Citation for final published version:

Wan, Zhenzhu, Algeo, Thomas J., Gensel, Patricia G., Scheckler, Stephen E., Stein, William E., Cressler, Walter L., Berry, Christopher M. , Xu, Honghe, Rowe, Harold D. and Sauer, Peter E. 2019. Environmental influences on the stable carbon isotopic composition of Devonian and Early Carboniferous land plants. Palaeogeography, Palaeoclimatology, Palaeoecology 531 (A) , 109100. 10.1016/j.palaeo.2019.02.025

Publishers page: http://dx.doi.org/10.1016/j.palaeo.2019.02.025

Environmental influences on the stable carbon isotopic composition of Devonian and Early Carboniferous land plants

Zhenzhu Wan[a], Thomas J. Algeo[a,b,c,*], Patricia G. Gensel[d], Stephen E. Scheckler[e], William E. Stein[f], Walter L. Cressler III[g], Christopher M. Berry[h], Honghe Xu[i], Harold D. Rowe[j], and Peter E. Sauer[k]

[a] *Department of Geology, University of Cincinnati, Cincinnati, OH 45221-0013, USA*

[b] *State Key Laboratory of Biogeology and Environmental Geology, China University of Geosciences, Wuhan 430074, China*

[c] *State Key Laboratory of Geological Processes and Mineral Resources, China University of Geosciences, Wuhan 430074, China*

[d] *Department of Biology, University of North Carolina, Chapel Hill, NC 27599 USA*

[e] *Depts. of Biological Sciences and Geosciences, Virginia Polytechnic Institute and State University, Blacksburg, VA 24061-0406, USA*

[f] *Department of Biological Sciences, SUNY Binghamton, Binghamton, NY 13901, USA*

[g] *Francis Harvey Green Library, West Chester University, West Chester, PA 19383, USA*

[h] *Department of Earth Sciences, Cardiff University, Cardiff CF10 3YE, Wales, UK*

[i] *State Key Laboratory of Palaeobiology and Stratigraphy, Nanjing Institute of Geology and Palaeontology, Nanjing 210008, China*

[j] *Bureau of Economic Geology, University of Texas at Austin, Austin, TX 78713-8924 USA*

[k] *Department of Earth and Atmospheric Sciences*, *Indiana University*, *Bloomington*, *IN 47405-2208*, *USA*

*Corresponding author: thomas.algeo@uc.edu

Abstract

Systematic analysis of the stable carbon isotopic composition of fossil land plants ($\delta^{13}C_p$) has the potential to offer new insights regarding paleoclimate variation and plant-environment interactions in early terrestrial ecosystems. $\delta^{13}C_p$ was measured for 190 fossil plant specimens belonging to 10 genera of Early to Late Devonian age (*Archaeopteris*, *Drepanophycus*, *Haskinsia*, *Leclercqia*, *Pertica*, *Psilophyton*, *Rhacophyton*, *Sawdonia*, *Tetraxylopteris*, and *Wattieza*) and 2 genera of Early Carboniferous age (*Genselia* and *Rhodeopteridium*) collected from sites located mainly in the Appalachian Basin (22-30 °S paleolatitude). For the full carbon-isotopic dataset (*n* = 309), $\delta^{13}C_p$ ranges from −20.3 ‰ to −30.5 ‰ with a mean of −25.5 ‰, similar to values for modern C_3 land plants. In addition to a secular trend, $\delta^{13}C_p$ exhibits both intra- and intergeneric variation. Intrageneric variation is expressed as a small (mean 0.45 ‰) ^{13}C-enrichment of leaves and spines relative to stems that may reflect differential compound-specific compositions. Intergeneric variation is expressed as a much larger (to ~5 ‰) spread in the mean $\delta^{13}C_p$ values of coeval plant genera that was probably controlled by taxon-specific habitat preferences and local environmental humidity. Among Early Devonian taxa, *Sawdonia* yielded the most ^{13}C-depleted values (−27.1±1.7 ‰), reflecting lower water-use efficiency that was probably related to growth in wetter habitats, and *Leclercqia*, *Haskinsia*, and *Psilophyton* yielded the most ^{13}C-

enriched values (–23.0±1.6 ‰, –22.3±1.3 ‰, and –24.8±1.6 ‰, respectively), reflecting higher water-use efficiency probably related to growth in drier habitats.

Keywords: water-use efficiency; early plants; canopy effect; *Archaeopteris*; *Drepanophycus*; *Sawdonia*

1. Introduction

The Devonian was a period of revolutionary changes in terrestrial floras, landscapes, and continental climates (Algeo et al., 1995, 2001; Gibling and Davies, 2012; Gibling et al., 2014). Although bryophytes (non-vascular plants) had appeared during the mid-Ordovician and tracheophytes (vascular plants) by the late Silurian (Wellman et al., 2003; Steemans et al., 2009), the continents remained sparsely vegetated until the Devonian. That period witnessed major morphological adaptations and diversifications among vascular land plants as well as order-of-magnitude increases in the complexity and geographic extent of terrestrial ecosystems (Gensel and Edwards, 2001; Taylor et al., 2009). Early Devonian land plants were mostly small (heights of no more than a few tens of centimeters), shallowly rooted where known, and limited to moist lowland habitats (Xue et al., 2016). By the end-Devonian, vascular land plants had evolved leaves, complex root systems, secondary supporting tissues (leading to tree stature), and desiccation-resistant reproductive propagules (seeds) that facilitated their spread into drier upland habitats (Mosbrugger, 1990; Driese et al., 1997; Gerrienne et al., 2004; Meyer-Berthaud et al., 2010; Hao and Xue, 2013a; Silvestro et al., 2015). [Note: “upland” is a general term signifying land lying above the level of local stream flow and flooding.] These changes resulted in an increase in organic carbon burial, a rise of O_2 and a drawdown of CO_2 in the atmosphere, and global climatic cooling, setting the stage for the Late Paleozoic Ice Age that began in the Early Carboniferous (Algeo et al., 1995, 2001; Berner, 1997; Le Hir et al., 2011).

The stable carbon isotopic composition of plants ($\delta^{13}C_p$) differs from that of their carbon source (atmospheric CO_2) owing to photosynthetic fractionation (Farquhar et al., 1982, 1989). Modern C_3 plants fix carbon via the Calvin-Benson cycle with an average fractionation of ca. –20 ‰, yielding organic material with a $\delta^{13}C$ value of –26±5 ‰ (Raven et al., 2004). Devonian vascular land plants are thought to have been exclusively C_3 plants (Taylor et al., 2009), and their carbon isotopic compositions are therefore likely to have been controlled by the same climatic and environmental factors as for modern C_3 plants (e.g., temperature, humidity, and atmospheric $p\mathrm{CO}_2$ and $\delta^{13}C_{CO2}$). To date, the carbon isotopic compositions of specific Devonian terrestrial plant fossils (as opposed to bulk sedimentary organic carbon) have been investigated in only a few studies of fungi (Boyce et al., 2007), lichens (Fletcher et al., 2004), liverworts (Graham et al., 2010), and vascular plants (Beerling et al., 2002; Feng et al., 2014).

The present study is the first systematic investigation of carbon isotopic variation among a large group of Devonian and Early Carboniferous vascular land plants and represents by far the largest such analytical dataset to date. We made 309 measurements of $\delta^{13}C_p$ (p = plant) on 190 unique specimens of fossil plants belonging to 12 widespread genera of Early Devonian to Early Carboniferous age (Table S1). Our goals in this study were to (1) characterize $\delta^{13}C_p$ variation for each fossil plant taxon, (2) investigate time-independent $\delta^{13}C_p$ variation within and among taxa that might reflect large-scale geographic (e.g., paleolatitudinal) or local environmental influences, and (3) use these patterns of variation to draw inferences concerning the ecology of individual early land plant taxa. Here, we show that there are significant differences in $\delta^{13}C_p$ between the 12 genera of this study that are probably of primary (*in vivo*) origin, and that were most likely related to local humidity and habitat preferences among early vascular land plants.

2. Fossil plant taxonomy and paleoecology

2.1. Fossil plant taxa

The fossil plants analyzed in this study belong to 12 different genera of tracheophytes, representing early members of the lycophyte and euphyllophyte lineages (Fig. 1A). All of the fossil plants in this study have been the subject of at least partial morphological reconstructions (Fig. 1B). Although many fossil specimens were identified to the species level, we conducted our evaluation of $\delta^{13}C_p$ variation at the genus level owing to (1) lack of species assignments for about half of the individual specimens, (2) uncertainty in some species-level assignments, and (3) the impracticability of analyzing carbon-isotopic patterns in a dataset with excessive splitting into species. Among the 12 genera analyzed, 10 have overlapping ranges extending through part or all of the Devonian (*Archaeopteris*, *Drepanophycus*, *Haskinsia*, *Leclercqia*, *Pertica*, *Psilophyton*, *Rhacophyton*, *Sawdonia*, *Tetraxylopteris*, and *Wattieza*) and two are entirely of Early Carboniferous age (*Genselia* and *Rhodeopteridium*; Fig. 2). A brief summary of the key anatomical features and paleoecologic/geographic characteristics of each genus follows:

Sawdonia (Dawson) Hueber is one of the early zosterophyllopsids, belonging to the Lycophyta. This plant is pseudomonopodially branched with circinate tips; reniform sporangia are borne along the sides of some stems and anatomy consists of an exarch haplostele (Hueber, 1971; Rayner, 1983; Gensel et al., 1975; Taylor et al., 2009; Gensel and Berry, 2016). It was widespread during the Early Devonian (Gensel, 1992; Kenrick and Crane, 1997; Gensel and Berry, 2016) but is also found in the Late Devonian (Hueber and Grierson, 1961). Our dataset includes both *S. ornata* and *S. acanthotheca*, although most specimens were not distinguished at the species level.

Drepanophycus Göppert belongs to the Drepanophycaceae, sister to other lineages of Lycopsida (Kenrick and Crane, 1997). Stems range from several millimeters to centimeters in diameter and from several centimeters to a meter in length. Leaves are unbranched falcate microphylls up to several millimeters long with a single prominent middle vascular thread, and they were arranged spirally or randomly on the stem. Reniform sporangia occur on short stalks between the leaves (Li and Edwards, 1995; Li et al., 2000). However, this genus exhibits considerable variation in leaf morphology and may be in need of revision. This taxon had a creeping to erect and sometimes dichotomizing rhizomatous growth habit. It existed through most of the Devonian Period and is found in eastern Canada, northeastern USA, China, Russia, and Europe (Stubblefield and Banks, 1978; Gensel and Berry, 2001; Xu et al., 2013). About half of the specimens used in the present study belong to *D. spinaeformis*, and the remainder were not distinguished at the species level.

Leclercqia Banks, Bonamo and Grierson is a member of the Protolepidodendraceae (Lycophyta) (Gensel and Kasper, 2005; Gensel and Albright, 2006). It is a slender, herbaceous plant, with distinctive leaves that typically exhibit five divisions and that bear sporangia on their upper surface. At least some species exhibit hook-shaped leaves that may have allowed a vine-like climbing habit (Xu et al., 2011). *Leclercqia* is distinguished in being a homosporous plant with a ligule; ligules otherwise are found only in heterosporous taxa (Grierson and Bonamo, 1979; Bonamo et al., 1988; Taylor et al., 2009; Benca et al., 2014). It existed from the late Early to the Middle Devonian and has a particularly wide distribution, being found in North America, Europe, Africa, Australia, and China (Banks et al., 1972; Bonamo et al., 1988; Meyer-Berthaud et al., 2003; Taylor et al., 2009; Xu et al., 2011; Prestianni et al., 2012; Benca et al., 2014). It was present mainly in warm, dry climates (Berry, 1994; Meyer-Berthaud et al., 2003), although some studies have claimed a wider environmental range (Xu and Wang, 2008). Our dataset includes both *L. complexa* and *L. andrewsii*, although half of the specimens were not distinguished at the species level.

Haskinsia Grierson and Banks is a herbaceous lycopsid (Bonamo et al., 1988; Xu et al., 2008; Yang et al., 2008; Taylor et al., 2009). It was once regarded as a species of *Drepanophycus* but was later placed with the Protolepidodendrales because of the presence of petiolate deltoid-shaped sporophylls (Grierson and Banks, 1983; Berry and Edwards, 1996; Xu and Berry, 2008). This taxon was widespread during the Middle to early Late Devonian age, being known from North America, South America, Russia, and China; it had wide environmental tolerances, thriving in both warm tropical and cool temperate conditions (Xu et al., 2008). The relatively small number of specimens in our dataset includes *H. hastata*, *H. sagittata*, and specimens not identified at the species level.

Psilophyton (Dawson) Hueber is one of the best-known members of the trimerophyte grade of basal euphyllophytes (Andrews et al., 1968; Banks et al., 1975; Doran et al., 1978; Trant and Gensel, 1985; Stein, 1993; Gerrienne 1995, 1997). The plant consists of a main axis and multiply divided lateral branches, some of which terminate in pairs of fusiform sporangia.

Stem anatomy, where known, is a centrarch haplostele. Intrageneric variation in sporangial length and in the presence and type of emergences is significant (Doran, 1980; Gerrienne, 1995; Taylor et al., 2009). *Psilophyton* was widespread in the Early to earliest Middle Devonian and has been found in the northeastern United States (Maine and New York), Canada (Quebec and New Brunswick), Belgium, the Czech Republic, and Yunnan, China (Taylor et al., 2009; Gensel, 2017). The most common species in our dataset is *P. princeps*, although specimens of *P. crenulatum*, *P. dawsonsii*, and *P. forbesii* are also present, as well as some specimens not identified at the species level.

Pertica Kasper and Andrews is also a basal euphyllophyte (Kasper and Andrews, 1972; Granoff et al., 1976). It exhibits a dominant main stem with regularly arranged lateral branches, organized either in a tight spiral or a four ranked/decussate pattern. Lateral branch morphology varies from equally dichotomous to a more pseudomonopodial pattern. These plants bore numerous fusiform sporangia in dense clusters on ultimate branchlets (Kasper and Andrews, 1972; Granoff et al., 1976; Taylor et al., 2009). *Pertica* existed from the Early to the Middle Devonian, with occurrences known from northern Maine, New Brunswick, and Quebec. Our dataset consists mainly of the species *P. varia* along with some specimens not identified at the species level.

Wattieza refers to compression fossils belonging to the order Pseudosporochnales within the class Cladoxylopsida (Leclercq and Banks, 1962; Gensel et al., 2001; Berry and Fairon-Demaret, 2002). Pseudosporochnaleans are all similar in their adult forms, consisting of small to large trees with a main trunk bearing a dense crown of frond-like branches. Instead of leaves, the branches bear densely ramified ultimate appendages with either terminally erect

(*Pseudosporochnus*) or reflexed (*Wattieza*) sporangia. The specimens analyzed for this study, which came from the West Cave Mountain and Steenberg/South Mountain quarries in New York, possessed key characteristics of Pseudosporochnaleans including distinctive clusters of sclerotic cells (Leclercq and Banks, 1962; Gensel et al., 2001; Berry and Fairon-Demaret, 2002) but lacked sporangia. However, a recent study of new South Mountain specimens identified them as *Wattieza* and linked them to the fossil tree trunks known as *Eospermatopteris* (Stein et al., 2007). Pseudosporochnaleans existed from the Middle to early Late Devonian in Europe, Venezuela, North America, and possibly China (Gensel and Andrews, 1984; Taylor et al., 2009).

Rhacophyton Crépin belongs to the order of Rhacophytales *sensu* Taylor et al. (2009; n.b., assigned to Rhacophytaceae or Protopteridales by others). According to Andrews and Phillips (1968), this plant grew to ~1-m-tall bushes, and its foliage consisted of a main axis bearing pairs of lateral branches that fork twice basally. Two of the lateral branches were multiply divided and terminated in elongate sporangia, and the other two extended further and bore higher-order branches. In some cases, the fertile branches were produced as second-order units on first-order laterals. The stem anatomy of the lateral branches consisted of clepsydroid primary xylem surrounded in some instances by a limited amount of secondary xylem. *Rhacophyton* existed during the Late Devonian and has been found in North America, Europe, possibly Bear Island (New Brunswick), and western Siberia (Andrews and Phillips, 1968; Cornet et al., 1977; Dittrich et al., 1983; Cressler, 1999, 2006; Taylor et al., 2009). Our dataset includes a few specimens identified as *R. ceratangium* although the majority were not identified at the species level.

Rhodeopteridium (Presl) Zimmermann (formerly *Rhodea* Presl) is a genus based mainly on foliage compression/impression fossils that is thought to represent an early seed plant (Jennings, 1976). Pinnately compound leaves terminate in barely laminate ends (Read, 1955). Similar fossil leaves with well-preserved petioles are known from early seed ferns of Late Mississippian (Chesterian) age in Illinois (Jennings, 1976). Most specimens are of Early Carboniferous (Tournaisian-Namurian A) age, but it may extend into the basal late Carboniferous. None of the small number of *Rhodeopteridium* specimens in our dataset was identified at the species level.

Tetraxylopteris Beck is an aneurophytalean progymnosperm (Beck, 1957; Bonamo and Banks, 1967; Scheckler and Banks, 1971; Hammond and Berry, 2005). It is characterized by a main stem with extensive secondary xylem and bearing several orders of laterals in a decussate pattern, each with a four-armed vascular strand. In fertile regions, second-order laterals dichotomize twice, and each division bears pinnately arranged clusters of sporangia. *Tetraxylopteris* existed during the Givetian and early Frasnian. To date, it is known from only two areas: the Catskill Delta in New York, and the Campo Chico Formation in northwestern Venezuela. A few of our specimens are assigned to *T. schmidtii* but most were not identified at the species level.

Archaeopteris (Dawson) Stur belongs to the Archaeopteridalean progymnosperms. It was a tall tree similar to some modern conifers, but it had pseudomonopodial branching in the lateral branch system (Beck, 1962) and laminate leaves with helices or decussate patterns of leaf attachment (Scheckler, 1978). Some ultimate clusters of leaves appear cone-like, bearing sporangia on their adaxial surfaces, but all species are thought to have been free-sporing and heterosporous. *Archaeopteris* is usually preserved as impression and compression fossils, and its fossilized trunks, named *Callixylon* when found separately, exhibit extensive secondary xylem. It ranged from the latest Middle to Late Devonian, with a number of different species being known from localities in North America, Russia, Europe, Morocco, China, Australia, and Colombia (Beck and Wight, 1988; Cressler, 1999, 2006; Meyer-Berthaud et al., 1999; Berry et al., 2000). By the middle of the late Frasnian, monospecific archaeopterid forests had become the dominant vegetation type in lowland areas and coastal settings over a vast geographic area (Algeo et al., 2001). About one third of the 51 specimens in our dataset are assigned to various species, including *A. fissilis*, *A. halliana*, *A. jacksonii*, *A. macilenta*, and *A. rogersii*, although most specimens were not identified at the species level.

Genselia Knaus may have been a seed fern (Knaus, 1995; Taylor et al., 2009). This genus has bipinnate compound leaves, non-bifurcate rachises, and in some, bifurcations at the tip of stems, terminating in elongated sporangia (Knaus, 1994, 1995; Skog and Gensel, 1980; Taylor et al., 2009). It is found in the Lower Carboniferous Pocono and Price Formations of Pennsylvania, Maryland, West Virginia and Virginia, in the Appalachian Basin of North America. None of the *Genselia* specimens in our dataset was identified at the species level.

2.2. Paleoclimate and paleoenvironments

The locales from which fossil plants in this study were collected have a quasi-global distribution (Fig. 3A), although a large majority of the analyses (n = 289 of 309, or 93 % of the total dataset) are of specimens from the Appalachian Basin or other areas in eastern North America, at locales ranging from southwestern Virginia in the south to the Gaspé Peninsula of

Québec in the north (Fig. 3B). Laurentia (Paleozoic North America) drifted northward during the Devonian Period (416-359 Ma), causing the Appalachian Basin to shift from ~40–35 ˚S in the Early Devonian to ~30–25 ˚S in the Late Devonian (van der Voo, 1988, 1993). Thus, many of the present study locales moved from the humid temperate zone into the dry subtropical zone during this interval. However, this shift occurred at tectonic timescales (i.e., over a ~50-Myr interval) and was monotonic in direction (i.e., did not reverse), so any regional climate changes induced by plate motion during this interval are likely to have been slow and steady. More rapid climate changes (i.e., at timescales of a few million years or less) are likely to have had non-plate-tectonic causes. Most of the remaining fossil plant specimens (n = 20, 7 % of total) are from a few sites in northwestern China (Xu et al., 2015; Zheng et al., 2016) and Venezuela (Berry, 1994; Berry and Edwards, 1996).

The depositional environments in which the 12 fossil plant taxa of the present study grew ranged from coastal deltas to upper floodplains, with the former being the most common environment. Coastal deltas are represented, for example, by most of the fossil-plant-bearing strata of the Emsian Battery Point Formation in Gaspé Bay, Quebec, and the more western portions of the Emsian-lower Eifelian Campbellton Formation in New Brunswick (Griffing et al., 2000; Hotton et al., 2001; Kennedy et al., 2012, 2013). Especially at Gaspé Bay, paleocurrent data document mainly seaward transport of sediment by rivers, and multistory sandstone bodies are interpreted as deposits of high-sinuosity river channels (Griffing et al., 2000). Closer to the coastline, bidirectional cross-bedding records tidal influence (Bridge, 2000; Griffing et al., 2000) and some trace fossils (e.g., *Diplocraterion*) may represent brackish conditions within estuaries (Lawrence, 1986). Deltaic processes are evidenced by a complex stratigraphic architecture of levees and crevasse splays, freshwater lakes and marshes, lacustrine deltas, brackish marshes and interdistributary bays, and sandy and muddy tidal flats (Griffing et al., 2000).

Coastal wetlands apparently developed frequently during the late Givetian to early Frasnian, especially at times of relatively high sea level, which resulted in marine incursions into generally terrestrial environments (Baird and Brett, 2008). Multiple wetland subenvironments are represented by stacked dark meter-thick sandstones at Riverside Quarry, Gilboa (Stein et al., 2012), dark siltstones and shales at Blenheim, Gilboa (Banks et al., 1972), and lighter gray to yellowish-brown sandstones at Manorkill Falls, Gilboa (Driese et al., 1997). Similar environments developed in the Late Devonian, as in the Famennian Hampshire Formation at Elkins, West Virginia (Scheckler, 1986a). In these settings, trees and understory plants appear to have had limited root penetration. During the Late Devonian, wetland floral diversity was relatively limited: *Archaeopteris* and *Rhacophyton* were endemic, although other taxa (especially arborescent lycophytes) occupied peri-swamp habitats and were washed in during floods and storms (Scheckler, 1986a). By the Tournaisian, coastal swamp habitats were increasingly dominated by lycophytes (Scheckler, 1986b; Rygel et al., 2006; Gensel and Pigg, 2010).

Upper floodplain settings are represented by fine-grained, multistory sandstone bodies and red mudstones in fining-upward successions lacking any evidence of marine influence. These deposits represent wide, meandering upland river channels and overbank (floodplain) deposits that were subject to frequent wet-dry cycles, e.g., through seasonal flooding (Cressler, 1999, 2006; Cressler et al., 2010). Examples of this type of environment are found in the

Famennian Duncannon Member of the Catskill Formation at Red Hill, Pennsylvania. Taphonomic observations indicate that both fossil plants and animals lived close to their sites of deposition. Fossil plant remains are often found in reduced lenses of greenish-gray mudstone, probably representing floodplain pond deposits related to major channel avulsion events (Cressler, 1999, 2006; Cressler et al., 2010).

3. Methods

The study specimens consist of well-preserved compression fossils of whole plants or parts of plants (i.e., stems, branches, and/or leaves) that are intact or nearly so (Fig. 4). All specimens were inspected using a binocular microscope to verify the state of fossil preservation, and a subset was examined using scanning electron microscopy. We initially bulk macerated each plant fossil in acid baths, yielding bulk organic material that was undifferentiated by plant anatomical parts. These specimens were digested successively in hydrochloric acid (HCl) and hydrofluoric acid (HF), with three rinses in distilled water between dissolution steps. We subsequently sampled the plant fossils by peeling pieces of organic material from the surface of each compression fossil, permitting us to analyze specific anatomical parts of each plant (e.g., stems, leaves, spines, and sporangia). These specimens were treated individually with HCl and HF, as needed, to remove rock matrix adhering to the organic material. Organic fragments were picked out of the residue, rinsed in distilled water, dried, and inspected under a binocular microscope to verify the removal of all impurities.

Carbon isotopic analyses were performed using EA-IRMS systems at the Stable Isotope Research Facility (SIRF) of the Department of Earth and Atmospheric Sciences of Indiana

University, Bloomington (IU) and the Department of Earth and Environmental Sciences of the University of Texas at Arlington (UTA). Analytical protocols were similar in the two laboratories: powdered samples were weighed into silver capsules that were acidified repeatedly with 6% sulfurous acid (H_2SO_3) in order to remove carbonate phases. Samples were analyzed using a Costech 4010 elemental analyzer interfaced with a Thermo Finnigan Conflo III device to a Thermo Finnigan Delta Plus XP isotope ratio mass spectrometer (IRMS). All isotopic results are reported in per mille (‰) variation relative to VPDB. At UTA, the average standard deviation was 0.04 ‰ for both the USGS-24 standard ($\delta^{13}C$ = –16.049 ‰) and unknowns. At IU, the average standard deviation was 0.06 ‰ for Acetanilides #1, #2, and #3, and Corn starch #1 (Schimmelmann et al., 2016) and unknowns. Interlaboratory calibration was undertaken by analyzing a common set of 12 samples at both IU and UTA. The co-analyzed samples yielded a correlation (r^2) of 0.997 with an offset of 0.21 ‰ between the two labs, which was corrected for in the integrated C-isotope dataset by averaging the difference (i.e., by reducing UTA values by 0.10 ‰ and increasing IU values by 0.10 ‰).

4. Results

4.1. General results

In this study, we generated a total of 309 analyses of plant $\delta^{13}C$ from 190 unique plant specimens (Table S1). The number of analyses per taxon varied considerably: the four most analyzed taxa were *Archaeopteris* (80 analyses of 51 unique specimens), *Drepanophycus* (40/21), *Rhacophyton* (33/18), and *Tetraxylopteris* (31/21) (Table 1). About half of the specimens were analyzed twice, mostly to generate paired $\delta^{13}C$ values for different anatomical parts (e.g., stems and leaves) but sometimes to check on the reproducibility of results for the same anatomical part of a single specimen. A handful of specimens were analyzed three or more times. The full dataset of 309 analyses yielded a mean $\delta^{13}C_p$ of –25.5 ‰ with a standard deviation of 1.8 ‰ and a range from –20.3 ‰ to –30.5 ‰. Individual taxa yielded mean $\delta^{13}C_p$ ranging from –22.3±1.3 ‰ for *Haskinsia* to –27.3±1.7 ‰ for *Sawdonia* (Table 1).

The full $\delta^{13}C_p$ dataset exhibits a distinct secular pattern (as reflected in a LOWESS curve) that is dominated by a long-term shift toward higher $\delta^{13}C_p$ values through most of the Devonian and Early Carboniferous (note: this secular pattern is not shown and will not be considered here because it is the focus of a companion paper that is in preparation). To determine the non-secular component of total $\delta^{13}C_p$ variation, we calculated the difference between each $\delta^{13}C_p$ value and the age-equivalent mean for the full $\delta^{13}C_p$ dataset as given by the LOWESS curve, i.e., $\Delta^{13}C_p$ (Fig. 5). Thus, a fossil specimen with a $\Delta^{13}C_p$ of 0 ‰ has a carbon-isotopic composition identical to the LOWESS mean for the $\delta^{13}C_p$ dataset at a given time, and positive and negative $\Delta^{13}C_p$ values represent ^{13}C enrichment and depletion of the specimen, respectively, relative to the age-equivalent LOWESS mean value. In this manner, the overall tendency of each fossil plant taxon toward ^{13}C enrichment or depletion can be evaluated independently of long-term secular variation in the $\delta^{13}C_p$ dataset (Fig. 6). The non-secular variance in our $\delta^{13}C_p$ dataset has two components: (1) variance among different plant taxa (‘intergeneric variation’), and (2) variance within individual taxa (‘intrageneric variation’). The proportion of total variance in the $\Delta^{13}C_p$ dataset represented by intrageneric variance (i.e., $\sigma^2_{intra} / \sigma^2_{total}$) was calculated as:

$$\left(\sum_{i=1}^{12} [\Sigma(X-\mu_i)^2 / N_i]\right) \; / \; [\Sigma(X-\mu_t)^2 / N_t] \qquad (1)$$

where X is the $\Delta^{13}C_p$ for a given fossil specimen, μ_i is the mean $\Delta^{13}C_p$ for a given plant genus *i*, and μ_t is the mean $\Delta^{13}C_p$ for all fossil specimens. The numerator of the equation sums the variances of the 12 fossil plant genera ($\sum_{i=1}^{12}$) calculated separately (i.e., $[\Sigma(X-\mu_i)^2 / N_i]$), and it is divided by the total variance for all fossil specimens (i.e., $[\Sigma(X-\mu_t)^2 / N_t]$) to yield the fraction of total variance attributable to intrageneric variance (70%); the remainder (30 %) represents intergeneric variance. Below, we analyze intrageneric and intergeneric patterns of variation as a function of possible taxonomic, anatomical, geographic, and environmental controls (Tables 2-3). Note that this analysis is conducted on the basis of $\Delta^{13}C_p$ values (Table S1), i.e., the deviations from the long-term secular $\delta^{13}C_P$ trend (Fig. 6), and is thus independent of the secular component of variance in the $\delta^{13}C_p$ dataset.

We considered the possibility of a geographic gradient in $\Delta^{13}C_p$, e.g., along a paleolatitudinal transect. Modern plants show a distinct latitudinal gradient in $\delta^{13}C_p$ that is a function of temperature and precipitation influences (Diefendorf et al., 2010) and/or light levels (Kohn, 2010). However, the high proportion (>90 %) of specimens from the Appalachian Basin in this study (Figs. 2-3) leads to a clustering of data within a relatively narrow range of paleolatitudes, resulting in our inability to identify any significant paleolatitudinal trends. A more robust test of geographic influences on Devonian fossil plant $\delta^{13}C$ will require a larger dataset covering a wider range of paleolatitudes.

4.2. Intrageneric variation in plant $\delta^{13}C$

For a subset of specimens (*n* = 32), we analyzed carbon from both the stem and the leaf or spine (not interpreted as a leaf homologue) of the fossil plant, allowing us to evaluate variation in $\delta^{13}C_p$ as a function of plant anatomy. The assessment of anatomical variation was made for 6 plant genera (Table 3). Among these genera, four showed higher $\delta^{13}C_p$ for leaves/spines than for stems, although the difference was statistically significant (based on a Student's t-test) only for *Sawdonia*, which yielded the largest $\Delta^{13}C_{(spine-stem)}$ value of any genus (1.6 ‰). The relatively small number of analyses per genus was a factor limiting the statistical significance of these results for individual plant taxa. All 6 genera together (*n* = 32) yielded a mean leaf/spine-vs-stem difference of 0.45 ‰, which is statistically significant ($p(\alpha) = 0.01$; Table 3). These results suggest that fossil plant leaves and spines are systematically slightly enriched in ^{13}C relative to stems of the same fossil plant specimen (as seen in 29 out of 32 of the paired analyses).

4.3. Intergeneric variation in plant $\delta^{13}C$

The 12 fossil plant genera of this study exhibit systematic differences in $\Delta^{13}C_p$ (Fig. 7). Three taxa yielded mean $\Delta^{13}C_p$ values that are significantly higher than the age-equivalent LOWESS mean: *Haskinsia* (+2.4±1.5 ‰; *n* = 4), *Leclercqia* (+1.8±1.0 ‰; *n* = 24), and *Psilophyton* (+0.9±1.5 ‰; *n* = 25), and one taxon yielded a significantly lower mean value: *Sawdonia* (–1.2±1.8 ‰; *n* = 28) (note: for all, $p(\alpha) \leq 0.01$; Student's t-test; Table 2). The remaining eight taxa have mean $\Delta^{13}C_p$ values within ±0.5 ‰ of the average for the dataset as a whole, among which only *Archaeopteris* yielded a significantly different mean (–0.4±1.2 ‰; $p(\alpha) = 0.002$), mainly as a result of the exceptionally large number of analyses of this taxon (n = 80; Table 2). However, the result for *Archaeopteris* must be viewed cautiously because there are relatively few coeval Late Devonian taxa in our dataset from which to construct the LOWESS mean trend.

Thus, we consider only *Haskinsia*, *Leclercqia*, *Psilophyton*, and *Sawdonia* to deviate significantly in $\Delta^{13}C_p$ from age-equivalent LOWESS mean values.

The critical time interval of our dataset for evaluating interspecific $\Delta^{13}C_p$ variation is the late Early through late Middle Devonian (~405-385 Ma), during which 6 of the 12 plant taxa analyzed in this study have largely overlapping stratigraphic ranges. The overlap in stratigraphic ranges facilitates intergeneric comparisons, and, not coincidentally, all four of the taxa showing large deviations from mean LOWESS $\delta^{13}C_p$ values are present in this interval, with three taxa showing ^{13}C-enriched values (*Haskinsia*, *Leclercqia*, and *Psilophyton*) and one showing ^{13}C-depleted values (*Sawdonia*) (see above). Importantly, the offsets in $\delta^{13}C_p$ between these taxa are relatively constant despite an overall increase in mean LOWESS $\delta^{13}C_p$ of ~3 ‰ from the late Early Devonian to the late Middle Devonian (Algeo et al., in preparation). We attribute these taxon-specific differences in ^{13}C enrichment to local environmental controls (see Section 5.3). In contrast, the 6 plant genera analyzed from the early Late Devonian to Early Carboniferous (~385-345 Ma) have only partially overlapping stratigraphic ranges, with no more than 2 taxa present in any given time window. The more limited dataset for this time interval renders uncertain evaluation of deviations in $\Delta^{13}C_p$ from the mean LOWESS trend (thus our caution with regard to interpreting *Archaeopteris* $\Delta^{13}C_p$ values).

5. Discussion

5.1. Robustness of plant $\delta^{13}C$ proxy

The fossil plant specimens used in this project were preserved as compressions of stems, branches, leaves and spines (Fig. 4A-B). During the carbonization process, most water and volatile organic compounds in the plant tissue were lost, and the remaining material was preserved as a black organic film that consists largely of carbon (Guo et al., 2010). Even though compressed as flat layers, many fossils show well-preserved cell-scale anatomical features such as tracheids (Fig. 4C) and cuticle (Fig. 4D). These observations confirm that pre-burial bacterial decay of these relatively resistant tissues was limited in the specimens that were selected for carbon-isotopic analysis in this study.

Vascular land plants exhibit internal carbon isotopic variation, commonly with a >5 ‰ range in δ^{13}C among their constituent compounds (O'Leary, 1988; Pate and Arthur, 1998). Although most types of compounds decay rapidly in the burial environment, the surviving compounds generally retain their characteristic carbon isotopic compositions (Benner et al., 1987). Cellulose and lignin are among the most resistant compound types and commonly dominate the terrestrial organic fraction preserved in sedimentary rocks (Arens et al., 2000), especially in compression fossils of the type analyzed in this study. Cellulose is up to ~3 ‰ heavier and lignin up to ~4 ‰ lighter than bulk plant δ^{13}C (Benner et al., 1987; O'Leary, 1988; Marino and McElroy, 1991; Badeck et al., 2005). Biomarker analysis of plant compression fossils typically yields a range of alkanes, alkenes, benzenes, pristenes, and polysaccharides derived in large part from decay of these primary compounds (e.g., Gupta et al., 2007), so a part of the variance in the $\Delta\delta^{13}C_P$ of an individual taxon in our dataset may reflect specimen-specific variation in the proportions of cellulose versus lignin and their derivatives (Arens et al., 2000).

Burial diagenesis can potentially modify the δ^{13}C composition of organic material through thermal cracking and release of methane at high temperatures (i.e., >160 ˚C, or beyond the 'oil window'; Faure and Mensing, 2005). The released methane is depleted in ^{13}C by 10-

30 ‰, resulting in a small increase in the $\delta^{13}C$ of the remaining kerogen (generally <1 ‰). The thermal maturity of Devonian sediments in the central Appalachian Basin exhibits a strong lateral gradient: maturity is high on the eastern basin margin (R_o >2.0; T >160 ˚C) but low to moderate on the western basin margin (R_o <1.0; T <100 ˚C) (Obermajer et al., 1997; Milici and Swezey, 2006; Rowan, 2006; Repetski et al., 2008). In our study areas in eastern New York and central West Virginia, the maximum burial depths of Upper Devonian strata were from ~4 to 10 km, with peak burial temperatures estimated to have been between ~150 and 170 °C (Friedman and Sanders, 1982; Dorobek, 1989; Repetski et al., 2008). Thermal maturity is also variable in New Brunswick and the Gaspé Peninsula of Québec in eastern Canada. There, some Devonian strata have been buried just a few kilometers (Heroux et al., 1979), whereas other units have experienced maximum burial depths up to 12 km (Utting and Hamblin, 1991), yielding peak burial temperature estimates ranging from ~50 to 280 °C (Bertrand and Malo, 2001; Chi et al., 2001). We tested the possibility of burial thermal effects on our dataset by plotting $\delta^{13}C_p$ against inferred maximum burial depths, but this yielded almost no correlation (r = +0.03; n = 309; $p(\alpha)$ >0.5). We are cognizant that the carbon isotopic compositions of our plant fossils may have been influenced by bacterial and thermal decay. However, the $\delta^{13}C_p$ values of a single taxon at a specific outcrop tend to be relatively consistent in our dataset (mostly showing <1 ‰ variation), indicating a general lack of random diagenetic effects on the plant fossils during burial.

5.2. Sources of anatomical variation in plant $\delta^{13}C$

Among modern plants, different plant parts exhibit systematic differences in carbon isotopic compositions. Based on a compilation of >400 plant taxa, leaves are on average 0.96 ‰ and 1.91 ‰ more depleted in ^{13}C than roots and woody stems, respectively (Badeck et al., 2005). This pattern is strong but not universal as ~10 % of the taxa examined in that study yielded leaf δ^{13}C greater than stem δ^{13}C. This pattern appears to be independent of climatic conditions, as plants from semi-arid habitats also exhibit leaf δ^{13}C lower than stem δ^{13}C (by an average of 1.51±0.42 ‰) (Nilson and Sharifi, 1997). The origin of these differences is not known with certainty: two leading ideas relate it to carbon-isotopic fractionation in generating different types of organic compounds (see Section 5.1), or preferential release of ^{13}C-enriched respiratory CO_2 from leaves (Badeck et al., 2005).

In our dataset, leaves and spines are somewhat ^{13}C-enriched relative to stems (by 0.45 ‰ on average; Table 3; see Section 4.2). If the δ^{13}C values of leaves and spines were originally lower than that of stems by ~1-2 ‰ in Devonian plants, then burial decay must have caused a relative shift of +1.5 to +2.5 ‰ in leaf/spine δ^{13}C relative to stem δ^{13}C. Such a shift might have occurred owing to a greater loss of lipids and/or increase in cellulose-derived compounds in the carbonized residue of leaves and spines relative to stems during the fossilization process. Further investigation will be needed to validate the existence and significance of internal δ^{13}C variation in Devonian and Carboniferous fossil plants.

5.3. Sources of intergeneric variance in plant δ^{13}C

5.3.1. Water-use efficiency

One mechanism that can lead to systematic differences in $\delta^{13}C$ values between plant taxa is differences in water-use efficiency (WUE): plants that grow in wet habitats do not need to limit water loss and, hence, tend to open their stomata more widely in order to maximize CO_2 uptake at the cost of low water-use efficiency. This process leads to maximum internal-leaf pCO_2 and maximum carbon isotope discrimination (i.e., lower $\delta^{13}C_p$) (Farquhar et al., 1989). In contrast, plants that grow in drier habitats are forced to limit water loss through their stomata (i.e., higher water-use efficiency), leading to less CO_2 uptake, lower internal-leaf pCO_2, and reduced carbon-isotope discrimination (i.e., higher $\delta^{13}C_p$) as a result of a larger fraction of internal-leaf CO_2 being utilized in photosynthesis (Farquhar et al., 1989).

The carbon isotopic composition of modern C_3 land plants exhibits a strong negative relationship to mean annual precipitation (MAP) (Miller et al., 2001; Liu et al., 2005; Roden et al., 2005; Diefendorf et al., 2010; Kohn, 2010). An increase in MAP from 0 to 1000 mm yr^{-1} correlates with a ~4-5 ‰ depletion in $\delta^{13}C_p$, although this relationship is non-linear because $\delta^{13}C_p$ becomes less sensitive to changes in precipitation at high MAP (Diefendorf et al., 2010). This relationship exists because land plants are more commonly water-limited than carbon-limited and have a strong incentive to engage in greater water-use efficiency where water is scarce (Bacon, 2004; Macfarlane et al., 2004). Plants tend to reduce stomatal conductance in arid regions in order to conserve water, which causes a larger proportion of the CO_2 that diffuses into the leaf to be used in photosynthesis, resulting in reduced fractionation (relative to the atmospheric carbon source) and heavier $\delta^{13}C_p$ values (Farquhar et al., 1989; Brugnoli and Lauteri, 1991; Poss et al., 2000). The opposite pattern is found in humid regions, causing $\delta^{13}C_p$

values to become lighter. This relationship accounts for ~55 % of $\delta^{13}C$ variance among modern C_3 plants (Diefendorf et al., 2010).

A fundamental divide exists between species with high hydraulic conductivity and low embolism resistance and those with low hydraulic conductivity and high embolism resistance (Wilson, 2016). Mapping of hydraulic ecospace shows that the high-conductivity/low-safety-margin domain was occupied by many early vascular land plants, including trimerophytes, lycophytes, cladoxylopsids, and sphenopsids, all of which developed tracheids with scalariform pits that maximized hydraulic conductivity (Wilson and Knoll, 2010; Wilson and Fischer, 2011; Wilson, 2013, 2016). For some plants, this strategy was associated with perennially wet habitats in which the danger of desiccation was limited, e.g., as for arborescent lycophytes and other plants that inhabited tropical lowland swamps during the Carboniferous Period. For other plants, this hydraulic mechanism was associated with an ecological strategy based on rapid growth and generational overturn (known as 'live fast, die young'), which possibly included all of the zosterophyllophytes and basal euphyllophytes of the present study. For example, *Psilophyton dawsonii* may have grown rapidly, as suggested by xylem with large pits similar to that of some modern small ferns whose aerial vegetative structures rarely persist for more than a year (Wilson, 2016). An additional factor in the high conductivity of early vascular plants was high atmospheric CO_2 levels, which permitted smaller and less numerous stomata (thus minimizing some types of cavitation hazards) and generally higher water-use efficiency (Sperry, 2003).

It has been suggested that early land plants were mostly generalists, occupying a wide range of habitats (Spicer, 1989; Meyer-Berthaud et al., 2003). However, the systematic variation in $\Delta^{13}C_p$ among Devonian plant taxa observed in this study (Fig. 7), if due to environmental controls, implies that some early vascular land plants may have been more specific in their habitat preferences than previously thought. Wetter habitats may have been favored by most zosterophyllophytes, which typically had small, shallow root systems (Gensel et al., 2001; Xue, 2012). *Sawdonia* exhibits the relatively most ^{13}C-depleted compositions among the 12 study taxa (Fig. 7). It occupied a wide range of environments but was most common along the margins of interdistributary basins in lower delta plain settings (Griffing et al., 2000; Hotton et al., 2001; Kennedy et al., 2012). Sedimentologic investigations have not established for certain whether these basins were inundated mainly with brackish (Hotton et al., 2001) or fresh waters (Kennedy et al., 2012). The low $\Delta^{13}C_p$ compositions of *Sawdonia* documented in the present study are more consistent with fresh waters, because C_3 plants growing in coastal salt marshes or in areas of elevated groundwater salinity tend to become ^{13}C-enriched rather than ^{13}C-depleted (Brugnoli and Lauteri, 1991; Malamud-Roam and Ingram, 2001).

Drier habitats may have been occupied by some of the plant taxa examined in this study. In particular, *Leclercqia*, *Haskinsia*, and, to a lesser degree, *Psilophyton* exhibit ^{13}C-enriched compositions consistent with higher water-use efficiency (Fig. 7). This inference is supported by the anatomical features of these plants. The morphologies of *Leclercqia* and *Haskinsia* are similar to modern *Lycopodium* s.l., which often grows in seasonally dry habitats (Fernandez et al., 2008). In these taxa, densely crowded and overlapping leaves can create a boundary layer effect that is effective at minimizing water loss during gas exchange, and which is therefore favored in dry habitats. In addition, the leaves of *Leclercqia* and *Haskinsia* partially enclosed the sporangia, protecting them from drying out (Meyer-Berthaud et al., 2003; Gensel and Kasper, 2005; Gensel and Albright, 2006). Certain features in *Psilophyton* may have adapted it to sunny, seasonally dry habitats, e.g., forked cylindrical lateral branchlets and a prominent outer cortex of axial fibers that provided mechanical support (possibly against wilting) and UV protection (via the high lignin content of fibrous cell walls) (Trant and Gensel, 1985; Gerrienne, 1995, 1997). These ^{13}C-enriched taxa may have grown where soils were better drained, e.g., on river levees or in upper floodplain settings that were further inland and somewhat drier than coastal environments, as proposed for *Psilophyton* (Hotton et al., 2001).

In areas of limited precipitation, increases in soil salinity can exert a strong influence on $\delta^{13}C_p$. Experimental and in-situ studies of modern C_3 plants have shown that $\delta^{13}C_p$ increases by 2-5 ‰ with rising soil salinity (Brugnoli and Lauteri, 1991; Poss et al., 2000; Winter and Holtum, 2005). A similar effect is observed in coastal salt marshes, where the $\delta^{13}C_p$ of C_3 marsh plants increases by several per mille with rising watermass salinity (Malamud-Roam and Ingram, 2001; Cloern et al., 2002). This effect is linked to the need to conserve water in more saline environments, leading to reduced stomatal conductance and photosynthetic fractionation and, thus, higher $\delta^{13}C_p$ values (Farquhar et al., 1989; Brugnoli and Lauteri, 1991; Poss et al., 2000). However, there is no evidence that any early vascular land plant grew in saline soils or brackish-water environments (Kennedy et al., 2012), and our results are consistent with this inference. The apparent water-use efficiencies of our fossil plant taxa suggested by their $\Delta^{13}C_p$ values (Fig. 7) correspond well with predictions of habitat preference based on paleoecological analyses (see Section 5.4). Further carbon isotopic study of early vascular land plants may prove invaluable in helping to define their habitat preferences.

5.3.2. Forest understory $\delta^{13}C$ gradient

A second environmental factor that can lead to systematic differences in $\delta^{13}C$ values between plant taxa is vertical $\delta^{13}C_{CO2}$ variation in the forest understory atmosphere. Forests are characterized by a higher rate of soil respiration than most other vegetation biomes (Raich and Tufekciogul, 2000), as well as by more limited airmass exchange than in open landscapes owing to the sheltering effects of a closed canopy (Feigenwinter et al., 2004). This combination of factors results in elevated pCO_2 in the forest understory atmosphere along with a significant vertical $\delta^{13}C_{CO2}$ gradient (Jackson et al., 1993). At 0.5 m above the forest floor, up to ~20 % of CO_2 is soil-derived (Da Silveira et al., 1989), exposing low-growing plants to strongly ^{13}C-depleted (–28 to –25 ‰) soil-respired CO_2 (Farquhar et al., 1989; Jackson et al., 1993). As a consequence, CO_2 in the forest understory atmosphere may be up to ~3-5 ‰ depleted relative to CO_2 in the open atmosphere, and this isotopic signature can be transferred to understory plants (Schleser and Jayasekera, 1985; Flanagan et al., 1996). In modern forests, >50 % of $\delta^{13}C$ variance in understory plants is linked to local variations in $\delta^{13}C_{CO2}$ (Da Silveira et al., 1989).

'Canopy effects' are unlikely to have played any role in the intergeneric differences in $\Delta^{13}C_p$ of the present study. The largest differences in $\Delta^{13}C_p$ values are seen among Early and early Middle Devonian plants that existed prior to the first forests (Fig. 6). These early plants grew in patchy floral communities consisting largely of clonal vegetation whose shoots would have lacked any capacity to limit airmass exchange (Gensel and Edwards, 2001; Edwards and Richardson, 2004), and whose limited productivity did not generate high excess soil pCO_2 (Mora et al., 1996; Elick et al., 1998). The appearance of the first forests during the late Givetian to early Frasnian (Decombeix et al., 2011; Stein et al., 2012; Berry and Marshall, 2015), an event known as 'afforestation' (Scheckler, 2001), resulted in denser floral communities with closed canopies formed by large leafy trees that, for the first time, would have generated understory niches with varying environmental characteristics. In our dataset, the only co-existing Late Devonian taxa that have the potential to exhibit $\Delta^{13}C_p$ differences due to a 'canopy effect' are *Archaeopteris*, a canopy tree, and *Rhacophyton*, a much smaller woody bush (Fig. 1). However, *Rhacophyton* yields $\Delta^{13}C_p$ values that are on average ~1.5 ‰ heavier than those of *Archaeopteris* (Fig. 6), which is the opposite of the pattern expected for canopy-versus-understory plants. We infer that the higher $\Delta^{13}C_p$ values of *Rhacophyton* are more likely due to its preference for dry habitats, leading to enhanced water-use efficiency (see Section 5.3.1).

5.4. Relationship of $\delta^{13}C$ to habitat preferences of early vascular plants

Although it has been proposed that early land plants were mostly generalists (Spicer, 1989; Meyer-Berthaud et al., 2003), the carbon-isotopic evidence of the present study suggests that Devonian and Early Carboniferous plant taxa varied considerably in their habitat preferences. Systematic interspecific differences in $\Delta^{13}C_p$ among the 12 taxa examined (Fig. 7) were probably controlled mainly by water-use efficiency, related to humidity levels at their sites of growth (see Section 5.3.1). To illustrate inferred differences in habitat, we have generated reconstructions of Early, Middle, and Late Devonian terrestrial ecosystems (Fig. 8A-C). These reconstructions focus on the distributions of the 12 plant genera of the present study, with the spatial distribution of each taxon shown to conform to its relative water-use efficiency as inferred from taxon-specific differences in $\Delta^{13}C_p$.

Our reconstructions of Devonian land-plant habitats were informed by the findings of earlier studies of terrestrial ecosystems. One consideration in habitat reconstruction is the in-situ versus transported character of land plant fossils. Most plant fossils have been transported prior to final deposition and burial (e.g., Pratt and van Heerde, 2017), although fossils that consist of intact stems and leaves and are well-preserved suggest limited transport distances (Hotton et al., 2001; Allen and Gastaldo, 2006; Kennedy et al., 2012; Gastaldo, 2016). Nearly all of the fossil plant specimens of the present study are well-preserved and relatively intact, often consisting of complete stems, twigs or leaves, and more rarely of dense intertangled mats of vegetation. Thus, it is unlikely that they were transported long distances prior to burial (cf. Berry and Edwards, 1996; Gastaldo, 2016; Pratt and van Heerde, 2017). Such fossils are considered to be ‘parautochthonous’, i.e., transported to only a limited degree and thus useful for environmental interpretations of their growth habitat (Hotton et al., 2001; Allen and Gastaldo, 2006; Kennedy et al., 2012).

During the Early to Middle Devonian, land plants were concentrated in coastal delta and lower floodplain habitats (Hotton et al., 2001). From the Middle Devonian, land plants began to penetrate more deeply into continental interiors and increasingly occupied upper floodplain settings (Algeo et al., 1995; Berry et al., 2000; Berry and Fairon-Demaret, 2001; Cressler et al., 2010; Retallack and Huang, 2011). However, the upland regions of continental interiors remained mostly devoid of vegetation until the advent of seed plants in the latest Devonian to Early Carboniferous (Algeo and Scheckler, 1998; Decombeix et al., 2011). Early to Middle Devonian plants appear to exhibit a wider range of $\Delta\delta^{13}C_p$ variation than Late Devonian to Early Carboniferous plants. This pattern may reflect greater environmental variation in Early-Middle

Devonian terrestrial habitats. During this early stage of landscape colonization, higher land plants had probably densely colonized some areas (e.g., delta plains) and sparsely colonized others (e.g., upland floodplains), while leaving broad continental interiors largely unvegetated. As a consequence of this mosaic floral distribution, land areas may have exhibited substantial spatial variation in environmental characteristics (e.g., humidity, soil development, albedo, etc.) (cf. Edwards and Richardson, 2004).

Early Devonian terrestrial ecosystems (Fig. 8A) have been analyzed in studies of the Emsian-age Battery Point Formation on the Gaspé Peninsula of Québec and the Campbellton Formation in New Brunswick. Facies A of the Cap-aux-Os Member of the Battery Point Formation contains unimodal paleocurrent indicators in coastal delta-plain facies that have been interpreted as crevasse splay or storm washover deposits (Hotton et al., 2001). The most common taxon in this setting is *Sawdonia*, which is inferred to have lived close to the shoreline on the margins of interdistributary basins that were subject to frequent flooding. Hotton et al. (2001) inferred that flooding events introduced brackish or marine waters, but Kennedy et al. (2012) inferred that similar occurrences of *Sawdonia* in the Campbellton Formation existed in fully freshwater habitats. Facies B of the Cap-aux-Os Member represents prograding fluvial channel deposits, hence mainly freshwater conditions, although some of the sandstones contain asymmetric bimodal cross-bedding indicative of tidal influence (Hotton et al., 2001). This facies contains abundant *Pertica* and *Drepanophycus* (as well as other taxa not analyzed in this study) that are thought to have grown along lower floodplain channel margins, whereas *Psilophyton* was inferred to have occupied an equivalent habitat somewhat further upstream. Similar assemblages containing *Pertica*, *Drepanophycus*, *Psilophyton*, and *Leclercqia* have been reported from coastal delta plain and freshwater fluvial deposits of the Campbellton Formation in New Brunswick (Kennedy et al., 2012) and the Trout Valley Formation of Maine (Allen and Gastaldo, 2006).

Early Devonian vascular plants were dominantly ground-hugging rhizomatous lycophytes (*Sawdonia*, *Drepanophycus*, *Leclercqia*, *Haskinsia*) and shrubby trimerophytes (*Psilophyton*, *Pertica*), mostly with heights of <1 m (Fig. 1B; Algeo and Scheckler, 1998). Many Early Devonian plants grew in dense monospecific stands, facilitated by predominantly rhizomatous growth strategies, allowing local resource domination (Fig. 8A; DiMichele and Hook, 1992; Xue, 2012). However, mixtures of plant fossil debris in many formations imply that such patchiness was relatively local, and that there was considerable taxonomic heterogeneity in Early Devonian landscapes at a slightly larger spatial scale (Allen and Gastaldo, 2006; Cressler et al., 2010; Gastaldo, 2016). This pattern may reflect habitat specialization ('floral partitioning') on the basis of environmental variability in soil moisture, nutrient levels, or environmental characteristics such as frequency of flooding or other disturbances (Allen and Gastaldo, 2006; Greb et al., 2006).

By the late Middle Devonian (Givetian; Fig. 8B), the development of secondary supporting tissues resulted in an increase in average plant heights and the development of the earliest forests in coastal wetland regions (DiMichele and Hook, 1992; Greb et al., 2006; Mintz et al., 2010; Stein et al., 2012; Berry and Marshall, 2015). Shrubby stands of aneurophytalean progymnosperms, lycopsids, cladoxylopsids, and zosterophyllophytes were widespread in lowland areas (Fig. 8B; Berry and Fairon-Demaret, 2001; Xue et al., 2018). Early forests consisted of the cladoxylopsid tree *Eospermatoperis* with an understory flora that included lycopsids and aneurophytalean progymnosperm shrubs such as *Tetraxylopteris* (Driese et al., 1997; Stein et al., 2007, 2012; Mintz et al., 2010; Xu et al., 2017). The rapid global spread of forests may have been linked to archaeopteridalean progymnosperm trees with laminate leaves and deep root systems. The appearance of early members of this clade in the early Givetian is signaled by the microspore *Geminospora lemurata* and the megaspore *Contagisporites optivus* (Marshall, 1996; Turnau, 2014), and their widespread presence in mid to late Givetian landscapes is attested by specimens of *Svalbardia* or proto-*Archaeopteris* (Berry, 2008).

Late Devonian landscapes were dominated by dense monospecific stands of *Archaeopteris*, with *Rhacophyton* and other shrubby plants either occupying open terrain or present as understory elements in forests (Fig. 8C). Archaeopterid progymnosperms formed extensive forests in both lower and upper floodplain environments (Scheckler, 1986a; Beck and Wight, 1988; Meyer-Berthaud et al., 1999), and *Archaeopteris* is thought to have generally favored dry riparian habitats (Cressler, 1999, 2006; Retallack and Huang, 2011). *Rhacophyton* grew as a bushy understory plant in forests subject to frequent ground fires (Cressler, 2001, 2006), although it also flourished in peat wetlands (Greb et al., 2006). The earliest known seed plants became established in the mid-Famennian as small bushy colonizers in disturbed habitats, but they did not become important members of terrestrial ecosystems until the Carboniferous (Scheckler, 1986b; Streel et al., 2000; Decombeix et al., 2011).

5.5. Middle/Late Devonian floral turnover

Floral turnovers were a regular feature of early Devonian plant communities owing to the rapid pace of evolutionary innovation at that time (Cascales-Miñana et al., 2010). The rhyniophytes, which, as the earliest tracheophyte clade, were morphologically simple, peaked in dominance during the Late Silurian and earliest Devonian (Lochkovian). By the Early Devonian, the zosterophyllophytes had evolved lateral sporangia, pseudomonopodial branching, and a rhizomatous habit, establishing them as the most morphologically complex, diverse, and abundant clade (Gensel, 1992; Edwards and Richardson, 2004; Cascales-Miñana and Meyer-Berthaud, 2015). They peaked during the mid to late Early Devonian (Pragian-Emsian) but gradually declined as lycophytes and basal euphyllophytes gained in importance during the early Middle Devonian (Eifelian) (Hao and Xue, 2013b; Cascales-Miñana and Meyer-Berthaud, 2015). Among other innovations, basal euphyllophytes developed planate or laminate lateral branched systems resembling proto-leaves (Gerrienne et al., 2014). The turnovers among these clades appear to have been competitive replacements that were not clearly linked to any specific climatic or tectonic event (Cascales-Miñana and Meyer-Berthaud, 2015).

The Givetian/Frasnian (Middle/Late Devonian) transition appears to mark another major floral turnover (Raymond and Metz, 1995; Silvestro et al., 2015). During this event, primitive spore-bearing plants such as zosterophyllophytes experienced high extinction rates (Cascales-Miñana and Meyer-Berthaud, 2014) and euphyllophytes, including both lignophytes and non-lignophytes, underwent a major diversification (Xue et al., 2018). Among the taxa of the present study, the euphyllophytes *Psilophyton* and *Pertica* declined in the Emsian-Eifelian, and the zosterophyllophytes and lycopsids *Drepanophycus*, *Sawdonia*, *Leclercqia*, and *Haskinsia* declined in the Givetian (Fig. 2). Although some of these taxa (e.g., *Drepanophycus*) persisted through the Famennian, their relative importance in the Late Devonian was greatly reduced. This floral turnover is even more apparent in palynomorph assemblages, which exhibit a sharp decline in diversity and rapid turnover at the Middle/Late Givetian boundary across Russia and eastern Europe and, to a lesser degree, in France, although it has not reported to date from North America (Obukhovskaya, 2000; Turnau, 2014).

One important aspect of the Givetian-Frasnian floral turnover was the appearance and diversification of arborescent (tree-sized) lycophytes and lignophytes (Wang et al., 2005; Galtier and Meyer-Berthaud, 2006; Meyer-Berthaud et al., 2010; Decombeix et al., 2011). In this study, the Givetian/Frasnian transition is marked by short-lived arborescent taxa such as *Wattieza* and *Tetraxylopteris*, after which longer-ranging arborescent taxa such as *Archaeopteris* became dominant in the Late Devonian (Fig. 2). The evolutionary development of trees led directly to the first forests, which were of Givetian (late Middle Devonian) to earliest Frasnian (early Late Devonian) age. They were dominated by tree-sized pseudosporochnalean cladoxylopsids such as *Wattiezia/Eospermatoperis* with an understory that included aneurophytaleans such as *Tetraxylopteris*, as well as possibly herbaceous and arborescent lycopsids (Driese et al., 1997; Stein et al., 2007, 2012; Mintz et al., 2010). The known distribution of pseudosporochnalean forests is mostly limited to the Euramerican continental block, but they were succeeded by archaeopterid-dominated forests on all continents during the later Frasnian to Famennian (Scheckler, 1986a, 2001). The advent of forests probably generated selective pressures on terrestrial plants either to evolve larger size or to adapt to a shaded understory. The near-simultaneous appearance of woody supporting tissues and tree-sized stature among arborescent lycophytes, sphenopsids, and lignophytes may imply the first strategy (Mosbrugger, 1990; Meyer-Berthaud et al., 2010). Less certain, due to difficulties in sampling, is what might be inferred about plant evolution in the understory. Nevertheless, the spread of forests likely had multifaceted, profound and irreversible consequences for both terrestrial floral communities and global climate conditions.

6. Conclusions

Carbon-isotopic variation among 12 genera of Devonian-Early Carboniferous vascular land plants provides insights into their habitat preferences and water-use efficiencies. A total of 309 analyses of 190 unique specimens yielded $\delta^{13}C_p$ values ranging from −20.3 ‰ to −30.5 ‰ with a mean of −25.5 ‰, similar to the range and mean of $\delta^{13}C$ values for modern C_3 land plants. *Sawdonia* yielded the most ^{13}C-depleted values (mean −27.1±1.7 ‰; $n = 28$), reflecting lower water-use efficiency that was probably related to growth in wetter habitats such as lower delta plains. Because salt-marsh vegetation is typically ^{13}C-enriched, the strongly ^{13}C-depleted composition of *Sawdonia* is suggestive of growth in freshwater habitats. In contrast, *Leclercqia*, *Haskinsia*, and *Psilophyton* yielded relatively ^{13}C-enriched values (means −23.0±1.6 ‰, −22.3±1.3 ‰, and −24.8±1.6 ‰, respectively), reflecting higher water-use efficiency related to growth in drier habitats such as upper floodplains. This inference is supported by anatomical adaptations to drier conditions exhibited by these taxa, e.g., related to leaf arrangement and structure. A large majority of our specimens (>90 %) are from the Appalachian Basin of eastern North America, precluding effective assessment of large-scale geographic patterns of carbon-isotopic variation in these fossil plant taxa. This study demonstrates that investigations of the carbon-isotopic composition of well-preserved plant fossils have the potential to yield insights regarding habitat preferences and ecosystem structure in paleofloral communities.

Acknowledgments

Thanks to the Peabody Museum of Yale University, the State Museum of New York, and the Smithsonian Institution for access to their paleobotanical collections. ZZW thanks Xinda Hu, Teng Teng, and N. Doug Rowe for field assistance. This study was supported by graduate research grants to ZZW from the American Association of Petroleum Geologists, the Geological Society of America, the Paleontological Society, the Society for Sedimentary Geology, and the University of Cincinnati Research Council.

References

Algeo, T.J., Scheckler, S.E., 1998. Terrestrial-marine teleconnections in the Devonian: links between the evolution of land plants, weathering processes, and marine anoxic events. Phil. Trans. Royal Society (B) 353, 113-130.

Algeo, T.J., Berner, R.A., Maynard, J.B., Scheckler, S.E., 1995. Late Devonian oceanic anoxic events and biotic crises: “rooted” in the evolution of vascular land plants? GSA Today 5(3), 45 & 64-66.

Algeo, T.J., Scheckler, S.E., Maynard, J.B., 2001. Effects of early vascular land plants on weathering processes and global chemical fluxes during the Middle and Late Devonian. In:

Gensel, P., Edwards, D. (Eds.), Plants Invade the Land: Evolutionary and Environmental Perspectives. Columbia Univ. Press, New York, pp. 213-236.

Allen, J.P., Gastaldo, R.A., 2006. Sedimentology and taphonomy of the Early to Middle Devonian plant-bearing beds of the Trout Valley Formation, Maine. In: Greb, S.F., DiMichele, M.A. (Eds.), Wetlands Through Time. Geol. Soc. Am. Spec. Publ. 399, pp. 57-78.

Andrews, H.N., Phillips, T.L., 1968. *Rhacophyton* from the Upper Devonian of West Virginia. Bot. J. Linn. Soc. 61, 37-64.

Andrews, H.N., Kasper, A., Mencher, E., 1968. *Psilophyton forbesii*, a new Devonian plant from northern Marine. Bulletin of the Torrey Botanical Club 95(1), 1-11.

Arens, N.C., Jahren, A.H., Amundson, R., 2000. Can C3 plants faithfully record the carbon isotopic composition of atmospheric carbon dioxide? Paleobiology 26(1), 137-164.

Bacon, M.A., 2004. Water-use efficiency in plant biology. In: Bacon, M.A. (Ed), Water-Use Efficiency in Plant Biology. Blackwell, Oxford, pp. 1-26.

Badeck, F.W., Tcherkez, G., Nogues, S., Piel, C., Ghashghaie, J., 2005. Post-photosynthetic fractionation of stable carbon isotopes between plant organs—a widespread phenomenon. Rapid Comm. Mass Spectr. 19(11), 1381-1391.

Baird, G.C., Brett, C.E., 2008. Late Givetian Taghanic bioevents in New York State: New discoveries and questions. Bulletin of Geosciences (Czech Geological Survey) 83(4), 357-370.

Banks, H.P., Bonamo, P.M., Grierson, J.D., 1972. *Leclercqia complexa* gen. et sp. nov., a new lycopod from the Late Middle Devonian of eastern New York. Rev. Palaeobot. Palynol. 14(1-2), 19-40.

Banks, H.P., Leclercq, S.M, Hueber, F.M., 1975. Anatomy and morphology of *Psilophyton dawsoni*, sp. n. from the Late Lower Devonian of Quebec (Gaspé) and Ontario, Canada. Palaeontogr. Amer. 8(48), 77-127.

Beck, C.B., 1957. *Tetraxylopteris schmidtii* gen. et sp. nov., a probable pteridosperm precursor from the Devonian of New York. Am. J. Bot. 42, 743-749.

Beck, C.B., 1962. Reconstruction of *Archaeopteris*, and further consideration of its phylogenetic position. Am. J. Bot. 49(4), 373-382.

Beck, C.B., Wight, D.C., 1988. Progymnosperms. In: Beck, C.B. (Ed.), Origin and Evolution of Gymnosperms. Columbia Univ. Press, New York, pp. 1-84.

Beerling, D.J., Lake, J.A., Berner, R.A., Hickey, L.J., Taylor, D.W., Royer, D.L., 2002. Carbon isotope evidence implying high O_2/CO_2 ratios in the Permo-Carboniferous atmosphere. Geochim. Cosmochim. Acta 66, 3757-3767.

Benca, J.P., Carlisle, M.H., Bergen, S., Strömberg, C.A., 2014. Applying morphometrics to early land plant systematics: A new *Leclercqia* (Lycopsida) species from Washington State, USA. Am. J. Bot. 101(3), 510-520.

Benner, R., Fogel, M.L., Sprague, E.K., Hodson, R.E., 1987. Depletion of ^{13}C in lignin and its implications for stable carbon isotope studies. Nature 329, 708-710.

Berner, R.A., 1997. The rise of plants and their effect on weathering and atmospheric CO_2. Science 276(5312), 544-546.

Berry, C.M., 1994. First record of the Devonian lycophyte *Leclercqia* from South America. Geological Magazine 131(2), 269-272.

Berry, C. M., 2008. The Middle Devonian plant collections of François Stockmans reconsidered. Geologica Belgica 12, 25-30.

Berry, C.M., Edwards, D., 1996. The herbaceous lycophyte *Haskinsia* Grierson and Banks from the Devonian of western Venezuela, with observations on the leaf morphology and fertile specimens. Bot. J. Linn. Soc. 122, 103-122.

Berry, C.M., Fairon-Demaret, M., 2001. The Middle Devonian flora revisited. In: Gensel, P., Edwards, D. (Eds.), Plants Invade the Land: Evolutionary and Environmental Perspectives. Columbia Univ. Press, New York, pp. 120-139.

Berry, C.M., Fairon-Demaret, M., 2002. The architecture of *Pseudosporochnus nodosus* Leclercq et Banks: A Middle Devonian cladoxylopsid from Belgium. Int. J. Plant Sci. 163, 699-713.

Berry, C.M., Marshall, J.E.A., 2015. Lycopsid forests in the early Late Devonian paleoequatorial zone of Svalbard. Geology 43, 1043-1046.

Berry, C.M., Morel, E., Mojica, J., Villarroel, C., 2000. Devonian plants from Colombia, with discussion of their geological and palaeogeographical context. Geol. Mag. 137, 257-268.

Bertrand, R., Malo, M., 2001. Source rock analysis, thermal maturation, and hydrocarbon generation in the Siluro-Devonian rocks of the Gaspé Belt basin, Canada. Bull. Can. Petr. Geol. 49, 238-261.

Bonamo, P.M., Banks, H.P., 1967. *Tetraxylopteris schmidti*: its fertile parts and its relationships within the Aneurophytales. Am. J. Bot. 54(6), 755-768.

Bonamo, P.M., Banks, H.P., Grierson, J.D., 1988. *Leclercqia*, *Haskinsia*, and the role of leaves in delineation of Devonian lycopod genera. Bot. Gaz. 149(2), 222-239.

Boyce, C.K., Hotton, C.L., Fogel, M.L., Cody, G.D., Hazen, R.M., Knoll, A.H., Heuber, F.M., 2007. Devonian landscape heterogeneity recorded by a giant fungus. Geology 35, 399-402.

Bridge, J.S., 2000. The geometry, flow patterns and sedimentary processes of Devonian rivers and coasts, New York and Pennsylvania, USA. In: Friend, P.F., Williams, B.P.J. (Eds.), New Perspectives on the Old Red Sandstone. Geol. Soc. Lond. Spec. Publ. 180, pp. 85-108.

Brugnoli, E., Lauteri, M., 1991. Effects of salinity on stomatal conductance, photosynthetic capacity, and carbon isotope discrimination of salt-tolerant (*Gossypium hirsutum* L.) and salt-sensitive (*Phaseolus vulgaris* L.) C3 non-halophytes. Plant Physiol. 95, 628-635.

Cascales-Miñana, B., Meyer-Berthaud, B., 2014. Diversity dynamics of Zosterophyllopsida. Lethaia 47(2), 205-215.

Cascales-Miñana, B., Meyer-Berthaud, B., 2015. Diversity patterns of the vascular plant group Zosterophyllopsida in relation to Devonian paleogeography. Palaeogeogr. Palaeoclimatol. Palaeoecol. 423, 53-61.

Cascales-Miñana, B., Muñoz-Bertomeu, J., Ros, R., Segura, J., 2010. Trends and patterns in the evolution of vascular plants: macroevolutionary implications of a multilevel taxonomic analyses. Lethaia 43, 545-557.

Chi, G., Lavoie, D., Bertrand, R., 2001. Hydrocarbon inclusions entrapped in diagenetic minerals in Paleozoic basins in eastern Canada—Implications for hydrocarbon exploration. Geol. Surv. Canada Contr. No. 2000252.

Cloern, J.E., Canuel, E.A., Harris, D., 2002. Stable carbon and nitrogen isotope composition of aquatic and terrestrial plants of the San Francisco Bay estuarine system. Limnol. Oceanogr. 47(3), 713-729.

Cornet, B., Phillips, T.L., Andrews, H.N., 1977. The morphology and variation in *Rhacophyton ceratangium* from the Upper Devonian and its bearing on frond evolution. Palaeontographica (B) 158, 105-129.

Cressler, W.L., III, 1999. Site-analysis and floristics of the Late Devonian Red Hill locality, Pennsylvania, an *Archaeopteris*-dominated plant community and early tetrapod site. Unpubl. dissertation, U. of Pennsylvania, Philadelphia, Pennsylvania, 156 pp.

Cressler, W.L., III, 2001. Evidence of earliest known wildfires. Palaios 16, 171-174.

Cressler, W.L., III, 2006. Plant paleoecology of the Late Devonian Red Hill locality, north-central Pennsylvania, an *Archaeopteris*-dominated wetland plant community and early tetrapod site. In: Greb, S.F., DiMichele, M.A. (Eds.), Wetlands Through Time. Geol. Soc. Am. Spec. Publ. 399, pp. 79-102.

Cressler, W.L., III, Daeschler, E.B., Slingerland, R., Peterson, D.A., 2010. Terrestrialization in the Late Devonian: a palaeoecological overview of the Red Hill site, Pennsylvania, USA. In: Vecoli, M., Clément, G., Meyer-Berthaud, B. (eds.), The Terrestrialization Process: Modelling Complex Interactions at the Biosphere-Geosphere Interface, Geological Society of London, Spec. Publ. 339, pp. 111-128.

Da Silveira Lobo Sternberg, L., Mulkey, S.S., Wright, S.J., 1989. Ecological interpretation of leaf carbon isotope ratios: influence of respired carbon dioxide. Ecology 70(5), 1317-1324.

Decombeix, A.-L., Meyer-Berthaud, B., Galtier, J., 2011. Transitional changes in arborescent lignophytes at the Devonian-Carboniferous boundary. J. Geol. Soc. London 168, 547-557.

Diefendorf, A.F., Mueller, K.E., Wing, S.L., Freeman, K.H., 2010. Global patterns in leaf ^{13}C discrimination and implications for studies of past and future climate. Proc. Nat. Acad. Sci. USA 107, 5738-5743.

DiMichele, W.A., Hook, R.W., 1992. Paleozoic terrestrial ecosystems. In: Behrensmeyer, A.K., Damuth, J.D., DiMichele, W.A., Potts, R., Sues, H.-D., Wing, S.L. (Eds.), Terrestrial Ecosystems Through Time. Univ. Chicago Press, Chicago, pp. 205-325.

Dittrich, H.S., Matten, L.C., Phillips, T.L., 1983. Anatomy of *Rhacophyton ceratangium* from the Upper Devonian (Famennian) of West Virginia. Rev. Palaeobot. Palynol. 40, 127-47.

Doran, J.B. 1980. A new species of *Psilophyton* from the Lower Devonian of northern New Brunswick, Canada. Can. J. Bot. 58, 2241-2262.

Doran, J.B., Gensel, P.G., Andrews, H.N., 1978. New occurrences of trimerophytes from the Devonian of eastern Canada. Can. J. Bot. 56, 3052-3068.

Dorobek, S., 1989. Migration of orogenic fluids through the Siluro-Devonian Helderberg Group during late Paleozoic deformation: constraints on fluid sources and implications for thermal histories of sedimentary basins. Tectonophysics 159, 25-45.

Driese, S.G., Mora, C.I., Elick, J.M., 1997. Morphology and taphonomy of root and stump casts of the earliest trees (Middle to Late Devonian), Pennsylvania and New York, USA. Palaios 12, 524-537.

Edwards, D., 2003. Xylem in early tracheophytes. Plant Cell and Environment 26, 57-72.

Edwards, D., Richardson, J.B., 2004. Silurian and Lower Devonian plant assemblages from the Anglo-Welsh Basin: A palaeobotanical and palynological synthesis. Geol. J. 39(3-4), 375-402.

Elick, J.M., Driese, S.G., Mora, C.I., 1998. Very large plant and root traces from the Early to Middle Devonian: Implications for early terrestrial ecosystems and atmospheric pCO_2. Geology 26(2), 143-146.

Farquhar, G.D., O'Leary, M.H., Berry, J.A., 1982. On the relationship between carbon isotope discrimination and the intercellular carbon dioxide concentration in leaves. Austral. J. Plant Phys. 9, 121-137.

Farquhar, G.D., Ehleringer, J.R., Hubick, K.T., 1989. Carbon isotope discrimination and photosynthesis. Ann. Rev. Plant Phys. Plant Mol. Biol. 40, 503-537.

Faure, G., Mensing, T.M., 2005. Isotopes: Principles and Applications, 3rd ed. John Wiley & Sons, Hoboken, New Jersey, 897 pp.

Feigenwinter, C., Bernhofer, C., Vogt, R., 2004. The influence of advection on the short term CO_2-budget in and above a forest canopy. Boundary-Layer Meteorology 113(2), 201-224.

Feng, J., Xu, H., Jiang, Q., 2014. Study on the stable carbon isotope of the Mid-Late Devonian plants from north Xinjiang. Bot. Res. 3, 227-237 (in Chinese with English abstract).

Fernandez, N., Messuti, M.I., Fontenla, S., 2008. Arbuscular mycorrhizas and dark septate fungi in *Lycopodium paniculatum* (Lycopodiaceae) and *Equisetum bogotense* (Equisetaceae) in a valvidian temperate forest of Patagonia, Argentina. Am. Fern J. 98(3), 117-127.

Flanagan, L.B., Brooks, J.R., Varney, G.T., Berry, S.C., Ehleringer, J.R., 1996. Carbon isotope discrimination during photosynthesis and the isotope ratio of respired CO_2 in boreal forest ecosystems. Global Biogeochemical Cycles 10(4), 629-640.

Fletcher, B.J., Beerling, D.J., Chaloner, W., 2004. Stable carbon isotopes and the metabolism of the terrestrial Devonian organism *Spongiophyton*. Geobiology 2, 107-119.

Friedman, G.M., Sanders, J.E., 1982. Time-temperature-burial significance of Devonian anthracite implies former great (~6.5 km) depth of burial of Catskill Mountains, New York. Geology 10, 93-96.

Gastaldo, R.A., 2016. New paleontological insights into the Emsian–Eifelian Trout Valley Formation, Baxter State Park's Scientific Forest Management Area, Aroostock County, Maine. Palaios 31(7), 339-346.

Gensel, P.G., 1992. Phylogenetic relationships of the zosterophylls and lycopsids: evidence from morphology, paleoecology, and cladistics methods of inference. Ann. Missouri Bot. Gard. 79(3), 450-473.

Gensel, P.G., 2017. Silurian origins of ferns and lycophytes - what we know, what we need to find out. Fern Gazette 20(6), 217-242.

Gensel, P.G., Albright, V.M., 2006. *Leclercqia complexa* from the Early Devonian (Emsian) of northern New Brunswick, Canada. Rev. Palaeobot. Palynol. 142, 103-121.

Gensel, P.G., Andrews, H.N., 1984. Plant Life in the Devonian. Praeger, New York, 380 pp.

Gensel, P.G., Berry, C.M., 2001. Early lycophyte evolution. Am. Fern J. 91(3), 74-98.

Gensel, P.G., Berry, C.M., 2016. Sporangial morphology of the Early Devonian zosterophyll *Sawdonia ornata* from the type locality (Gaspé). Internat. J. Plant Sci. 177(7), 618-632.

Gensel, P.G., Edwards, D. (Eds.), 2001. Plants Invade the Land: Evolutionary and Environmental Perspectives. Columbia Univ. Press, 304 pp.

Gensel, P.G., Kasper, A.E., Jr., 2005. A new species of the Devonian lycopod genus, *Leclercqia*, from Emsian of New Brunswick, Canada. Rev. Palaeobot. Palynol. 137, 105-123.

Gensel, P.G., Pigg, K.B., 2010. An arborescent lycopsid from the Lower Carboniferous Price Formation, southwestern Virginia, USA and the problem of species delimitation. Intern. J. Coal Geol. 83(2-3), 132-145.

Gensel, P.G., Andrews, H.N., Forbes, W.H., 1975. A new species of *Sawdonia* with notes on the origin of microphylls and lateral sporangia. Bot. Gaz. 136(1), 50-62.

Gensel, P.G., Kotyk, M., Basinger, J.M., 2001. Morphology of above- and below-ground structures in Early Devonian plants. In: Gensel, P.G., Edwards, D. (Eds.), Plants Invade the Land: Evolutionary and Environmental Perspectives. Columbia Univ. Press, New York, pp. 83-102.

Gerrienne, P., 1995. Les fossils végétaux du Dévonien inférieru de Marchin (bord nor du Synclinorium de Dinant, Belgique). III. *Psilophyton parvulum* nov. sp. Geobios 28(2), 131-144.

Gerrienne, P., 1997. The fossil plants from the Lower Devonian of Marchin (northern margin of Dinant Synclinorium, Belgium). V. *Psilophyton genseliae* sp. Nov., with hypotheses on the origin of Trimerophytina. Rev. Palaeobot. Palynol. 98, 303-324.

Gerrienne, P., Meyer-Berthaud, B., Yang, N., Steemans, P., Li, C.S., 2014. *Planatophyton* gen. nov., a Middle Devonian euphyllophyte from Xinjiang, north-west China. Rev. Palaeobot. Palynol. 208, 55–64.

Gibling, M.R., Davies, N.S., 2012. Palaeozoic landscapes shaped by plant evolution. Nature Geoscience 5(2), 99-105.

Gibling, M.R., Davies, N.S., Falcon-Lang, H.J., Bashforth, A.R., DiMichele, W.A., Rygel, M.C., Ielpi, A., 2014. Palaeozoic co-evolution of rivers and vegetation: a synthesis of current knowledge. Proc. Geol. Assoc. 125(5-6), 524-533.

Gradstein, F.M., Ogg, J.G., Schmitz, M., Ogg, G. (Eds.), 2012. The Geologic Time Scale 2012. Elsevier, Amsterdam, 2 vols.

Graham, L.E., Gray, J., 2001. The origin, morphology, and ecophysiology of early embryophytes: neontological and paleontological perspectives. In: Gensel, P.G., Edwards, D. (Eds.), Plants Invade the Land: Evolutionary and Environmental Perspectives, Columbia Univ. Press, New York, pp. 140-159.

Graham, L.E., Cook, M.E., Hanson, D.T., Pigg, K.B., Graham, J.M., 2010. Structural, physiological, and stable carbon isotopic evidence that the enigmatic Paleozoic fossil *Prototaxites* formed from rolled liverwort mats. Am. J. Bot. 97(2), 268-275.

Granoff, J.A., Gensel, P.G., Andrews, H.N., 1976. A new species of *Pertica* from the Devonian of Canada. Palaeontographica (B) 155, 119-128.

Greb, S.F., DiMichele, M.A., Gastaldo, R.A., 2006. Evolution and importance of wetlands in earth history. In: Greb, S.F., DiMichele, M.A. (Eds.), Wetlands Through Time, Geol. Soc. Am. Spec. Publ. 399, pp. 1-40.

Grierson, J.D., Banks, H.P., 1983. A new genus of lycopods from the Devonian of New York State. Bot. J. Linn. Soc. 86, 81-101.

Grierson, J.D., Bonamo, P.M., 1979. *Leclercqia complexa*: Earliest ligulate lycopod (Middle Devonian). Am. J. Bot. 66, 474-476.

Griffing, D.H., Bridge, J.S., Hotton, C.L., 2000. Coastal-fluvial palaeoenvironments and plant palaeoecology of the Lower Devonian (Emsian), Gaspé Bay, Quebec, Canada. In: Friend, P.F., Williams, B.P.J. (Eds.), New Perspectives on the Old Red Sandstone. Geol. Soc. London Spec. Publ. 180, pp. 61-84.

Guo, J., Tan, H., Ding, S., Wang, S., 2010. Study on continuous carbonization process of biomass. www.igu.org/html/wgc2009/papers/docs/wgcFinal00123.pdf.

Gupta, N.S., Briggs, D.E., Collinson, M.E., Evershed, R.P., Michels, R., Pancost, R.D., 2007. Molecular preservation of plant and insect cuticles from the Oligocene Enspel Formation, Germany: evidence against derivation of aliphatic polymer from sediment. Org. Geochem. 38(3), 404-418.

Hammond, S.E., Berry, C.M., 2005. A new species of *Tetraxylopteris* (Aneurophytales) from the Devonian of Venezuela. Bot. J. Linn. Soc. 148, 275-303.

Hao, S.G., Xue, J.Z., 2013a. Earliest record of megaphylls and leafy structures, and their initial diversification. Chinese Science Bulletin 58(23), 2784-2793.

Hao, S.G., Xue, J.Z., 2013b. The Early Devonian Posongchong Flora of Yunnan. Science Press, Beijing, 366 pp.

Heroux, Y., Chagnon, A., Bertrand, R., 1979. Compilation and correlation of major thermal maturation indicators. Am. Assoc. Petrol. Geol. Bull. 63(12), 2128-2144.

Hilton, J., Bateman, R.M., 2006. Pteridosperms are the backbone of seed-plant phylogeny. J. Torrey Bot. Soc. 133(1), 119-168.

Hotton, C.L., Hueber, F.M., Griffing, D.H., Bridge, J.S., 2001. Early terrestrial plant environments: An example from the Emsian of Gaspé, Canada. In: Gensel, P.G., Edwards, D. (Eds.), Plants

Invade the Land: Evolutionary and Environmental Perspectives, Columbia Univ. Press, New York, pp. 179-212.

Hueber, F.M., 1971. *Sawdonia ornata*: A new name for *Psilophyton princeps* var. *ornatum*. Taxon 20, 641-642.

Hueber, F.M., Grierson, J.D., 1961. On the occurrence of *Psilophyton princeps* in the early Upper Devonian of New York. Am. J. Bot. 48, 473-479.

Jackson, P.C., Meinzer, F.C., Goldstein, G., Holbrook, N.M., Cavelier, J., Rada, F., 1993. Environmental and physiological influences on carbon isotope composition of gap and understory plants in a lowland tropical forest. In: Ehleringer, J.R., Hall, A.E., Farquhar, G.D. (Eds.), Stable Isotopes and Plant Carbon-Water Relations, Academic Press, pp. 131-140.

Jennings, J., 1976. The morphology and relationships of *Rhodea*, *Telangium*, *Telangiopsis*, and *Heterangium*. Am. J. Bot. 63(8), 1119-1133.

Kasper, A.E., Jr., Andrews, H.N., Jr., 1972. *Pertica*, a new genus of Devonian plants from northern Maine. Am. J. Bot. 59(9), 897-911.

Kennedy, K., Gensel, P.G., Gibling, M.R., 2012. Paleoenvironmental inferences from the classic Early Devonian plant-bearing locality of the Campbellton Formation, New Brunswick, Canada. Palaios 27, 424-438.

Kennedy, K.L., Gibling, M.R., Eble, C.F., Gastaldo, R.A., Gensel, P.G., Werner-Zwanziger, U., Wilson, R.A., 2013. Lower Devonian coaly shales of northern New Brunswick, Canada: plant accumulations in the early stages of terrestrial colonization. J. Sed. Res. 83(12), 1202-1215.

Kenrick, P., Crane, P.R., 1997. The origin and early evolution of plants on land. Nature 389, 33-39.

Knaus, M.J., 1994. *Triphyllopteris collombiana*: a clarification of the generic concepts based on rediscovered specimens from Kossberg bei Plauen, Germany, and a reassignment of the North American species of *Triphyllopteris* to *Genselia* gen. nov. Int. J. Plant Sci. 155(1), 97-116.

Knaus, M.J., 1995. The species of the Early Carboniferous fossil plant genus *Genselia*. Int. J. Plant Sci. 156(1), 61-92.

Kohn, M.J., 2010. Carbon isotope compositions of terrestrial C3 plants as indicators of (paleo)ecology and (paleo)climate. Proc. Nat. Acad. Sci. USA 107(46), 19691-19695.

Lawrence, D., 1986. Sedimentology of the Lower Devonian Battery Point Formation, eastern Gaspé Peninsula. Ph.D. thesis, University of Bristol, Bristol, U.K.

Leclercq, S., Banks, H.P., 1962. *Pseudosporochnus nodosus* sp. nov., a Middle Devonian plant with cladoxylalean affinities. Palaeontographica (B) 110, 1-34.

Le Hir, G., Donnadieu, Y., Goddéris, Y., Meyer-Berthaud, B., Ramstein, G., Blakey, R.C., 2011. The climate change caused by the land plant invasion in the Devonian. Earth Planet. Sci. Lett. 310, 203-212.

Li, C.S., Edwards, D., 1995. A re-investigation of Halle's *Drepanophycus spinaeformis* Göpp from the Lower Devonian of Yunnan Province, southern China. Bot. J. Linn. Soc. 118, 163-192.

Li, C.S., Hueber F.M., Hutton C.L., 2000. A neotype for *Drepanophycus spinaeformis* Goppert 1852. Can. J. Bot. 78, 889-902.

Liu, W., Feng, X., Ning, Y., Zhang, Q., Cao, Y., An, Z., 2005. δ^{13}C variation of C3 and C4 plants across an Asian monsoon rainfall gradient in arid northwestern China. Glob. Change Biol. 11, 1094-1100.

Macfarlane, C., Adams, M.A., White, D.A., 2004. Productivity, carbon isotope discrimination and leaf traits of trees of *Eucalyptus globulus* Labill in relation to water availability. Plant Cell Environ. 27, 1515-1524.

Malamud-Roam, F., Ingram, B.L., 2001. Carbon isotopic compositions of plants and sediments of tide marshes in the San Francisco estuary. J. Coast. Res. 17(1), 17-29.

Marino, B.D., McElroy, M.B., 1991. Isotopic composition of atmospheric CO_2 inferred from in C_4 plant cellulose. Nature, 127-131.

Marshall, J.E.A., 1996. *Rhabdosporites langii*, *Geminospora lemurata* and *Contagisporites optivus*: An origin for heterospory within the progymnosperms. Rev. Palaeobot. Palynol. 93, 159-189.

Meyer-Berthaud, B., Scheckler, S.E., Wendt, J., 1999. *Archaeopteris* is the earliest known modern tree. Nature 398, 700-704.

Meyer-Berthaud, B., Fairon-Demaret, M., Steemans, P., Talent, J., Gerrienne, P., 2003. The plant *Leclercqia* (Lycopsida) in Gondwana: implications for reconstructing Middle Devonian palaeogeography. Geol. Mag. 140, 119-130.

Meyer-Berthaud, B., Soria, A., Decombeix, A.L., 2010. The land plant cover in the Devonian: A reassessment of the evolution of the tree habit. In: Vecoli, M., Clément, G., Meyer-Berthaud, B. (eds.), The Terrestrialization Process: Modelling Complex Interactions at the Biosphere-Geosphere Interface, Geological Society of London, Spec. Publ. 339, pp. 59-70.

Milici, R.C., Swezey, C.S., 2006. Assessment of Appalachian basin oil and gas resources; Devonian Shale-Middle and Upper Paleozoic total petroleum system: U.S. Geological Survey Open-File Report 2006-1237, 70 pp. (http://pubs.usgs.gov/of/2006/1237/).

Miller, J.M., Williams, R.J., Farquhar, G.D., 2001. Carbon isotope discrimination by a sequence of Eucalyptus species along a subcontinental rainfall gradient in Australia. Funct. Ecol. 15, 222-232.

Mintz, J.S., Driese, S.G., White, J.D., 2010. Environmental and ecological variability of Middle Devonian (Givetian) forests in Appalachian Basin paleosols, New York, United States. Palaios 25, 85-96.

Mora, C.I., Driese, S.G., Colarusso, L.A., 1996. Middle to late Paleozoic atmospheric CO_2 levels from soil carbonate and organic matter. Science 271(5252), 1105-1107.

Nilsen, E., Sharifi, M., 1997. Carbon isotopic composition of legumes with photosynthetic stems from mediterranean and desert habitats. Am. J. Bot. 84(12), 1707-1707.

Obermajer, M., Fowler, M.G., Goodarzi, F., Snowdon, L.R., 1997. Organic petrology and organic geochemistry of Devonian black shales in southwestern Ontario, Canada. Org. Geochem. 26, 229-246.

Obukhovskaya, T., 2000. Miospores of the Givetian-Frasnian boundary deposits in Belarus. Acta Palaeobotanica 40(1), 17-23.

O'Leary, M.H., 1988. Carbon isotopes in photosynthesis. BioScience 38(5), 328-336.

Pate, J., Arthur, D., 1998. δ^{13}C analysis of phloem sap carbon: Novel means of evaluating seasonal water stress and interpreting carbon isotope signatures of foliage and trunk wood of *Eucalyptus globulus*. Oecologia 117, 301-311.

Playford, G., Dino, R., 2005. Carboniferous and Permian palynostratigraphy. In: Koutsoukos, E.A.M. (Ed.), Applied Stratigraphy. Springer, Amsterdam, pp. 101-121.

Poss, J.A., Grattan, S.R., Suarez, D.L., Grieve, C.M., 2000. Stable carbon isotope discrimination: an indicator of cumulative salinity and boron stress in *Eucalyptus camaldulensis*. Tree Phys. 20, 1121-1127.

Pratt, B.R., van Heerde, J., 2017. An arborescent lycopsid stem fragment from the Palliser Formation (Famennian) carbonate platform, southwestern Alberta, Canada, and its paleogeographic and paleoclimatic significance. Can. J. Earth Sci. 54(2), 141-145.

Prestianni, C., Meyer-Berthaud, B., Blanchard, R., Rücklin, M., Clément, G., Gerrienne, P., 2012. The Middle Devonian plant assemblage from Dechra Aït Abdallah (Central Morocco) revisited. Rev. Palaeobot. Palynol. 179, 44-55.

Raich, J.W., Tufekciogul, A., 2000. Vegetation and soil respiration: correlations and controls. Biogeochemistry 48(1), 71-90.

Raven, P.H., Evert, R.F., Eichhorn, S.E. (Eds.), 2004. Biology of Plants, 7th ed. W.H. Freeman, New York, 686 pp.

Raymond, A., Metz, C., 1995. Laurussian land-plant diversity during the Silurian and Devonian: mass extinction, sampling bias, or both? Paleobiology 21, 74-91.

Rayner, R.J., 1983. New observations on *Sawdonia ornata* from Scotland. Trans. Roy. Soc. Edinburgh (Earth Sciences) 74, 79-93.

Read, C.B., 1955. Floras of the Pocono Formation and Price Sandstone in parts of Pennsylvania, Maryland, West Virginia, and Virginia. U.S. Geol. Surv. Prof. Paper 263, 30 pp.

Repetski, J.E., Ryder, R.T., Weary, D.J., Harris A.G., Trippi, M.H., 2008. Thermal maturity patterns (CAI and %R_0) in Upper Ordovician and Devonian rocks of the Appalachian Basin:

a major revision of USGS Map I-917-E using new subsurface collections. U.S. Geol. Survey, Sci. Inv. Map 3006.

Retallack, G.J., Huang, C., 2011. Ecology and evolution of Devonian trees in New York, USA. Palaeogeogr. Palaeoclimatol. Palaeoecol. 299, 110-128.

Richardson, J.B., McGregor, D.C., 1986. Silurian and Devonian spore zones of the Old Red Sandstone continent and adjacent regions. Geol. Surv. Can. Bull. 384, 1-79.

Roden, J.S., Bowling, D.R., McDowell, N.G., Bond, B.J., Ehleringer, J.R., 2005. Carbon and oxygen isotope ratios of tree ring cellulose along a precipitation transect in Oregon, United States. J. Geophys. Res. 110, G02003, 11 pp., doi: 10.1029/2005JG000033.

Rowan, E.L., 2006. Burial and thermal history of the Central Appalachian Basin, based on three 2-D models of Ohio, Pennsylvania, and West Virginia. U.S. Geol. Surv., Open-File Report 2006-1019.

Rygel, M.C., Calder, J.H., Gibling, M.R., Gingras, M.K., Melrose, C.S.A., 2006. Tournaisian forested wetlands in the Horton Group of Atlantic Canada. In: Greb, S.F., DiMichele, M.A. (Eds.), Wetlands Through Time. Geol. Soc. Am. Spec. Publ. 399, pp. 103-126.

Scheckler, S.E., 1978. Ontogeny of progymnosperms II. Shoots of Upper Devonian Archaeopteridales. Can. J. Bot. 56, 3136-3170.

Scheckler, S.E., 1986a. Geology, floristics and paleoecology of Late Devonian coal swamps from Appalachian Laurentia (U.S.A.). Ann. Soc. Géol. Belg. 109, 209-222.

Scheckler, S.E., 1986b. Floras of the Devonian–Mississippian transition. In: Gastaldo, R.A. (Org.) & Broadhead, T.W. (Ed.), Land Plants: Notes for a Short Course. Univ. Tenn. Dept. Geol. Sci., Knoxville, Tenn., pp. 81-96.

Scheckler, S.E., 2001. Afforestation—The first forests. In: Briggs, D.E.G., Crowther, P. (Eds.), Palaeobiology II. Blackwell, Oxford, pp. 67–71.

Scheckler, S.E., Banks, H.P., 1971. Anatomy and relationships of some Devonian progymnosperms from New York. Am. J. Bot. 58(8), 737-751.

Schimmelmann, A., Qi, H., Coplen, T.B., Brand, W.A., Fong, J., Meier-Augenstein, W., Kemp, H.F., Toman, B., Ackermann, A., Assonov, S., Aerts-Bijma, A.T., 2016. Organic reference materials for hydrogen, carbon, and nitrogen stable isotope-ratio measurements: caffeines, n-alkanes, fatty acid methyl esters, glycines, L-valines, polyethylenes, and oils. Anal. Chem. 88(8), 4294-4302.

Schleser, G.H., Jayasekera, R., 1985. δ^{13}C-variations of leaves in forests as an indication of reassimilated CO_2 from the soil. Oecologia 65(4), 536-542.

Silvestro, D., Cascales-Miñana, B., Bacon, C.D., Antonelli, A., 2015. Revisiting the origin and diversification of vascular plants through a comprehensive Bayesian analysis of the fossil record. New Phytol. 207(2), 425-436.

Skog, J.E., Gensel, P.G., 1980. A fertile species of *Triphyllopteris* from the Early Carboniferous (Mississippian) of Southwestern Virginia. Am. J. Bot. 67(4), 440-451.

Sperry, J.S., 2003. Evolution of water transport and xylem structure. Internat. J. Plant Sci. 164(S3), S115-S127.

Spicer, R.A., 1989. Physiological characteristics of land plants in relation to environment through time. Trans. Roy. Soc. Edinburgh (Earth Sci.) 80, 321-9.

Stein, W., 1993. Modeling the evolution of stelar architecture in vascular plants. Int. J. Plant Sci. 154(2), 229-263.

Stein, W.E., Mannolini, F., Hernick, L.V., Landing, E., Berry, C.M., 2007. Giant cladoxylopsid trees resolve the enigma of the Earth's earliest forest stumps at Gilboa. Nature 446(7138), 904-907.

Stein, W.E., Berry, C.M., VanAller Hernick, L., Mannolini, F., 2012. Surprisingly complex community discovered in the mid-Devonian fossil forest at Gilboa. Nature 483, 78-81.

Stewart, W.N., Rothwell, G.W., 1993. Paleobotany and the Evolution of Plants, 2nd ed. Cambridge Univ. Press, Cambridge, 521 pp.

Streel, M., Caputo, M.V., Loboziak, S., Melo, J.H.G., 2000. Late Frasnian-Famennian climates based on palynomorphs analyses and the question of the Late Devonian glaciations. Earth-Sci. Rev. 52, 121-173.

Stubblefield, S., Banks H.P., 1978. The cuticle of *Drephanophycus spinaeformis*, a long-ranging Devonian Lycopod from New York and Eastern Canada. Am. J. Bot. 65(1), 110-118.

Taylor, T.N., Taylor, E.L., Krings, M. (Eds.), 2009. Paleobotany: The Biology and Evolution of Fossil Plants, 2nd ed. Academic Press, London, 1230 pp.

Trant, C.A., Gensel, P.G., 1985. Branching in *Psilophyton*: a new species from the Lower Devonian of New Brunswick, Canada. Am. J. Bot. 72(8), 1256-1273.

Turnau, E., 2014. Floral change during the Taghanic Crisis: spore data from the Middle Devonian of northern and south-eastern Poland. Rev. Palaeobot. Palynol. 200, 108-121.

Utting, J., Hamblin, A.P., 1991. Thermal maturity of the Lower Carboniferous Horton Group, Nova Scotia. Int. J. Coal Geol. 19, 439-456.

van der Voo, R., 1988. Paleozoic paleogeography of North America, Gondwana, and intervening displaced terranes: Comparisons of paleomagnetism with paleoclimatology and biogeographical patterns. Geol. Soc. Am. Bull. 100, 311-324.

van der Voo, R., 1993. Paleomagnetism of the Atlantic, Tethys, and Iapetus Oceans. Cambridge Univ. Press, Cambridge, 411 pp.

Wellman, C.H., Osterloff, P.L., Mohiuddin, U., 2003. Fragments of the earliest land plants. Nature 425(6955), 282-285.

Wilson, J.P., 2013. Modeling 400 million years of plant hydraulics. Paleontol. Soc. Pap. 19, 175–194.

Wilson, J.P., 2016. Hydraulics of *Psilophyton* and evolutionary trends in plant water transport after terrestrialization. Rev. Palaeobot. Palynol. 227, 65-76.

Wilson, J.P., Fischer, W.W., 2011. Hydraulics of *Asteroxylon mackiei*, an early Devonian vascular plant, and the early evolution of water transport tissue in terrestrial plants. Geobiology 9, 121–130.

Wilson, J.P., Knoll, A.H., 2010. A physiologically explicit morphospace for tracheid-based water transport in modern and extinct seed plants. Paleobiology 36, 335–355.

Winter, K., Holtum, J.A.M., 2005. The effects of salinity, crassulacean acid metabolism and plant age on the carbon isotope composition of *Mesembryanthemum crystallinum* L., a halophytic C3-CAM species. Planta 222, 201-209.

Xu, H.H., Berry, C.M., 2008. The Middle Devonian lycopsid *Haskinsia* Grierson et Banks from the Ruppert Coast, Marie Byrd Land, West Antarctica. Rev. Palaeobot. Palynol. 150, 1-4.

Xu, H.H., Wang, Y., 2008. The palaeogeographical significance of specimens attributed to *Protolepidodendron scharyanum* Krejči (Lycopsida) from the Middle Devonian of North Xinjiang, China. Geological Magazine 145(2), 295-299.

Xu, H.H., Wang, Y., Berry, C.M., Cai, C.Y., 2008. Two species of *Haskinsia* Grierson & Banks (Lycopsida) from the Middle Devonian of Xinjiang, China, and consideration of their palaeogeographical significance. Bot. J. Linn. Soc. 157, 633-644.

Xu, H.H., Berry, C.M., Wang, Y., Marshall, J.E.A., 2011. A new species of *Leclercqia* Banks, Bonamo et Grierson (Lycopsida) from the Middle Devonian of North Xinjiang, China, with a possible climbing habit. Int. J. Plant Sci. 172, 836-846.

Xu, H.H., Feng, J., Jiang, Q., Wang, Y., 2013. Report of *Drepanophycus* Göppert (Lycopsida) from the Middle Devonian of Xinjiang, China. J. Syst. Evol. 51(6), 765-772.

Xu, H.H., Jiang, Q., Zhang, X.L., Wang, Y., Feng, J., 2015. On the Mid Devonian Hujiersite flora from West Junggar, Xinjiang, China, its characteristics, age, palaeoenvironment and palaeophytogeographical significances. Acta Palaeontol. Sin. 54, 230-239.

Xu, H.H., Berry, C.M., Stein, W., Wang, Y., Tang, P., Fu, Q., 2017. Unique growth strategy in the Earth’s first trees revealed in silicified fossil trunks from China. Proc. Nat. Acad. Sci. (U.S.A.) 114, 12009-12014.

Xue, J., 2012. Lochkovian plants from the Xitun Formation of Yunnan, China, and their palaeophytogeographical significance. Geol. Mag. 149(2), 333-344.

Xue, J., Deng, Z., Huang, P., Huang, K., Benton, M.J., Cui, Y., Wang, D., Liu, J., Shen, B., Basinger, J.F., Hao, S., 2016. Belowground rhizomes in paleosols: The hidden half of an Early Devonian vascular plant. Proc. Nat. Acad. Sci. (U.S.A.) 113(34), 9451-9456.

Xue, J., Huang, P., Wang, D., Xiong, C., Liu, L., Basinger, J.F., 2018. Silurian-Devonian terrestrial revolution in South China: Taxonomy, diversity, and character evolution of vascular plants in a palaeogeographically isolated, low-latitude region. Earth-Sci. Rev. 180, 92-125.

Yang, N., Edwards, D., Ablaev, A., Li, C.S., 2008. The discovery of fertile *Haskinsia colophylla* (Haskinsiaceae) in Far East Russia. Taxon 57(2), 588-593.

Zaiontz, C., 2014. Real statistics using Excel. Online. Tersedia di: http://www. realstatistics. com/tests-normality-and-symmetry/statistical-tests-normalitysymmetry/kolmogorov-smirnov-test/[diakses pada 03 Juni 2014] Lampiran, 1.

Zheng, D., Xu, H., Wang, J., Feng, C., Zhang, H., Chang, S.C., 2016. Geochronologic age constraints on the Middle Devonian Hujiersite flora of Xinjiang, NW China. Palaeogeogr. Palaeoclimatol. Palaeoecol. 463, 230-237.

Ziegler, W., Sandberg, C.A., 1990. The Late Devonian Standard Conodont Zonation. Cour. Forschungsinst. Senckenberg 121, 1-115.

Tables

Table 1. Summary of carbon isotope analyses by taxon and plant part

Table 2. Mean differences in $\Delta^{13}C_p$ by taxon

Table 3. Differences in $\delta^{13}C_P$ between anatomical parts of individual specimens

Table S1. Stable carbon isotope compositions of Devonian-Carboniferous fossil plant specimens

Table 1. Summary of carbon isotope analyses by taxon and plant part

	Number of specimens	Number of carbon isotope analyses					Mean $\delta^{13}C$				
		Stem	Leaf	Spine	Indet.	Total	Stem	Leaf	Spine	Indet.	Avg.
Archaeopteris	51	36	17		27	80	-25.53	-25.32		-27.16	-26.04
Drepanophycus	21	23		7	10	40	-26.08		-25.52	-25.90	-25.94
Genselia	9	2	1		7	10	-22.66	-23.08		-23.72	-23.44
Haskinsia	4	2			2	4	-22.27			-22.31	-22.29
Leclercqia	13	16		3	5	24	-22.96		-23.91	-22.71	-23.02
Pertica	11	12			8	20	-25.55			-25.26	-25.43
Wattieza	8	6			5	11	-26.21			-26.74	-26.45
Psilophyton	17	13			12	25	-24.46			-25.27	-24.85
Rhacophyton	18	19	4		10	33	-24.51	-25.28		-25.82	-25.00
Rhodeopteridium	3	2	1			3	-23.71	-22.82			-23.41
Sawdonia	14	18		4	6	28	-27.46		-27.08	-25.85	-27.07
Tetraxylopteris	21	19	2		10	31	-25.56	-26.09		-27.01	-26.06
Total / Average	190	168	25	14	102	309	-25.30	-25.18	-25.62	-25.87	-25.50

Notes: Indet. = indeterminate. Avg. = average (of stem + leaf + spine + indet. mean values).

Table 2. Mean differences in $\Delta^{13}C_p$ by taxon

	Number of analyses	mean (‰)	st. dev. (‰)	s.e. (‰)	Student's t	p(α)
Archaeopteris	80	**-0.38**	**1.18**	**0.13**	**3.05**	**0.002**
Drepanophycus	40	-0.26	1.08	0.17	1.32	0.189
Genselia	10	0.37	1.12	0.36	0.69	0.492
Haskinsia	4	**2.41**	**1.48**	**0.74**	**3.31**	**0.001**
Leclerqcia	24	**1.81**	**1.02**	**0.21**	**6.57**	**0.001**
Pertica	20	0.40	0.56	0.12	1.06	0.292
Wattieza	11	-0.51	0.49	0.15	1.28	0.201
Psilophyton	25	**0.94**	**1.55**	**0.31**	**3.22**	**0.001**
Rhacophyton	33	0.23	1.32	0.23	0.65	0.517
Rhodeopteridium	3	0.41	0.89	0.51	0.42	0.677
Sawdonia	28	**-1.22**	**1.81**	**0.34**	**5.02**	**0.001**
Tetraxylopteris	31	-0.05	1.35	0.24	0.37	0.714

Notes: bold type denotes significant values (= $p(\alpha) < 0.05$); st. dev. = standard deviation; s.e. = standard error of the mean. Statistical software from Zaiontz (2014).

Table 3. Differences in $\delta^{13}C_P$ between anatomical parts of individual specimens

	No. of pairs	Stem $\delta^{13}C$ (‰)	Leaf/spine $\delta^{13}C$ (‰)	Difference $\Delta^{13}C$ (‰)	s.e. $\delta^{13}C$ (‰)	Student's t	p(α)
Archaeopteris	13	-0.08	0.33	0.42	0.20	1.54	0.137
Drepanophycus	6	-0.81	-0.12	0.69	0.30	1.15	0.277
Leclerqcia	3	1.75	1.96	0.21	0.22	0.45	0.676
Rhacophyton	4	0.51	0.05	-0.46	0.23	1.54	0.173
Sawdonia	4	**-2.77**	**-1.22**	**1.56**	**0.21**	**2.85**	**0.029**
Tetraxylopteris	2	0.10	-0.06	-0.16	0.13	1.25	0.337
Total	32	**-0.30**	**0.15**	**0.45**	**0.14**	**2.64**	**0.010**

Notes: bold type denotes significant values (=$p(\alpha) < 0.05$); s.e. = standard error of the mean.

Figure captions

Figure 1. Devonian plant taxa of the present study: (A) phylogenetic relationships, and (B) morphological reconstructions. Reconstructions show full shoot system of plant except for *Tetraxylopteris* (lateral branch); open scale bars equal 10 cm and hachured scale bars equal 1 m. Sources: (A) Kenrick and Crane (1997), Hilton and Bateman (2006), and Taylor et al. (2009); (B) *Sawdonia* (http://www.ucmp.berkeley.edu/IB181/VPL/Lyco/Lyco1.html, *Drepanophycus* (Stewart and Rothwell, 1993), *Leclercqia* (Bonamo et al., 1988), *Haskinsia* (http://dinoera.com/tags/haskinsia), *Psilophyton* (https://openclipart.org/detail/231687/psilophyton-princeps), *Pertica* (Kasper and Andrews, 1972), *Wattieza* (Stein et al., 2007) [note: reconstructions of European *Pseudosporochnus* are slightly different, e.g., Berry and Fairon-Demaret, 2002], *Rhacophyton* (Cornet et al., 1977), *Rhodeopteridium* (Jennings, 1976), *Tetraxylopteris* (Bonamo and Banks, 1967), *Archaeopteris* (www.devoniantimes.org), and *Genselia* (Knaus, 1995).

Figure 2. Stratigraphic ages of fossil plant locales and ranges of fossil plant taxa used in this study. Marine conodont and terrestrial palynomorph zonations are from Richardson and McGregor (1986), Ziegler and Sandberg (1990), Streel et al. (2000), and Playford and Dino (2005); the timescale is from Gradstein et al. (2012).   Locales: Gaspe Peninsula, Quebec, 1 = Haldimand Head, 2 = Aiguillon, Cap-aux-Os, and L'Anse-a-brilliant, and 3 = Seal Rock; New Brunswick, 1 = south shore Chaleur Bay (localities A, B, F), Dalhousie, Pin Sec Point, and

Peuplier Point, and 2 = Atholville and Restigouche; Hujiersite Fm (Xinjiang, China), 1 = Gannaren, 2 = Hwy 217, 3 = 251 Hill, and 4 = Hujiersite. Specimens of the Oneonta, Sonyea, Escuminac, and Pocono formations (italicized) are from multiple locales, but all fall within the narrow age ranges shown for each unit. For plant taxa, solid bars indicate peak abundance, open bars the full established biorange, and dashed bars uncertain extensions of the biorange.

Figure 3. Geographic distribution of fossil plant locales, shown on Late Devonian paleogeographic maps of (A) the world and (B) the Appalachian Basin. (A) Inset rectangle represents area of map B; abbreviations: A = Alberta, G = Germany, O = Ontario, V = Venezuela, X = Xinjiang, and Y = Yunnan; base map courtesy of Ron Blakey (Deep-Time Maps). Note that Laurentia (Paleozoic North America) was rotated ~30˚ clockwise relative to its present-day orientation (van der Voo, 1988, 1993). (B) Main fossil plant locales in Appalachian Basin; site names are positioned either directly above or below site locales (black dots); county names are given where multiple locales are within a small area. Dark brown field shows approximate area of basinward progradation of shoreline from the Early to the Late Devonian. N = present-day north; N_D = Devonian north.

Figure 4. Representative study specimens: (A) *Psilophyton forbesii* stems and leaves, Emsian, Gaspé Peninsula, Québec; (B) *Archaeopteris* sp. stems and leaves, Frasnian, West Virginia; (C) tracheids of *Psilophyton charientos*, Emsian, New Brunswick; (D) cuticle of *Sawdonia* sp., Emsian, Gaspé Peninsula, Québec. All specimens were inspected using a binocular scope to

verify the state of fossil preservation, and a subset was examined using scanning electron microscopy.

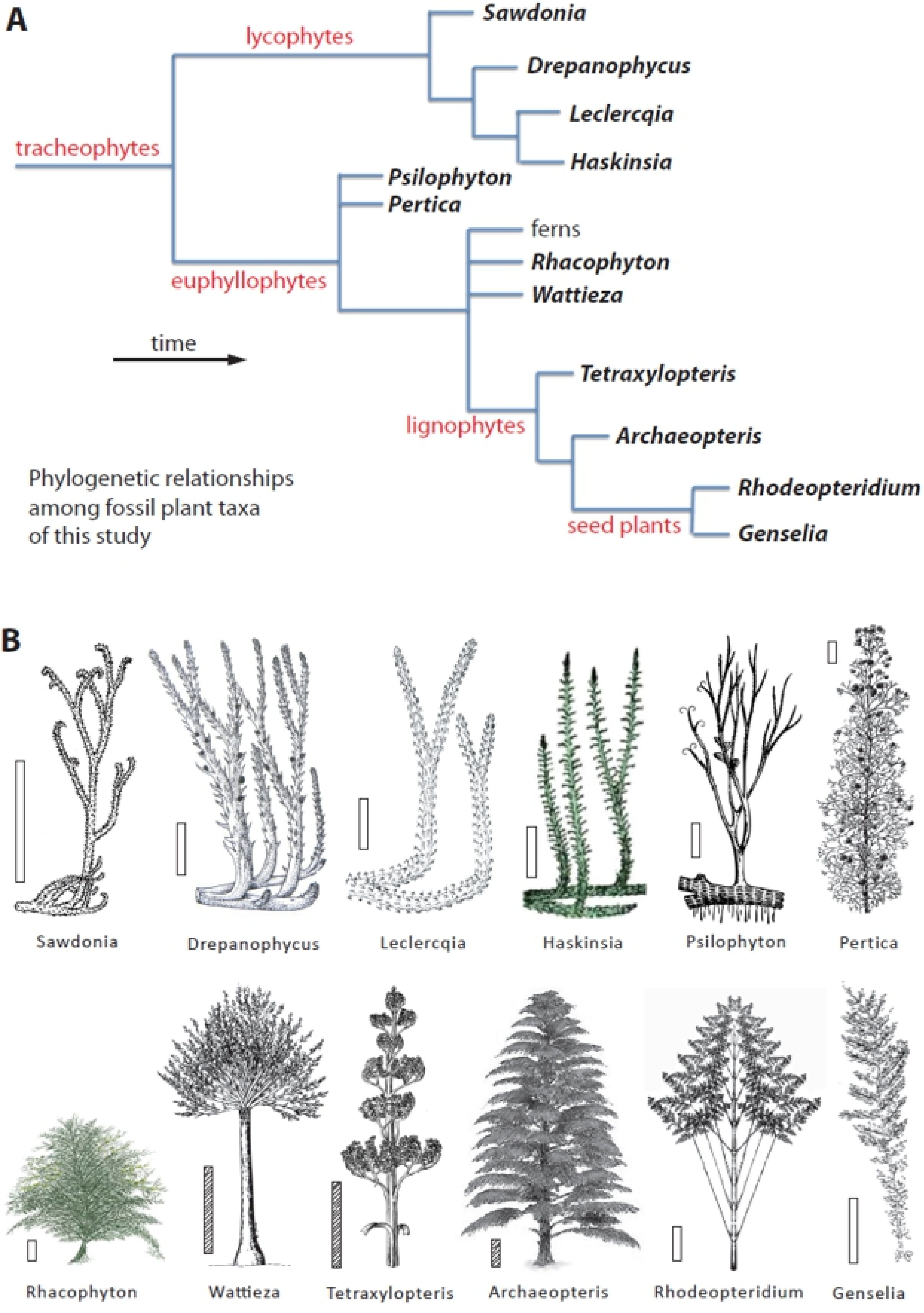
Figure 5. Model of $\Delta\delta^{13}C_{P-X}$ calculations. Relative to the LOWESS curve generated from the full plant $\delta^{13}C$ dataset, some fossil plant taxa exhibit systematically higher or lower $\delta^{13}C$ values. In this example, Taxon A exhibits higher-than-average $\delta^{13}C$ (positive $\Delta\delta^{13}C_{P-X}$), Taxon B exhibits average $\delta^{13}C$ (near-zero $\Delta\delta^{13}C_{P-X}$), and Taxon C exhibits lower-than-average $\delta^{13}C$ (negative $\Delta\delta^{13}C_{P-X}$). The complete LOWESS curve for the $\delta^{13}C_P$ dataset will be published in a companion study (Algeo et al., in preparation).

Figure 6. Deviations of $\delta^{13}C$ of Devonian fossil plant specimens from the LOWESS reference curve ($\Delta\delta^{13}C_{P-X}$) plotted as a function of specimen age. For each genus, the regression of $\Delta\delta^{13}C_{P-X}$ against time is shown as a thin line; these regression lines are shown not to suggest that the $\Delta\delta^{13}C_{P-X}$ of a given taxon changes significantly with time (for most taxa, there is not enough data to generate a statistically significant time-dependent trend) but, rather, to show which taxa yield $\Delta\delta^{13}C_{P-X}$ values that are systematically higher or lower than the age-specific mean $\Delta\delta^{13}C_{P-X}$ values (0 ‰). Note that some taxa are systematically ^{13}C-enriched and others are systematically ^{13}C-depleted.

Figure 7. Deviations of $\delta^{13}C$ of Devonian fossil plants from the LOWESS reference curve ($\Delta\delta^{13}C_{P-X}$) by taxon. Open circles represent individual C-isotope analyses; red diamond, open rectangle, and horizontal bar represent the mean, standard error of the mean, and

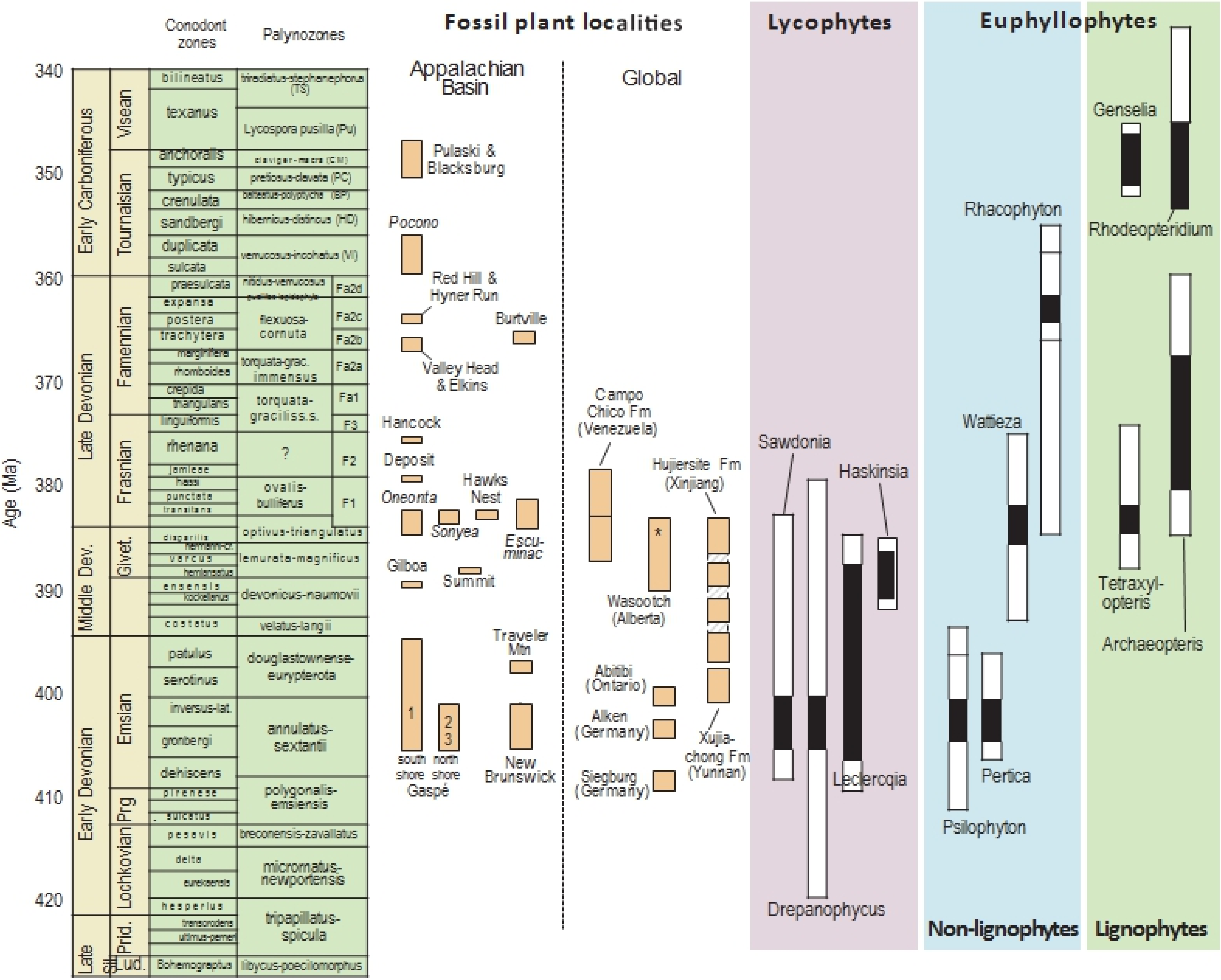
standard deviation range, respectively, for each taxon. The dashed vertical line represents no offset from the LOWESS trend, and the yellow field represents +/-1 standard deviation for the dataset as a whole. Note that some taxa are systematically ^{13}C-enriched and others are systematically ^{13}C-depleted.

Figure 8. Reconstructions of the ecological distribution of the 10 Devonian vascular land plant taxa of the present study: (A) Early Devonian (Emsian), (B) Middle Devonian (Givetian), and (C) Late Devonian (late Frasnian-early Famennian). Note that actual Devonian landscapes included other vascular and non-vascular plant taxa not shown here; these reconstructions are intended to show only the relative habitat preferences of the 10 Devonian taxa of the present study. Note further that the proposed paleo-environmental distributions of these taxa shown here are consistent with, but not proven by, the available isotopic data, and that alternative models are possible. Further study of Devonian fossil plant taxa will be needed to accurately constrain their actual habitat preferences.



Phylogenetic relationships
among fossil plant taxa
of this study

Figure 1


Conodont zones
Palynozones
Fossil plant localities
Appalachian Basin
Global
Lycophytes
Euphyllophytes
Non-lignophytes
Lignophytes
Age (Ma)
340
350
360
370
380
390
400
410
420
Early Carboniferous
Late Devonian
Middle Dev.
Early Devonian
Late Sil.
Visean
Tournaisian
Famennian
Frasnian
Givet.
Emsian
Prg
Lochkovian
Prid.
Lud.
bilineatus
texanus
anchoralis
typicus
crenulata
sandbergi
duplicata
sulcata
praesulcata
expansa
postera
trachytera
marginifera
rhomboidea
crepida
triangularis
linguiformis
rhenana
jamieae
hassi
punctata
transitans
disparilis
hermanni-cr.
varcus
hemiansatus
ensensis
kockelianus
costatus
patulus
serotinus
inversus-lat.
gronbergi
dehiscens
pirenese
sulcatus
pesavis
delta
eurekaensis
hesperius
transcrodera
ultimus-pemer
Bohemograptus
triradiatus-stephanephorus (TS)
Lycospora pusilla (Pu)
claviger-macra (CM)
pretiosus-clavata (PC)
balteatus-polyptycha (BP)
hibernicus-distinctus (HD)
verrucosus-incohatus (VI)
nitidus-verrucosus
pusillites-lepidophyta
flexuosa-cornuta
torquata-grac. immensus
torquata-gracilis s.s.
?
ovalis-bulliferus
optivus-triangulatus
lemurata-magnificus
devonicus-naumovii
velatus-langii
douglastownense-eurypterota
annulatus-sextantii
polygonalis-emsiensis
breconensis-zavallatus
micrornatus-newportensis
tripapillatus-spicula
libycus-poecilomorphus
Fa2d
Fa2c
Fa2b
Fa2a
Fa1
F3
F2
F1
Pulaski & Blacksburg
Pocono
Red Hill & Hyner Run
Valley Head & Elkins
Burtville
Hancock
Deposit
Oneonta
Sonyea
Hawks Nest
Escuminac
Gilboa
Summit
Traveler Mtn
1
2
3
south shore
north shore
Gaspé
New Brunswick
Campo Chico Fm (Venezuela)
Hujiersite Fm (Xinjiang)
*
Wasootch (Alberta)
Abitibi (Ontario)
Alken (Germany)
Siegburg (Germany)
Xujiachong Fm (Yunnan)
Sawdonia
Drepanophycus
Haskinsia
Leclercqia
Rhacophyton
Wattieza
Psilophyton
Pertica
Gensella
Rhodeopteridium
Tetraxylopteris
Archaeopteris

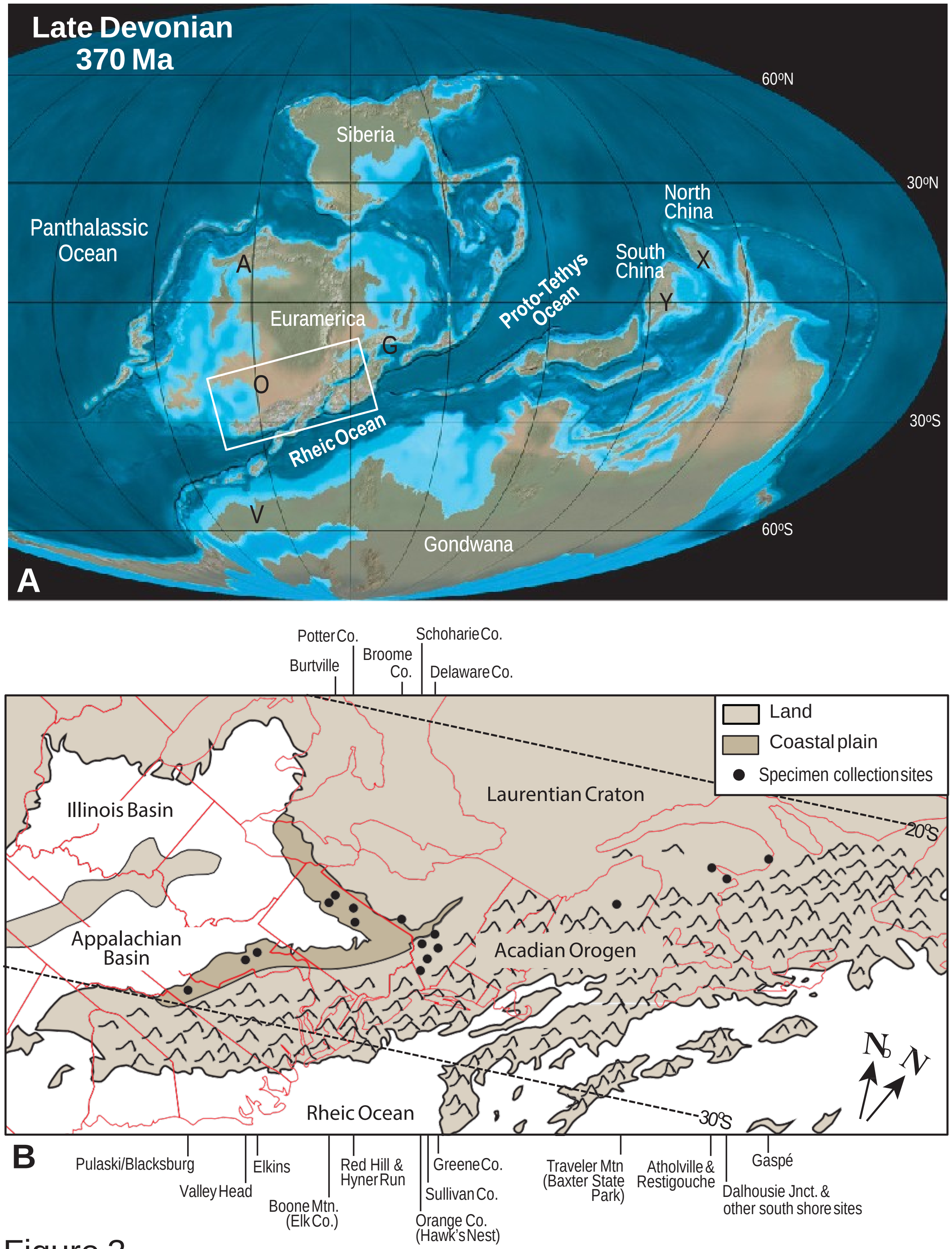

Late Devonian
370 Ma
60ºN
Siberia
30ºN
North China
Panthalassic Ocean
A
South China
X
Proto-Tethys Ocean
Y
Euramerica
G
O
Rheic Ocean
30ºS
V
60ºS
Gondwana
A
Potter Co.
Schoharie Co.
Burtville
Broome Co.
Delaware Co.
Land
Coastal plain
● Specimen collection sites
Illinois Basin
Laurentian Craton
20°S
Appalachian Basin
Acadian Orogen
N
N
Rheic Ocean
30°S
B
Pulaski/Blacksburg
Elkins
Valley Head
Red Hill & Hyner Run
Boone Mtn. (Elk Co.)
Greene Co.
Sullivan Co.
Orange Co. (Hawk's Nest)
Traveler Mtn (Baxter State Park)
Atholville & Restigouche
Gaspé
Dalhousie Jnct. & other south shore sites

Figure 3

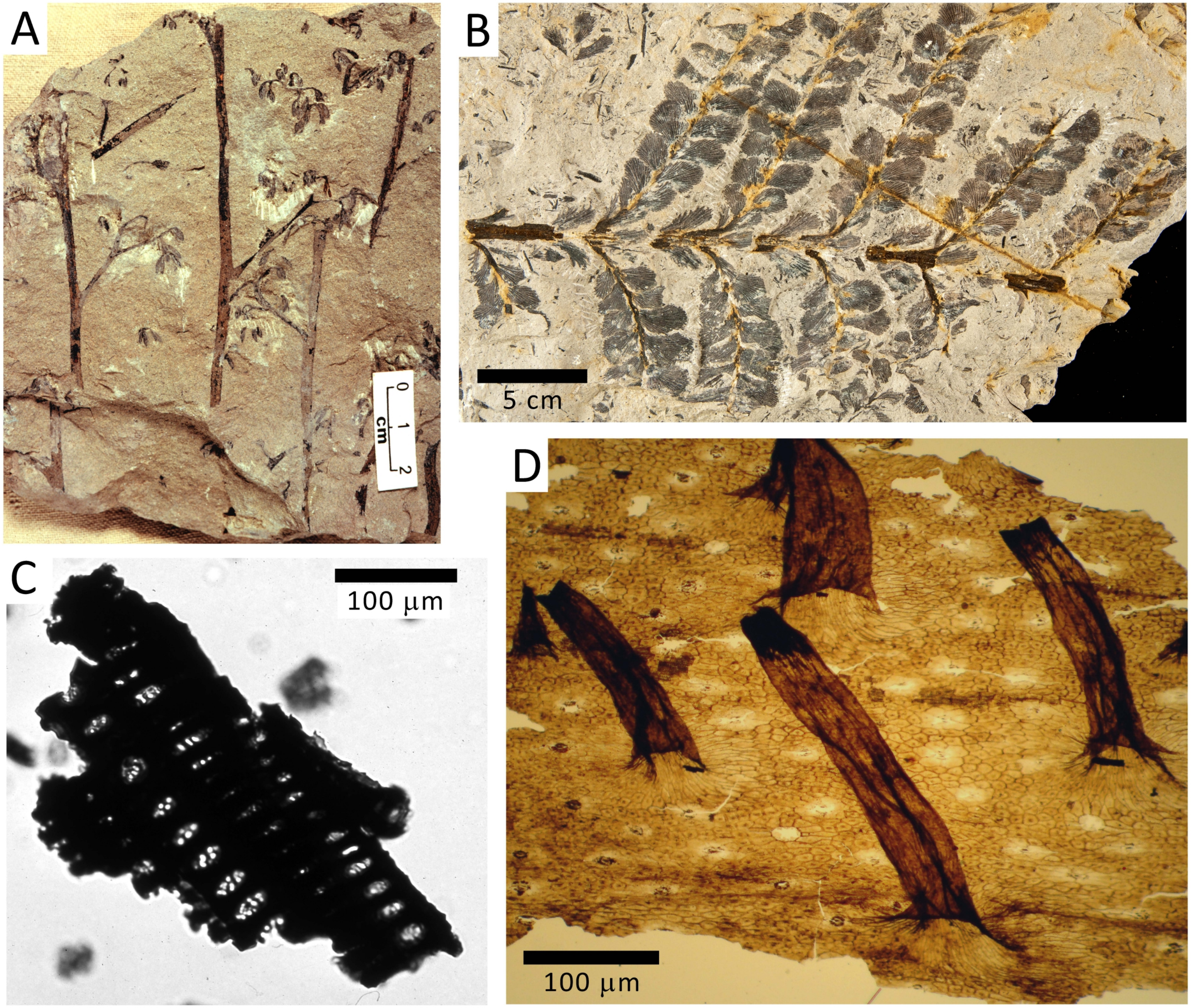


Figure 4

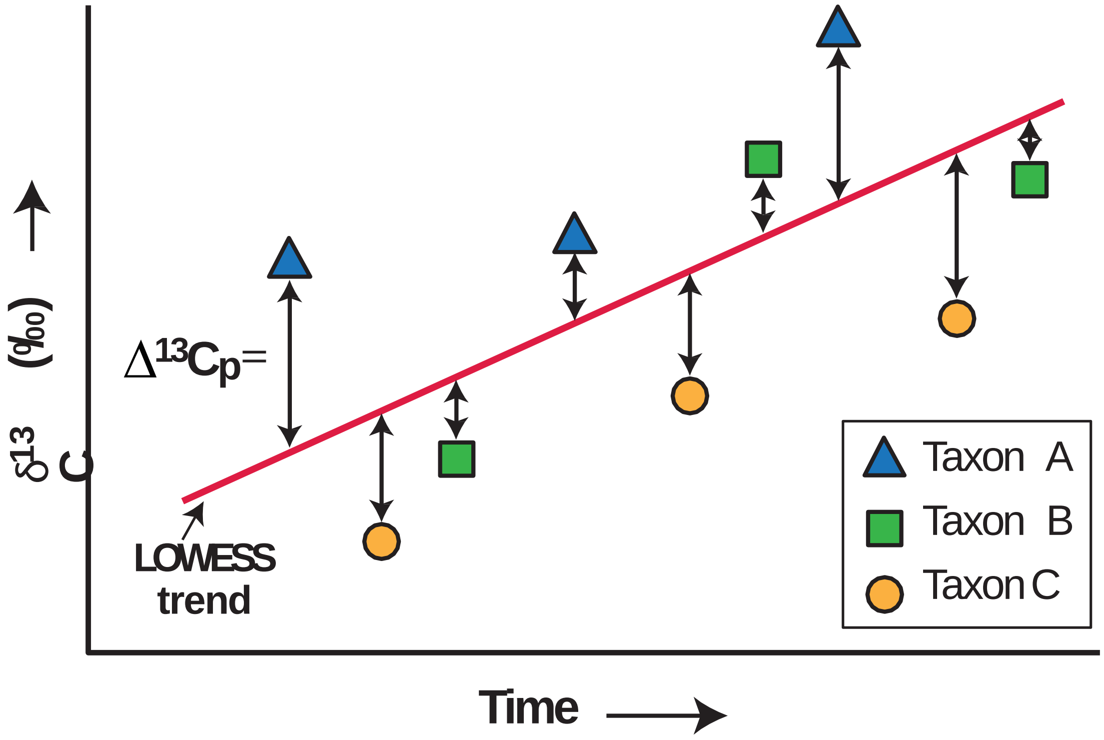
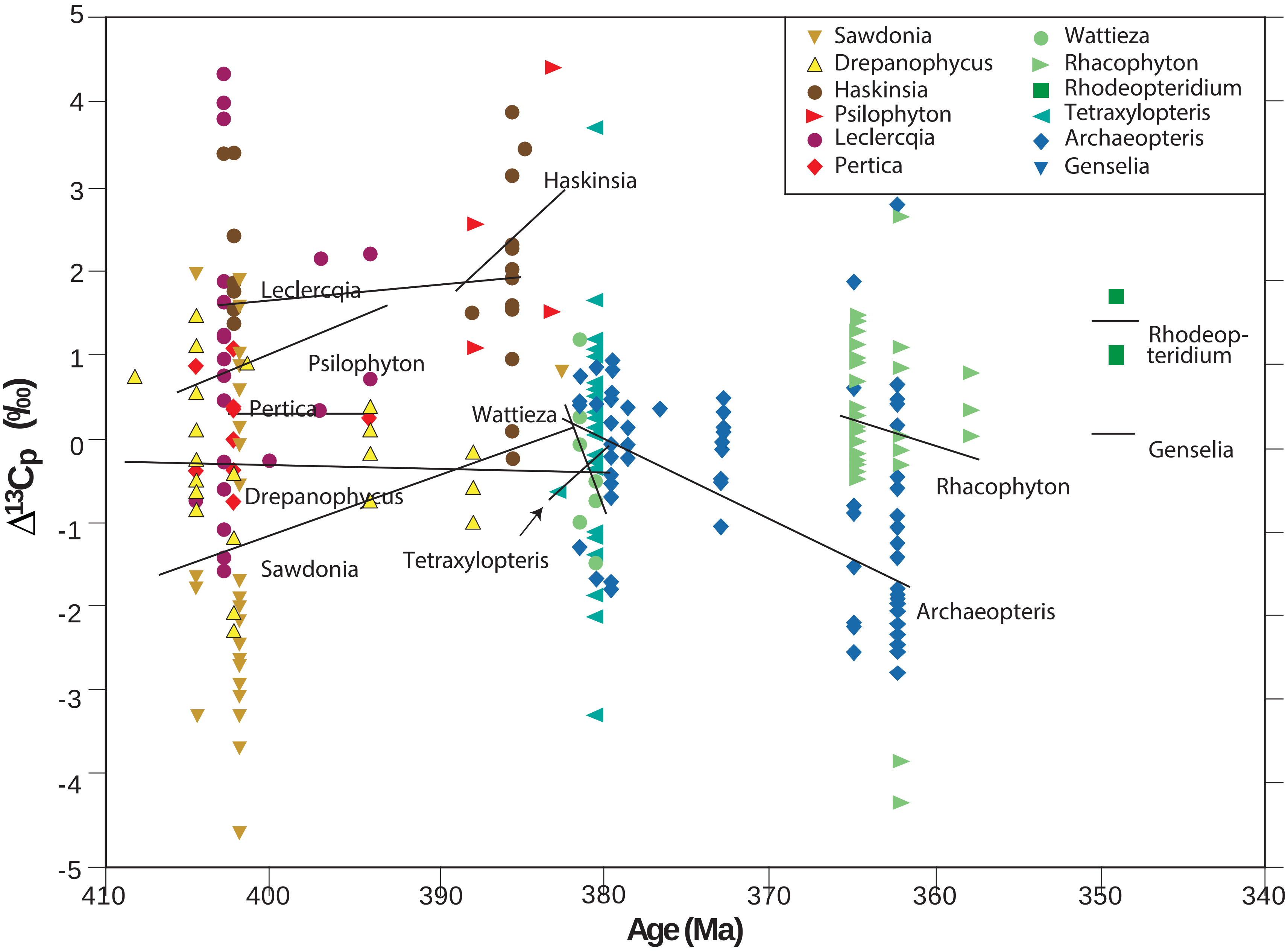


Figure 5



5
4
3
2
1
0
-1
-2
-3
-4
-5
Δ^{13}Cp (‰)
410
400
390
380
370
360
350
340
Age (Ma)
Sawdonia
Drepanophycus
Haskinsia
Psilophyton
Leclercqia
Pertica
Wattieza
Rhacophyton
Rhodeopteridium
Tetraxylopteris
Archaeopteris
Genselia
Haskinsia
Leclercqia
Psilophyton
Pertica
Wattieza
Drepanophycus
Sawdonia
Tetraxylopteris
Rhodeop-teridium
Genselia
Rhacophyton
Archaeopteris

Figure 6

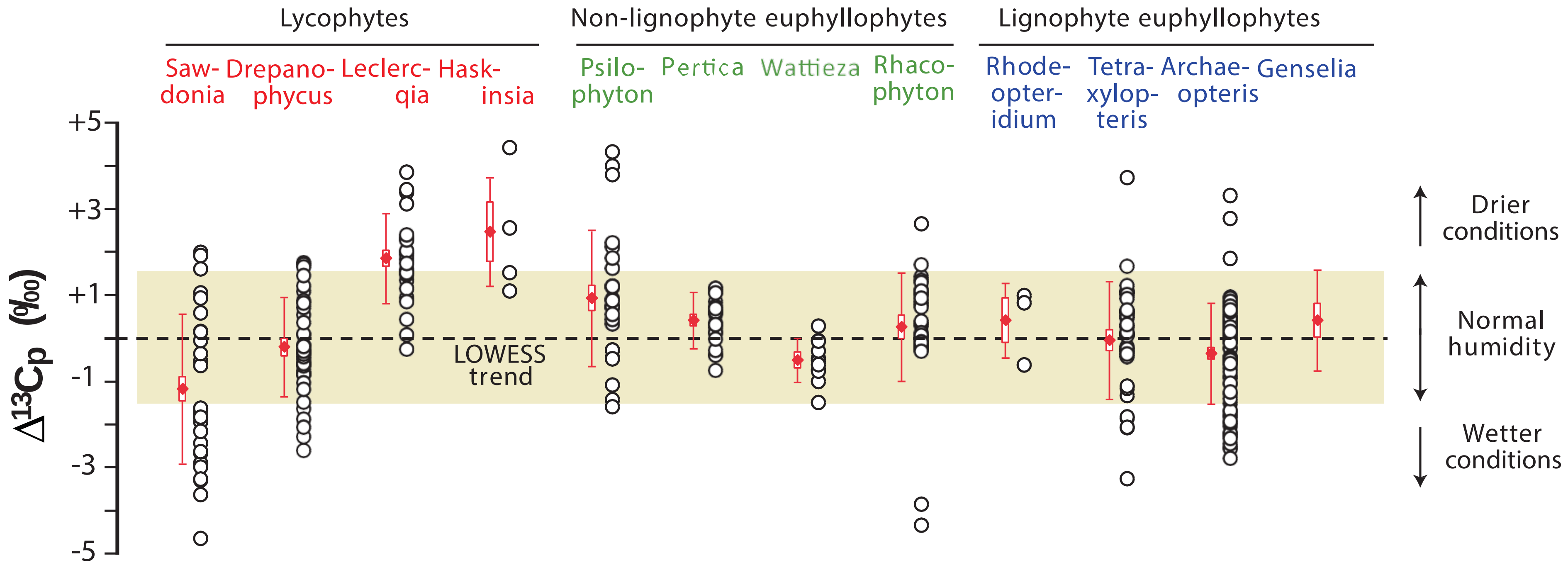
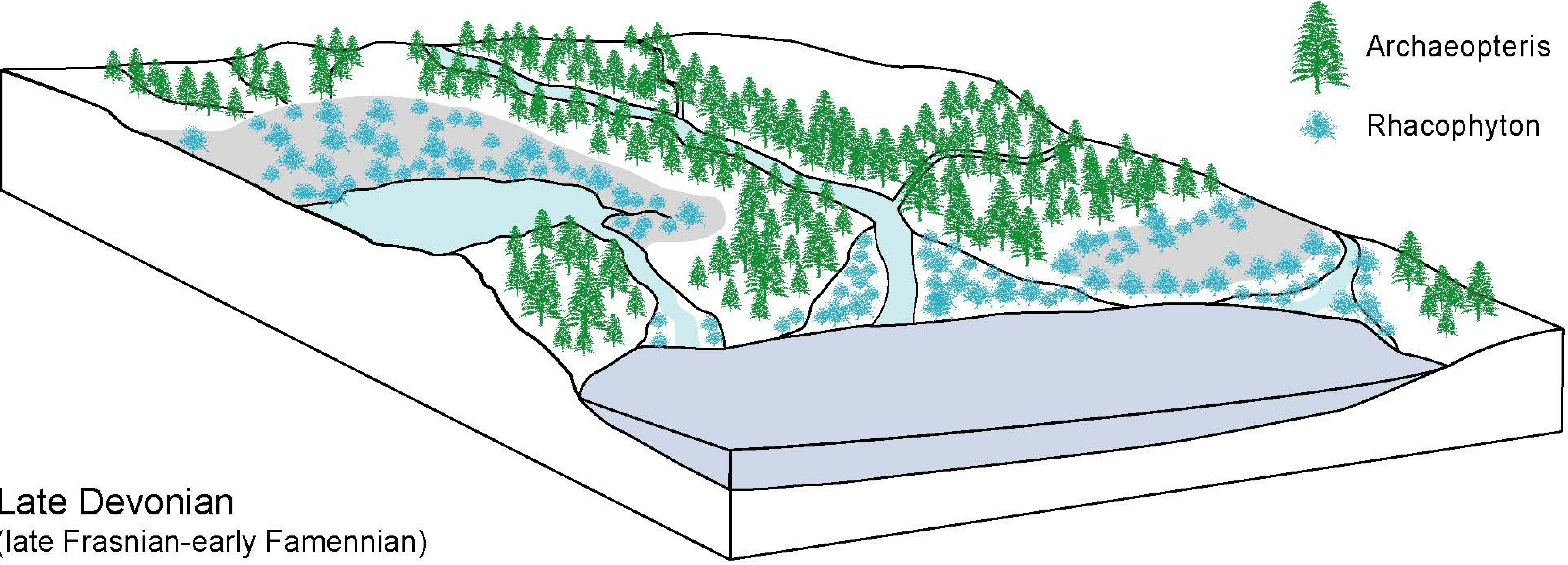
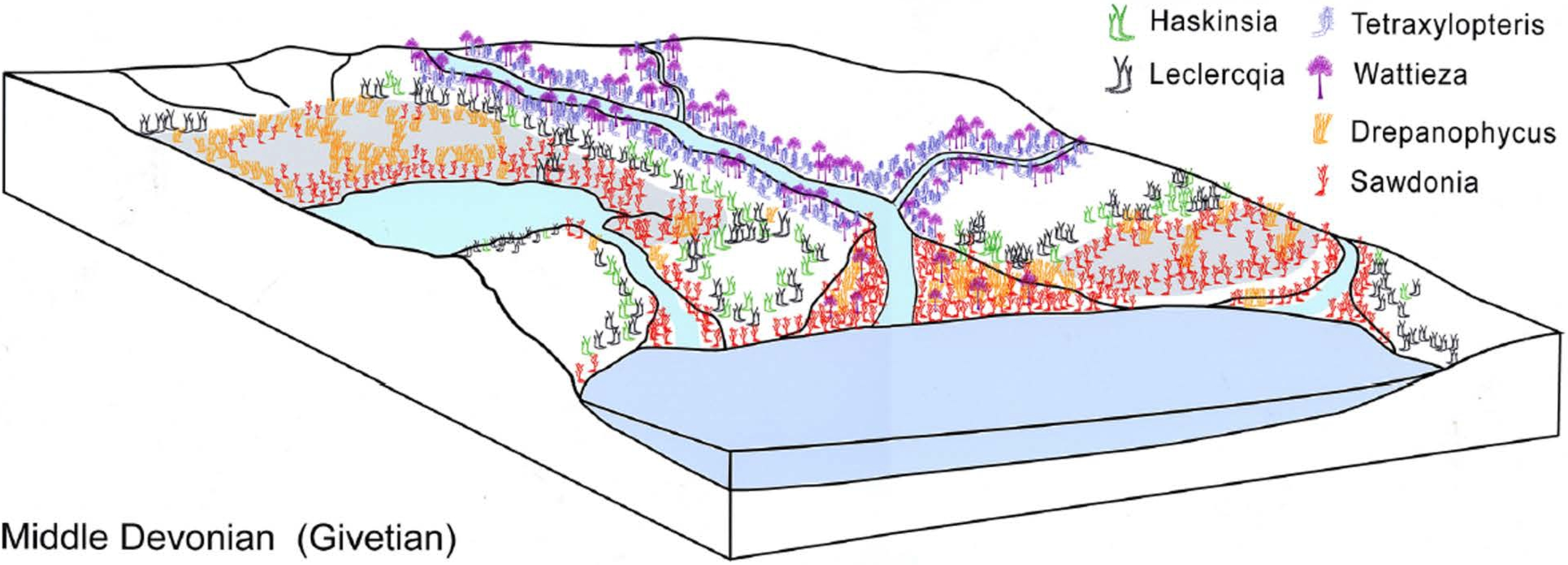

Lycophytes
Non-lignophyte euphyllophytes
Lignophyte euphyllophytes
Sawdonia
Drepanophycus
Leclercqia
Haskinsia
Psilophyton
Pertica
Wattieza
Rhacophyton
Rhodeopteridium
Tetraxylopteris
Archaeopteris
Genselia
+5
+3
+1
-1
-3
-5
$\Delta^{13}C_p$ (‰)
LOWESS trend
Drier conditions
Normal humidity
Wetter conditions

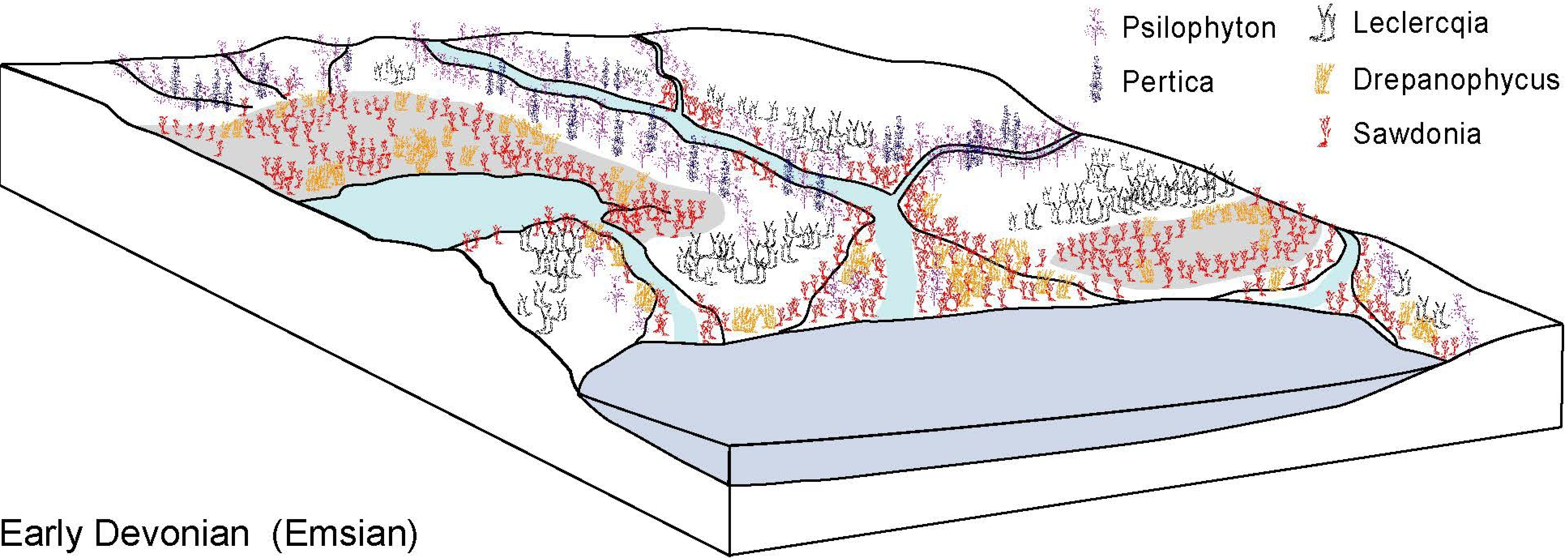
Figure 7



Late Devonian
(late Frasnian-early Famennian)



Middle Devonian (Givetian)



Early Devonian (Emsian)