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## Stable carbon isotopes and bulk-sediment geochemistry as indicators of relative sea-level change in tidal marshes, mangroves and isolation basins: Application and developments

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#### ARTICLE INFO

Handling editor Melanie Leng

Keywords: Relative sea-level Stable carbon isotope Salt marsh Mangrove Isolation basin

#### ABSTRACT

Bulk organic stable carbon isotope ( $\delta^{13}$ C) and accompanying bulk organic geochemical measurements have been increasingly used as a relative sea-level (RSL) indicator over the last two decades. Their utility as a RSL indicator is premised on the ability of bulk organic  $\delta^{13}$ C and bulk organic geochemistry to distinguish between organic matter (OM) sources in coastal environments, and to identify changes in OM source contributions in sediment sequences in response to RSL change. We evaluate the performance of bulk  $\delta^{13}$ C and bulk organic geochemistry as a RSL indicator in tidal marsh, mangrove and isolation basin environments. The interpretation of isotope measurements from these environments requires knowledge of the processes controlling contemporary OM  $\delta^{13}$ C, and the influence of decomposition on bulk values. We review in detail the controls on the  $\delta^{13}$ C composition of OM in tidal marshes, mangroves and isolation basins, and advocate wherever possible for the collection of contemporary geochemical datasets corrected for the <sup>13</sup>C Suess effect to help inform interpretations. From the wide range of case studies considered, an emerging principle is that the degree of isotopic distinctiveness between OM sources is key in determining how the technique can be deployed as a RSL indicator. This can range in use from the provision of qualitative information on changes in marine influence over time, to the identification of sea-level index points at lithostratigraphic contacts, and most powerfully to the recognition of inter-tidal sub-environments with isotopically well-constrained vertical limits.

#### 1. Introduction

The application of stable isotope geochemical techniques to derive Late Quaternary relative sea-level (RSL) records from coastal lowland deposits has developed over the past two decades. The potential of bulk organic stable carbon isotope ( $\delta^{13}$ C) and organic carbon/total nitrogen (C/N) ratios as paired RSL indicators was first demonstrated in UK tidal wetland sediments (Wilson et al., 2004, 2005a) and isolation basin deposits (Mackie et al., 2005). Use of  $\delta^{13}$ C and C/N analyses to derive RSL records from tidal wetlands has since undergone methodological refinement and become increasingly popular (e.g., Kemp et al., 2010; Khan et al., 2015a). Advancements include: the use of bulk organic  $\delta^{13}$ C and C/N, measured alongside total organic carbon (TOC) content, as geochemical signatures of past depositional environments (Khan et al., 2015b; Wilson, 2017); incorporating  $\delta^{13}$ C as an additional (to microfossil assemblages) proxy in Bayesian transfer functions to reduce the vertical uncertainty and improve the accuracy of reconstructed RSL (Cahill et al., 2016); developing procedures to correct for the <sup>13</sup>C Suess effect (Wilson, 2017); and assessing the potential of bulk organic  $\delta^{13}$ C, C/N and TOC as RSL indicators in mangrove environments (e.g., Khan et al., 2019). Here we explore the development and application of bulk organic  $\delta^{13}$ C and C/N in the context of RSL research, with a particular focus on tidal wetland (tidal marsh and mangrove) and isolation basin deposits. We consider the mechanisms controlling  $\delta^{13}$ C suess effect and post-deposition decomposition on bulk-sediment  $\delta^{13}$ C values.

https://doi.org/10.1016/j.quascirev.2024.108855

Received 30 April 2024; Received in revised form 18 July 2024; Accepted 22 July 2024 Available online 9 August 2024

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#### 2. Tidal wetlands and isolation basins

Tidal wetland and isolation basin deposits provide a natural archive of past movements in RSL (e.g., Barlow et al., 2013; Lloyd, 2000; Woodroffe et al., 2005). Tidal wetlands occupy sheltered, low-energy coastal environments, with salt marsh ecosystems prevailing in the temperate and high latitudes (Chapman, 1977) and mangroves prevalent in tropical and sub-tropical latitudes (Kathiresan and Bingham, 2001). Tidal wetlands receive sediment from several different sources, including riverine- and marine-derived minerogenic sediment and particulate organic matter (POM), organic matter (OM) derived from vascular vegetation growing on wetland surfaces, and biota such as bacteria inhabiting the surface sediments (Section 4.1; Khan et al., 2019; Luternauer et al., 1995). In tidal wetland deposits, the amount of OM derived from these different sources varies among sites and with tidal altitude within a site. Isolation basins are natural rock depressions that at different times may be connected to, or isolated from, the sea due to RSL change. The elevation of the basin rock sill controls the level of tidal inundation and is used to calculate the altitude of former sea level (Long et al., 2011). When isolated (during periods of low RSL), freshwater conditions prevail in the basin. When RSL rise carries high tides above the sill freshwater-brackish conditions characterise the basin, and then eventually fully marine conditions where the basin sill lies below lowest astronomical tide. During periods of RSL fall isolation basins will record the reverse trend, changing from fully marine through a brackish phase to freshwater conditions when the sill is higher than all stages of the tidal cvcle.

In tidal wetlands and isolation basins, RSL change influences the source of OM contributions. In tidal wetlands, RSL rise increases the frequency and duration of tidal flooding, and therefore greater delivery of tidal-derived POM (Section 4). In isolation basins, RSL rise causes a change in dominance from OM derived from freshwater algae and terrigenous input during isolation, to OM increasingly derived from marine POM (Section 5). OM  $\delta^{13}$ C and C/N values can vary between sources (Megens et al., 2002), allowing the provenance of OM in tidal wetland and isolation basin deposits to be determined (e.g., Chmura and Aharon, 1995; Ember et al., 1987; Haines, 1976; Mackie et al., 2005). To be of value, RSL indicators (which can include stable isotope and element geochemistry of bulk OM, as well as macro- and microfossils) should have a quantifiable relationship to a reference water level (RWL) within the tidal frame (Van de Plassche, 1986). The RWL is the elevation at which the indicator occurs, defined with reference to a tidal datum (Woodroffe and Barlow, 2015). The quantifiable relationship between the indicator and a RWL is known as the 'indicative meaning', and the elevational envelope in which the indicator occurs within the tidal frame is termed the 'indicative range'. The indicative meaning and range are determined from contemporary observation (e.g., Long et al., 1999; Shennan, 1982). RSL records generated from tidal wetland and isolation basin deposits often comprise of a series of sea-level index points (SLIPs) - dated sediment horizons with a measured altitude (relative to a local datum) and indicative meaning. In tidal marsh deposits, for example, SLIPs are traditionally derived from transgressive (organogenic to minerogenic) or regressive (minerogenic to organogenic) lithostratigraphic contacts in intercalated coastal sediment sequences (Tooley, 1982). The indicative range at lithostratigraphic contacts is minimal (typically  $\pm 20$  cm, depending on tidal range) and the availability of OM for radiocarbon dating enables lithostratigraphic contacts to be placed in a chronological framework, thus permitting the reconstruction of RSL time-altitude changes with defined temporal and vertical errors. Methodological advances have focused on developing records of tidal-marsh elevation changes through time, permitting sub-millennial scale RSL changes to be identified (Barlow et al., 2013). This approach is underpinned by the recognition of elevation-dependent zones in the sediment record. This can be achieved by establishing the presence of well-constrained elevation-dependent microflora and/or microfauna groups in contemporary tidal-marsh environments and using microfossil

transfer functions to identify equivalent depositional environments in core material (e.g., Edwards and Horton, 2000). In isolation basin deposits, SLIPs are located at 'isolation contacts', which in the sediment record are represented as changes in lithology and accompanying changes in microfossil assemblages associated with changes in water salinity (e.g., Kjemperud, 1981; Long et al., 1999) and/or stable isotope and element geochemistry (Mackie et al., 2005; Taylor et al., 2023) in response to hydrological isolation of the basin.

Fossil diatom and foraminifera assemblages preserved in tidal wetland deposits and fossil diatom, foraminifera and thecamoebian assemblages preserved in isolation basin deposits are used extensively to identify RWLs and to ensure a conformable facies sequence at lithostratigraphic and isolation contacts (for method details and example applications, see Cahill et al., 2016; Edwards and Wright, 2015; Kemp et al., 2009; Lloyd, 2000; Long et al., 1999; Zong and Sawai, 2015). These approaches are well-established, and although labour intensive, can yield detailed RSL reconstructions (e.g., Horton et al., 2006). The application of microfossils as sea-level indicators has several limitations. however. These include: (i) an assumption of unchanged optima and tolerance of tidal wetland biological indicators in relation to a RWL (Gehrels, 2002); (ii) the vertical range restriction of different tidal wetland biological indicators within the tidal frame (Gehrels et al., 2001); (iii) multiple environmental variables that may influence species distributions (Mackie et al., 2005); (iv) the susceptibility of biological indicators to chemical and mechanical damage during burial (e.g., Birks and Birks, 1980; Kato et al., 2003). The use of microfossils to quantitatively estimate past RWLs using transfer functions may suffer additional drawbacks, including the absence of modern analogues (e.g., Wilson and Lamb, 2012), problems of spatial autocorrelation (e.g., Telford and Birks, 2005) and a large amount of unexplained variability in contemporary training sets (e.g., Zong and Horton, 1999).

Geochemical RSL indicators (principally bulk OM  $\delta^{13}$ C and C/N) may be used in combination with biological indicators to provide robust records of RSL change (e.g., Kemp et al., 2013). When OM geochemical and biological indicators have been combined, an improvement in the accuracy of reconstructed RSL has been achieved (e.g., Section 4.2; Cahill et al., 2016; Kemp et al., 2013). Used in isolation, OM geochemical indicators can be a viable alternative to biological sea-level indicators (e.g., Khan et al., 2015b; Wilson, 2017) and, in the event of poor microfossil preservation, can prevent redundancy of potential SLIPs at lithostratigraphic and isolation contacts. Because OM is almost ubiquitous in tidal wetland and isolation basin sediments, and the quantity of OM required for bulk isotope and element geochemical analysis is small (typically <0.5 g dry weight), continuous palaeoenvironmental records can be derived using these techniques. Nevertheless, OM geochemical indicators also have limitations, including the ability to accurately identify palaeodepositional environments in some tidal wetland contexts, and OM degradation which can lead to shifts in bulk geochemical values (Section 6; Lamb et al., 2006). The application of OM geochemistry as RSL indicators in tidal wetlands and isolation basins is explored in Sections 4 and 5. Underpinning the application of bulk OM  $\delta^{13}$ C and C/N to indicate past RSL movements is the ability to distinguish between OM sources in tidal wetland and isolation basin deposits. The controls on, and variability in,  $\delta^{13}$ C and C/N values of OM encountered in tidal wetlands and isolation basins is explored in Sections 3, 4.1 and 5.1. While this review concentrates on the use of bulk OM  $\delta^{13}$ C and C/N analyses, other stable isotope and geochemical techniques have been used in coastal reconstruction and RSL research. For example, the stable oxygen isotope ( $\delta^{18}$ O) analysis of carbonate material from ostracods and benthic foraminifera living in isolation basins, and  $\delta^{18}$ O,  $\delta^{13}$ C and Sr/Ca analysis of ostracods in lagoon deposits, has been used to reconstruct past changes in salinity and RSL (Hammarlund, 1999; Lloyd and Evans, 2002; Marco-Barba et al., 2013). More recently, Taylor et al. (2023) investigated the potential of osmium isotopes  $(^{187}\text{Os}/^{188}\text{Os})$  from bulk sediment as a proxy for sediment provenance associated with changes in marine influence within an isolation basin

due to changes in RSL. In tidal marshes, analytical Rock-Eval pyrolysis has been investigated as a potential RSL indicator (Kemp et al., 2017a), whilst the use of biomarkers to identify mangrove sediment has been explored by Sefton et al. (2024). For more details of these techniques, see the above references.

## 3. $\delta^{13}$ C and C/N

## 3.1. $\delta^{13}C$ measurement and notation

Carbon has two stable isotopes (Nier, 1950). Carbon-12 (<sup>12</sup>C) is the most abundant (98.89%) with carbon-13 (13C) present in smaller amounts (1.11%). Measurements of isotopic content require the conversion of the material being analysed to CO<sub>2</sub> prior to measurement in a mass spectrometer. The ratio (R) of  ${}^{13}CO_2/{}^{12}CO_2$  in a sample is then measured and expressed relative to an internationally agreed reference standard. The primary reference standard is CO<sub>2</sub> obtained from the calcium carbonate from the rostrum of a Cretaceous belemnite, Belemnitella americanum, an extinct marine cephalopod from the Peedee formation in South Carolina (Craig, 1957), and referred to as 'PDB'. The primary reference standard material has long been exhausted and has been replaced by the Vienna Peedee Belemnite (VPDB) reference standard (Coplen, 1994). The VPDB isotope reference defines the zero point of the delta ( $\delta$ ) scale. Abundances of stable isotopes are generally expressed in parts per thousand (% or per mille) difference from the standard (Equation (1)). Therefore, VPDB has a  $\delta^{13}$ C value of 0%; a  $\delta^{13}$ C sample value of -1% indicates that the sample has 1% less of the heavier <sup>13</sup>C isotope relative to VDPB, whereas a  $\delta^{13}$ C value of +1% indicates that the sample has 1‰ more of the heavier <sup>13</sup>C isotope than VDPB.

$$\delta^{13}C = \left[ \left( R_{\text{sample}} / R_{\text{standard}} \right) - 1 \right] \times 1000$$
 (Equation 1)

where  $R_{(Sample)}$  and  $R_{(Standard)}$  is the ratio  ${}^{13}CO_2/{}^{12}CO_2$  measured by mass spectrometery of the sample and standard (VPDB), respectively.

## 3.2. $\delta^{13}C$ fractionation

Organic  $\delta^{13}C$  values reflect primarily the dynamics of carbon assimilation during photosynthesis and the isotope composition of the carbon source (e.g., Hayes, 1993; Meyers, 1997). Carbon isotopes are fractionated during photosynthesis (Park and Epstein, 1960). The amount of fractionation can be determined by calculating the difference between the isotopic value of the inorganic carbon (IC) source and the product (organic material) (O'Leary et al., 1986). Expressed in  $\delta$  notation relative to VPDB, the IC source value for terrestrial vascular vegetation (atmospheric CO<sub>2</sub>) is currently c.-8.5‰ (2021 Antarctic air sample values: NOAA Global Monitoring Laboratory, https://gml.noaa. gov), whilst vascular plant OM has  $\delta^{13}$ C values between -37% and -9‰ (Deines, 1980; Kohn, 2010, Table 1) depending on the photosynthetic pathway used for carbon fixation. The depletion in <sup>13</sup>C relative to the source IC is due to kinetic isotope effects during diffusion of CO2 in air through the stomatal pore (Craig, 1954) and discrimination against <sup>13</sup>C during the initial carboxylation reaction (Farquhar et al., 1982) resulting in concentration of the light isotope (<sup>12</sup>C) in the synthesised organic material. Vascular plants that utilise the C<sub>3</sub> photosynthetic pathway have  $\delta^{13}$ C values that generally range between c. -37‰ and -20% (Deines, 1980; Kohn, 2010), with globally-averaged C<sub>3</sub> plant biomass  $\delta^{13}C$  values estimated to be between -28.5% and -27.0%(Kohn, 2010; O'Leary, 1988). The discrimination against <sup>13</sup>C largely occurs during carboxylation of ribulose bisphosphate via the enzyme Ribulose 1,5- bisphosphate carboxylase/oxygenase (RuBisCO). This enzyme is the first step in CO<sub>2</sub> fixation (Park and Epstein, 1960) and causes a carbon isotope fractionation of 29‰ (e.g., Roeske and O'Leary, 1984). Variable rates of diffusion and carboxylation will jointly limit the uptake of CO<sub>2</sub> in C<sub>3</sub> plants, leading to a range in C<sub>3</sub> plant  $\delta^{13}$ C values

#### Table 1

Inorganic carbon source, organic matter and bulk organic sediment  $\delta^{13}$ C value ranges reported in the literature from selected environments. Note that ranges are inherently the extreme ends of the scale and there is often less overlap in typical  $\delta^{13}$ C values between the listed environments. See Sections 3.2, 4.1, and 5.1 for further details, typical range values and spatial variability in tidal wetland, estuarine and isolation basin settings. References: <sup>1</sup>2021 Antarctic air samples (NOAA Global Monitoring Laboratory, http://gml.noaa.gov);<sup>2</sup>Rubino et al. (2013);<sup>3</sup>Francey et al. (1999);<sup>4</sup>Schmitt et al. (2012);<sup>5</sup>Ge et al. (2022);<sup>6</sup>Gruber et al. (1999);<sup>7</sup>Quay et al. (2003);<sup>8</sup>Filipsson et al. (2017);<sup>9</sup>Papadimitriou et al. (2005);<sup>10</sup>He and Xu (2017);<sup>11</sup>Hellings et al. (1999);<sup>12</sup>Ray et al. (2018);<sup>13</sup>Samanta et al. (2015);<sup>14</sup>Bade et al. (2004) \*;<sup>15</sup>Keeley and Sandquist (1992)\*;<sup>16</sup>Finlay (2003);<sup>17</sup>Mook and Tan (1991);<sup>18</sup>Raymond et al. (2004);<sup>19</sup>Sackett and Moore (1966);<sup>20</sup>Tan (1989);<sup>21</sup>Tan and Edmond (1993);<sup>22</sup>Deines (1980)\*;<sup>23</sup>Kohn (2010)\*;<sup>24</sup>O'Leary (1988);<sup>25</sup>Bouillon et al. (2004);<sup>26</sup>Chmura and Aharon (1995)\*;<sup>27</sup>Fry et al. (1977);<sup>28</sup>Hemminga and Mateo (1996)\*;<sup>29</sup>Kennedy et al. (2010)\*;<sup>30</sup>Khan et al. (2019);<sup>31</sup>Mackie et al. (2005);<sup>32</sup>Raven et al. (2002)\*;<sup>33</sup>Smeaton et al. (2022);<sup>34</sup>Velázquez-Ochoa et al. (2022);<sup>35</sup>Cloern et al. (2002);<sup>36</sup>France (1995) \*,<sup>37</sup>Goericke and Fry (1994)\*,<sup>38</sup>Taipale et al. (2016);<sup>39</sup>Kemp et al. (2010);<sup>40</sup>Khan et al. (2015b)\*;<sup>41</sup>Wilson et al. (2005b);<sup>42</sup>Chmura et al. (1987);<sup>43</sup>Kemp et al. (2012);<sup>44</sup>Andrews (2008);<sup>45</sup>Weiss et al. (2016);<sup>46</sup>Barber et al. (2017);<sup>47</sup>Bauer et al. (2002);<sup>48</sup>Fry et al. (1998);<sup>49</sup>Han et al. (2022);<sup>50</sup>Lamb et al. (2006)\*;<sup>51</sup>Lewis et al. (2024);<sup>52</sup>Yamashita et al. (2015);<sup>53</sup>Goñi et al. (2003);<sup>54</sup>Peterson et al. (1994);<sup>55</sup>Wang et al. (2012);<sup>56</sup>Bade et al. (2007);<sup>57</sup>Havas et al. (2023);<sup>58</sup>Bouillon et al. (2007);<sup>59</sup>Liu et al. (2022);<sup>60</sup>Nakatsuka et al. (2004);<sup>61</sup>Sackett et al. (1965);<sup>62</sup>Verwega et al. (2021) \*;<sup>63</sup>Holding et al. (2017);<sup>64</sup>Sepúlveda et al. (2011)\*;<sup>65</sup>McGovern et al. (2020);<sup>66</sup>Gearing et al. (1984);<sup>67</sup>Middelburg and Herman (2007);<sup>68</sup>Yu et al. (2010);<sup>69</sup>Hebbeln et al. (2000);<sup>70</sup>Hunt (1970);<sup>71</sup>Naidu et al. (2000)\*;<sup>72</sup>Kim et al. (2020);<sup>73</sup>Kumar et al. (2016);<sup>74</sup>Smeaton and Austin (2022);<sup>75</sup>Shultz and Calder (2020), remain et al. (2010), sincation and retain (2022), sincation and retain (2022), sincation and retain (2010), reference of the relation of t (2007);<sup>85</sup>Middelburg et al. (1997);<sup>86</sup>Bouillon et al. (2003);<sup>87</sup>Kemp et al. (2019);<sup>88</sup>Manju et al. (2016);<sup>89</sup>Sanders et al. (2010);<sup>90</sup>Tue et al. (2011);<sup>91</sup>Wilson (2017)\*;<sup>92</sup>Wilson et al. (2005a);<sup>93</sup>Mackie et al. (2007);<sup>94</sup>Watcham et al. (2011);<sup>95</sup>Stearns et al. (2023);<sup>96</sup>Kemp et al. (2013). \*Includes compilations of existing data.

	$\delta^{13}$ C (‰)	Reference			
Source inorganic carbon					
Atmospheric CO <sub>2</sub>					
Contemporary	-8.5	1			
Pre-industrial	$-6.5^2$ , $-6.4^3$	2, 3			
Late Holocene	-6.35	4			
Early Deglacial (16–12 ka BP)	-6.65	4			
Last Glacial Maximum (24–19ka BP)	-6.45	4			
Dissolved Inorganic					
Carbon (DIC)					
Marine (surface ocean)	-0.17 to +2.4 (global average <sup>7</sup> : +1.51)	5–7			
Fjord	-5.4 to +0.8	8–9			
Estuarine	-20.7 to 0	10-13			
Freshwater (lake)	-31 to +2.6	14–15			
Freshwater (river)	-26 to 0	10, 15-21			
Photoautotrophs					
C <sub>3</sub> Plants	-37 to -20 (global average <sup>23,24</sup> : -28.5 to -27)	22–24			
C <sub>4</sub> Plants	-17 to -9	22			
CAM Plants	-31 to -11	22			
Marine macrophytes	-34.6 to -2.2	9, 25-34			
Freshwater macrophytes	-50 to -11	15, 31			
Phytoplankton	-45 to -9	22, 26, 35-38			
Tidal wetland vegetation					
Salt marsh (C <sub>3</sub> )	-31.3 to -22.1	26, 33, 35, 39-41			
Salt marsh (C <sub>4</sub> )	-16.9 to -11.7	26, 33, 35, 39, 40,			
		42, 43			
Reed swamp	-30.7 to -24.6	26, 35, 40, 43			
Wet woodland (Fen carr)	-34.8 to -26.6	40, 44			
Mangrove	-32.7 to -24.1	25, 30, 45			
	(	continued on next page)			

#### Table 1 (continued)

	δ <sup>13</sup> C (‰)	Reference		
Dissolved Organic Carbon (DOC)				
Marine	-25.4 to -18.8	46–51		
Fjord	-31.8 to -21.3	46, 52		
Estuarine	-30.56 to -22	10, 12, 46, 53, 54		
River	-32.1 to -23.1	10, 18, 50, 55,		
Lake	-29.3 to -8.7	56, 57		
Particulate Organic Carbon (POC)				
Marine	-35 to $-9$ (global average <sup>62</sup> .	22 35-37 47		
	-23.96)	58-61, see also 62		
Fiord	-29.3 to $-18.3$	63. 64		
Estuarine	-41.8 to -19	11–13, 17, 35, 66– 68		
Freshwater	-45 to -21	17, 18, 21, 35, 36, 50, 56, 58, 68		
Sedimentary Organic Matter				
Bulk surface sediment				
Marine	-27.6 to $-16.0$	58, 66, 69-71		
Fjord	-29.69 to -19.1	64, 72-74		
Estuary	-26.6 to -17.7	53, 66, 68, 75, 76		
River	-30.1 to -24.2	53, 64, 70, 71		
Isolation basin	-29.3 to -20.8	31		
Tidal Flat	-25.53 to -19.2	41, 77, 78, 79, 80,		
		81, 91		
Saltmarsh ( $C_3$ veg. dominance)	-29.5 to -19.84	39-42, 77-84, 91		
Saltmarsh ( $C_4$ veg. dominance)	-20.4 to -14.1	39, 42, 43, 85, 95		
Mangrove	-30 to -20.6	25, 30, 45, 86–90,		
Seagrass	-26.6 to -7.3	25, 27, 29		
Buried sediment (Late Quaternary)				
Tidal marsh (native C <sub>3</sub> veg.)	-31.44 to -15.86	33, 78, 82, 84, 91, 92		
Tidal marsh (native C <sub>3</sub> and C <sub>4</sub> yeg.)	-29.5 to -13.1	43, 79, 95, 96		
Mangrove	-28.1 to $-18.5$	30, 87		
Isolation basin	-30 to -16	31, 93, 94		
		, >0, > ,		

(O'Leary, 1988). For example, changes in plant stomatal aperture mediates CO<sub>2</sub> diffusion and can lead to variations in internal CO<sub>2</sub> concentrations and plant  $\delta^{13}$ C values (Farguhar et al., 1982); this may arise in response to drought stress (Farquhar et al., 1989) and salinity stress (Seeman and Critchley, 1985). Plants utilising the C<sub>4</sub> photosynthetic pathway have  $\delta^{13}$ C values of around -13% (range = -17% to -9%; Deines, 1980), principally reflecting the smaller isotope fractionation by the enzyme phosphoenolpyruvate carboxylase and the fractionation associated with diffusion (Farguhar, 1983; O'Leary et al., 1992). The Crassulacean acid metabolism (CAM) cycle occurs mostly in succulents. CAM plants can incorporate IC using both the C3 photosynthetic pathway and a C4-like pathway, typically resulting in a wide range of  $\delta^{13}$ C values (-31‰ to -11‰; Deines, 1980; Schleser, 1995), depending on which pathway is dominant. In addition to physiological and ambient environmental conditions, genetic differences between species and annual cycles of plant growth and senescence (Cloern et al., 2002) can also lead to plant  $\delta^{13}$ C variability.

Aquatic plants and photosynthetic algae utilise dissolved inorganic carbon (DIC) as the IC source. Total DIC (also referred to as  $\sum$ CO<sub>2</sub>) comprises aqueous CO<sub>2</sub> (CO<sub>2(aq)</sub>), bicarbonate (HCO<sub>3</sub><sup>-</sup>) and carbonate (CO<sub>3</sub><sup>2-</sup>). At pH values of 6 and below CO<sub>2(aq)</sub> is the dominant IC species, with HCO<sub>3</sub><sup>-</sup> dominating at pH 7–9, and CO<sub>3</sub><sup>2-</sup> dominating above pH 9.5. Carbon isotopes fractionate between solid, dissolved and gaseous phases. The equilibrium isotope fractionation effect for each DIC species with respect to gaseous CO<sub>2</sub> (CO<sub>2(g)</sub>) is detailed in Zhang et al. (1995); see also Yumol et al. (2020) for kinetic isotope effects. The equilibrium isotope fractionation effect values ( $\epsilon$ ), which are

temperature-dependent, have been measured at -1.29% to -1.19%between aqueous and gaseous CO<sub>2</sub>, 7.9% to 10.2% between HCO<sub>3</sub> and  $CO_{2(g)}$  (i.e.  $HCO_3^-$  is enriched in <sup>13</sup>C relative to  $CO_{2(g)}$ ), and 6% to 7% between  $CO_3^{2-}$  and  $CO_{2(g)}$  (Zhang et al., 1995; measured over a temperature range of 5°C–25°C). Based on current atmospheric CO<sub>2</sub>  $\delta^{13}$ C values (-8.5‰), expected  $\delta^{13}$ C values for CO<sub>2(aq)</sub> in air-equilibrated water are c. -9.5%, and c. -0.5% for HCO<sub>3</sub>. It follows that photosynthetic algae and submerged aquatic plants utilising  $HCO_3^-$  as the IC source can have  $\delta^{13}$ C values several per mille higher than photoautotrophs utilising CO<sub>2</sub>. Most species of submerged aquatic plants and algae use the C<sub>3</sub> photosynthetic pathway (Falkowski and Raven, 2007). In the species analysed to date, most algae can utilise dissolved CO<sub>2</sub> and HCO<sub>3</sub> as the IC source (Giordano et al., 2005) and fix IC (in the form of CO<sub>2</sub>) via RuBisCO. Carbon concentration mechanisms (CCMs) are widespread in aquatic photoautotrophs (Falkowski and Raven, 2007). The diffusion coefficient of  $CO_2$  in water is  $10^4$  times slower than that in the atmosphere (Falkowski and Raven, 2007). Furthermore, RuBisCO has a low affinity for its substrate CO<sub>2</sub> and evolved at a time when atmospheric CO<sub>2</sub> concentrations were much higher than present levels (Raven et al., 2017). CCMs actively transport  $CO_2$  and/or  $HCO_3^-$  from the surrounding water into cells; where  $HCO_3^-$  is the IC source, intracellular carbonic anhydrase can rapidly convert HCO<sub>3</sub><sup>-</sup> to CO<sub>2</sub>. CCMs raise the concentration of CO<sub>2</sub> at the site of carboxylation, thus increasing the rate of photosynthesis in environments where CO2 concentrations are low (Badger et al., 1980). Diffusive CO<sub>2</sub> intake occurs in a very small fraction of marine photoautotrophs (e.g. Velázquez-Ochoa et al., 2022) and in a slightly larger fraction of freshwater photoautotrophs (Falkowski and Raven, 2007). CCM fixation of IC tends to reduce discrimination against <sup>13</sup>C (Giordano et al., 2005).

The  $\delta^{13}$ C values of aquatic photoautotrophs vary considerably (Table 1) due to environmental and physiological factors, including: DIC source (CO<sub>2</sub> or HCO<sub>3</sub><sup>-</sup>); CO<sub>2</sub> concentration (Burkhardt et al., 1999; Laws et al., 1995); temperature (Liu et al., 2022; Sackett et al., 1965); thickness of the stagnant boundary layer (France, 1995); growth rate (Burkhardt et al., 1999); cell physiology (e.g. IC acquisition via diffusion or via a CCM (e.g., Velázquez-Ochoa et al., 2022); RuBisCO form (Brandenburg et al., 2022; Tabita et al., 2008)). Marine phytoplankton can have a wide range of  $\delta^{13}$ C values (-35‰ to -16‰; Deines, 1980; France, 1995; Guericke and Fry, 1994; Sackett et al., 1965) or sometimes greater (Deines, 1980; Falkowski and Raven, 2007, Table 1), although frequently fall within a narrower range of between -22% and -18%(Falkowski and Raven, 2007; France, 1995; Goericke and Fry, 1994; Hunt, 1970) which is several per mille higher than typical  $\delta^{13}$ C values for C<sub>3</sub> vascular vegetation (Deines, 1980). This difference in typical  $\delta^{13}$ C values reflects the contrasting IC source values and IC acquisition modes in aquatic and terrestrial photoautotrophs. HCO<sub>3</sub><sup>-</sup> is the dominant DIC source for marine photoautotrophs (at current ocean pH levels of 8.1, 98% of DIC is in the form of HCO<sub>3</sub><sup>-</sup>). DIC  $\delta^{13}$ C values of the world oceans is between -0.5% and +2.4% (Kroopnick, 1985; Quay et al., 2003), which is much higher than terrestrial vascular vegetation IC source values (present atmospheric CO<sub>2</sub>  $\delta^{13}$ C = c. -8.5‰). This difference in  $\delta^{13}$ C values of marine phytoplankton and terrestrial vascular vegetation permits the use of  $\delta^{13}$ C to determine OM provenance (Lamb et al., 2006). Marine macrophyte  $\delta^{13}$ C values range between -35% and -2% (Raven et al., 2002; Velázquez-Ochoa et al., 2022, Table 1). At the lower end of this range (<-30‰) are photoautotrophs which rely on dissolved CO<sub>2</sub> entering cells via diffusion (Maberly et al., 1992; Raven et al., 2002; Velázquez-Ochoa et al., 2022), as found in specimens of marine red macroalgae occupying sub-tidal and shaded inter-tidal habitats, for example (Raven et al., 2002). Much higher  $\delta^{13}$ C values (>-10‰) have been measured in specimens of seagrass and in specimens of brown, red and green macroalgae; their high  $\delta^{13}$ C values likely reflect HCO<sub>2</sub> uptake as part of a CCM (Raven et al., 2002). Marine macrophytes with intermediate  $\delta^{13}$ C values reflect a mixture of CCM active uptake of HCO<sub>2</sub><sup>-</sup> and diffusive uptake of CO<sub>2</sub> (Raven et al., 2002; Velázquez-Ochoa et al.,

2022). Freshwater phytoplankton have  $\delta^{13}$ C values ranging between -45% and -21% (Table 1), with a mean value of -32% (SD  $\pm 3\%$ ) (France, 1995). At pH < 6, dissolved CO<sub>2</sub> becomes the more important IC source.  $CO_{2(aq)}$  has a  $\delta^{13}$ C value of around -9.5% in air-equilibrated water, which is only slightly lower than atmospheric  $CO_2 \delta^{13}C$  values. As a result, there is considerable overlap in the  $\delta^{13}$ C values of C<sub>3</sub> vascular plants and C<sub>3</sub> phytoplankton (Lamb et al., 2006).  $\delta^{13}$ C values for freshwater aquatic plants vary between -50% and -11% (Keeley and Sandquist, 1992). This wide variation reflects previously discussed environmental and physiological factors, including the presence of C<sub>3</sub>, C4 and CAM photosynthetic pathways (Keeley and Sandquist, 1992). DIC  $\delta^{13}$ C ( $\delta^{13}$ C<sub>DIC</sub>) values as low as -31‰ (Bade et al., 2004, Table 1) have been observed in freshwater environments. Such low  $\delta^{13}C_{DIC}$ values most likely reflect the dominance of IC derived from respiration of autochthonous aquatic photoautotrophs or receipt of allochthonous IC derived from decomposition of terrestrial detritus. Uptake of this 'recycled' CO<sub>2</sub> by aquatic photoautotrophs can result in extremely low  $\delta^{13}$ C values.

## 3.3. Temporal changes in $CO_2 \delta^{13}C$ and the <sup>13</sup>C Suess effect

Changes in IC source values will in turn influence terrestrial and aquatic photoautotroph  $\delta^{13}$ C values. Of temporal relevance to  $\delta^{13}$ Cbased RSL reconstruction in tidal wetland and isolation basin deposits is changes in Late Quaternary atmospheric CO<sub>2</sub>  $\delta^{13}$ C values. Polar ice core records reveal similar atmospheric CO<sub>2</sub>  $\delta^{13}$ C values during the Last Glacial Maximum (mean atmospheric CO<sub>2</sub>  $\delta^{13}$ C value of -6.45‰ between 24 and 19 ka BP) and during the late Holocene (mean atmospheric CO<sub>2</sub>  $\delta^{13}$ C value of -6.35‰), with slightly lower  $\delta^{13}$ C values during the early deglacial (atmospheric CO<sub>2</sub>  $\delta^{13}$ C value of c. -6.65‰ between 16 and 12 ka BP) (Schmitt et al., 2012). More recently, fossil fuel burning and deforestation has introduced <sup>12</sup>C-enriched CO<sub>2</sub> into the atmosphere, which decreased atmospheric CO<sub>2</sub>  $\delta^{13}$ C values from a pre-industrial average of -6.5% (Rubino et al., 2013) to approximately -8.5% today. The fall in atmospheric CO<sub>2</sub>  $\delta^{13}$ C values during the industrial period is called the <sup>13</sup>C Suess effect (Keeling et al., 1979). Air-water gas exchange resulted in an associated decrease in  $\delta^{13}$ C-DIC values in the ocean mixed layer (Quay et al., 1992). The influence of the  $^{13}$ C Suess effect on ocean  $\delta^{13}$ C-DIC values is spatially variable (unlike the atmospheric effect); as of 2018 globally-averaged surface ocean  $\delta^{13}\text{C-DIC}$  values may have declined by  $-1.1\pm0.2\text{\%}$  relative to 1780 CE (Kwon et al., 2022). This reduction in IC source  $\delta^{13}$ C values is expressed in the  $\delta^{13}$ C values of terrestrial and aquatic photoautotrophs (e.g., McCarroll and Loader, 2004; McMahon et al., 2015; Schelske and Hodell, 1995; Verburg, 2007; Zimmerman and Canuel, 2002). It follows that contemporary tidal wetland and isolation basin bulk OM sediment  $\delta^{13}$ C values could be up to c. 2‰ lower (based on the date of measurement) than pre-industrial equivalent samples. Corrections for the <sup>13</sup>C Suess effect are applied where contemporary  $\delta^{13}$ C values are used as modern analogues of past environments (e.g., Stearns et al., 2023; Wilson, 2017) or processes (e.g., Verburg, 2007).

Procedures to correct post-industrial tidal wetland bulk sediment  $\delta^{13}$ C values for the  $^{13}$ C Suess effect are detailed in Wilson (2017), and this approach is also applicable to isolation basin sediments. Correction for the  $^{13}$ C Suess effect is achieved by adjusting the post-industrial tidal wetland or isolation basin bulk OM  $\delta^{13}$ C sample value (termed  $\delta^{13}$ C<sub>ad-justed</sub>) to account for the difference between the atmospheric CO<sub>2</sub>  $\delta^{13}$ C value at the time of sample collection (derived from instrumental air sample and Antarctic ice core records: Francey et al. (1999)) and a pre-industrial standard value of -6.4% (McCarroll and Loader, 2004). Because surface sediment samples accrete over several years (depending on sample thickness and site sedimentation rate), a decade-average  $\delta^{13}$ C value can be calculated to account for this. For example, Stearns et al. (2023) adjusted contemporary tidal marsh bulk OM  $\delta^{13}$ C measurements by adding 1.86‰ (the decadal-averaged correction for 2010–2020 CE during which the contemporary samples were collected). Correction for

the <sup>13</sup>C Suess effect is expected to improve the analogy between  $\delta^{13}$ C values measured in modern and fossil samples. This correction is premised on the assumption that bulk OM in the modern sample is contemporary, and thus contains recently sequestered carbon (e.g., OC derived from the overlying vegetation, from aquatic photoautotrophs, and/or from contemporary organic detritus introduced by tidal inundation). The contemporary bulk OM sample may also contain tidal-derived refractory OM that is millennia old (Komada et al., 2022; Van de Broek et al., 2018). Where tidal-derived OM is expected to be a major component of the sample (e.g., lower inter-tidal areas of minerogenic tidal marshes), a<sup>13</sup>C Suess effect correction may be deemed either as unnecessary or may be viewed as introducing a source of bias if applied. However, our recommendation is to apply a<sup>13</sup>C Suess effect correction to post-industrial samples for the following reasons. Firstly, the relative proportion of recent OM and old (pre-industrial) recalcitrant OM in contemporary samples is a source of uncertainty and will vary among sites and with tidal altitude within a site (Section 4.1; Komada et al., 2022; Van de Broek et al., 2018). Secondly, in tidal wetlands, minerogenic lower inter-tidal samples (which would be expected to contain a higher proportion of tidal-derived POM, including an unknown quantity of millennia-old POM) typically have higher  $\delta^{13}$ C values than samples derived from organogenic upper inter-tidal and supra-tidal areas (Section 4.1). Whilst applying a correction could potentially introduce bias (an 'erroneous'  $\delta^{13}C_{adjusted}$  value of up to +2% higher), it will not result in sample convergence of  $\delta^{13}$ C values from contrasting environments. Finally, as the <sup>13</sup>C Suess effect continues (Kwon et al., 2022), the  $\delta^{13}$ C 'signature' values of fossil and modern-equivalent samples will be increasingly dissimilar as time progresses.

#### 3.4. C/N variability

The weight of nitrogen and organic carbon is normally measured alongside  $\delta^{13}$ C and can provide an additional indication of the relative contributions of OM from contrasting sources (often quantified as C/N). The weight ratio can be multiplied by 1.167 (the ratio of the atomic weights of nitrogen and carbon) to convert into atomic ratios although the ratio used is not always stated in the literature. Here, C/N refers to the weight ratio unless stated otherwise. Terrestrial vegetation normally has relatively high C/N ratios (range of 20-1340; Tyson, 1995) as it is composed predominantly of lignin and cellulose, which are nitrogen poor. Algae, which tends to be nitrogen rich (molar ratio of C:N:P of 106:16:1 among marine plankton (Redfield et al., 1963)), has relatively low C/N ratios (<10; Meyers, 1994) reflecting the absence of cellulose and a higher protein content (mean protein content of 35%) compared with vascular plants (mean protein content of 5%) (Romankevich, 1990). Bacteria contain about 15 wt% protein (Brock and Madigan, 1991) and typically have low C/N ratios of 4-6 (Tyson, 1995). Bulk sediment C/N values can be influenced by particle size, whereby bulk organic C/N ratios in fine-grained sediment deposits are often lower than those in coarse-grained deposits (Keil et al., 1994; Monrozier et al., 1981; Prahl, 1994). Coarse-grained sediment deposits can contain a larger proportion of intact terrestrial plant detritus, which will elevate bulk sediment C/N ratios. In fine-grained deposits, the higher proportion of clay minerals and associated ammonia adsorption can lead to lower bulk sediment C/N ratios. Hydrodynamic sorting of sediment by grain size can therefore influence C/N ratios and, by extension, potentially bias OM source assignments (Meyers, 1994).

## 4. Geochemical RSL indicators in tidal wetland environments

## 4.1. Controls on $\delta^{13}C$ and C/N in tidal wetland environments

Tidal wetland surface sediment bulk organic  $\delta^{13}$ C and C/N values are influenced by (i) the  $\delta^{13}$ C and C/N values of autochthonous OM sources (principally the overlying marsh or mangrove vegetation, but also edaphic algae and bacteria); (ii) allochthonous OM (principally tidal-

derived POM); and (iii) decomposition. At the landward limit of tidal wetlands, infrequent tidal inundation allows for extensive vegetation cover, resulting in organic-rich surface sediments. In contrast, tidal flats are frequently inundated for lengthy periods, vascular vegetation is largely absent, and allochthonous, tidal-derived minerogenic and POM, dominate the surface sediments. The sediment composition of tidal wetland surfaces is therefore primarily determined by wetland surface elevation within the tidal frame (Allen, 1990), with the relative contribution of allochthonous and autochthonous material changing in response to changes in the frequency and duration of tidal inundation (Adame et al., 2010; Wilson, 2017). These two contrasting sources of (autochthonous vascular vegetation and allochthonous OM tidal-derived POM) can have distinctly different  $\delta^{13}$ C and C/N compositions (Sections 4.1.1 and 4.1.2; Lamb et al., 2006). Measurement of  $\delta^{13}$ C and C/N in bulk sediments should, theoretically, directly reflect the relative contribution of organic carbon (OC) originating from these sources (Fry and Sherr, 1989). Consequently, bulk organic  $\delta^{13}$ C and C/N analysis of tidal wetland deposits can be used to determine changes in the relative contribution of OC source over time, which will vary as a function of RSL change, for example (Wilson et al., 2005a). Sediments accumulating at similar elevational zones with respect to the tidal frame can have  $\delta^{13}$ C and bulk geochemical values that differ among sites owing to variability in physiographic conditions (e.g., Engelhart et al., 2013). The relative scale of within and among site variability should be considered in designing a sampling regime to quantify the relationship between elevation and sea-level indicators.

#### 4.1.1. Autochthonous organic carbon

Marsh and mangrove vegetation is an important source of OC in tidal wetland sediments (e.g., Bouillon and Boschker, 2006; Bouillon et al., 2003; Chmura et al., 1987). The tidal elevation of the wetland surface and consequent frequency and duration of tidal submergence is a key control governing vegetation density and species distributions, which can result in clearly defined vegetation zones (Crase et al., 2013; Gray, 1992). Although there is geographical variability in the distribution of salt marsh and mangrove plant species (e.g., Adam, 1978; Tomlinson, 1986), vegetation belonging to a few cosmopolitan genera are common to these tidal wetlands (Chapman, 1977; Ricklefs and Latham, 1993). For example, in western and northern Europe tidal flats are locally covered by seaweeds and eelgrass (Zostera), with terrestrial halophytes (grasses, sedges, rushes, and succulents) colonising surfaces above the mean high water neap (MHWN) tide level. Species of Salicornia and Spartina typically characterise the low inter-tidal pioneer communities, with Puccinellia maritima and Halimione portulacoides commonly found in low marsh areas between mean high water (MHW) and mean high water spring (MHWS) tidal levels. Closed vegetation cover is found on upper marsh areas above the MHWS tide level, and these areas may be colonised by a mixture of halophytic and non-halophytic species. Characteristic taxa include Armeria maritima, Festuca rubra and Juncus gerardi (Beeftink, 1977). Similarly, in eastern North American tidal marshes, Spartina alterniflora typically dominates low marsh communities (Reimold, 1977), with Spartina patens, Distichlis spicata, Juncus roemerianus and Juncus gerardi often found in higher marsh areas (Chmura and Aharon, 1995). In (sub)tropical regions, mangrove vegetation communities occupy a similar environmental niche as salt marshes do in temperate climates (Saintilan et al., 2014). Distinctive mangrove taxa are found in Atlantic-East Pacific (North and South America and western Africa) and Indo-West Pacific (east coast of Africa, Asia, and Oceania) regions (Duke, 1992). In the Atlantic-East Pacific, Rhizophora mangle typically occupies the low elevation mangrove fringe, and Avicennia germinans and Laguncularia racemosa dominate inland forested basins not regularly flushed by high tides (Lugo and Snedaker, 1974). In comparison, Indo-West Pacific mangroves exhibit greater species diversity and regional variability in species zonation (Duke, 1992). The hydrogeomorphic classification recognised in the Atlantic-East Pacific is less distinguishable in the Indo-West Pacific due

to the complex structure of its mangrove forests (Woodroffe et al., 2016). Tidal marsh and mangrove isotope mixing models show that surface sediment bulk organic  $\delta^{13}$ C values are often strongly influenced by the  $\delta^{13}$ C values of the overlying vegetation (Bouillon et al., 2003; Chmura and Aharon, 1995; Malamud-Roam and Ingram, 2001). In tidal marsh areas dominated by C<sub>4</sub> plants (e.g., S. alterniflora, S. patens, *D. spicata*) observed bulk organic  $\delta^{13}$ C values of the underlying surface sediment are typically > -20% (Chmura and Aharon, 1995), whilst in C<sub>3</sub> vegetated tidal marshes and mangroves, bulk organic  $\delta^{13}$ C values of the underlying surface sediment are usually <-20% (Wilson, 2017; Xia et al., 2015, Table 1). This difference reflects the contrasting isotopic value of the source OM. Tidal wetland bulk C/N values reflect the proportion of OM derived from vascular vegetation (which typically have C/N ratios >20) and algae and bacterial (C/N ratios <10) (Section 3.4). As such, bulk surface sediment C/N ratios tend to be higher in upper inter-tidal and supra-tidal zones compared with lower inter-tidal and sub-tidal zones (e.g., Wilson, 2017).

The presence of large quantities of bacteria and edaphic algae can also influence tidal wetland surface bulk organic  $\delta^{13}$ C and C/N values (Peterson et al., 1980). Bacteria are an abundant component of coastal microbial communities; fungi also populate salt marsh sediments but can be orders of magnitude less abundant than bacteria in these systems (Leadbeater et al., 2021). Bouillon and Boschker (2006) examined bacterial  $\delta^{13}$ C values in sediments from a range of coastal settings, including C<sub>3</sub> and C<sub>4</sub> salt marshes and mangroves, and found large variability in bacteria  $\delta^{13}$ C (c. -35% to -14%). This illustrates that in most settings sedimentary bacteria may depend on carbon from various origins, although they found that tidal-derived carbon often formed a significant source, with local macrophyte production becoming the major bacterial carbon source in salt marsh and mangrove environments where the OC content of the sediment was >10 wt%. The algal flora of tidal wetlands can be diverse and the  $\delta^{13}$ C values of algae wide ranging (e.g., between -30.9‰ and -4.9‰; Chmura and Aharon, 1995). Freshwater algae in C<sub>3</sub>-dominated environments tend to have lower  $\delta^{13}$ C values (-30% to -26%; Meyers, 1994; Schidlowski et al., 1983) than marine algae (-23% to -16%; Haines, 1976; Meyers, 1994) but algae in C<sub>4</sub>-vegetated marshes can have relatively high  $\delta^{13}$ C values (<16‰) (Chivas et al., 2001). Compared with vascular plants, algae contain fewer refractory compounds and will therefore decompose more rapidly (Lamb et al., 2006).

#### 4.1.2. Allochthonous organic carbon

Tidal-derived POM is a major source of allochthonous OC delivery to tidal wetlands (Bouillon and Boschker, 2006; Lamb et al., 2006; Wilson, 2017). In estuarine waters, suspended and deposited POM can potentially contain terrestrial OM (plant detritus and eroded soil), phytoplankton, zooplankton, and heterotrophic bacteria (Lamb et al., 2006). Estuarine suspended and surface sediment particulate organic carbon (POC)  $\delta^{13}$ C ( $\delta^{13}$ C<sub>POC</sub>) values (see Table 1 for range) have been found to increase systematically towards the mouth of an estuary. For example, in the Mersey and Tay estuaries (UK),  $\delta^{13}C_{POC}$  values increase from -27.2‰ and -26.2‰ in the inner estuary to -22.0‰ and -23.2‰ in the outer estuary, respectively (Thornton and McManus, 1994; Wilson et al., 2005b). Similarly, in the Pearl River estuary (China),  $\delta^{13}C_{POC}$ values increase from a freshwater environment average value of -25.0% to an average value of -21.0% in the marine environment (Yu et al., 2010). Lower  $\delta^{13}C_{POC}$  values in inner estuarine environments reflect the contribution of terrestrial-derived OM entering the estuary and mixing with estuarine plankton (Hellings et al., 1999; Middelburg and Nieuwenhuize, 1998; Yu et al., 2010), and this is supported by the elevated C/N ratios of POM in inner estuarine environments (e.g., Hellings et al., 1999; Lamb et al., 2006; Middelburg and Nieuwenhuize, 1998; Wilson et al., 2005b; Yu et al., 2010). In estuaries situated in catchments with a large river input, river-proximal estuarine  $\delta^{13}C_{POC}$ and C/N values can be expected to largely reflect terrestrial-derived POM (e.g., Wilson et al., 2005b; Yu et al., 2010). Outer estuary

 $δ^{13}C_{POC}$  values will mainly be influenced by marine phytoplankton, as revealed by low C/N ratios of POM in these environments (Middelburg and Nieuwenhuize, 1998; Thornton and McManus, 1994; Wilson et al., 2005b; Yu et al., 2010). In light of the systematic shift in suspended  $δ^{13}C_{POC}$  values observed along the axis of many estuaries, it follows that tidal wetlands proximal (distal) to a freshwater source or situated in estuaries with a large (small) riverine input will receive POC with lower (higher)  $δ^{13}C$  values (e.g. Goslin et al., 2017).

 $\delta^{13}C_{DIC}$  values (see Table 1 for range) can also vary considerably along the axis of an estuary (e.g., Samanta et al., 2015), which will in turn affect phytoplankton  $\delta^{13}$ C values. The potential sources of DIC in river water include atmospheric CO<sub>2</sub>, CO<sub>2</sub> derived from catchment soil respiration and the decay of OM, and from the dissolution of carbonate rocks (Raymond et al., 2004; Tan and Edmond, 1993) with in situ processes (CO<sub>2</sub> evasion, aquatic productivity and respiration) further altering  $\delta^{13}C_{DIC}$  values (Finlay, 2003). Raymond et al. (2004) measured  $\delta^{13}C_{DIC}$  values ranging from -15.5% to -7% in a group of rivers in the northeast U.S., and a similar range in  $\delta^{13}C_{DIC}$  values was found by Finlay (2003) in the South Fork Eel River, California, whilst Tan and Edmond (1993) observed an even greater range (-23% to -8.1%) in waters of the Orinoco basin, Venezuela. Mixing of river water (low  $\delta^{13}C_{DIC}$ ) with marine waters (higher  $\delta^{13}C_{DIC}$ ; Section 3.2) results in intermediate  $\delta^{13}C_{DIC}$  values in estuarine environments, and often a gradient of increasing  $\delta^{13}C_{DIC}$  with distance from the freshwater source can be discerned (e.g., Hellings et al., 1999; Ray et al., 2018; Samanta et al., 2015).

In rivers and estuaries, on average 60% of carbon transported is in the dissolved phase, whilst 40% is in the particulate phase (Raymond and Bauer, 2001). The axial estuary trend in dissolved organic carbon (DOC)  $\delta^{13}C$  ( $\delta^{13}C_{DOC}$ ) is typically similar to that of  $\delta^{13}C_{POC}$ , as DOC is principally derived from phytoplankton in the marine environment and a mixture of terrestrial OM and freshwater phytoplankton in the riverine environment (Rashid, 1985). For example, in a study of  $\delta^{13}C_{DOC}$  in several estuaries in the U.S., Peterson et al. (1994) found that the marine end-members had  $\delta^{13}C_{DOC}$  values of between -25.0% and -22.0%whereas the freshwater end-members had  $\delta^{13}C_{DOC}$  values of between -28.0% and -26.0% (see also Table 1). This pattern of  $\delta^{13}C_{DOC}$  in estuarine waters, reflecting riverine and marine source material, has been observed by others (e.g., Barber et al., 2017; Goñi et al., 2003; Otero et al., 2003; Van Heemst et al., 2000). Humic substances constitute a significant proportion of DOC. Otero et al. (2003) found that humic substances accounted for 50%-80% of riverine DOC and 5%-15% of marine DOC in the Altamaha and Satilla Estuaries in southeastern U.S. A. The provenance of humic substances may be determined by their  $\delta^{13}$ C values. Terrestrial humic substances will retain  $\delta^{13}$ C values in the C<sub>3</sub> plant range for  $C_3$  plant dominated catchments, whilst the  $\delta^{13}C$  value of marine humic substances will reflect their phytoplankton precursors (Harvey and Boran, 1985). The systematic increase in  $\delta^{13}C_{DOC}$  values with proximity to the ocean is indicative of conservative DOC mixing and this is related to the relatively short residence times (days to months) of estuarine waters (Raymond and Bauer, 2001). The humic acid fraction of DOC, however, may be partly removed in the estuarine zone by flocculation, or adsorption onto inorganic colloids (Mayer, 1985); clay minerals are the most abundant inorganic colloids in estuarine environments (Andrews et al., 1996), whilst humic substances represent a significant component of organic colloids (Rashid, 1985). Humic substances, either adsorbed to clay particles or present as organic aggregates in estuarine waters, therefore represent a further source of allochthonous OC in tidal wetlands. Possibly due to the removal of a large component of terrestrial OM at the head of the estuary, there is evidence to suggest that colloidal material in estuaries appears to be dominated by marine-derived OM (Mayer, 1985), although other studies have found a significant terrestrial-derived DOM component in the marine environment (e.g., Lee and Wakeman, 1992). Apart from marine and riverine sources of DOM, in situ estuary DOM may be important in some estuaries. Estuarine DOM may originate from excretion by

estuarine dwelling organisms, autolysis of dead organisms and microbial decomposers (Van Heemst et al., 2000).

4.2. Bulk sediment geochemical RSL indicators in tidal marshes with  $C_3$  and  $C_4$  vegetation

Salt marshes in most regions are vegetated by plants utilising the C<sub>3</sub> photosynthetic pathway (Section 4.3; Goslin et al., 2017; Milker et al., 2015; Wilson, 2017). Mangrove communities are also comprised of C<sub>3</sub> plants (Section 4.4; Kemp et al., 2019; Khan et al., 2019; Sun et al., 2021). In contrast, salt marshes in the U.S. mid-Atlantic and New England regions are unusual because they are often (and natively) dominated by the C<sub>4</sub> plants S. alterniflora, S. patens, and D. spicata (Eleuterius, 1976; Niering et al., 1977; Redfield, 1972). These communities occupy the vegetated (above approximately mean tide level; MTL) low marsh and peat-forming high-marsh between approximately mean high water (MHW) and mean higher high water (MHHW). Between MHHW and the upper limit of marine influence there is a botanical zone occupied by C<sub>3</sub> plants that are tolerant of brackish conditions and intermittent marine inundation such as Phragmites australis, Typha augustifolia, Schoenoplectus spp., and Iva fructescens. Foraminifera are present in both environments. Above the upper limit of marine influence is a terrestrial, C<sub>3</sub> community which lacks foraminifera.

Where salt marshes are dominated by C<sub>4</sub> plants, the application of bulk sediment  $\delta^{13}$ C values as a sea-level proxy is premised on several key environmental characteristics and assumptions. (1) The botanical subcommunities dominated by C<sub>4</sub> plants occupy a discrete elevational range with a functional relationship to tidal datums. It is not necessary that this range be distinctive from zones of C<sub>3</sub> plants, although in practice the MHHW tidal datum approximates the elevation dividing vertically-sequential zones (Walker et al., 2021). (2) Plants using the C3 and C<sub>4</sub> photosynthetic pathways have distinctive  $\delta^{13}$ C values in tissue. Empirical observations show that the isotopic difference between these two groups is systematic and large in coastal environments (Johnson et al., 2007; Lamb et al., 2006; Middelburg et al., 1997, Table 1). (3) The principal source of OC in bulk sediment is in-situ accumulation of plant tissue (and by extension that influx of allochthonous carbon from adjacent or distal environments is relatively small). In the organic-rich salt marshes of eastern North America this assumption has been validated across a range of climatological and oceanographic settings (Chmura and Aharon, 1995; Gebrehiwet et al., 2008). Therefore, bulk sediment  $\delta^{13}$ C values in the stratigraphic record are a proxy for the dominant plant community at the time of sediment accumulation. (4) Post-depositional fractionation of carbon isotopes does not cause C<sub>3</sub> and C4 zones to become indistinguishable. Preferential decay of more refractory cellulose increases the relative concentration of lignin, which is associated with a systematic shift toward more negative  $\delta^{13}$ C values (Benner et al., 1987, 1991). However, the magnitude of this change is modest compared to the underlying difference between C<sub>3</sub> and C<sub>4</sub> plants (Section 5). In contrast, bulk sediment C/N ratios from different depositional zones can display a tendency to converge during early diagenesis (Gebrehiwet et al., 2008; Kemp et al., 2010). (5) Marine-influenced and terrestrial zones occupied by C3 plants can be readily distinguished using a supporting proxy such as identification of plant macrofossils (Niering et al., 1977), or the presence of foraminifera (Edwards and Wright, 2015; Scott and Medioli (1978). (6) Bulk sediment  $\delta^{13}$ C values in the stratigraphic record are interpreted through comparison to a modern dataset from analogous depositional environments. Surface bulk sediment inherently captures short term (seasonal and annual) variability, early post-depositional modification, and the balance of OC inputs from autochthonous and allochthonous sources.

There are two approaches for using bulk sediment  $\delta^{13}$ C values to reconstruct relative sea level, which are examined using a case study. In southern New Jersey (Kemp et al., 2013), northern New Jersey (Walker et al., 2021), and Rhode Island (Stearns et al., 2023), bulk sediment  $\delta^{13}$ C was measured along intertidal transects to generate a modern dataset of

129 surface samples (Fig. 1a). After correction for the <sup>13</sup>C Suess effect,  $\delta^{13}$ C ranged from -27.5% to -13.7% (Fig. 1b). The spatial consistency of  $\delta^{13}$ C values among the three study areas indicates they are likely to be temporally robust under late Holocene climate variability. The highest occurrence of foraminifera was estimated from samples at the same sites. There are 70 samples with  $\delta^{13}$ C values greater than -17%, of which 62 (86%) accumulated below MHHW in botanical zones dominated by *Spartina* spp. and *D. spicata*. There are 41 samples with  $\delta^{13}$ C values less than -20‰, of which 34 (83%) formed above MHHW (including 12 from above the highest occurrence of foraminifera) at locations occupied by C<sub>3</sub> plants. The remaining 18 samples had  $\delta^{13}$ C values of -20% to -17%, with 14 occurring below MHHW. These values are used to divide the dataset into four groups. (1) Samples with  $\delta^{13}$ C values lower than -20% and lacking foraminifera likely accumulated above the highest occurrence of foraminifera and represent a terrestrial environment. Such samples can produce limiting data (i.e., RSL must have been below this elevation), but not RSL reconstructions. (2) Samples with  $\delta^{13}$ C values less than -20% and containing





foraminifera likely accumulated at elevations between MHHW and the highest occurrence of foraminifera. (3) Samples with intermediate  $\delta^{13}$ C values (-20‰ to -17‰) with foraminifera present may have accumulated in the low marsh, high marsh, or brackish botanical zones from approximately MTL to the highest occurrence of foraminifera. (4) Samples with  $\delta^{13}$ C values greater than -17‰ represent deposition between MTL and MHHW. The presence of foraminifera is not required to identify this group (although in practice they are almost always found in this type of sediment). We note that the threshold  $\delta^{13}$ C values used to delineate groups are established subjectively and could change among researchers, in response to the availability of additional modern datasets (or indeed when using a subset of the available data), and to adjust confidence intervals.

The first approach to reconstructing RSL is assigning a sample from the stratigraphic record to one of these four groups. Consequently, elevation is treated as a discrete variable. The second approach also assigns core samples to one of the four categories, but the elevation range is used as an informative prior for a Bayesian transfer function that estimates the elevation at which a sample accumulated from foraminiferal assemblages (Cahill et al., 2016; Kemp et al., 2018). In this approach elevation is treated as a continuous variable and the priors do not set rigid bounds for the transfer function. Stearns et al. (2023) collected a sediment core from a salt marsh in Rhode Island (Fig. 1a) that records approximately 3.25 m of RSL rise during the past 3000 years (Fig. 1c). An age-depth model estimated the age of 1-cm thick intervals of the core. The lower part of the core provides a representative example of reconstructing RSL. Below 340 cm, foraminifera were absent, but shallower samples preserved assemblages (high abundances of Jadammina macrescens, Trochammina inflata, and Tiphotrocha comprimata) that are typical of high salt marsh and brackish environments (Edwards et al., 2004). At 351-336 cm,  $\delta^{13}$ C values are lower than -20% indicating accumulation above MHHW. Intermediate  $\delta^{13}$ C values (-20 to -17‰) occur from 331 cm to 306 cm, and for 301-226 cm all  $\delta^{13}C$  values are greater than -17‰ (mean -14.6‰), which is indicative of accumulation below MHHW. This trend in  $\delta^{13}$ C values is observed regularly in the basal sections of cores from the study region (Kemp et al., 2012; Walker et al., 2023). It is characteristic of a transgressive stratigraphy as a terrestrial site is converted briefly into a brackish environment and then a salt marsh, which subsequently tracks rising RSL closely through sediment accumulation.

Irrespective of the approach employed, the lowest section of the core lacking foraminifera and yielding bulk-sediment  $\delta^{13}$ C values characteristic of an environment dominated by C<sub>3</sub> plants cannot produce a RSL reconstruction because this sediment likely accumulated above marine influence. When using  $\delta^{13}$ C values as the primary proxy the most precise reconstruction ( $\pm 17$  cm) is associated with the narrow (above MHHW and below the highest occurrence of foraminifera), brackish zone of C<sub>3</sub> plants where foraminifera are present to indicate tidal inundation. The least precise reconstruction ( $\pm$ 43 cm with a conservative lower bound of MTL) comes from sediment with intermediate  $\delta^{13}$ C values. Samples with  $\delta^{13}$ C values (greater than -17‰) indicating accumulation in a C<sub>4</sub> botanical zone (i.e. a salt marsh between MTL and MHHW) have an uncertainty of  $\pm 27$  cm. Transitions between groups in this transgressive sequence would result in step changes in the subsequent RSL reconstruction. A Bayesian transfer function trained on a regional-scale data set of modern foraminifera suggests that the core sediment accumulated in the high salt marsh (above MHW) or brackish zones. The mean uncertainty (66% credible interval) was  $\pm 15$  cm. When adopting priors from  $\delta^{13}$ C values the Bayesian transfer function provides markedly different estimates for samples that likely accumulated below MHHW. At 302-227 cm, the priors cause reconstructed elevation to be 11 cm lower and uncertainty is reduced by an average of 84%. For samples that likely accumulated above MHHW (below 302 cm in the core), adopting informative priors resulted in elevation reconstructions that are on average 6 cm higher, with a mean reduction in uncertainty of 37%.

Outside of the U.S. mid-Atlantic and New England regions, Spartina

spp. can be common today, but is a recent introduction (Craven et al., 2017; Hubbard and Stebbings, 1969; Lee, 2001). In these regions it may be possible to leverage bulk sediment  $\delta^{13}$ C values as a sea-level proxy (as described previously) for the relatively recent past. The introduction of a non-native C<sub>4</sub> plant such as Spartina spp. to salt marshes with native vegetation dominated by C3 taxa may result in a recognizable shift in sediment  $\delta^{13}$ C values at the time of arrival. If the time of introduction is known (for example from historical records), this horizon could be used as a constraint or input to age-depth models. Similarly, the  $^{13}\!\mathrm{C}$  Suess effect has a well-established history (Kwon et al., 2022); in an otherwise stable depositional environment (as evidenced for example by foraminifera), recent up-core trends in  $\delta^{13}$ C values (increasingly negative) may reflect the change in atmospheric  $\delta^{13}$ C values caused by fossil fuel combustion, the onset (and possibly later features) of this trend could also provide a constraint for age-depth models. Evaluating bulk-sediment  $\delta^{13}\text{C}$  values as a chronological tool may broaden the utility of this isotope system for reconstructing RSL.

## 4.3. Bulk sediment geochemical RSL indicators in $C_3$ -vegetated tidal marshes

In the context where coastal marsh C<sub>4</sub> vegetation is absent or rare, bulk organic sediment  $\delta^{13}$ C, measured alongside other geochemical parameters (typically C/N and TOC), may be applied to distinguish between upper inter-tidal or supra-tidal environments (OM derived predominantly from C<sub>3</sub> vascular vegetation) and lower inter-tidal or subtidal environments (OM derived predominantly from tidal-derived C<sub>3</sub> POM) (e.g., Bender et al., 2015; Craven et al., 2013, 2017; Goslin et al., 2017; Milker et al., 2015; Wilson et al., 2005a). Changes in the relative contribution of autochthonous OM (C<sub>3</sub> vascular vegetation, edaphic algae and bacteria) and allochthonous OM (tidal-derived POM and DOM) can occur in response to RSL change. Therefore, in C<sub>3</sub>-vegetated tidal marshes, bulk organic  $\delta^{13}$ C analysis of marsh deposits, measured alongside additional geochemical parameters, may be used to detect changes in RSL tendency (Section 4.3.1) or to distinguish between depositional environments (Section 4.3.2).

### 4.3.1. Relative sea-level tendency

The  $\delta^{13}$ C value of OC from C<sub>3</sub> vascular vegetation ranges between -37% and -20%, with C<sub>3</sub> tidal-derived POM values ranging between -33% and -18% (Section 3.2; Table 1). Because the  $\delta^{13}$ C range of these two sources (C<sub>3</sub> vascular vegetation and C<sub>3</sub> tidal-derived POM) overlap, additional geochemical measurements such as C/N, TOC and total nitrogen (TN) are necessary to help differentiate OM sources and depositional environments. For example, C/N analysis can distinguish between OM from vascular vegetation (C/N typically >20) and phytoplankton (C/N typically <10) (Tyson, 1995). Contemporary  $\delta^{13}$ C, C/N and TOC values from northwest European C3 coastal wetland sediments (tidal flat, salt marsh, reedswamp, fen carr) are shown in Fig. 2  $\delta^{13}$ C values range from -30.1‰ to -21.7‰, C/N 2.3 to 26.4 and TOC 0.4%-47.6%. There are clear relationships between all three variables, e.g.,  $\delta^{13}$ C is inversely correlated with C/N (r = -0.68, p = 0.00) and TOC (r = -0.71, p = 0.00) (Wilson, 2017). The progressive transition from tidal-derived POM dominance in sub-tidal and lower inter-tidal environments, to C<sub>3</sub> vascular vegetation dominance in high inter-tidal and supra-tidal areas, is reflected in falling surface sediment bulk organic  $\delta^{13}C$  and rising C/N and TOC values (Wilson, 2017). This pattern has also been observed in C<sub>3</sub> coastal wetland sediments outside of northern Europe (e.g., Bender et al., 2015; Milker et al., 2015).

The relationship between bulk organic  $\delta^{13}$ C, C/N and TOC values and ground elevation within the tidal frame can be retained in Holocene sediment deposits (Wilson et al., 2005a). Therefore, within-core trends in  $\delta^{13}$ C, C/N and TOC can be used qualitatively to identify changing RSL tendency. The integrity of SLIPs partly relies on the evidence of increasing or decreasing marine influence across transgressive or regressive contacts, respectively.  $\delta^{13}$ C, C/N and TOC analysis can



Fig. 2. Bi-plots of contemporary bulk organic sediment  $\delta^{13}$ C, C/N and TOC samples from C<sub>3</sub> estuaries and coastal wetlands in northwest Europe (adapted from Wilson, 2017).  $\delta^{13}$ C values in this figure are unadjusted for the <sup>13</sup>C Suess effect. Partitioning Around Medoids analysis distinguishes two geochemical groups in the northwest Europe  $\delta^{13}$ C<sub>adjusted</sub>, C/N and TOC dataset: Group 0: tidal flat/salt marsh (represented by star-shaped symbols) and Group 1: reedswamp/fen carr (represented by diamond-shaped symbols).

identify changing marine influence prior to lithostratigraphic contacts (e.g., Goslin et al., 2017; Khan et al., 2015b; Kemp et al., 2017a; Lamb et al., 2007; Wilson, 2017). For example, increasing  $\delta^{13}$ C and decreasing C/N across a transgressive contact could indicate increased tidal-derived POM and a diminishing contribution of OM from C<sub>3</sub> vascular vegetation. Therefore,  $\delta^{13}$ C, C/N and TOC may be used to help verify SLIPs (Goslin et al., 2017; Khan et al., 2015b; Kemp et al., 2017a; Lamb et al., 2006, 2007; Wilson et al., 2005a).

#### 4.3.2. Geochemical classification of tidal environments

The use of bulk organic  $\delta^{13}\text{C},$  C/N and TOC analysis of C\_3 vegetated tidal marsh sediments to distinguish between depositional environments for the purposes of RSL reconstruction has been attempted in northwest Europe (Goslin et al., 2015, 2017; Khan et al., 2015b; Wilson, 2017), Eurasia (Kemp et al., 2017a), northwestern USA (Bender et al., 2015; Engelhart et al., 2013; Milker et al., 2015) and eastern Canada (Kemp et al., 2017b). The approach involves establishing the contemporary variability of bulk organic  $\delta^{13}$ C, C/N and TOC, usually along tidal marsh transects, and utilising quantitative techniques to identify statistically significant geochemical groups. This allows past depositional environments to be recognised based on the similarity of contemporary and sediment core geochemical values, and the indicative meaning and associated RWLs to be identified. Problems encountered with this approach include: (i) the potential for a lack of geochemical distinctiveness between tidal marsh zones (Kemp et al., 2017b); (ii) selective decomposition of OM, which can alter bulk  $\delta^{13}$ C and C/N values (Section 6; Kemp et al., 2017a; Lamb et al., 2006; Wilson, 2017); (iii) the influence of the <sup>13</sup>C Suess effect on contemporary  $\delta^{13}$ C values (Section 3.3; Wilson, 2017). Although these issues are not unique to C<sub>3</sub>-vegetated tidal marshes, the smaller range in bulk sediment  $\delta^{13}$ C values encountered in C<sub>3</sub> vegetated tidal environments (typically <10‰, Table 1) may mean that relatively small shifts in bulk organic  $\delta^{13}$ C values as a result of decomposition or in response to the <sup>13</sup>C Suess effect, are of sufficient magnitude to potentially complicate source OM identification. This is less of an issue in tidal marshes supporting C3 and C4 plants, where the range in  $\delta^{13}C$  values is much greater (>10‰) and vegetation zones isotopically distinctive (Section 4.2; Kemp et al., 2010).

Several studies have explored and successfully applied bulk  $\delta^{13}$ C and geochemical indicators to reconstruct past RSL change in tidal marsh environments that are vegetated exclusively, or mainly, by C<sub>3</sub> plants (e. g., Bender et al., 2015; Engelhart et al., 2013; Goslin et al., 2017; Milker et al., 2015). At Bandon Marsh, Oregon, USA, Milker et al. (2015) found that contemporary bulk organic  $\delta^{13}$ C, C/N and TOC values significantly

correlated with elevation, with no significant inter-annual variations in values. The generally decreasing  $\delta^{13}$ C values from the tidal flat to upland environments (c. -21% to -30%), along with increasing C/N and TOC values, indicate the increasing incorporation into sediments of OC from C<sub>3</sub> terrestrial plants and a decrease in the influence of allochthonous POM and DOC derived from marine sources. Engelhart et al. (2013) measured contemporary bulk sediment  $\delta^{13}$ C, C/N and TOC in four transects across Siletz Bay, located further north along the Oregon coastline.  $\delta^{13}$ C values here ranged from c. -31% to -21%, and three geochemical groups were identifying using Partitioning Around Medoids (PAM) analysis (Kaufman and Rousseeuw, 1990). The three groups broadly correspond to those identified in a foraminifera dataset collected along the same transects. Bulk sediment  $\delta^{13}\text{C}\text{, C/N}$  and TOC distinguished between highest marsh ( $\delta^{13}$ C of  $-29.6 \pm 0.8$ %; C/N of 20.4  $\pm$  3.7; TOC of 30  $\pm$  4.6%), high to middle marsh ( $\delta^{13}$ C of -27.3  $\pm$ 1.4‰; C/N of 13.6  $\pm$  1.4; TOC of 12.4  $\pm$  4.0%) and low marsh to tidal flat environments (  $\delta^{13}C$  of  $-24.1\pm1.7$  %; C/N of 10.4  $\pm$  1.4; TOC of 2.5  $\pm$  1.8%). Based on the contemporary  $\delta^{13}C$  and bulk geochemical elevation-dependent distributions, Engelhart et al. (2013) were able to identify palaeomarsh environments and associated changes in palaeomarsh elevation across a stratigraphic contact marking the 1700 CE great Cascadia earthquake. The  $\delta^{13}$ C and bulk geochemical-inferred magnitude of coseismic subsidence across the contact was comparable to the value calculated using fossil foraminifera assemblages. In this proof-of-concept study, Engelhart et al. (2013) found that  $\delta^{13}$ C and bulk geochemistry can be used to quantify the magnitude of RSL change, in this example in response to tectonic activity. This finding was supported by Bender et al. (2015), who successfully applied  $\delta^{13}$ C and bulk geochemistry to estimate the magnitude of coseismic RSL changes associated with the 1964 great Alaskan earthquake.

Goslin et al. (2017) generated a contemporary bulk sediment  $\delta^{13}$ C and geochemical (C/N, TOC, TN) dataset from tidal marshes in Western Brittany, France. They also found that  $\delta^{13}$ C, C/N, TOC and TN values were correlated with elevation. Contemporary bulk sediment  $\delta^{13}$ C values generally decreased from tidal flat to upland environments (c. -18% to -30.5%), with a corresponding overall increase in C/N (5.6 to 15.74), TOC (0.22 to 48.5%) and TN (0.03 to 3.5%), reflecting a relative increase in OM derived from C<sub>3</sub> terrestrial photoautotrophs, and a diminishing influence of allochthonous POM and DOC derived from marine sources. PAM analysis of the contemporary dataset distinguished three elevation-dependent geochemical groups. Based on the contemporary  $\delta^{13}$ C and bulk geochemical elevation-dependent distributions, Goslin et al. (2017) generated Holocene SLIPs using  $\delta^{13}$ C and bulk

geochemistry. Nevertheless, Goslin et al. (2017) found that C/N ratios proved unreliable as a source OM indicator owing to post-depositional losses in TN. This issue has also been noted in other studies (e.g., Engelhart et al., 2013; Kemp et al., 2017a).

Research by Kemp et al. (2017a, 2017b) highlighted some of the limitations and complications of using bulk sediment  $\delta^{13}C$  and geochemistry as RSL indicators in tidal marsh environments vegetated exclusively, or mainly, by C<sub>3</sub> plants. Kemp et al. (2017a) measured  $\delta^{13}$ C, C/N and TOC (alongside additional geochemical and biological (diatom and foraminifera) analyses) along transects incorporating tidal flat, salt marsh, transitional reed swamp and upland Taiga environments in several sub-arctic marshes of the White Sea, Russia. PAM analysis identified two elevation-dependent groups in the geochemical dataset, distinguishing between samples originating from below MHHW (tidal flat and salt marsh environments, characterised by high  $\delta^{13}$ C, low C/N and TOC) and above MHHW (predominantly from the Phragmites and upland Taiga forest zone, characterised by low  $\delta^{13}$ C, high C/N and TOC). This dichotomy in contemporary samples was replicated in the contemporary foraminifera assemblages. However, contemporary bulk sediment  $\delta^{13}$ C and C/N values in supra-tidal and inter-tidal zones overlapped and therefore the upper inter-tidal limit could not be constrained based on  $\delta^{13}$ C and C/N values alone. Kemp et al. (2017a) then evaluated the use of bulk sediment geochemistry to identify past changes in RSL in a late Holocene sediment core capturing a regressive sediment succession sequence. C/N values exhibited a clear systematic increase in values as tidal flat sediments were replaced by salt marsh deposits and then by a freshwater peat.  $\delta^{13}$ C changed more conservatively across the regressive contact, falling from c. -25% to c. -27%, together with changing C/N attesting to a reduction in marine-derived OC. Nevertheless, because elevation-dependent zones in the contemporary environment were poorly constrained in the  $\delta^{13}C$  and C/N dataset, Kemp et al. (2017a) were unable to use absolute  $\delta^{13}$ C and C/N values to identify elevation-dependent environments in the sediment core, and this limited the capability of  $\delta^{13}$ C and C/N as RSL indicators in this location. C/N values experienced notable post-depositional modification, with C/N values higher in core sediments compared with their modern equivalent counterparts. Kemp et al. (2017a) also cited post-depositional modification as a potential issue in using absolute  $\delta^{13}$ C values to infer past depositional environments. They assert that bulk sediment from the *Phragmites* and Taiga forest zones did not appear to undergo significant post-depositional modification in  $\delta^{13}$ C values, but tidal flat and salt marsh sediments did on the basis that core sediment values of microfossil-inferred tidal flat and salt marsh deposits (-26%)were on the whole lower than the contemporary equivalent environments (-27% to -19%). The absence of significant post-depositional modification may reflect the slow rates of OM decomposition in terrestrial subarctic environments. POM is also susceptible to decomposition, which would result in a post-depositional shift to lower  $\delta^{13}C$ values (Section 6). However, other factors can influence POM  $\delta^{13}$ C values, particularly water temperature (Liu et al., 2022; Sackett et al., 1965) and it is interesting to note that the sediment sequence analysed by Kemp et al. (2017a) and radiocarbon dated to 2804  $\pm$  52 cal yr BP was deposited around the time of a rapid climate event, with North Atlantic ice rafting and alpine glacier advances attesting to cold high-latitude climates (Mayewski et al., 2004). Therefore, a scenario of cooler regional sea surface temperatures resulting in the receipt of tidal-derived POM with lower  $\delta^{13}$ C values than that measured in the contemporary environment is also plausible.

In a similar study, Kemp et al. (2017b) explored the utility of bulk sediment  $\delta^{13}$ C (used in isolation) as a RSL indicator in several salt marshes located in Newfoundland, Canada. Surface sediment samples from transects at two sites were collected from salt marsh and supra-tidal environments for  $\delta^{13}$ C analysis. Contemporary bulk sediment  $\delta^{13}$ C values ranged between -29% and -24%, with no discernible trend in  $\delta^{13}$ C values with ground elevation. They concluded that bulk sediment  $\delta^{13}$ C cannot be used as a RSL indicator in Newfoundland.

Moreover, they advised that the effective use of  $\delta^{13} C$  as a RSL indicator is restricted along the Atlantic coast of North America to regions between Chesapeake Bay to southern Nova Scotia. Here, high salt marsh zones are colonised extensively by C<sub>4</sub> plants, resulting in bulk sediment  $\delta^{13}$ C values that are isotopically distinctive from freshwater C3 marsh and upland deposits (Section 4.2). More research from C<sub>3</sub> dominated marshes along the Atlantic coastline would be needed to confirm this because  $\delta^{13}$ C, measured alongside other bulk geochemical parameters, has been used successfully in other (European) C<sub>3</sub> tidal-dominated marshes (e.g., Goslin et al., 2017; Khan et al., 2015b). The coastal configuration of the Newfoundland study sites investigated by Kemp et al. (2017b) may well explain the conservative variability in salt marsh bulk sediment  $\delta^{13}$ C values. Their study sites are situated in areas with a restricted connection to the open ocean. Coastal waterbodies with a restricted connection to the ocean along the Atlantic seaboard exhibit POM  $\delta^{13}$ C values several per mille lower than water bodies without a restricted connection (Hunt, 1970). For example, Albemarle Sound (North Carolina, USA) is protected by extensive barrier islands and has POM  $\delta^{13}$ C values in the range of -27.9% to -24.8%. This contrasts with Narragansett Bay (Rhode Island), which does not have a restricted connection to the open ocean and has POM  $\delta^{13}$ C values several per mille higher (-22.7% to -22.2%) (Hunt, 1970). The lower POM  $\delta^{13}$ C values in Albemarle Sound reflect the influence of terrestrial-derived OM via river input, and the restricted influence of marine POM. Indeed, almost all C/N ratios in the surface sediments of the marshes studied by Kemp et al. (2017b) remain elevated above 12, demonstrating the mixing of terrestrially-derived OM in the sample.

It is important to analyse contemporary tidal marsh bulk sediment  $\delta^{13} C$  and geochemistry in order to distinguish elevation-dependent geochemical groups and to use such observations to infer indicative meanings and RWLs. However, where the collection of local contemporary geochemical datasets is restricted or prohibitive, use of regional contemporary datasets could be a viable alternative. Khan et al. (2015b) developed a regional (England) dataset of tidal marsh bulk  $\delta^{13}$ C and geochemical values (n = 132). They identified four elevation-dependent groups in the regional dataset. These were: tidal flat/low marsh ( $\delta^{13}$ C:  $-24.9 \pm 1.2$ %; C/N: 9.9  $\pm$  0.8; TOC: 3.6  $\pm$  1.7%); mid marsh/high marsh ( $\delta^{13}$ C: -26.2 ± 1.0%; C/N: 12.1 ± 1.8; TOC: 9.8 ± 6.7%); reedswamp ( $\delta^{13}$ C: -27.9 ± 0.7%; C/N: 13.9 ± 1.2; TOC: 36.5 ± 11.5%) and fen carr ( $\delta^{13}$ C:  $-29.0 \pm 0.6$ %; C/N: 17.4  $\pm$  3.1; TOC: 41.6  $\pm$  5.7%). Khan et al. (2015b) were able to apply this dataset to identify new SLIPs in a Holocene sediment sequence from the Thames Estuary, UK. Changes in  $\delta^{13}$ C and bulk geochemical values across stratigraphic contacts were used to demonstrate changes in marine tendency (Section 4.3.1), with RWLs inferred from characteristic changes in  $\delta^{13}$ C and bulk geochemistry across threshold values. For example, MHWS-20 cm was assigned to transgressive contacts comprising a Phragmites or monocot peat deposit situated directly below a clastic salt marsh deposit, associated with  $\delta^{13}$ C values increasing from <-27% to >-27%, and C/N and TOC values decreasing from >14 to <14, and >18% to <18%, respectively (Khan et al., 2015b).

Wilson (2017) compiled a dataset of bulk organic sediment  $\delta^{13}$ C, C/N and TOC drawn from published studies of contemporary C<sub>3</sub>-vegetated northwest European coastal wetlands (datasets from England, Ireland, Scotland and France). The contemporary  $\delta^{13}$ C measurements were corrected for the <sup>13</sup>C Suess effect (termed  $\delta^{13}$ C<sub>adjusted</sub>) to allow comparison of contemporary and pre-industrial  $\delta^{13}$ C values (Section 3.3), thus permitting a more accurate use of contemporary  $\delta^{13}$ C values as analogues for Holocene deposits. PAM analysis distinguished two geochemical groups: tidal flat/salt marsh (Group 0) and reedswamp/fen carr environments (Group 1) (Fig. 2). Salt marsh and reedswamps are part of the natural coastal vegetation zonation of temperate northwest Europe. As such, the salt marsh and reedswamp transition (Group 0–1 transition) will often form part of a conformable lithostratigraphy in temperate northwest European coastal wetland deposits. RWLs at salt marsh and reedswamp transitions are well established (e.g., Shennan,

1982). Group 0–1 mid-point  $\delta^{13}C_{adjusted}\!,$  C/N and TOC values of -26.0‰, 13.3 and 22.8%, respectively, generally separate spatially adjacent mid/high saltmarsh and reedswamp samples in the contemporary northwest European dataset. By comparison, Khan et al. (2015b) calculated  $\delta^{13}$ C, C/N and TOC threshold values of -27‰, 14 and 18% respectively to distinguish between mid/high saltmarsh and reedswamp environments in the English dataset and applied these threshold values to assign RWLs at lithostratigraphic contacts in Thames Estuary Holocene deposits. Their use of  $\delta^{13}$ C values uncorrected for the  $^{13}$ C Suess effect mostly accounts for the observed difference in the  $\delta^{13}\text{C}$  threshold values between mid/high saltmarsh and reedswamp environments in the two datasets. The application of the northwest European  $\delta^{13}$ C, C/N and TOC dataset in RSL reconstruction studies was exemplified using Holocene sediment deposits of the Mersey Estuary, UK (Fig. 3); the range in core  $\delta^{13}$ C, C/N and TOC values here is typical of the variability encountered in Holocene sediment sequences from temperate C<sub>3</sub> tidal wetlands (e.g., Goslin et al., 2015; Khan et al., 2015b; Lamb et al., 2007). Group membership of sediment deposits was identified using binary logistic regression, which models the correspondence between sediment core  $\delta^{13}$ C, C/N and TOC values and the contemporary northwest European dataset. Group membership was predicted with high probability in the various depositional contexts studied and the accuracy of group prediction was verified by microfossil evidence. Group 0-1 transitions were found to coincide with changes in litho- and bio-stratigraphy. Transitions between depositional group membership in sediment sequences was accompanied by a progressive increase or removal of marine conditions qualitatively inferred by trends in  $\delta^{13}$ C, C/N and TOC (Section 4.3.1). This demonstrates the transition between spatially adjacent environments in core samples, thereby narrowing the altitudinal envelope of the indicative meaning of the sample and allowing a more confident distinction of RWLs. This approach proved robust because (i) the difference in medoid  $\delta^{13}C$  and C/N values between contemporary depositional groups is greater than the typical post-depositional shift in bulk  $\delta^{13}$ C and C/N resulting from the selective

degradation of organic compounds (Section 6); (ii) coastal depositional environments are identified in sediment records based on a probabilistic measure of correspondence with the contemporary geochemical dataset, thus uncertainty introduced as a result of an unusual combination of values will yield a lower probability of group allocation; (iii) agreement between all co-variates is required to achieve a high probability of group membership. Therefore, without a corresponding change in C/N and TOC, any shift in  $\delta^{13}$ C due to selective OM degradation, for example, should not lead to a change in group allocation of core samples – a fall in the probability of group allocation is more likely.

#### 4.4. Geochemical RSL indicators in mangrove environments

Mangrove vegetation communities are dominated by C3 plants and therefore as in C<sub>3</sub>-vegetated tidal marshes (Section 4.3), bulk organic sediment  $\delta^{13}\mbox{C},$  measured alongside C/N and TOC, may be applied to distinguish between upper inter-tidal and supra-tidal environments, where sedimentary organic matter is primarily derived from in situ C3 vegetation, and lower inter-tidal and sub-tidal environments, where tidal-derived POM and marine macrophytes (e.g., seagrass or algae) substantially contribute to sedimentary OM (see Table 1 for  $\delta^{13}$ C value ranges). Based on this premise, numerous recent studies have used  $\delta^{13}$ C and C/N geochemistry to examine the response of mangrove ecosystems to RSL changes in South (Kumar et al., 2018; Sarkar et al., 2009), Southeast (Bird et al., 2010; Chua et al., 2023; Tue et al., 2011, 2019; Xu et al., 2024), and East Asia (Meng et al., 2016; Sun et al., 2021; Xia et al., 2015, 2019; Xiong et al., 2018; Zhang et al., 2021, 2023), Oceania (Jacotot et al., 2018), the circum-Caribbean (Castañeda-Posadas et al., 2022; Monacci et al., 2009; Jones et al., 2019) and Pacific (Seddon et al., 2011) and Atlantic South America (Bozi et al., 2021; Cohen et al., 2012, 2016, 2021, 2016; França et al., 2013, 2014, 2015, 2016, 2019; Martins et al., 2021; Nunes et al., 2023; Ribeiro et al., 2018). Fewer studies, however, have applied  $\delta^{13}$ C, TOC and C/N to produce records of past RSL change, at least in part because of some of the unique challenges



Fig. 3. Mersey Estuary (UK) Holocene diatom assemblages together with  $\delta^{13}$ C (inverted axis), C/N, TOC sample values and group classification (-0.98 to +2.42 m OD). Core lithostratigraphy and chronology are also shown (1 = 7350 ± 60 <sup>14</sup>C yr BP; 2 = 6800 ± 50 <sup>14</sup>C yr BP; 3 = 5940 ± 40 <sup>14</sup>C yr BP). Modified from Wilson (2017).

### these environments present.

Mangroves are considered to be the low-latitude counterpart of tidal marshes, although they differ in vegetation type, relation to the tidal frame, and climate regime, which may affect the use of  $\delta^{13}\text{C}$  and C/N in palaeoenvironmental and RSL reconstruction (Khan et al., 2019). Herbaceous vegetation such as grasses, rushes, and sedges are found in tidal marshes, whereas trees, shrubs, and ferns predominate in tropical mangroves (Robertson and Alongi, 1992).  $\delta^{13}$ C and to a much greater extent C/N varies between herbaceous and woody materials, and these vegetation types may differ in resistance to microbial attack (Benner et al., 1987) given the recalcitrance of mangrove-derived OM (Adame et al., 2024; Baker et al., 2021). Furthermore, similarities between the  $\delta^{13}$ C and C/N values of mangrove and other upland/terrestrial vegetation communities complicate the use of  $\delta^{13}C$  and C/N as a standalone sea-level indicator and often necessitates the use of complementary indicators (e.g., microfossils). For example, Khan et al. (2019) found that brackish to freshwater, supra-tidal environments dominated by Avicennia and Pterocarpus in Puerto Rico have similar vegetation and sediment  $\delta^{13}$ C and C/N signatures to saline, inter-tidal mangroves occupied by Rhizophora, which required the use of foraminifera and thecamoebians to robustly distinguish between these environments. Similarly, Kemp et al. (2019) observed that *Rhizophora* can occupy both saline and freshwater habitats in Bermuda, although bulk surface sediments from these environments were characterized by distinct ranges in  $\delta^{13}$ C, TOC and C/N values between the saline mangroves and freshwater environments likely due to greater import of marine OM by tides in the saline mangroves.

Mangroves, particularly in microtidal settings, can colonize elevations lower in the intertidal zone down to mean low water (MLW) (Dawes, 1998; Khan et al., 2022) compared to tidal marsh vegetation, which typically grows at elevations greater than MTL or above (Davis and Fitzgerald, 2003; Engelhart et al., 2011). Due to the greater inundation period by tides and stronger bottom friction effects created by aerial root systems, mangroves may incorporate greater amounts of allochthonous marine organic material into their sediments (Wolanski et al., 2013). This ability to rapidly trap and accumulate sediments may cause mangrove sedimentation to occur at a rate that is out of pace with changes in RSL, which can complicate RSL reconstruction from mangrove archives especially when using  $\delta^{13}C$  and C/N proxies (e.g., Kemp et al., 2019; Khan et al., 2019). The greater belowground biomass (i.e., roots) generated by mangrove ecosystems compared to salt marshes (Alongi, 2020) may further confound the interpretation of sediment archives by contaminating deeper (older) sediment layers with mangrove-derived carbon (e.g., Tue et al., 2012). This may lead to the (erroneous) interpretation that these deeper sediment layers accumulated within mangrove environments in the absence of other detailed litho-, bio-, and chrono-stratigraphic evidence (Khan et al., 2019).

In the (sub)tropics, higher temperatures and precipitation promote faster rates of OM breakdown and may alter OM stability in the mangrove surface and subsurface (Coûteaux et al., 1995; Franzluebbers et al., 2001; Malhi et al., 1999). This may complicate the interpretation of  $\delta^{13}$ C and C/N OM signatures from source (e.g., in situ vegetation, marine phytoplankton) to incorporation and burial in sediments. For example, Tue et al. (2011) and Xia et al. (2015) used end-member mixing models to estimate the contribution of mangrove-derived OM to sediments and found that diagenetic effects caused sediment  $\delta^{13}$ C and C/N to differ from their source. Therefore, while source  $\delta^{13}$ C and C/N values are useful for interpreting their relative contributions to sediments, measurements of the  $\delta^{13}$ C and C/N of local surface sediments are more suitable for interpreting sediment archives for the purpose of palaeoenvironmental and RSL reconstruction. Furthermore, while some studies have found that post-depositional alteration of surface sediment signatures appears to be minimal in late Holocene sediments (e.g., Khan et al., 2019; Sun et al., 2021), others found convergence of  $\delta^{13}$ C and C/N values in the sedimentary archives (Kemp et al., 2019). Fewer studies considered these effects over early to mid Holocene timescales (e.g., Tue

et al., 2019). Finally, elevated temperatures and precipitation also promote higher rates of microbial activity that contribute to poor preservation or absence of microfossil indicators (e.g., Berkeley et al., 2007) that are often necessary complementary indicators to  $\delta^{13}$ C and C/N in mangrove environments.

Studies that tested the use of  $\delta^{13}$ C, TOC and C/N of bulk mangrove sediments to reconstruct palaeoenvironmental and RSL change have typically focused on first characterizing the  $\delta^{13}$ C, TOC and C/N of elevation-dependent environmental zones and second assigning samples from sedimentary archives to one of these environmental zones on the basis of  $\delta^{13}$ C, TOC and C/N signatures (i.e., treating elevation as a discrete variable). Both Atlantic-East Pacific and Indo-West Pacific mangroves have shown distinct  $\delta^{13}$ C values of elevation-dependent environmental zones, with mangrove sediments ranging from  $\sim -27\%$ to -24% and freshwater/upland environments characterized by values <-28% (Table 2). Variability in the  $\delta^{13}$ C of sub-tidal and tidal flat sediments can be attributed to the local geomorphic setting of each site. For example,  $\delta^{13}$ C values of  $-18.6 \pm 2.8\%$  from a tidal flat adjacent to an open marine coast in Puerto Rico reflect the incorporation of marine macrophytes and POM, whereas lower  $\delta^{13}$ C values of  $-24.3 \pm 0.5$ % and  $-24.2 \pm 1.0\%$  from tidal flats of Hong Kong and Vietnam, respectively, can be explained by their proximity to large river estuaries with high freshwater discharge and terrestrial OM supply. TOC and C/N values tend to increase across tidal flat, mangrove, and freshwater/upland sediments due to the decreasing contributions of tidally-imported OM, although as with tidal flat  $\delta^{13}$ C, there are considerable differences across sites, with the estuarine sites in Vietnam and Hong Kong exhibiting overall lower TOC and C/N values at all environmental zones due to the greater delivery of mineral sediment from river discharge (Sun et al., 2021; Tue et al., 2012).

Assigning samples from sedimentary archives to vegetation zones by qualitative comparison to modern  $\delta^{13}$ C, TOC and C/N observations has yielded variable results. Kemp et al. (2019) found that the  $\delta^{13}$ C, TOC, and C/N of late Holocene bulk freshwater and mangrove sediment converged after incorporation into the stratigraphic record, becoming difficult to distinguish from one another. They attributed the decrease in mangrove sediment  $\delta^{13}$ C values over time to the preferential decay of cellulose over lignin (Benner et al., 1987), and the increase in  $\delta^{13}$ C of freshwater sediment in the core to physical mixing (e.g., bioturbation from mangrove roots) with an overlying marine algal layer more depleted in  ${}^{13}$ C (~-20‰) or limited sampling of the modern freshwater environment that did not adequately capture the full degree of variability in  $\delta^{13}$ C. Conversely, Sun et al. (2021) deemed trends in bulk sediment  $\delta^{13}$ C, TOC and C/N were sufficient to identify mudflat, mangrove, and mixed forest environments and a pattern of coastal progradation from three late Holocene cores.

On the northeast coast of Puerto Rico, Khan et al. (2019) attempted to reconstruct palaeomangrove elevation using modern observations of  $\delta^{13}$ C, TOC and C/N values (Fig. 4). They identified four vertically-zoned environments (tidal flat, mangrove, brackish transition, freshwater swamp (Table 2; Fig. 4a)). These environments (including two mangrove sub-zones: a basin stand dominated by the species Avicennia and a riverine stand occupied by mixed mangrove species) had distinct  $\delta^{13}$ C, TOC and C/N values, with the exception of the brackish transition and freshwater swamp zones that were difficult to distinguish on a geochemical basis alone (Fig. 4b and c). However, the ratio of foraminifera to thecamoebians (F/T) were able to aid the distinction between these environments. Linear discriminant functions developed from the modern training set of bulk sediment  $\delta^{13}$ C, TOC, C/N, and F/T values (Fig. 4d) were applied to a core, which began accumulating at  $\sim$ 1650–1950 CE, to estimate the probability that each core sample belonged to each of the environmental zones (Fig. 4e). From 1.70 to 1.40 m in the core, samples from a mud unit with respective  $\delta^{13}$ C, TOC, and C/N values between -23.8% and -18.5%, 6.9-18.1%, 14.3 to 19.4, and a 100% calcareous foraminiferal assemblage were assigned to the tidal flat environment forming below MTL.  $\delta^{13}$ C sharply decreased to

#### Table 2

 $\delta^{13}$ C, TOC and C/N values of modern elevation-dependent environmental zones typical of (sub)tropical coastlines. The inclusion of data was limited to studies with a sampling regime designed to use modern observations of bulk sediment  $\delta^{13}$ C, TOC and C/N to interpret paleoenvironmental and relative sea-level changes from sedimentary archives. Mean  $\pm 1\sigma$  are given for  $\delta^{13}$ C, TOC and C/N values unless otherwise specified. MSL: mean sea level; MTL: mean tide level; MLW: mean low water; MHW: mean high water; HAT: highest astronomical tide.

Location	Environment/ vegetation zone	δ <sup>13</sup> C (‰)	TOC (%)	C/N	Salinity	Species present	Tidal range (m)	Elevation range (m MSL)	Indicative meaning	Sample size	Reference
Hungry Bay, Bermuda	Saline mangrove*	-24.3 to -26.0	23.7 to 27.9	11.5 to 15.3	Saline	Rhizophora mangle, Avicennia germinans	0.83–0.86	-	-	4	Kemp et al. (2019)
	Freshwater* wetland	-26.9 to -29.7	42.2 to 45.4	17.7 to 24.9	Brackish (<2)	Cladium jamaicae, Juniperus bermudiana, Myrica cerifera (wax myrtle), and R. mangle		-	-	4	
Northeastern Puerto Rico	Tidal flat	$\begin{array}{c} -18.6 \\ \pm \ 2.8 \end{array}$	$\begin{array}{c} 10.2 \\ \pm \ 5.7 \end{array}$	$\begin{array}{c} 12.7 \\ \pm \ 3.1 \end{array}$	High salinity (26–32)	Unvegetated or colonized by seagrasses	0.2-0.3	-0.49 to 0.10	< MTL	11	Khan et al. (2019)
	Mangrove	$\begin{array}{c} -26.4 \\ \pm \ 1.0 \end{array}$	$33.9 \pm 13.4$	24.3 ± 6.2	Intermediate to high salinity (6–32)	R. mangle, A. germinans, Laguncularia racemosa		-0.06 to 0.46	MLW to HAT	24	
	Avicennia basin	$\begin{array}{c} -25.1 \\ \pm \ 0.4 \end{array}$	14.7 ± 6.5	$\begin{array}{c} 16.0 \\ \pm \ 3.9 \end{array}$	High salinity (26–32)	A. germinans with patches of Batis maritima		0.23 to 0.46	MHW to HAT	7	
	Mixed species riverine	$\begin{array}{c} -26.9 \\ \pm \ 0.5 \end{array}$	41.7 ± 3.8	$\begin{array}{c} \textbf{27.7} \\ \pm \textbf{ 2.6} \end{array}$	Intermediate salinity (6–26)	R. mangle at the channel edge and R. mangle with A. germinans and L. racemosa moving away from the channel		-0.06 to 0.24	MLW to MHW	17	
	Brackish transition	$\begin{array}{c} -28.8 \\ \pm \ 0.7 \end{array}$	$40.8 \pm 11.7$	21.7 ± 3.7	Low salinity (<4)	A. germinans, L. racemosa, and the fern Acrostichum aureum		-0.02 to 0.66	> MHW	16	
	Freshwater swamp	$\begin{array}{c} -28.4 \\ \pm \ 0.4 \end{array}$	$\begin{array}{c} 42.8 \\ \pm \ 4.8 \end{array}$	$\begin{array}{c} 17.0 \\ \pm \ 1.1 \end{array}$	Freshwater (<0.5)	Pterocarpus officinalis and A gureum		0.60 to 0.77	> HAT	8	
Ba Lat Estuary, Red River, Vietnam	Subtidal habitat	$\begin{array}{c} -22.8 \\ \pm \ 1.0 \end{array}$	0.24 $\pm$ 0.21	8.8 ± 2.9	-	Unvegetated	2.6	-	-	27	Tue et al. (2012)
	Tidal flat	$\begin{array}{c} -24.2 \\ \pm \ 1.0 \end{array}$	0.86 ± 0.27	$\begin{array}{c} 12.8 \\ \pm \ 3.3 \end{array}$	-	Unvegetated		-	-	13	
	Natural mangroves	$\begin{array}{c} -25.9 \\ \pm \ 1.4 \end{array}$	$\begin{array}{c} 1.45 \\ \pm \\ 0.45 \end{array}$	$\begin{array}{c} 11.6 \\ \pm \ 2.5 \end{array}$	-	Sonneratia caseolaris, Kandelia obovata, A. corniculatum, and A. marina		-	-	12	
	Planted mangroves	$\begin{array}{c} -24.6 \\ \pm 1.1 \end{array}$	1.09 $\pm$ 0.32	$\begin{array}{c} 12.3 \\ \pm \ 3.1 \end{array}$	-	K. obovata		-	-	20	
Mai Po, Hong Kong	Tidal flat	$\begin{array}{c} -24.3 \\ \pm \ 0.5 \end{array}$	$\begin{array}{c} 1.6 \\ \pm \ 0.2 \end{array}$	$\begin{array}{c} \textbf{4.4} \\ \pm \ \textbf{1.3} \end{array}$	-	Unvegetated or colonized by sparse algal mats	1.8	0.3 to 0.44	-	10	Sun et al. (2021)
	Mangrove	$\begin{array}{c} -26.8 \\ \pm \ 0.4 \end{array}$	3.1 ± 0.7	8.6 ± 1.6	-	Kandelia candel, Acanthus ilicifolius, Aegiceras corniculatum, Avicennia marina		0.44 to 1.3	-	15	
	Mixed forest	$\begin{array}{c} -28.5 \\ \pm \ 0.5 \end{array}$	8.4 ± 2.2	11.9 ± 2.0	-	Mangrove forest mixed with upland species		1.3 to 1.6	_	3	

 $^*$  The range of  $\delta^{13}$ C, TOC and C/N values are provided for each vegetation zone.

values between -26.5‰ and -23.7‰ and TOC and C/N respectively increased from 6.9 to 18.1% and 14.3 and 19.4 in a transitional unit between 1.39 and 1.24 m composed of organic-rich mud with wood and shell fragments dominated by calcareous foraminifera. This unit, which lacked an analogue in the modern training setting (Fig. 4b and c), could not be confidently assigned to either the tidal flat or mangrove environment by the linear discriminant analysis. The upper 1.24 m of the core consisted of a red muddy mangrove peat (identified by its coarse, fibrous texture and the presence of *R. mangle* roots) with relatively uniform  $\delta^{13}$ C, TOC, and C/N values between -27.5% and -25.6%, 28.2–47.4%, and 20.2 to 39.0, respectively. In contrast, foraminiferal assemblages changed within the peat unit, transitioning from a calcareous-dominated assemblage from 1.24 to 0.67 m to an assemblage dominated by agglutinated taxa characteristic of mangrove environments from 0.66 m to the top of the core. Linear discriminant functions assigned samples from this unit to mangrove environments forming



**Fig. 4.** Case study from northeastern Puerto Rico using bulk sediment  $\delta^{13}$ C, TOC, C/N, and microfossils to reconstruct RSL (Khan et al., 2019). (a) The elevation distribution of environmental zones identified across study sites. Lightly-shaded gray boxes represent sub-environments within the mangrove zone. Elevations are expressed relative to local mean tide level (MTL) and tidal datums are shown for reference. HAT, highest astronomical tide; MHHW, mean higher high water; MHW, mean high water; MTL, mean tide level. (b) Comparison of  $\delta^{13}$ C and C/N values from modern and core bulk organic sediment. Shaded boxes represent the range of  $\delta^{13}$ C and C/N values from modern sediments of the dominant environmental zones. Core sediment  $\delta^{13}$ C and TOC values from modern sediments of the dominant environmental zones. Core sediment  $\delta^{13}$ C and TOC values from modern sediments of the dominant environmental zones. Core sediment  $\delta^{13}$ C and TOC values from modern sediments of the dominant environmental zones. Core sediment  $\delta^{13}$ C and TOC values from modern sediments of the dominant environmental zones. (d) Modern samples positioned on the first two discriminant axes based on linear discriminant analysis of  $\delta^{13}$ C, TOC, C/N and F/T are shown on the discriminant axes. (e) Lithology, chronology,  $\delta^{13}$ C, TOC, C/N, fora-miniferal abundance, absence-presence counts of microfossil groups (calcareous foraminifera, agglutinated foraminifera, thecamoebians), and reconstructions of palaeomangrove elevation from linear discriminant analysis applied to from core BC7 collected from Espiritu Santo, Puerto Rico. Assignment of palaeoenvironmental zones.

between MLW to highest astronomical tide (HAT). Using a second training set that differentiated between mangrove subzones, the assignment of core samples by the linear discriminant analysis revealed a palaeoenvironmental succession from an A. germinans basin from 1.3 to 1.24 m, to predominantly A. germinans basin and mixed species riverine mangroves from 1.24 to 0.67 m, and finally to a mixed species riverine mangrove from 0.66 m to the core top – a pattern consistent with shoreline progradation. Together with the core chronology,  $\delta^{13}$ C, TOC, C/N, and microfossil assemblages revealed rapid accumulation likely due to enclosure by a barrier formed from increased anthropogenic sediment delivery and shoreline progradation - at a rate out of equilibrium with regional RSL rise, making the core unsuitable for RSL reconstruction. This finding is consistent with a similar disequilibrium between the rate of mangrove peat accumulation and regional-scale RSL rise following initial mangrove colonization of a shallow basin ~1200–700 years ago observed by Kemp et al. (2019) in Hungry Bay, Bermuda.

archives to reconstruct RSL have illustrated several challenges, these difficulties may not solely be attributable to the  $\delta^{13}C$  and C/N approach itself. Rather, these challenges (poor preservation of microfossils, bioturbation from mangrove roots, lack of accommodation space, etc.) are inherent in producing any proxy-based RSL reconstruction from mangrove environments (e.g., Khan et al., 2022; Sefton and Woodroffe, 2021; Woodroffe et al., 2015). However, it has been demonstrated that modern observations of bulk sediment  $\delta^{13}C$  and C/N can differentiate elevation-dependent environmental zones, particularly when used in combination with simple microfossil metrics, and these zones can be identified in sedimentary records to indicate palaeoenvironmental changes. Consequently, this approach holds promise as a valuable tool for future mangrove palaeoecological and palaeoenvironmental investigations.

## 4.5. Emerging principles and conditions of application

Although attempts to use bulk sediment  $\delta^{13}$ C and C/N of mangrove

Evaluating the performance of  $\delta^{13}$ C and bulk organic geochemistry

as a sea-level indicator in contrasting coastal environments reveals that the degree of isotopic distinctiveness between OM sources determines how the proxy can be deployed. Bulk organic  $\delta^{13}$ C can be used as the primary (or only) sea-level indicator in tidal marshes occupied by C<sub>3</sub> and C<sub>4</sub> vegetation (Section 4.2). Here, the isotopic contrast between depositional environments found at distinctive tidal elevations is very large. This difference between elevation zones within a single salt marsh exceeds variability of the same environment among sites and remains readily identifiable even with post-depositional modification. Bulk organic  $\delta^{13}C$  cannot be the sole sea-level indicator in tidal wetlands occupied exclusively by native  $C_3$  plants because the range in  $\delta^{13}C$ values encountered in these environments is small (Sections 4.3, 4.4), and contrasting sources of OM can have overlapping  $\delta^{13}$ C values (Table 1). Therefore, additional information is necessary to reconstruct RSL, which can include additional geochemical data (C/N, TOC, TN) to help distinguish between OM sources. For example, in northwest Europe  $\delta^{13}$ C values are used alongside C/N and TOC to generate SLIPs at lithostratigraphic contacts (Section 4.3.2). Such contacts separate depositional environments with contrasting OM sources, specifically densely vegetated higher inter-tidal and supra-tidal environments (OC principally derived from autochthonous C<sub>3</sub> photoautotrophs) and sparsely vegetated or unvegetated lower inter-tidal and sub-tidal environments (OC principally derived from allochthonous (tidal-derived) POM). The elevational range of these two contrasting environments is broad; the upper marine limit can be geochemically poorly constrained, but the transition between the two environments in contemporary datasets and sediment sequences is geochemically distinctive. In this context  $\delta^{13}C$ combined with other bulk organic geochemical measurements may be viewed as a primary RSL indicator for use in identifying SLIPs at lithostratigraphic contacts. Similarly, RSL reconstructions generated to estimate coseismic subsidence on the Cascadia subduction zone focuses on  $\delta^{13}$ C values across peat-mud couplets that record a stratigraphic change indicative of rapid RSL rise.

Where the isotopic contrast between depositional environments used to reconstruct RSL are modest there is the potential that factors other than RSL change may be the cause of  $\delta^{13}$ C values through time. Such controls could include, for example, the effect of sea surface temperature on planktonic  $\delta^{13}$ C values, or a coastal configuration promoting a greater retention of terrestrial-derived OM in coastal waters (Section 4.3.2). In each example, the inter-tidal gradient in bulk  $\delta^{13}$ C values could be greatly diminished, resulting in geochemically indistinguishable inter-tidal sub-environments. In drawing parallels with the discussion around the relative utility of applying local or regional microfossil datasets to reconstruct RSL (Horton and Edwards, 2005; Khan et al., 2015b), it is noteworthy that the number of inter-tidal sub-environments recognised in a contemporary geochemical dataset can increase when local datasets are combined (e.g., Engelhart et al., 2013). This is because regional datasets comprise sample measurements from a range of different sites, capturing a broader range of physiographic conditions. As such they are useful in providing a greater range of modern analogue measurements (Khan et al., 2015b). More research is needed to investigate the potential role of regional geochemical datasets to distinguish between elevation-dependent inter-tidal sub-environments. This includes the collection of a greater number of contemporary sample measurements from a wider range of locations to facilitate the generation of more regional datasets. Regional geochemical datasets may prove useful in distinguishing inter-tidal sub-environments in sediment records from locations where contemporary geochemical surveys are not possible, or where the full range of physiographic conditions that might be encountered in core records is not represented in the local contemporary environment. The use of probabilistic measures of correspondence to identify inter-tidal sub-environments in sediment deposits should be used to guard against the misidentification of palaeoenvironments (e.g., Goslin et al., 2017; Wilson, 2017) owing to, for example, site-specific environmental contexts lacking a modern regional analogue, changeable environmental

variables such as water temperature, or the impact of decomposition, each of which might influence one or more of the geochemical proxies and yet be unrelated to RSL change. Additional research directions could include evaluating the ability of other OM provenance methods in distinguishing elevation-dependent inter-tidal sub-environments, such as bulk  $\delta^{15}$ N (Smeaton et al., 2024), bulk  $\delta^{34}$ S (Lamb et al., 2023) and analytical pyrolysis (Lamb et al., 2007). In mangrove environments, organic geochemical biomarkers that are less sensitive to diagenetic processes, such as the abundance of the lipid compound taraxerol, show promise in future palaeoenvironmental and RSL reconstruction applications (e.g., Sefton et al., 2024).

#### 5. Geochemical RSL indicators in isolation basins

## 5.1. Controls on $\delta^{13}C$ and C/N in isolation basins

Isolation basin bulk organic sediment  $\delta^{13}$ C (Table 1) and C/N values are primarily controlled by the source of OM to the basin. This includes autochthonous in situ productivity and allochthonous material from the terrestrial catchment and also from the marine environment when an isolation basin is connected to the sea. The various proportions of these sources and their impact on the actual sedimentological signal has not been investigated in any detail but will be dependent on the degree of connection/isolation of the basin from marine inundation. Indeed, there have been very few studies of modern analogues of isolation basins. Mackie et al. (2005) were the first to investigate using  $\delta^{13}$ C and C/N from isolation basins as an alternative way of reconstructing palaeosalinity within a basin controlled by changes in RSL. They measured surface samples from a fully isolated basin (Loch nan Corr, LNC, a freshwater loch) and a tidal lagoon (Rumach tidal pond). Rumach tidal pond has a rock sill close to mean tide level so is connected to the sea for approximately half of the tidal cycle. The freshwater loch sample returned a relatively low  $\delta^{13}$ C value of -28.4% and a high C/N of 14.6 (Mackie et al., 2005) which fits with other surface lake bulk samples  $(\delta^{13}C \text{ of } -30 \text{ \low to } -26 \text{\ of } 8-16; \text{ Hodell and Schelske, } 1998;$ Meyers and Horie, 1993; Thornton and McManus, 1994). The tidal pond returned a higher  $\delta^{13}$ C value of -20.8% and lower C/N of 7.7 reflecting the changing source of OM to the basins (Mackie et al., 2005).

#### 5.1.1. Autochthonous organic carbon

In isolation basins the nature of the autochthonous OM from *in situ* productivity will vary depending on the connectivity of the basin to the sea. When the basin is completely isolated from the sea autochthonous productivity will be exclusively freshwater algae and aquatic plants. As a basin becomes connected to the sea there is a transitional phase when the basin is only connected during parts of the tidal cycle and forms a brackish water tidal lagoon. During this phase *in situ* productivity will shift to algae and aquatics that can tolerate brackish water conditions. Eventually, as RSL rises, the basin will be connected to the sea at all stages during the tidal cycle and will form a marine embayment. *In situ* productivity will shift to marine algae and marine plants.

C/N values for algae in general tends to be relatively low, <10. Freshwater algae tend to have relatively lower  $\delta^{13}$ C values (-30‰ to -26‰; Meyers, 1994; Schidlowski et al., 1983) while coastal lagoon plankton (brackish conditions) have intermediary values (-25‰ to -24‰; Müller and Voss, 1999) in comparison to higher values for marine algae (-23‰ to -16‰; Meyers, 1994; Prahl et al., 1980). Yu et al. (2010) identified a trend in the  $\delta^{13}$ C from lower to higher values in POC from plankton tow nets in the Pearl River estuary along a gradient from freshwater through fresh/brackish water to brackish/marine water. This trend was more pronounced during winter when input of fine-grained terrestrial OM is at a minimum (due to low rainfall) hence the POC is dominated by *in situ* algal productivity.  $\delta^{13}$ C<sub>POC</sub> values range from -30‰ to -26‰ in the freshwater section, -26‰ to -22‰ in the fresh/brackish water section to -23‰ to -22‰ in the brackish/marine section of the estuary. The *in situ* vascular plant communities from basins

show similar patterns in terms of  $\delta^{13}$ C values. Mackie et al. (2005) report  $\delta^{13}$ C values from freshwater aquatic plants ranging from -32% to -25% (C/N ranging from 12 to 18) while marine plants have  $\delta^{13}$ C values ranging from -22% to -17% (C/N ranging from 8 to 40).

#### 5.1.2. Allochthonous organic carbon

An isolation basin will receive allochthonous OC via freshwater runoff from the basin catchment, typically from a fluvial source, and, depending on the degree of isolation, from a marine source through tidal flooding. During the freshwater phase when a basin is totally isolated from the sea, allochthonous OM will be derived from the terrestrial system and will tend to have a lower  $\delta^{13}C$  value and higher C/N. However, there may be some variability depending on the type of vegetation in the catchment and background geology of the catchment (Section 4.1.2). During the transitional brackish phase an isolation basin becomes inundated through a gradually increasing proportion of the tidal cycle. Through this transition the allochthonous OM within the basin will become increasingly dominated by a marine source. This tidally derived POC will have a higher  $\delta^{13}$ C value, similar to that seen in outer estuaries of -23% to -22%, and a low C/N < 10 (e.g., Section 4.1.2; Thornton and McManus, 1994). The combined effect of allochthonous OM from a terrestrial and marine source during the transition from freshwater to fully marine conditions will be a gradual increase in the  $\delta^{13}$ C value of OM to the isolation basin sediments. While no studies have actually investigated the relative contribution of allochthonous vs autochthonous OM to isolation basin sediments the impacts of changing source of allochthonous OM alongside changes in the autochthonous OM will both lead to an increase in  $\delta^{13}$ C values over the transition from freshwater to marine conditions.

#### 5.2. Application in isolation basins

There are relatively few studies that have used bulk organic sediment  $\delta^{13}C$  and C/N in isolation basins to investigate palaeoenvironmental changes, specifically palaeosalinity changes associated with changes in RSL. This technique was first highlighted as a potential alternative to microfossil-based studies by Mackie et al. (2005), who then went on to apply the technique to a series of isolation basins in northwest Scotland (Mackie et al., 2007). Watcham et al. (2011) also included organic sediment  $\delta^{13}$ C analysis in a multi-proxy study from a series of isolation basins from the South Shetland Islands, Antarctica. More recently Taylor et al. (2023) investigated a range of proxies, including  $\delta^{13}$ C and C/N, for RSL reconstruction from Loch Duart also in northwest Scotland. Mackie et al. (2005) present  $\delta^{13}$ C and C/N data from a set of modern marine, freshwater and terrestrial plants along with sediment samples from a freshwater and marine isolation basin end member. They present a biplot of  $\delta^{13}$ C vs. C/N for this data alongside published datasets from a range of coastal environments including freshwater, coastal lagoon (brackish water) and marine plankton, tidal marsh and estuarine surface sediment samples. A series of data fields were then identified as follows: marine samples with a  $\delta^{13}$ C range from -23% to -17% and C/N range from 5 to 40; brackish samples (including coastal lagoon, estuarine and saltmarsh surface sediment samples) with a  $\delta^{13}$ C range from -26.5% to -23% and C/N range from 5 to 28; freshwater aquatic samples with a  $\delta^{13}C$  range from -33% to -24.5% and C/N range from 5 to 18; and terrestrial samples with a  $\delta^{13}$ C range from -33% to -24.5% and C/N range from 12 to >60. This approach is then applied to a previously studied isolation basin, Upper Loch nan Eala, that records clear shifts in RSL identified based on lithostratigraphy and diatom flora from c. 13 ka cal. yr BP through to the late Holocene (Shennan et al., 1994). The variability of  $\delta^{13}C$  and C/N through the sediment core can be used to construct a core zonation that can be interpreted based on the modern data fields from the  $\delta^{13} C$  vs. C/N biplot reproduced here in Fig. 5. The interpretation of environmental conditions based on these data fields is generally consistent with reconstructions based on the diatom flora (Fig. 6). Zones 1, 4 and 6 plot clearly in the marine field and have a



**Fig. 5.** C/N and  $\delta^{13}$ C from bulk organic material from a sediment core collected from the isolation basin, Upper Loch nan Eala. The core has been split into eight zones based on variations in C/N and  $\delta^{13}$ C (see Fig. 6). The marine, brackish, freshwater aquatics and terrestrial 'fields' of data are taken from modern data presented in Mackie et al. (2005) and references therein. Adapted from Mackie et al. (2005).

diatom flora dominated by marine species. Zones 2, 5 and 8 plot within the terrestrial field with most samples within the freshwater aquatic field. However, there is a clear trend of decreasing  $\delta^{13}$ C values and increasing C/N values from zone 2 through zone 5 to zone 8. These sections within the core are dominated by freshwater diatom flora. Samples from zones 3 and 7 sit within the brackish field with some zone 7 samples within the marine field. The shift to lower  $\delta^{13}$ C values and high C/N in zone 8 is suggested to be driven by an increase in terrestrial plant material content as the basin starts to become infilled. This is also supported by the significant increase in TOC over the transition into zone 8. The results from this study show that changes  $\delta^{13}$ C values and C/N from Upper Loch nan Eala isolation basin follow, and are consistent, with trends in the diatom flora highlighting the potential of this approach.

Following the initial investigation this approach was tested on a series of 6 additional isolation basins with sediment sequences dating from the Lateglacial through the Holocene (Mackie et al., 2007). All basins had previously been studied with detailed lithostratigraphy, microfossil and radiocarbon dating to provide RSL reconstructions (Lloyd, 2000; Shennan et al., 1998, 2000). Based on results from two isolation basins (Loch nan Corr and Rumach VI) this study highlighted the suitability of  $\delta^{13}$ C and C/N for palaeosalinity reconstruction associated with changes in RSL from Holocene through to the Lateglacial Interstadial (c. 13 ka cal. yr BP) aged sediments. However, when applied to basins containing older sediment sequences dating back to the early Lateglacial the relationship between  $\delta^{13}$ C and C/N variability and palaeosalinity/RSL changes breaks down. Indeed Mackie et al. (2007) conclude that  $\delta^{13}$ C and C/N cannot be used to interpret changes in palaeosalinity associated with RSL changes from sites with older sequences dating from the Lateglacial (>c. 14 ka cal. yr BP). Results from three isolation basins (Torr a' Bheithe, Loch Torr a'Bheithe and Loch a'Mhuilinn) are presented with lithostratigraphic and biostratigraphic shifts from marine to freshwater conditions dating from 14.5 to 15 ka cal. yr BP and an additional basin (Upper Allt Dail An Dubh-Asaid) from above the marine limit dating from 15.5 ka cal. yr BP. When the data are plotted on  $\delta^{13}$ C vs. C/N biplot all samples from the three sites with a clear isolation contact (with transition from marine to freshwater conditions interpreted from diatom flora) plot within the marine field. Data for the basin above the marine limit (freshwater throughout) mostly plot within the marine field, with several samples within the brackish field. No samples plot clearly within the freshwater aquatics or terrestrial



Fig. 6. Diatom salinity reconstruction of the sediment sequence from Upper Loch nan Eala from Shennan et al. (1994) alongside C/N,  $\delta^{13}$ C and %TOC from additional core collected nearby (Mackie et al., 2005). The sequence has been divided into eight zones based on variation in C/N and  $\delta^{13}$ C. These zones correlate well with changes in diatom flora and environmental interpretation based on the diatom flora of Shennan et al. (1994). Adapted from Mackie et al. (2005).

plant fields. A similar issue was identified by Taylor et al. (2023) based on analyses from Loch Duart isolation basin, NW Scotland. They report a multiproxy study from Loch Duart combining geochemistry with microfossil analyses (foraminifera and previous diatom analyses from Hamilton et al. (2015)). Based on diatom and foraminiferal records, the basin records Lateglacial marine conditions with a transition to freshwater conditions at c. 14.1 ka cal. yr BP followed by marine conditions due to re-inundation at 10.6 ka cal. yr BP, then gradual reduction in marine conditions to a brackish lagoonal environment by c. 0.3 ka cal. yr BP. All samples from the Lateglacial marine unit ( $\delta^{13}$ C values: -20% to -15%, C/N: 11 to 12) and Lateglacial through Younger Dryas freshwater unit ( $\delta^{13}$ C values: -22% to -17%, C/N: 15 to 18) plot within the marine fields on the  $\delta^{13}$ C vs. C/N biplot. While the  $\delta^{13}$ C values from these sections of the core overlap, the C/N values are higher for the freshwater section.

Mackie et al. (2007) suggest the breakdown in relationship during the Lateglacial could be due to lower atmospheric  $CO_2$  concentrations at this time and/or poor soil/vegetation development associated with changes in climatic conditions. During the early part of the Lateglacial (c. 15 ka BP) atmospheric  $CO_2$  concentration was lower than pre-industrial levels (approx. 240 ppmv in comparison to 280 ppmv). While the isotopic composition of the  $CO_2$  during the Lateglacial was higher than modern values (the <sup>13</sup>C Suess effect; Section 3.3) there is very little variation from the Lateglacial through the Holocene (Schmitt et al., 2012, Table 1). Changing climatic conditions, however, may have more significant impact. Increased temperature dependent fractionation between  $CO_{2(aq)}$  and  $HCO_3^-$  during the colder Lateglacial conditions, would lead to increased soil bicarbonate  $\delta^{13}$ C values to +2.8‰ during the Lateglacial. Poor vegetation cover during the relatively harsh conditions of the Lateglacial would lead to increased runoff of soil bicarbonate into basins. This would enrich the pool of total DIC in the basin taken up by aquatic productivity and, hence, preserved in the sediment record. In addition, terrestrial plants growing in a stressed environment either due to lack of water or nutrients, likely conditions during the cold and harsh climate of the early Lateglacial, have been shown to record higher  $\delta^{13}$ C values (Heaton, 1999). Taylor et al. (2023) suggest that the relatively high  $\delta^{13}$ C values from the Lateglacial freshwater unit of Loch Duart isolation basin that overlap with the Lateglacial marine unit may also be influenced by changes in vegetation. Pollen analysis from the freshwater organic unit identified *Myriophyllum alterniflorum*, a freshwater aquatic plant with  $\delta^{13}$ C values ranging from -22 to -12‰ (Chappuis et al., 2017).

While the  $\delta^{13}$ C values from Lateglacial samples interpreted as being from marine and freshwater environments based on microfossil assemblages completely overlap (ranging from -22% to -17%) samples from the freshwater sections show an increase in C/N from 6 to 8 in the marine sections of the core to 8–15 in the freshwater sections. This most likely reflects a decrease in proportion of phytoplankton to the OC pool replaced by aquatic plants or terrestrial plant material washed into the basin. While this might indicate a transition to freshwater conditions, with freshwater aquatics and increased terrestrial plant in wash, the C/N of marine aquatics will also produce a similar trend. Therefore, it is not possible to make a firm palaeoenvironmental interpretation basin on C/ N ratios alone.

Watcham et al. (2011) also analysed bulk organic  $\delta^{13}$ C from a series of isolation basins from the South Shetland Islands, Antarctica through the Holocene. The basins in general are characterised by relatively low OC content and extremely low nitrogen content making it impossible to generate C/N data. The diatom flora identify clear marine, brackish and freshwater units that are used, in combination with radiocarbon dating, to identify changes in RSL and ultimately generate a RSL curve for the region. However, the interpretation of the bulk organic  $\delta^{13}$ C data is not always consistent with the palaeoenvironmental interpretation based on the diatom flora with some basins showing no clear relationship between  $\delta^{13}$ C and other proxies. For some of the transitions between marine, brackish and freshwater conditions identified from the diatom flora the  $\delta^{13}$ C trends are consistent with the diatom flora but for a significant number of transitions they do not agree with interpretations based on the diatoms. The authors conclude that there are other palaeoenvironmental factors, such as variations in storminess as well as climate, that also influence the  $\delta^{13}$ C signature of the sediments. Interpreting the geochemical signature is also hampered by the lack of C/N data due to low nitrogen concentrations.

The majority of the relatively small number of isolation basin studies show that bulk organic sediment  $\delta^{13}$ C and C/N is potentially a useful additional proxy to identify changes in OC source to isolation basins associated with changes in palaeosalinity through Holocene sequences. The studies from Scotland show the combination of these measurements can be used to identify changes in in situ productivity from marine, brackish lagoonal and freshwater lagoonal environments and also associated changes in OM delivered from the marine and terrestrial environments. However, this relationship breaks down in older sequences, particularly through the early Lateglacial but also through to the Younger Dryas in some locations. This is most likely associated with major climatic shifts impacting temperature dependent isotopic fractionation and also fractionation linked to vegetation productivity under stressed environmental conditions. Changes in soil and vegetation development within the catchments of isolation basins will also impact the amount of in-washed OM and soil bicarbonate. Under such conditions the bulk organic sediment  $\delta^{13}C$  signature cannot be used as an indicator of shifts in palaeosalinity within the basin. In order to better understand some of the processes affecting the  $\delta^{13}$ C and C/N from isolation basin palaeo-records more studies of modern-day analogues are required and particularly analogues across a range of climatic settings.

# 6. Influence of decomposition on the integrity of geochemical RSL indicators

The potential for post depositional changes in bulk organic sediment  $\delta^{13}$ C and C/N is an important consideration, particularly when using contemporary geochemical values to distinguish between depositional environments (Kemp et al., 2019; Wilson, 2017). A detailed discussion of OM degradation and associated influences on bulk organic  $\delta^{13}$ C and C/N values is provided in Lamb et al. (2007, 2006). The rate and extent of decomposition depends on the chemical and structural composition of plant material (Carrasco-Barea et al., 2022; Cornwell et al., 2008), environmental factors such as temperature, hydroperiod, sediment structure, aeration and drainage (e.g., Halupa and Howes, 1995; Kirwan et al., 2014; Mueller et al., 2018; Puppin et al., 2023; Wang et al., 2019), and the community of the decomposer organisms (Gessner et al., 2010; Leadbeater et al., 2021). These factors are spatially variable, and therefore intra- and inter-site variability in the rate and magnitude of decomposition is expected.

Dead vascular vegetation is a major source of autochthonous OM in higher inter-tidal marsh, brackish and freshwater marsh and mangrove environments (Section 4.1). Lignin, cellulose and hemi-cellulose together comprise up to 90% of plant material. Relative to the whole plant, cellulose and hemi-cellulose are enriched in  $^{13}$ C by 1‰–2‰, with lignin depleted in  $^{13}$ C by between 2‰ and 6‰ (Benner et al., 1987). It

follows that changes in the relative concentrations of different plant compounds during decomposition can result in changes in bulk organic sediment  $\delta^{13}$ C values (Kemp et al., 2019; Lamb et al., 2006). For example, salt marsh monitoring studies showed the preferential decay of hemi-cellulose and cellulose (e.g., Fogel et al., 1989; Leadbeater et al., 2021; Maccubbin and Hodson, 1980), with lignified macrophyte detritus the most refractory organic component of sediments (Leadbeater et al., 2021; Tyson, 1995). The loss of labile OM during decomposition and the selective preservation of <sup>13</sup>C-depleted refractory lignin in sediments can result in a decrease in bulk sediment  $\delta^{13}C$  by several per mille (Benner et al., 1987, 1991; DeLaune, 1986; Fogel et al., 1989; Kemp et al., 2019; Wilson et al., 2005b). Post-depositional changes in bulk C/N values may also occur, particularly during the early stages of decomposition (Valiela et al., 1985). Initial leaching of soluble compounds can lead to a loss of mass in vascular plant detritus within days (Leadbeater et al., 2021), which can result in an increase in C/N as nitrogen is lost more rapidly than carbon (e.g., Benner et al., 1991; Rice and Tenore, 1981; White and Howes, 1994). A fall in C/N may follow during microbial degradation of OM (principally by bacteria: Benner et al., 1984; Carrasco-Barea et al., 2022; Leadbeater et al., 2021), due to an increase in nitrogen associated with higher microbial colonization (Benner et al., 1991; Carrasco-Barea et al., 2022; Glenn, 1976; Rice, 1982) alongside continued carbon loss through respiration, leaching, or particulate transport. The rates of decomposition may then slow during the refractory phase, owing to the concentration of recalcitrant material such as lignin (Leadbeater et al., 2021; Valiela et al., 1985).

Tidal-derived POM is an important source of allochthonous OM in sub-tidal and lower inter-tidal areas of tidal wetlands, and in isolation basins (Sections 4.1, 5.1; Lamb et al., 2006). Much of the terrigenous component of tidal-derived POM may have already been extensively degraded on land or further upstream and should be relatively resistant to further degradation (Hedges and Keil, 1995; Van de Broek et al., 2018). Phytoplankton undergoes considerable degradation in the water column, with C/N ratios of POM tending to increase as degradation proceeds resulting from the breakdown of nitrogen-rich compounds of plankton (Lamb et al., 2006). Furthermore, the preferential degradation of <sup>13</sup>C-enriched compounds such as carbohydrates and proteins, and the subsequent increasing concentration of the refractory <sup>12</sup>C enriched lipid fraction (Deines, 1980) can also result in a decrease in bulk organic sediment  $\delta^{13}$ C (Wilson et al., 2005b). Although degradation can lead to marked differences in bulk surface sediment C/N values compared to that of the principal organic source material (i.e. overlying tidal marsh vascular vegetation (Wang et al., 2003; Wilson et al., 2005b) or phytoplankton (Cifuentes, 1991; Middelburg and Nieuwenhuize, 1998)), C/N values tend to stabilise in the refractory phase, as carbon and nitrogen are lost at approximately the same rate (Melillo et al., 1989; White and Howes, 1994). This results in relatively unchanged bulk sediment C/N ratios in recent coastal wetland sediment deposits (Arzayus and Canuel, 2004; Wang et al., 2003), although conservative shifts in C/N (<3) over millennial timescales may result from further decomposition of OM and the concentration of carbon-rich refractory organic compounds (Wilson et al., 2005a).

Post-depositional changes in bulk organic sediment properties do not affect the integrity of using bulk organic  $\delta^{13}$ C or C/N qualitatively to detect increasing or decreasing marine influence in tidal wetland deposits (Chua et al., 2023; Khan et al., 2019; Lamb et al., 2007; Wilson et al., 2005a), or in isolation basin sequences (Mackie et al., 2005). This is because within-core trends in  $\delta^{13}$ C and C/N, rather than absolute values, are the focus. The ability to identify precise coastal depositional environments in sedimentary records based on absolute values of  $\delta^{13}$ C and C/N (Sections 4.2–4.4) may be compromised by the post-depositional alteration in some circumstances. Kemp et al. (2017a) found C/N to be unreliable when used to identify depositional environments in a core taken from a Eurasian subarctic saltmarsh. They found that C/N values experienced notable post-depositional modification, with C/N values higher in core sediments compared with their modern equivalent counterparts. For example, core sediments identified by microfossil analysis as being deposited in a tidal flat environment had C/N values of 13–20, whereas modern tidal flats had C/N ratios of 7–15. Furthermore, core sediment deposited in *Phragmites* and freshwater environments had C/N ratios of 28–44, which is higher than the modern equivalent range (11–26). Goslin et al. (2017) highlighted that even small changes in TN content can result in large changes in C/N values. Additionally, they noted that the magnitude of TN loss was greater in sediments with higher organic content, such as those originating in high-marsh environments, compared with organic poor sediments originating in lower inter-tidal settings.

The potential magnitude of change in bulk sediment  $\delta^{13}\!C$  values during decomposition (e.g. up to 3.7‰; Wilson et al., 2005b) is small compared with differences in  $\delta^{13}$ C values of C<sub>3</sub> plants ( $\delta^{13}$ C values of around -27%) and C<sub>4</sub> plants ( $\delta^{13}$ C values of around -13%) (Section 3.2). Therefore, the influence of decomposition on bulk sediment  $\delta^{13}$ C values does not prevent the distinction between C4-vegetated salt marsh and C3-vegetated freshwater marsh/upland deposits (e.g., Section 4.2; Malamud-Roam and Ingram, 2004). In C<sub>3</sub> dominated coastal environments, where OM sources (autochthonous C3 vascular vegetation and allochthonous C<sub>3</sub> POM and DOM) are isotopically less distinctive (Section 4.1), post-depositional shifts in bulk sediment  $\delta^{13}$ C values can have the potential to introduce uncertainty when identifying depositional environments. For example, Kemp et al. (2019) found that bulk organic  $\delta^{13}$ C could not be used to distinguish between freshwater peat and saline mangrove peat deposits in a sediment sequence collected from Hungry Bay, Bermuda. Although the modern equivalent environments were isotopically distinctive (freshwater marsh sediment  $\delta^{13}C < -26\%$ ; saline mangrove sediment > -26%), preferential decay of cellulose and the selective preservation of <sup>13</sup>C-depleted refractory lignin in sediments may have led to a shift to lower bulk organic sediment  $\delta^{13}$ C values over time, and the resultant convergence of freshwater and mangrove peat  $\delta^{13}$ C values (Kemp et al., 2019). Combining  $\delta^{13}$ C measurements with other analyses (e.g., TOC, C/N, TN) is recommended to help identify distinctive depositional zones in tidal wetland deposits when using a bulk sediment geochemical approach (Goslin et al., 2017; Kemp et al., 2019; Khan et al., 2015b). Wilson (2017) evaluated the potential for decomposition to complicate the use of bulk sediment  $\delta^{13}$ C and C/N measurements in identifying tidal marsh elevation-dependent environments in Holocene coastal sediment sequences in a C3 estuary. In a contemporary dataset of northwest European tidal marsh bulk sediment  $\delta^{13}C_{adjusted},$  C/N and TOC, the difference in medoid  $\delta^{13}C_{adjusted}$  and C/N values between the two elevation-dependent groups (tidal flat/salt marsh (Group 0) and reedswamp/fen carr environments (Group 1); 2.9‰ and 5.3, respectively) was greater than the observed average post-depositional changes in bulk organic  $\delta^{13}$ C and C/N in the Holocene coastal deposits analysed. Moreover, coastal depositional environments were identified in the Holocene sediment record based on a probabilistic measure of correspondence with the contemporary geochemical dataset. Using this approach, uncertainty introduced by an unusual combination of  $\delta^{13}$ C, C/N and TOC values would yield a lower probability of group allocation. It follows that any shift in bulk organic  $\delta^{13}$ C values due to selective OM degradation would not result in an incorrect group allocation of core samples, but rather a fall in the probability of group allocation.

In summary, although the influence of decomposition on bulk sediment geochemistry does not affect the use of  $\delta^{13}$ C and C/N to track increasing or decreasing marine influence in Holocene tidal wetland and isolation basin deposits, it must be considered when using modern bulk organic geochemical values to identify elevation-dependent depositional zones in core material. This is particularly important in environments occupied exclusively by C<sub>3</sub> plants, and receiving tidal-derived C<sub>3</sub> particulate OM, owing to the reduced range in  $\delta^{13}$ C between these contrasting organic sources. Employing multiple geochemical measurements to characterise specific depositional environments can help to overcome this potential issue (Goslin et al., 2017; Khan et al., 2015b; Wilson, 2017), as well as using statistical techniques to assign probabilistic estimates of depositional group allocation of Holocene deposits based on measures of correspondence with a contemporary geochemical dataset (e.g. Khan et al., 2019; Wilson, 2017).

#### 7. Conclusion

Bulk organic  $\delta^{13}$ C and accompanying bulk organic geochemical measurements have been increasingly used as a RSL indicator over the last two decades, specifically in tidal marsh, isolation basins and more recently in mangrove environments. The development of this technique has been driven by the potential of  $\delta^{13}$ C and bulk organic geochemistry to offer independent and continuous records of RSL change that can be generated more quickly compared with established microfossil techniques.  $\delta^{13}$ C and bulk organic geochemistry is an established method for determining organic matter (OM) provenance in a wide range of environmental settings. In tidal marsh, mangrove and isolation basins, OM is received from a range of sources with contrasting  $\delta^{13}$ C and geochemical values. Of these environments the greatest range in  $\delta^{13}$ C is encountered in tidal wetlands occupied by C3 and C4 plants, where inter-tidal subenvironments can have very distinctive  $\delta^{13}$ C signature values. This permits the use of bulk  $\delta^{13}$ C as a primary RSL indicator, capable of resolving geochemically distinct inter-tidal sub-environments with wellconstrained vertical limits. In tidal wetland and isolation basin environments occupied natively by  $C_3$  plants, the range in OM  $\delta^{13}$ C is smaller and accompanying geochemical analyses are required to help distinguish OM sources. In these environments,  $\delta^{13}$ C and bulk geochemistry have been used successfully to detect changes in marine influence over time, to identify SLIPs at lithostratigraphic contacts, and in tidal marshes to distinguish elevation-dependent inter-tidal sub-environments. However, the reduced range in OM  $\delta^{13}$ C in environments occupied natively by C3 plants means that additional environmental parameters influencing  $\delta^{13}$ C, including decomposition, can obscure the detection of elevation-dependent depositional environments. We find that the degree of isotopic distinctiveness between OM sources is key in determining how  $\delta^{13}$ C and bulk geochemistry can be deployed as a RSL indicator. Wherever possible, the collection of contemporary  $\delta^{13}C$  (corrected for the <sup>13</sup>C Suess effect) and bulk geochemical data is recommended to help inform the interpretation of core measurements. Moreover, a growing dataset of contemporary measurements will facilitate the generation of regional datasets; these capture a diversity of physiographic conditions, provide a greater range of modern analogues, and can be used to reconstruct RSL in locations where the geochemical characterisation of the contemporary environment is not possible. Future research efforts could explore the potential of other isotope and geochemical OM provenance methods to help refine the identification of elevationdependent depositional environments, particularly in tidal wetland environments occupied natively by C3 plants.

#### 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.

## Data availability

No data was used for the research described in the article.

### Acknowledgements

We thank the two anonymous reviewers for their time in reading this manuscript and for providing helpful suggestions.

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