Hydroclimate variability in the United States continental interior during the early Eocene Climatic Optimum

Amy L. Elson a,⁎, Megan Rohrssen b, John Marshall a, Gordon N. Inglis a, Jessica H. Whiteside a

a School of Ocean and Earth Science, University of Southampton, National Oceanography Centre Southampton, UK
b Geology and Environmental Sciences, Central Michigan University, USA

A R T I C L E   I N F O
Editor: Shucheng Xie
Keywords:
Eocene
Green River Formation
EECO
Hydrogen cycle
Compound-specific hydrogen isotope analysis
Lacustrine

A B S T R A C T
The early Eocene (56.0 to 47.8 million years ago) was punctuated by a series of transient episodes of rapid global warming superimposed on the long-term early Cenozoic warming trend, culminating in the early Eocene Climatic Optimum (EECO; 53.3 to 49.1 million years ago). Details of the hydroclimate regime operating during the EECO are poorly constrained, especially for continental interior sites. The Green River Formation (GRF) of Utah and Colorado was deposited in a suite of large, unusually productive lakes that offer an ideal opportunity to study the hydrological response to warming. Here we report the hydrogen isotopic composition (δ2H) of leaf wax (long-chain n-alkanes) and algal (phytane) lipids preserved in the organic-rich Mahogany Zone (49.3 to 48.7 Ma) and use these data to reconstruct precipitation and lake water δ2H records, respectively. We observe large inter-site variations in algal and leaf wax δ2H values (~50 to 75‰), suggesting that additional local controls influence precipitation and/or lake water δ2H (e.g., salinity). Intriguingly, leaf wax and algal lipid δ2H values show little variation through the Mahogany Zone, implying a relatively stable hydrological regime during the latter phase of the EECO. This contrasts with the more variable hydrological regime that prevailed during early Eocene hyperthermals. Unlike the EECO, the early Eocene hyperthermals in the Uinta region do not coincide with the deposition of organic-rich sediments. This suggests that a stable hydrological regime during the EECO may enable the preservation of organic matter within continental-interior lake systems, potentially leading to an important negative climate feedback during the early Eocene and other greenhouse climates.

1. Introduction

The Green River Formation (GRF) was deposited between ~53 and 44 million years ago (Ma) and represents a series of intermittently interconnected terminal continental-interior basins that extended across north-eastern Utah, north-western Colorado, and south-western Wyoming (Bradley, 1929; Tissot et al., 1978; Dyni, 1987; Smith et al., 2008, 2010) (Fig. 1). Lake Uinta was deposited in the Uinta Basin (north-eastern Utah) and is characterized by at least three hypersaline intervals (Vanden Berg and Birgenheier, 2017). The first of these coincides with the richest interval of organic carbon (OC) content in the Green River Formation, the Mahogany Zone (MZ). Deposited over ~400 thousand-years (kyr), the MZ comprises sediments deposited in the deepest part of the paleo-lake (Tissot et al., 1978). This unusually organic-rich section is found throughout the basin and contains a thin (0.5 meter; m) marker bed of peak total organic carbon (TOC; 43 weight percent%; wt %; Whiteside and Van Keuren, 2009), referred to as the Mahogany Bed marker.

The MZ was deposited towards the end of the early Eocene Climatic Optimum (EECO; 53.3 to 49.1 Ma) and is constrained by radioisotopic dating (49.32 ± 0.30 to 48.66 ± 0.23 Ma; Smith et al., 2008, 2010). The EECO was an interval of sustained global warmth, with global surface temperatures reaching ~10–16 °C above pre-industrial levels (Zachos et al., 2001; Inglis et al., 2020). The EECO is also characterized by an intensified hydrological cycle (Carmichael et al., 2016) with evidence for enhanced rainfall in high-latitudes (Carmichael et al., 2016; Inglis et al., 2022). However, the hydrological response within the low to mid-latitude continental interiors has been assessed in only a few studies (Hyland and Sheldon, 2013; Carmichael et al., 2016). A better understanding of hydrological cycle perturbations during the early Eocene could provide important insights into a range of biogeochemical processes, including soil erosion rates, methane cycling and organic carbon burial (Carmichael et al., 2017 and references therein). These processes may have acted as positive or negative feedbacks to global warming and

⁎ Corresponding author.
E-mail address: ale1u16@soton.ac.uk (A.L. Elson).

https://doi.org/10.1016/j.palaeo.2022.110959
Received 27 April 2021; Received in revised form 20 March 2022; Accepted 24 March 2022
Available online 27 March 2022
0031-0182/© 2022 The Authors. Published by Elsevier B.V. This is an open access article under the CC BY license (http://creativecommons.org/licenses/by/4.0/).
thus, may have played an important role in early Cenozoic greenhouse conditions. However, understanding these processes requires better constraints on the hydrological cycle of these large early Eocene lakes.

Here we determine compound-specific hydrogen isotope ($\delta^2$H) values in leaf waxes and algal lipids to reconstruct changes in the hydrological cycle over the mid-latitudes during the deposition of the Mahogany Zone. In modern settings, leaf waxes record the $\delta^2$H of the source water from the surrounding vegetative environment, whereas phytane is commonly derived from autotrophic aquatic microorganisms and provides insights into the $\delta^2$H value of lake water (Volkman et al., 1998; Eglinton and Eglinton, 2008; Sachse et al., 2004, 2012). We use lipid $\delta^2$H values to infer changes in the $\delta^2$H value of precipitation through the latest EECO and post-EECO interval (~49.3 to 48.7 Ma) and assess the stability of the hydrological cycle during this event. We also explore the role of the hydrological cycle in the deposition of the organic-rich Mahogany Zone during the termination of the EECO.

2. Background and methods

2.1. Site description

Continual industry interest in the oil shales of the Green River Formation has resulted in numerous boreholes, particularly in the Uinta Basin. We sampled a centre to margin transect of the profundal zone through three Mahogany Zone sections (Fig. 1). The main facies represented are continuous parallel- to undulose laminated mudstone with little bioturbation. Drilled by TOSCO, the Utah State 1 core represents a basin-centre during the EECO and post-EECO interval (~49.3 to 48.7 Ma) and contains a relatively condensed section of the Mahogany Zone (39.870658°N, 109.112281°W). The P-4 Chevron White Shale Project core (P-4) was drilled in the eastern section of the Uinta Basin (reported as 40.010576°N, 109.511638°W). Drilled by the Utah Geological Survey, the Skyline 16 core represents a proximal basin-margin, lacustrine setting and contains a relatively condensed section of the Mahogany Zone (39.931419°N, 109.134227°W). Drilled by TOSCO, the Skyline 16 core represents a basin-margin during this time. Locations of the cores are indicated in red circles. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

2.2. Age model

The Green River Formation is punctuated by numerous tuff layers from the northeastern Absaroka Volcanic Province. Two dated tuff horizons - the Curly and Wavy tufts (49.32 ± 0.33 Ma and 48.66 ± 0.27 Ma respectively; Smith et al., 2008) – are found below and above the Mahogany Zone of the P-4 drill core, respectively. These tuff layers have been dated with $^{40}$Ar/$^{39}$Ar, measured on single crystal analysis of biotite (Smith et al., 2008) and used to develop an age model for the P-4, Skyline 16, and Utah State 1 Mahogany Zone sections. Detailed sedimentological analysis supports a nearly constant accumulation rate of 100–200 μm/yr (Smith et al., 2008; Whiteside and Van Keuren, 2009; Walters et al., 2020).

2.3. Organic geochemistry

Rock plugs through the Mahogany Zone from Utah State 1, P-4 and Skyline 16 (2274–2376 ft., 683–785 ft. and 430–513 ft. respectively) were removed with a water-cooled drill press (Delta DF300L) and powdered by agate mortar and pestle. Molecular extraction and fractionation were conducted at the University of Southampton. Total lipid extracts (TLE) were isolated from powdered rock using a Thermo 350 Accelerated Solvent Extractor with the following program: preheat = 5 min; heat = 5 min; static = 5 min; pressure = 1500 psi; flush = 70%, purge = 300 s.; cycles = 3; solvent = dichloromethane:methanol (9:1, v/v). Solvent extracts were evaporated using a Genevac EZ-2 vacuum centrifuge and subsequently fractionated using silica gel columns. The TLE was eluted with hexane, hexane:dichloromethane (DCM) (4:1, v/v), and DCM:methanol (MeOH) (1:1, v/v), yielding the aliphatic, aromatic and polar fractions, respectively. Activated copper was added to each fraction to remove elemental sulphur.

Biomarker identification was performed using a Thermo Trace 1310 Gas Chromatograph (GC) coupled to a Thermo TSQ8000 Triple Quadrupole Mass Spectrometer (MS). The GC used a DB-5 column (30 m ×
0.25 mm i.d, 0.25-μm film thickness) with the following oven program: 40 °C (held for 2 min), increased at a rate of 6 °C/min to 310 °C, and then held for 20 min. Compound identification of n-alkanes and pristane/phytane was made using mass spectra and comparison with an in-house reference oil (North Sea Oil-1).

Compound-specific hydrogen isotope analysis was conducted using a Thermo Scientific Trace 1310 GC with a DB-5 column (30 m x 0.25 i. d.25-μm film thickness) coupled to a Delta V Plus Isotope Ratio Mass Spectrometer via a Thermo GC Isolink and Conflo IV. Samples were injected splitless and the GC program was as follows: 40° for 2 min then 6 °C/min to 310 °C, and then held for 15 min. Results are reported in delta notation (%o) and normalized to a suite of n-alkanes with a known isotopic composition (n-C16 to n-C30; i.e., the A7 standard reference material obtained from Arndt Schimmelmann). Standards were run in triplicate before and after each sample batch (n = 5). The standard deviation was typically between ~2 and 5%. Samples were rejected when the standard deviation exceeded 5%. Error bars represent the standard deviation of the A-7 mix run in concert with samples. The H2 values through the Mahogany Zone samples in Skyline 16.

Several methods for characterising the n-alkane distribution of a sample have been developed, including the carbon preference index (CPI), average chain length (ACL) and odd-over-even predominance (OEP). In fresh plant material, the CPI is high (>.5 to 40; Bush and McInerney, 2013), but decreases over time due to thermal maturation, approaching values of ~1 in mature oils and sediments. The CPI is calculated following Marzi et al. (1993) as:

\[
\text{CPI} = \frac{(2 \times (C_{23} + C_{25} + C_{27} + C_{29}))}{(C_{22} + 2 \times (C_{24} + C_{26} + C_{28}) + C_{30})}
\]

ACL represents the weighted averages of carbon chain lengths and can be calculated as (Ficken et al., 2000; Bush and McInerney, 2013):

\[
\text{ACL} = \frac{(25 \times C_{23}) + (27 \times C_{25}) + (29 \times C_{27}) + (31 \times C_{29}) + (33 \times C_{31})}{C_{23} + C_{25} + C_{27} + C_{29} + C_{31} + C_{33}}
\]

Vascular plants synthesize hydrocarbons with a strong predominance of odd-over-even numbered n-alkanes. The formula to determine OEP used here (following Scalan and Smith, 1970) is:

\[
\text{OEP} = \frac{(C_{27} + 6 \times C_{29}) + C_{31}}{(C_{29} + C_{31} + 4 \times C_{33})}
\]

3. Results

3.1. Leaf wax distributions

In the most distal site (Utah State 1), CPI values are relatively low and range from 1.4 and 2.4, with little observed variation apart from slight fluctuations towards the base and top of the Mahogany Zone. The P-4 core exhibits variable CPI values ranging from 0.7 to 3.5 with larger variations towards the base and top of the section. In the most proximal site (Skyline 16), CPI values are relatively high (1.9 to 4.5) with larger variations towards the base and top. As expected, the OEP values for each site display similar patterns. In the most distal site (Utah State 1), the OEP ranges between 1.5 and 3.9. The P-4 core exhibits variable OEP values (ca. 0.5 to 10.6), with more variation at the base and top. In the most proximal site (Skyline 16), OEP values fluctuate from 4.1 up to 10.2, with higher values observed towards the upper and the lower portions of the Mahogany Zone.

In the most distal site (Utah State 1), the ACL is characterized by very similar values through the section (27.7 to 30.0), with stronger variation in values towards the base and top of the section. ACL values in P-4 range from 26.3 to 30.8, with more variation observed in the deepest samples (781.7–763.15 ft). In the most proximal core (Skyline 16), ACL values vary from 28.5 to 30.2, showing minor variability with values slightly higher than those in the Utah State 1 and P-4 cores (Tables 1–3, see Supplementary Information).

3.2. Compound-specific hydrogen isotope values

3.2.1. Utah State 1

C29 n-alkane δ2H values range from −111.2 to −169.4‰ (Fig. 2). The highest δ2H values (−111.2‰) are found at 2371.5 ft. (correlating to the Curly Tuff bed, an ash layer deposited in a tuffaceous debris flow; Smith et al., 2008) and the lowest δ2H values (−169.4‰) are found at 2290.15 ft. δ2H values are particularly low at 2314.1 and 2311.45 ft. (−165.2 and −166.8‰ respectively) and represent beds that are rich in organic matter, including the Mahogany Bed at 2314 ft. Phytane δ2H values range from −245.7 to −292.4‰, with the highest δ2H values present at the base of the section. Lower δ2H values are observed at 2314.1 and 2308.3 ft. (−289.6 and −291.4‰ respectively), coinciding with lower δ2H wax values.

3.2.2. P-4

δ2H wax values from the more proximal P-4 range from −89.4 to −143.0‰, which are higher than values in Utah State 1 and Skyline 16. Lower variability is observed in this proximal core (typically ±10‰) and no clear trend is observed upwards through the section. The lowest δ2H wax value is seen at 731 ft. (−143.0‰) and the second lowest δ2H wax recorded is −110.1‰, immediately below the deposition of the Mahogany Bed. In the proximal P-4 core, δ2H wax values are relatively low and vary from −247.0 to −224.6‰. We observe an upwards trend of increasingly lower δ2H wax values in the middle and upper sections with higher δ2H wax values near the base (748.5–738.0 ft).

3.2.3. Skyline 16

δ2H wax values in the Skyline 16 core vary from −164.1 to −208.5‰ (472.1 and 437.9 respectively). There is a trend of progressively lower δ2H values through the Mahogany Zone samples in Skyline 16. δ2H phytane values in the Skyline 16 core vary from −295.6 to −265.4‰. Much of the section, however, shows much more limited variation. However, the top of section exhibits higher δ2H phytane values.

4. Discussion

4.1. Controls on phytane δ2H values within the Uinta Basin

The hydrogen isotopic composition of phytane is affected by multiple controls, including changes in source organism and/or variations in source water δ2H. These factors need to be considered before interpreting the isotopic record (see discussion below).

4.1.1. Non-algal sources

Phytane is mostly derived from the side chain of chlorophyll-a. As such, it is thought to average the entire phytoplankton community (Witkowski et al., 2018). However, phytane can have multiple sources (e.g. methanogens and halophiles; ten Haven et al., 1987), which may influence δ2H phytane values. We argue that inputs from methanogens and halophiles are relatively minor, due to the extreme productivity of the lake autotrophs within the photic zone and evidence for predominantly microbial organic matter found in petrographical studies (Elson et al., 2022). This would have vastly outweighed the potential input from these alternate sources.

4.1.2. Changes in source water δ2H

The input of δ2H-depleted snowmelt into the lake basin may affect δ2H phytane values during the growing season. Early Paleogene
Fig. 2. (Top) Utah State 1 core results: Stratigraphic column through the Mahogany Zone, Utah State 1, through the basin center (a) Left: $\delta^2$H of phytane values. Right: $\delta^2$H of C$_{29}$ n-alkane. (b) CPI measurements with higher values indicating an increased input of vascular plant material (calculated from Marzi et al., 1993.) (Middle) P-4 core results: Stratigraphic column through the Mahogany Zone, P-4, through the basin margin (a) Left: $\delta^2$H of phytane and C$_{29}$ n-alkane. Right: $\delta^2$H. (b) CPI measurements. (Bottom) Skyline 16 core results: Stratigraphic column through the Mahogany Zone, Skyline 16, through the basin margin (a) Left: $\delta^2$H of phytane. Right: $\delta^2$H of C$_{29}$ n-alkane. (b) CPI measurements. MB = Mahogany Bed Marker. (* Smith et al., 2008). Error bars represent 1σ uncertainties. Depth is reported in both meters (stratigraphic log) and feet (geochemical results).
topography reconstructed for the Uinta Mountains reached at least 3000 m high with a basin floor paleoelevation of, at most, 1000 m high (Gao and Fan, 2018) and may have supplied snowmelt to the surrounding lake basins (Norris et al., 1996; Sewall and Sloan, 2006). However, proxy estimates from the basin have placed mean annual air temperature (MAAT; Wing, 1998) and warm month mean temperature (WMMT) estimates at ~16 °C and ~40 °C (Snell et al., 2013), respectively. Combined with floral and faunal studies (Wing, 1998), this suggests that temperatures would rarely drop below zero. Well-preserved palm trees (e.g. Phoenix windmills; Snell et al., 2013) and fossil crocodilians (Markwick, 1994), have also been found through the Green River Formation and wider region, indicating CMMT >5 °C (cold month mean temperature) and MAT >10 °C. Thus, it is unlikely that input of 2H-depleted snowmelt into the lake basin changed dramatically over our timescales (e.g., ~10^8–10^9 kys).

The δ^2H value of the lake surface water would have been particularly sensitive to water balance changes in the basin, either through precipitation-evaporation (P-E) and/or the addition of isotopically distinct water from other sources. Previous work in the Uinta Basin has identified three hypersaline zones within the upper Green River Formation (Vanden Berg and Birgenheier, 2017). The first of these hyper-saline phases coincides with deposition of the Mahogany Zone and is restricted to the eastern side of the basin, where the depocenter of the lake was located. Drivers of hypersalinity have been attributed to the input of Lake Gosiute water and associated high-density brines from the Greater Green River Basin to the north, which was far more evaporitic in nature and was undergoing north-south infilling (Smith et al., 2008).

Water transported to Lake Uinta from the shallower, evaporitic basin would have been 2H-enriched and associated with relatively high δ^2Hphytane values (Fig. 5). Skyline 16 – which is further to the south and shallower than the other locations – has the lowest δ^2Hphytane values and may have received less input of 2H-enriched water. The evaporation of river channels and adjustment of regional fluvial systems to a decreasingly energetically hydrological regime may have also resulted in different water sources being delivered to P-4 and Skyline 16, despite their relative proximity in location (Gall et al., 2017; Birgenheier et al., 2019). Although further work is required, it is likely that input of 2H-enriched, saline water exerted an important control on δ^2Hphytane values in the hypersaline portions of the Uinta Basin.

4.2. Controls on leaf wax δ^2H values within the Uinta Basin

δ^2Hwax values primarily reflect the δ2H value of the plant’s source water and – by extension – the hydrogen isotopic composition of precipitation (δ^2Hprecip; Sachse et al., 2012). However, a fractionation factor (εprecip) is required to estimate δ^2Hprecip. Here we employ a net fractionation factor of 110 ± 20% as this captures the variability in modern C3 gymnosperms and angiosperms (Sachse et al., 2012; Pedentchouk et al., 2008). This yields average δ^2Hprecip values of ~43‰ (Utah State 1), +7‰ (P-4) and ~67‰ (Skyline 16). These values are 2H-enriched (~30 to 100‰), relative to modern values of ~96 to ~100‰ (Bowen and Revenaugh, 2003). This can be explained by two mechanisms. Firstly, in a warmer climate, warmer air temperatures will yield more 2H-enriched water vapor (a temperature effect) (Poulsen et al., 2007; Speelman et al., 2010). Secondly, in a warmer world, there will be decreased rainfall efficiency in the subtropics, resulting in more 2H-enriched precipitation at the mid-to-high latitudes (e.g., Pagani et al., 2006; Speelman et al., 2010). However, there are large (~50 to 75%) inter-site variations that suggest additional controls on δ^2Hprecip values (see below).

4.2.1. Diagenesis

Hydrogen exchange processes can alter lipid δ2H values slowly over time and overprint the original environmental signature (Sessions et al., 2004). The most common way to assess isotopic exchange is to compare long-chain n-alkanes and isoprenoid (e.g. phytane) δ2H values (Pedentchouk et al., 2006). In immature modern sediment samples, isoprenoids (e.g. phytol) are 2H-depleted (up to 150 to 200‰) relative to long-chain n-alkanes (Li et al., 2009). However, with increasing maturation, the offset between isoprenoids and long-chain n-alkanes diminishes to zero (Sessions, 2016 and references therein). In the Uinta Basin, the offset between isoprenoids (phytane) and n-alkanes is consistent and large (~100 to 140‰; Fig. 3), suggesting minimal hydrogen exchange. The Green River Formation is also known for the thermal immaturity of its vast oil shale resources (Birgenheier and Vanden Berg, 2011). Relatively low thermal maturity is supported by relatively high OEP (~4.1 to 10.4) and relatively high CPI values (>1.5–4) recorded through the Mahogany Zone. Overall, this suggests that the impact of thermal maturity on δ^2H values is minimal.

4.2.2. Plant type

Changes in the plant community can influence the apparent fractionation between δ^2Hwax and δ^2Hprecip values (εprecip). In modern C3 plants, εprecip values range between ca. ~80 to ~150‰ (Sachse et al., 2012 and references therein) however, accounting for changes in plant community in ancient settings is challenging (Feakins, 2013). Recent work has used pollen assemblages to calculate plant-specific fractionation factors (e.g. Feakins, 2013; Inglis et al., 2022). However, this was not possible here because of the very high content of amorphous organic matter (AOM) in the samples. By volume this comprises the bulk of the samples so that any attempt to dissolve the sample in hydrochloric acid (HCl) or hydrofluoric acid (HF) is near impossible. The AOM both shields the mineral content which is also not sufficiently abundant that removing it disaggregates the sample to release the palynomorphs. An attempt was made to disaggregate the AOM with a tunable ultrasonic probe after the HF treatment but this had little effect. Alternatively, a shift in the average chain length (ACL) could potentially reveal a change in the higher plant community, as shown during the PETM (e.g. Schouten et al., 2007). Although modern plant surveys cast doubt as to whether the ACL can discriminate between key plant types (with the exception of mosses; Bush and Mclnerney, 2013), invariant ACL values in the Uinta Basin sediments imply no significant change in vegetation during the EECO.

4.2.3. Soil and/or leaf water δ^2H-enrichment

In modern sediments, δ^2Hwax values are correlated with source water δ2H (i.e., precipitation). However, evaporative δ2H-enrichment of soil and/or leaf water can modify δ^2Hwax values, especially in (semi-arid) settings (Kahmen et al., 2012). Subsequently, the isotopic difference between terrestrial and aquatic biomarkers can be used to constrain soil and/or leaf water evaporative enrichment (see Rach et al., 2017 and references therein) and is typically calculated using long-chain n-alkanes (C20-C28; i.e. higher plants) and mid-chain n-alkanes (C23-C25; i.e. submerged macrophytes). However, other algal biomarkers (e.g. phytane) can be used (Rach et al., 2017). In sites where evaporative δ2H-enrichment of soil and/or leaf water is minimal, there should be a linear relationship between δ^2Hwax and δ^2Hphytane. With increasing evaporative δ2H-enrichment of soil and/or leaf, the relationship between δ^2Hwax and δ^2Hphytane will weaken. In the Uinta Basin, we find a significant positive linear relationship between δ^2Hwax and δ^2Hphytane (r^2 = 0.78; p < 0.001). This suggests minimal evaporative δ2H-enrichment of soil and/or leaf water within the Uinta Basin (Fig. 4).

4.2.4. Orogenic effects

Intercontinental basins are particularly sensitive to tectonic controls that can have numerous consequences for basin accommodation, sediment supply and lake stratification (Carroll and Bohacs, 1999). Tectonic upheaval leading to rising regional land elevation and dropping temperatures may result in more 2H-depleted precipitation being delivered to the basin (c.f., Dansgaard, 1964; Sachse et al., 2012).

During deposition of the Green River Formation, steady subsidence of the Uinta Basin increased accommodation space, while the sediment
supply fluctuated, often paced by the Eocene hyperthermals (Gall et al., 2017). At the time of deposition of the Mahogany Zone, this long-term tectonic subsidence continued as a result of flexure from Laramide uplifts, including the Uinta Mountains towards the north and the Uncompaghre Uplift and San Rafael Swell towards the south. Paleocurrent data suggest the Douglas Creek Arch, a key structural high controlling connectivity between the Uinta and Piceance Creek basins, was uplifted during the Sunnyside Delta interval of the middle Green River Formation and prior to Mahogany Zone deposition (Birgenheier et al., 2019). In addition to the relatively short time span studied and the presence of several topographic highs surrounding the Uinta Basin, it is unlikely that changes in tectonic upheaval, resulting in a potential amount effect (Dansgaard, 1964) was a driver for the minor upwards depletion of $^{2}$H observed in this section. However, at higher elevation, precipitation becomes progressively $^{2}$H-depleted (e.g., Bai et al., 2015). As such, variations in $^{2}$H$_{\text{wax}}$ may be due to changes in plant wax source regions (e.g., low vs. high elevation).

4.3. Stable hydrological cycle promoted OC burial during the EECO

Our leaf wax and phytane $^{2}$H data indicate relatively gradual changes in source water $^{2}$H in the Uinta Basin during deposition of the Mahogany Zone. This could indicate a stable hydrological cycle during the latter stages of the EECO, which differs from the more variable and dynamic hydrologic response observed during transient climatic events in the region, i.e., the PETM and early Eocene hyperthermals (Hyland et al., 2018). The latter are associated with high seasonality and ephemeral fluvial discharge, leading to high siliciclastic inputs (Gall et al., 2017; Birgenheier et al., 2019), whereas the EECO is characterized by the low siliciclastic input and a lower energy highstand lake (Gall et al., 2017; Birgenheier et al., 2019).

Reduced siliciclastic input during the EECO would have also limited dilution from inorganic (clastic) sediments and thus promoted rich organic matter accumulation within the lake. Indeed, the hyperthermal phase lacks evidence for OC-rich deposits (Birgenheier et al., 2019). Collectively, this suggests that during the termination of the EECO, the development of a stable hydrological cycle may have been critical to the development of the OC-rich (>35%) Mahogany Zone (~49.3 to 48.7 Ma). These large highly productive saline lakes possibly acted as carbon sinks, providing a negative climate feedback mechanism during intervals between hyperthermals (Birgenheier et al., 2019).

Intriguingly, the deposition of the Mahogany Zone is roughly coincident with the onset of long-term Eocene cooling (~48–49 Ma; Inglis et al., 2015; Smith et al., 2010; Gall et al., 2017). Large amounts of organic carbon would have been sequestered in Lake Uinta (~76 Gt; Elson et al., 2022), similar to the OC-rich (>5 wt%) deposition observed in deposits from the same time period in the High Arctic (Brinkhuis et al., 2006) and Nordic Seas (Brinkhuis et al., 2006; Barke et al., 2012). This suggests a link between carbon cycling and global climate evolution. Unlike the Uinta Basin, the high Arctic is characterized by low salinity conditions. However, both are strongly stratified, largely anoxic basins (Brinkhuis et al., 2006; Vanden Berg and Birgenheier, 2017) characterized by limited siliciclastic input (Birgenheier et al., 2019). Both suggest a stable hydrological cycle during the EECO and imply a
causal relationship between the hydrological cycle, organic matter burial and carbon cycling during the early Eocene. However, additional work from other sites is required to test this hypothesis.

5. Conclusions

Here we use leaf wax and algal lipid δ²H analysis to study the hydrological regime in the mid-latitude continental interior during the latest EECO and post-EECO interval (~49.3 to 48.7 Ma). We find large inter-site variation in both leaf wax and algal lipid δ²H values. This may have arisen from various factors, including the input of δ²H-enriched, saline water from Lake Gosiute. Unlike the more variable hydrological regime of the early Eocene hyperthermals, limited variations in δ²H values during the deposition of the Mahogany Zone suggest a relatively stable hydrological cycle during the latest EECO. We interpret this to indicate that the hydrologic cycle responds differently during rapid vs. gradual climatic perturbations. A stable hydrological regime may provide conditions that promote organic matter productivity and preservation within large lacustrine systems and may serve as an important negative climatic feedback during intervals of sustained global warmth.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgements

This contribution represents a portion of A. Elson’s doctoral dissertation at the University of Southampton, funded by the Natural Environment Research Council Centre for Doctoral Training in Oil & Gas. Oliver Bench assisted with field work and sample collection and P. Sargent Bray is acknowledged for help with organic geochemistry. Discussions with Mike Vanden Berg contributed to the development of this manuscript. Funding was provided by CASP and the NSF for fieldwork and workshop attendance for A. Elson, and the Donors to the Petroleum Research Fund of the American Chemical Society to J. H. Whiteside. G. N. Inglis was funded by a GCRF Royal Society Dorothy Hodgkin Fellowship (DHF/R1/191178).

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.palaeo.2022.110959.

References


