Elevated CO₂, increased leaf-level productivity and water-use efficiency during the early Miocene

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Abstract. Rising atmospheric CO₂ is expected to increase global temperatures, plant water-use efficiency, and carbon storage in the terrestrial biosphere. A CO₂ fertilization effect on terrestrial vegetation is predicted to cause global greening as the potential ecospace for forests expands. However, leaf-level fertilization effects, such as increased productivity and water-use efficiency, have not been documented from fossil leaves in periods of heightened atmospheric CO₂. Leaf gas-exchange rates reconstructed from early Miocene fossils which grew at southern temperate and tropical latitudes, when global average temperatures were 5–6°C higher than today reveal that atmospheric CO₂ was ~450–550 ppm. Early Miocene CO₂ is similar to projected values for 2040AD, and consistent with Earth System Sensitivity of 3–7°C to a doubling of CO₂. While early Miocene leaves had photosynthetic rates similar to modern plants, southern temperate leaves were more productive than modern due to a longer growing season. This higher productivity was likely mirrored at northern temperate latitudes as well, where a greater
availability of landmass would have led to increased carbon storage in forest biomass relative to today.

Intrinsic water-use efficiency of both temperate and tropical forest trees was high, toward the upper limit of the range for modern trees, which likely expanded the habitable range in regions that could not support forests with high moisture demands under lower atmospheric CO\(_2\). Overall, early Miocene elevated atmospheric CO\(_2\) sustained globally higher temperatures and our results reveal the first empirical evidence of concomitant enhanced intrinsic water-use efficiency, indicating a forest fertilization effect.

1 Introduction

Terrestrial plants comprise 450 Gt of carbon, representing 80% of Earth’s dry carbon (C) biomass (Bar-on et al., 2018). Globally, plants draw down ~120 Gt of atmospheric C per year through photosynthesis, representing the largest annual C flux on Earth (Beer et al., 2010). Total plant biomass is believed to be determined in large part by atmospheric carbon dioxide concentrations (C\(_a\)), and it is predicted that future increases in C\(_a\) will have a three-pronged effect on the terrestrial biosphere: 1) increased global temperatures will shift the boundaries of climate zones and thereby the potential forest expanse (Rubel and Kottek, 2010); 2) productivity will increase because global photosynthesis is C limited and increased C\(_a\) will make more C available to the terrestrial biosphere (Zhu et al., 2016); and 3) elevated C\(_a\) will increase plant water-use efficiency and reduce the threshold for physiological drought (Cernusak, 2020), making more land area available for biosphere expansion (Zhou et al., 2017). Plant fossils record the effect of past changes in climate, including CO\(_2\) enrichment, and thus fossil floras provide insight into changes in the carbon cycle and their effects on the terrestrial biosphere from a natural, whole-ecosystem perspective.

The Miocene has been considered problematic for our understanding of Earth System Sensitivity (ESS) to C\(_a\), because most proxy-based C\(_a\) estimates indicate concentrations near 300 ppm (Foster et al., 2017), close to pre-industrial values, yet global temperatures were 5–6 °C higher than modern (Hansen et al., 2013). Enhanced radiative forcing is required to maintain such elevated early Miocene temperatures (Herold et al., 2010; Hansen et al., 2013), and without elevated C\(_a\), climate models cannot achieve such
high global temperatures in the Miocene (Henrot et al., 2010). The early Miocene also had an expanded biosphere compared to today, including woody vegetation in locations that are currently too cold and/or too dry for forests (e.g. Askin and Raine, 2000; Herold et al., 2010). A biosphere of the magnitude observed in the early Miocene fossil record requires elevated temperatures and plant water-use efficiency, suggesting C₄ levels higher than estimated by most existing proxy reconstructions (Herold et al., 2010; Henrot et al., 2010). Importantly, plant-based C₄ reconstructions have challenged the consensus of low C₄ in the early Miocene (Kürschner and Kvaček, 2009; Reichgelt et al., 2016; Tesfamichael et al., 2017; Londoño et al., 2018) and previous interpretations of alkenone-based C₄ proxies are being disputed (Bolton et al., 2016; Witkowski et al., 2019).

We applied plant gas-exchange modeling (Franks et al., 2014) to fossil leaves from the early Miocene (~23 Ma) rainforest ecosystem from southern New Zealand preserved in the Foulden Maar deposit (Bannister et al., 2012; Reichgelt et al., 2013; Conran et al., 2014; Lee et al., 2016) to reconstruct carbon assimilation rates (Aₙ), intrinsic water-use efficiency (iWUE; the ratio between carbon assimilation and stomatal conductance to water), and the C₄ levels required to maintain these values. The same analyses were performed on previously published leaf δ¹³C and micromorphological measurements from two early Miocene fossil floras from Ethiopia (Tesfamichael et al., 2017) and Panama (Londoño et al., 2018).

Because Aₙ, iWUE, and C₄ are interdependent (Farquhar et al., 1980; Drake et al., 1997), we reconstructed these variables in concert for each fossilized leaf recovered from these forest ecosystems. This allows us to make inferences concerning carbon availability, productivity, and water balance in the forest.

2 Methods

2.1 Site Description

Foulden Maar (Fig. 1a) is a unique Konservat-Lagerstätte with abundantly preserved plants and insects (Kaulfuss et al., 2015; Lee et al., 2016) in southern New Zealand (45.527°S, 170.219°E). It was formed in an ancient maar-diatreme lake (Fig. 1b) at the Oligocene/Miocene boundary (Fox et al., 2015; Kaulfuss,
2017) and consists of ~100 kyr of annually laminated diatomite (Lindqvist and Lee, 2009; Fox et al., 2016). The Foulden maar-diatreme complex is part of the larger late Oligocene – late Miocene Waipati Volcanic Field that produced a variety of maar volcanoes and scoria cones (Németh and White, 2003). Plant fossils used in this study were collected from a ~183 m long drill core (Fig. 1c). The Lauraceae-dominated rainforest (Bannister et al., 2012) surrounding the lake grew at ~50°S (Fig. 1a). The climate was marginally subtropical with a mean annual temperature of ~18°C, similar to modern day climates at 30°S (Reichgelt et al., 2019). The length of the growing season in this climate was ~10 months, compared to 5–6 months today, as reconstructed from the surface exposure macrofossil assemblage using the Climate Leaf Analysis Multivariate Program (Reichgelt et al., 2013).

2.2 Fossil leaf anatomy and paleoecology

Mummified fossil leaves were extracted from turbidite deposits that occur frequently within the Foulden Maar diatomite core (Fox et al., 2015). The diatomite is loosely consolidated allowing mummified leaves to be extracted using a combination of water and scalpels. After extraction, the leaves were cut into three pieces: one for bulk δ¹³C analysis, one for stomatal conductance measurements, and a third as a reference specimen. Leaf δ¹³C was measured using a Costech elemental combustion system (EA) coupled to a Delta V Plus IRMS (Thermo). In order to place measured δ¹³C on the VPDB scale, we calibrated measurements using a two-point isotope calibration based on the USGS40 and USGS41 standards. Measurement uncertainty was calculated by replicating ~15 samples, and applying averaged uncertainty to the remaining leaves. For conductance measurements the leaves were soaked in hydrogen peroxide (H₂O₂) with up to 40% dilution, with tetra-sodium pyrophosphate salt crystals (Na₄P₂O₇ · 10[H₂O]), on a boiling plate at 40–50 °C for 1–2 hours (Bannister et al., 2012). When the adaxial and abaxial cuticle layer could be separated, the leaf layers were cleaned of mesophyll cell debris using small paintbrushes and both layers were stained with <0.5% Crystal Violet (C₂₃N₆H₉O₂Cl) and mounted on glass slides with glycerin jelly. Conductance measurements were made on 5–8 pictures at 100× magnification using TSVView 7.1.1.2 microscope imaging software on a Nikon Optiphot. Each picture was given bounding boxes (0.3 ×
0.3 mm) on which cells were counted, to calculate stomatal density, and stomatal size measurements were made using ImageJ 1.48v software (Schneider et al., 2012).

18 distinct leaf morphotypes were identified from the Foulden Maar drill core. Descriptions and justification for identification are found in the Supplementary Information. Species identifications are provided, where possible, based on paleobotanical studies from the Foulden Maar surface exposures.

Known species recovered from the Foulden Maar drill core are *Litsea calicarioides* (Fig. S1a), *Cryptocarya taieriensis* (Fig. S1b), *C. maarensis* (Fig. S1i), *Beilschmiedia otagoensis* (Fig. S2a) (Lauraceae) (Bannister et al., 2012), *Laurelia otagoensis* (Fig. S2h) (Atherospermataceae) (Conran et al., 2013), and *Hedycarya pluvisilva* (Fig. S2i) (Monimiaceae) (Conran et al., 2016). Otherwise, tentative genus or family identifications are provided, or unspecified morphotypic qualifiers, for leaves that could not be assigned a plant group. These will be henceforth be referred to as “C” (Fig. S1c), cf. Myrtaceae (Fig. S1d), cf. *Ripogonum* (Fig. S1e), cf. *Myrsine* (Fig. S1f), “H” (Fig. S1g), cf. Elaeocarpaceae/Cunoniaceae (Fig. S1h), cf. *Dysoxylum* (Fig. S2b), cf. *Cryptocarya* (Fig. S2c), “O” (Fig. S2d), “P” (Fig. S2e), “Q” (Fig. S2f) and cf. *Endiandra* (Fig. S2g).

We made 375 anatomical and 80 carbon isotope measurements on 72 organically preserved fossil leaves representing the 18 species collected from the Foulden Maar deposit (Fig. 1a). The affinities of modern living relatives of the plant types at Foulden Maar strongly suggest that during the Miocene the site was characterized by a multi-layered closed canopy rainforest ecosystem (Reichgelt et al., 2013; Conran et al., 2014). In order to determine atmospheric carbon (C_a), intrinsic water-use efficiency (iWUE), and carbon assimilation rates (A_n), the ecological strategies of the individual fossil species at Foulden Maar need to first be established (Reichgelt and D’Andrea, 2019). Understory species rarely experience light saturation and utilize respired CO_2 that has already undergone isotopic fractionation; both conditions influence gas-exchange modelling results (Royer et al., 2019). Therefore, C_a reconstructed from understory species cannot be considered indicative of true global C_a. Here, we determine whether a fossil leaf type was likely in the canopy or the understory, based on 1) leaf δ^{13}C, 2) leaf cell density, and 3) sinuosity of the epidermal cell walls. A large range of leaf δ^{13}C in a single species...
is indicative of different levels of light saturation, which indicates that this species may preferentially occur in the subcanopy or in the understory (Graham et al., 2014). Leaves in the canopy, experiencing light saturation, divide epidermal cells rapidly compared to leaves in the shade, leading to high cell densities and relatively high leaf mass per areas in sun-exposed leaves (Šantrůček et al., 2014). Finally, a high level of anticlinal cell wall sinuosity has been interpreted as indicative of low-light conditions (Kürschner, 1997; Bush et al., 2017). We consider these three lines of evidence occurring in concert as indicative of a canopy or subcanopy ecological preference.

2.3 Modelling gas-exchange

Atmospheric carbon dioxide (\(C_a\)), plant photosynthesis (\(A_n\)), and intrinsic water-use efficiency (iWUE) are tightly linked (Farquhar et al., 1980; Drake et al., 1997), which allows us to solve for these parameters iteratively, through anatomical and carbon isotope (\(\delta^{13}C\)) measurements of the fossil leaves. The Franks et al. (2014) gas-exchange model solves for \(C_a\), by iteratively reconstructing \(A_n\) and leaf conductance to atmospheric carbon (\(G_c\)), using a Monte Carlo approach. This means that every \(C_a\) reconstruction has an associated \(A_n\) and \(G_c\) value.

\[
C_a = \frac{A_n}{G_c \times \left(1 - \frac{C_i}{C_a}\right)} \tag{1}
\]

In which \(C/C_a\) represents the ratio of intercellular carbon to atmospheric carbon, which can be reconstructed using known leaf fractionation processes: fractionation caused by diffusion (\(a\)), carboxylation (\(b\)), and fractionation caused by the preferential uptake of \(^{12}\)C to \(^{13}\)C in photosynthesis (\(\Delta\)), which is also influenced by the rate at which the leaf is photosynthesizing (Farquhar et al., 1982).

\[
\frac{C_i}{C_a} = \frac{\Delta - a}{b - a} \tag{2}
\]
Here, \( a = 4.4\% \text{oo} \) and \( b = 29\% \text{oo} \) (Farquhar et al., 1982; Roeske and O’Leary, 1984). \( \Delta \) can be calculated from the \( \delta^{13}C \) of the air, derived from Tipple et al. (2010) and measurements of leaf \( \delta^{13}C \) (Farquhar and Richards, 1984; Farquhar et al., 1989). Leaf and air \( \delta^{13}C \) used in the Franks et al. (2014) model are presented in Table S1.

\[
\Delta = \frac{\delta^{13}C_{\text{air}} - \delta^{13}C_{\text{leaf}}}{1 + \delta^{13}C_{\text{leaf}}} (3)
\]

\( G_c \) is determined by the maximum capacity for conductance of a leaf surface \( (G_{\text{max}}) \), the ratio of operational conductance to \( G_{\text{max}} \) (\( \zeta \)), boundary layer conductance \( (G_b) \), and mesophyll conductance \( (G_m) \) (Franks et al., 2014).

\[
G_c = \left( \frac{1}{G_b} + \frac{1}{\zeta \times G_{\text{max}}} + \frac{1}{G_m} \right)^{-1} (4)
\]

\( G_b, \zeta, \text{and} G_m \) are all changeable under natural conditions (e.g. Schuepp, 1993; Niinemets et al., 2009; Londoño et al., 2018) and it is highly disputed if these variables can be determined from fossil leaf material at all (e.g. McElwain et a., 2016; Soh et al., 2017). However, we adopt a standardized approach put forward by Franks et al. (2014) to obtain input for these variables. \( G_b = 2 \pm 0.1 \text{ mol m}^{-2} \text{ s}^{-1}, \zeta = 0.2 \pm 0.02 \) (Franks et al., 2009; Dow et al., 2014), and \( G_m \) is determined using an empirical calibration (Evans and Von Caemmerer, 1996).

\[
G_m = 0.013 \times A_n (5)
\]
G_{\text{max}} is determined using predominantly measurable anatomical features of the fossil leaf cuticle (Franks and Beerling, 2009): stomatal density (SD), maximum aperture surface area (a_{\text{max}}), pore depth (p_d), and the ratio of diffusivity of CO_2 in air over the molar volume of air (d/v), here taken as 0.000714 mol m^{-3} s^{-1} (Marrero and Mason, 1972).

\[
G_{\text{max}} = \frac{d}{v} \times SD \times \frac{a_{\text{max}}}{p_d + \frac{\pi}{2} \sqrt{a_{\text{max}}/\pi}} \quad (6)
\]

In this equation, SD can be measured directly from the leaf, p_d is assumed to be the same as guard cell width (gcw), and a_{\text{max}} is determined assuming a circular opening for the aperture, with the measurable pore length (p_l) as the diameter (Franks et al., 2014).

\[
a_{\text{max}} = \pi \times \frac{p_l}{4} \quad (7)
\]

Measurements of SD, gcw and p_l used in the Franks et al. (2014) gas-exchange model are presented in Table S1.

Hence, it is clear that though G_c is largely determined by measurable anatomical parameters, because A_n is a component of calculating G_{\text{max}}, G_c is solved iteratively. However, A_n is also solved iteratively, as it is dependent on C_a and the carbon saturation value (\Gamma), set at 40 ppm (Franks et al., 2013).

\[
A_n \approx A_0 \times \frac{(C_a - \Gamma) \times (C_{a0} + 2\Gamma)}{(C_a + 2\Gamma) \times (C_{a0} - \Gamma)} \quad (8)
\]

In which A_0 is the photosynthetic rate of a modern model species that can represent the fossil species’ photosynthetic rate, and C_{a0} is the atmospheric carbon dioxide level at which A_0 was measured. A_0 for
each fossil species was derived from the compilation of photosynthetic rates presented in Reichgelt and D’Andrea (2019). For fossil leaves with known modern relatives, we constrained the possible $A_0$ range by only including modern relatives within the same family or order, i.e. Lauraceae for *Litsea calicarioides*, *Cryptocarya taieriensis*, *C. maarensis*, cf. *Cryptocarya*, *Beilschmiedia otagoensis* and cf. *Endiandra*, Myrtaceae for cf. Myrtaceae, Liliales for cf. *Ripogonum*, Primulaceae for cf. *Myrsine*, Elaeocarpaceae and Cunoniaceae for cf. Elaeocarpaceae/Cunoniaceae, Meliaceae for cf. Meliaceae, Atherospermataceae for *Laurelia otagoensis*, and Laurales for *Hedycarya pluvisilva*. Then, following the method of constraining $A_0$ of modern living relatives presented in Reichgelt and D’Andrea (2019), only $A_0$ values of plants with similar growth forms to the fossil plants, and growing in similar light environments as Foulden Maar were included. $A_0$ and $C_a$ used in the Franks et al. (2014) model, and associated ecology of fossil leaf types is shown in Table S2.

The Franks et al. (2014) gas-exchange model thus iteratively solves for $C_a$, $A_n$, and $G_c$. However, only leaves derived from canopy trees are likely to represent these values at light saturation. Moreover, plants in the understory assimilate a mix of atmospheric and respired $CO_2$, which has therefore already undergone fractionation processes, making the calculated $C_i/C_a$ problematic. Therefore, we present the results for $C_a$, $A_n$, and $G_c$ of leaf types most likely to be derived from canopy trees separately, as they are more likely to not have a systematic skew.

$iWUE$ is defined as the ratio between $A_n$ and stomatal conductance to water (Feng, 1999).

\[ iWUE = \frac{A_n}{G_w} \]  

Due to the different rates at which carbon dioxide and water vapor diffuse in air, a transformation of $G_c$ is required to calculate $G_w$. 


Finally, cumulative annual carbon uptake through photosynthesis ($A_{\text{tot}}$) can be calculated in gC m$^{-2}$ yr$^{-1}$, by transferring from moles to grams, including a measure for the relative time the leaf is assimilating carbon ($\zeta$), and a measure for the absolute amount of time that the leaf was assimilating carbon.

$$A_{\text{tot}} = (2.6 \times \zeta \times A_n \times GS) \times 12 \quad (11)$$

In which GS is the length of the growing season, which we can derive from the fossil plant assemblage (Reichgelt et al., 2019), using the method of Spicer et al. (2009). $G_w$, $A_{\text{tot}}$, and iWUE values for *Litsea calicarioides*, *Cryptocarya taieriensis*, *C. maarenisis*, cf. Elaeocarpaceae/Cunoniaceae, and cf. Myrtaceae are presented in Table S3. The modern reference $A_n$ and $G_w$ data is derived from Maire et al. (2015), which included coordinates, habit, $A_n$ and $G_w$ data, from which we could then calculate iWUE and $A_{\text{tot}}$.

### 2.4 Comparison to Earth System Sensitivity

Earth System Sensitivity to $C_a$ (ESS) is the amount of temperature increase expected under a doubling of atmospheric CO$_2$. This sensitivity is likely not static in Earth’s history and is dependent on, among other aspects, continental configuration and ocean circulation patterns (Royer, 2016). ESS is likely to have been between 3–7°C for the Neogene (Hansen et al., 2013; Royer, 2016), meaning that a doubling of $C_a$ compared to pre-industrial levels would have led to an increase of global average surface temperatures ($T_s$) of 3–7°C compared to modern. Using this expected ESS, we calculated the expected $C_a$ using the methodology of Hansen et al. (2013).

Compiled deep-sea benthic foraminifera $\delta^{18}$O data of the last 30 million years (Zachos et al., 2001) were averaged into 20 kyr time bins. Deep-sea temperatures ($T_d$) were then calculated using the linear transfer functions of Hansen et al. (2013), which depend on the presence of sea-ice.
\[ T_d = 5 - 8 \times \frac{\delta^{18}O - 1.75}{3} \ IF (\delta^{18}O < 3.25) \] (12)

\[ T_d = 1 - 4.4 \times \frac{\delta^{18}O - 3.25}{3} \ IF (\delta^{18}O > 3.25) \] (13)

T_s was then calculated for post-Pliocene using:

\[ T_s = 2 \times T_d + 12.25 \] (14)

For the Pliocene:

\[ T_s = 2.5 \times T_d + 12.15 \] (15)

And for pre-Pliocene we assumed that T_s changed linearly with T_d, by a factor of 1.5.

\[ \Delta T_s = 1.5 \times \Delta T_d \] (16)

C_s based on an ESS range of 3–7°C was then calculated using the resulting T_s.

\[ C_s = 310 \times \frac{T_s[x] - T_s[0]}{2 \times ESS} + 310 \] (17)

In which T_s[x] is the calculated average global surface temperature at time x, and T_s[0] is the modern day average global surface temperature.

3 Results and Discussion

3.1 Southern Temperate Rainforest Paleocology

Modern day Lauraceae rainforests in New Zealand have a single dominant canopy tree, *Beilschmiedia tawa*, and their farthest southern extent is ~42°S (Leathwick, 2001), the farthest southern occurrence of
any arborescent Lauraceae species in the world. Rainforests at higher latitudes in New Zealand are
usually dominated by Nothofagaceae or Podocarpaceae, and the only modern-day forests at ~50°S are the
Magellanic Subpolar Forests in southern South America. Low-growing Podocarpaceae/Nothofagaceae
forests, similar to modern forests in southern New Zealand and southern South America, dominated
Antarctic vegetation during the early Miocene (Askin and Raine, 2000) and the Foulden Maar rainforest
included at least ten Lauraceae species (Bannister et al., 2012), emphasizing the expanded biosphere
potential in the early Miocene compared to today (Herold et al., 2010).

We identify *L. calicarioides*, *C. maarensis*, *C. taieriensis*, cf. Elaeocarpaceae/Cunoniaceae, and cf.
Myrtaceae as the most probable canopy components because they lack 1) the large range of leaf δ13C
values, relatively low overall leaf δ13C values (Graham et al., 2014), 2) low cell densities typical of
understory components (Kürschner, 1997; Bush et al., 2017) (Fig. 2a,b), and 3) the undulating or sinuous
cell walls typical of understory components (Kürschner, 1997; Bush et al., 2017). The most likely
subcanopy or understory taxa were cf. *Ripogonum*, cf. *Myrsine*, “O”, and cf. *Dysoxylum*, because leaf
fossils of these types have both low leaf δ13C and sinuous or undulating cells (Fig. S1e,f, S2b,d). *H. pluvisilva*, *L. otagoensis*, cf. *Cryptocarya*, cf. *Endiandra*, *B. otagoensis*, “C”, “H”, “P”, and “Q”, all
displayed some variation in these features and occurred in relative low abundance, and are therefore
considered of uncertain ecological affinity.

### 3.2 Earliest Miocene CO2

Gas-exchange modeling (Franks et al., 2014) of canopy leaves throughout the Foulden Maar core
indicates that *C*<sub>a</sub> was 445 ±618 / -100 ppm, whereas understory elements experienced a higher *C*<sub>a</sub> of 622
+3017 / -161 ppm (Fig. 2c), consistent with understory plants assimilating respired CO2 that has
undergone prior fractionation processes, as well as experiencing elevated levels of *C*<sub>a</sub> under the canopy
(Graham et al., 2014; Royer et al., 2017). Prior work on the Foulden Maar core established three different
phases based on bulk organic δ13C (Fig. 1c), fatty acid δ13C, and fatty acid δD: Phase I (80–105 m depth)
with high δ13C and low δD, Phase II (55–65 m depth) with low δ13C and high δD, and Phase III (0–45 m
depth) with high δ\textsuperscript{13}C and low δD (Reichgelt et al., 2016). Phase III can be further subdivided into Phase IIIa (30–45 m depth) and IIIb (0–20 m depth), as Phase IIIa exhibits a period of low fatty acid δ\textsuperscript{13}C and high δD, which is not expressed in bulk organic δ\textsuperscript{13}C (Reichgelt et al., 2016). Gas-exchange modelling on leaves from these phases (Fig. 1c) suggest that during Phase II and IIIa C\textsubscript{a} may have been elevated (C\textsubscript{a} = 529 +1159/ -125 and C\textsubscript{a} = 538 +769/ -181 ppm, respectively) compared to Phase I and Phase IIIb (C\textsubscript{a} = 444 +572/ -95 and 442 +1219/ -110 ppm, respectively) (Fig. 3).

The advantage of using gas-exchange modeling to reconstruct C\textsubscript{a} from multiple species is that the uncertainty is quantified and constrained, greatly reducing the potential for systematic error in the final estimate (Reichgelt and D’Andrea, 2019; Royer et al., 2019). Along with the enhanced accuracy comes a more comprehensive appraisal of uncertainty than is achieved using other proxy approaches (Fig. 4).

Proxy error propagation is based on mechanistic variability, grounded in known physical and physiological limits of plant gas-exchange that are understood to be universal (Franks et al., 2014). This differs from empirical proxies, whose uncertainty representation is based on calibration error of modern-day observations without mechanistic constraints. Our canopy C\textsubscript{a} estimate (445 +618/ -100 ppm, Fig. 2c) is independent of calibration error, based on universal gas-exchange mechanisms, and represents plant vegetative organs of multiple plant species that directly interacted with the available pool of atmospheric carbon dioxide. Previous C\textsubscript{a} estimates from the Oligocene/Miocene boundary based on boron isotopes and paleosol carbonates are generally lower than our estimates (Ji et al., 2018; Greenop et al., 2019) (Fig. 4b), whereas C\textsubscript{a} estimates based on stomatal index and recent alkenone-based C\textsubscript{a} estimates are more similar to our results (Kürschner et al., 2008; Super et al., 2018).

Reconstructions of globally elevated temperatures of 5–6 °C in the early Miocene (Hansen et al., 2013) with a C\textsubscript{a} of ~300 ppm (Ji et al., 2018; Greenop et al., 2019) upsets the expected ESS to C\textsubscript{a} during this period (Henrot et al., 2010). Geochemical C\textsubscript{a} proxy estimates consistently produce C\textsubscript{a} estimates that are too low to satisfy ESS to C\textsubscript{a} prior to the Pliocene (Royer, 2016) (Fig. 4a,b). Estimates from the fossil leaf-based stomatal index proxy for C\textsubscript{a} (Kürschner et al., 2008) on the other hand do indicate a positive correlation between temperature and C\textsubscript{a} in the Neogene (Fig. 4a). At present, there are too few studies that
reconstruct C<sub>a</sub> using gas-exchange modeling to allow for a full comparison to other C<sub>a</sub> proxies; however, our C<sub>a</sub> estimates of ~450–550 ppm are in line with the ESS to C<sub>a</sub> in the early Miocene (Fig. 4a,b), based on modelling experiments (Herold et al., 2010; Henrot et al., 2010). Moreover, thus far, Neogene C<sub>a</sub> estimates reconstructed using gas-exchange methods (Reichgelt et al., 2016; Tesfamichael et al., 2017; Londoño et al., 2018; Moraweck et al., 2019) appear to agree with the suggested ESS to C<sub>a</sub> (Fig. 4a,b).

Bulk organic and leaf wax δ<sup>13</sup>C values reveal a ~4‰ decrease at Foulden Maar over a 10-meter interval at the beginning of Phase II (55–65 m depth), likely representing a time period of <10 kyr (Fox et al., 2016). This shift in isotopic composition suggests a substantial change in the global carbon cycle (Reichgelt et al., 2016). Results presented here show a ~100 ppm C<sub>a</sub> increase (from ~450 to 550) from Phase I to Phase II (Fig. 3). The C<sub>a</sub> values stay near 550 ppm throughout Phase II and Phase IIIa, representing a 20–40 kyr time period (Fig. 3). Absolute dating of Foulden Maar based on paleomagnetic reversals in the core, annual lamination of lake sediments, and basalt-derived Ar/Ar dates indicates that the deposition of the Foulden Maar sediment coincided with the termination of the earliest Miocene (Mi-1) glaciation of Antarctica (Fox et al., 2015). An increase in C<sub>a</sub> from ~450 to ~550 ppm at the termination of Mi-1 is consistent with modeling studies indicating that C<sub>a</sub> > 500 ppm is necessary to terminate a large-scale Antarctic glaciation (DeConto et al., 2008).

### 3.3 Elevated CO<sub>2</sub> and the early Miocene biosphere

The Foulden Maar Miocene rainforest was primarily evergreen (Lee et al., 2016). The main Miocene canopy trees at Foulden Maar, *Litsea calicarioides*, *Cryptocarya taieriensis*, *C. maarensis*, cf. Elaeocarpaceae/Cunoniaceae and cf. Myrtaceae, had relatively high iWUE (Miocene iWUE first quartile [Q<sub>1</sub>] – third quartile [Q<sub>3</sub>] = 70–101) compared to modern evergreen trees (evergreen iWUE Q<sub>1</sub>–Q<sub>3</sub> = 31–73) (Fig. 5a). Reconstructed iWUE from tropical early Miocene plants (Tesfamichael et al., 2017; Londoño et al., 2018) is slightly higher (Q<sub>1</sub>–Q<sub>3</sub> = 80–125) (Fig. 5a). The difference between reconstructed Miocene iWUE and that of modern deciduous trees is greater still (deciduous iWUE Q<sub>1</sub>–Q<sub>3</sub> = 27–52), consistent with the expectation that increased C<sub>a</sub> favors evergreen trees (Niinemets et al., 2011; Soh et al., ...
2019). In contrast to iWUE, reconstructed conductance to water ($G_w$) for Miocene trees is similar to the modern-day range at the same latitude (Fig. 5b), a somewhat surprising result because $G_w$ is expected to be reduced in high $C_a$ climates (Franks and Beerling, 2009). Increased atmospheric evaporative demand in combination with a longer growing season in a warmer climate would facilitate a relatively high hydraulic flux to the atmosphere, despite higher iWUE and similar $G_w$ (Frank et al., 2015). Furthermore, a longer growing season also results in increased total annual carbon flux ($A_{tot}$) to the biosphere (Fig. 5c).

Early Miocene trees at 50°S likely assimilated $A_{tot}$ $Q_1$–$Q_3 = 265$–696 g C m$^{-2}$ yr$^{-1}$, in comparison to $A_{tot}$ $Q_1$–$Q_3 = 108$–182 g C m$^{-2}$ yr$^{-1}$ in modern evergreen forests, and $A_{tot}$ $Q_1$–$Q_3 = 249$–410 g C m$^{-2}$ yr$^{-1}$ in modern deciduous forests at the same latitude (Fig. 5c). Tropical trees appear to have slightly higher total annual carbon flux ($A_{tot}$ $Q_1$–$Q_3 = 596$–1220 g C m$^{-2}$ yr$^{-1}$) than today ($A_{tot}$ $Q_1$–$Q_3 = 329$–721 g C m$^{-2}$ yr$^{-1}$).

Although this estimate cannot take the number of leaves per unit area into account, the results provide strong evidence for enhanced productivity in “greener” worlds.

4 Conclusions

Leaf-level gas-exchange derived $C_a$ estimates suggest that early Miocene atmospheric CO$_2$ was higher than pre-industrial levels at 450–550 ppm, further solidifying the growing consensus of relatively high early Miocene global temperatures maintained by high atmospheric CO$_2$ (Kürschner et al., 2009; Tesfamichael et al., 2017; Super et al., 2018; Londoño et al., 2018; Moraweck et al., 2019). A relatively high $C_a$ in the early Miocene also satisfies an Earth System Sensitivity of 3–7°C (Hansen et al., 2013; Royer, 2016). A potential shift in atmospheric CO$_2$ from 450 to 550, and back to 450, is recorded in the 100 kyr of sedimentation and leaf deposition at Foulden Maar. A disruption of the regional carbon and hydrological cycle was also recorded in leaf-wax $\delta^{13}C$ and $\delta$D (Reichgelt et al., 2016), and may be linked to the Antarctic deglaciation at the termination of the Mi-1 (DeConto et al., 2008; Fox et al., 2015; Liebrand et al., 2017).

The first record is provided of increased Miocene leaf-level intrinsic water-use efficiency in both temperate New Zealand and the tropics, and we provide evidence for increased leaf-level productivity in
temperate New Zealand. Enhanced productivity and water-use efficiency on other landmasses in temperate latitudes during the early Miocene, such as North America, Australia, and Asia, would have had a major impact on the global carbon and water cycles. Our gas-exchange results from New Zealand, supplemented with results from Ethiopia (Tesfamichael et al., 2017) and Panama (Londoño et al., 2018) provide empirical evidence for high water-use efficiency in the globally warmer world of the early Miocene, associated with elevated C₄. Tropical trees with high water-use efficiency compared to modern, would have likely facilitated forest survival in climates where currently tropical savannas and grasslands exist. An overall higher water-use efficiency in the early Miocene tropics is corroborated by the spread of C₄ grasslands in the late Miocene, when reduced CO₂ levels started favoring the more efficient C₄ photosynthetic pathway (Strömberg, 2011; Polissar et al., 2016).

Emission scenarios suggest that atmospheric CO₂ will reach our reconstructed early Miocene values of 450 ppm by 2030–2040 CE. While the global temperature response may lag the C₄ increase, and forest habitat expansion is hampered by the slow dispersal and growth rate of climax forest trees and anthropogenic influence (e.g., forest fragmentation and fire), early Miocene water-use efficiency and productivity estimates provide insight into the near future-biosphere potential, as well as into selective pressures that influence the types of plants that may proliferate under future elevated C₆.

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Author contributions. TR and WJD conceived of the idea and performed data analyses. BRSF and DEL collected sediment core, BRSF and TR sampled the sediment core, JGC and JMB identified fossil leaf
taxa. ACVM and TR gathered data from fossil leaves. TR and WJD wrote the paper and all authors contributed to the final manuscript.

**Competing interests.** The authors declare no competing interests.

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Figure 1. Foulden Maar site information. (a) Location of the Foulden Maar deposit and paleogeographic reconstruction of early Miocene New Zealand (Boyden et al., 2011; Lee et al., 2014). (b) Schematic reconstruction of the Foulden Maar depositional environment. (c) Stratigraphic column of the Foulden Maar core (Fox et al., 2015), with sample locations and bulk organic δ¹³C (Reichgelt et al., 2016).
Figure 2. Foulden Maar paleoecology and atmospheric CO₂ reconstructions. (a) δ¹³C values and stomatal density from the fossil leaves of Foulden Maar provide constraints to distinguish canopy leaf types from understory, because understory leaves tend to have a high range of δ¹³C and low cell density (Graham et al., 2014; Bush et al., 2017). (b) Paleoecological reconstruction of the dense rainforest at Foulden Maar with a canopy comprising *Litsea calicarioides* (Lcal), *Cryptocarya taieriensis* (Ctaï), *C. maarensis* (Cmaa), cf. Elaeocarpaceae/Cunoniaceae (E/C), and cf. Myrtaceae (Myrt), and an understory comprising cf. *Myrsine* (Myrs), cf. *Ripogonum* (Rip), cf. *Dysoxylum* (Dys), and leaf type “O”. *Hedycarya*
pluvialva (Hplu), Laurelia otagoensis (Lota), Beilschmiedia otagoensis (Bota), cf. Cryptocarya (cfC), cf. Endiandra (cfE), and leaf types “C”, “H”, “P”, and “Q” could not be ecologically placed with certainty.

(e) Probability density distributions of $C_a$ reconstructions from canopy (thick light green line) and understory components (thick dark green line) using a gas-exchange model (Franks et al., 2014). Grey curves represent 1000 Monte Carlo reconstructions on a single fossil leaf.

**Figure 3.** Probability density distributions of $C_a$ reconstructions using a gas-exchange model (Franks et al., 2014), divided by bulk carbon isotope phases (Fig. 1c).

Fig. 4
Figure 4. Neogene Earth System Sensitivity (ESS) and C₄ reconstructions. Calculated C₄ levels for an ESS range of 3–7 °C (orange shaded area) for the last 30 Ma (a), and for the interval between 22.5–23.5 Ma (b), the red dashed line in (b) indicates the global average surface temperature (Tₛ) in the earliest Miocene (Hansen et al., 2013). The ESS envelope was determined using deep-sea δ¹⁸O of benthic foraminifera (Zachos et al., 2001) and the transform function approach from Hansen et al. (2013) (Supplementary Information). Proxy-based Neogene C₄ reconstructions are derived from a previously published compilation (Foster et al., 2017) and are supplemented with more recently published data (Ji et al., 2019; Londoño et al., 2018; Super et al., 2018; Greenop et al., 2019; Moraweck et al., 2019, Steinthorsdottir et al., 2019). Error bars on gas-exchange based proxy estimates represent ±1σ.
**Figure 5.** Early Miocene leaf-level physiological parameters of canopy trees. (a) Intrinsic water-use efficiency (iWUE) of evergreen (green circles) and deciduous trees (red circles) based on modern leaf-level measurements (Maire et al., 2015), and fossil reconstructions (green triangles and diamonds). Error bars on fossil-derived data indicate ±1σ, box-and-whisker plots indicate median, first and third quartile (Q1 and Q3), and 95% confidence interval of modern leaves of canopy trees. Individual datapoints are randomized on the x-axis for a clearer depiction of the distribution. (b) Conductance to water ($G_w$) from modern evergreen (Ev) and deciduous (Dec) trees (Maire et al., 2015) and fossils (EM) from different latitudes. The shaded red and green areas indicate the Q1–Q3 range of modern evergreen and deciduous trees, respectively, and the dashed lines indicate the overall linear trend with latitude. Text in panel is the Q1–Q3 range for each group, grouped in 5° latitude bins. (c) Total annual carbon flux per unit leaf area ($A_{tot}$) from modern evergreen (Ev) and deciduous (Dec) trees (Maire et al., 2015) and fossils (EM) from different latitudes. The shaded red and green areas indicate the Q1–Q3 range of modern evergreen and deciduous trees, respectively, and the dashed lines indicate the overall exponential trend with latitude. Text in panel is the Q1–Q3 range for each group, grouped in 5° latitude bins.

**Data availability.** All raw measurement data on fossil leaves generated for this paper is available in the online supplementary information. Raw measurements on fossil leaves from Ethiopia (Tesfamichael et al., 2017) and Panama (Londoño et al., 2018), δ18O measurements (Zachos et al., 2001), and iWUE, $G_w$, $A_{tot}$.
and $A_h$ measurements on modern plants (Maire et al., 2015) are available through the cited original works.