This file contains authors responses to review comments by **Anonymous Referee #1** and **Anonymous Referee #2** (responses R1.. in green); and marked-up manuscript

Anonymous Referee #1

The study by Eldrett and colleagues tries to understand and constrain the nature and timing of water-mass evolution in the southern gateway to the Cretaceous Western Interior Seaway (KWIS) and define the associated paleoenvironmental and paleoclimatic processes. This article presents detailed palynological and geochemical analyses from Cenomanian-Conacian interval from southwest Texas,USA. They also analysed materials from the central KWIS, to the south in the Tropical Atlantic and Southern Ocean in order to do correlations and reconstruct N-S water mass circulation. This work proposes another model of ocean circulation in the KWIS to explain the presence of normal oxygenation conditions during CIE (OAE-2) in this zone. It shows, inter alia, a link between sea level variations and N-S oceanic circulation reorganization. Finally, authors discuss and try to explain the global trace metal drawdown during OAE-2.

This is an interesting article with many new data. It provides opportunities to discuss the impact of N-S water mass exchanges on regional paleoenvironmental changes during a period with a predominant latitudinal water mass circulation. However, many ortions/sections should be clarified or developed (- discussion about trace-metal significance (detrital versus authigenic origin); - discussion about sequential stratigraphy; - discussion about tethyan water mass in the western part of the Central Atlantic ocean: ::) and some paragraphs must be organised differently in order to help the reader (result and interpretation presentation with the same stratigraphic subdivision that is chronostratigraphic subdivision).

- 1. â A c p.4 ligne 36 : change "paleonenvironmental" by paleoenvironmental R1. Thankyou for pointing this error, it is now corrected
 - 2. â A c p.5 ligne 25 : "previously published organic and inorganic geochemical analyses" put bibliographic references
- R2. We have inserted the appropriate bibliographic references as suggested.
 - 3. â A c 2. Material and Method; In the supplementary data there is a lack of discussion on the authigenic origin of the trace metals used. To validate their types of source, the major and trace element abundances have to cross-correlated with Al abundances (indicators of detrital influx). See for example Lebedel et al., 2013 (Pal, Pal., Pal.)
- R3. We had not included a discussion on the authigenic origin of the trace metals as we felt this had been addressed in previous publications. We refer the reviewer to Eldrett et al. 2015b, Earth Planetary Science Letters 423, 98-113, in particular Figure 5 and discussion in section 3.3. The relevant trace metals and associated litho-facies are cross-correlated with Al and discussed in that paper. We have inserted a sentence into the supplementary information referring the readers to Eldrett et al. 2015b and have provided all data in the supplementary datafile where the data can be cross-plotted and interrogated.

4. You can also distinguish between redox proxies, Mo, V, U (Calvert and Pedersen, 1993) and palaeoproductivity proxies, Zn, Ni (Hatch and Leventhal, 1992).

R4. We do not believe that Zn and Ni can be distinguished from redox control and used solely as palaeo-productivity proxies. See also reply R11.

- 5. Finally, there is no Measurement accuracy.
- R5. Measurement accuracy table added to the supplemental.
- 6. â A c 3.1 Organic carbon-isotope Stratigraphy; In this section I think it is important to confront geochemical correlations with biostratigraphic correlations. This section seems to suggest that the identification of the Middle Cenomanian event and the Cenomanian-Turonian CIE is not based on age, but only on the magnitude of _13 Corg positive excursion.
 R6. Clarification statement added. Carbon isotope stratigraphy is not just based on magnitude of the positive excursions, but also constrained by biostratigraphy (nannofossils, foraminifera, palynology).

positive excursions, but also constrained by biostratigraphy (nannofossils, foraminifera, palynology), geochronology and astrochronologic calibration. These details were presented in Eldrett et al. 2015a and a clarification statement referring the readers to that publication for detailed discussion is inserted.

- 7. â A c 3.2 Geochemistry p.6; It is necessary to re-organize this section. Use chronostratigraphic subdivision instead of lithostratigraphic subdivision. It lacks a description of the diverse sedimentary series that is the description of the main facies because the paleoenvironmental perturbations are recorded in the litho and biofacies. Are there cherts in these series?
- R7. Re-organizing this section according to chronostratigraphy would in the authors opinion create unnecessary complexity and confusion. The results and majority of the text is presented in lithostratigraphic units and in depth as a presentation of results per core/outcrop section. Most of the geochemical and environmental signals are reflected in the deposition of lithology, not age. Therefore, presenting in chronostratigraphy, particularly the PCA results would be entirely confusing as the main lithological trends would be obscured. The extra interpretative step of regional trends in the discussion of water-masses is presented in chronostratigraphic age as an attempt at synthesis of the regionally important trends. To help clarify, age assignments are provided next to the associated depth in core (in parenthesis) so that the reader can orientate themselves in age as well as depth; also being consistent with the figures. We feel the main lithologic units are adequately presented in this contribution with focus on larger scale trends. Detailed environmental trends related to lithofacies (limestone-marlstone and bentonites) is presented by Eldrett et al. 2015b (Earth Planetary Science Letters 423, 98-113).bed-scale lithofacies descriptions are beyond the scope and is presented by Minisini et al. (Sedimentology, in review).
 - 8. â A c 3.3 Palynology p.7; In order to help the reader, it's better to present the meaning of the different parameters studied and presented in figures 4-10 (for example T/M ratio, Dinocyst P/G ratio, Shannon-Wiener diversity and Simpson Hunter Diversity). This information should not only appear in the supplementary data.
- R8. The meaning of the parameters are now presented in the methods (page 5)
 - 9. â A c 4.1 n Principal Component Analyses z (p. 8), Use chronostratigraphic subdivision instead of lithostratigraphic subdivision (same subdivision as geochemistry section).
- R9. The authors see this would add complexity and confusion obscuring the main environmental signals. These are results and not presented in an interpreted chronostratigraphic context. We have amended the text from 'stratigraphically' to 'lithostratigraphically'; In. 25, and inserted, "The groups are discussed in a chronostratigraphic context in Section 4.2."

- 10. â A 'c p.8 line 19 "Gallium (Ga)-Al2O3 [kaolinite]" I don't understand the direct link between Al2O3 and Kaolinite. Illite also contains Al2O3
- R10. Gallium is generally associated with Al2O3 associated with detrital clays and agree that the direct link with kaolinite is difficult to establish.
 - 11. â Ă ć p. 8 line 27 n redox sensitive trace metal concentrations z. Zn and Ni are also paleoproductivity proxies
- R11. Although we agree with the reviewer in part, the main paleo-productivity elemental proxies are Barium and Phosphorus which have only one oxidation state. Zn, Cu, Ni are redox sensitive elements that are thought to be delivered to the sediment-water interface through the sinking of organic matter and could be used as proxies of organic matter flux. However, solubility and oxidation state of these elements are still a function of redox state and thus not solely an organic matter flux proxy. The delivery mechanisms and the calibration to surface water productivity are debated, so due to these uncertainties we have not split the element preferences from the primary category of redox sensitive. We retain the use of redox sensitive as it encompasses the main elements discussed in the paper (i.e. Mo, V, U).
- 12. â A 'c p.8 line 35 change "paleonenvironmental" by paleoenvironmental R12. We do not see this typo in the text.
 - 13. â A c p. 9 lines 19- 23 "In addition, it is interesting to note that although phytoclasts plot negatively along eigen axis 2 and may represent a reduced masking effect of AOM during oxygenated conditions (Tyson, 1995); they also plot positively along Eigen score 1 (noncarbonate/volcaniclastic trend) alongside freshwater algae and Areoligeracean dinocysts suggestive of a more nearshore environment (Brinkhuis and Zachariasse 1988; Harker et al. 1990; Li and Habib 1996)". This last interpretation must be confirmed by the calculation of a correlation coefficient which, in my opinion, will not show a correlation. Here, you do only a suggestion but not a real interpretation because theposition of the phytoclasts is not at all correlated to the axis 1
- R13. We agree with the reviewer. The suggestion was that freshwater algae and areoligeracean dinocysts are suggestive of a more nearshore environment by Brinkhuis and Zachariasse, 1988, Harker et al. 1990 and Li and Habib, 1996 rather than the relationship solely with axis 1. The statement has been amended to clarify the suggestion. It remains a suggestion as we state and believe other factors that we state (such as the masking effect and potential recycling) would complicate the relationship along axis 1 and reduce any correlation co-efficient.
- 14. â A c p. 9 line 25 change "Figures 4-11" by Figures 4-6, 8-10 R14. Done, thankyou for the observation.
 - 15. â A c p.9 "Paleoenvironment Interpretation" Use the same chronostratigraphic subdivision as geochemistry section (3.2); If the sequence stratigraphy interpretation was not published before it's important to explain it.
- R15. Chronostratigraphic age assignment for each of the main lithostratigraphic units presented and discussed in the text is provided in parentheses and we believe this as the optimal structure of the text. The sequence stratigraphic interpretation has not been previously published and is now included in the supplemental information.
 - 16. â A c p.11, line 16-19 "The sporomorph assemblages during OAE-2 mainly record a relative increase in gymnosperms, in particular during the PCE interval, and thus any increase in T:M ratio may reflect transition from mega-thermal to meso-thermal vegetation (perhaps also reflecting increased pollen production by wind dispersed gymnosperms) in response to

climate cooling episode rather than increased hydrologic cycle". Ti/Al can be used as a proxy for eolian versus fluvial input. What does this proxy show?

R16. The Ti/Al ratio shows an increase during the Plenus Marl event corresponding with recorded increase in gymnosperms. However, during this interval there is also an increase in mafic trace metal abundances, REE and Eu anomaly perhaps reflecting an igneous source, such as the High Arctic Large Igneous Province (see Eldrett et al. 2014; and supplemental figures). The increase in Ti/Al may reflect the emplacement and weathering of a LIP as well as alteration of biotite from the ubiquitous felsic volcanic ash beds rather than solely eolian versus fluvial inputs. We have included a sentence to discuss these uncertainties in Ti/Al as a proxy in the text and the relationship to increased gymnosperms and the PCE.

- 17. â A c P.11 "Regional water-mass evolution"; "In this study we infer three main water-mass properties: i) a restricted suboxic-anoxic marine water-mass characterized by low diversity dinocyst assemblages interpreted to represent a tethyan source; ii) an unrestricted/open marine oxygenated water-mass characterized by high diversity dinocyst assemblages interpreted to represent a boreal source and iii) a partially restricted dysoxic water-mass interpreted to represent a more local central KWIS source."

 It's too direct, you have to explain!! Why do you talk about a tethyan source and not an Atlantic-tethyan source? You specify Â'n a restricted suboxic-anoxic marine watermass of tethyan source" before and after CIE. However Tethyan marine waters are generally well-oxygenated during these time intervals which are not the case of the Atlantic marine waters (see ref Monteiro et al., 2012 Paleoceanography for example: ::)". It would be better to write water mass of Atlantic-Tethyan source. Section organization: use the same chronostratigraphic subdivision than previously.
- R17. This sentence is direct, but we feel appropriate. The reviewer is correct in that the definition of tethyan source was loosely applied here as part of the eastern Tethys was oxygenated during this time. As suggested we have amended the text and specifically refer to an equatorial-Atlantic tethyan source
 - 18. â A c p. 12 lines 8-12 "Lower Cenomanian sediments from the Equatorial Atlantic (ODP Site 1260) are interpreted as being deposited in a stratified suboxic-anoxic marine environment as indicated by laminated organic rich mudrock deposition and positive PCA-2 scores. This interpretation is consistent with a southern tethyan water-mass and a circulation controlled nutrient trap fuelling surface water productivity and anoxic depositional environment (Jiménez Berrocoso et al. 2010; Trabucho-Alexandre, 2010)". I don't understand why it's consistent with a southern tethyan water-mass. According Trabucho-Alexandre et al., 2010, this zone is the seat of upwelling of deep waters coming from the Pacific, no southern tethyan water-mass is mentioned by these authors.
- R18. We agree with the reviewer. Our findings are consistent with the depositional conditions presented by Trabucho-Alexandre, 2010 and Jiménez Berrocoso et al. 2010 and not necessarily linked with a southern tethyan water mass. We have amended the text accordingly.
- 19. â A 'c p.12 line 17: "with mixed dinocyst assemblages". List of the genera R19. We have amended the text to read: "In the Central KWIS (Portland-1 core), the early Cenomanian is characterized by relatively dysoxic depositional conditions, with frequent to common prasinophyte phycomata and mixed dinocyst assemblages, including taxa more typical of the northern and central KWIS such as *Senoniasphaera microreticulata* and *Palaeoperidinium cretaceum*, and the first consistent but mainly rare occurrence of *Bosedinia* cf. sp 1 & 3 which is common in coeval deposits at Demerara Rise; along with low diversity agglutinated benthic foraminfera (**Figure 8**) and the occasional occurrence of rare tethyan calcareous planktonic taxa, together suggestive of a mainly Western Interior Seaway source (Eicher and Diner, 1985)."

20. â A c p.12, line 28 "In the Portland-1 core there is a clear shift from agglutinated to calcareous benthic foraminifera near the top of the MCE interval suggestive of a tethyan influence" why? Is there no agglutinated and calcareous benthic foraminifer in shallow water environment in the Atlantic and Pacific oceans?

R20. We agree with the comment of the reviewer and have clarified this sentence. The transition from agglutinated to calcareous foraminifera had been interpreted as reflecting the incursion of carbonate rich tethyan water into the KWIS, which in part is supported by occurrence planktonic foraminifera, ammonites, nannofossils of tethyan influence (see previous sentence). Detailed discussion of the foraminiferal assemblages we feel is beyond the scope of this contribution and have also inserted "for detailed discussion of tethyan-boreal foraminiferal distribution within Colorado, see Eicher and Diner, 1989)"

21. â A c p.13, lines 35-39. "Furthermore, at ODP Site 1261, the recorded shift towards a more diverse and open-marine dinocyst assemblage is also associated with an increase in the abundance of organic foraminiferal test linings; re-population by calcareous benthic foraminifera (Friedrich et al., 2011), which combined with a reduction in redox sensitive trace metals is indicative of an improvement in environmental conditions and a reduction on the oxygen minimum zone". Warning! There are few samples analysed; 4 samples in 10 m, it's little. This interpretation does not seem really justified because the organic matter concentration is very high.

R21. Indeed there are only four samples, however trace metal data and palynological data (including foraminiferal test linings) are from the same samples which show correspondence. In addition, much higher resolution is available from Friedrich et al. (2011) who recorded re-population of benthic foraminifera. Therefore, we believe it reasonable to suggest improved environmental conditions. However, the wording may be too strong given some of the uncertainty and potential for high frequency variations not captured in the sampling resolution. We have therefore amended the text from "is indicative" to "may indicate" and also inserted "However it should be noted that organic matter concentration remains high".

22. â A c p.15 paragraph n Global trace metal draw-down during OAE-2" line 3, "During the Cenomanian, sediments that have been influenced by tethyan waters"

R22. Inserted "equatorial Atlantic"

Anonymous Referee #2

The study by Eldrett et al is a very nice contribution about the mid Cretaceous Western interior seaway and how WIS sediments are influenced by different water masses that are supposed to originate both from the north and from the south. The authors present a large amount of data that makes it sometimes very hard for the reader to follow the arguments because many data are only shown in the supplements but are discussed in the main text in length and are sometimes very important for the interpretation of the data. Here, it would be good to have some more information in the main figures (maybe one or two additional figures. Overall, I think that this is a nice contribution that is worth to be published in Climate of the Past. However, there are quite a few points

that should be clarified by the authors to increase readibility of the text and the data interpretation. Furthermore, available datasets from the sites studied should be taken into account by the authors to support their statements and interpretation. Therefore, I recommend moderate to major revision. My main points are (in the order they appear in the manuscript):

- 1) Abstract: why is the abbreviation for the Cretaceous WIS written with a K instead of a C?
- R1. This nomenclature for abbreviation follows conventional usuage and previous publications on the topic from the 1990's onwards; either shortening Cretaceous Western Interior Seaway to KWIS or Cretaceous Western Interior Basin to KWIB. As for the K-T boundary event the K is for Cretaceous and T for Tertiary; at the level of system/period, C is usually reserved for the Carboniferous. Within the Cretaceous, at the level of stage, C is used in this paper to abbreviate Cenomanian; if we abbreviated Cretaceous to C it may be confused with the C-T (Cenomanian-Turonian) boundary. The early workers of the Western Interior Seaway did not abbreviate; however to be more concise with fewer words we abbreviate so consistent with other publications.
- 2) The first paragraph and especially the first sentence of the introduction needs much more references to be included. Same is true for lines 20-30 on page 2.
- R2. Our intention for the first sentence was to set the scene; with the detailed references relating to Large Igneous, sea level etc... being stated in lines 20-26. We agree with the reviewer that currently the first sentence reads light on the references, but our concern if inserted is that they will be duplicated in the following sentence. Lines2-30, page 2. We believe the key literature is referenced here; but will review available literature to assess whether additional references are warranted.
- 3) line 11 on page 3: make clear that the benthic zone is only a small part of OAE 2. R3. We agree with the reviewer that the benthic zone as defined by Keller and Pardo (2004) is only a small part of OAE-2; however we also note that the original benthonic zone definition of Eicher and Worstell (1970) is much broader and in northerly KWIS localities spans the entire OAE-2 and post-OAE-2 interval. We have amended the text as follows:
- "..tethyan water during OAE-2 was interpreted to have cumulated in the abrupt oxygenation of the seafloor as recorded by development and persistent abundance of benthic fauna (i.e. Elderbak and Leckie, 2016). However, this interval of benthic faunal abundance in the KWIS was originally termed the benthonic zone by Eicher and Worstell, (1970) who demonstrated that the benthonic foraminifera zone was best expressed in northerly sections where it spanned the entire Cenomanian-Turonian Bridge Creek Limestone, and is less developed in the central KWIS sections where it is stratigraphically restricted to the uppermost Cenomanian (i.e. beds 68-78 at Rock Canyon, Pueblo, Colorado; Eicher and Worstell, 1970), and where it spans only part of the OAE-2 interval and subsequently termed the Benthic Zone by Keller and Pardo (2004).
- 4) line 11 on page 4: where is the connection between figures 1 and 2 and the text? R4. We agree with the reviewers comment. We have deleted "(Figures 1-2)" from the text and inserted reference to the figures more appropriately at the beginning of this section and in the methods. E.g. "The Eagle Ford Gr. was deposited during the Cenomanian to Turonian across the broad Comanche Platform in the southern intersection of the KWIS and northern Gulf of Mexico (Figure 1), "
- 5) chapter 3.2.2 needs a reference to figure 8 R5. We have inserted reference to Figure 8 as suggested.

6) chapter 3.3: at least present the most important features of the palyno-dataset that are used in the following discussion so that the reader has not to go back to the supplements every time.

R6. We utilized the supplementary information in order to reduce the size of the manuscript. We would like to refer to the editor whether the palynological results section should be re-instated to the main text.

- 7) line 25 on page 9: eigen scores are not shown in figures 7 and 11.
- R7. This was also identified by reviewer #1; we have corrected.
- 8) lines 29-31 on page 10: would delete this statement from the text. R8. We have deleted this comment
- 9) lines 14-15 on page 11: since there is no increase in pollen and spores, there is no support for an increased hydrological cycle

R9. In the sections from Texas and Demerara Rise there is not an increase in absolute abundance of pollen spores. However; in the Portland-1 core there is a slight increase (2,000-5,000 c.p.g) so we disagree with the reviewers observation and a discussion is warranted. Regardless of absolute abundance, our data show a pollen assemblage shift to gymnosperm dominance; something that is highly relevant when comparing with other palynological records discussing increased hydrological cycle (e.g. Van Helmond et al. 2014) and subsequent citations.

- 10) line 19 on same page: what is the indication for climate cooling? Only the PCE is cooler, under background values that are much warmer than before or after OAE2! R10. We agree with the reviewer that the primary evidence for climate cooling is linked with the PCE interval that elsewhere is associated with a sea surface temperature cooling (e.g. Forster et al. 2007; Van Helmond et al. 2014, 2016); climate cooling and drop in PCO² (See Jarvis et al. 2011) as well as influx of boreal fauna. The increase in gymnosperms may therefore reflect expansion of conifer forests during this "cool snap". Additional evidence for climate cooling for the Turonian (i.e. what were background levels in the Turonian) are not well constrained. The persistence of boreal fauna in the sections presented here may indicate the presence of additional cooling episodes throughout the Turonian. Further work is required on this topic.
- 11) Chapter 4.2.2: why should the tethyan water mass be suboxic-anoxic? This is inferred by the authors at the beginning of this chapter and then used in the following interpretation but it is never shown convincingly to the reader that this is the case. What is the independent evidence for this? Same holds true for the boreal water mass. Are there any other indications other than own data and interpretations? If yes, present them in detail. So far, the main problem with this chapter is that there is no prove that the suggested water masses existed and are characterized by the suggested data in the way they are presented here.
- R11. This point was also raised by reviewer #1. In our initial response to reviewer #1 were thought the introduction section was adequate/to the point. However, as both reviewers have raised this issue we have expanded the introduction and detail the independent evidence
- 12) line 18 on page 12: why is this indicative for an WIS source? Explain and justify in detail. Same with the argument in lines 28-29 with the shift from agglutinated to calcareous forams. Why is this a water mass characteristic and not simply a matter of preservation or changing food availability? Technically I wonder how the foram data were produced. Are the based on the linings in the palynological samples as indicated by the figure headings? I am not aware of a single study that has shown this to work.
- R12. This point was also raised by reviewer #1. (comment 20 and response). As per previous response "We agree with the comment of the reviewer and have clarified this sentence. The transition from agglutinated to calcareous foraminifera had been interpreted as reflecting the

incursion of carbonate rich tethyan water into the KWIS, which in part is supported by occurrence planktonic foraminifera, ammonites, nannofossils of tethyan influence (see previous sentence). Detailed discussion of the foraminiferal assemblages we feel is beyond the scope of this contribution and have also inserted "for <u>detailed discussion of tethyan-boreal foraminiferal distribution within</u> Colorado, see Eicher and Diner, 1989)"

The benthic foram data was produced through two methods that we shall make clearer in the methods and figure captions; i) test linings from the palynological residues; ii) micropalaeontological analyses from sieved residues and thin sections. As previously mentioned, detailed inclusion of the micropalaeontological data we feel is beyond the scope of the paper and we primarily discuss the benthic abundances; inclusion would also lengthen the paper significantly as every sample analysed for palynology has an associated micropalaeontological dataset; as well as ~40 outcrops and we believe this requires a dedicated contribution. The occurrences of benthic foraminifera in the micropaleontological analyses and test linings in palynological assemblages are in general good agreement (as presented in Figure 4; quantified as c.p.g; and Figure 8 as relative abundance) and the method is demonstrated to work in this study. As below (comment 14) we shall also include published benthic foraminiferal data for sites 1260 and 1261.

- 13) line 31 on same page: at this point in the succession, there are no benthic forams, so the statement above in lines 28-29 cannot be valid!
- R13. We disagree with the observation of the reviewer; there are foraminifera test linings (including for outside the initial 100 count specimens) in almost all samples throughout the Graneros to Hartland (with the exceptions of the 160.07m and 185.53m samples [column GQ in the Portland-1 datafile]). In addition, the statement above in lines 28-29 refers to the transition near the top of the MCE interval that is within the Graneros Shale and thus are also two completely different intervals of the core.
- 14) lines 32ff on page 13: there are benthic foraminiferal assemblage data available form these sites, how do they compare to the data produced by linings? This would be a good test to show if the presented foram data of this study are of any value.

 R13. We agree with the reviewer and shall include benthic foraminifera abundances for Sites 1260 and 1261 in figures 9-10; and are in good agreement with the foram lining data.
- 15) lines 2-3 on page 14: I am not aware that there are any data e.g. Nd isotopes from these sites that support a boreal influence. Furthermore, the authors state that this water mass should only influence shallow water settings. However, a cold boreal watermass should be denser than wormer waters near the tropics and therefore influencing bottom waters and not surface waters as suggested here.
- R15. As we state "The southern expression of this boreal influence is therefore limited in duration and extent". There is a Nd isotope excursion at this horizon and the nature of Nd signal is complex and requires additional localities from the KWIS., We state that our findings "indicating more complex interaction between water-masses and the oxygen minimum zone" and requires further work to resolve. It is an assumption by the reviewer that a boreal watermass would be denser as it would be colder; currently thermal gradients are not well constrained and salinity variations, particularly in the Western Interior Seaway are debated with suggestions of relatively freshwater (although we find no evidence of freshwater algae in our dataset). Our principal data are dinocysts that are mostly indicative of the photic zone so the watermasses are inferred to reflect surface water; however the vertical expression is apparent with sediment-water interface becoming oxygenated during OAE-2 associated with boreal taxa/watermass. Whether this is direct evidence for intermediate or deep water is unclear and we have included a statement in the text to raise this point see also reviewer comment 22

16) line 35-36 on page 14: nowadays, nobody thinks anymore that the Cretaceous had a equitable climate!

R16. We agree and that is why it is stated .." than previously thought". In addition, this is an important point to make as not many studies have investigated this topic; the contribution of Gambacorta et al., (2016) is notable and our data is supportive of the proposition.

- 17) lines 23-24 on page 15: but the red dots are all over the place in figure 15 and quite a few from Demerara Rise even above 3. What is the r2 for these data? Further in this chapter, Mo/TOC is used to say something about a silled basin situation at this site. Why not simply cite the papers that show that there was no sill during that time (e.g. seismic evidence)? R16. In unrestricted settings such as the Namibian Shelf and the OAE-2 interval presented here, there is a positive but generally weak co-variance between Mo and TOC and so would expect poor r2 values. In addition, preferential enrichment of Mo in euxinic conditions and in particular recycling in the particulate shuttle usually results in a TOC threshold for enhanced Mo enrichment; so the basic relationship is non-linear. We have clarified this in the text and inserted "weak positive co-variance between Mo and U..". Public domain evidence for absence of a sill across the US Gulf Coast is limited and along with the Demerara Rise ambiguous as reflect present-day geometries.
- 18) last paragraph page 15: wouldn't be the absolute amount of refractory terrestrial organic matter (RTOM) an even more important factor than the T:M ratio alone? The ration could be high even when there is less RTOM and therefore a lower influence on Mo! Since this is not quantified, this is a weak justification and discussion.

R18. We agree with the reviewers comment. The absolute amount of refractory terrestrial organic matter (RTOM) would be more significant than the T:M ratio alone. However; properly quantifying RTOM as far as we are aware is not currently possible. The major component of the organic matter is amorphous organic matter (AOM) and as stated in the text its origin is relatively poorly constrained; some part can be degraded terrestrial and/or marine in origin. Given that we cannot yet adequately identify and quantify the origin and composition of AOM and thus RTOM; the T:M ratio is the best approximation of terrestrially derived palynomorphs (and thus a component of RTOM). The text is slightly amended to include "refractory" and feel this uncertainty is now adequately captured, "We cannot determine the impact of this observation as the T:M ratio reflects a relatively small proportion of the total *refractory* organic matter, however further investigation is warranted into the variable origin of the more dominant and relatively unknown component, namely AOM; and whether Mo is preferentially incorporated within different organic matter components".

- 19) first paragraph on page 16: How do these factors deplete Mo? Please explain the details. R19. The processes and controls have been included as suggested by the reviewer.
- 20) lines 8-9 on same page: but isn't that what you are proposing above?
 R20. In part we agree, but the actual relationship between refractory organic matter and Mo is not documented by Dickson et al. (2016); as such this is the first integrated data of terrigeneous organic matter and Mo enrichments.
- 21) lines 21-23: check benthic assemblage data for these sites if available and see if there are benthic forams occurring in these intervals. If yes, these are oxygenation events, if not, it was anoxic. This would be an independent proof of the statements made here.

 R21. Benthic foraminifera are present (Friedrich et al. 2006, 2011) and reflect re-population events associated with improved oxygenation. This data will be included in Figures 9-10 (see comment 14) and the following text has been inserted ". This interpretation is also supported by the occurrence of benthic foraminifera during the OAE-2 interval in both organic rich and organic lean sediments from

Demerara Rise (Friedrich et al. 2006, 2011); Texas (Lowery et al. 2014; Dodsworth, 2016; this study) and central KWIS (e.g. Eicher and Worstell, 1970; Keller and Pardo, 2004; Elderbeck and Leckie, 2016 and references therein).

- 22) point 1 in the conclusions: are these water masses be interpreted to be surface and bottom-water masses at once? This has to be clarified in the discussion.
- R22. See comment R15. This has been clarified in the discussion.
- 23) Figure 2: in part "a" it is hard to figure out the core locations.
- R23. Core locations shall be made bigger and unique symbology
- 24) Figure 3: what are the horizontal red lines?

R24. Horizontal red wavy lines are hiatal surfaces. The captions shall be expanded to better explain the symbology in the figure

Cenomanian to Coniacian Water-mass Evolution in the

Cretaceous Western Interior Seaway of North America and Equatorial Atlantic

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Abstract. The Late Cretaceous Epoch was characterized by major global perturbations in the carbon cycle, the most prominent occurring near the Cenomanian-Turonian (CT) transition marked by Oceanic Anoxic Event/OAE-2 at 94.9 – 93.7 Ma. The Cretaceous Western Interior Seaway (KWIS) was one of several epicontinental seas in which a complex water-mass evolution was recorded in widespread sedimentary successions. This contribution integrates new data on the main components of organic matter, geochemistry, and stable isotopes along a North-South transect from the KWIS to the equatorial western Atlantic and Southern Ocean. In particular, cored sedimentary rocks from the Eagle Ford Group of West Texas (~90-98 Ma) demonstrate subtle temporal and spatial variations in paleoenvironmental conditions and provide an important geographic constraint for interpreting water-mass evolution. High latitude (boreal-austral), equatorial Atlantic tethyan and locally sourced Western Interior Seaway water-masses are distinguished by distinct palynological assemblages and geochemical signatures. The northward migration of an equatorial Atlantic tethyan water-mass into the KWIS occurred during the early-middle Cenomanian (98-95 Ma) followed by a major re-organization during the latest Cenomanian-Turonian (95-94 Ma) as a full connection with a northerly- boreal water-mass was established during peak transgression. This oceanographic change promoted de-stratification of the water column and improved oxygenation throughout the KWIS and as far south as the Demerara Rise off Suriname. In additionaddition, the recorded decline in redox-sensitive trace metals during the onset of OAE-2 likely reflects a

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WIS written with a K instead of
a C?

R1. This nomenclature for abbreviation follows conventional usuage and previous publications on the topic from the 1990's onwards; either shortening Cretaceous Western Interior Seaway to KWIS or Cretaceous Western Interior Basin to KWIB. As for the K-T boundary event the K is for Cretaceous and T for Tertiary; at the level of system/period, C is usually reserved for the Carboniferous. Within the Cretaceous, at the level of stage, C is used in this paper to abbreviate Cenomanian; if we abbreviated Cretaceous to C it may be confused with the C-T (Cenomanian-Turonian) boundary. The early workers of the Western Interior Seaway did not abbreviate; however to be more concise with fewer words we abbreviate so consistent with other publications.

genuine oxygenation event related to open water-mass exchange and may have been complicated by variable contribution of organic matter from different sources (e.g. refractory/terrigenous material), requiring further investigation.

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Author Contributions: P.D. conducted palynological analyses; M.W. principal investigator for trace metal analyses; D.M. principal investigator for core description; J.E., and S.B. and D.M, integrated and analysed the datasets; all authors co-wrote the paper.

10 The authors declare no conflict of interest.

KEYWORDS: Cenomanian, Turonian, palynology, trace metal, anoxia, water-mass, boreal, tethyan, OAE, Western Interior Seaway

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15 1. Introduction

The Late Cretaceous Epoch was characterized by sustained global warming, emplacement of several Large Igneous Provinces (LIPs), global extinctions, global sea-level highstands leading to several epicontinental seaways, and major global perturbations in the carbon cycle termed Oceanic Anoxic Events (OAE's), the most prominent occurring at the Cenomanian-Turonian transition, and termed OAE-2 (Schlanger and Jenkins, 1976). This event is globally recognized by a positive carbon isotope excursion (CIE) reflecting the widespread sequestration of ¹²C-enriched organic matter in marine sediments under global anoxic conditions (see Jenkyns, 2010 and references therein). Proposed hypotheses for initiation of global anoxia and enhanced carbon sequestration include long term triggers such as changes in ocean circulation and eustatic sea level rise, flooding large areas of continental shelves promoting global stratification and stagnation in greenhouse climates (Erbacher et al. 2001), abrupt episodes of volcanogenic activity and emplacement of large igneous provinces (LIPs; Orth et al. 1993; Snow et al. 2005; Turgeon and Creaser, 2008; Kuroda et al. 2007; Duvivier et al. 2014) releasing large quantities of CO2 in the atmosphere and increasing delivery of hydrothermally-derived and weathered nutrients into the photic zone enhancing primary production (e.g., Adams et al., 2010); or a combination of both (e.g., Kidder and Worsley, 2010). However, increasing evidence indicates a decoupling in the precise timing of the CIE (hence OAE-2) and the location of organic-rich sediment deposition, reflecting that deposition of organic-rich sediment was modulated and ultimately dependent on local and regional processes (basin restriction, water stratification, bottom currents, sediment input) although favoured by global phenomena (sea level change, orbital forcing) (e.g., Trabucho-Alexandre et al., 2010). This is particularly apparent within shallow epicontinental seaways such as the southern Cretaceous Western Interior Seaway (KWIS) where parts of

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The first paragraph and especially the first sentence of the introduction needs much more references to be included. Same is true for lines 20-30 on page 2.

Our intention for the first sentence was to set the scene; with the detailed references relating to Large Igneous, sea level etc... being stated in lines 20-26. We agree with the reviewer that currently the first sentence reads light on the references, but our concern if inserted is that they will be duplicated in the following sentence. Lines 2-30, page 2. We believe the key literature is referenced here; but will review available literature to assess whether additional references are warranted.

the seaway experienced anoxia and recorded organic-rich black shales prior to the OAE-2 interval, and in contrast to other globally recognized sections that recorded organic-rich sediments during the OAE-2 interval (e.g. Plenus Marl and Bonarelli intervals in Europe); the KWIS recorded relatively organic-lean and oxygenated sediments (*e.g.*, Meyers, 2007; Eldrett et al., 2014). Thus, the oceanographic regime of the southern KWIS and its influence on the geologic expression of OAE-2 in this shallow epicontinental sea has been considered unique. It has been proposed that ocean circulation in the KWIS was restricted during the Cenomanian, promoting anoxia due to a sill in the southern gateway (i.e. Texas/Mexico); and that late Cenomanian sea level rise (Greenhorn cyclothem of Kauffman, 1977, 1984) was sufficient to reach a critical sill depth allowing a breach of the southern end of the seaway allowing rapid incursion of warm, normal saline tethyan waters (Arthur and Sageman, 2005). Previous publications have characterized the inflow of tethyan waters at this time by the i) improved environmental conditions as indicated by the sharp increase in abundance and diversity of foraminiferal/molluscan and ammonite assemblages reaching far north into the KWIS (McNeil and Caldwell, 1981; Kauffman, 1984, 1985; Eicher and Diner, 1985; Elder, 1985; Leckie et al., 1998; Caldwell et al., 1993; Kauffman and Caldwell, 1993, Elderbak and Leckie, 2016) and ii) the lithologic transition from organic-rich mudrocks to a highly bioturbated limestone dominated facies (Corbett et al. 2014; Lowery et al. 2014).

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The inflow of southern more saline waters into the southern KWIS during the latest Cenomanian has been proposed to either promote overturning of the water-column or mixing with a northern water-mass resulting in "caballing" and the production of a third more dense water-mass (Hay et al., 1993; Slingerland et al., 1996). In either scenario, the inflow of tethyan water during OAE-2 was interpreted to have cumulated in the abrupt oxygenation of the seafloor as recorded by development and persistent abundance of benthic fauna (i.e. Elderbak and Leckie, 2016). However, this interval of benthic faunal abundance in the KWIS was originally termed the benthonic zone by Eicher and Worstell, (1970) who demonstrated that the benthonic foraminifera zone was best expressed in northerly sections where it spanned the entire Cenomanian-Turonian Bridge Creek Limestone, and is less developed in the central KWIS sections where it is stratigraphically restricted to the uppermost Cenomanian (i.e. beds 68-78 at Rock Canyon, Pueblo, Colorado; Eicher and Worstell, 1970), where it spans only part of the OAE-2 interval and was subsequently termed the Benthic Zone by Keller and Pardo (2004) tethyan water during OAE 2 cumulated in the abrupt oxygenation of the seafloor as recorded by development of benthic fauna, termed the benthonic zone (Eicher and Worstell, 1970) or Benthic Zone (Keller and Pardo, 2004). However, it should be noted that the benthonic/benthic zone in the KWIS has also been shown to correspond with the equatorial migration of boreal dinoflagellate cyst (dinocyst) taxa (Eldrett et al. 2014; Van Helmond et al. 2016) and has been correlated with a short-lived climate cooling episode termed the Plenus Cold Event (PCE; Eldrett et al. 2014; Van Helmond et al. 2014; 2016; Elderbak and Leckie, 2016), whereby similarly cool boreal waters invaded northern and central Europe (Jefferies, 1963; Gale and Christensen, 1996; Voigt et al., 2006; Jarvis et al., 2011) and equatorial waters cooled by up to 4°C (Forster et al. 2007). It is therefore difficult to reconcile the late Cenomanian northerly inflow of a warm tethyan water-mass into the southern KWIS, at a time of global cooling, the southerly restriction in benthic fauna and the coeval southerly equatorial migration of boreal taxa and associated water-mass in the KWIS and Europe. It is plausible that a much more complex oceanographic system existed in the KWIS; such as that modelled by Slingerland et al. (1996) and

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Kump and Slingerland (1999), whereby a strong cyclonic gyre developed in the central KWIS drawing both tethyan waters northward along the eastern margin and boreal waters southward along the western margin of the seaway (see also discussions in Elderbak and Leckie, 2016).

In order to better understand and constrain the nature and timing of water-mass evolution in the southern gateway to the KWIS, and the associated paleoenvironmental and paleoclimatic processes, this contribution presents detailed palynological and geochemical analyses within a multidisciplinary framework from the Eagle Ford Group (Gr.) and bounding formations of the Buda Limestone and Austin Chalk from southwest Texas, USA. Furthermore, to place the southern gateway of the KWIS into a more supra-regional understanding we also analysed correlative materials from the central KWIS (Portland-1 core, Colorado) and to the south in the Tropical Atlantic (ODP Leg 207, Demerara Rise) and Southern Ocean (ODP Leg 183, Kerguelen Plateau).

1.1. Geological Setting

The Eagle Ford Gr. was deposited during the Cenomanian to Turonian across the broad Comanche Platform in 15 the southern intersection of the KWIS and northern Gulf of Mexico (Figure 1), which represented part of the >3,000 km long foreland basin that formed behind the greater Cordilleran retro-arc fold-and-thrust belt during Late Mesozoic through Eocene times along the inboard side of the Cordilleran magmatic arc and accreted allochthonous terranes of North America (e.g., Burchfiel et al. 1992; Dickinson, 2004). The regional tectonic setting was influenced during the Cretaceous by the subduction of the conjugate oceanic plateau to the Shatsky 20 Rise (Liu et al., 2010) dynamic topography from the subducting Farallon Plate (Liu, 2014) and thermally subsiding Gulf of Mexico margin, resulting in the development of broad ramp-shelves including the Comanche Platform with reactivated basement structures defining intra-shelf basins, such as the Maverick Basin and structural highs such as the Terrell Arch. The Cenomanian-Turonian Trans-Pecos region (Figure 2) was deposited in a distal sediment starved setting >500 km from the nearest shoreline during a locally quiescent 25 tectonic period resulting in stable platform conditions and gradual subsidence ideal for the preservation of mudstones, limestones and bentonites of the Eagle Ford Gr., the underlying Buda Limestone and overlying Austin Chalk. (Figures 1-2).

1.2. Previous Palynological Studies

Previous Cenomanian-Turonian palynological studies of the KWIS include those of Brown and Pierce (1962), Christopher (1982), Courtinat (1993), Li and Habib (1996), Cornell (1997), Dodsworth (2000; 2016), Harris and Tocher (2003), Eldrett et al (2014, 2015a, b) and Van Helmond et al. (2016). Brown and Pierce (1962) first reported dinocysts and other palynomorphs from the Eagle Ford Gr. in northeast Texas, whereas terrestrial sporomorphs were recovered from the Woodbine interval of the Eagle Ford Gr. by Christopher (1982).
 Subsequently, most studies have focused on the central part of the KWIS and in particular the Global boundary Stratotype Section and Point (GSSP, "golden spike") for the base of the Turonian at Rock Canyon, Pueblo,

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line 11 on page 4: where is the connection between figures 1 and 2 and the text?

R4. We agree with the reviewers comment. We have deleted "(Figures 1-2)" from the text and inserted reference to the figures more appropriately at the beginning of this section and in the methods.

Colorado (e.g., Courtinat, 1993; Li and Habib, 1996; Dodsworth, 2000; Harris and Tocher, 2003). For the southern sector of the KWIS relatively few sections have been published, notably Cornell (1997) reporting diversified mainly gonyaulacacean dinocyst assemblages from the massive limestones of the Buda Formation (Fm.) in Dona Ana County, New Mexico; and more recently Eldrett et al (2014) reporting the incursion of boreal dinocysts in the Eagle Ford Gr. during the OAE-2 CIE interval corresponding with improved bottom-water oxygenation. This equatorial migration of the boreal dinocyst complex Cyclonephelium compactum-C. membraniphorum has subsequently been recorded across the KWIS and northern Tethys (see Van Helmond et al. 2016). Many of the regional and globally recognized age diagnostic biostratigraphic events encountered in the KWIS were calibrated against an astronomically tuned and geochronologically constrained age model for the Cenomanian-Turonian and early Coniacian based on a relatively expanded section of the Eagle Ford Gr. and bounding units of the Austin Chalk and Buda Limestone that were recovered from the Shell Iona-1 research core, in west Texas (Eldrett et al. 2015a). Paleoenvironmental reconstructions of the Eagle Ford Gr based on the biostratigraphic assemblage data presented by Eldrett et al. (2015a) were beyond the scope of that paper; but some aspects were presented as part of an integrated multidisciplinary contribution demonstrating that obliquity and precession forcing on the latitudinal distribution of solar insolation may have been responsible for the observed lithological and environmental variations through the Cenomanian, Turonian and Coniacian in midlatitude epicontinental sea settings (Eldrett et al. 2015b). Subsequently, Dodsworth (2016) provided more detailed paleonenvironmental interpretations from palynological assemblages primarily from the Eagle Ford Gr. exposed at the Lozier Canyon cliff section, Terrell County, west Texas, which is similar to the Innes-1 core (see below) and contains a significant unconformity (duration $> 2 \frac{\text{million years; } M \text{myrs}}{\text{myrs}}$) within the Turonian. This unconformity is much less intense (duration_<0.2 Mmyrs) in the more distal Iona-1 core.

This paper aims to build onep and expand these previous contributions by: i) providing detailed palynological interpretations of the Eagle Ford Gr. based on several locations enabling a more widespread geographic understanding; ii) integrating multidisciplinary datasets including organic and inorganic geochemistry allowing a greater understanding of the main paleoenvironmental controls; and iii) presenting new data analysed from sections further to the north in the KWIS (Portland-1 core) and to the south in the Tropical Atlantic (ODP Leg 207, Demerara Rise; Figures 1-2) and Southern Ocean (ODP Leg 183, Kerguelen Plateau) to provide insights into supra-regional ocean circulation and water-mass evolution during the Cenomanian-Turonian Greenhouse climate state.

2. Material & Methods

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Core material from the Eagle Ford Gr., and bounding units in the Maverick Basin, West Texas, U.S.A., were analysed for visual kerogen analyses (palynology, palynofacies). The core material was sampled along a physiographic transect from the main Comanche carbonate shelf (Innes-1) towards the edge of the Maverick intra-shelf basin (Iona-1) and central part of the intra-shelf basin (well 'X'); Figure 1-2). Outcrop sections were also analysed in the San Marcos Arch area near Austin along the Bouldin Creek section. In order to compare

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data collected from the Eagle Ford Gr., with any regional trends, data were also collected further to the North near the base of the Turonian GSSP (Kennedy et al. 2005; Sageman et al. 2006) in Colorado (USGS Portland-1) and to the South in the Tropical Atlantic off Suriname (ODP Leg 207; Sites 1260 and 1261, Demerara Rise; Shipboard Scientific Party, 2004a, 2004b). Site locations are presented in Figures 1-2. - Material was also collected from Site 1138, Kerguelen Plateau (Shipboard Scientific Party, 2000). Palynological analyses on these sites were conducted under both transmitted and fluorescence microscopy and the results supplemented and compared to newly collected and previously published organic and inorganic geochemical analyses including total organic carbon (TOC) and major, minor and trace elements (Erbacher et al. 2005; Forster et al. 2007; Friedrich et al. 2008; Hetzel et al. 2009; Joo and Sageman, 2014; Duvivier et al. 2014; Lowery et al. 2014; Eldrett et al. 2014; 2015a, b; Sun et al. 2016; Dickson et al. 2016; Minisini et al. in review). Palynological parameters presented include i) the ratio between terrestrial (T) and marine (M) palynomorphs (T:M ratio) as a proxy for terrestrial input; ii) the ratio between peridinioid or P-cysts and gonyaulacoid or G-cysts (P:G ratio) of the dinocyst assemblage as a proxy of nutrient input. Diversity of the dinocyst assemblage was also calculated using both Shannon-Wiener (H) and Simpson-Hunter (D) indexes. Detailed palynological methods and associated discussion of paleoenvironmental parameters are provided in the supplemental information. In addition to the quantification of organic foraminifera test linings from palynological residues, benthic foraminifera abundances were also counted from a combination of micropaleontological picked residues and thin sections following the methods detailed in Eldrett et al. (2015a) and supplemented by published benthic foraminiferal records (e.g. Friedrich et al. 2006, 2008, 2011). Principal Component Analyses (PCA) were run on this integrated palynological-geochemical dataset to elucidate the primary controls for paleoenvironmental and paleoclimatic interpretations. Details of site locations and methodologies employed are presented in the supplemental information.

25 3. Results

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The following section describes some of the key results that are referred to in the discussion section and presented in **Figures 3-11**. Detailed results and data are presented in the supplemental information and datafile.

3.1. Organic carbon-isotope Stratigraphy

Significant organic carbon isotopic differences are observed throughout the studied interval. The Buda Limestone record $\delta^{13}C_{org}$ values ~-26‰ and become more negative within the Eagle Ford Gr. where background values are ~-27 to -28‰ for Iona-1, Innes-1 and well 'X'. From these background levels, five notable positive-;

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Palynology p.7; In order to help the reader, it's better to present the meaning of the different parameters studied and presented in figures 4-10 (for example T/M ratio, Dinocyst P/G ratio, Shannon-Wiener diversity and Simpson Hunter Diversity). This information should notonly appear in the supplementary data.

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and three negative- carbon isotope excursions (CIE) of varying magnitudes were recognized. These excursions can be correlated with the English Chalk reference section of Jarvis et al. (2006; see Eldrett et al. [2015a_for detailed discussion of biostratigraphic calibration of CIE's in the KWIS) and compared with the previously published data for ODP Leg 207 and KWIS as presented in **Figure 3**. Two of these CIE's have specific relevance for this contribution: i) the ~2‰ positive $\delta^{13}C_{org}$ excursion in Iona-1 (143.73m-139.27m) and Innes (76.88m-74.63m) that corresponds with the Middle Cenomanian Event (MCE) and ii) the Cenomanian-Turonian CIE that is clearly expressed with a positive CIE of up to 4‰ occurring in Iona-1 (112.4505.96m-92.73m), Innes-1 (55.74 m- 42.51 m) and well 'X' (1639.9 m – top not recorded as above the cored interval). It should be noted that the definition of the base of the Cenomanian-Turonian CIE has been re-interpreted to include the precursor events presented in Eldrett et al. (2014, 2015a); as such the base of the CIE at Iona-1 is moved from 105.96m to 112.45m and is assigned an age of 95.01±0.12 Ma based on the obliquity age model presented in Eldrett et al. (2015).

3.2. Geochemistry

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3.2.1. Southern KWIS (Texas)

The Buda Limestone (early Cenomanian; ca 98-97.5 Ma) is characterized in the Iona-1 core by high percentage of Cealcium Oexide (CaO, >45 wt %) and low percentages of Aluminium Oxide (Al₂O₃: <2 wt %), Silicon Dioa Oxide (SiO₂: <6 wt %) and Titanium DioOxide (TiO₂: <1 wt %). Exceptions occur in the interbedded bentonite layers that record higher values of Al₂O₃ (>10 wt %); SiO₂ (>25 wt %) and TiO₂ (~1 wt %). Redox sensitive trace metal concentrations and enrichment factors (EF) in the limestones are low with Molybdenum (Mo); Uranium (U) and Vanadium (V) recording <1ppm/<2 EF; <2ppm/<4 EF and <20ppm/<2 EF respectively. The lower Eagle Ford Gr. (ca. 97.2-94.9 Ma) is characterized by high TOC and concentrations of redox sensitive trace metals that are enriched compared to average shale. In particular, the basal most part of the lower Eagle Ford Gr. at Iona-1 (153m - 144m) records the highest trace metal enrichments with Mo_{EF} = ave. 400, U_{EF} = 12 and $V_{EF} = 17$. These trace metal enrichments decline sharply at the base of the MCE interval (143.37m – 142.27m) after which they increase again and become relatively enriched (Mo_{EF} = ave. 140, U_{EF} = 7 and V_{EF} = 13). A similar trend is also recorded in both Innes-1 and well 'X' albeit at overall lower values. In all three cores redox sensitive trace metal concentrations and enrichments start to decline prior to the lower - upper Eagle Ford Gr. boundary (~94.6 Ma). The upper part of the Eagle Ford Gr. is characterized by generally low redox sensitive trace metal concentrations and enrichments in all three cores, with minima associated with the interpreted enthonic benthic foraminiferal oxic zone (see Eldrett et al. 2014). However, within the Benthic Oxic Zone, there is a recorded increase in Ti/Al, maffic trace elements and europium anomaly (Eu/Eu*; see Eldrett et al. 2014; Figure DR5). Variations in redox trace metals occur between the limestone-marlstone couplets; with marlstones recording slightly elevated enrichments compared to the limestones (see Eldrett et al. 2015b for details). The uppermost part of the Eagle Ford Gr. (corresponding with the Langtry Member; sensu Pessagno,

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Organic carbon-isotope Stratigraphy; In this section I think it is important to confront geochemical correlations with biostratigraphic correlations. This section seems to suggest that the identification of the Middle Cenomanian event and the Cenomanian-Turonian CIE is not based on age, but only on the magnitude of _13 Corg positive excursion.

Clarification added

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1969; Mb. Mb.~92.5-90.3 Ma) record a slight increase in redox sensitive trace metals in mostly marlstone lithologies in both Innes-1 and Iona-1 cores (**Figures 4-5**). The overlying Austin Chalk is generally not enriched in redox sensitive trace metals with the exception of the interbedded marlstones. The Bouldin Creek outcrop locality was not analysed for geochemistrytrace metals as part of this study.

3.2.2. Central KWIS (USGS Portland-1, Colorado)

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The Dakota Sandstone and Graneros Shale interval in the USGS Portland-1 core (213m to 175m; ~100-95.8 Ma; Figure 8) is predominantly comprised -of high relative concentrations of SiO₂ (ave. ~60 wt %) and Al₂O₃ (ave. ~15 wt %). The Thatcher Limestone interval (~185m; ~96.5 Ma) is represented by a single sample that records a slight relative increase in CaO (~8 wt %), but it is likely that this sample _does adequately represents or resolves the varied lithological end-members recorded in this interval. A relative increase in CaO is recorded from the base of the Lincoln Shale Mb. to near the top of the Hartland Shale Mb. (175.29m – 150m; 95.8-94.7Ma) reaching up to 25 wt %; and then increase substantially to ~40 wt % CaO marking the lithological break of the base of the Bridge Creek Limestone Mb. (~148.6m).

Throughout the Dakota Sandstone and Graneros Shale intervals the majority of the redox sensitive trace metals record low concentrations and are not significantly enriched compared to average shales (e.g. U, V, Ni, Cu, Zn) and Mo is only slightly enriched (Mo_{EF}= ave. 3; ppm= ave. 4 ppm; Figure 8). Redox sensitive trace metals concentrations increase slightly in the Lincoln Shale Mb. and only become significantly enriched in the Porltand-1 core during the deposition of the Hartland Shale Mb. The overlying Bridge Creek Limestone Mb. and in particular the interval recording the OAE-2 CIE is characterized by an overall reduction in redox sensitive trace metals and lack of significant enrichments compared to average shale values. However, variations in redox sensitive trace element enrichments are recorded betweenin the marlstone and limestone couplets of the Bridge Creek Limestone Mb.

3.2.3. Demerara Rise, Atlantic Ocean (ODP Leg 207, Sites 1260 and 1261).

The pre-OAE-2 sediments recovered from ODP sites 1260 and 1261 record high and variable values of CaO (25-50 wt %); which decline substantially during the OAE-2 interval as SiO₂ values increase with maximum values of 47 wt % and 23 wt % recorded for each site respectively (see supplemental datafile). At Site 1260 in the lower part of the studied interval (445.19 and 462.7 meters composite depth; m.c.d) high concentrations of redox sensitive trace metals are recorded that are significantly enriched compared to average shale (see supplemental datafile; **Figure 9**). Concentrations and enrichments of the redox sensitive trace metals decline from these high values around ~443 m.c.d and reach minima during part of the OAE-2 CIE interval (426-423 m.c.d), but still

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remain enriched compared to average shale ($U_{EF} = 10$; $M_{OEF} = 25$ -; $V_{EF} = 4$). This same trend is mirrored in the sedimentary record from Site 1261; with minima enrichment values recorded during the OAE-2 CIE interval (albeit in lower resolution) that even though are relatively low compared with the pre-OAE-2 interval are still enriched compared to average shale (minima values: $U_{EF} = 12$; $M_{OEF} = 46$; $V_{EF} = 5$; Figure 10).

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3.3. Palynology

Detailed palynological results and data tables are presented in the supplemental dataset and displayed in **Figures** 4-11.

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4. Discussion

4.1. Principal Component Analyses (PCA)

In order to provide additional understanding into the main controls on organic matter composition and paleoenvironmental significance, PCA was <u>initially</u> conducted on the palynological and geochemical datasets separately and then on the integrated palynological and geochemical dataset. All three analyses showed the similar principal components and clusters and in support of the discussion only the PCA results run on the combined dataset are presented. The combined dataset PCA was run on cores that had multiple detailed analyses from the same core sample/depth, -including Innes-1, Iona-1, well 'X', USGS Portland-1 and ODP Sites 1260 and 1261 using the statistical software C2 (http://www.staff.ncl.ac.uk/stephen.juggins/software/C2Home.htm). Variables within the dataset include palynology, major and trace elements, δ¹³C_{org} and TOC. The PCA results show a clear grouping of samples along the two primary axes (Figure 12).

Eight groups were identified and are lithostratigraphically defined: Group I = lower Eagle Ford; Group II =

upper Eagle Ford; Group III = Buda Limestone and Austin Chalk; Group IV = Bridge Creek Limestone; Group

V= Hartland Shale Mb.; Group VI = Graneros Shale; Group VII = Dakota Sandstone and Group VIII = Demerara

Rise. The groups are discussed in a chronostratigraphic context in Section 4.2. Along the principle axis/eigen

score 1, samples with high negative scores comprise high CaO contents and are associated with the Buda

Limestone and Austin Chalk formations_samples, whereas high positive scores correspond with high values of

elements enriched in heavy minerals (Zirconium-Hafnium; Zr-Hf), silicates (quartz, feldspar),

phyllosilicates/clay minerals (Gallium-Potassium Oxide-Rubidium-Titanium; Ga-K2O-Rb-Ti [eg., illite, biotite,

smectite, kaolinite], and increased terrigenous contributions (T:M ratio) locally corresponding with the clastics

of the Dakota Sandstone Gr. The high positive score along axis 1 may not solely represent terrestrial/detrital

riverine dilution, but may also reflect diagenetic alteration of the abundant volcanic ash from atmospheric fallout

of western Cordillera plinian eruptions (SiO2, Al2O3, TiO2, heavy minerals) that were transformed into smectite—

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chapter 3.3: at least present the most important features of the palyno-dataset that are used in the following discussion so that the reader has not to go back to the supplements every time.

"I think it is better to keep the palynological results in the supplemental text, and to keep your main text concise. I will mention this to the reviewer before his final check of your ms. So leave the structure as it is" editorial comment

illite-Fe-Ti oxides (see Eldrett et al. 2015b).elays (Potassium Oxide- Rubidium; K2O-Rb [illite/k-feldspar], Gallium [Ga) Al2O3 [kaolinite]) and increased terrigenous contributions (T:M ratio) corresponding with the elastics of the Dakota Sandstone Gr. The high positive score along axis 1 may not solely represent terrestrial/detrital riverine dilution, but also reflect diagenetic alteration of the volcanic ash (SiO2-and heavy minerals) into smectite-illite (see Eldrett et al. 2015b). Therefore, the clustering of the environmental variables along the Axis/Eigen score 1 is interpreted as representing the carbonate - non carbonate/volcaniclastic mixing trend

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Axis/Eigen score 2 is interpreted to represent a restricted/eutrophic/anoxic to open marine/oligotrophic/oxic

marine trend influencing both surface and deep-waters/the water column and/or sediment-water interface; with high positive scores associated with high values in TOC, redox sensitive trace metal concentrations and associated with preservation of amorphous organic matter (AOM)AOM and assemblages dominated by deprasinophyte algae phycomata, the latter are —indicative of eutrophic and stratified water column conditions (cf. Prauss, 2007)

It is also noted that high eigen scores are associated with dinocyst assemblages dominated by peridinioid (P-) cysts and particularly taxa comparable to those informally described from Tarfaya, Morocco by Prauss (2012a, b); Bosedinia cf. sp 1 & 3 of Prauss (2012b) (photographic illustrations are provided in the supplemental information). Sporadically common/abundant occurrences of Bosedinia sp. 1 & sp. 3 were recorded in the Cenomanian and Turonian of Tarfaya by Prauss (2012a, b), where it is often associated with common occurrences of the colonial green alga Botryococcus; high concentrations of Botryococcus had previously only been reported from fresh/brackish-water paleoenv+ironnomnements (e.g. Zippi, 1988) and this led Prauss to infer a freshwater affinity for the Bosedinia, suggesting it probably represents episodic salinity stratification in the marine paleoenvironment at Tarfaya (Prauss, 2012c). However, in all the localities investigated in this study and also Lozier Canyon (Dodsworth, 2016), abundances of Bosedinia cf. sp 1 & 3 are not associated with an increase in Botryococcus or terrigenous pollen and spores, or with the presence of fresh-water algal genera such as Concentricystes, Ovoidites (Schizophacus), Pediastrum and Schizosporis. The latter genera are common components at Cenomanian - Turonian continental to fluvial-deltaic settings in Utah (Akyuz et al., 2016) and east Texas (Dodsworth, 2016). In the present study, the persistent occurrence of these freshwater algal genera are restricted to the Dakota Sandstone in the Portland-1 core where they are associated with the only frequent occurrences of Botryococcus and super-abundant (>50%) occurrences of terrigenous spores and pollen reported here. Influxes of Bosedinia do not co-occur in any of these proximal KWIS settings. The PCA results presented in this study indicates that Bosedinia cf. sp 1 & 3 plots positively along eigen score 2, associated with indicators for eutrophic and anoxic marine conditions; potentially representing the enhanced nitrite/nitrate availability in the photic zone (see Dodsworth, 2016). During the Cenomanian Epoch, abundances of Bosedinia cf. sp 1 & 3 are confined to middle-lower latitudes (Prauss 2012a, b; this study), being absent from higher latitude records (e.g. Foucher, 1980; Jarvis et al. 1988, 2011; Fitzpatrick, 1995; Pearce et al. 2003, 2009; Lignum 2009), and is thus interpreted to have a tethyan equatorial Central Atlantic affinity (see supplemental information).-

Commented [11]:

Anonymous Referee #1. 10

"Gallium (Ga)-Al2O3 [kaolinite]" I don't understand the direct link between Al2O3 and Kaolinite. Illite also contains Al2O3.

Gallium is generally associated with Al2O3 associated with detrital clays and agree that the direct link with kaolinite is difficult to establish.

Commented [12]:

Anonymous Referee #1. 11 redox sensitive trace metal concentrations Â'z. Zn and Ni are also paleoproductivity proxies

Reply:

Although we agree with the reviewer in part, the main paleo-productivity elemental proxies are Barium and Phosphorus which have only one oxidation state. Zn, Cu, Ni are redox sensitive elements that are thought to be delivered to the sediment-water interface through the sinking of organic matter and could be used as proxies of organic matter flux. However, solubility and oxidation state of these elements are still a function of redox state and thus not solely an organic matter flux proxy. The delivery mechanism and the calibration to surface water productivity is debated, so due to these uncertainties we have not split the element preferences from the primary category of redox sensitive. We retain the use of redox sensitive as it encompasses the main elements discussed in the paper (i.e. Mo, V, U).

The high negative eigen scores along axis 2 correspond with proxies associated with open marine oligotrophic conditions such as gonyaulacoid dinocysts (G-cysts) including *Spiniferites* spp., and *Pterodinium* spp.; the latter shows affinity with modern-day *Impagadinium* spp. which is found in oceanic and open marine conditions (Wall et al. 1977; Zonneveld et al. 2013). High negative eigen scores are also associated with proxies indicative of more oxygenated water conditions such as higher concentration of the redox sensitive trace metal Manganese Oxide (MnO) and foraminiferal test linings. The negative eigen scores are therefore interpreted as representing an open marine, oxygenated and oligotrophic depositional environment. In addition, it is interesting to note that although phytoclasts plot negatively along eigen axis 2 and may represent a reduced masking effect of AOM during oxygenated conditions (Tyson, 1995); they also plot positively along Eigen score 1 (non-carbonate/volcaniclastic trend) alongside freshwater algae and Areoligeracean dinocysts; the latter are more suggestive of a more nearshore environment (see Brinkhuis and Zachariasse 1988; Harker et al. 1990; Li and

carbonate/volcaniclastic trend) alongside freshwater algae and Areoligeracean dinocysts; the latter are more suggestive of a more nearshore environment (see Brinkhuis and Zachariasse 1988; Harker et al. 1990; Li and Habib 1996). In addition, a component of this trend may also represent recycling and transportation of nearshore palynomorphs to relativelymore distal environments. The eigen scores are plotted against depth for each of the sites in (Figures 4-11-6, 8-10) to further support the paleoenvironmental interpretation as discussed below.

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4.2. Paleoenvironmental Interpretation

4.2.1. Southern KWIS

The Buda Limestone (early Cenomanian; ca 98-97.5 Ma) comprises highly diverse and low abundance dinocyst assemblages (see also Cornell, 1997) indicative of open marine oligotrophic conditions. This interpretation is supported by the i) PCA results reflecting the lack of enrichment in redox sensitive trace metals, low TOC and poor preservation of AOM; dominance of G-cysts and ii) sedimentological and micropaleontological evidence for abundant and diverse benthos indicative of a healthy carbonate factory. The Buda Limestone dinocyst assemblages are comparable to those reported from marine limestone facies of early Cenomanian age in western Europe, including England (e.g. Cookson and Hughes, 1964) and France (e.g. Foucher, 1980).

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The overlying lower Eagle Ford Gr. interval (ca. 97.2-94.9 Ma) is generally characterized by a decline in dinocyst species diversity, with the palynological assemblages comprising high absolute abundance of prasinophyte phycomata with P-cysts being the major component of the dinocyst community indicative of eutrophic and stratified water column conditions (cf. Prauss, 2007; Sluijs et al. 2005). This interpretation is supported by PCA results reflecting the enrichment and co-variance in redox sensitive trace metals and high TOC values combined with low, but sporadic occurrences of benthic foraminifera indicative of restricted and suboxic-anoxic depositional conditions (see also Eldrett et al. 2014). Furthermore, the presence of aryl isoprenoids in the lower Eagle Ford Gr. section from Iona-1 has been demonstrated as originating from *Chlorobi* (green sulphur bacteria) and thus evidence for at least temporary and/or partial photic zone euxinia at this time (Sun et al. 2016). Bed- scale variations are also identified in redox conditions recording greater water-mass ventilation and current activity during the deposition of limestone beds compared to deposition of marlstone beds due to combined obliquity and precessional forcing on solar insolation (Eldrett et al. 2015b). Comparable

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Anonymous Referee #1. 13

"In addition, it is interesting to note that although phytoclasts plot negatively along eigen axis 2 and may represent a reduced masking effect of AOM during oxygenated conditions (Tyson, 1995); they also plot positively along Eigen score 1 (noncarbonate/volcaniclastic trend) alongside freshwater algae and Areoligeracean dinocysts suggestive of a more nearshore environment (Brinkhuis and Zachariasse 1988; Harker et al. 1990; Li and Habib 1996)". This last interpretation must be confirmed by the calculation of a correlation coefficient which, in my opinion, will not show a correlation. Here, you do only a suggestion but not a real interpretation because the position of the phytoclasts is not at all correlated to the axis 1

Text amended so clarified and response detailed

Commented [14]:

Anonymous Referee #1. 14 Anonymous Referee #2. 7 c p. 9 line 25 change "Figures 4-11" by Figures 4-6, 8-10

Done

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Anonymous Referee #1. 15

c p.9 "Paleoenvironment Interpretation" Use the same chronostratigraphic subdivision as geochemistry section (3.2); If the sequence stratigraphy interpretation was not published before it's important to explain it. Reply:

Chronostratigraphic age assignment for each of the main lithostratigraphic units presented and discussed in the text is provided in parentheses and we believe this as the optimal structure of the text. The sequence stratigraphic interpretation has not been previously published and is now included in the supplemental information.

palynological assemblages have not been reported from the middle to upper Cenomanian deposits in Europe but high relative abundances of prasinophyte phycomata and the peridinioid genus *Bosedinia* have been documented further south in the Cenomanian and Turonian organic-rich shale facies at Tarfaya (Prauss 2012a, 2012b) in north Africa (Prauss 2012a, 2012b).

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Within the middle part of the lower Eagle Ford Gr. (~96.1-95.4 Ma) the relative abundance contribution of prasinophyte phycomata to the palynological assemblage decreases as the absolute abundance of the P-cyst *Bosedinia cf.* sp 1 & 3 significantly increases. This shift in the palynological assemblage is recorded in all the studied sections including the San Marcos Arch outcrop; also in the nearby Lozier Canyon outcrop section (Dodsworth, 2016). As discussed in **section 4.1**, the abundance of *Bosedinia* sp. 1 & sp. 3 had previously been interpreted as reflecting the occurrence of freshwater/brackish water conditions in the photic zone at Tarfaya and episodic salinity stratification there (Prauss 2012a, 2012b, 2012c), but in the absence of freshwater algae and the combined PCA results for the studied sections this interpretation is not supported here for the KWIS or Demerara Rise. Alternatively, Dodsworth (2016) proposed that the introduction of waters from respectively deeper denitrification zones into photic zone by vertical expansion of the oxygen-minimum zone or by upwelling may be a controlling factor, with reduced nitrogen/ammonium favouring prasinophyte algal production (cf. Prauss 2007) and availability of nitrite/nitrate promoting P-cyst (e.g. *Bosedinia* sp. 1 & sp. 3) productivity in the surface waters. In order to resolve the contribution and variation in the nitrogen cycle and its impact on community structure of primary producers stable nitrogen isotope data are currently being acquired and it is the authors' intention to publish this data when available.

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The upper Eagle Ford Gr. associated with first peak ("A") of the OAE-2 CIE (~94.65 Ma) is characterized by a sharp change in palynological assemblages with increase abundance of G-cysts, in particular open marine forms including Pterodinium, Spiniferites ramosus and Nematopshaeropsis spp. Prasinophyte phycomata become rare and overall dinocyst diversity increases. This assemblage shifts along with the reduction in redox sensitive trace metal enrichments, i lowered TOC; increased bioturbation and occurrence of benthic foraminifera indicate deposition within an open marine meso-oligotrophic and oxygenated depositional environment (also see Eldrett et al. 2014). By contrast, in some European depositional basins, e.g. eastern England, northern Germany and Crimea, uppermost Cenomanian to lower Turonian deposits associated with OAE-2 are characterized by intervals of interbedded organic-rich shales-mudrocks with limestones, the formerthat contain isolated influxes of prasinophyte phycomata and higher numbers of P-cysts than under- and overlying formations (Marshall and Batten, 1988; Dodsworth, 1996; 2004; Prauss, 2006); whereas limestone deposition, lean in organic matter, is continuous in other areas, e.g. southern England - northern France. The relative increase in the T:M ratio during OAE-2 in the studied sections presented here likely reflects the closed-sum effect as absolute abundance of marine palynomorphs decreases during the upper Eagle Ford (transition from eutrophic to meso-oligotrophic conditions). Absolute abundance of terrestrial palynomorphs does not increase significantly, although this may partly reflect greater distance from shoreline during the Cenomanian-Turonian transgression and potentially diluted concentration of all palynomorphs including pollen during increased biogenic carbonate productivity

Commented [16]:

Anonymous Referee #2. 8

lines 29-31 on page 10: would delete this statement from the text.

R8. We have deleted this comment

associated with the limestones of the upper Eagle Ford – Lower Bridge Creek. An increase in absolute abundance of terrestrial palynomorphs is recorded at the Pueblo GSSP, in uppermost Cenomanian beds 82-lower 85 (2000-5000 counts per gram; 30-60%; Dodsworth, 2000). It is unclear whether palynological assemblages in the studied sections support increased hydrological cycle during OAE-2 (see Van Helmond et al. 2014). The sporomorph assemblages during OAE-2 mainly record a relative increase in gymnosperms, in particular during the PCE interval, and thus any increase in T:M ratio may reflect transition from mega-thermal to meso-thermal vegetation (perhaps also reflecting increased pollen production by wind dispersed gymnosperms) in response to climate cooling episode (see Forster et al. 2007; Jarvis et al. 2011) rather than increased hydrologic cycle. The recorded increase in Ti/Al in the PCE interval may reflect increased fluvial and/or eolian inputs, however the association of Ti/Al with trace metal enrichments in Cobalt, Chromium, Scandium as well an increase in heavy-to-light rare earth elements and a positive europium anomaly, together are indicative of a hydrothermal or mafic influence and suggest emplacement and weathering of a LIP during the PCE interval (see Eldrett et al. 2014). In addition, the Ti/Al record may also reflect alteration of biotite from the ubiquitous felsic volcanic ash beds rather than solely eolian versus fluvial inputs, and the links with gymnosperm abundances could have resulted from changes in climate and/or oceanographic conditions resulting from these large scale and regional igneous events.

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The PCA results indicate occasional dysoxia-anoxia within the upper Eagle Ford Gr. interval; which becomes more persistent in the upper-part of the Langtry Mb. as indicated by slight enrichment in redox sensitive trace metals and palynological assemblages recording reduced dinocyst diversity and enhanced abundances of prasinophyte phycomata. This interpretation seems to be in contrast to the sedimentological evidence that preserves bioturbated marlstones and nodular limestones along with shell hash horizons forming symmetric ripples and abundant in situ macrofauna (i.e. echinioids); all indicative of a high energy dynamic environment (Minisini et al. in review). It may be the case that short-lived storm and oxic events are not resolved at our sampling resolution as bulk geochemical and palynological data intergrates longer time periods; or alternatively we resolve short-lived periods of dysoxia-anoxia that are subsequently smeared and re-distributed by bioturbation within a background environment characterized by high energy and well-oxygenated conditions. It should be noted that Ssimilar abundance increases in prasinophyte phycomata are recorded in the upper part of the South Bosque Formation at the Bouldin Creek outcrop and recorded in the Lozier Canyon outcrop section (Dodsworth, 2016). The occurrences of prasinophyte phycomata in the Lozier Canyon outcrop were thought to represent an artiefact of their preferential preservation in weathered material (Dodsworth, 2016), but-however. compared to the regional trends identified in well-preserved core alongside trace metal enrichments suggests a (and even in the upper part of the South Bosque Formation at the Bouldin Creek outcrop) indicates that this trend reflects a genuine transition to more restricted and at least partially and/or episodically dysoxic-anoxic depositional environment. The limestones of the Austin Chalk- record a return to open marine and persistently oxygenated conditions supported by the PCA results reflecting very diverse dinocyst assemblages, comprised mainly G-cysts with abundant foraminiferal test linings, -reduced AOM and no enrichments in redox sensitive trace metals.

Commented [17]:

Anonymous Referee #2. 9

lines 14-15 on page 11: since there is no increase in pollen and spores, there is no support for an increased hydrological cycle

R9. In the sections from Texas and Demerara Rise there is not an increase in absolute abundance of pollen spores. However; in the Portland-1 core there is a slight increase (2,000-5,000 c.p.g) so we disagree with the reviewers observation and a discussion is warranted. Regardless of absolute abundance, our data show a pollen assemblage shift to gymnosperm dominance; something that is highly relevant when comparing with other palynological records discussing increased hydrological cycle (e.g. Van Helmond et al. 2014) and subsequent citations.

Commented [18]:

line 19 on same page: what is the indication for climate cooling? Only the PCE is cooler, under background values that are much warmer than before or after OAE2!

References added; also added TEX86 to Figure 13

Commented [19]:

Anonymous Referee #1. 16

Ti/Al can be used as a proxy for eolian versus fluvial input. What does this proxy show?

The Ti/Al ratio shows an increase during the Plenus Marl event corresponding with recorded increase in gymnosperms. However, during this interval there is also an increase in maffic trace metals, REE and recorded Eu anomaly interpreted as reflecting a maffic source, possible High Arctic Large Igneous Province (see Eldrett et al. 2014; and supplemental figures). The increase in Ti/Al may also contain a component or reflect the emplacement and weathering of a LIP rather than solely eolian versus fluvial input. Due to the uncertainty we have excluded this possible proxy in this publication.

4.2.2. Regional water-mass evolution

In this study we infer three main water-masses-properties: i) eEquatorial-Atlantic tethyan source; ii) a northern 5 boreal source and iii) a more local central KWIS source. The eEquatorial-Atlantic sourced water is primarily based on the occurrence of dinocysts and other microfauna (i.e. calcareous nannofossils and foraminifera) that are geographically restricted to low latitudes and specifically Eequatorial Atlantic with inferred tethyan affinities (e.g. Bosedinia cf. sp 1 & 3; see discussion below). The surface waters of this watermass is characterized by low diversity dinocyst assemblages dominated by P-cysts and prasinophyte phycomata indicative of stratified and 10 hydrographically restricted and eutrophic conditions (this study); whilst the correspondence with the presence of isorenieratene derivatives (i.e. Sinninghe Damsté and Köster, 1998; Kuypers et al. 2002; Kolonic et al. 2005; Van Bentum et al, 2009; Sun et al. 2016) demonstrate at least temporary and/or partial photic zone euxinia. The underlying bottom-water; or at least sediment-water interface was predominantly dysoxic-anoxic as evidenced by the enrichment in redox sensitive trace metals and relatively sparse benthic foraminifera (i.e. Hetzel et a. 15 2009; Eldrett et al., 2014). The northerly sourced watermass is constrained by the occurrence of dinocysts with a boreal affinity (see Eldrett et al. 2014; Van Helmond et al. 2014; 2016); combined with the increased planktonic diversity and dominance of G-cysts that are associated with more oligotrophic/hydrographically unrestricted conditions. The absence of prasinophyte algae point towards a mixed water-column (cf. Prauss, 2007); and the overall low values in redox sensitive elements and increased abundance and diversity of benthic fauna along 20 with enhanced bioturbation indices (e.g. Meyers et al. 2007; Eldrett et al. 2014) indicate frequent ventilation of the bottom-waters and/or sediment-water interface. A more local and partially restricted dysoxic watermass $\underline{sourced\ by\ the\ central\ KWIS\ is\ interpreted\ based\ on\ the\ occurrence\ of\ mixed\ moderate\ diversity\ dinocyst}$ assemblages, including taxa more typical of the northern and central KWIS, with rare tethyan components combined with limited enrichment in redox sensitive elements and abundance of low diversity agglutinated 25 benthic foraminfera assemblages (see discussion below). It should be noted that within these regional watermass regimes; higher frequency variations in redox state are documented; in part a response to obliquity and precession forcing on the latitudinal distribution of solar insolation (see Eldrett et al. 2015b). a restricted suboxic-anoxic marine water-mass characterized by low diversity dinocyst assemblages interpreted to represent a tethyan source; ii) an unrestricted/open marine oxygenated water mass characterized by high diversity dinocyst 30 assemblages interpreted to represent a boreal source and iii) a partially restricted dysoxic water mass interpreted to represent a more local central KWIS source. Comparing the paleoenvironmental trends recorded in the Cenomanian - Coniacian sections from SW Texas to those recorded further north within the central KWIS (USGS Portland-1; Pueblo GSSP outcrop) and to the south in the equatorial North Atlantic (ODP sites 1260, 1261, Demerara Rise) allows the reconstruction of their water-mass evolution allows an attempt at 35 reconstructing the evolution of these various water masses (Figure 13-14). A discussion of dinocyst and pollen paleo-latitudinal provinicialism across these regions during Cenomanian and Turonian times is given in the supplemental information.

Commented [20]:

Anonymous Referee #1, 17

It's too direct, you have to explain!! Why do you talk about a tethyan source and not an Atlantic-tethyan source? You specify Â'n a restricted suboxic-anoxic marine watermass of tethyan source" before and after CIE. However Tethyan marine waters are generally well-oxygenated during these time intervals which are not the case of the Atlantic marine waters (see ref Monteiro et al., 2012 Paleoceanography for example: : :)". It would be better to write water mass of Atlantic-Tethyan source. Section organization: use the same chronostratigraphic subdivision than previously.

As suggested we have amended the text and specifically refer to an equatorial-Atlantic tethyan source

Eldrett, James S GSNL-PTI/EG

Anonymous Referee #2, 11

11) Chapter 4.2.2: why should the tethyan water mass be suboxic-anoxic? This is inferred by the authors at the beginning of this chapter and then used in the following interpretation but it is never shown convincingly to the reader that this is the case. What is the independent evidence for this? Same holds true for the boreal water mass. Are there any other indications other than own data and interpretations? If yes, present them in detail. So far, the main problem with this chapter is that there is no prove that the suggested water masses existed and are characterized by the suggested data in the way they are presented here.

ammended

Commented [21]:

Anonymous Referee #2. 11

11) Chapter 4.2.2: why should the tethyan water mass be suboxic-anoxic? This is inferred by the authors at the beginning of this chapter and then used in the following interpretation but it is never shown convincingly to the reader that this is the case. What is the independent evidence for this? Same holds true for the boreal water mass. Are there any other indications other than own data and interpretations? If ves. present them in detail. So far, the main problem with this chapter is that there is no prove that the suggested water masses existed [

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Anonymous Referee #1. 17

c P.11 "Regional water-mass evolution"; "In this study we infer three main water-mass properties: i) a restricted suboxic-anoxic marine water-mass characterized by low diversity dinocyst assemblages interpreted to represent a tethyan source; ii) an unrestricted/open marine oxygenated water-mass characterized by high diversity dinocyst assemblages interpreted to represent a boreal source and iii) a

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Lower Cenomanian sediments from the e-Equatorial Atlantic (ODP Site 1260) are interpreted as being deposited in a stratified suboxic-anoxic marine environment as indicated by enrichment in redox sensitive trace metals; low diversity dinocyst assemblages with abundance of prasinophyte phycomata; preservation of laminated organic rich mudrocks deposition and positive PCA-2 scores. This interpretation is consistent with a southern 5 tethyan water mass and a circulation controlled nutrient trap fuelling surface water productivity and anoxic depositional environment-as proposed by (Jiménez Berrocoso et al. (2010) and; Trabucho-Alexandre et al. (7 2010). Further to the North in the southern part of the KWIS, the early Cenomanian is characterized by the deposition Buda Limestone, which is primarily characterized by well oxygenated conditions. The first incursion of an equatorial-Atlantic tethyan water-mass into southern Texas occurred at 97.2 Ma (t1; Figure 14). 10 and is marked by the deposition of hummocky cross stratified limestones and mass transport deposits interbedded with the organic rich sediments that characterize the lowermost -Eagle Ford Gr., above the top Buda limestone submarine unconformity (see Minisini et al. in review). In the Central KWIS (Portland-1 core), the early Cenomanian is characterized by relatively dysoxic depositional conditions, with frequent to common prasinophyte phycomata and mixed dinocyst assemblages, including taxa more typical of higher paleolatitudes of 15 the northern and central KWIS such as Senoniasphaera microreticulata and Palaeoperidinium cretaceum, and the first consistent but mainly rare occurrence of Bosedinia cf. sp 1 & 3 which is common in coeval deposits at Demerara Rise; along with low diversity agglutinated benthic foraminfera (Figure 8) and the occasional occurrence of rare tethyan calcareous planktonic taxa, together suggestive of a mainly Western Interior Seaway source (Eicher and Diner, 1985). that are not recorded further south in the coeval Buda Limestone such as 20 Senoniasphaera microreticulata and Palaeoperidinium cretaceum, along withand high abundance of agglutinated benthic foraminfera (Figure 8) indicative of a Western Interior Seaway source. The initial northward expression of an equatorial-Atlantic tethyan water-mass is only evidenced in the central KWIS (Portland-1 core) during the deposition of the Thatcher Limestone Mb. (ca 96.5-96.6 Ma), with slight increases in prasinophte phycomata, decrease in dinocyst diversity, reduced benthic foraminfera and supported by the 25 occurrence of ammonites, calcareous nannofossils and an increase in planktonicie foraminifera with tethyan affinities (see Eicher and Diner, 1985; Cobban, 1993; Hancock et al., 1993, Bralower and Bergen, 1998). Watermass characteristics fluctuate during the middle Cenomanian in the Portland-1 core, but are interpreted as being predominately sourced locally from the Western Interior Seaway as evidenced by mixed dinocyst assemblages; relatively low enrichments in redox sensitive trace metals and dominant occurrence of agglutinated foraminifera. 30 This study is limited by the relatively low sampling resolution from the MCE interval; in order to better resolve the detail and timing of these paleoceanographic variations higher resolution multi-proxy analyses are required. In the Portland-1 core there is a clear shift from agglutinated to calcareous benthic foraminfera near the top of the MCE interval; which combined with the increased abundances of calcareous planktonic foraminifera (e.g. Rotalipora cushmani) and dinocysts (e.g. Bosedinia cf. sp. 1 & 3) with tethyan affinities interval is suggestive of 35 an equatorial-Atlantic tethyan influence (Figure 8; for detailed discussion of tethyan-boreal foraminiferal distribution within Colorado, see Eicher and Diner, 1989). However However, it is only during the deposition of the middle part of the Lincoln Shale (~95.6 Ma) that abundant prasinophyte phycomata, increased trace metal enrichments and reduced dinocyst diversity point towards a persistent influence of an equatorial-Atlantic tethyan water-mass. These proxies reach maxima along with the abundance of Bosedinia cf. sp. 1 & 3 during the deposition of the Hartland Shale Mb. (~95 Ma; t2: Figure 14) indicating the marine transgression of equatorial-

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Anonymous Referee #1, 18

c p. 12 lines 8-12 "Lower Cenomanian sediments from the Equatorial Atlantic (ODP Site 1260) are interpreted as being deposited in a stratified suboxic-anoxic marine environment as indicated by laminated organic rich mudrock deposition and positive PCA-2 scores. This interpretation is consistent with a southern tethyan water-mass and a circulation controlled nutrient trap fuelling surface water productivity and anoxic depositional environment (Jiménez Berrocoso et al. 2010; Trabucho-Alexandre, 2010)". I don't understand why it's consistent with a southern tethyan water-mass. According Trabucho-Alexandre et al., 2010, this zone is the seat of upwelling of deep waters coming from the Pacific, no southern tethyan water-mass is mentioned by these authors.

We agree with the reviewer. Our findings are consistent with the depositional conditons presented by Trabucho-Alexandre, 2010 and Jiménez Berrocoso et al. 2010

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Anonymous Referee #1. 19 c p.12 line 17 : "with mixed dinocyst assemblages". List of the genera

We have amended the text

Eldrett, James S GSNL-PTI/EG why is this indicative for an WIS source? Explain and justify in detail.

done

Commented [25]:

Anonymous Referee #2. 13

line 31 on same page: at this point in the succession, there are no benthic forams, so the statement above in lines 28-29 cannot be valid!

We disagree with the observation of the reviewer; there are foraminifera test linings (including for outside the initial 100 count specimens) in almost all samples throughout the Graneros to Hartland (with the exceptions of the 160.07m and 185.53m samples [column GQ in the Portland-1 datafile]).

Commented [26]:

Anonymous Referee #1. 20

c p.12, line 28 "In the Portland-1 core there is a clear shift from agglutinated to calcareous benthic foraminifera near the top of the MCE interval suggestive of a tethyan influence" why? Is there no agglutinated and calcareous benthic foraminifer in shallow water environment in the Atlantic and Pacific oceans?

Reply: We agree with the comment of the reviewer and have clarified this sentence. The transition from agglutinated to calcareous foraminifera had been

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Atlantic tethyan waters into the KWIS and mirrors the 3rd order eustatic Greenhorn Cycle (Kauffman, 1977) reaching the maximum flooding at ~94.7 Ma (see supplemental information). The long term (3rd order) trends in water-mass evolution reported here are therefore likely driven by variations in global eustasy related to regional tectonic and/or mantle plume-lithosphere dynamics associated with the emplacement of LIPs (i.e. High Arctic; Caribbean) during this Greenhouse period lacking polar continental ice-sheets.

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During peak transgression (~94.7 Ma, t3; Figure 14) a significant oceanographic re-organization was marked initially by increased bottom-water current activity resulting in widespread hiatal surfaces across the KWIS and with the rapid onset of more oligotrophic, oxygenated and open marine conditions. These trends are evidenced by increased diversity of dinocyst assemblages dominated by G-cysts; reduction in trace metals abundances, reduced TOC and isorenieratene derivatives (see Sun et al, 2016) and as well as increased bioturbation and diverse /benthic foraminfera abundances cumulating in the development of the Benthonic/Benthic Oxic Zone (Eicher and Worstell, 1970; Keller and Pardo, 2004). This environmental shift is interpreted to reflect the rapid southward incursion of a northerly sourced water-mass into the central and southern KWIS and is supported by the equatorial migration of boreal dinocyst taxa (e.g. Cyclonephelium compactum-membraniphorum morphological plexus; Eldrett et al. 2014; Van Helmond et al. 2016), occurring during a period of climate cooling with a minimaminima (3-5°C cooling) during the PCE (Jarvis et al. 2011; Van Helmond et al. 2014; 2016; Elderbak and Leckie, 2016). Therefore, the increase in abundance and diversity of fforaminiferal/molluscan and ammonite assemblages in the KWIS (McNeil and Caldwell, 1981; Kauffman, 1984, 1985; Eicher and Diner, 1985; Elder, 1985; Leckie et al., 1998; Caldwell et al., 1993; Kauffman and Caldwell, 1993, Elderbak and Leckie, 2016) and the lithologic transition from organic-rich mudrocks to limestone dominated facies (Corbett et al. 2014; Lowery et al. 2014) in the latest Cenomanian does not reflect the incursion of tethyan water, but instead the southward flow of a boreal water-mass at this time. Comparable dinocyst assemblages including diversified G-cysts are recorded i) in the Canadian KWIS throughout the Cenomanian and Turonian (e.g. Singh, 1983; Bloch et al. 1999), albeit with higher numbers of boreal P-cyst taxa including Isabelidinium magnum and Eurydinium glomeratum, and ii) coeval sediments from European shelves f (e.g. Foucher, 1980; Jarvis et al. 1988, 2011; Fitzpatrick, 1995; Pearce et al. 2003, 2009; Lignum 2009) which also correspond with the influx of boreal macro-fauna (Jefferies, 1962, 1963; Gale and Christensen, 1996; Voigt et al., 2006; Jarvis et al., 2011). Evidence for the southward incursion of a northerly sourced water-mass during the latest Cenomanian is recorded in all the studied sections spanning the central and eastern part of the KWIS and therefore does not support a more complex oceanographic system such as that modelled by Slingerland et al. (1996) and Kump and Slingerland (1999). The data presented here indicates that the initial inflow of equatorial-Atlantic tethyan water across the southern gateway is completely replaced by the almost simultaneous southward flow of boreal water (see also Van Helmond et al. 2016), with no evidence for a contemporaneously counterflowing tethyan water-mass along the eastern margin. However, based on the limited geographic extent of the studied sections these findings are considered tentative, with additional palynological and geochemical investigations on the eastern margin of the KWIS being required to constrain the lateral variability in possible water-mass properties.

Although the migration of boreal dinocyst taxa (namely the Cyclonephelium compactum-membraniphorum morphological plexus; van Helmond et al. 2016) during the PCE is not recorded further to the south in the equatorial North Atlantic; a similar shift in palynological assemblages, including a marked decline in prasinophyte algae, is recorded in the shallower shelf settings on Demerara Rise (ODP Site 1261). Furthermore, at ODP Site 1261, the recorded shift towards a more diverse and open-marine dinocyst assemblage is also associated with an increase in the abundance of organic foraminiferal test linings; re-population by calcareous benthic foraminifera (Friedrich et al., 2006; 2011), which combined with a reduction in redox sensitive trace metals is may indicate ive of an improvement in environmental conditions and a reduction on the oxygen minimum zone. However, it should be noted that organic matter concentration remains high. This shift in environmental conditions also corresponds with a sea surface temperature minimum in the lower part of the OAE-2 CIE (up to 4°C; Forster et al. 2007) on the flank of Demerara Rise (ODP Site 1260) and perhaps marks the southernmost influence of a boreal water-mass. However, in Site 1260 Bosedinia cf. sp.1 & 3 remains abundant despite the decline in both prasinophyte algae, redox trace metals and the associated increase in benthic foraminifera indicating more complex interaction between water-masses and the oxygen minimum zone along the slope. The southern expression of this boreal influence is therefore limited in duration and extent, mostly affecting shallower water settings. These findings are consistent with Neodymium (Nd) isotope data from Demerara Rise indicative of circulation pattern change in the Atlantic-Tethys at this time (Zheng et al., 2016; Martin et al 2012).

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20 The early Turonian to Coniacian (~94 Ma - 89 Ma, t4-t5: Figure 14) interval iswas characterized by two further proposed incursions of both boreal and equatorial-Atlantic tethyan waters into the KWIS. The first Turonian incursion of boreal water into the southern KWIS (~92.5 Ma) was marked by increased bottom water currents and a 125-210 kyr hiatal interval in Iona-1 (Eldrett et al. 2015a, b). This hiatal interval correlates with that of much greater duration in shallower shelf setting of Innes-1 (> 2 Mmyrs) and is coeval with the regional middlelower Turonian hiatus identified throughout the KWIS (e.g. Ewing, 2013). In the central KWIS, at the Pueblo GSSP, the upper part of the lower Turonian and lower part of the middle Turonian substages contain a marked increase in the boreal P-cyst taxa Isabelidinium magnum and Eurydinium glomeratum. The most notable incursion of equatorial-Atlantic tethyan water in Texas in the Turonian corresponds with the Langtry Mb. of the Eagle Ford Gr. and is characterized by greater abundance of prasinophyte phycomata; greater dinocyst P: G 30 ratio; reduced dinocyst diversity; enrichments in redox sensitive trace metals and enhanced preservation of TOC: all indicative of increased surface water organic matter production and preservation due to partially or episodically anoxic bottom-water or sediment-water conditions. The second main incursion of boreal water into Texas occurs in the Turonian-Coniacian and is marked by the development of chalk facies (e.g. Austin Chalk), diverse dinocyst assemblages and a reduction in redox sensitive trace metals and TOC indicative of oxygenated 35 and oligotrophic marine conditions. Although, the transition from the Eagle Ford Gr., to Austin Chalk is conformable in both Iona-1 and Innes-1, an unconformity is identified on the San Marcos Arch (Lowery et al., 2014; this study). With the exception of the Buda-Eagle Ford unconformity that marks the initial flooding of the southern KWIS by tethyan waters; Therefore, the other major unconformitieshiatal surfaces identified throughout the KWIS also-appear to be associated with increased bottom-water current activity linked to

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Anonymous Referee #1. 21

c p.13, lines 35-39. "Furthermore, at ODP Site 1261, the recorded shift towards a more diverse and open-marine dinocyst assemblage is also associated with an increase in the abundance of organic foraminiferal test linings; re-population by calcareous benthic foraminifera (Friedrich et al., 2011), which combined with a reduction in redox sensitive trace metals is indicative of an improvement in environmental conditions and a reduction on the oxygen minimum zone". Warning! There are few samples analysed; 4 samples in 10 m, it's little. This interpretation does not seem really justified because the organic matter concentration is very high.

Reply:

Indeed there are only four samples, however trace metal data and palynological data (including foraminiferal test linings) are from the same samples which show correspondence. In addition, much higher resolution is available from Friedrich et al. (2011) who recorded re-population of benthic foraminifera. Therefore, we believe it reasonable to suggest improved environmental conditions. However, the wording may be too strong given some of the uncertainty and potential for high frequency variations not captured in the sampling resolution. We have therefore amended the text from "is indicative" to "may indicate" and also inserted "However it should be noted that organic matter concentration remains high".

Eldrett, James S GSNL-PTI/EG

Anonymous Referee #2. 14 lines 32ff on page 13: there are benthic foraminiferal assemblage data available form these sites, how do they compare to the data produced by linings? This would be a good test to show if the presented foram data of this study are of any value.

R13. We agree with the reviewer and shall include benthic foraminifera abundances for Sites 1260 and

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Anonymous Referee #2. 15

lines 2-3 on page 14: I am not aware that there are any data e.g. Nd isotopes from

these sites that support a boreal influence. Furthermore, the authors state that this water mass should only influence shallow water settings. However, a cold boreal watermass should be denser than wormer waters near the tropics and therefore influencing bottom waters and not surface waters as suggested here

R15. As we state "The southern expression of this boreal influence is therefore limited in duration and extent". There is a Nd isotope excursion at this horizon and the nature of Nd signal is complex and requires additional localities from the KWIS., We state that our findings "indicating more complex interaction between water-masses and the oxygen minimum zone" and requires further work to resolve. It is an assumption by the reviewer that a boreal watermass would be denser as it would be colder; currently thermal gradients are

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regional climatic and/or tectonic subsidence-induced incursions of boreal water that ventilated the seaway rather than solely by global eustatic drivers that are difficult to reconcile with a greenhouse world without significant polar ice. The development of coeval hiatal intervals are also recorded -near the top and base of the OAE-2 CIE in the Bonarelli interval, Italy, where vigorous bottom currents were proposed to be induced by warm and dense saline deep waters that originated on tropical shelves in the Tethys and/or proto-Atlantic Ocean (Gambacorta et al. 2016). Although our contribution proposes an alternative mechanism for invigorated circulation, it further supports the suggestion by Gambacorta et al. (2016) that enhanced current activity and associated oceanographic circulation was much more dynamic than previously thought during times of Greenhouse climates when conditions were thought to be more equitable.

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4.2.3. Global trace metal draw-down during OAE-2

During the Cenomanian, sediments that have werebeen influenced by equatorial-Atlantic tethyan waters record redox sensitive trace metals that are enriched compared to average shale, in particular Molybdenum (Mo) which requires free H₂S for authigenic sedimentary enrichment (-(Helz et al. 1996) and is interpreted as reflecting anoxic to euxinic depositional conditions favouring organic matter preservation (see Algeo and Lyons, 2006). The relative enrichment of Mo compared to the other redox elements such as U and V in the pre-OAE-2 lower Eagle Ford Gr., suggests an active cycling of Mn–Fe particulates (especially Mn-oxyhydroxides) between the water column and sediment water interface characteristic of a "particulate shuttle" (Algeo and Tribovillard, 2009; Tribovillard et al. 2012; Figure 15a). The presence of aryl isoprenoids in this interval from the Iona-1 core also supports at least temporary and partial water column (photic zone) euxinia at this time (Sun et al. 2016)

During the onset of OAE-2 the recorded depletion of sedimentary redox sensitive trace metals has been proposed to reflect the draw-down of the global trace metal sea-water inventory due to sequestration in sediments under expanded anoxic/euxinic conditions (Hetzel et al. 2009; Dickson et al. 2016; Goldberg et al. 2016). These interpretations are based primarily on the exceptionally low Mo/TOC gradients compared to those documented in modern-day anoxic silled marine basins (Algeo and Lyons, 2006), whereby removal of aqueous Mo concentrations under anoxic/euxinic conditions to the sediment is in excess to resupply by inter-basinal transfer of water masses. This contribution also documents exceptionally low Mo/TOC gradients for sediments spanning the OAE-2 interval from Demerara Rise and SW Texas (Figure 15b; also see Hetzel et al. 2009; Eldrett et al. 2014 Fig. DR3), but also from those recovered from the upper Eagle Ford Gr., and from the Dakota Sandstone interval from the Portland-1 core (supplemental datafile).

However, there are three main challenges to the application of Mo/TOC gradients to infer global trace metal inventory drawdown during OAE-2. Firstly, during the OAE-2 interval in the KWIS, Demerara Rise and Kerguelen Plateau, the weak positive co-variance between Mo and U-is indicates d-of deposition along the unrestricted open marine trend (Figure 15a; also Eldrett et al. 2014); an interpretation supported by the palynological assemblages presented here. Therefore, during the OAE-2 interval, these localities were not subject to significant hydrographic restriction (e.g., silled basins), being deposited instead along continental margins. Thus the application of modern-day Mo/TOC gradients to infer hydrographic restriction and trace metal

Commented [29]:

Anonymous Referee #2. 16

line 35-36 on page 14: nowadays, nobody thinks anymore that the Cretaceous had a equitable climate!
R16. We agree and that is why it is stated .." than previously thought". In addition, this is an important point to make as not many studies have investigated

this topic; the contribution of Gambacorta et al., (2016)

is notable and our data is supportive of the proposition.

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Anonymous Referee #2. 17

lines 23-24 on page 15: but the red dots are all over the place

We have clarified this in the text and inserted "weak positive co-variance between Mo and U..".

drawdown for OAE-2 may not be valid and requires additional studyis not valid (see discussion in Algeo and Lyons, 2006).

Secondly, as Mo is mostly incorporated into organic matter, we propose that the various contributions of different types of organic matter would affect Mo/TOC gradients. Whereas sulfurized organic matter is known to enhance Mo uptake by the sediment (Tribovillard et al. 2004), the impact of variable contributions of labile versus refractory organic matter is not well constrained. The palynological analyses presented here demonstrate increased contribution of refractory terrigenous organic matter (>T:M ratio) in the samples with low Mo/TOC values from the KWIS and Demerara Rise (Figure 15b). We cannot determine the impact of this observation as the T:M ratio reflects a relatively small proportion of the total refractory organic matter, however further investigation is warranted into the variable origin of the more dominant and relatively unknown component, namely AOM; and whether Mo is preferentially incorporated within different organic matter components.

Thirdly; in all the sections presented here the depletion in redox sensitive trace metals during OAE-2 is either associated with oxygenated depositional conditions, or an improvement in redox state. Therefore Therefore, the recorded depletion in redox sensitive trace metals, in particular Mo may instead -record a genuine environmental response to benthic oxygenation preventing authigenic sedimentary enrichments as under oxic conditions, Mo is present in seawater as the stable and largely unreactive molybdate oxyanion. These observations are possibly related interpreted to be related to the equatorial migration of boreal water-masses promoting water column destratification and partial ventilation/re-oxygenation of the surface - intermediate deep waters.

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These challenges were partly addressed by Dickson et al. (2016) who proposed that the recorded low Mo/TOC gradients from Site 1138, Kerguelen Plateau reflected the widespread and global nature of trace-metal depletion during OAE-2 by demonstrating that i) localized oxygenation was also not responsible due to Mo isotope compositions and ii) organic matter type was not a control due to the low abundance of terrigenous material. Palynological investigation of these sediments (this study) confirms the low abundance of terrigenous material at Site 1138. In addition, the dinocyst assemblage recovered from this site is typical of a well-oxygenated boreal equivalent (austral) water-mass with high diversity and abundance of G-cysts, notably Cyclonephelium compactum- membraniphorum dinocyst plexus, and outer neritic to open marine taxa such as Spiniferites ramosus and Pterodinium cingulatum. These findings would suggest deposition under the influence of an oxygenated and open marine/hydrographically unrestricted austral (Southern Hemisphere) surface water-mass on the Kerguelen Plateau; consistent with the relatively low $\delta^{98,95}$ Mo values and Mo-U co-variance trends reflecting oxic-suboxic depositional conditions; with at most sulphidic pore-waters (Dickson et al. 2016). Furthermore, conditions at the sediment-water interface were sufficient for the presence of a low diversity, high carbon-flux benthic foraminifera biofacies (Holbourn and Kuhnt, 2002) as also indicated by the rare occurrence of organic linings of benthic foraminifera throughout the OAE-2 interval at Site 1138 (this study)...-Therefore, in addition to trace metal drawdown as an explanation to reconcile low Mo/TOC gradients and hydrologically unrestricted regime, it is possible that even in the organic-rich laminated sedimentary intervals (i.e. Sites 1138 and 1260) a slight improvement increase in redoxic conditions related to the influence of oxygenated high-latitude australboreal water-masses, may result in aqueous H2S concentrations at the sediment- water interface being below the

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Anonymous Referee #2. 18

last paragraph page 15: wouldn't be the absolute amount of refractory terrestrial organic matter (RTOM) an even more important factor than the T:M ratio alone? The ration could be high even when there is less RTOM and therefore a lower influence on Mo! Since this is not quantified, this is a weak justification and discussion.

The text is slightly amended to include "refractory" and feel this uncertainty is now adequately captured

Commented [32]:

Anonymous Referee #2. 19

last paragraph page 15: wouldn't be the absolute amount of refractory terrestrial organic matter (RTOM) an even more important factor than the T:M ratio alone? The ration could be high even when there is less RTOM and therefore a lower influence on Mo! Since this is not quantified, this is a weak justification and discussion.

Text amended and processes added

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Anonymous Referee #2. 20

lines 8-9 on same page: but isn't that what you are proposing above?

In part we agree, but the actual relationship between refractory organic matter and Mo is not documented by Dickson et al. (2016); as such this is the first integrated data of terrigeneous organic matter and Mo enrichments.

critical threshold for conversion of molybdate to thiomolybdate (Helz et al. 1996), resulting in the observed depletion in Mo sedimentary concentrations and low Mo/TOC gradients during OAE-2. This interpretation is also supported by the occurrence of benthic foraminifera during within the OAE-2 interval in both organic rich and organic lean sedimentary sections from Demerara Rise (Friedrich et al. 2006, 2011); Kerguelen Plateau (Holbourn and Kuhnt, 2002); Texas (Lowery et al. 2014; Dodsworth, 2016; this study) and central KWIS (e.g. Eicher and Worstell, 1970; Keller and Pardo, 2004; Elderbeck and Leckie, 2016 and references therein).

5. Conclusions

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This integrated palynological and geochemical study withinusing a multidisciplinary approach has provided insights into depositional environments of the Cenomanian-Turonian Eagle Ford Gr. and bounding formations of the Buda Limestone and Austin Chalk from southwest Texas, USA. The study spans a physiographic transect across the Comanchean Shelf from the shallow setting of the San Marcos Arch (AU2); through the main shelf (Innes-1), the slope (Iona-1) to the intra-shelf basin (well X). Furthermore, comparison of the paleoenvironmental trends recorded in the Cenomanian – Coniacian section from SW Texas to those recorded further north within the central KWIS (USGS Portland-1) and to the south in the equatorial North Atlantic (ODP sites 1260, 1261, Demerara Rise) and Kerguelen Plateau (ODP Site 1138) allows the interpretation of the evolution of various water-masses along this north-south transect. The main findings are:

- High latitude boreal and austral (Northern and Southern Hemisphere) and equatorial <u>Atlantic</u> tethyan water-masses can be distinguished based on their distinct palynological assemblages and geochemical signatures.
- 2. The northward flow of a suboxic-anoxic equatorial Atlantic tethyan water-mass into the Western Interior Seaway occurred during the early-middle Cenomanian; followed by a major re-organisation of the oceanographic regime during the latest Cenomanian- Turonian as a full connection of the Western Interior Seaway with a northerly- boreal water-mass was established during peak transgression. This oceanographic change promoted de-stratification of the water column and improved oxygenation throughout the KWIS and as far south as the Demerara Rise.
- These long term trends in water-mass evolution are tentatively linked to third order eustatic
 transgression-regression cycles driven by regional <u>Cordilleran</u> tectonic and/or mantle plume-lithosphere
 dynamics associated with the emplacement of LIPs during this time as well as shorter term variations in
 climate (i.e. Plenus Cold Event).
- 4. Low Mo/TOC ratios in the equatorial North Atlantic in comparison to other oceanic basins during the onset of OAE-2 argue for partial restriction and draw-down of global trace metal sea-water inventories. However, this study demonstrates that the recorded decline in redox-sensitive trace metals during the onset of OAE-2 likely reflect either a genuine oxygenation event related to open water-mass exchange

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Anonymous Referee #2, 21

lines 21-23: check benthic assemblage data for these sites if available and see if there are benthic forams occurring in these intervals. If yes, these are oxygenation events, if not, it was anoxic. This would be an independent proof of the statements made here.

Benthic foraminifera are present, text ammended

Commented [35]:

Anonymous Referee #2. 22

point 1 in the conclusions: are these water masses be interpreted to be surface and bottom-water masses at once? This has to be clarified in the discussion. R22. See comment R15. This has been clarified in the discussion.

at this time and/or is further complicated by variable contribution of organic matter from different sources (e.g. refractory/terrigenous material) that requires further evaluation.

6. Acknowledgements

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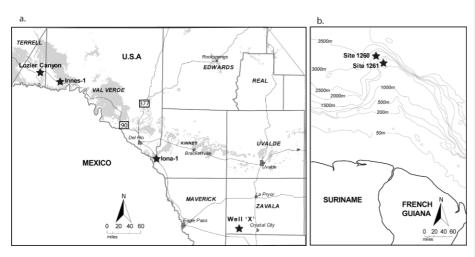
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Figure 1: Site Locations. a. Present-day position of the study areas; b. Turonian paleogeographic reconstruction with site locations; gray shaded area= landmass; dotted line = paleo-shelf; CLIP = Caribbean Large Igneous Province; HALIP = High Arctic Large Igneous Province. Boxes show study area as presented in Fig. 2

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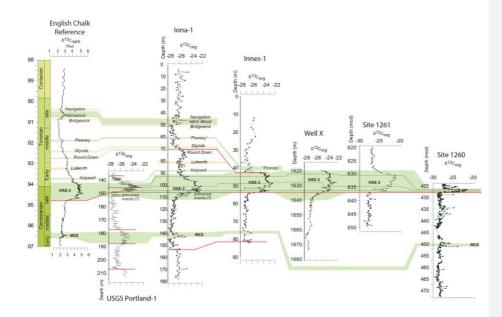


Figure 3: Isotope data and correlation between studied sites and the English Chalk Reference section. Data sources: δ^{13} C for English Chalk (Jarvis et al. 2006); USGS Portland-1 (Joo and Sageman, 2014; Eldrett et al. 2014; Duvivier et al. 2014; Eldrett et al. 2015a, this study) ODP sites 1260-1261 (Erbacher et al. 2005; Friedrich et al. 2008) Note variable depth scales. Wavy horizontal red lines represent hiatal surfaces. Green shading = positive δ^{13} C isotope events; Orange shading = negative

 δ^{13} C isotope events. Isotopic events nomenclature follows that of Jarvis et al. (2006); with the exception of precursor events (after Eldrett et al 2014); which are now though to occur within the OAE-2 CIE; but in the section not recorded at Portland-1 core or the Pueblo GSSP due to the hiatal surface at the base of the Bridge Creek Limestone.

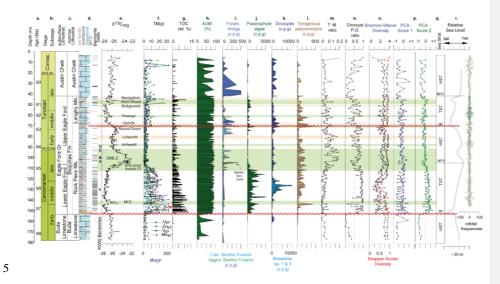


Figure 4: Iona-1 core data. a. depth/age; b. chronostratigraphy; c. lithostratigraphy; d. lithology; e. $\delta^{13}C_{org}$; f. redox sensitive trace metal enrichments (TM_{EF}); g. TOC; h. AOM; i. foraminiferal test linings; micropalaeontological abundance data as counts per gram (c.p.g) for both calcareous (Calc.) and agglutinated (Agglut.) benthic foraminifera; j. prasinophyte algae; k. dinocysts, Bosedinia cf. sp. 1 & 3; l. Terrigenous palynomorphs; m. T:M ratio; n. dinocyst P:G ratio; o. Shannon-Wiener and Simpson-Hunter dinocyst diversity; p. Principle Component results; eigen score 1, eigen score 2; q. Sequence stratigraphic interpretations (see supplemental information); r. inferred relative sea level history (after Minisini et al. in review), with Milankovitch orbital frequencies (after Eldrett et al. 2015a, b).

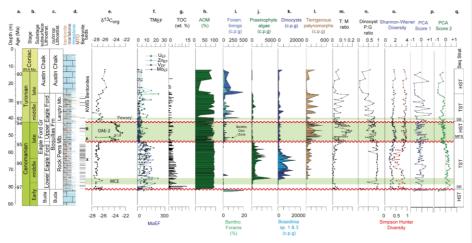


Figure 5: Innes-1 core data. Legend same as Figure 4. Lithology after Minisini et al. (in review).

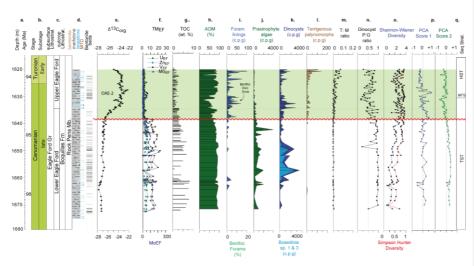


Figure 6: Well 'X' core data. Legend same as Figure 4.

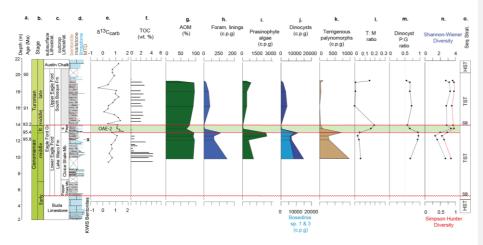


Figure 7: Bouldin Creek outcrop (AU-2) data. Legend same as Figure 4. Lithology after Minisini et al. (in review). $\delta^{13}C_{org}$ and TOC (wt. %) from Lowery et al. (2014)

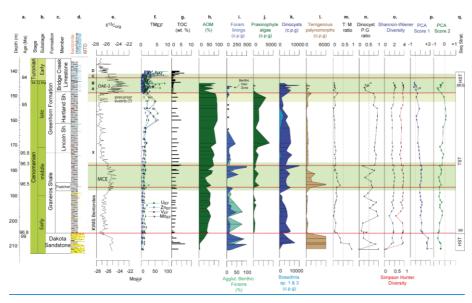


Figure 8: USGS Portland-1 core data. Legend same as Figure 4. $\delta^{13}C_{org}$ data from Joo and Sageman (2014); Eldrett et al. (2014); Duvivier et al. (2014); Eldrett et al. (2015a), this study.

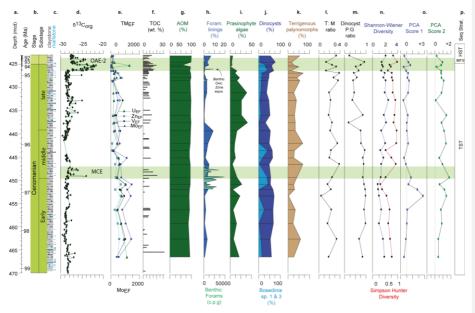


Figure 9: ODP Site 1260 data. Legend same as Figure 4. $\delta^{13}C_{org}$ data from Erbacher et al. (2005); Friedrich et al. (2008).

Benthic foraminiferal abundance data from Friedrich et al. (2006),

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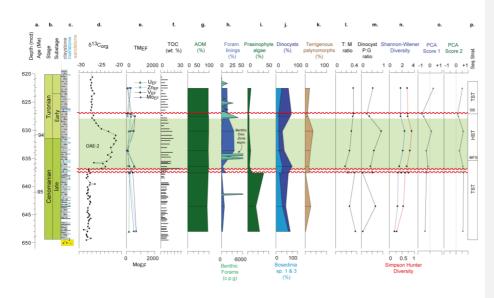
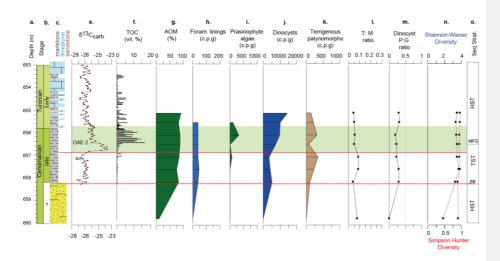


Figure 10: ODP Site 1261 data. Legend same as Figure 4. $\delta^{13}C_{org}$ data from Erbacher et al. (2005) and Friedrich et al. (2008). <u>Benthic foraminiferal abundance data from Friedrich et al. (2006).</u>



 $\textbf{Figure 11:} \ ODP \ Site \ 1138 \ data. \ Legend \ same \ as \ Figure \ 4. \ \ \delta^{13}C_{org} \ and \ TOC \ data \ from \ Dickson \ et \ al. \ (2016).$

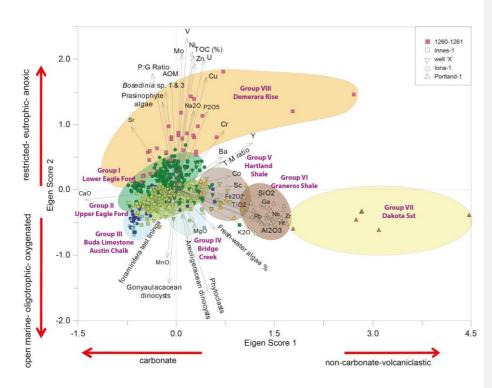


Figure 12: PCA results showing principle axis/eigen scores; selected environmental variables. Samples plotted as a function of site location (see insert) and lithostratigraphic interval (colours; groups in figure).

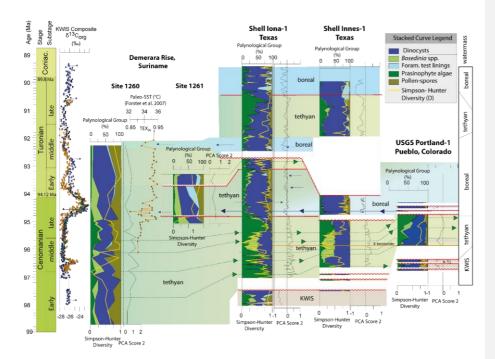


Figure 13: Cenomanian-Coniacian chronostratigraphic correlation of interpreted water-masses along a South-North transect from the equatorial western Atlantic to the central Cretaceous Western Interior Seaway (KWIS). The KWIS composite δ¹³Corg after Joo and Sageman (2014) and including data from Duvivier et al. (2014). Eldrett et al. (2015) for USGS Portland-1 (orange), plus data from Iona-1 (blue); Innes-1 (black) and well 'X' (green) cores calibrated using the age model of Eldrett et al. (2015a). Stacked curves showing principal palynological components (legend on figure). Paleo-SST and TEX₈₆ record from ODP Site 1260 from Forster et al. (2007). Colour shading: green = tethyan sourced water from the eEquatorial Atlantic; blue = northerly boreal watermass: brown = locally sourced KWIS watermass. Green dashed arrows indicate tethyan incursions; blue arrows indicate boreal influence. Principal Component Score (PCA) axis 2; positive indicative of eutrophic/anoxic waters; negative indicative of oxygenated oligotrophic waters (see Figure 12).

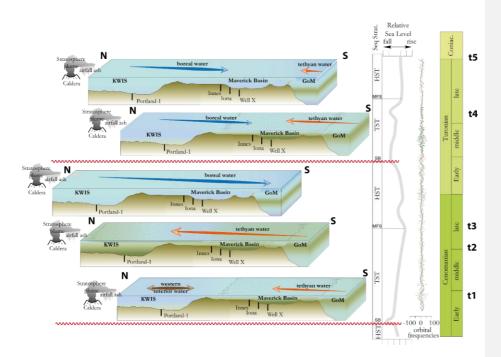


Figure 14. Regional water-mass depositional model of the KWIS. Time_selices (t1-t5) showing the northward flow of eutrophic/anoxic tethyan equatorial Atlantic water into the Western Interior Seaway from the proto-Atlantic and southward flow of a boreal watermass. Milankovitch orbital frequencies of eccentricity, obliquity and precession from the Iona-1 core (Eldrett et al. 2015a) resulting in shorter-term (<0.1 Ma) development of limestone-marlstone couplets (Eldrett et al. 2015b). GoM = Gulf of Mexico. Sea level curve after Minisini et al., (in review).

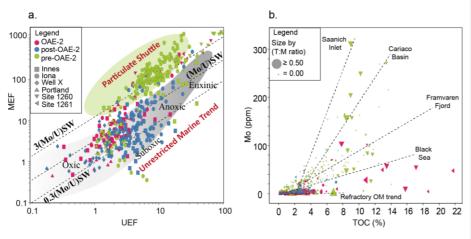


Figure 15. a, Logarithmic plot of Mo:U covariance for the studied sections. Diagonal dashed lines represent multiples (0.3, 1, and 3) of Mo:U ratio (3) of present-day seawater (SW). Values greater than x3 SW are generally associated with the particulate shuttle; whereas those trending with modern day sea water reflect an unrestricted marine trend with a weak positive Mo:U covariance. b, Mo-TOC ccovariance data for studied sections compared to modern anoxic silled-basin

environments (after Tribovillard et al. 2012). Regression of the modern datasets are shown as solid lines with MO/TOC regression slopes displayed (Tribovillard et al. 2012). The proposed refractory organic matter trend added as dashed line.