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- Subsurface sediment organic geochemical composition mainly controlled by catchment vegetation properties in eastern Australian lakes

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Contemporary Controls on Terrestrial Carbon Characteristics in Temperate and Sub-Tropical Australian Wetlands

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Abstract A thorough understanding of controls over terrestrial sedimentary organic carbon characteristics in both the present and the past is pivotal to better understand atmospheric CO_2 pathways into depositional sinks such as peats, swamps, and lakes. We explored the relationship between wetland sediment organic matter storage, climate (precipitation, temperature) and catchment vegetation data (catchment vegetation cover in percent; leaf carbon content in g/m^2) by means of multivariate statistical analyses to investigate patterns of carbon deposition in modern wetlands and to provide a more robust framework for interpreting sediment bulk organic geochemistry as a proxy for past carbon cycling. Carbon and nitrogen elemental concentration and stable isotope composition were analyzed from sub-surface sediments at 18 wetlands in eastern Australia. The statistical analyses indicate that variability in geochemical organic matter data in wetland sediments is best explained by geographic differences in catchment vegetation cover and, by inference, the balance of terrestrial versus aquatic organic matter input to the sediment. TOC/TN of aquatic matter may be additionally driven toward higher (terrestrial) values by nitrogen limitation in the catchment and the lakes. These processes explain up to ~40% of the total variance in the sediment geochemistry (redundancy analyses). Up to ~10% of the total variance may be attributed to post-depositional processes and organic matter remineralization. The remaining ~50% of total variance in the data may be attributed to local conditions across the sites, geochemical processes that were not captured in this study, or to the different timescales covered by the sediments at each site.

Plain Language Summary The accumulation of organic material in wetlands depends on the wetland productivity by aquatic plants and algae and on supply of (decomposed) leaf litter and soil material from the catchment. Sedimentary elemental concentrations and ratios (TOC, TN, TOC/TN) and stable isotope compositions of nitrogen and carbon are often used as proxy for the relative contribution of catchment and lake organic matter accumulation in the geological history of wetlands. This information is then used to reconstruct past environments and climates, as well as terrestrial carbon pathways from the atmosphere to a depositional sink. Here, we statistically link the geochemical composition (TOC, TN, TOC/TN, carbon and nitrogen isotopes) of subsurface sediments (upper 4 cm) to catchment vegetation characteristics and climatic data in 18 wetlands from temperate and subtropical Australia. The data provides an interpretative framework of sedimentary organic geochemical data by calibration against contemporary climates and vegetation characteristics. Catchment vegetation cover, nitrogen limitation, runoff mobilization of leaf litter and soil organic matter, and lake productivity are the major control of subsurface geochemical data.

1. Introduction

Terrestrial carbon stored in soils and freshwater environments is estimated to have absorbed about 29% of global anthropogenic CO_2 emissions that occurred between 2008 and 2017 (IPCC, 2021). However, this carbon is only removed from the contemporary atmosphere gas balance if it is effectively stored in a depositional sink such as the sediments of lakes, reservoirs, swamps and estuaries—collectively described as wetlands (Mendonca et al., 2017). In contrast, mineralization and respiration during temporary storage in soils may promote carbon remobilization into the atmosphere (Doetterl et al., 2016). Wetlands in both natural and agricultural landscapes thus play an essential role in atmospheric CO_2 sequestration, despite their low spatial extent in comparison to land

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and oceans (Dean & Gorham, 1998). The efficiency of wetlands as continental CO_2 sinks is particularly dependent on sedimentary accumulation rates: high sedimentation rates enable a higher fraction of settling organic carbon to escape remineralization (Doetterl et al., 2016; Mendonca et al., 2017). These processes are strongly influenced by the type and flux of organic matter to the wetland sediments, including whether the organic matter is composed of higher plant or microbial detritus, and thus the relative productivity and biomass within the wetland compared to that of the surrounding catchment. The processes and controls of CO_2 sequestration in terrestrial wetlands are, however, still difficult to quantify by modern observations (IPCC, 2021). This is even further complicated when considering terrestrial carbon storage under future or past climate conditions beyond the range of those observed today.

The vast temperate and semi-arid to arid regions of the Southern Hemisphere, and in particular Australia, play a particularly intriguing role in global terrestrial carbon sequestration. In Australia, high modern day soil carbon stocks per hectare are found in soils of the temperate east coast where annual precipitation is higher, whilst carbon stocks in the continent's dry interior are significantly lower (Viscarra Rossel et al., 2014). Central parts of Australia however still contribute to terrestrial carbon storage particularly during La Niña climatic events. For example, the exceptional La Niña event between 2010 and 2012, which promoted high precipitation across Australia, resulted in a major expansion of terrestrial biomass and a globally significant increase in soil organic carbon storage (Cleverly et al., 2016; Poulter et al., 2014). More than 60% of the 2010–2012 increase in global carbon sequestration is attributed to Australia alone (Poulter et al., 2014). However, the fate of carbon following subsequent drying is poorly understood, as is the potential response of the Australian landscape during prolonged (decadal to centennial timescale) periods of elevated precipitation. Understanding the relationship between terrestrial carbon sequestration, and unraveling the source (terrestrial vs. aquatic) and characteristics of deposited organic matter in wetlands is therefore important, both for modeling carbon storage under contemporary and future climates, but also to enable reconstructions of past carbon cycling and storage under past wetter and drier climates.

Carbon accumulation in continental wetlands is controlled by both authigenic productivity (e.g., phytoplankton and macrophytes) and allogenic supply (terrestrial soil and plant material from the catchment). Total wetland carbon accumulation thus depends on a variety of factors including climate, catchment vegetation cover, erosion, mixing, and nutrient cycling and availability (e.g., Stallard, 1998). Geological analogs for freshwater carbon accumulation under conditions similar to those predicted for the coming century (IPCC, 2021) can aid assessments of the future role of freshwater systems in the global CO_2 cycle (Tranvik et al., 2009). Lake sediments provide an outstanding archive to simultaneously study terrestrial carbon accumulation, climate, and landscape change using sediment geochemistry, microfossils and molecular and biomarker remains (Cohen, 2003; Francke et al., 2020; Giguet-Covex et al., 2014; Holtvoeth et al., 2019; Thomas et al., 2020). A fundamental prerequisite for such studies, however, is estimates of the relative contribution of authigenic and allogenic matter to the bulk carbon pool in the sedimentary basin. Various analytical methods have been used to estimate the authigenic versus allogenic origin of sediment organic matter, including the concentrations and ratios of total organic carbon and total nitrogen (TOC, TN and TOC/TN; Meyers & Teranes, 2002), the stable isotope ratios of $^{13}C/^{12}C$ and $^{15}N/^{14}N$ ($\delta^{13}C$, $\delta^{15}N$; e.g., Leng et al., 2005), the concentration, molecular structure and isotope composition of specific organic compounds (*n*-alkanes; glycerol dialkyl glycerol tetraethers, e.g., Holtvoeth et al., 2017, 2019), and the molecular evolution of sedimentary organic matter, as evaluated by rock eval pyrolysis (oxygen and hydrogen indices, e.g., Mayr et al., 2008; Lacey et al., 2015). Combining these tracers of organic-geochemical catchment cycling with independent, quantitative estimates of past catchment erosion of detrital matter can then be used to assess the complex interplay between soil-carbon erosion, soil erosion, climate, catchment vegetation cover and final (soil-) carbon deposition in a sedimentary sink (e.g., Chappell et al., 2015; Francke et al., 2020, 2022).

Of the methods to assess catchment-wide carbon cycling, bulk organic geochemical properties—namely elemental concentrations (TOC, TN) and their ratios (TOC/TN), as well as $\delta^{13}C$ and $\delta^{15}N$—are relatively simple and hence commonly applied proxies for the accumulation and origin of organic matter in wetland sediments. However, despite widespread use, very few studies have attempted to critically test the assumptions that underpin this application of bulk organic geochemistry. These indices are controlled by a variety of catchment and internal lake processes including catchment vegetation density and composition (C_3 vs. C_4 vegetation), soil carbon and nitrogen cycling and decomposition, wetland productivity and mixing-driven oxygenation (summarized in Cohen, 2003). For example, selective decomposition of nitrogen during organic matter mineralization may promote higher TOC/TN ratios in sedimentary deposits, and many studies have already elaborated potential overlaps in carbon-nitrogen elemental ratios of aquatic and (soil-) terrestrial plant material (Francke et al., 2016; Holtvoeth et al., 2016). Lake-internal carbon cycling can significantly alter the lake sediment $\delta^{13}C$ if organic matter is composed of a mixture of aquatic and terrestrial sources. For example, selective incorporation of the lighter isotope ^{12}C into phytoplankton may promote increasingly lower lake water $\delta^{13}C$ (and thus lower sedimentary $\delta^{13}C$). In contrast, in settings with (a) strong primary productivity and low dissolved carbon supply, or (b) with high supply of ^{13}C by atmospheric exchange or limestone dissolution, ^{12}C assimilation may be outpaced by replenishment from the atmosphere or catchment, leading to increasingly high $\delta^{13}C$ in lake sediment organic matter. These processes are even more complex in relation to $\delta^{15}N$ where isotope fractionation during nitrogen assimilation is less, but where processes such as denitrification or nitrification may lead to higher or lower lake sediment $\delta^{15}N$ (e.g., Leng & Marshall, 2004; Priestley et al., 2022).

Given the complexities involved, interpretation of organic carbon and nitrogen concentrations and isotopic composition in the sedimentary archive should ideally be supported by modern calibrations and process studies. However, despite hundreds of studies, which apply these tracers to reconstruct past carbon cycling and deposition in sediments, remarkably few investigations have been conducted on the modern environment (Talbot, 1990; Teranes & Bernasconi, 2000; Lehmann et al., 2002; Teranes & Bernasconi, 2005; Moschen et al., 2009; Cadd et al., 2018; Maxson, Tibby, Barr, et al., 2021 to name a few exceptions).

We investigated the association between climate, vegetation and sediment organic matter composition (TOC, TN, TOC/TN, $\delta^{13}C$, $\delta^{15}N$) in the surficial sediments of a series of wetlands across eastern Australia to assess the extent to which broad assumptions regarding the interpretation of these tracers, particularly as proxies for terrestrial carbon input, hold true on a regional scale. The overall aim of this study was to develop a better understanding of the broad controls over organic matter composition and preservation in Australian wetland sediments, to provide a platform for more detailed, process-based studies. We compared sub-surface sediment organic geochemical data with remote sensing-based estimates of vegetation cover, leaf carbon content and local climate data (precipitation, temperature). The selected sites span a wide climate and environmental gradient; from sub-tropical Queensland to semi-arid and temperate sites in New South Wales, Victoria, and Tasmania. The results of this study will provide a more robust basis for interpreting bulk organic geochemistry data from wetland sediments as records of past carbon cycling. By providing insights into the source and characteristics of deposited organic matter in different environmental settings, we provide an important framework of information that will aid understanding long-term carbon storage in Australian wetlands in future studies.

2. Material and Methods

2.1. Material

Geochemical and remote sensing data were obtained for 18 sites located along a roughly north-south transect in eastern Australia, comprising temperate to sub-tropical climates (in Tasmania, Murray-Darling Basin (MDB), North Stradbroke Island Table 1, Figure 1). Eight of the 18 selected sites are located within the MDB. The MDB is Australia's largest exorheic river basin, covering approximately 14% of the continent's land mass (Lu et al., 2006). The basin can be described as predominantly flat and dry, with greater relief and precipitation toward the southern and eastern divides. Most of the catchment's runoff is generated along the highlands along Australia's east coast since aridity generally increases inland (SILO database, see Bureau of Meteorology & Queensland Government, 2021). Additional sites were selected from outside the MDB to provide a broader coverage of both climate and catchment vegetation density.

Subtropical sites with no distinct dry season are located on North Stradbroke Island, near Brisbane, Queensland (QLD). Subtropical sites with moderately dry winters were selected along the Macintyre River that forms the border between QLD and New South Wales (NSW) approximately 340 km inland from the coast. Sites located in temperate climates are located midway between Brisbane and Sydney (NSW) approximately 70 km from the coast (Bishops Swamp, no dry season, warm summer), and along the Murray River at the border between NSW and Victoria (VIC) approximately 300 km inland (no dry season, hot summer). Sites located on Tasmania (TAS) are characterized by temperate climates with no dry season and mild summers. Structural forms of vegetation (following the classification of Specht, 1970) in the lake catchments comprise closed forests for sites on North Stradbroke Island, woodlands for sites in the sub-tropical MDB, open forests for temperate sites in the MDB

Table 1
Location and Characteristics of Wetlands From North Stradbroke Island (NSI), Murry Darling Basin (MDB), and Tasmanian (TAS)

Site	Region	Location	Climate	BOM climate station
Swallow Lagoon	NSI-QLD	27°29′55.63″S 153°27′17.41″E	Subtropical, no dry season	Cape Moreton Lighthouse (40043)
Blue Lake	NSI-QLD	27°31′57.62″S 153°28′36.49″E	Subtropical, no dry season	Cape Moreton Lighthouse (40043)
18 Mile Swamp	NSI-QLD	27°36′5.18″S153°27′55.29″E	Subtropical, no dry season	Cape Moreton Lighthouse (40043)
Pungbougal Lagoon	MDB-NSW (north)	28°39′40.08″S 150°16′14.84″E	Subtropical, moderately dry winter	Mungindi Post Office (52020)
Whynot Billabong	MDB-QLD	28°34′47.08″S 149°29′27.60″E	Subtropical, moderately dry winter	Mungindi Post Office (52020)
Booberoi Lagoon	MDB-QLD	28°29′30.25″S 149°34′44.78″E	Subtropical, moderately dry winter	Mungindi Post Office (52020)
Macintyre Downs Billabong	MDB-QLD	28°36′22.14″S 149°39′11.92″E	Subtropical, moderately dry winter	Mungindi Post Office (52020)
Bishop Swamp	NSW	31°8′33.36″S 152°13′15.63″E	Temperate, no dry season, warm summer	Yarras, Mount Seaview (60085)
Moira Lake	MDB-NSW (south)	35°56′17.25″S 144°56′11.06″E	Temperate, no dry season, hot summer	Echuca Aerodrome (80015)
2 Carp Billabong	MDB-VIC	36°10′35.55″S 146°15′20.42″E	Temperate, no dry season, hot summer	Rutherglen Research (82039)
Dairy Billabong	MDB-NSW (south)	35°58′14.66″S 146°34′44.76″E	Temperate, no dry season, hot summer	Rutherglen Research (82039)
Lake Spicer	TAS	41°58′46.64″S 145°39′39.26″E	Temperate, no dry season, mild summer	Marrrawah (91223)
Dove Lake	TAS	41°39′35.35″S 145°57′42.54″E	Temperate, no dry season, mild summer	Marrrawah (91223)
Lake Basin	TAS	41°58′49.01″S 145°32′54.74″E	Temperate, no dry season, mild summer	Marrrawah (91223)
Lake Gwendolen	TAS	42°15′44.77″S 145°49′24.84″E	Temperate, no dry season, mild summer	Marrrawah (91223)
Lake Godwin Tarn	TAS	42°14′35.71″S 146° 8′3.05″E	Temperate, no dry season, mild summer	Marrrawah (91223)
Lake Rolleston	TAS	41°55′18.59″S 145°37′29.15″E	Temperate, no dry season, mild summer	Marrrawah (91223)
Lake Tahune	TAS	42°16′0.24″S 145°50′8.97″E	Temperate, no dry season, mild summer	Marrrawah (91223)

Note. Three-letter codes refer to the states of Queensland (QLD), New South Wales (NSW), Victoria (VIC), and Tasmania (TAS). Climate classifications are inferred from the Bureau of Meteorology (BOM, Australia). BOM climate stations refer to the stations used to infer climate data reported in Table 3.

and at Bishops Swamp, and more densely vegetated catchments for Tasmanian sites. Natural vegetation may be significantly disturbed by agricultural use, particularly for the sites in the MDB.

2.2. Geochemical Analysis

A total of 70 sub-surface sediment samples were analyzed for all sites down to 4 cm depth (Table S1). All samples were collected as part of previous research projects over the last 10 years, and stored at 4°C, following standard procedures in national and international lake drilling projects (e.g., Wilke et al., 2016). While sample storage might affect lipid biomarker and complex organic compounds, it is rather unlikely to affect C and N isotopic composition and elemental concentrations. Sediments were subsampled at 0.5 cm intervals for sites in Tasmania, 1 cm intervals for sites from North Stradbroke Island, and 4 cm for sites from the MDB depending on availability of sediment material. Samples with significant quantities of moisture were centrifuged for approximately 2 min to collect the sediment at the bottom of the tube, and the excess water was decanted. All samples were freeze-dried and ground to fine powder using a ball mill prior to further analyses.

An Elementar elemental analyser linked by continuous flow to a Nu Horizon isotope ratio mass spectrometer was used for elemental (TOC, TN) and stable isotope ($\delta^{13}C$, $\delta^{15}N$) analysis of organic matter. TOC/TN ratios are reported as mass organic carbon over mass nitrogen. Standardisation and quality control was obtained against

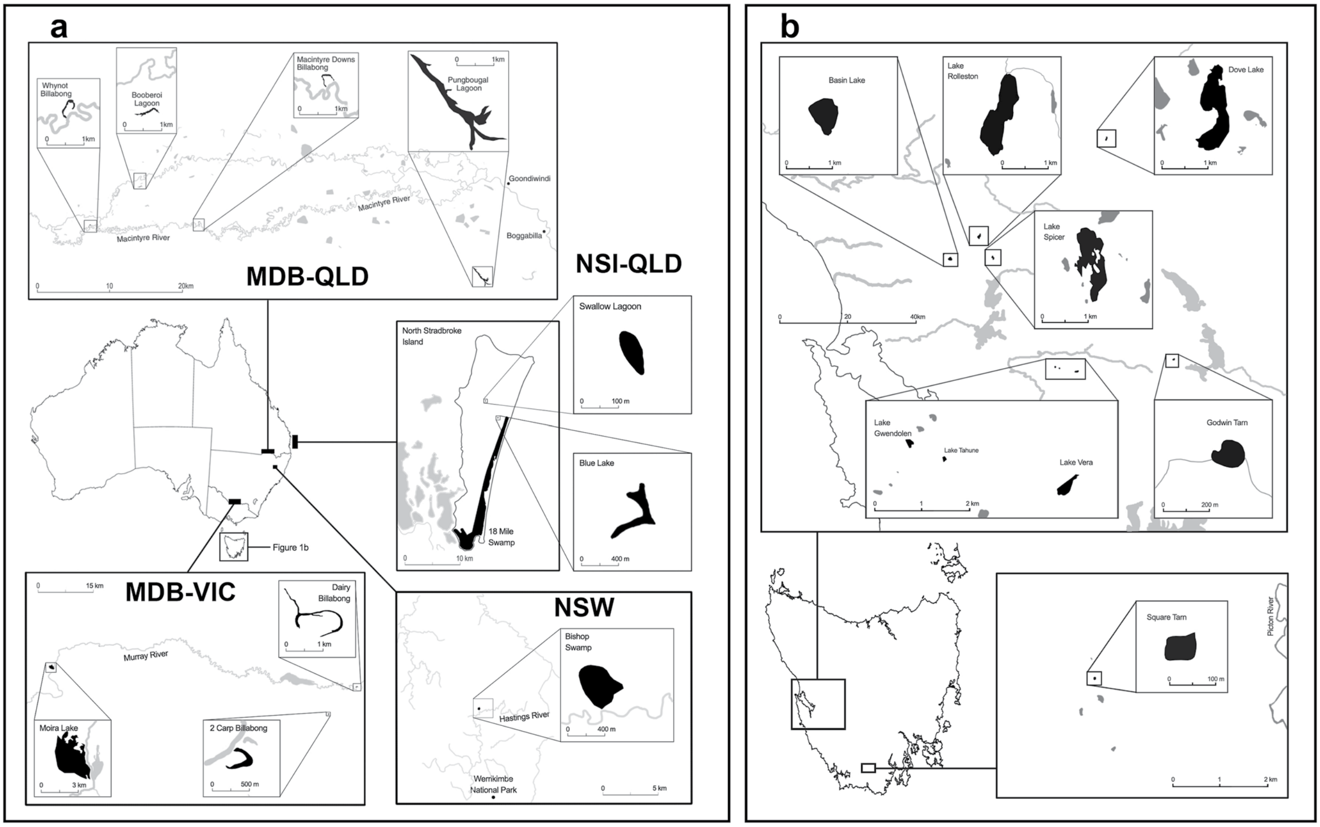

Figure 1. Locations of sites investigated in this study on mainland Australia (1a) and Tasmania (1b). Acronyms in 1a summarize different regions of the studied lakes as listed in Table 1.

in-house standards glycine, glutamic acid, and triphenylamine, calibrated against external standards. Carbon and nitrogen isotope ratios are reported in per mille units relative to the Vienna Pee Dee Belemnite standard (VPDB) for carbon and standard AIR for nitrogen. Acid fumigation (Harris et al., 2001) was applied prior to carbon isotope and TOC analyses to remove any inorganic carbon. Geochemical data were averaged for the upper 4 cm sediment depth prior to multivariate statistical analyses and comparison to meteorological and vegetation data.

2.3. Modern Climate and Vegetation Data

Climate data for each site were inferred for the period 2000 to 2013 using continuous climate data series for temperature and precipitation retrieved from the nearest climate station operated by the Australian Bureau of Meteorology, exported via the SILO database (Jeffrey et al., 2001; Bureau of Meteorology, 2021, see Table S2). The distances between the studied sites and the climate stations are <60 km for sites at North Stradbroke Island; <80 km for sites in MDB-Queensland; <30 km for sites in MDB-Victoria and southern NSW, <30 km for Bishops Swamp, and <150 km for sites in Tasmania. Seasonal and annual average precipitation and temperature data were calculated prior to further statistical analyses.

Catchment vegetation characteristics at each site were quantified as total vegetation cover (in percent of total catchment area, %) and leaf carbon content (in gram per meter square catchment area, g/m^2, see Table S3). Total vegetation cover at each site was inferred from the "MODIS Vegetation Fractional Cover Algorithm" (Guerschman, 2021), courtesy of the Commonwealth Scientific and Industrial Research Organisation (CSIRO) Australia, Terrestrial Ecosystem Research Network (TERN) Australia, and the Australian Government Department of Agriculture. The Vegetation Fractional Cover represents a raster-map of the exposed proportion of photosynthetic vegetation (PV), non-photosynthetic vegetation (NPV) and bare soil. The MODIS algorithm calculates the vegetation cover as the sum of PV and NPV as seasonal (summer, winter) and annual mean raster data. MODIS annual mean vegetation cover between 2000 and 2013 was estimated for each site by constraining the lakes' catchments against a digital elevation model (DEM) obtained from the Elevation Information System (ELVIS, Geoscience Australia). The accuracy of the estimated catchment outline is dependent on the surrounding topography. Catchments located within more pronounced topographies (predominantly sites located in Tasmania) are likely to have better defined catchment area outlines. Using ArcGIS software, monthly data for each catchment was obtained by converting the DEM raster into contour raster using ArcGIS contour tool. Site-specific remote sensing data was then extracted using a catchment polygon as mask and the pixel values (of the remote sensing data) and their catchment mean is extracted for each month and each year.

Leaf carbon content at each site was inferred from the BIOS2 coupled carbon and water cycling modeling system, which was developed for the Australian Water Availability Project (Haverd, Raupach, Briggs, Canadell, et al., 2013; Haverd, Raupach, Briggs, Davis, et al., 2013; King et al., 2009; Raupach et al., 2009). The BIOS2 modeling environment is forced using gridded meteorological data, soil properties and vegetation cover at 0.05° spatial resolution. Leaf carbon content (reported as g/m^2) are modeled as the difference between "net primary production" (NPP) and "leaf decay" over time as part of BIOS2's carbon sub model. By accounting for carbon decay, the modeled data provides a theoretically more accurate estimate of catchment carbon contents that may be delivered to the wetlands. Monthly leaf carbon content between 2000 and 2013 was inferred for each catchment constrained by the DEM. Seasonal and annual average leaf carbon contents were calculated prior to further statistical analyses. The time window covered by the climate and catchment vegetation data is likely shorter compared to the time covered by the top 4 cm at each site. The resulting implications and limitations are discussed in more detail below.

2.4. Statistical Analyses

We used principal component (PCA) and redundancy analysis (RDA) to examine the relationship between the geochemical, climate and vegetation data from North Stradbroke Island, the MDB, and Tasmania. Significance of climatic and vegetation predictor data on each geochemical variable was tested prior to RDA analyses. This was achieved by computing importance measures in a tree by summing changes in the node risk due to splits on every predictor, and then dividing the sum by the total number of branch nodes. Averaged geochemical data for the upper 4 cm at each site were used as response variables, and average annual climate and vegetation data were the predictor variables. All variables were standardised to have a mean of zero and a standard deviation of one prior to statistical analyses, and an ANOVA permutation test was carried out alongside RDA to test the significance of all canonical axes. Data processing and statistical analyses were carried out using MATLAB R2020a and the Fathom toolbox (Jones, 2017).

3. Data

3.1. Geochemical, Vegetation, and Climate Data

The average elemental concentrations within the upper 4 cm in all wetlands range between 0.4% and 40.9% for TOC, and between 0.15% and 2.91% for TN, respectively (Table 2, see Table S1 for full data set). Elemental ratios (TOC/TN) fall between 2.8 and 27.2 (Figure 2, Table 2). $\delta^{13}C$ range between −32.10‰ and −22.33‰ and $\delta^{15}N$ range between −2.76‰ and 6.48‰ (Figure 2, Table 2). Overall low standard deviations (1σ) for TOC (<3.34%), TN (<0.21%), TOC/TN (<2.34), $\delta^{13}C$ (<0.47‰), and $\delta^{15}N$ (<0.51‰) indicate that the upper 4 cm of sediments are geochemically homogenous (Table 2).

Average monthly precipitation for 2000 to 2013 at all sites range between 33.1 and 197.5 mm, and the average annual average precipitation between 397.7 and 1,039.7 mm, respectively (Table 3, see Table S2 for complete data set). More average summer precipitation was recorded for North Stradbroke Island sites and those located in the northern part of the MDB and in NSW (MDB-QLD, MDB-NSW (north), Tables 1 and 3). Sites located within the southern MDB (MDB-VIC, MDB-NSW (south)) and in Tasmania had more winter precipitation (Table S3). Average annual temperatures ranged between 16.3°C (Tasmania) and 28.4°C (MDB-QLD), with average winter and summer temperatures between 19.9°C (in Tasmania) and 34.8°C (in MDB-QLD), and 12.9°C (in Tasmania) and 20.8°C (in MDB-QLD), respectively (Table S2).

Average annual catchment vegetation cover (for the years 2000–2013) was <50% at most sites located in the MDB (Table 4, Table S3). Higher average annual catchment vegetation cover >50% was recorded for sites located

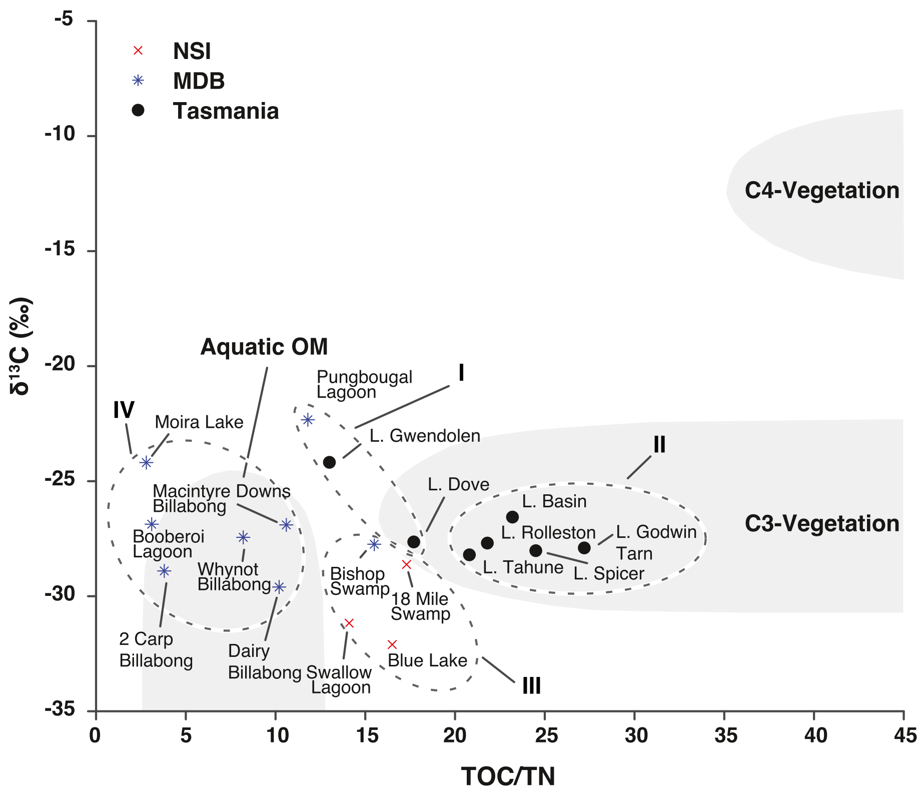

Figure 2. Scatter plot for $\delta^{13}C$ versus TOC/TN to identify OM sources in the investigated sediments. Gray-shaded areas indicate C-4, C-3, and aquatic dominated OM sources and are inferred from Leng et al. (2005). Circles with dotted lines summarize sites that plot in RDA-sectors I-IV as shown in Figure 3.

at North Stradbroke Island and Tasmania, as well as for Bishop Swamp (NSW) and Moira Lake (MDB-NSW (south)). Average catchment vegetation cover during summer and winter was overall similar to average annual catchment vegetation cover for most sites (Table S3). Only one site, 2 Carp Billabong (MDB-VIC), yielded notably lower catchment vegetation cover during summer compared to annual and winter catchment vegetation cover. This is also supported by similar annual, summer, and winter leaf carbon content at all sites, generally ranging between 108 and 479 g/m^2 (mean annual), between 109 and 485 g/m^2 (summer), and between 104 and 471 g/m^2 (winter), respectively (Table S3).

3.2. Statistical Analyses

PCA analyses using geochemical data (TOC, TN, TOC/TN, $\delta^{13}C$, $\delta^{15}N$) yielded significant variance for principal component (PC) I (62.07%) and PC II (16.26%, together 78.33%, Figure 3). Positive loadings on PC I are reported for TOC, TN, and TOC/TN, negative loadings for $\delta^{13}C$ and $\delta^{15}N$. Only $\delta^{13}C$ and TOC/TN show strong loadings on PC I, and TN shows negative loadings on PC II; TOC and $\delta^{15}N$ show weak loadings on PC II only. Significance tests of predictor variables showed that precipitation has only weak predictive importance on the geochemical data, and a co-correlation of precipitation, leaf carbon content, and vegetation cover (Table S4). RDA was therefore carried out with and without precipitation as predictor variable (Figure 3). Both RDA analyses carried out with and without precipitation, as well as with temperature, leaf carbon content and catchment vegetation cover as explanatory variables, and organic-geochemical data (TOC, TN, TOC/TN, $\delta^{13}C$, $\delta^{15}N$) as response variables yielded statistically robust results (Figure 3, explanatory variance of canonical axis I and II ~40% and ~10%, p-value < 0.05). Strong positive loadings on canonical axes I are reported for catchment vegetation cover, leaf carbon content, and precipitation (if included in RDA)) as explanatory variables, and for TOC, TN, and TOC/TN as response variables (Figure 3). $\delta^{15}N$, TN, TOC as response variables, and temperature as explanatory variable show a strong negative loading on canonical axis II for both RDA, whilst $\delta^{13}C$ and TOC/TN yield strong positive loadings on canonical axis II. Precipitation has a negative loading on canonical axis II

Table 2
Geochemical Results

					$\delta^{13}C$		$\delta^{15}N$		TOC		TN		TOC/TN	
Site	Region	Sample depth (cm)	Sample res. (cm)	Number of samples	Avg. (‰)	1σ (‰)	Avg. (‰)	1σ (‰)	Avg. (%)	1σ (%)	Avg. (%)	1σ (%)	Avg.	1σ
Swallow Lagoon	NSI-QLD	0–5	1	4	−31.17	0.067	−2.76	0.087	41.0	0.83	2.91	0.061	14.1	0.5
Blue Lake	NSI-QLD	0–5	1	4	−32.10	0.473	2.15	0.506	11.2	3.25	0.68	0.183	16.5	1.83
18 Mile Swamp	NSI-QLD	0–5	1	4	−28.62	0.391	3.3	0.221	18.7	3.34	1.09	0.208	17.3	0.95
Pungbougal Lagoon	MDB-NSW (north)	0–4	4	1	−22.33	na	5.59	na	4.3	na	0.36	na	11.8	na
Whynot Billabong	MDB-QLD	0–4	4	1	−27.44	na	3.51	na	1.4	na	0.17	na	8.2	na
Booberoi Lagoon	MDB-QLD	0–4	4	1	−26.87	na	5.5	na	0.6	na	0.18	na	3.1	na
Macintyre Downs Billabong	MDB-QLD	0–4	4	1	−26.90	na	6.48	na	3.9	na	0.37	na	10.6	na
Bishop Swamp	NSW	0–4	4	1	−27.74	na	3.27	na	27.2	na	1.75	na	15.5	na
Moira Lake	MDB-NSW (south)	0–4	4	1	−24.19	na	3.86	na	0.4	na	0.15	na	2.8	na
2 Carp Billabong	MDB-VIC	0–4	4	1	−28.90	na	3.29	na	0.7	na	0.19	na	3.8	na
Dairy Billabong	MDB-NSW (south)	0–4	4	1	−29.60	na	2.68	na	3.8	na	0.37	na	10.2	na
L. Spicer	TAS	0–5	0.5	8	−28.02	0.107	1.91	0.069	24.3	1.75	0.99	0.082	24.5	0.67
Dove L.	TAS	0–5	0.5	8	−27.64	0.2	1.15	0.376	4.2	1.72	0.25	0.114	17.7	2.34
L. Basin	TAS	0–5	0.5	8	−26.56	0.093	1.58	0.552	11.9	1.81	0.52	0.105	23.2	3.94
L. Gwendolen	TAS	0–5	0.5	8	−24.18	0.103	0.7	0.219	9.0	1.86	0.7	0.16	13	0.82
L. Godwin Tarn	TAS	0–5	0.5	8	−27.90	0.057	1.25	0.312	16.6	3.48	0.61	0.12	27.2	1.37
L. Rolleston	TAS	0–5	0.5	5[a]	−27.69	0.146	1.09	0.231	17.1	1.03	0.78	0.068	21.8	0.65
L. Tahune	TAS	0–5	0.5	5[a]	−28.20	0.055	2.56	0.08	11.1	1.05	0.54	0.014	20.8	2.48

Note. Average and 1σ refers to the number of samples analyses for the top 4 cm at each site. See Supporting Information S1 for full data set.
[a]See Supporting Information S1 for horizons with missing data.

if included in redundancy analyses (Figure 3). Correlations (R, R^2, and *p*-values) between all explanatory and response variables are reported in the Supplementary Information (Table S4).

The PCA and both RDA-biplots highlight the clear separation of sites originating from the MDB (incl. lakes from QLD, NSW, and VIC), North Stradbroke Island, and Tasmania into sectors I to VI (Figure 3). Bishops Swamp, located in the eastern tablelands of NSW, plots in sector III within the North Stradbroke Island cluster for all tested climate scenarios (Figure 3). All sites in Tasmania except for Lake Gwendolen and Dove Lake plot within sector II for all three RDA analyses. All sites from the MDB except for Pungbougal Lagoon plot within sector IV. Pungbougal Lagoon, Lake Gwendolen, and Dove Lake plot within sector I for all RDA biplots. The described clustering of sites within our RDA analyses can be observed in Figure 2. Sites in sector II generally plot alongside organic matter characterized by $\delta^{13}C < -22$‰ and TOC/TN > 20 (Figure 2). Sites in sector IV generally exhibit TOC/TN < 13 and $\delta^{13}C < -25$‰ (Figure 2). Sites within sectors I and III exhibit geochemical values which fall between the classically defined C3 terrestrial and aquatic organic matter sources.

RDA analyses were also carried out on regional subsets of data from North Stradbroke Island, the MDB, and Tasmania, but yield results that are statistically not robust (*p*-value >> 0.05, not shown). This can probably be attributed to the low number of sites or to the low variability within climate and vegetation data for each region.

In order to visualize the relationship between organic matter content and catchment vegetation cover, as implied by the RDA analysis, we plotted biplots of these values. Simple linear regressions indicate correlation coefficient of $R^2 = 0.48$ between vegetation cover and TOC, and $R^2 = 0.5$ between vegetation cover and TOC/N (Figure 4). The correlation coefficient increases to $R^2 = 0.64$ for TOC versus vegetation cover if Swallow Lagoon (North Stradbroke Island) is excluded. Furthermore, the distribution of values falls along a relatively even continuum, with no notable outliers exerting leverage over these relationships. Similarly, the linear models determined for regional subsets are similar to those defined for the whole data set (Table S1), suggesting that the relationship

Table 3
Climate Data

		Monthly averaged precipitation (mm)		Monthly averaged temperatures (°C)	
Site	Region	Year	Year 1σ	Annual	Annual 1σ
Swallow Lagoon	NSI-QLD	80.7	18	24.1	0.3
Blue Lake	NSI-QLD	80.7	18	24.1	0.3
18 Mile Swamp	NSI-QLD	80.7	18	24.1	0.3
Pungbougal Lagoon	MDB-NSW (north)	41.2	13.7	28.4	0.9
Whynot Billabong	MDB-QLD	41.2	13.7	28.4	0.9
Booberoi Lagoon	MDB-QLD	41.2	13.7	28.4	0.9
Macintyre Downs Billabong	MDB-QLD	41.2	13.7	28.4	0.9
Bishop Swamp	NSW	197.5	58	24.6	0.7
Moira Lake	MDB-NSW (south)	33.1	12	23	0.6
2 Carp Billabong	MDB-VIC	45.9	15.7	22.4	0.7
Dairy Billabong	MDB-NSW (south)	45.9	15.7	22.4	0.7
L. Spicer	TAS	86.6	10.6	16.3	0.3
Dove L.	TAS	86.6	10.6	16.3	0.3
L. Basin	TAS	86.6	10.6	16.3	0.3
L. Gwendolen	TAS	86.6	10.6	16.3	0.3
L. Godwin Tarn	TAS	86.6	10.6	16.3	0.3
L. Rolleston	TAS	86.6	10.6	16.3	0.3
L. Tahune	TAS	86.6	10.6	16.3	0.3

Note. Average and 1σ data of each year analyzed (2000–2013). See Supplement for full data set.

between sediment organic matter and catchment vegetation is continuous, and not solely driven by differences between regions.

4. Discussion

4.1. Sediment-Geochemical Characteristics

Where multiple analyses were feasible for sites on North Stradbroke Island and Tasmania, low 1σ standard deviations in TOC, TN, TOC/TN, $\delta^{15}N$, and $\delta^{13}C$ imply that sediment organic geochemical composition remained broadly constant with depth throughout the analyzed deposits (Table 2). The top 4 cm of sediments likely account for several centuries for sites in Tasmania and Blue Lake on North Stradbroke Island (e.g., Mariani et al., 2017; Maxson, Tibby, Barr, et al., 2021; Schneider et al., 2020), and for several decades for sites located in the MDB and Swallow Lagoon and 18 Mile Swamp on North Stradbroke Island (Barr et al., 2019; Kattel et al., 2016; Mettam et al., 2011; Reid, Chilcott, & Thoms, 2017) as inferred from previous studies on the sites studied herein and other sites from their vicinity. The homogenous nature of the sediments despite the extended time cover for the Tasmanian sites may be due to physical or biological mixing of the uppermost sediments or to their (partly remote) location in relatively protected national parks, preventing major post-colonial anthropogenic disturbance in recent decades, both in the catchment and wetlands. This presents a sharp contrast to the wetlands in the MDB, which has been subjected to intensive agriculture and water extraction (Gell et al., 2005; Reid, Fluin, et al., 2017).

Using the conventional interpretative framework (e.g., Meyers and Lallier Verges, 1999), average TOC/TN ratios between 2.8 and 27.2 and $\delta^{13}C$ values between −32.1‰ and −22.3‰ imply that sub-surface sediments at all sites predominantly comprise either aquatic or C3 terrestrial organic matter (Table 2, Figure 2, e.g., Meyers & Lallier-Vergès, 1999; Leng et al., 2005). Organic matter derived from terrestrial C4 vegetation is not implied by any of the $\delta^{13}C$ data (Figure 2). C3-dominated organic matter sources are consistent with catchment vegetation compositions at all sites (Biodiversity Assessment Working Group, 2009; Munroe et al., 2022).

Table 4
Catchment Vegetation Data

		Monthly averaged leaf carbon content (g/m^2)		Monthly averaged vegetation cover (%)	
Site	Region	Year	Year 1σ	Year	Year 1σ
Swallow Lagoon	NSI-QLD	310.73	8.46	71.69	3.25
Blue Lake	NSI-QLD	341.11	32.96	73.94	1.58
18 Mile Swamp	NSI-QLD	341.11	32.96	82.85	3.96
Pungbougal Lagoon	MDB-NSW (north)	143.90	20.02	21.13	7.54
Whynot Billabong	MDB-QLD	113.81	13.71	36.68	6.78
Booberoi Lagoon	MDB-QLD	136.98	24.85	29.80	6.03
Macintyre Downs Billabong	MDB-QLD	108.39	14.52	29.59	8.40
Bishop Swamp	NSW	478.96	24.66	87.38	5.44
Moira Lake	MDB-NSW (south)	150.65	20.95	46.45	5.51
2 Carp Billabong	MDB-VIC	163.27	23.74	39.70	11.09
Dairy Billabong	MDB-NSW (south)	158.23	28.06	62.79	6.17
L. Spicer	TAS	210.06	19.01	79.15	3.98
Dove L.	TAS	251.94	17.80	69.92	2.27
L. Basin	TAS	221.62	34.75	76.10	4.58
L. Gwendolen	TAS	254.43	25.46	47.81	6.08
L. Godwin Tarn	TAS	256.34	22.69	76.36	2.47
L. Rolleston	TAS	190.86	14.98	76.73	4.26
L. Tahune	TAS	239.69	18.09	52.67	8.09

Note. Average and 1σ reports data of each years analyzed (2000–2013). See Supporting Information S1 for full data set.

Sediment organic geochemistry in Tasmanian lakes implies that they are mostly dominated by C3 vegetation sources. The only exception is Lake Gwendolen, where the subsurface sediments are characterized by overall lower TOC/TN and higher δ^{13}C compared to the other Tasmanian sites, possibly indicating somewhat higher contributions of aquatic algae and macrophyte-derived organic matter. Western Tasmanian lakes are thought to receive substantial amounts of terrestrially derived organic matter (Buckney & Tyler, 1973; Mariani et al., 2018). However, acidic soils poor in nutrients may be the main source for nitrogen and other nutrient supplied to the lakes (Fletcher et al., 2014; Mariani et al., 2018). This may complicate the interpretative framework of TOC/TN ratios in Tasmania, since nitrogen depletion may have artificially shifted aquatic-derived TOC/TN toward more terrestrial endmembers (Fletcher et al., 2014). However, Mariani et al. (2018) demonstrated that a shift from rainforest to sclerophyll dominated catchment vegetation characteristics likely promoted a reduction in peat depth, and a drop in lake sediment TOC/TN from ∼20 to 18, demonstrating that the elemental ratio still records a terrestrial signal.

Sub-surface sediments with TOC/TN < 12, from sites located within the MDB in QLD, NSW, and VIC, would be interpreted to reflect a higher contribution from aquatic organic matter, derived from either algae or submerged aquatic macrophytes (Table 2, Figure 2, e.g., Meyers & Lallier-Vergès, 1999; Leng et al., 2005). TOC/TN and δ^{13}C from Bishops Swamp, located in the vicinity of the MDB along the east coast of NSW, suggest a more terrestrial dominated organic matter source (Figure 2). Values from the three North Stradbroke Island sites plot outside the traditionally defined range for both aquatic and C3 dominated organic matter, which may reflect a mixture of aquatic and terrestrial-derived organic sources as well as additional processes affecting TOC/TN and δ^{13}C. Previous studies demonstrated that organic matter accumulated in Blue Lake and other North Stradbroke Island sites (Cadd et al., 2018; Maxson, Tibby, Barr, et al., 2021; Maxson, Tibby, Marshal, et al., 2021) is predominantly of aquatic origin, despite an elemental and carbon isotope signature more characteristic for terrestrial sources. This has been attributed to N-limitation artificially increasing TOC/TN, similar to the process described for Tasmania (Maxson, Tibby, Barr, et al., 2021; Maxson, Tibby, Marshal, et al., 2021).

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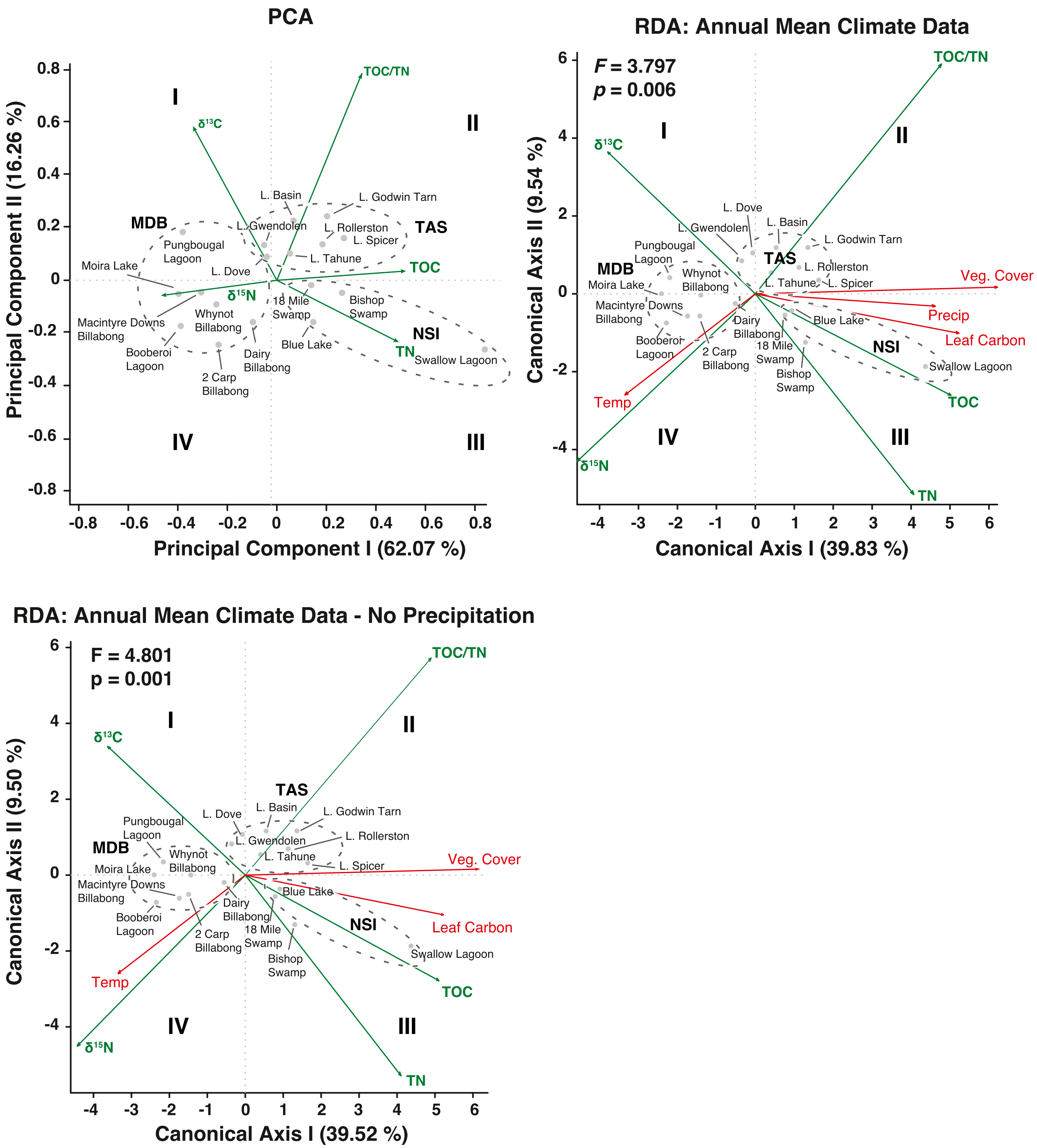

Figure 3. Top panel: PCA-biplot for organic-geochemical data (TOC, TOC/TN, TN, $\delta^{13}C$, and $\delta^{15}CN$). Middle and bottom panel: RDA-biplots for annual mean climate data with and without precipitation. Organic-geochemical data are used as response variables (green), catchment vegetation and climate data are used as explanatory data (red). Roman numbers refer to RDA sectors I-IV. MDB, NSI, and TAS summarize sites located in the Murray Darlin Basin, on North Stradbroke Island, and on Tasmania, respectively. Canonical axes I and II account for approximately 50% of the total variance recorded in the data.

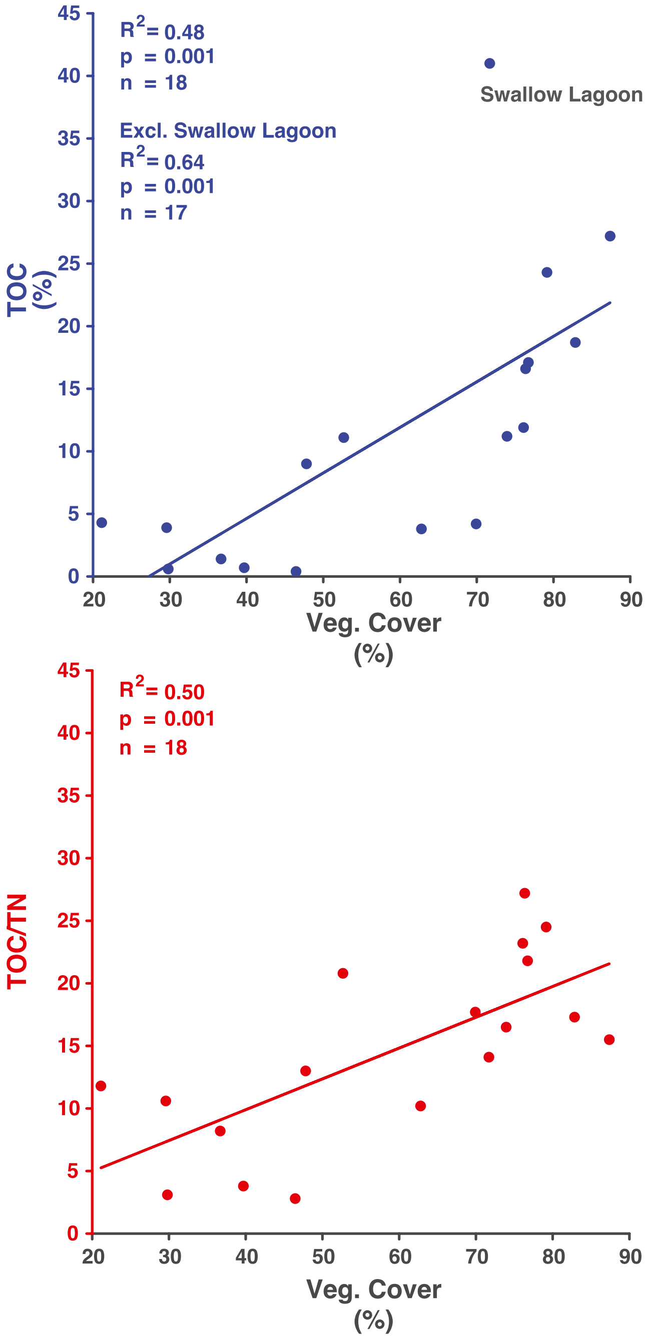

Figure 4. Simple linear regression models between total organic carbon (TOC) content and catchment vegetation cover, and between total organic carbon to total nitrogen ratios (TOC/TN) and catchment vegetation cover, respectively. The correlation coefficient increases to $R^2 = 0.64$ for TOC versus vegetation cover if Swallow Lagoon (North Stradbroke Island) is excluded.

4.2. Climatic and Vegetation Controls on Sediment Biogeochemistry

The positive loadings of vegetation cover and leaf carbon content as explanatory variables, and of TOC, TN and TOC/TN as response variables, and for both RDAs on canonical axis I imply a strong control of catchment vegetation characteristics on the organic geochemical characteristics of contemporary sediments at all wetlands. Overall, denser catchment vegetation cover, as reflected by vegetation and leaf carbon content, promotes higher organic matter concentration in sediments (high TOC and TN), of which a greater proportion is derived from terrestrial sediments (higher TOC/TN). This is also supported by the PCA results and the linear regression models between TOC and TOC/TN and vegetation cover (Figures 3 and 4, Table S4). These observations support the conventional interpretation of TOC/TN in wetland sediments as a proxy for terrestrial organic matter input, and further suggest that catchments with higher terrestrial productivity result in a greater flux of carbon to wetland sediments, proportional to dilution by inorganic matter and loss to remineralization. The finding that denser catchment vegetation cover corresponds to increased organic carbon deposition in wetlands may have important implications about the significance of Australian wetlands' role in the terrestrial carbon cycle. The finding implies that increased terrestrial biomass production on geological time scales has the potential to increase atmospheric carbon sequestration and permanent carbon burial in Australian landscapes, which was previously discussed for annual to decadal timescales from modern observations (Chappell et al., 2015; Cleverly et al., 2016; Poulter et al., 2014). However, more detailed research on the interplay between vegetation cover, catchment erosion, and final carbon burial is needed to validate the underlying chemical and physical processes at geological timescales (Francke et al., 2022).

The positive response of TOC/TN to increased catchment vegetation cover could additionally be promoted by nitrogen depletion in more vegetated sites on Tasmania and North Stradbroke Island, as outlined above. Furthermore, both the terrestrial biomass indices TOC and TOC/TN plot alongside precipitation if considered (Figure 3). However, significance tests prior to RDA analyses revealed only weak effects of precipitation upon geochemical data. The weak effect of precipitation is also supported by low correlation coefficients ($R^2 < 0.41$) of precipitation versus all geochemical parameters (Table S4). It is well established that higher precipitation promotes greater terrestrial productivity in Australian catchments (Cleverly et al., 2016), probably increasing leaf litter and soil organic carbon content (Viscarra Rossel et al., 2014). This is also supported by correlations of precipitation to leaf carbon content ($R^2 = 0.77$) and vegetation cover ($R^2 = 0.5$, Table S4), suggesting that the weak RDA significance of precipitation is probably attributed to co-correlations between the three variables. This likely implies a complex, indirect control of precipitation, by (a) increasing vegetation cover, and terrestrial biomass accumulation, and (b) increased runoff in the studied sites. Conversely, negative loadings on canonical axis I are probably indicative of more aquatic-dominated depositional environments (lower TOC/TN). The negative loadings of $\delta^{13}C$ and $\delta^{15}N$ on canonical axis I would consequently imply that part of the variance in carbon and nitrogen isotope signatures in these aquatic-dominated systems is controlled by ongoing selective assimilation and deposition of the lighter isotopes ^{12}C and ^{14}N during algae and macrophyte growth (Rayleigh fractionation), promoting higher $\delta^{13}C$ and $\delta^{15}N$ values relative to terrestrial sources (see also Figure 2, Talbot, 1990; Leng & Marshall, 2004; Leng et al., 2005).

Rayleigh fractionation during aquatic primary productivity was proposed to explain Holocene $\delta^{15}N$ variability at Dove Lake, Tasmania (Mariani et al., 2018). It may also explain relatively high $\delta^{13}C$ between −25.5‰ and −22‰ and $\delta^{15}N$ between 2‰ and 4‰ (compared to values reported in this study, Figure 2) for sub-surface sediments from Kings Billabong in the Murray River floodplain (MDB-VIC), where OM is thought to mainly consist of phytoplankton and aquatic macrophyte remains (Kattel et al., 2016). Higher sedimentary $\delta^{15}N$ in areas of intensive agriculture, such as in the MDB, may additionally be affected by land use and nitrogen pollution. Only up to 41% of nitrogen introduced as fertilizer is efficiently used by agricultural plants (Chen et al., 2008; Kattel et al., 2016), and (mineralized animal waste and sewage (grazing) can drive ^{15}N-enriched inorganic nitrogen supply, as demonstrated for Lake Alexandria in South Australia (Herczeg et al., 2001).

Whilst there is evidently a geographic influence on the associations between land cover and sediment organic geochemistry, close inspection of the sites in the PCA and RDA analysis indicates that patterns within the regional sub-divisions further highlight the overriding dominance of land cover. For example, Lake Gwendolen (Tasmania), plot in sector I (Figure 3) and exhibit TOC/TN and $\delta^{13}C$ values commonly associated with aquatic OM dominance (Figure 2). This can probably be attributed to the high-altitude location resulting in a somewhat more open and less dense catchment vegetation cover (Table 4, 47.8% vegetation cover) and thus, lower supply of C-3 derived terrestrial OM to the lakes. Bishop's Swamp (NSW), by contrast, has higher TOC/CN and lower $\delta^{13}C$, consistent with a higher proportion of C3 terrestrial OM contribution, contrasting with other sites in the MDB and exhibiting greater similarity to wetlands in Tasmania and NSI (Table 4, Figures 2 and 3, 87.4% vegetation cover).

Evidence that the sedimentary bulk stable isotope compositions are controlled by additional processes than simple aquatic primary productivity versus terrestrial input may come from the poor correlation ($R^2 = 0.2$, $p = 0.06$) between $\delta^{15}N$ and $\delta^{13}C$. This poor correspondence is also supported by the RDA and PCA analyses, where $\delta^{15}N$ and $\delta^{13}C$ show negative and positive loadings on canonical axis II and principal component axis II (Figure 3). The poor correlation may be attributed to the complex behavior of $\delta^{15}N$ in general, and further amplified by the wide range of sites investigated herein. For example, wetlands on North Stradbroke Island and Tasmania are generally nitrogen limited (Cadd et al., 2018; Fletcher et al., 2014; Mariani et al., 2018; Maxson, Tibby, Barr, et al., 2021). Nitrogen limitation should result in $\delta^{15}N$ values reflecting those of source nitrogen rather than isotope fractionation within the wetland linked to primary productivity (Cadd et al., 2018; Leng et al., 2005; Talbot, 2002). Catchment $\delta^{15}N$ values can have a wide range of values between −2 and +10‰ for terrestrial plants, 0‰ for atmospheric N, and ∼+5‰ for soil derived N (Leng et al., 2005; Talbot, 2002). Nitrogen export from the catchment has been reported to be particularly low in Australian catchments that are more densely vegetated (sites on Tasmania, North Stradbroke Island, Bishops Swamp) as outlined above for TOC/TN; conditions that might promote N-limitation at sites with long lake-water residence times (Harris, 2001). Moreover, terrestrial vegetation tends to support low $\delta^{15}N$ (<0‰) in N-poor high precipitation environments such as at North Stradbroke Island (Koopmans et al., 1997; Schulze et al., 2014). This may explain the low $\delta^{15}N < 0$‰ (Table 2) at Swallow Lagoon, where organic matter accumulation is mainly dominated by accumulation of leaf litter (Barr et al., 2019).

The positive loadings of $\delta^{13}C$ and negative loadings for $\delta^{15}N$ on canonical axis II and principal component axis II, which accounts for approximately 10% and 16% of the total variance recorded in all RDAs and the PCA, probably implies that a secondary process has additional significant control on the organic geochemical characteristics of sediments at all sites. This may be a process that cannot be attributed to site-specific conditions, such as the leaf litter dominance in OM at Swallow Lagoon. This may also be supported by the positive loading of TOC/TN, and negative loadings of TOC and TN, indicating that a common feature controls elemental and stable isotope compositions at all sites. A positive correspondence between $\delta^{13}C$ and TOC/TN, which simultaneously promotes a negative correspondence between these variables and $\delta^{15}N$, TN, and TOC (as inferred from RDA results; Figure 3) may be attributed to lake mixing and organic matter mineralization before, during or after sediment deposition. Oxygenation of water columns and sediments by mixing accelerates respiration of organic matter and results in the loss of carbon to the atmosphere. Preferential respiration of ^{12}C would result in an increase in the $\delta^{13}C$ of residual organic matter (Leng et al., 2005). For south-eastern Australia, this interpretation is supported by monitoring of the Murray River in the MDB, which highlighted that the river's dissolved and particulate carbon pool is mainly controlled by organic matter remineralization (Cartwright, 2010). Organic matter remineralization may also explain the positive loading of TOC/TN (on canonical axis and principal component axis II) when driven toward higher values by early digenetic selective loss of N (e.g., Cohen, 2003). A directional change toward higher $\delta^{13}C$ and TOC/TN may also be controlled by soil-organic matter decomposition in the catchment (Krull et al., 2006), which is often neglected in palaeo-studies. This is likely due to difficulties in assessing past soil properties such as soil texture, structure, pH, and composition, all of which affect soil-organic matter decomposition in the catchment, and the difficulties in untangling soil- and depositional decomposition in the palaeo-archive. Whereas respiration of organic matter can result in marked changes in $\delta^{13}C$, the equivalent effects upon $\delta^{15}N$ are thought to be relatively small (Leng et al., 2005). However, microbial reworking and subsequent nitrification of ammonium can lead to denitrification in the water column and loss of N to the into the atmosphere, which strongly discriminates against ^{15}N. This process would result in higher $\delta^{15}N$ of the residual dissolved nitrogen pool (Talbot, 2002). However, a positive correspondence between $\delta^{15}N$ and $\delta^{13}C$ is not supported by canonical component axis II. This may imply that organic matter mineralization processes have little control on $\delta^{15}N$, and that the negative relationship between $\delta^{15}N$ and $\delta^{13}C$ along canonical component axis II is mainly controlled by variability in $\delta^{13}C$ and TOC/TN.

If canonical axis II can be interpreted to reflect the effects of mixing and OM mineralization, then the relatively high significance is surprising in the light of the contrasting limnic settings of the studied sites and also the marked difference in sediment organic geochemical properties (Figure 2). Dove Lake, for example, is up to 60 m deep and oligotrophic (Mariani et al., 2018), billabongs across the MDB are predominantly shallow and eutrophic, and sites on North Stradbroke Island can be relatively shallow, up to 10 m deep (Blue Lake) and oligotrophic (Cadd et al., 2018; Maxson, Tibby, Barr, et al., 2021). This may imply a generalized framework for bulk geochemical evolution during OM mineralization, however, unraveling these processes is complicated by the variable controls both over initial values and subsequent microbial activity. This complexity is reflected in the remaining ~50% of total variance which is not accounted for by canonical axes I and II. Some of the processes not accounted for in this study may be attributed to TOC/TN ratios of topsoil and leaf litter in the range of aquatic OM (<10–20, Holtvoeth et al., 2016; Forbes et al., 2021; Maxson, Tibby, Barr, et al., 2021), as well as different degrees of soil-organic matter decomposition in the catchments across the sites studied herein. Most significantly, TOC/TN < 4 in combination with low TOC, as reported for Booberoi Lagoon (MDB-QLD), Moira Lake (MDB-NSW (south)), and 2 Carp Billabong (MDB-VIC), can be attributed to decomposition controlling C-loss and clay-bound ammonium supply from the catchment promoting increasing N concentrations (Francke et al., 2016; Holtvoeth et al., 2016). Additionally, $\delta^{15}N$ can be controlled by a number of additional processes including nitrification (oxidation of ammonia to nitrate or nitrite) or nitrogen burial during sedimentation (Talbot, 2002). Finally, the unexplained ~50% of total variance may also be attributed to the different timescales covered by the sediment geochemical and catchment data, although the homogenous nature of the sub-surface geochemical data (Section 4.1) suggests that the analyzed sediments provide a good representation of recent subsurface geochemical properties.

5. Conclusion

Sedimentary organic geochemical analyses are commonly used to interpret past variability in both terrestrial and aquatic organic matter accumulation in wetland sediments. Few studies, however, have investigated the validity of those interpretations on a continental scale. For wetland sediments in eastern Australia, multivariate statistical analyses indicate that ~40% variance of sub-surface organic-geochemical properties (TOC, TN, TOC/TN, $\delta^{13}C$, $\delta^{15}N$) in wetland sediments spanning sub-tropical to temperate environments can be explained by catchment vegetation characteristics (vegetation cover in percent, leaf carbon content in g/m^2), and additional indirect controls of the amount of runoff and precipitation, since precipitation promotes vegetation growth in eastern Australia. These data imply that higher catchment vegetation cover, leaf carbon content, and more catchment runoff foster the accumulation of terrestrially (C3) derived organic matter. Additionally, TOC/TN of aquatic matter may be artificially enhanced in densely vegetated catchments promoting N limitation, further contributing to the observed response of elemental ratios to catchment vegetation cover. Water mixing, oxygenation and remineralization appears to play a secondary role over OM storage in the analyzed sediments, corresponding to a second mode of variability which accounts for up to ~10% of the total variance recorded in the data. The results may also imply that additional factors, probably attributed to local conditions such as human land use, local limnologic settings, and a varying age of the investigated sediments account for the remaining ~50% of total variance in the data. Overall, these observations support the use of bulk organic geochemistry as a first order proxy for the amount of terrestrially derived organic carbon accumulation in wetland sediments, in particular when combined with complimentary proxies for catchment vegetation (e.g., pollen, plant macrofossils), organic matter composition (biomarkers) and erosion (mineralogy, inorganic chemistry, uranium isotopes). However, further site-specific and process-based investigations are required to further substantiate such interpretations at local scales, particularly in order to quantitatively infer past fluxes in the carbon cycle.

Conflict of Interest

The authors declare no conflicts of interest relevant to this study.

Data Availability Statement

The geochemical and remote sensing data used for geochemical characterisation of sub-surface sediments and of climatic and catchment vegetation properties in the study are provided in Tables S1–S4 and Supporting Information S1, and at EarthChem (https://doi.org/10.26022/IEDA/112698).

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Erratum

In the originally published article, the plots in Figure 3 were incorrect. A new figure for Figure 3 with the plots rearranged has been inserted, and this may be considered the authoritative version of record.