

Response and additions/changes performed following the Referee 1 (Eric Barefoot) comments: R1Cx.

R1C-1. Synopsis

Response: Thank you for the general appreciation of our old manuscript (OMS) and the summary of the reported research. The concern pointed in the (2) "main findings": "it is not made clear why oxygen isotopes are not affected by these processes." was thoroughly addressed in the revised manuscript (RMS) and in the responses to the reviewer **Minor comments**.

Added/Changed: none.

R1C-2. Overall Comments

Response: Thank you for the very positive comments on our study of the MECO in a well-chosen field site.

Added/Changed: none.

R1C-3. Minor Comments; comments on: Primary vs Diagenetic Signals & MECO isotopic record "I appreciate... then follow up with evidence."

Response: Thank you for this comment. We agree that while our writing reflects how we have worked and thought initially, it does not ease the reading. We have followed the reviewer's suggestion and switched the order of these two sections.

Added/Changed:

Section 5.1 in OMS is Section 5.2 in RMS (lines 391 to 432).

Section 5.2 in OMS is Section 5.1 in RMS (lines 352 to 390).

Both sections were reorganized, several sentences were rewritten, and some explanations and references were added.

R1C-4. Minor Comments; comments on: Primary vs Diagenetic Signals & MECO isotopic record "Additionally, I found the language in these two particular subsections was less clear than some of the other text in the paper..."

Response: Yes, we agree. The discussion on the processes affecting the carbon and oxygen isotope composition ($\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ values) of the dissolved inorganic carbon (DIC) and carbonates were reorganized and reworded in the RMS.

Added/Changed:

"5.2 Primary versus diagenetic signals

The carbonate primary carbon and oxygen isotope compositions may be affected by postdepositional processes, including the neoformation of authigenic and diagenetic phases. Therefore, before the paleoenvironmental interpretation of $\delta^{13}\text{C}_{\text{carb}}$ and $\delta^{18}\text{O}_{\text{carb}}$ records from shallow marine environments, it is necessary to determine primary versus diagenetic signal components. This discrimination requires understanding the factors controlling the primary marine isotopic composition and an evaluation of potential diagenetic overprints on the original geochemical signatures (e.g., Marshall, 1992; Schrag et al., 1995).

Oxygen isotopes in carbonates are controlled by the temperature of formation, the $\delta^{18}\text{O}$ value of the carbonate-precipitating fluid ($\delta^{18}\text{O}_w$), the mineralogy (e.g., higher $\delta^{18}\text{O}$ in dolomite vs. calcite), and any environmental parameter (e.g., pH, salinity) affecting the rate of carbonate precipitation (Swart, 2015). The effect of diagenetic alteration is more pronounced in the case of oxygen isotopes than carbon isotopes due to the high amount of oxygen relative to carbon present in postdepositional fluids and their variable $\delta^{18}\text{O}$ values (e.g., Marshall, 1992; Schrag et al., 1995; Fio et al., 2010). Carbonate with low $\delta^{18}\text{O}$ values can be produced by increasing temperature, freshwater input, and meteoric diagenesis, whereas ^{18}O enrichment could indicate either lower temperature or evaporation (e.g., Marshall, 1992; Patterson and Walter, 1994; Schrag et al., 1995). In contrast, carbon isotopes are not thought to be directly influenced by temperature and are generally more resistant to diagenetic processes (Patterson and Walter, 1994; Schrag et al., 1995; Swart, 2015). However, $\delta^{13}\text{C}$ values are also controlled by kinetic effects, mineralogy, and mainly by the $\delta^{13}\text{C}$ value from the DIC (Wendler, 2013). The primary diagenetic process that affects the

$\delta^{13}\text{C}$ values of the DIC is the oxidation of the organic matter, which produce CO_2 (and DIC species) depleted in ^{13}C (low $\delta^{13}\text{C}$ values). Therefore, the $\delta^{13}\text{C}$ values of the DIC and derived carbonates indicate the source of carbon, including the type of degraded/oxidized organic matter (OM) of different types, original seawater carbon, skeletal and non-skeletal carbonate sources (e.g., Swart, 2015). In proximal depositional environments, however, the $\delta^{13}\text{C}$ values could be modified by (1) OM source, productivity, and burial rate, (2) extrabasinal carbonate input, (3) water circulation/stratification and evaporation, (4) terrestrial runoff and weathering (Saltzman and Thomas, 2012, Läuchli et al., 2021). Considering this, $\delta^{13}\text{C}$ is usually used as a global correlation tool since it can register eustatic sea-level fluctuations, changes in weathering flux, or significant perturbations in the global carbon cycle (e.g., volcanic CO_2 input; Wendler 2013 and references therein).

The degree of diagenetic alteration was assessed through three different approaches. First, was evaluated the relationship between $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ values (Brasier et al., 1996). Statistically, a non-significant correlation (Pearson correlation coefficient; $r < 0.6$) indicates that a diagenetic overprint of the primary isotopic signature can be excluded (e.g., Fio et al., 2010). In both sections, no statistical significant correlation ($r < 0.3$) was found between the $\delta^{18}\text{O}_{\text{carb}}$ and $\delta^{13}\text{C}_{\text{carb}}$ values. This lack of relationship suggests that no or minor diagenetic modifications affected the primary isotopic compositions (Fig. 10). The second approach used to assess the degree of alteration uses clay mineralogy. Kübler and Jaboyedoff (2000) defined four diagenetic zones by comparing illite crystallinity with mineral assemblages and organic matter type. The Belsué and Yebra de Basa samples have 20–30% smectite within the illite-smectite (IS) mixed layers and are within the 3rd diagenetic zone of Kübler and Jaboyedoff (2000), i.e., shallow diagenesis (*ca.* 60–80°C). Another diagenetic indicator is the maximum temperature (T_{max}) reached during the Rock-Eval Pyrolysis (S2), which marks the maturity of the OM. The T_{max} values obtained in samples with relatively high OM content (TOC > 0.5 wt.%; S2 > 0.2) were < 440°C (Fig. 8), which corresponds to the beginning of the oil window (*ca.* 60°C; Espitalié et al., 1985). This maturity level of the organic matter agrees with vitrinite reflectance and Raman measurements in the studied area (Labaume et al., 2016). In summary, the three approaches for assessment of the diagenetic degree, i.e., carbonate $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ values, illite crystallinity, and thermal maturation of the organic matter (T_{max}), suggest that the diagenetic overprint in the studied Belsué and Yebra de Basa rocks is low. The primary isotopic signal is preserved largely in both sections. It can be safely used to study paleoenvironmental conditions and be compared to global key isotopic curves during the MECO event."

R1C-2. "Figure 5, 6, 7, & 10..."

Response:

Agree.

There are four main gaps in data: 1) on Belsué-E section between ~55 and ~65 m, 2) on Belsué-E section between ~85 and ~115 m, and 3) on Yebra de Basa (HR, figure 4) section between ~100 and ~120 m, and 4) on Yebra de Basa (HR, figure 4) section between ~180 m and ~200 m.

In section 3.1 "A total of 101 samples in BS and 157 samples in YB were collected, each of them was composed by *ca.* 200 g of fine-grained and fresh rock from below the weathering depth to avoid alteration and grain size bias", we did not emphasize enough that we tried to sample in the most carbonate rich and as homogeneous as possible fine-grained material. This corresponds to the marls. They represent similarly deep environments, and are carbonate, organic and clay rich, which is what the type of proxies we required. Sampling the sandy clastic intervals can be performed when one only looks at the organic matter, but is less than ideal to explore primary signals in carbonates. Although the exposure conditions are usually ideal for this work, difficulties in sampling in this field area can arise because of either 1) a dominance of sandy facies at the outcrop, or 2) insufficient exposure due to the fine-grained nature of marls (marly intervals in steep topography are usually providing excellent outcrops, but if situated in topographic depressions they can also be more vegetated and lacking exposure).

Samples free intervals 1, 3 and 4 correspond to the most sandy intervals at the moments of maximum deltaic progradation. Sample free interval 2 results of both coarse-grained outcrops at the level of the second progradation in Belsué, and of lack of sufficient exposure in the marlier interval above this progradation. To be more fair in the representation, we highlighted the poor exposure zones and data gaps in Figure 4. Moreover, we erased in Figures 5,6,7, and 10 the line that connect the different intervals of the four data gaps, and we added a fine dashed line of light grey to differentiate the sample and non-sample intervals.

It remains important to note that, as explained by reviewer 1, given our magnetostratigraphic constraints and the fit with global curves, the absence of data in these three intervals has fortunately no impact on our results/conclusions.

Added/Changed:

Added, line 164 to 165 in the RMS: [The samples were mostly marls, corresponding to rocks rich in carbonate, OM, and clays.]

Added, lines 168 to 171 in the RMS: [The exposure conditions were usually ideal for sampling in both sections. However, there were difficulties in four intervals, resulting in gaps in the data. The problems were due to a dominance in sandy facies at the outcrop, corresponding to moments of maximum deltaic progradation, or to poor exposure because of the fine-grained nature of the marls (e.g., Quaternary cover).]

Added, lines 252 to 256 in the RMS: [Three of these data gaps were due to the dominance of sandy facies. In YB, the sandy intervals correspond to the Sabiñánigo sandstone deltaic bodies located approximately at 100–120 m and 180–200 m (YB-HR section; Fig. 4). In BS, the Belsué sandstone interval is placed between 55 and 60 m (Belsué-E section; Fig. 4). The fourth data gap located at 85–115 m in Belsué-E, results of lack of sufficient exposure within the marls and the presence of a coarse-grained sandy interval (Fig. 4).].

Change in figure 4: **Highlighted poor exposure zones**

Change in figures 5, 6, 7, and 10: **Erased connecting lines of data gaps, and added a fine dashed line of light grey to differentiate the sample and non-sample intervals.**

R1C-3. “Age model in Figure 10...”

Response:

Agree. This is missing. Indeed, we simply scaled our sections and the corresponding proxy records based on the magnetostratigraphic tie points (with repositioning our data on Garcés et al., 2014 and Vinyoles et al., 2021 magnetostratigraphic sections).

Added/Changed:

Added in the legend of figure 9 (RMS), lines 366 to 367: [Figure 9: Oxygen isotope ($\delta^{18}\text{O}_{\text{carb}}$) correlation panel for the studied sections (Belsué and Yebra de Basa) with MECO target curves from Alano (Italy, Tethys Ocean, Spofforth et al., 2010), ODPS 1051 (N Atlantic Ocean; Edgar et al., 2010), ODPS 702 (S Atlantic Ocean; Bohaty et al., 2009) and ODPS 738 (S Indic Ocean; Bohaty et al., 2009). Data from the bulk and fine sediments fractions. Highlighted in red the OM rich interval (TOC peak) in Yebra de Basa. The two progradation-retrogradation cycles referred in the text are drawn with grey and white triangles. The data are scaled according to magnetostratigraphic tie points between C18r-18n.2n and C18n.2n-C18n.1r chronos.]

Typos/Misspelling/Style

R1C-4. “I noticed several places where there were copyediting issues. For example: The legend of figure 1 says “Litostratigraphy”, not “Lithostratigraphy”...”

Response: Thanks for pointing out these.

Added/Changed:

Corrected Figure 2. We changed “Litostratigraphy” by “Lithostratigraphy” and “Depositional” by “Depositional”.

We double-checked the RMS and have corrected typos, grammatical mistakes, and bad choose of words. The English was revised.

R1C-5. “I also noticed a few instances where the authors could improve their style. For example:

1. “starving” of what? I assume you mean oxygen, but it could mean food as well. line 60...”

Response: Corrected.

Added/Changed:

Lines 61 to 65 in the RMS:

We added new information and changed the final sentence: [However, while the temperature increase in the oceans has been inferred in multiple sites, the MECO environmental perturbation affected differently the fauna communities (Arimoto *et al.*, 2020). In some locations, the warmer conditions reduced nutrient availability, decreasing the benthic productivity (Arimoto *et al.*, 2020; Bijl *et al.*, 2010, Galazzo *et al.*, 2014; Moebius *et al.*, 2015). In contrast, the Southern Ocean (Moebius *et al.*, 2014) or the Neo-Tethys Ocean (Galazzo *et al.*, 2013) record increased productivity during the MECO.]

The style of the text was improved and several sentences were reworded, which can be easily find in the ms version with "track-changes".

R1C-6. “2. You use “key” twice in this sentence, which gives me the impression that you have awhole keyring. Moreover, the actual meat of the sentence is a little vague, and the reader comes away with no concrete idea of what you mean. Rather than declaring that the MECO can teach us something about the Earth system, I would re-write this to just specifically state what it can teach us. line 52...”

Response: Agree.

Added/Changed:

Lines 50 to 55 in RMS:

We modified the “key” elements and provide additional text to explain more specifically why we care about the MECO:

[Therefore, considering the unresolved MECO driving mechanism(s), and how the Earth system responded to this carbon cycle perturbation, the MECO poses a significant challenge to understanding carbon cycle variations on timescales of several hundreds of thousands of years (Sluijs *et al.*, 2013; Henehan *et al.*, 2020; Sternai *et al.*, 2020). Addressing this challenge requires extensive documentation of the MECO in a range of environments and geodynamic contexts, as well as documentation of its effect on Earth surface dynamics.]

R1C-7. “suffered” is a strange word here. It applies a bit too much humanity to the oceans, for my taste. line 55”

Response: Corrected

Added/Changed: Line 56: We changed “suffered” by “experienced”.

R1C-8. “The (2.4 My) in parentheses is redundant. line 50”

Response: Corrected

Added/Changed: Line 50: We erased the parentheses