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RESEARCH ARTICLE

TITLE: ***Biogeochemical responses to Holocene catchment-lake dynamics in the Tasmanian World Heritage Area, Australia***

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KEY POINTS:

- Aquatic dynamics at Dove Lake are modulated by climate- and fire- driven terrestrial vegetation changes
- A period of high rainforest cover prior ca. 6 ka is linked to changing dystrophic conditions, lower light penetration depths and anoxic conditions in the lake bottom waters
- Increasing sclerophyll cover after ca. 6 ka is associated with lower nutrient input, lower dystrophy, more oxic conditions and higher light availability for aquatic organisms

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ABSTRACT

Environmental changes such as climate, land-use, and fire activity affect terrestrial and aquatic ecosystems at multiple scales of space and time. Due to the nature of the interactions between terrestrial and aquatic dynamics, an integrated study using multiple proxies is critical for a better understanding of climate- and fire- driven impacts on environmental change. Here we present a synthesis of biological and geochemical data (pollen, spores, diatoms, µXRF-scanning, CN content and stable isotopes) from Dove Lake, Tasmania, allowing us to disentangle long-term terrestrial-aquatic dynamics through the last 12 kyrs. We found that aquatic dynamics at Dove Lake are tightly linked to vegetation shifts dictated by regional hydro-climatic variability in western Tasmania. A major shift in the diatom composition was detected at ca. 6 ka and it was likely mediated by changes in regional terrestrial vegetation, charcoal and iron accumulation. High rainforest abundance prior ca. 6 ka is linked to increased terrestrially-derived organic matter delivery into the lake, higher dystrophy, anoxic bottom conditions and lower light penetration depths. The shift to a landscape with a higher proportion of sclerophyll species following the intensification of El Niño Southern Oscillation since ca. 6 ka corresponds to a decline in terrestrial organic matter input into Dove Lake, lower dystrophy levels, higher oxygen availability and higher light availability for algae and littoral macrophytes. This record provides new insights on terrestrial-aquatic dynamics that could contribute to the conservation management plans in the Tasmanian World Heritage Area and in temperate high-altitude dystrophic systems elsewhere.

1. INTRODUCTION

Terrestrial and aquatic ecosystem dynamics are influenced by climatic change [e.g. *Ball et al.*, 2010; *Fritz and Anderson*, 2013; *Massaferro et al.*, 2013], land-use/vegetation alteration [e.g. *Cooper et al.*, 2015; *Kissman et al.*, 2017] and fire regime shifts [e.g. *Araneda et al.*, 2013; *Bixby et al.*, 2015; *Brown et al.*, 2014] at multiple scales of space and time. Given the importance and the interconnectedness of both climate and fire [e.g. *Emelko et al.*, 2016; *Fletcher et al.*, 2014; *Power et al.*, 2016], and terrestrial and aquatic ecosystems [*Beck et al.*, 2018a; *Kissman et al.*, 2017; *Strock et al.*, 2017], the paucity of research on how climate and fire drive terrestrial and aquatic ecosystem dynamics constitutes a critical knowledge-gap that potentially undermines effective management and conservation endeavours. This lack of cross-cutting research is particularly salient in Australia, where fires are key ecological agent that has both shaped the unique flora of the region [*Bowman*, 2000] and which threatens relict fire-sensitive plant systems with extinction [*Holz et al.*, 2014]. Little is known about how fire-driven vegetation dynamic influence aquatic ecosystems in this region, exposing a concerning lack of understanding. This knowledge-gap is important in the context of a predicted increase in fire activity over the upcoming centuries [*Moritz et al.*, 2012], and it constitutes a significant impediment to effective management of aquatic systems in fire-prone environments. Here, we integrate datasets of terrestrial and aquatic ecosystem dynamics spanning the past 12,000 years (12 kyrs; kyrs = duration in thousands of years) within the Tasmanian World Heritage Area, Australia, in an attempt to understand how these systems respond to inferred shifts in climate and fire activity through this time.

Fires can drive substantial changes in terrestrial vegetation and landscapes processes, such as soil alteration and erosion, which can mobilise nutrients from soils, consequently increasing nutrient loading into streams, rivers, lakes and reservoirs [*Lane et al.*, 2008; *Moody and Martin*, 2001; 2009; *Reneau et al.*, 2007; *Sheridan et al.*, 2007; *Sherson et al.*, 2015; *Wilkinson et al.*, 2009]. Fire can also affect soil chemistry by altering organic matter content, cation exchange capacity, pH, electrical conductivity and nutrient concentration [e.g. *Bixby et al.*, 2015; *Cerrato et al.*, 2016; *Fletcher et al.*, 2014; *González-Pérez et al.*, 2004; *Knoepp et al.*, 2009; *Kutiel and Inbar*, 1993; *Raison*, 1979; *Robinne et al.*, 2018] . These fire-driven processes can influence the behaviour of micronutrients, such as redox-sensitive iron (Fe) and manganese (Mn) [*Calvert and Pedersen*, 2007; *Engstrom and Wright*; 1984; *Davison*, 1993; *Mackereth*, 1966; *Och et al.*, 2012; *Wersin et al.*, 1991], potentially leading to anoxia of bottom waters through reductive dissolution and (re)oxidation, which progressively leads to geochemical focusing. This process transfers and enriches these elements in deeper waters and can drive the development of anoxic bottom waters [*Naeher et al.*, 2013; *Schaller and Wehrli*, 1996]. Anoxia within aquatic systems is associated with biodiversity loss, nutrient availability and food web alterations [*Diaz*, 2001; *Hughes et al.*, 2015; *North et al.*, 2014]. While the influence of redox conditions over lake water oxygen dynamics is well understood [e.g. *Balistrieri et al.*, 1992; *Davison*, 1993; *Sigg*, 2000], little is known about how long-term changes in terrestrial processes influences within lake dynamics, especially in temperate Australia.

Long-term disturbance from fire in fire-sensitive vegetation, such as rainforest, can result in substantial ecosystem transformation, such as altered species composition, soil formation and nutrient loss that can have important implications for associated water bodies [*Ball et al.*, 2010; *Huvane and Whitehead*, 1996; *Korhola et al.*, 1996; *Leys et al.*, 2016; *Morris et al.*, 2015; *Smith et al.*, 2011]. Fires in temperate environments, such as western Tasmania, mostly occur in the period from September to March (from spring to early autumn) and are often followed by heavy rain events [*Bridle et al.*, 2003; *Pemberton*, 1988], which can remove the soil layer into water bodies, altering water geochemistry and nutrient availability [*Beck et al.*, 2018a; *Boerner*, 1982] and precipitating an aquatic ecosystem response [*Beck et al.*, 2018b]. Disturbance from repeated fire in the rainforest of Tasmania, for example, are associated with the destruction and complete erosion of highly organic soil profiles, localised plant species extinctions and invasion by fire-promoting vegetation that can radically alter fire-vegetation-soil dynamics. Investigation in to the effects of these changes in the dystrophic aquatic environments of Tasmania has revealed a tight coupling between fire-driven changes in the terrestrial environment and aquatic ecosystem dynamics, via the influence of organic soils over aquatic trophic status [*Beck et al.*, 2018a; *Fletcher et al.*, 2014] and the influence of terrigenous input over lake water pH [*Beck et al.*, 2018b]. While the recent work by *Beck et al*. (2018b) reveals a tight coupling between terrestrial and aquatic ecosystem changes in response to fire, there is little empirical evidence for the influence of long-term changes in fire and vegetation over important within-lake processes, such as lake mixing, light availability changes and redox conditions.

Here we present a new lake sediment geochemical (µXFR-scanning, CN content and stable isotopes) and biological (diatoms) proxy dataset from a relatively large and deep dystrophic lake (Dove Lake) located in cool temperate west of Tasmania, Australia. We compare our new proxy dataset from Dove Lake to a recent pollen-based quantitative vegetation reconstruction from the same sediment sequence that reveals marked climate and fire driven changes in vegetation land-cover over the last 12 kyrs [*Mariani et al.,* 2017]. These changes are characterised by shifts in the importance of high biomass rainforest vegetation and lower biomass sclerophyll-dominated vegetation. Peat soils predominate in this cool temperate and oligotrophic environment, with rainforest typically developing atop deep peat profiles, while sclerophyll vegetation typically produces shallow to skeletal peat profiles with a lower nutrient content [*Beadle*, 1966; *Jackson*, 2000]. Like other temperate blanket peat environments [*Bergström et al.*, 2001; *Hansen*, 1962; *Steinberg et al.*, 2003], water bodies in western Tasmania receive substantial amounts of terrestrially derived organic matter and are, consequently acidic, oligotrophic and dystrophic [*Buckney and Tyler*, 1973; *Tyler*, 1992], with an elevated content of reactive Fe and Mn [*Mackey et al.*, 1996]. Given the high nutrient limitation in this region, the entire nutrient pool supporting terrestrial vegetation development only derives from the accumulation of organic debris. This implies that the nutrient input into water bodies is almost entirely derived from these acid peat soils, producing low-nutrient, tea-coloured waters (i.e. oligotrophic and dystrophic/humic lakes) [*Tyler*, 1974; 1992]. We hypothesise that (i) phases of maximum rainforest cover around Dove Lake will be associated with deeper catchment peat profiles that deliver elevated amounts of terrestrial organic matter, along with carbon, nitrogen, Fe and Mn into the lake; (ii) the increase in sclerophyll vegetation cover will be associated a shift toward shallower peat profiles and a concomitant decrease in nutrient supply into the lake; and (iii) changes in terrestrially-derived organic matter will drive changes in within-lake redox conditions, light and oxygen availability and diatom compositional changes.

1.1 Western Tasmania

Tasmania is a large cool temperate island located between 41-44°S (Figure 1) characterised by a temperate maritime climate with mild winters and cool summers [*Gentilli*, 1972]. A rain-shadow is produced by the prevailing westerly winds that rise over the northwest to southeast trending mountain range bisecting Tasmania. The regional geology of western Tasmania is dominated by highly resistant and inert quartz-dominated rock types which contribute little mineral matter to soils, resulting in the predominance of poor acidic soils (organosols/peats) [*di Folco*, 2007; *Tyler*, 1992]. Indeed, the ionic composition of Tasmanian lakes water reflects a minimal geological contribution [*Buckney and Tyler*, 1973]. The cool temperate climate, extreme oligotrophy and predominance of organosols result in ubiquitously moderately to highly dystrophic and acidic waterbodies that a thermal behaviour from warm monomictic to polymictic, with some evidence of meromixis (or partial meromixis) [*Bowling and Tyler*, 1984; *Buckney and Tyler*, 1973; *King and Tyler*, 1981; *Tyler*, 1974].

The modern vegetation landscape of western Tasmania is characterised by a mosaic of rainforest, moorland and sclerophyll vegetation [*Kirkpatrick and Dickinson*, 1984] that has resulted from the long-term application of fire by people through the Late Pleistocene and Holocene [*Fletcher and Thomas*, 2010; *Jackson*, 1968; *Mount*, 1979; *Thomas*, 1995]. The type of extant vegetation is intimately linked to peat development and nutrient availability for aquatic systems [Bowman and Jackson, 1981; Bridle et al., 2003; Beck et al., 2018a]. For instance, soils formed under sclerophyll vegetation in Tasmania are thinner and have significantly lower nutrient content compared to soils developed under rainforest vegetation [Bowman and Jackson, 1981; Jackson, 1968; Wood et al., 2011] and fire-driven shifts from rainforest to more sclerophyll dominant of vegetation in Tasmania is associated with a shift to a more oligotrophic aquatic environment [*Beck et al.*, 2018a; *Fletcher et al.*, 2014].

1.2. Study site

Dove Lake (41°39'34.27"S, 145°57'35.14"E) is a large (90 ha) dystrophic subalpine lake (940 m a.s.l.) located within the Tasmanian World Heritage Area in Australia (Figure 1). Average total annual rainfall at the nearest meteorological station (Cradle Valley; 2.7 km from Dove Lake; 41°38'24.23"S, 145°56'24.28"E; 903 m a.s.l.) is c. 2700 mm/yr (Figure 1a). The lake has a relatively steep bathymetric profile characterised by two deep basins (Figure 1b; Supporting Information Figure S1). Maximum depth was recorded c. 60 m in the south basin, whereas, a maximum depth of c. 45 m was recorded in the northern basin (core location). The geology of the watershed is represented by two dominant types: quartzite and Quaternary glacial deposits (33.5% and 45.9%, respectively) (Figure 1c). The modern catchment vegetation is dominated by wet sclerophyll forest on the eastern and western flanks, treeless fire-promoted moorland on the northern edge and rainforest in the southwest corner (Figure 1d). Above c. 1000 m a.s.l. the landscape is dominated by highland and treeless vegetation, which represents 45.1% of the Dove Lake catchment. Important species in the modern landscape are *Eucalyptus coccifera*, *Lophozonia cunninghamii* (syn. *Nothofagus cunninghamii)*, *Gymnoschenous sphaerocephalus, Athrotaxis selaginoides* and a variety of ericaceous shrubs. Within the lake catchment and across the surrounding plateau, typical montane rainforest trees are also found, such as *Fuscospora gunnii* (syn. *Nothofagus gunnii*), *Athrotaxis selaginoides* and *A. cupressoides* (Cupressaceae).

A quantitative vegetation reconstruction was previously carried out at this location [*Mariani et al.*, 2017a], which indicated the dominance of fire-promoted treeless moorland over the last 12 kyrs. Forest cover was at c. 40% between 10 and 4 ka (ka= specific point in time in thousands of years), peaking at c. 50% of the surrounding landscape between 8-7 ka in response to a shift to a wetter hydro-climate and a reduction in regional fire activity [*Fletcher and Moreno*, 2011; 2012; *Mariani and Fletcher*, 2017; *Wilkins et al.*, 2013; *Xia et al.*, 2001]. A shift to a drier and more variable hydro-climate after ca. 5.5 ka [*Fletcher and Moreno*, 2012; *Mariani and Fletcher*, 2017] drove an increase in regional biomass burning and a replacement of areas of rainforest by fire-promoting sclerophyll plant taxa, such as the fire-adapted *Eucalyptus [Beck et al., 2017; Beck et al., 2018a; Fletcher et al., 2014; Mariani et al., 2017b; Stahle et al., 2017; Stahle et al., 2016]*. Subsequently, during the late Holocene, a decrease in moisture availability and a synchronous phase of increased fire activity across western Tasmania [*Fletcher et al.*, 2015; *Mariani and Fletche*r, 2017] resulted in a rainforest decline around Dove Lake [*Mariani et al.*, 2017a]. These hydro-climatic changes are likely the long-term (millennial scale) effect of increasingly frequent El Niño events in the tropical Pacific [*Donders et al.*, 2008; *McGlone et al.*, 1992; *Moy et al.*, 2002], which would have resulted in overall cooler than normal waters over the western tropical Pacific and high pressure systems driving negative moisture anomalies in eastern Australia. Today, El Niño events are associated with drier conditions and increased fire activity in south-east Australia today [*Mariani et al.*, 2016; *Nicholls and Lucas*, 2007].

2. METHODS

The 122 cm-long organic sediment core from Dove Lake was extracted in December 2015 using a 6.8 cm- diameter polycarbonate chamber attached to a Universal Gravity Corer (http://www.aquaticresearch.com/universal_core_head.htm). The core chronology was obtained using ^{210}Pb and ^{14}C dating methods (Supporting Information Table S1a,b) and the age-depth model was previously published in *Mariani et al.* (2017) (Supporting Information Figure S2).

2.1 Charcoal and pollen analysis

Macroscopic (>125µm) and microscopic (<125µm) charcoal content was analysed to document the local fire history and the results were previously presented in *Mariani et al*. [2017]. Macroscopic charcoal content was measured using 1.25 cm^3 taken from 5-mm thick (median time resolution= ca. 48 years) contiguous samples and microscopic charcoal content was extracted from 0.5 cm^3 samples taken at 1 cm-resolution. The macroscopic and microscopic charcoal counts were converted into accumulation rates (CHAR, particles cm^2 yr^{-1}) and for the remainder of this manuscript they will be referred as macroscopic-CHAR and microscopic-CHAR, respectively.

On the same sequence, pollen analysis was performed at 1 cm-resolution to investigate the vegetation history and the results were previously shown in *Mariani et al*. (2017). The ratio between rainforest and sclerophyll plant taxa (Rainforest:Sclerophyll) was calculated from the land-cover data published in *Mariani et al*. (2017) prior to the elaboration of statistics (see below). From the same dataset, *Isoëtes,* a littoral macrophyte, spore abundances were considered as proxy for aquatic conditions, such as lake-level and light penetration variation [*Bogotá-A et al.*, 2016; *Chappuis et al.*, 2015; *Finkenbinder et al.*, 2014; *Pesce and Moreno*, 2014; *Simi et al.*, 2017].

2.2 µXRF core scanning

The micro X-Ray Fluorescence (µXRF) core scanning undertaken at the Australian Nuclear Science and Technology Organisation (ANSTO) provided 0.5 mm-interval, non-destructive elemental analyses of the Dove Lake core. Manganese:Iron (Mn:Fe) and Iron:Titanium (Fe:Ti) ratios were calculated to document changes in redox conditions at the water-sediment interface [*Corella et al.*, 2012; *Cuven et al.*, 2010; *Haberzettl et al.*, 2007].

2.3 Carbon and Nitrogen

Analysis for percent Carbon (%C), percent Nitrogen (%N), $\delta^{13}C$ and $\delta^{15}N$ for Dove Lake were performed at a resolution of 1 cm (median time resolution= c. 96 years) at the ANSTO facility for stable isotope analyses using standard procedures [*Ohlsson and Wallmark*, 1999]. Briefly, the crushed and dried samples were weighed into tin capsules and introduced sequentially into an elemental analyser (Thermo Fisher Flash 2000 HT EA) using an autosampler. No pre-treatment was performed as no carbonates were detected in preliminary tests executed on the pollen samples from *Mariani et al*. [2017].

2.4 Diatoms

Diatom analysis was performed at an irregular resolution and volumes throughout the Dove Lake core due to sediment availability. Samples were processed using standard methods [*Battarbee*, 1986]. Samples were placed in a hot bath and a series of hydrogen peroxide additions were used to remove all organic matter. Sample dilutions were mounted with Naphrax® and at least 300 diatom valves were identified to genus level using an oil immersion DIC light microscope at 1000X magnification. Diatoms were identified to genera rather than species level in order to observe the changes in diatom habitat to shifts in climate and terrestrial environmental dynamics, rather than a detailed ecological analysis of the lake.

2.5 Statistical analyses

µXRF core scanning and diatom data from Dove Lake were summarised using Principal Curves (PCs) in R 3.3.1 [*R Core Development Team*, 2013]. PCs [*Hastie and Stuetzle*, 1989] are a generalised form of the first principal component axis as a smooth, one-dimensional curve fitted through the data in multiple dimensions such that the curve best fits the data. Thus, distances of the samples to the PC are somewhat minimised [*Simpson and Birks*, 2012]. PCs are considered to perform substantially better than Principal Component Analysis (PCA) and Correspondence Analysis (CA) in many cases, especially when data are dominated by a single dominant gradient [*Simpson and Birks*, 2012]. Prior the PC analysis, µXRF data were normalised by dividing all elemental counts by the total counts per second (cps) [*Croudace and Rothwell*, 2015] and binned to 50-years intervals to facilitate the statistical performance. All elements, but not ratios, were included in the PC analysis. Diatom data were square-root transformed to correct for taxon skewness.

Generalized Additive Models (GAMs) were used to identify significant trends in the Dove Lake vegetation, charcoal influx, µXRF core scanning, diatoms and carbon and nitrogen data. The additive models do not involve a-priori parameter setting, instead allowing the shape of the relationship to be determined from the data using penalised regression [*Hastie and Tibshirani*, 2006]. All GAMs were fitted using the Mixed GAM Computation Vehicle (mgcv) package [*Wood and Wood*, 2007] for R 3.3.1 [*R Core Development Team*, 2013]. To highlight periods of significant shifts in all time-series analysed with GAMs, the first derivative of the GAM splines were calculated (https://github.com/gavinsimpson/tsgam). This approach allows measurement of the slope at each point throughout a time-series, thus enabling us to extract trends in the palaeodata [*Bunting et al.*, 2016]. When slope exceeds confidence intervals, the relative time period is considered as 'periods of significant change' of a certain time-series. Significance level for the derivative calculations was set to 0.95.

3. RESULTS

3.1 Charcoal and pollen analysis

The macroscopic- and microscopic- CHAR record reveal substantial changes in fire regimes throughout the record (Figure 2). In both records, relatively high accumulation rates are observed prior to ca. 6 ka with a peak occurring at 7.3 ka. A gradual decline is visible since ca. 6 ka and interrupted by an important peak at 3.7 ka, then the data reach minimum values between 2.5-0.5 ka. A total of 109 samples from Dove Lake were analysed for pollen and spores content and results were presented in *Mariani et al.* (2017). The ratio Rainforest:Sclerophyll vegetation calculated from the data of *Mariani et al.* (2017) shows high values between 10-6 ka, followed by a pronounced decline occurring between 6 and 4 ka. The littoral macrophyte *Isoëtes* displays relatively low and stable abundances between 11.7-6.5 ka, then increases substantially (up to three times the initial values) during the period between 6.5 and 3 ka. A decline is evident since ca. 3 ka, but the percent abundance is maintained higher than the Early to mid-Holocene values.

3.2 µXRF core scanning

A total of 4578 data points was recorded on the Dove Lake core and results are presented in Figure 2 (red curves). Selected elemental data obtained by this analysis revealed high counts of iron (Fe), the most abundant element in the spectrum. A gradual decline in Fe is detected between 11.7-5.2 ka with a sharper decline between ca. 7-5.2 ka, when Fe counts reach minimum values until >0.1 ka (Figure 2). Titanium (Ti) displays very low counts and the opposite trend of Fe, whereas manganese (Mn) counts are persistently low until ca. 3.5 ka, then gradually increase (Figure 2). The Mn:Fe ratio is an indicator for reducing conditions at the water-sediment interface [*Corella et al.*, 2012; *Cuven et* *al.*, 2010; *Haberzettl et al.*, 2007] and shows fairly consistent high values prior to ca. 6 ka with a gentle decline evident between ca. 7.5-6 ka. The Fe:Ti ratio has been employed with the same scope as the Mn:Fe [*Aufgebauer et al.*, 2012] as they show virtually identical trends, although specular (Figure 2).

3.3 Carbon and Nitrogen

A total of 92 samples were analysed for %C, %N, $\delta^{13}C$, and $\delta^{15}N$ (Figure 2). The percentages of C, N and the C:N ratio show a slow gradual increase from ca. 11.7 to 8 ka, then become more stable until around 6 ka. At this point, a substantial, although gradual, decline occurs in these proxies, alongside a marked variation in the $\delta^{13}C$ and $\delta^{15}N$ values. The amount of C declines from 19% to 12%, whereas %N drops from 0.9% to 0.6%. Analogously, the ratio C:N follows a gradual decline from values clustered around 22 to around 18. The C:N has a mean value of 19.46 suggesting a high terrestrial component of this metric [*Meyers and Teranes*, 2002]. $\delta^{13}C$ is gradually increases between 6.2 to 3.6 ka from values around −26.7‰ to values around −26.3‰. Given the extremely low variation in $\delta^{13}C$, this proxy was not object of discussion. Meanwhile, $\delta^{15}N$ gradually increases from 6.1 to 3.5 ka with values ranging from 2‰ to 3.7‰. Minerogenic flux was roughly estimated by mass difference using the summed C and N content, then converted into accumulation rates (cm^{-2} yr^{-1}) (Supporting Information Figure S4).

3.4 Diatoms

A total of 37 diatoms samples were counted and selected genera are presented in Figure 3. The most abundant taxon in the spectrum is *Frustulia* spp., displaying higher abundances before ca. 9.8 ka and after ca. 5 ka (c. 40%). Although less abundant in the record (<5%), *Aulacoseira* spp. and

Gomphonema spp. show important trends analogous to *Frustulia* spp. *Eunotia* spp., *Brachysira* spp. and *Actinella* spp. display relatively low abundances between ca. 11.7-9.8 ka and post ca. 5 ka, whereas higher percentages are recorded between ca. 9.8-5 ka. *Eunotia* spp. is the second most abundant taxon after *Frustulia* sp., reaching maximum values around 30% at ca. 6 ka. *Tabellaria* spp. shows relatively high percentages (8-10%) prior to ca. 5 ka, dropping to values around 5% afterwards. An extended diatom stratigraphic plot is displayed in Supporting Information Figure S3, alongside the CONISS (stratigraphically constrained cluster analysis) zonation description (Document S1).

3.5 Statistical analyses

Results from the PC analyses on the µXRF scanning and diatoms data are presented in Figures 2 and 3 (respectively). The µXRF data PC explains approximately 46% of the total variance in the dataset. Iron (Fe) shows the highest positive correlation with the PC (r = 0.93), whereas, Ti displays a significant negative correlation with this curve (r = −0.83), alongside other detrital elements, such as potassium (K) and zirconium (Zr) (Supporting Information Table S2). Fe:Ti and Mn:Fe show very high positive correlation values with the µXRF data PC (0.85 and −0.49 respectively).

The diatoms PC explains c. 31% of the variance of the assemblage dataset. *Gomphonema* spp., *Aulacoseira* spp. and *Frustulia* spp. show the three highest negative correlations with the PC (−0.79, −0.75 and −0.65 respectively). On the positive end of the correlation values spectrum, *Eunotia* spp., *Brachysira* spp. and *Actinella* spp. show the three highest values (0.77, 0.69 and 0.62 respectively). *Tabellaria* spp. displays a significant positive correlation of 0.3 with the diatoms PC. A table with the Pearson correlation values for all taxa is presented in Supporting Information Table S3.

The GAM splines produced for selected proxies from Dove Lake highlight the significant trends in the data by efficiently removing the noise embedded in the time-series and, thus, allowing a better comparison of the palaeorecords (Figure 4). Similar trends in the GAM splines are especially observed in the diatoms PC, C:N and Rainforest:Sclerophyll ratios. The analysis of the first derivatives allowed us to identify significant shifts in the time-series and a better assessment of synchronicity between proxies (Figure 4). A significant increase in the Rainforest:Sclerophyll ratio between 11.7 and 9 ka was found at the same time as significant increase in the diatoms PC and the C:N ratio. A decrease in the Rainforest:Sclerophyll ratio, diatoms PC and C:N ratio between 6.5-3 ka is concurrent with a increase in the littoral macrophyte *Isoëtes*. The µXRF data PC follows a persistent declining trend from 11.7 to ca. 5 ka, reaching significant levels between 11.7-10 ka and 7-5 ka. Although there is a higher amount of noise in the data (i.e. peaks), macroscopic-CHAR appears to follow a trend analogous to the µXRF data PC, but reaches significance only between 6.5-1.7 ka.

4. DISCUSSION

4.1 Vegetation change and macronutrient dynamics at Dove Lake

Our data demonstrates that changes vegetation and fire dynamics around Dove Lake in response to long-term changes in hydroclimate and their influence over fire activity [*Mariani et al.*, 2017b; *Stahle et al.*, 2017; *Stahle et al.*, 2016] have played an important role in driving changes in within-lake dynamics at this site. The climate and fire-driven increase in sclerophyll-dominant vegetation cover (at the expense of rainforest) around Dove Lake at ca. 6 ka (Figure 5) is associated with a substantial decline in carbon and nitrogen content of lake sediment organic matter (Figure 4). This trend is mirrored elsewhere in Tasmania [*Fletcher et al.*, 2014; *Beck et al.*, 2018a] and is consistent with a reduction in peat depth in response to a reduction of rainforest cover around Dove Lake (Figure 5c) and lower nutrient status of peat soils that develop under sclerophyll vegetation, relative to rainforest [*Bowman and Jackson*, 1981; *Bridle et al.*, 2003].

A shift from high relative terrestrial nutrient input into the lake in response to the shift from high relative rainforest cover to high relative sclerophyll cover is supported by (1) the shift from high to low relative C:N ratios at ca. 6 ka [*Meyers and Teranes*, 2002] and (2) the concomitant shift from low to high relative $\delta^{15}N$ values (Figure 2). Aquatic organisms preferentially uptake the lighter ^{14}N isotope and N enriched lake water results in the production of organic matter low in ^{15}N [so called Rayleigh Distillation; *Talbot & Johannessen*, 1992; *Talbot,* 2002]. The high C:N values prior to ca. 6 ka indicate an increase in the delivery of terrestrial organic matter into Dove Lake from the well-developed peat soils within the catchment under high relative rainforest cover. This evidence is supported by an estimate of minerogenic flux (Supporting Information Figure S4), which shows relatively high fluxes prior to ca. 6 ka and a decline between 6 and 4 ka. The high relative input of terrestrial organic matter would have provided a constant input of ^{14}N and organic matter under an enriched ^{14}N regime, with the shift to increased sclerophyll vegetation and the resultant decrease in, and impoverishment of, terrestrial organic matter forcing the uptake of increased amounts of ^{15}N.

4.2 Changes in acidity, dystrophy, redox conditions and light availability at Dove Lake

Our data reveals a covariance between millennial-scale trends in diatom community composition within Dove Lake (diatoms PC; Figure 3) and trends in both the ratio of Rainforest:Sclerophyll vegetation and C:N throughout the last 12 kyrs (Figure 4 and 5). This covariance indicates a tight coupling between millennial-scale terrestrial and aquatic ecosystem dynamics through this time. A positive correlation between C:N and dystrophy has been identified in cool temperate systems elsewhere [*Hansen*, 1962] and we interpret the coupling between C:N ratio and the diatom PC as an indication of high relative levels of dystrophy and acidity prior to ca. 6 ka when increased rainforest cover and associated peat profiles delivered high amounts of humic acids and leached terrestrially-derived nutrients into the lake (Figure 4). Further, we contend that the high correlations found between the diatom PC with *Eunotia* spp., *Brachysira* spp. and *Actinella* spp. (positive) and with *Gomphonema* spp., *Aulacoseira* spp. and *Frustulia* spp. (negative) (Figure 3) tracks dystrophy and acidity levels. While most diatom taxa in western Tasmania are adapted to high acidity and dystrophic/oligotrophic waters [*Vyverman et al.*, 1996], the diatom compositional shifts represented by the diatom PC as reflect shifts between more acidic /more dystrophic to less acidic/less dystrophic conditions (Figure 5). Indeed, *Eunotia* spp., *Brachysira* spp. and *Actinella* spp., which are important components of the diatom community prior to ca. 6 ka, are associated with generally high dystrophy and lower pH in temperate southeast Australia and Tasmania [*Hodgson et al.*, 1996; *Philibert et al.*, 2006; *Sonneman et al.*, 1999; *Vyverman et al.*, 1995; *Vyverman et al.*, 1996], while taxa that increase in important after ca. 6 ka, such as *Gomphonema*, *Aulacoseira* and *Frustulia* species, are associated with slightly higher pH [*Philibert et al.*, 2006; *Vyverman et al.*, 1995; *Vyverman et al.*, 1996].

Our inference of changes in dystrophy is consistent with changes in the diatom type *Tabellaria* spp., which shifts gradually from high to low relative values at ca. 6 ka (Figure 3). *Tabellaria flocculosa* is the only member of the *Tabellaria* genus currently found in Dove Lake (<1% of modern composition:

Vyverman et al., 1995). *T. flocculosa* displays a marked preference for low-light conditions during the photosynthetic process [*Paul and Duthie*, 1989], indicating that this species is sensitive to colour changes in lake waters that influence light penetration, such as results from changes in levels of dystrophy. Indeed, *T. flocculosa* and *T. quadriseptata* are abundant in dystrophic environments [*van Dam et al.*, 1981], indicating a competitive advantage in low light conditions by members of the *Tabellaria* genus. Thus, we interpret the relatively higher abundance of *Tabellaria* spp. prior to ca. 6 ka as reflecting increased inputs of terrestrially derived organic material in to the lake (Figure 4). Moreover, our inferred changes in dystrophy resulting from hydroclimatic and fire-driven changes in terrestrial vegetation and organic soil development are consistent with trends in the littoral macrophyte, *Isoëtes*, which shifts from low to high relative values at ca. 6 ka (Figure 4b). While comparatively little is known about the ecology of Tasmanian *Isoëtes* species [*Garrett and Kantivalis*, 1992], evidence indicates an increase in mortality in response to a reduction in the penetration of light through the water column in the widespread European and North American species, *Isoëtes lacustris* [*Chappuis et al.*, 2015; *Riera et al.*, 2017]. Thus, in combination with the evidence for changes in dystrophy in Dove Lake, we interpret the low relative values of *Isoëtes* prior to ca. 6 ka as reflecting a reduction in suitable habitat due to poor light penetration through the highly dystrophic lake waters, with an increase in this aquatic macrophyte after ca. 6 ka reflecting increased light penetration through less dystrophic waters (Figure 5).

We observe a high correlation between the µXRF data PC with Fe:Ti and Mn:Fe ratios that we interpret as indicating changes in redox conditions at the water-sediment interface. Ratios of Mn:Fe have been used repeatedly to reconstruct changing redox conditions in lakes [e.g. *Dean and Doner*, 2012; *Koinig et al.*, 2003; *Loizeau et al.*, 2001; *Melles et al.*, 2012; *Naeher et al.*, 2013; *Wersin et al.*, 1991], with lower Mn:Fe ratios associated with lower O_2 concentrations in the water column [e.g.

Balistrieri et al., 1992; *Davison*, 1993; *Loizeau et al.*, 2001; *Mackereth*, 1966; *Wersin et al.*, 1991; *Nealson and Saffarini*, 1994; *O'Sullivan and Reynolds*, 2008]. Thus, the substantially lower Mn:Fe ratios prior ca. 6.5 ka indicate lower oxygen abundance at the water-sediment interface (i.e. anoxic bottom waters) during the phase of high relative terrestrial organic matter input into the lake – more organic matter was available for remineralisation which consumed more O_2 resulting in anoxia. The shift to lower terrestrial organic matter input after ca. 6 ka, thus, resulting in a reduction in O_2 consumption and the development of more oxic bottom waters (Figure 5).

4.3 Climate- or biomass- induced charcoal accumulation?

We observe high relative macroscopic charcoal influx into Dove Lake during the phase of high relative forest cover prior to ca. 6 ka, with an overall shift to lower charcoal influx between 6.2-1.7 ka in concert with a shift to increased sclerophyll vegetation and overall landscape openness (Figures 4 and 5) [*Mariani et al.*, 2017]. While inconsistent with the fire-ecology of rainforest (pyrophobic), and sclerophyll and open moorland (pyrophytic) vegetation in this region, our results are consistent with a primary control of biomass control over the production of macroscopic charcoal at Dove Lake. *Fletcher et al*. [2014] report similar reduction in sedimentary macroscopic charcoal in response to fire-driven shifts between pyrophobic and pyrophytic vegetation during the Holocene in Tasmania, concluding that the higher woody biomass and destructive nature of rainforest fires can produce high amounts of macroscopic charcoal relative to fire-promoting vegetation, where less woody material is usually burnt [*Fletcher et al.*, 2014]. Indeed, a reduction in macroscopic-CHAR was also observed in New Zealand following pyrophobic forest removal by , where a decline in biomass availability following burning and deforestation after the Maori [*McWethy et al.*, 2010].. Further afield, our data showing high charcoal accumulation in line with high forest cover is analogous to studies from Kenya [*Colombaroli et al.*, 2014], Spain [*Gil-Romera et al.*, 2014] and Arizona [*Brunelle et al.*, 2010] where biomass-limitation was indicated as the most plausible explanation for the link between high forest cover and high charcoal influx. Nevertheless, the hypothesis of biomass-limited fire occurrence is unlikely in the generally high biomass environments of western Tasmania [*McWethy et al.*, 2013], thus it is more likely that a variation in the fuel type, from rainforest to sclerophyll vegetation, would have modulated charcoal influx in the Dove Lake record.

5. CONCLUSION

A tight coupling between the terrestrial-aquatic systems was found at Dove Lake, reflecting terrestrial organic material inputs into the lake from the vegetation-soil system over the last 12 kyrs. The ratio between rainforest and sclerophyll vegetation cover shows synchronicity to shifts in carbon and nitrogen content and isotopic composition, alongside shifts in the diatoms assemblage. Phases of high rainforest cover with deep peat profiles are linked to high carbon, iron and nitrogen delivery prior ca. 6 ka at Dove Lake. Contrastingly, an increase in sclerophyll abundance in the landscape after the intensification of ENSO since ca. 6 ka corresponds to a decline in nutrient supply to the lake. Moreover, we found that the changes in terrestrially-derived organic matter inputs drove relatively synchronous changes in dystrophy and acidity levels, water redox conditions, light availability and algal ecosystem dynamics.

Light was found to be an important driver of changes in algal composition and aquatic macrophyte abundance. Trends in *Tabellaria* spp. and *Isoëtes* spp. suggest a variation in light penetration depth resulting from dystrophy level changes after ca. 6 ka. Furthermore, the µXRF core scanning results allowed us to reconstruct changes in redox conditions at the water-sediment interface: low Mn:Fe ratios prior ca. 6 ka probably indicate an oxygen depletion of the water column and anoxic lake bottom attributable to altered lake mixing and metabolism of nutrients. This record provides new insights on the importance of terrestrial inputs in driving light availability changes and aquatic ecosystems dynamics at Dove Lake in the World Heritage Area of Tasmania.

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Data availability

Data produced within the present work will be made available upon publication on NEOTOMA

(https://www.neotomadb.org/).

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FIGURE CAPTIONS

Figure 1 Introductory figure showing a) site location (upper panel) and climatic diagram (lower panel); b) 3D projection of Dove Lake watershed and bathymetric profile. Lower panel in b) represents the depth profile from point A to point B (direction as indicated in the map). Red star in b) indicates the location of the coring site. A detailed bathymetric map of Dove Lake is presented in Supporting Information Figure S1; c) 3D projection of the Dove Lake watershed overlayed with the geological map d) 3D projection of the Dove Lake overlayed with the vegetation map (data from https://www.thelist.tas.gov.au/app/content/home).

Figure 2 Charcoal and geochemical results for the Dove Lake core. Black step-like curves are the results from microscopic and macroscopic charcoal analyses. Black scatter lines represent show the results of carbon and nitrogen analyses; red curves represent selected µXRF elements and ratios (50-years binned). µXRF elements counts were normalised by total counts per second (tcps). µXRF Principal Curve (PC) includes all elements listed in Supplementary Table S2.

Figure 3 Results of the diatom analysis for selected taxa and diatoms Principal Curve (PC). Red bars identify taxa with positive Pearson correlation values with the PC. Blue bars identify taxa with negative Pearson correlation values with the PC. Dashed lines represent the significant zonations (more detail in Supporting Information Figure S3 and Document S1). The asterisk (*) indicates that *Tabellaria* does not show a strong correlation with the PC (r=0.3), but given its abundance in the record, its trend was considered important for the discussion.

Figure 4 Results from the application of Generalised Additive Models (GAMs) and first derivative calculations on selected time-series. Curve segments highlighted in red identify periods of significative increasing values , whereas blue highlights period of significant decrease in the time-series. Significance levels for derivative calculations were set to 0.95.

Figure 5 Summary figure showing a schematic representation of the environmental changes recorded at Dove Lake based on the time-series presented on the right: a) macroscopic-CHAR; b) Carbon to Nitrogen ratio (C:N); c) Rainforest to Sclerophyll ratio (R:S); d) Diatoms Principal Curve and e) µXRF scanning data Principal Curve. Drawings are not in scale.

FIGURES

Figure 1

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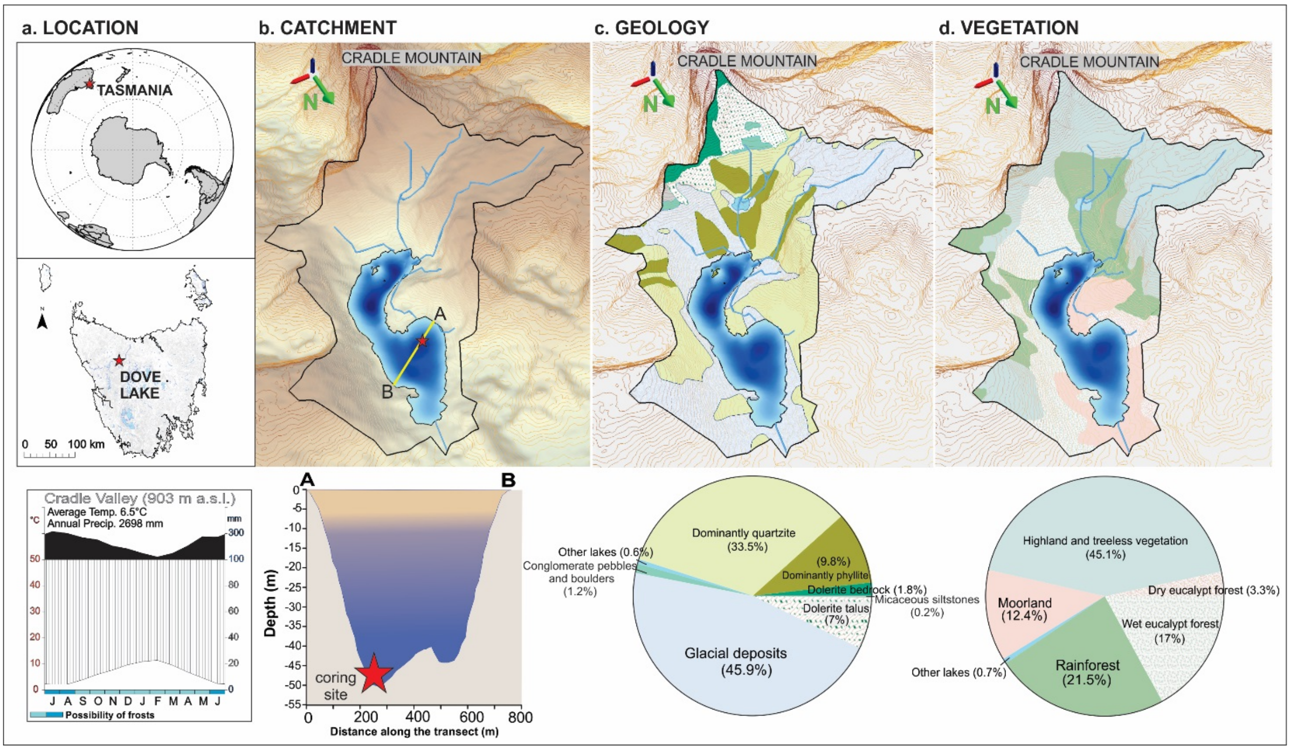

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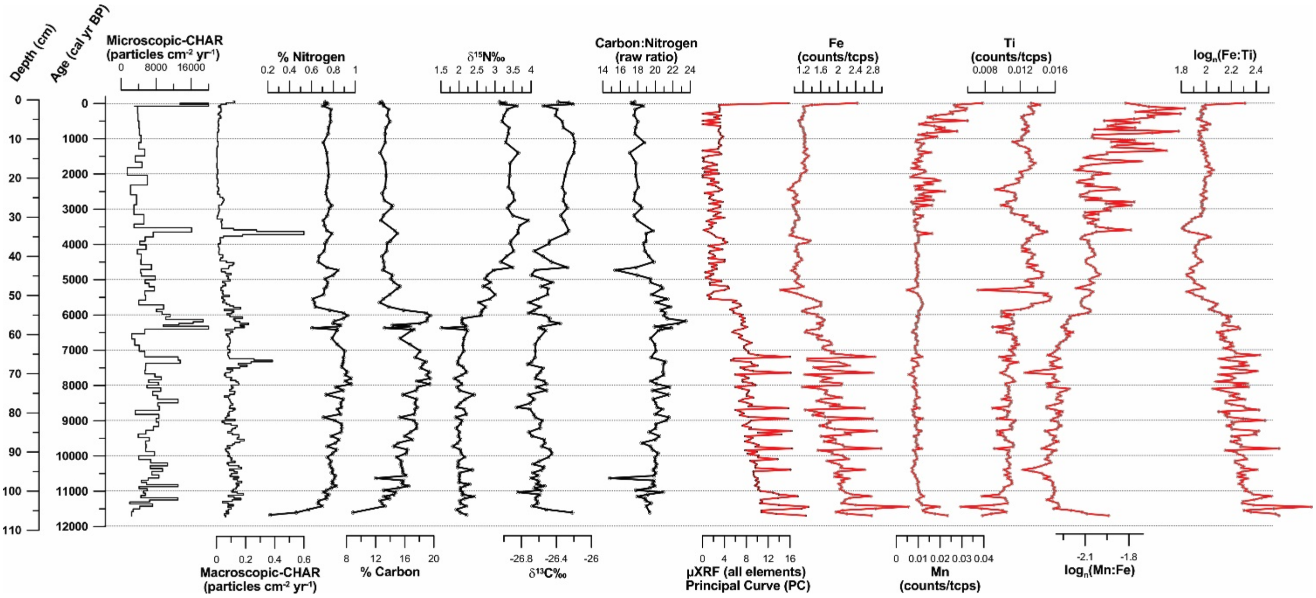

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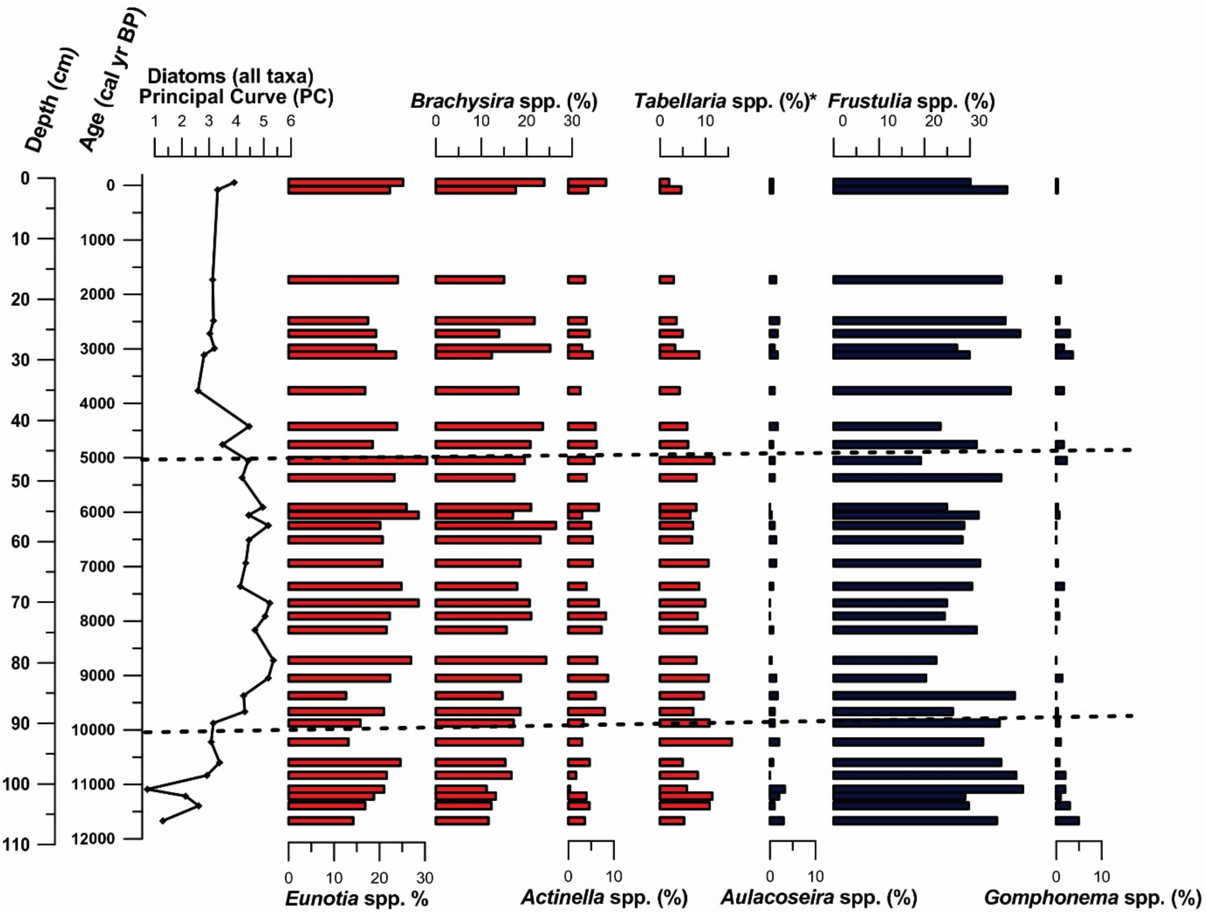

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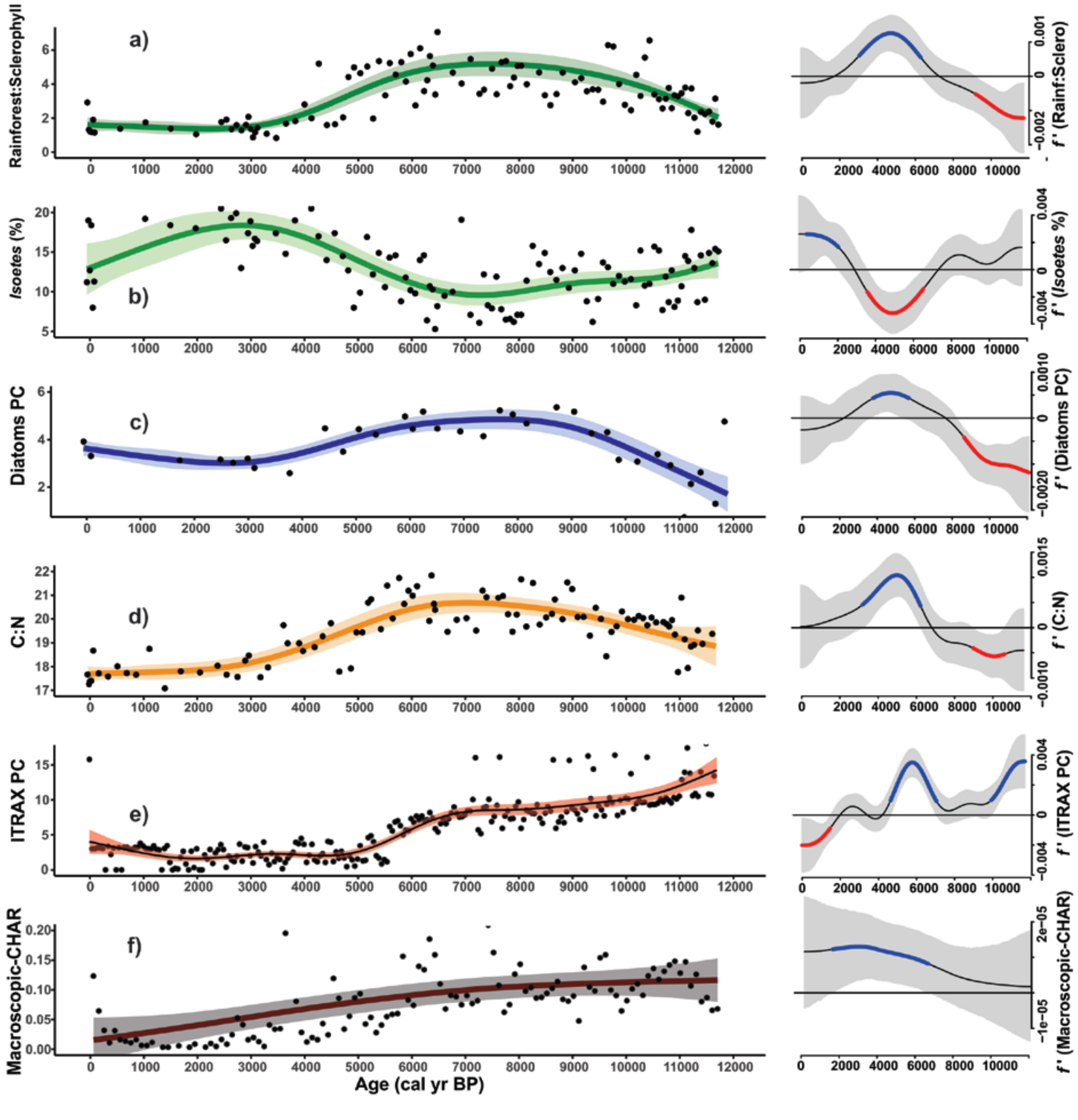

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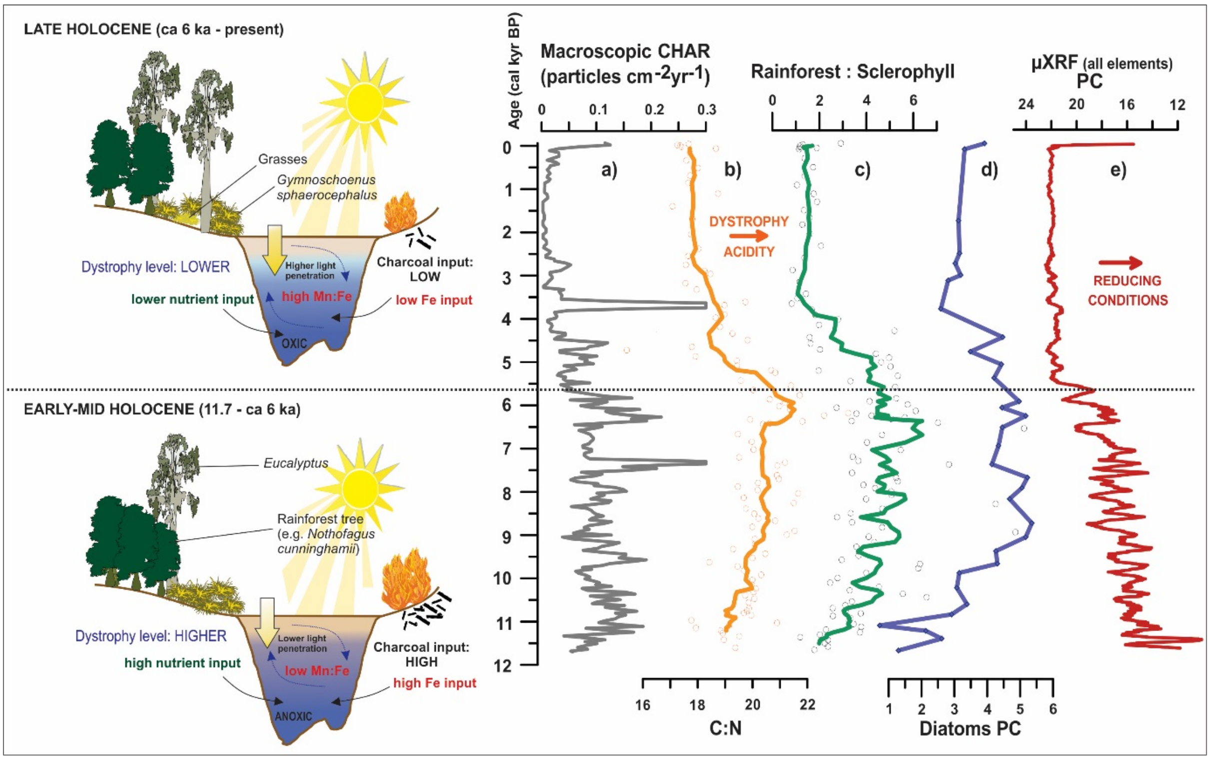

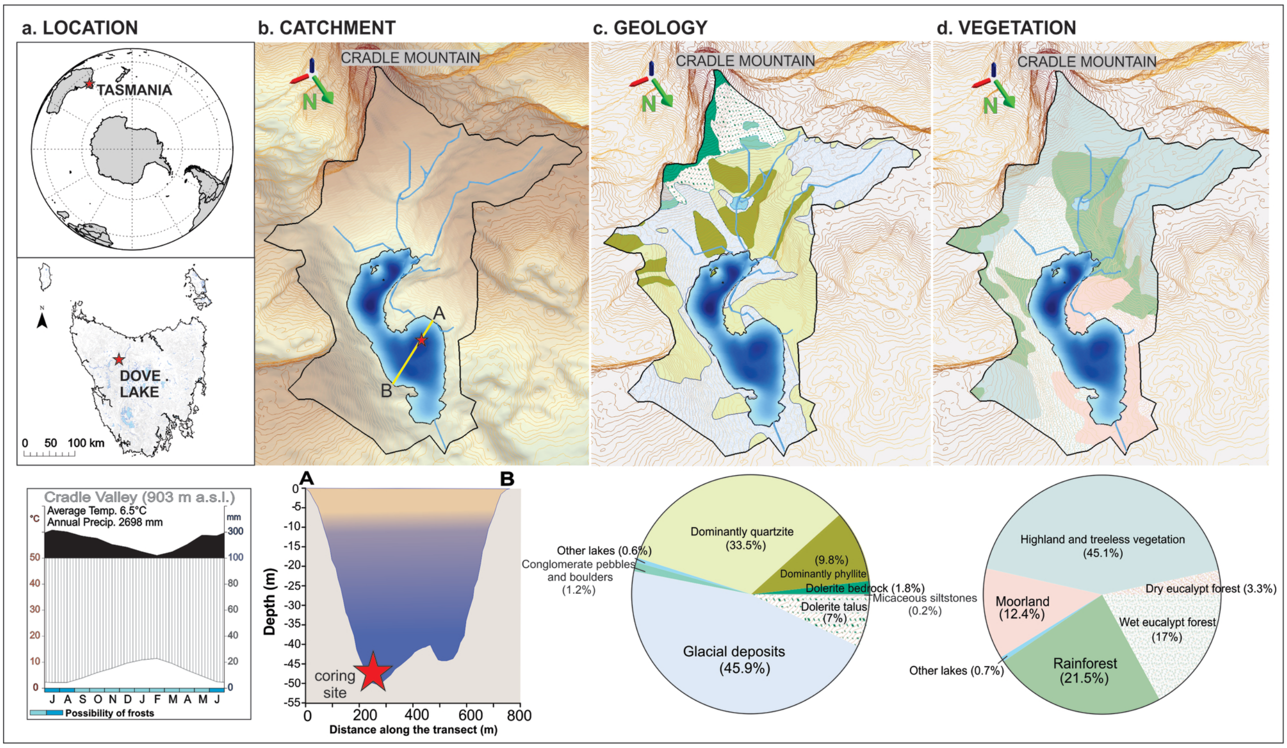

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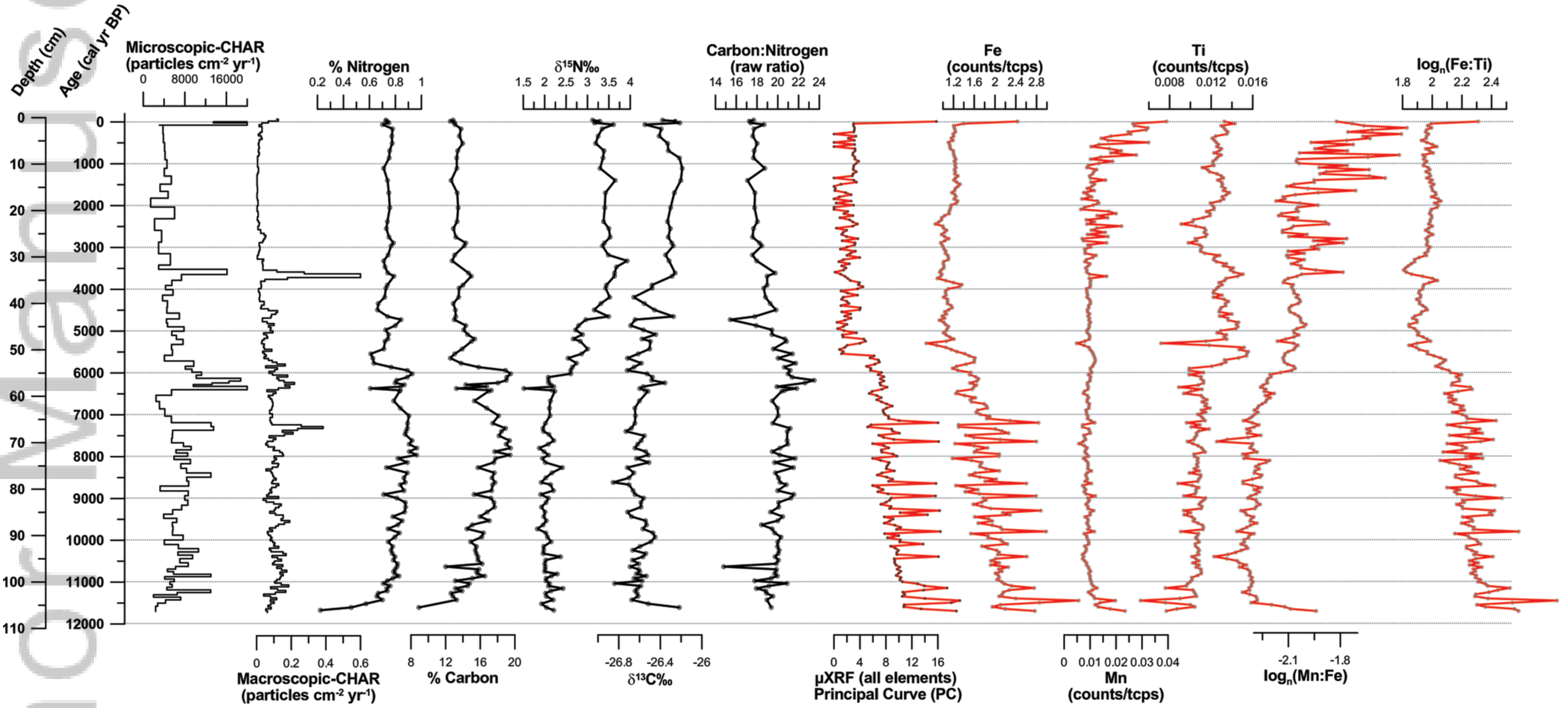

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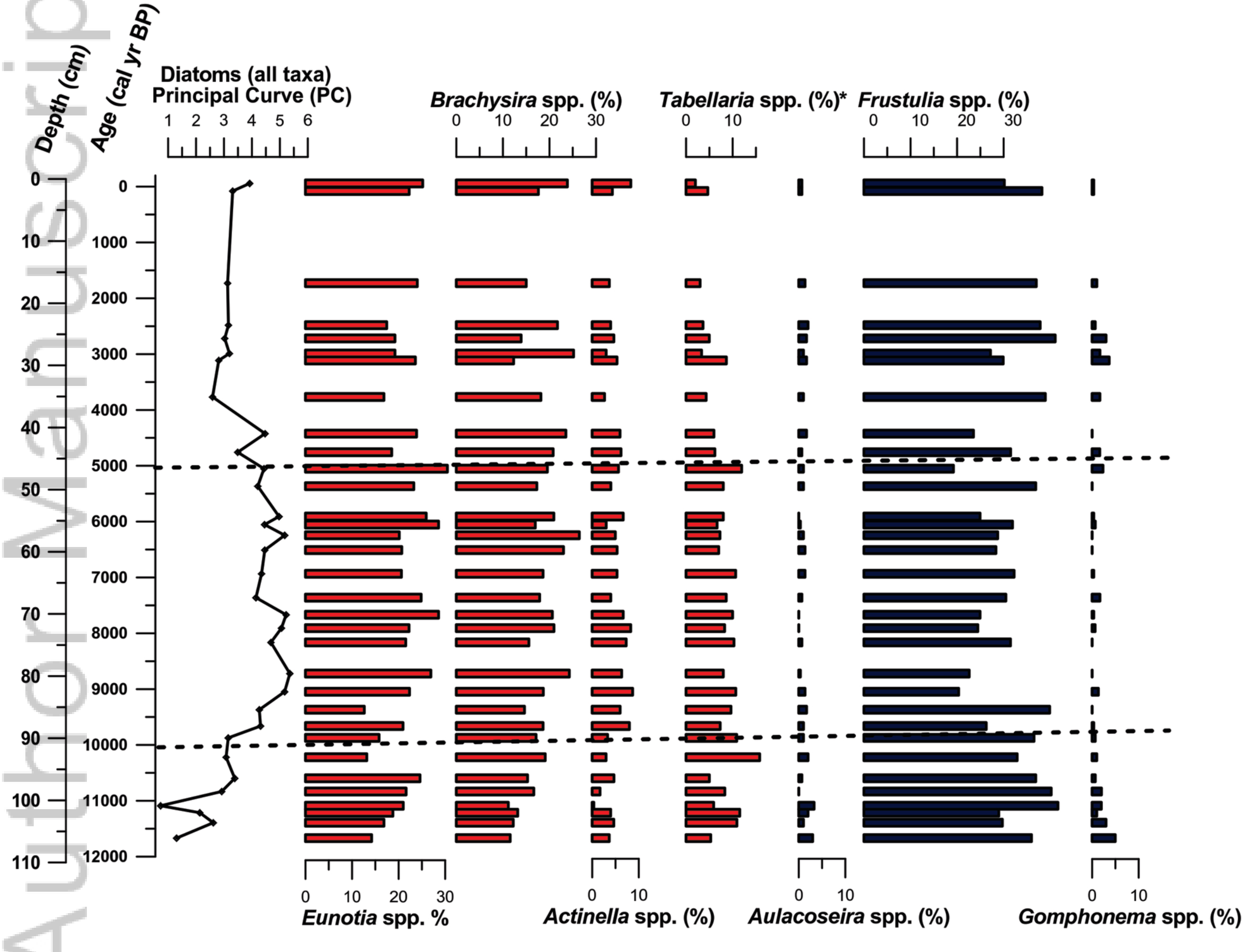

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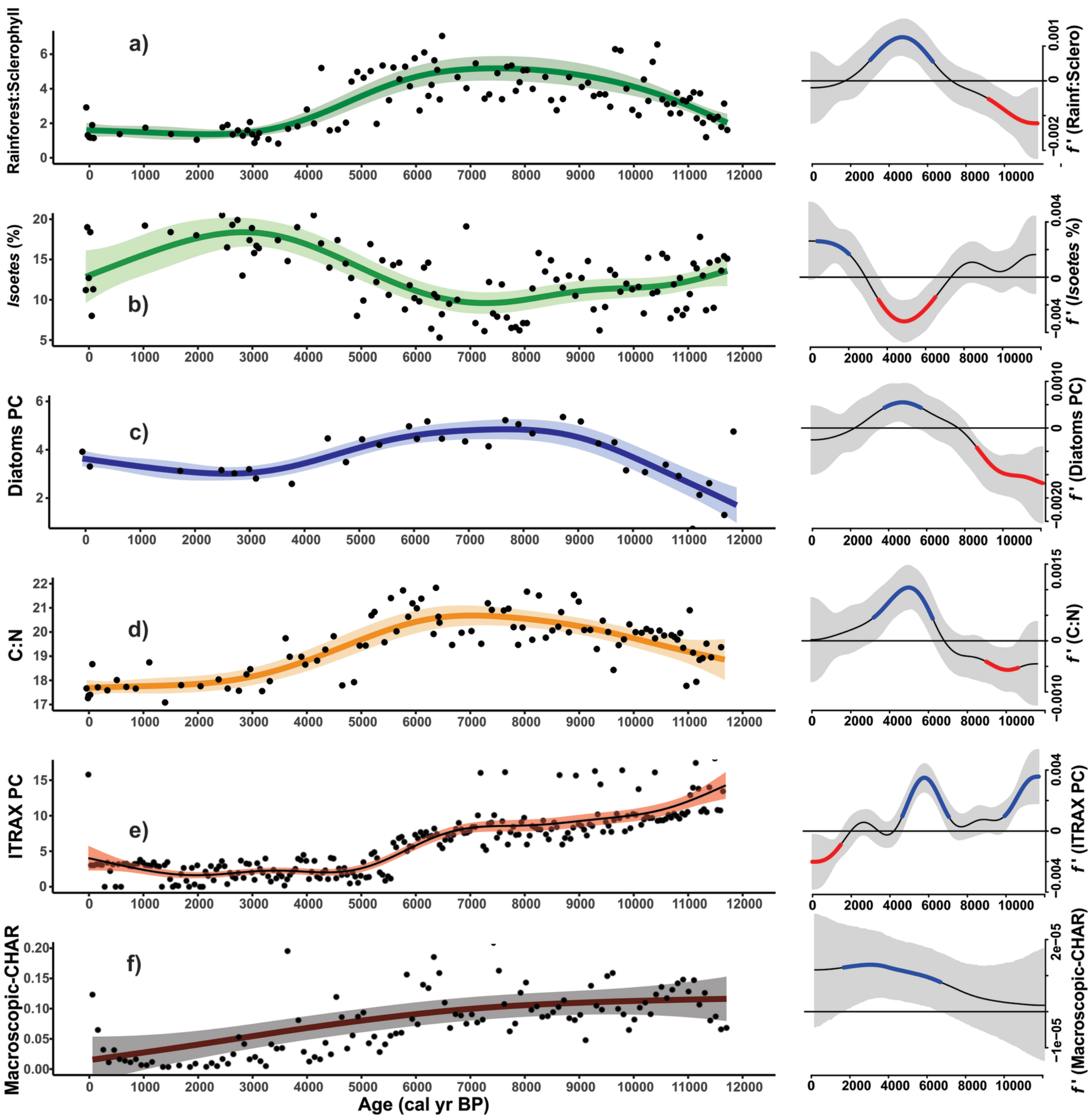

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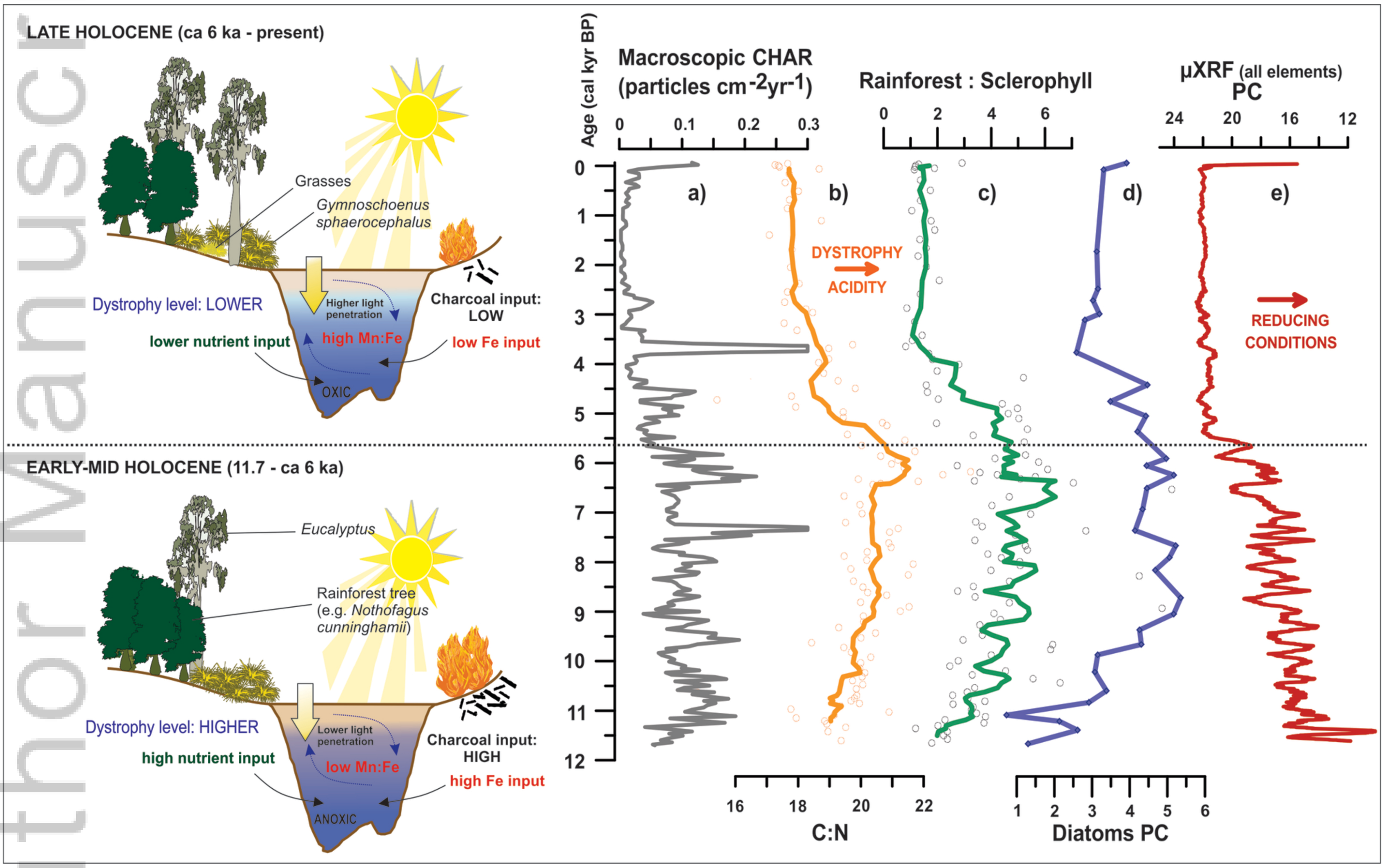

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