofossil TI fluctuations. 682 683 The warming at the onset of OAE 1a corresponds to an increase of 2-3°C and climate 684 variability during OAE 1a is marked by a cooling of $\sim 2^{\circ}$ C. The prominent cooling at the end of global anoxia corresponds to a decrease of $\sim 3^{\circ}$ C, followed by a warming of $\sim 3^{\circ}$ C and the 685 coolest interval in the late Aptian is marked by a further decrease of ~4°C. As far as OAE 1a 686 is concerned, the SST variability estimated from the TI (2–3 °C) differs little from the direct 687 TEX₈₆ estimates of 4–5 °C. 688

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690 Volcanically linked climate change seems closely connected to nutrient recycling and ocean 691 fertilization. Different eruption styles and duration, as well as magma composition and 692 quantity, presumably produced diverse weathering rates and introduction of biolimiting 693 metals. Although calcareous nannoplankton are but one group of primary producers and they thrive under oligotrophic-mesotrophic conditions, the nannofossil NI can be used to trace the 694 trophic levels of surface waters in the past. Figure 7 suggest \mathcal{P} at nutrient availability was 695 strongly coupled with climate change in the early Aptian, but less so in the late Aptian. 696 697 Fertility fluctuations could be due to differential weathering rates. During OAE 1a, 698 greenhouse conditions generated by repetitive volcanogenic CO₂ emissions (e.g. Méhay et al., 699 2009; Erba et al., 2010) might have increased weathering rates, and thereby the supply of nutrients. We see a correspondence between maximum warming and high Prface-water 700 701 fertility. In addition, the largest submarine volcanic pulses at the beginning of OAE 1a and in 702 the mid-late Aptian seem to have introduced biolimiting metals during submarine plateau 703 construction. The nutrients presumably stimulated primary productivity with consequent 704 consumption of oxygen through organic matter and metal oxidation, hence promoting anoxic 705 conditions. The upper part of the Selli Level has high TOC content, indicating that 706 productivity and/or preservation of organic matter was relatively high. The apparent oligotrophic conditions suggested by the NI are explained by biomarker data and nitrogen 707 stable isotopes, indicating N-fixing cyanobacteria as the likely main primary producers during 708

709 OAE 1a (Kuypers et al., 2004; Dumitrescu and Brassell, 2006). Their N-fixation potentially 710 provided N nutrients for the rest of the oceanic biota and presumably was the key-process in 711 the production of organic matter, maintaining higher productivity through OAE 1a. The 712 accumulation and burial of organic matter would have progressively acted as storage for excess CO₂, leading to lower temperatures and, possibly, to the termination of OAE 1a under 713 714 less active (or ceased) OJP volcanism. We notice that the two more intense cooling interludes 715 across OAE 1a correspond to levels with relatively high TOC content (>4%), suggesting that the burial of organic matter may have acted as a reservoir for excess CO₂, thus temporarily 716 717 mitigating greenhouse conditions.

Among Cretaceous calcare nannofloras, nannoconids are interpreted as specific to the 718 719 lower photic zone, associated with a deep nutricline, so that they thrived when surface waters 720 were characterized by oligotrophic conditions (Erba, 1994, 2004). The record of nannoconid 721 distribution compared with the nannofossil NI confirms this hypothesis for the entire Aptian 722 interval: the "nannoconid crisis" correlates with an increase of the NI, while the return of 723 nannoconids following deposition of the Selli Level and the N. truittii acme corresponds to 724 minima in the NI curve. We stress the fact that nannoconid abundance does not unequivocally 725 correlate with climate change, at least in the Aptian, because the "nannoconid crisis" 726 coincides with major warming while the final nannoconid disruption (the end of the N. truittii 727 acme) corresponds to the most severe cooling.

728 These data contradict the interpretation of McAnena et al. (2013) for the nannoconid failure 729 due to cold conditions in the late Aptian and imply a different explanation for abundance 730 changes of these rock-forming nannofossils. We believe that volcanically induced CO_2 731 concentrations played a key role for nannoconid calcification, regardless of climatic 732 conditions (Erba, 2006). Both the OJP and Kerguelen LIPs emitted huge quantities of CO₂ that arguably provoked ocean acidification. We emphasize that the prolonged cooling in the 733 734 late Aptian promoted CO₂ absorption in the ocean and acidification. The nannoconid crises, 735 including their final collapse in the latest Aptian, could thus be viewed as failures in 736 biocalcification. Similarly, the major reduction in size, decrease in abundance, and species 737 turnover documented for planktonic foraminifers (Huber and Leckie, 2011), which is coeval 738 with the final nannoconid decline and a nannofossil turnover, might be the response of 739 calcareous zooplankton to volcanically triggered ocean acidification.

740

741 6 Conclusions

• Quantitative study of calcareous nannofossils integrated with oxygen-isotope and TEX₈₆ records from the Tethys and Pacific Oceans has provided a reconstruction of the climatic evolution through the entire Aptian. The excellent stratigraphic time control on the studied sections coupled with high sampling density, allows confirmation of some of the climatic variations detected in previous work and highlights, during OAE 1a, temperature fluctuations not previously detected

- 748 The results from the Tethys and Pacific Oceans confirm climatic variability through the 749 Aptian, characterized by a warming trend that began prior to and reached a maximum 750 during OAE 1a, coincident in time with the negative carbon-isotope excursion. The rest 751 of OAE 1a was marked by subsequent cold snaps and a further cooling took place when 752 the uppermost part of the Selli Level was being deposited. A cooling marked the end of 753 global anoxia and another long-lasting cooling characterized the middle late Aptian, 754 culminating soon after the N. truittii acme. The latest Aptian was, instead, characterized by a gradual warming accorded by nannofossil assemblages and TEX_{86} data. SSTs from 755 TEX₈₆ are suggestive of 24–27 °C in the Tethys during OAE 1a, which are nevertheless 756 757 5°C to 8 °C lower than estimates from the Pacific Ocean and Boreal Realm, being probably affected by maturity levels or other factors. Although the earliest Aptian was 758 characterized by oligotrophic condition, the onset of OAE 1a was marked by increasing 759 760 fertility, which reached a maximum at a time corresponding to the core of the negative 761 carbon-isotope excursion. A decrease in surface-water fertility is recorded from the vounger part withe Selli Level. A shift to warm and meso- to eutrophic conditions is 762 763 detected after OAE 1a up to the beginning of the N. truittii acme interval, corresponding 764 to minimal fertility conditions. The latest Aptian was then characterized by intermediate 765 fertility, continuing into the earliest Albian.
- 766 Our data indicate that the beginning of the prolonged volcanic phase during OAE 1a coincided with the warmest temperatures and the highest surface-water fertility. 767 768 Weathering and hydrothermal activity were the main drivers of nutrient input, positively 769 affecting meso-to eutrophic taxa but having a negative impact on oligotrophic species 770 such as nannoconids, which were not greatly affected by climatic changes. Rapid 'cold 771 snaps' are detected when OJP volcanism apparently continued, suggestive of feedback 772 mechanisms, drawing down CO₂ and affecting the climate. The end of anoxia was in 773 phase with diminished OJP activity and global cooling. We hence see a direct

- relationship between OJP volcanism and climatic changes in the interval encompassingOAE 1a.
- We suggest that OJP volcanism directly caused general global warming, while the excess burial of organic matter acted as an additional and/or alternative process to weathering, causing CO_2 drawdown and consequent climate change during OAE 1a. Massive subaerial volcanism (Kerguelen Plateau LIP which took place during the late Aptian, was associated with relatively cool conditions, implying the dominant effect of atmospheric CO_2 drawdown via accelerated weathering.
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- 783

784 Appendix A: Taxonomy

- 785 Calcareous nannofossils cited in this work:
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- 787 Biscutum constans (Górka 1957) Black in Black and Barnes, 1959
- 788 Cretarhabdus Bramlette and Martini, 1964
- 789 Cretarhabdus surirellus (Deflandre, 1954) Reinhardt, 1970
- 790 Discorhabdus Noël, 1965
- 791 Discorhabdus rotatorius (Bukry 1969) Thierstein 1973
- 792 Eprolithus Stover, 1966
- 793 Eprolithus floralis (Stradner, 1962) Stover, 1966
- 794 Nannoconus Kamptner, 1931
- 795 Repagulum Forchheimer, 1972
- 796 *Repagulum parvidentatum* (Deflandre and Fert, 1954) Forchheimer, 1972
- 797 Rhagodiscus Reinhardt, 1967
- 798 Rhagodiscus asper (Stradner, 1963) Reinhardt, 1967
- 799 Staurolithites Caratini, 1963
- 800 Staurolithites stradneri (Rood et al., 1971) Bown, 1998

- 801 Watznaueria Reinhardt, 1964
- 802 Watznaueria barnesiae (Black, 1959) Perch-Nielsen, 1968
- 803 Zeugrhabdotus Reinhardt, 1965
- *Zeugrhabdotus diplogrammus* (Deflandre in Deflandre and Fert, 1954) Burnett in Gale etal., 1996
- 806 Zeugrhabdotus erectus (Deflandre in Deflandre and Fert, 1954) Reinhardt, 1965
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809 References

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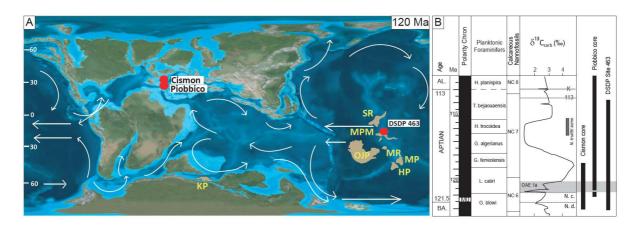
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Figure 1. A) Location map of studied sites at 120 Ma (modified after Erba et al. under final
review by the editor). OJP = Ontong Java Plateau; KP = Kerguelen Plateau; SR = Shatsky
Rise; MPM = Mid-Pacific Mountains; MR = Magellan Rise; MP = Manihiki Plateau; HP =
Hikurangi Plateau. B) Stratigraphic ranges of the studied sections. Latest Barremian to earliest
Albian chronologic framework is from Erba et al. (under final review by the editor).
Numerical ages are based on the timescale of Malinverno et al. (2012). K = Niveau Kilian;
1172 113 = 113 Level; N.c. = Nannoconid crisis; N.d.= Nannoconid decline.

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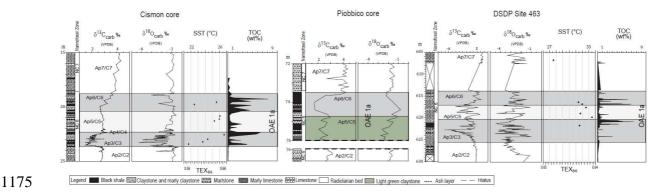
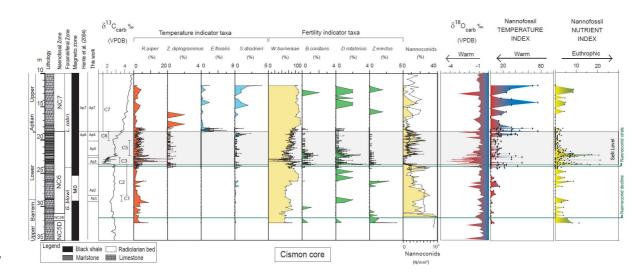


Figure 2. Correlation between the Cismon core, the Piobbico core and DSDP Site 463. δ^{13} C 1176 data after: Erba et al. (1999) and Méhay et al. (2009) for the Cismon core; Erba et al. (under 1177 final review by the editor) for the Piobbico core; Price (2003), Ando et al. (2008) and Bottini 1178 et al. (2012) for DSDP Site 463. Bulk δ^{18} O data after: Erba et al. (2010) for the Cismon core; 1179 Price (2003), Ando et al. (2008) and this work for DSDP Site 463. TOC after: Erba et al. 1180 1181 (1999) and Bottini et al. (2012) for the Cismon core; Ando et al. (2008) for DSDP Site 463. TEX₈₆ after: Schouten et al. (2003) for DSDP Site 463; this work for the Cismon core. For 1182 1183 both cores SST was calculated using the equation of Kim et al. (2010). Grey bands indicate 1184 intervals of higher (darker) and lower (lighter) TOC values.

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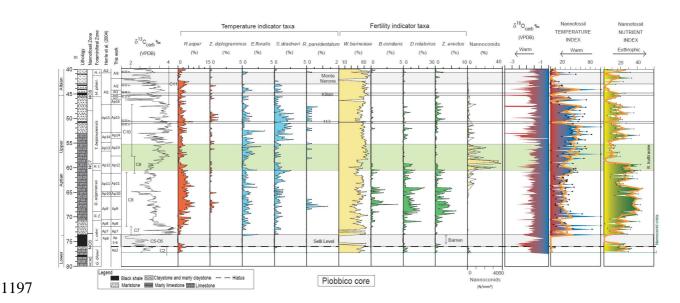


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Figure 3. Cismon core: fluctuations of calcareous nannofossil temperature and fertility indicator taxa. Temperature (TI) and Nutrient (NI) indices based on calcareous nannofossils (low values of the TI indicate high temperatures and vice versa; high values of the NI indicate

1191 high surface-water productivity and vice versa). δ^{13} C is from Erba et al. (1999) and Méhay et 1192 al. (2009). Nannofossil and foraminiferal biostratigraphy is from Erba et al. (1999). 1193 Magnetostratigraphy is from Channell et al. (2000). Bulk δ^{18} O data are from Erba et al. 1194 (2010).

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1198 Figure 4. Piobbico core: fluctuations of calcareous nannofossil temperature and fertility 1199 indicator taxa. Temperature (TI) and Nutrient (NI) indices based on calcareous nannofossils 1200 (low values of the TI indicate high temperatures and vice versa; high values of the NI indicate 1201 high surface-water productivity and vice versa). Orange curve indicates smoothed TI and NI 1202 records based on three-point moving average. δ^{13} C is from Erba et al. (under final review by 1203 the editor). Nannofossil and foraminiferal biostratigraphy is from Erba et al. (1988) and 1204 Tornaghi et al. (1989). Bulk δ^{18} O data are from this work.

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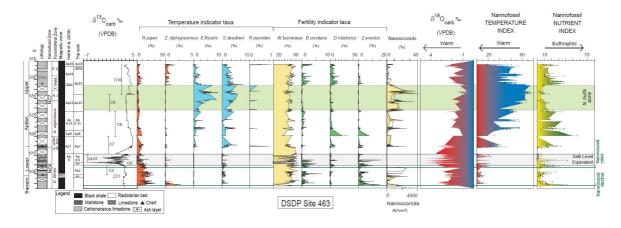
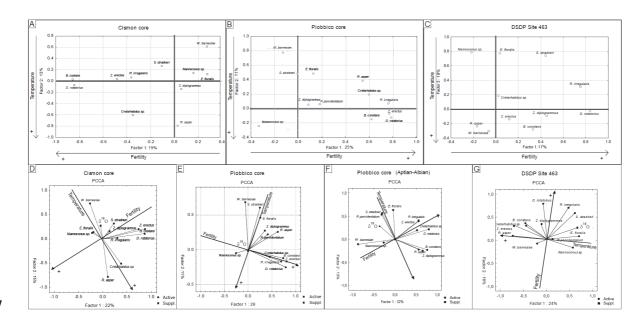


Figure 5. DSDP Site 463 Mid-Pacific Mountains: fluctuations of calcareous nannofossil temperature and fertility indicator taxa. Temperature (TI) and Nutrient (NI) indices based on calcareous nannofossils (low values of the TI indicate high temperatures and vice versa; high values of the NI indicate high surface-water productivity and vice versa). δ^{13} C is from Price (2003), Ando et al. (2008), Bottini et al. (2012). Nannofossil and foraminiferal biostratigraphy is from Erba, (1994) and Ando et al. (2008). Magnetostratigraphy is from Tarduno et al. (1989). Bulk δ^{18} O data are from Price (2003), Ando et al. (2008), and this work.

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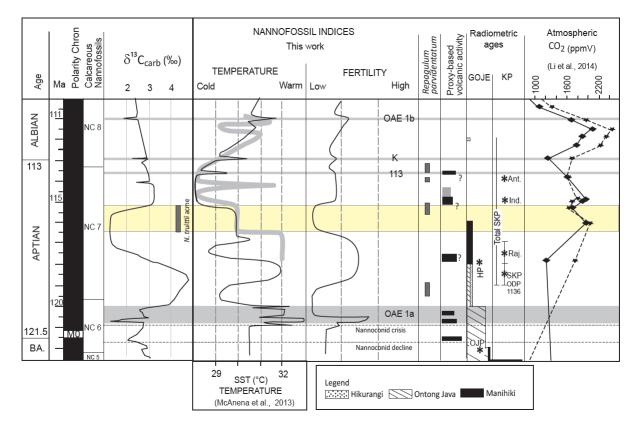
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Figure 6. On the top row, the results of Factor Analysis (R-mode) varimax normalized rotation with principal component extraction are presented for (A) Cismon core, (B) Piobbico core and (C) DSDP Site 463. On the bottom row, the results of the principal component and classification analysis (PCCA) are presented for (D) Cismon core, (E) Piobbico core, (F)

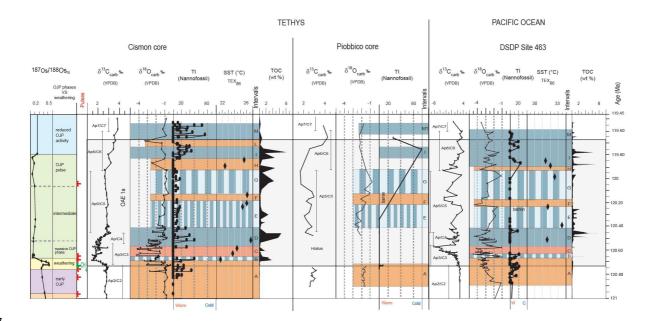
- 1222 Piobbico core, including the Albian dataset from Tiraboschi et al. (2009), (G) DSDP Site 463.
- 1223 The associated variable is the δ^{18} O.
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1227 Figure 7. Nannofossil-based temperature and nutrient variations across the Aptian 1228 reconstructed in this work and across the Albian (from Tiraboschi et al., 2009). The thick-1229 grey curve represents SST from McAnena et al. (2013). Bio-chemo-magneto stratigraphy 1230 after Erba et al. (under final review by the editor). Numerical ages are based on the timescale 1231 of Malinverno et al. (2012). Multiproxy-based volcanic phases and radiometric ages of the 1232 Greater Ontong Java Event (GOJE) and Kerguelen LIPs are from Erba et al. (under final 1233 review by the editor). Atmospheric CO₂: Li et al. (2014). Nannofossil data and isotopic data 1234 are integrated with nannofossil data from the Albian (Tiraboschi et al., 2009).

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1238 Figure 8. Nannofossil temperature index (TI), TEX_{86} , and oxygen-isotope values for the Cismon core, Piobbico core and DSDP Site 463 plotted against chemostratigraphy. The age 1239 1240 determination is based on the cyclocronology available for the Cismon core (Malinverno et al., 2010). δ^{13} C data after: Erba et al. (1999) and Méhay et al. (2009) for the Cismon core; 1241 1242 Erba et al. (under final review by the editor) for the Piobbico core; Price (2003), Ando et al. (2008) and Bottini et al. (2012) for DSDP Site 463. Bulk δ^{18} O data after: Erba et al. (2010) for 1243 1244 the Cismon core; Price (2003), Ando et al. (2008) and this work for DSDP Site 463. TOC 1245 after: Erba et al. (1999) and Bottini et al. (2012) for the Cismon core; Ando et al. (2008) for 1246 DSDP Site 463. TEX₈₆ after: Schouten et al. (2003) for DSDP Site 463; this work for the Cismon core (SST calculated using the equation of Kim et al., 2010). On the left is reported 1247 1248 the Os-isotope curve (Bottini et al., 2012) and the volcanogenic CO_2 pulses (red arrows) 1249 reconstructed by Erba et al. (2010). The intervals A to M represent the climatic interludes 1250 (warming and cooling) reconstructed in this work.

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