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Sensitivity of plants to changing atmospheric CO₂ concentration: from the geological past to the next century

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Summary

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The rate of CO₂ assimilation by plants is directly influenced by the concentration of CO₂ in the atmosphere, c_a . As an environmental variable, c_a also has a unique global and historic significance. Although relatively stable and uniform in the short term, global c_a has varied substantially on the timescale of thousands to millions of years, and currently is increasing at seemingly an unprecedented rate. This may exert profound impacts on both climate and plant function. Here we utilise extensive datasets and models to develop an integrated, multi-scale assessment of the impact of changing c_a on plant carbon dioxide uptake and water use. We find that, overall, the sensitivity of plants to rising or falling c_a is qualitatively similar across all scales considered. It is characterised by an adaptive feedback response that tends to maintain $1 - c_i/c_a$, the relative gradient for CO₂ diffusion into the leaf, relatively constant. This is achieved through predictable adjustments to stomatal anatomy and chloroplast biochemistry. Importantly, the long-term response to changing c_a can be described by simple equations rooted in the formulation of more commonly studied short-term responses.

I. Introduction

More than two centuries after the precise experiments by de Saussure (1804) confirmed the dependence of photosynthesis on atmospheric CO₂, important questions remain about the fundamental relationship between atmospheric CO₂ concentration (c_a), plant carbon uptake, and water use. As c_a has continued to increase in recent decades researchers have sought to predict the long-term impacts on plant growth. This pursuit is increasingly allied with understanding how large fluctuations of c_a in the past might have shaped present-day plant functional diversity (Ehleringer *et al.*, 2005).

Based on the steep positive response of CO₂ assimilation rate to c_a in short-term assays, early expectations were of greatly enhanced growth under elevated c_a (Radmer & Kok, 1977; Witter, 1979). It later became apparent that in the long term, increases in CO₂ assimilation rate and growth were likely to be more moderate due to various feedback responses and constraints (Kramer, 1981). The primary goal for many decades has been to understand the nature of these feedbacks at different scales within different vegetation systems in an effort to predict the general behaviour of plants under changing c_a . Several comprehensive reviews have advanced our knowledge in this area (Bowes, 1993; Ceulemans & Mousseau, 1994; Idso & Idso, 1994; Curtis, 1996; Drake *et al.*, 1997; Saxe *et al.*, 1998; Norby *et al.*, 1999; Amthor, 2001; Ainsworth & Rogers, 2007; Gerhart & Ward, 2010).

For any given set of environmental conditions, the rate of exchange of carbon dioxide for water vapour at the leaf surface is governed largely by the diffusive conductance of the epidermis (Farquhar & Sharkey, 1982), as determined by the density, size and dynamic properties of stomata. Of the many environmental signals to which stomata respond (Willmer & Fricker, 1996), the short-term sensitivity of stomata to CO₂ concentration has long been known to scientists. In the late 19th century Francis Darwin observed 'pure CO₂ slowly closes the stomata' and 'when illuminated, stomata remain open in the absence of CO₂' (Darwin, 1898). More than a century later it was demonstrated that prolonged exposure of plants to different CO₂ concentrations could induce permanent changes in stomatal density (Woodward, 1987). This fundamental sensitivity at the anatomical level, which translates to a shift in maximum leaf diffusive conductance, has now been observed across several geological time scales, suggesting a close coupling between long-term changes in c_a and the evolution of leaf gas exchange capacity (van der Burgh *et al.*, 1993; van de Water *et al.*, 1994; Wagner *et al.*, 1996; McElwain *et al.*, 1999; Royer *et al.*, 2001; Beerling *et al.*, 2002; Retallack, 2002; Roth-Nebelsick & Konrad, 2003; Franks & Beerling, 2009; de Boer *et al.*, 2011; Lammertsma *et al.*, 2011). With significant progress in the refinement of model genetic systems for analysis of stomatal development, and an ever increasing number of sequenced plant genomes, the opportunity now exists for an improved evolutionary perspective on stomatal developmental sensitivity to c_a (Vatén & Bergmann, 2012). Along with genomic diversity, recent evidence suggests that the evolution of plant genome size *per se* could be a central factor in the historic correlations between c_a and stomatal traits (Franks *et al.*, 2012a).

Here we evaluate the latest information on the sensitivity of plant carbon dioxide uptake and water use to sustained changes in c_a . These fundamental and intimately connected physiological responses are examined from two perspectives: time, ranging from decades to hundreds of millions of years; and magnitude of c_a , ranging from pre-industrial lows of *c.* 200 ppm, to > 10 times this, as occurred in ancient atmospheres. We present a concise synthesis and interpretation of the emerging pattern of historic trends in c_a , the capacity for CO₂ assimilation, and leaf diffusive conductance, and provide a basic, quantitative framework for characterising these trends.

This review is timely for several reasons. First, the maturation of several long-term forest tree Free Air CO₂ Enrichment (FACE) experiments (e.g. Aspen FACE (Darbah *et al.*, 2010), Duke FACE (Ellsworth *et al.*, 2012), and Oak Ridge FACE (Norby *et al.*, 2010), has allowed the effects of sustained changes in c_a on vegetation systems to be more fully assessed. In their design these long-term studies better account for transient effects associated with the imposition of a step change in c_a on established (Körner *et al.*, 2005) or newly developing systems (Ellsworth *et al.*, 1995; Uddling *et al.*, 2009). Second, there is an increasing need for better integration of observational data and the theory of underlying mechanisms of plant response to varying c_a over more extensive timescales, including the deep geological past. Third, land surface models used to simulate hydrological and climatic processes under different CO₂ forcing scenarios rely crucially upon accurate accounting of stomatal sensitivity to CO₂ (Betts *et al.*, 1997, 2007; Sellers *et al.*, 1997; Gedney *et al.*, 2006). These models have traditionally utilised simple empirical parameterisations of stomatal sensitivity developed for short-term responses (Collatz *et al.*, 1991). However, it is not known whether these short-term parameterisations adequately describe longer-term responses that may be modified by resource constraints, developmental processes or adaptation.

II. Atmospheric CO₂ concentrations through time

1. Direct measurements: 1958 to present

The longest continuous record of direct measurement of c_a is that from the Mauna Loa Observatory, Hawaii, which commenced in 1958 (Keeling *et al.*, 1976). Even after a few years it was clear that c_a was increasing, and Keeling *et al.* (1976) attributed this to industrial CO₂ emissions. This trend has continued to the present day. Over the last decade c_a has increased at the rate of *c.* 2 ppm yr⁻¹, and at the end of 2011 the estimated global annual mean was *c.* 391 ppm (Conway & Tans, 2012). The trend is not restricted to CO₂. Carbonyl sulphide (abbreviated COS, chemical formula OCS), for example, is an atmospheric trace gas with strong greenhouse properties (Ueno *et al.*, 2009). During photosynthesis, COS co-diffuses with CO₂ into leaves at a ratio of *c.* 1.4 pmol COS per μ mol CO₂ (Stimler *et al.*, 2010), making it a potentially important tracer for large-scale gross primary productivity estimates (Goldan *et al.*, 1988; Campbell *et al.*, 2008). Concentrations of COS have been increasing in a similar fashion to CO₂ since industrialisation, and this too has been attributed to industrial emissions (Montzka *et al.*, 2007; Aydin *et al.*, 2008).

2. Reconstructions: ice cores to geochemical models

The c_a record before 1958 comprises a number of indirect measurements and proxy estimates (Fig. 1). The uncertainty of these measures tends to increase with increasing time before present. Measurements of c_a in air occluded in ancient ice sheets extending in age to 800 000 yr are the most precise. The composition of the air in the bubbles trapped within the ice, formed over a period of several years as firn changed to ice, is assumed to closely reflect the local atmospheric composition at the time and to have remained unchanged until extraction for measurement (Oeschger *et al.*, 1984). These measurements have a precision of ± 5 –6 ppmv (Oeschger *et al.*, 1984; Neftel *et al.*, 1985). It should be noted that, due to the time taken for the transition from firn to ice, the trapped CO_2 in any bubble has an age distribution, rather than a discrete age. This also means that the amplitude of fast changes in atmospheric CO_2 concentration is attenuated in the signal extracted from ice cores (Spahni *et al.*, 2003). Estimates of c_a for periods older than the oldest ice sheets rely upon various proxies and biogeochemical models (Royer, 2006). These estimates have considerable uncertainties attached (Berner & Kothavala, 2001; Royer, 2006). As an overview for the Phanerozoic Eon, spanning the last *c.* 542 million yr (Ma),

the geochemical model GEOCARBSULF (Berner, 2006, 2008) is one of the most widely adopted, providing estimates of c_a for 10 Ma intervals (Fig. 1a).

Overall the evidence indicates that c_a has varied substantially through geological time. This point was first realised in the late nineteenth century by Ball (1879) and ran counter to the opinion of many at the time, including that of the foremost geologist Charles Lyell. After estimating the enormous volume of carbon in the Carboniferous coal measures, ‘all of which must have been extracted from the atmosphere’, Ball (1879) reasoned ‘...before the deposition of the [Carboniferous] coal measures, the atmosphere contained twenty times as much carbonic acid gas... than it does at present’. This is within the realm of current estimates of c_a for the early Palaeozoic Era (Berner, 2008; Fig. 1). Since the Carboniferous Period the fluctuations in c_a have been more moderate, with the concentration perhaps exceeding 1000 ppm in the mid Cretaceous Period *c.* 100 Ma before present, then trending steadily down through most of the Cenozoic Era (Hansen & Wallmann, 2003; Berner, 2008; Fig. 1a). For much of the last million years c_a has oscillated between 172 and 300 ppm and includes nine glacial periods in which c_a remained in the lower part of this range for tens of thousands of years (Luthi *et al.*, 2008).

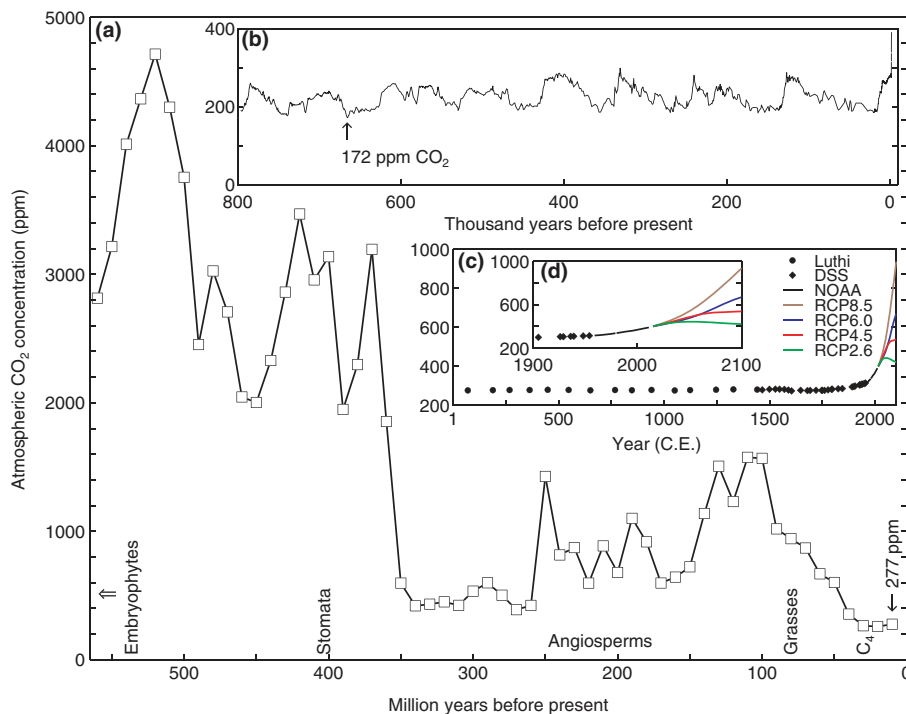


Fig. 1 Atmospheric CO_2 concentration (c_a) through time. (a) From 560 to 10 million yr ago simulated by the GEOCARBSULF model (Berner, 2008; parameterised for basalt/granite weatherability = 2, $\text{NV} = 0.015$, $\text{fB}0 = 5$) connecting discrete values (open squares) with straight lines. Also indicated are proposed timings of appearances of embryophytes and angiosperms (815–568 and 240–175 million yr ago, respectively; Clarke *et al.*, 2011), stomata (Edwards *et al.*, 1998), grasses (Strömberg, 2011) and C_4 plants (Sage *et al.*, 2012). (b) High-resolution atmospheric CO_2 concentration values from Antarctic ice cores as compiled by Luthi *et al.* (2008) for the period 800 000–1000 yr ago and from the DSS Antarctic ice core for the period 1006–1954 C.E. (Etheridge *et al.*, 1996). That ice core record is extended with annual mean measured *in situ* values at the Mauna Loa Observatory (1959–1979; Tans & Keeling, 2012) and globally averaged marine surface annual mean measurement data (1980–2011; Conway & Tans, 2012). Discrete values are connected with straight lines. The arrow indicates the lowest CO_2 concentration reported from an ice core measurement (171.6 ppm). (c) Measured values since year 1 C.E. as given in (b) but with ice core values shown as discrete points and with the Mauna Loa and global marine surface time series jointly labeled ‘NOAA’. The measured time series is continued (extrapolated) with the four ‘representative concentration pathways’ (RCPs) scenarios RCP2.6, RCP4.5, RCP6.0 and RCP8.5 (Moss *et al.*, 2010), for the period 2015–2100 C.E. (d) Values for the period 1900–2100 C.E. from (c), but at greater resolution.

3. The future: scenarios, not predictions

There are currently no firm predictions for the course of c_a over the remainder of this century. This uncertainty stems in large part from the unpredictable time course of future CO₂ emissions, dependent on unknown future economic, political and social factors. Thus, for climate modelling purposes the approach has been to model effects across a range of different feasible greenhouse gas emission scenarios. In 2001 the Intergovernmental Panel on Climate Change (IPCC) published several possible future trends for c_a , modelled from 1980 through to 2100, based on the IPCC Special Report on Emissions Scenarios (SRES; IPCC, 2001). These 'what if' trends are distinct from predictions because each is generated from a carbon cycle model using rates of CO₂ emission based on prescribed (but intrinsically unknowable) sets of future economic and political circumstances (IPCC, 2000). The IPCC Fourth Assessment Report on climate change adopted a core group of greenhouse gas emissions scenarios from SRES for use in climate simulations, but reiterated that 'no likelihood has been attached to any of the SRES scenarios' (IPCC, 2007a). The c_a resulting from these scenarios ranged between *c.* 550 and 850 ppm by the year 2100. It should be noted that the carbon cycle models used in generating these possible future c_a trends also contained additional hypothetical assumptions about rates of terrestrial system response and oceanic uptake of atmospheric CO₂ (Prentice *et al.*, 2001). The IPCC Fifth Assessment Report, due in 2013/14, will utilise a different approach to generate a set of possible future c_a trends for this century. Four 'Representative Concentration Pathways' (RCPs) have been devised from different emission trajectories (Moss *et al.*, 2010; Vuuren *et al.*, 2011) resulting in different intensities of radiative forcing by the year 2100 (2.6, 4.5, 6.0 and 8.5 W m⁻²) that correspond to a range of c_a between 421 and 936 ppm by the year 2100 (Fig. 1c,d).

At its current rate of increase of *c.* 2 ppm per year, c_a will exceed 500 ppm by the year 2067, but with rates of CO₂ emissions still steadily increasing (Peters *et al.*, 2012) it seems almost inevitable that c_a will exceed that value sooner. In the latest International Energy Statistics database compiled by the U.S. Energy Information Administration (EIA, 2011), the 2008–2035 reference case projects a doubling of CO₂ emissions from both China and India (largely due to power station construction) that together would comprise 38% of projected 2035 CO₂ emissions. As a guide, therefore, values of c_a between 500 and 1000 ppm should be considered as realistic experimental treatments or conditions for studying plant behaviour in the context of atmospheric CO₂ concentrations for the end of this century.

III. Plant sensitivity to CO₂ at geological timescales

1. The biochemical and anatomical apparatus

In all green plants the photosynthetic apparatus comprising the chloroplast appears to function much the same as it did in cyanobacteria approximately 2.3–3.5 billion yr ago (Blankenship, 1992; Farquhar *et al.*, 2011). However, recent molecular evidence suggests that the kinetic properties of the chloroplast enzyme

Rubisco have undergone substantial selection during periods of decreasing c_a over the last 1.5 billion yr (Young *et al.*, 2012). At the leaf and canopy scale, net CO₂ assimilation rate (A_n) on a per-unit area basis comprises a number of structural and compositional properties including leaf diffusive conductance (as determined by stomata and mesophyll arrangement), leaf area index and other gross morphological properties such as leaf shape and orientation, leaf surface properties and branching pattern (Smith *et al.*, 2004). All of these properties, together with those of the chloroplast itself, are potentially adaptable as part of the response of A_n to c_a .

2. The basic leaf-level CO₂ feedback loop

An immediate effect of a change in atmospheric carbon dioxide concentration is a change in the rate of carboxylation by Rubisco (in C₃ plants) or PEP carboxylase (in C₄ plants), each a crucial limiting step in the enzymatic reactions of these respective pathways of photosynthesis. This translates to an initial increase in A_n , albeit less pronounced in C₄ plants at ambient c_a and above because of their unique CO₂ concentrating mechanism (von Caemmerer & Furbank, 1999). The final response depends on the magnitude and direction of change in c_a , but at every scale there are negative feedbacks that counteract the initial effect of the change in c_a (Farquhar *et al.*, 1978; Stitt, 1991; Diaz *et al.*, 1993; Norby *et al.*, 2010). In both C₃ and C₄ plants, a fundamental component of this feedback response is the control of leaf diffusive (stomatal) conductance to CO₂ and water vapour. Following the initial change in c_a away from ambient, stomatal conductance seemingly adjusts back towards a value that will optimise carbon gain with respect to water loss (Cowan & Farquhar, 1977; Dubbe *et al.*, 1978; Farquhar *et al.*, 1978; Medlyn *et al.*, 2011). This involves reducing stomatal conductance under elevated c_a , or increasing it under reduced c_a , as widely observed (Ainsworth & Rogers, 2007). The short-term stomatal response to c_a is a core physiological trait across all vascular plant lineages (Mansfield & Willmer, 1969; Morison & Gifford, 1983; Brodribb *et al.*, 2009; Ruzsala *et al.*, 2011). It has been suggested (Gedney *et al.*, 2006) that over recent decades this feedback response to rising c_a at the landscape scale has been sufficient to suppress plant water loss via transpiration and increase river runoff.

3. Fossils and the adaptive feedback response

Many factors have contributed to the evolution and diversification of plants, but selection by environment has undoubtedly played a crucial role (Knoll & Niklas, 1987; Chapin *et al.*, 1993; Raven, 2002). Evolution of planate leaves as atmospheric CO₂ concentration fell during the Devonian Period (Beerling *et al.*, 2001) and the rise of C₄ photosynthesis in the increasingly arid and CO₂-starved late Cenozoic Era (Ehleringer *et al.*, 1991; Ehleringer, 2005; Osborne & Sack, 2012; Sage *et al.*, 2012) are striking examples of selection pressure exerted on leaf functional traits by large and sustained changes in c_a . Certainty about the timing and phylogenetic positioning of many of the formative steps in plant evolution is improving as a result of new molecular tools and approaches (Clarke *et al.*, 2011). Increasingly, it is the analysis of

the remains of fossilised plants that is revealing the underlying functional traits upon which selection has acted.

Over 400 Ma of evolution, plant adaptation to c_a could be viewed as one of adaptive feedback, involving both positive and negative feedback elements. The evolution of stomata, as well as rudimentary xylem and a primitive root system in the Silurian Period, coincident as they were with decreasing atmospheric CO_2 concentration, set in train a self-reinforcing evolutionary feedback loop that promoted an explosion in plant stature, coupled with transformation of the soil, hydrological and climatic environments (Algeo & Scheckler, 1998; Berry *et al.*, 2010).

Large changes in c_a through geological time correlate with major functional changes in the plant gas exchange and water transport (hydraulic) systems (Boyce *et al.*, 2009; Franks & Beerling, 2009; Brodribb & Feild, 2010; de Boer *et al.*, 2012). One of the most widely observed responses is the generally negative correlation between stomatal density and c_a in the fossil record (van der Burgh *et al.*, 1993; van de Water *et al.*, 1994; Royer, 2001; Lammertsma *et al.*, 2011). This pattern is also seen in the developmental response of stomatal density to experimental manipulation of c_a (Woodward & Kelly, 1995; Royer, 2001; Ainsworth & Rogers, 2007). There are exceptions to the overall negative trend in stomatal density with increasing c_a (e.g. Reid *et al.*, 2003), and the reason for this is not clear. Nevertheless, across fossil studies and CO_2 experiments there is an average reduction of 2–4% in stomatal density per 100 ppm increase in c_a (Franks *et al.*, 2012b). The tendency for stomatal density to decrease with increasing c_a appears to be part of a broader pattern of adaptation of plant gas exchange capacity to changing c_a . Fossils reveal that, across many different plant clades, coordinated changes in stomatal density and stomatal size over tens of millions of years altered the maximum leaf diffusive conductance in the direction that counteracted the effects of falling c_a (Fig. 2). In similar fashion to the short-term feedback adjustment of stomatal conductance to c_a (Farquhar *et al.*, 1978; Santrucek & Sage, 1996), this pattern is consistent with an adaptive negative feedback system of plant gas exchange control that operates across developmental and evolutionary timescales, moving maximum stomatal conductance towards a value that improves carbon gain with respect to water loss (Franks *et al.*, 2012b). The sensitivity or gain of the adaptive feedback process depends on the spatial and temporal scale of observation as well as the phylogenetic structure and history of the system. Overall, however, fossils reveal that the pattern of short-term, leaf-level physiological response of stomata to a change in c_a is reiterated over geological timescales via evolutionary changes in stomatal and leaf hydraulic traits. The full mechanism for this has yet to be determined, but its expression will be constrained by intrinsic physical, genomic and genetic limits on the plasticity of stomatal density and stomatal size (Franks *et al.*, 2009, 2012a; Vatén & Bergmann, 2012).

IV. Plant sensitivity to CO_2 over the last 200 yr

1. Increased growth?

As a direct result of increasing c_a , particularly in recent decades, forests may have absorbed and sequestered carbon (as woody

growth) at an increasing rate (LaMarche *et al.*, 1984; Hari & Arovaara, 1988). However, forest carbon cycles are complex and other contributing factors make it difficult to test this hypothesis, especially at the global scale (Malhi & Grace, 2000; Gedalof & Berg, 2010). Significant increases in the rate of forest carbon accumulation (i.e. ‘increasing biomass increment’) for sites across the USA and Europe have in many instances been attributed to land

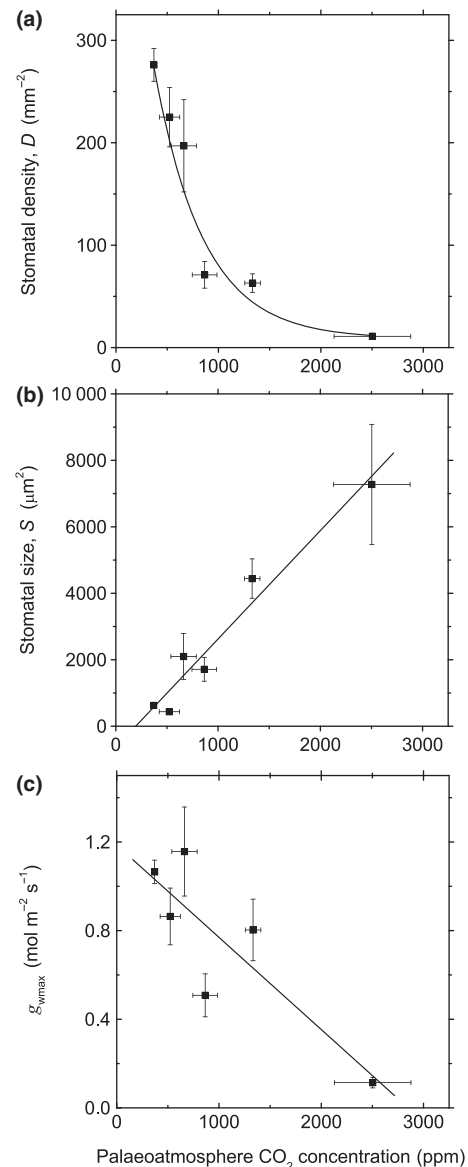


Fig. 2 Stomatal response to atmospheric CO_2 concentration over geological time, as inferred from stomatal dimensions in fossils and using the GEOCARBSULF model for palaeoatmosphere CO_2 concentration over the Phanerozoic Eon (Fig. 1; Berner, 2008). Data are means \pm SE for 50- or 100 Ma time slices, beginning at 400 Ma before present. (a) Stomatal density, D , is negatively correlated with palaeoatmosphere CO_2 concentration; (b) stomatal size, S , is positively correlated with palaeoatmosphere CO_2 concentration; (c) maximum diffusive (stomatal) conductance of the leaf epidermis to water vapour, $g_{w(\text{max})}$, derived from D and S , is negatively correlated with palaeoatmosphere CO_2 concentration. Adapted from Franks & Beerling (2009).

use change and recovery from degradation (Schimel *et al.*, 2000; Ciais *et al.*, 2008; Kauppi *et al.*, 2009; Hoover, 2011). Nonetheless, there is mounting evidence to suggest that, across a range of forest types, increasing c_a has contributed to increased rates of tree growth over the past two centuries (Graybill & Idso, 1993; Cannell *et al.*, 1998; Baker *et al.*, 2004; Voelker *et al.*, 2006; Martinez-Vilalta *et al.*, 2008; Lewis *et al.*, 2009; Cole *et al.*, 2010; Bellassen *et al.*, 2011).

If increasing c_a has contributed to accelerated tree growth in some ecosystems the question then becomes, can this be sustained? The world's forests have an immense capacity for sequestering increasing amounts of atmospheric CO₂ (Luyssaert *et al.*, 2008; Pan *et al.*, 2011) and models have predicted that future increases in c_a will enhance forest growth (Melillo *et al.*, 1993; Lloyd, 1999; Scheiter & Higgins, 2009). But there must be an upper limit to the biomass that any given forest can hold (Phillips *et al.*, 1998), and the CO₂ 'fertilisation effect' will diminish as this limit is approached. It is difficult to say when this might occur, but Canadell *et al.* (2007) have suggested that the strength of the large terrestrial carbon sink in the northern hemisphere and tropics, of which forests are a key component, could be considerably diminished by the end of this century. Long-term CO₂-enrichment experiments with forests also point to the possible slowing-down of the CO₂ fertilisation effect in the long term, highlighting nutrient availability as a significant constraint (see section V Plant sensitivity to long-term experimental manipulation of CO₂). In tropical forests the carbon sink strength could diminish more quickly if elevated c_a stimulates the growth of woody vines, which are known to enhance tree mortality and suppress tree growth (Phillips *et al.*, 2002).

2. Increased water-use efficiency?

Evaporative water loss from leaves during the uptake of CO₂ is unavoidable, so when water is limiting it is advantageous for plants to use it more 'efficiently'. Plant water-use efficiency (WUE) at any time during photosynthesis is typically defined as the ratio of the rates of net CO₂ assimilation to transpiration, A_n/E (Farquhar *et al.*, 1989). The greater the WUE, the more carbon is fixed per unit water lost, thereby improving the chance of survival in drier environments or under drought. Because c_a directly influences A_n , global changes in c_a will have some influence on WUE.

WUE may be written in terms of the ratio of concentrations (mole fraction) of CO₂ and water vapour on either side of the epidermis as (Farquhar *et al.*, 1989):

$$\frac{A_n}{E} = \frac{c_a \left(1 - \frac{c_i}{c_a}\right)}{1.6v} \quad \text{Eqn 1}$$

where c_i is the CO₂ concentration inside the leaf and v is the water vapour pressure difference between the saturated air in the leaf intercellular air spaces and the atmosphere. The discrimination against ¹³C during photosynthesis can be expressed in its simplest form, in terms of the ratio c_i/c_a , as (Farquhar *et al.*, 1989):

$$\Delta^{13}\text{C} = a + (b - a) \frac{c_i}{c_a} \quad \text{Eqn 2}$$

where a and b are fractionations occurring during diffusion through stomata and during carboxylation (as modified by mesophyll conductance), respectively. Eqns 1 and 2 may be combined to give WUE in terms of carbon isotope discrimination:

$$\frac{A_n}{E} = \frac{c_a \left(1 - \frac{\Delta^{13}\text{C} - a}{b - a}\right)}{1.6v} \quad \text{Eqn 3}$$

Eqn 3 reduces to

$$\frac{A_n}{g_w} = \frac{c_a \left(1 - \frac{\Delta^{13}\text{C} - a}{b - a}\right)}{1.6} \quad \text{Eqn 4}$$

where g_w is the leaf diffusive (stomatal) conductance to water vapour. The quantity A_n/g_w is often referred to as the 'physiological' or 'intrinsic' water-use efficiency (Feng, 1999) because it provides an indication of potential WUE based on its physiological components; that is, it excludes the direct influence of v , which is determined mainly by environmental conditions.

Caution should be used when comparing different measurements of A_n/g_w in the context of potential WUE, particularly across different species. This is because v also has an indirect effect on WUE via its role in the stomatal feedback control mechanism that determines g_w (Cowan, 1977; Franks & Farquhar, 1999; Buckley *et al.*, 2003; Franks, 2004). Higher v , for example, tends to reduce g_w and therefore E , increasing WUE. If A_n/g_w in one plant measured at high v is similar to that of another plant measured at low v , then the second plant is likely to have a higher inherent WUE than the first plant despite exhibiting the same A_n/g_w . Ideally, when considering A_n/g_w as an indicator of WUE, comparisons should be made only where it can be assumed that v is constant. It should also be noted that Eqn 2 is a simplified description of the isotopic effects occurring during photosynthesis. Leaf internal conductance to CO₂ (or 'mesophyll conductance', g_m) can impart a significant influence on $\Delta^{13}\text{C}$ (Flexas *et al.*, 2008). Although g_m is known to vary significantly across species and environments, usually in correlation with photosynthetic capacity (von Caemmerer & Evans, 1991; Warren *et al.*, 2003; Ethier & Livingston, 2004), the short-term and long-term effects of c_a on g_m remain unclear, with reports of both rapid adjustment of g_m to c_a (Flexas *et al.*, 2007) and no response (Tazoe *et al.*, 2009). The method of estimating A_n/g_w here will need to be reassessed when a stronger mechanistic understanding of g_m has been developed.

The stable carbon isotope composition of tree rings provides a valuable record of plant response to the environment over time (Francey & Farquhar, 1982). Numerous studies over the past few decades have constructed historic time sequences of A_n/g_w using Eqn 4 and wood $\Delta^{13}\text{C}$ from tree ring chronologies (McCarroll & Loader, 2004; Gagen *et al.*, 2011). Most of these studies have indicated a significant increase in average growing season A_n/g_w as atmospheric CO₂ concentration increased from the year 1900 to present. A recent study by Peñuelas *et al.* (2011) assessed changes in

A_n/g_w and stem growth between the early 1960s and the early 2000s for 47 tree ring studies around the world. Over the 40-yr period, they found that A_n/g_w increased by 20.5%, on average, while stem growth showed no significant change. As trees increase in height there are numerous structural and environmental changes that may result in increased A_n/g_w and decreased $\Delta^{13}C$ (Lloyd *et al.*, 2010; McDowell *et al.*, 2011b). These height-dependent changes can be reflected in a less negative $\delta^{13}C$ for the wood being laid down in older trees, this then giving rise to a potential ‘age effect’ (McCarroll & Loader, 2004). This ‘age effect’ could potentially confound the above interpretation of a widespread CO_2 mediated increase in A_n/g_w over recent decades as most dendro-isotope studies have not encompassed trees of many different ages and analysed them individually. This is required to allow any ontogenetic effects to be effectively removed from the longer-term climate change signal (McCarroll & Loader, 2004). Nevertheless, from a simple consideration of measured gradients in $\Delta^{13}C$ with height (Lloyd *et al.*, 2010; McDowell *et al.*, 2011b) and known height/dbh

allometries (e.g. Feldpausch *et al.*, 2011) it seems unlikely that this effect could account for all of the apparent changes in A_n/g_w shown in Fig. 3, especially for the most recent decades.

Using the same approach, we have calculated A_n/g_w from tree ring $\Delta^{13}C$ reported in a range of studies that have examined tree rings spanning approximately the last 200 yr. The results (Fig. 3) show little change in A_n/g_w between the late 1800s and 1920s ($4 \pm 2\%$ increase on average; not significantly different from zero) but there is an overall $16.9 \pm 2\%$ increase in A_n/g_w between 1920 and 1960, and (for the subset of the Peñuelas *et al.* (2011) data assessed here) an overall $17.9 \pm 2\%$ increase in A_n/g_w between 1960 and 2000. The same trend of increasing A_n/g_w was observed across each of the biomes sampled in this assessment, including boreal, high elevation, temperate, Mediterranean and tropical forests. If v at these study sites has not increased systematically in this time (and at a global scale there is little evidence to indicate it has; Roderick & Farquhar, 2002), then tree WUE has been increasing over the past 200 yr.

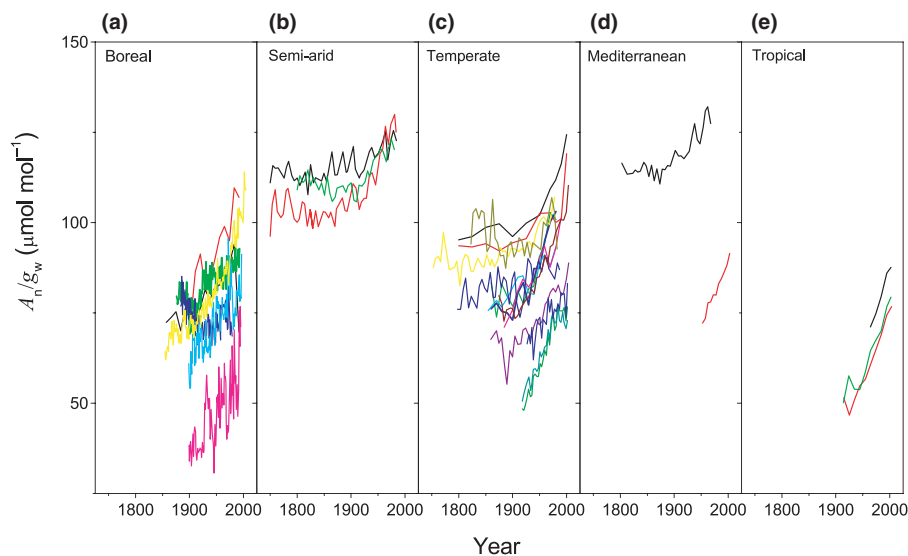


Fig. 3 The change in the ratio of net CO_2 assimilation rate to stomatal conductance, A_n/g_w , estimated from $\delta^{13}C$ of tree rings ($\delta^{13}C_p$), between 1750 and the present day for five climatic zones. Assuming there has not been a systematic increase in the evaporation potential in this time (Roderick & Farquhar, 2002) then the trend of increasing A_n/g_w indicates tree water-use efficiency is increasing. A_n/g_w was estimated by first calculating the atmospheric CO_2 concentration (c_a) and its $\delta^{13}C_a$ from the relationships $c_a = 282.23 + (\exp(\text{year}/51.35) \times 1.03 \times 10^{-15})$ and $\delta^{13}C_a$ (in per mil) = $-6.40 - (0.004 \times \exp(0.0197 \times (\text{year} - 1695.06)))$, then ^{13}C discrimination $\Delta^{13}C_p$ (in per mil) = $(\delta^{13}C_a - \delta^{13}C_p)/(1 + \delta^{13}C_p)$. A_n/g_w was then calculated from Eqn 4 using $a = 4.4$ and $b = 27$ (Francey & Farquhar, 1982; Farquhar *et al.*, 1989). Data are from published studies. (a) Boreal forest: black, *Pinus sylvestris*, Turuhansk, central Siberia (Arneth *et al.*, 2002); red, *Pinus sylvestris*, Krasnoyarsk, central Siberia (Arneth *et al.*, 2002); green, *Pinus sylvestris*, northern Finland (Berninger *et al.*, 2000); blue, *Picea sitchensis*, Alaska, (Stuiver *et al.*, 1984); cyan, *Larix gmelinii*, Taimyr Peninsula, northern Siberia (Sidorova *et al.*, 2010); magenta, *Larix gmelinii*, north-eastern Yakutia, northern Siberia (Sidorova *et al.*, 2010); yellow, *Larix gmelinii*, central Siberia (Sidorova *et al.*, 2009). (b) Semi-arid: all from Leavitt & Long (1986); black, *Pinus edulis*, Alton Utah; red, *Pinus edulis*, Dry Canyon Colorado; green, *Pinus edulis*, Cerro Colorado New Mexico. (c) Temperate: black, *Pinus nigra*, Iberian Peninsula (Andreu-Hayles *et al.*, 2011); red, *Pinus uncinata*, Iberian Peninsula (Andreu-Hayles *et al.*, 2011); green, *Abies alba*, selection forest, Jura Mountains, eastern France (Bert *et al.*, 1997); blue, *Abies alba*, even-aged forest, Jura Mountains, eastern France (Bert *et al.*, 1997); cyan, *Fagus sylvatica* coppiced, north-eastern France (Duquesnay *et al.*, 1998); magenta, *Fagus sylvatica* tall trees, north-eastern France (Duquesnay *et al.*, 1998); yellow, *Pinus ponderosa*, Prewitt, California (Leavitt & Long, 1988); dark yellow, *Pinus coulteri*, San Dimas Mountains, California (Feng & Epstein, 1995); purple, *Araucaria angustifolia*, grassland, Southern Brazil (Silva *et al.*, 2010); wine, *Araucaria angustifolia*, forest, Southern Brazil (Silva *et al.*, 2010); olive, central elevation, dark cyan, high elevation, and royal blue, low elevation, all *Fagus sylvatica*, Montseny Mountains, north-east Spain (Peñuelas *et al.*, 2008). (d) Mediterranean: black, *Juniperus phoenicea*, Sinai, (Epstein *et al.*, 1990); red, *Abies pinsapo*, southern Spain (Linares *et al.*, 2009). (e) Tropical: black, *Melia azedarach*, red, *Toona ciliata*, green, *Chukrasia tabularis*, all west-central Thailand (Nock *et al.*, 2011).

V. Plant sensitivity to long-term experimental manipulation of CO₂

1. Overview of the trend in long-term CO₂ enrichment experiments

Several decades of research with plants grown under elevated and sub-ambient c_a treatments have built an increasingly clear picture of the relationship between CO₂ assimilation rate and the c_a at which plants grow (see reviews listed in section I Introduction). Because some experimental results potentially contain artefacts of methodology (e.g. pot effects, pulsed vs continuous nutrient supply), the overall picture has improved as the number of studies has increased. Here we have compiled an updated summary of the relative response of net CO₂ assimilation rate (A_n) across a broad range of c_a , from *c.* 150 to 2500 ppm (Fig. 4). This includes data from plants grown in pots in climate-controlled chambers or in CO₂-enhanced natural systems where c_a was controlled at specified concentrations. The survey covers a wide range of growth forms, from grasses and herbs to forest trees (see data and sources in Supporting Information Table S1). For each treatment c_a , we report the relative net CO₂ assimilation rate, $A_{n(\text{rel})}$, calculated as the ratio of light-saturated A_n in plants grown and measured at the treatment c_a , relative to A_n in plants grown and measured at ambient c_a . Overall, plants grown and measured at elevated c_a exhibit higher A_n , and those grown and measured at sub-ambient c_a exhibit lower A_n , relative to plants growing and measured under ambient c_a . This relationship appears to be nonlinear, with a greater rate of change in $A_{n(\text{rel})}$ at sub-ambient c_a compared to elevated c_a .

2. An equation for long-term CO₂ assimilation vs c_a ?

Do long-term CO₂ manipulation experiments reveal an adaptive relationship between A_n and c_a ? There is at present no generally accepted model for predicting long-term $A_{n(\text{rel})}$ in response to c_a ,

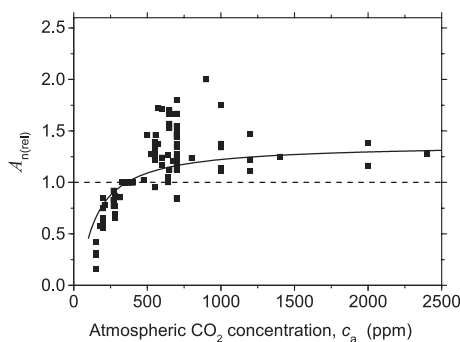


Fig. 4 Relative net CO₂ assimilation rate, $A_{n(\text{rel})}$, vs the atmospheric CO₂ concentration in which plants were grown (all C₃). $A_{n(\text{rel})}$ is calculated as the ratio of the assimilation rate in plants grown and measured at the growth CO₂ concentration, relative to that in plants grown and measured at ambient atmospheric CO₂ concentration. Data (black squares) are from numerous independent studies and surveys (see Table S1). The black line is the predicted relationship for long-term C₃ plant adaptation according to the hypothesis of optimal chloroplast nitrogen allocation under limiting nitrogen, using Eqn 5 with $c_{a0} = 360$ ppm and $\Gamma^* = 40$ ppm (see text for details).

partly because of the complexity of other mitigating ecological factors that can influence the relationship. Earlier studies, characterised by resource-abundant and/or rapidly developing systems, suggested that $A_{n(\text{rel})}$ could be sustained at much greater rates than what now appears to be the case, especially for natural systems (Körner, 2006). The study by Aoki & Yabuki (1977) with potted cucumber (*Cucumis sativus*) was one of the first to highlight the potentially unsustainable transient characteristics of a system exposed to a step increase in c_a . Their study showed a two-fold enhancement of $A_{n(\text{rel})}$ in the initial resource-abundant phase, but this response dissipated within weeks to a more modest enhancement in the resource-limited phase. More extensive, long-term studies on various plant systems, including perennial herbs (Tissue & Oechel, 1987) and forests (Körner *et al.*, 2005; Norby *et al.*, 2010) have reported qualitatively similar transient characteristics.

Recognising that plants have been equipped with most, if not all, of the fundamental physiological characteristics governing A_n (e.g. stomata, chloroplasts, leaves, roots, hydraulic systems) for at least 370 Ma (Kenrick & Crane, 1997; Kenrick *et al.*, 2012), and that c_a has fluctuated five- to ten-fold its current ambient concentration over the same period (and perhaps more; Berner, 2006), it is possible, even likely, that a generalised long-term A_n vs c_a relationship evolved early in the history of vascular plants. Given the absence, so far, of a method to directly quantify A_n in long extinct vegetation, the description of such a relationship must be speculative.

For the purpose of discussion, however, we consider the hypothetical case in which, over sufficiently long periods of time in a given system, net CO₂ assimilation rate (A_n) tends towards the condition of optimal nitrogen allocation. Nitrogen is a limiting resource in most natural systems (Vitousek & Howarth, 1991). In the Farquhar-von Caemmerer-Berry C₃ photosynthesis model (Farquhar *et al.*, 1980), $A_n = \min(W_e, W_c)$, where W_e is the light-limited (or ribulose 1,5-bisphosphate (RuBP) regeneration-limited) rate and W_c is the Rubisco capacity-limited rate. Strictly this holds only for $c_a > \Gamma^*$, where Γ^* is the CO₂ compensation point in the absence of dark respiration. In a nitrogen-limited system the distribution of protein to these co-limiting photosynthetic processes will be optimal when $W_c = W_e$ at the average incident light intensity. Solar luminosity has not changed by more than *c.* 2% since the origin of vascular plants (Bahcall *et al.*, 2001), so, except for catastrophic events, it can be assumed that photosynthesis-saturating irradiance has not changed appreciably either. Accordingly, the expression for W_e may be used to describe the relative change in A_n with long-term changes in c_a :

$$A_{n(\text{rel})} \approx \left[\frac{(c_a - \Gamma^*)(c_{a0} + 2\Gamma^*)}{(c_a + 2\Gamma^*)(c_{a0} - \Gamma^*)} \right] \quad \text{Eqn 5}$$

where c_{a0} is the reference ambient atmospheric CO₂ concentration. Although Γ^* is influenced by leaf temperature (Farquhar *et al.*, 1980) there is evidence to indicate that despite widely varying seasonal and latitudinal temperatures, much of the photosynthetic productivity of plants occurs within a relatively narrow band of

assimilation rate-weighted temperature ranging from *c.* 19°C in boreal systems through to 26°C in tropical systems (Helliker & Richter, 2008; Song *et al.*, 2011). Therefore, we assume a mean leaf temperature during photosynthesis of 25°C, giving Γ^* a mean value of 40 $\mu\text{mol mol}^{-1}$ as a first approximation. Using this parameterisation for Eqn 5 we have plotted $A_{n(\text{rel})}$ vs long-term c_a in Fig. 4 (solid line). This long-term adaptation model closely resembles the trend in the data compilation from many independent studies in which c_a was experimentally manipulated (black symbols in Fig. 4). Eqn 5 may therefore be a useful basis for reconstructing $A_{n(\text{rel})}$ for times in the geological past where c_a has been determined, or for future estimates of $A_{n(\text{rel})}$ based on c_a projections or scenarios.

3. The ecosystem approach: forest FACE studies

Long-term CO₂ enrichment of forests using FACE has provided insights into plant sensitivity to elevated atmospheric CO₂ concentration at the ecosystem scale. These studies, some running for over a decade, have yielded crucial quantitative information and challenged some initial assumptions (Norby & Zak, 2011).

Early CO₂ enrichment experiments with tree seedlings and saplings demonstrated that stomatal conductance is generally reduced in response to elevated CO₂ (mean reduction of 21%; Medlyn *et al.*, 2001). Meta-analyses of FACE experiments reported somewhat similar reductions of 16–19% (Ainsworth & Long, 2005; Ainsworth & Rogers, 2007). Persistent reductions in stomatal conductance in a forest canopy in a CO₂-enriched future atmosphere would increase the probability of reductions in whole-canopy water use (transpiration), and increases in soil moisture and water yield. In turn, these changes could increase drought resistance and influence many other secondary responses that are governed by plant or soil water status (Wullschlegel *et al.*, 2002). However,

increases in forest stand leaf area index (LAI, or total leaf area per unit ground area) of forest stands in a CO₂-enriched atmosphere could counteract leaf-level reductions in water use (Warren *et al.*, 2011). Furthermore, at the scale of whole forest canopies and growing seasons, relationships between stomatal conductance and atmospheric CO₂ concentration are confounded by many other factors. Understanding the connectivity between leaf-level physiological responses and higher-order ecological consequences of those responses was a primary rationale for long-term CO₂ enrichment experiments (Norby & Zak, 2011).

Most long-term experiments of CO₂ enrichment of forests have included measurements of both stomatal conductance and transpiration rate, permitting an analysis of the relationship between these variables and c_a (Fig. 5). Most of the data conform to the general pattern of reduced stomatal conductance in elevated CO₂ (7–19% reduction after normalisation to an elevated atmospheric CO₂ concentration of 550 ppm). While many of the individual differences were not statistically significant, Keel *et al.* (2007) argued that small, CO₂-induced decreases in stomatal conductance at the leaf level, although hard to measure and document, are nevertheless sufficient to cause cumulative increases in soil water content in the area exposed to elevated CO₂ concentration. An exception to the general response of a reduction in stomatal conductance under elevated CO₂ concentration came from the Aspen FACE experiment (Uddling *et al.*, 2009). Here, stomatal conductance in trees was 19% greater in elevated CO₂, although measurements earlier in the experiment when the trees were young and the canopy was still expanding had shown reduced stomatal conductance in elevated CO₂ (Noormets *et al.*, 2001). Uddling *et al.* (2008, 2009) suggest that increased leaf-level stomatal conductance and whole-tree sap flux in the Aspen FACE experiment could be attributed to increased hydraulic efficiency caused by long-term cumulative effects on plant and

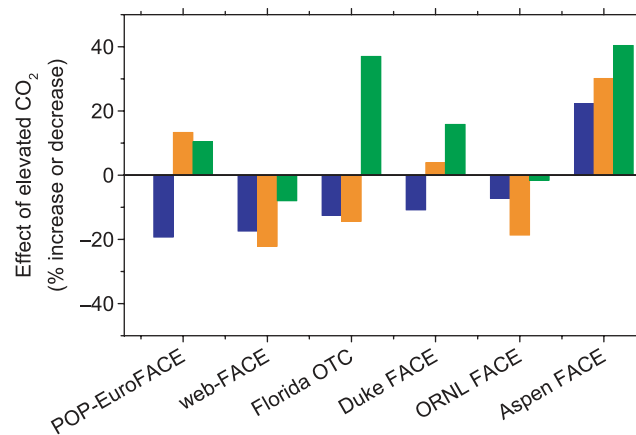


Fig. 5 Effect of elevated atmospheric CO₂ concentration on stomatal conductance (from leaf-level porometer measurements; blue bars), canopy transpiration (from sap flux measurements; orange bars), and leaf area index (LAI; green bars) in long-term Free Air CO₂ Enrichment (FACE) experiments in forests. Data were extracted from published reports from the latest year in which conductance and transpiration both were measured. Reported values were normalized to a CO₂ enrichment of 180 ppm (corresponding to a treatment c_a of *c.* 550 ppm) assuming a linear response to CO₂ enrichment over this range. Actual experimental enrichments during the year of measurement ranged from 151 to 198 ppm, except for the Florida OTC site (+350 ppm). Data sources: POPEuroFACE, *P. x euroamericana* in 2000 (Gielen *et al.*, 2003; Tricker *et al.*, 2009); webFACE, mixed deciduous species in 2005 (Keel *et al.*, 2007; Leuzinger & Körner, 2007); Florida OTC, *Quercus myrtifolia* in 2000 (Li *et al.*, 2003); Duke FACE, *Pinus taeda* in 1996 (conductance) and 1998–2000 (transpiration; Ellsworth, 1999; Schafer *et al.*, 2002); http://public.ornl.gov/face/npp_synthesis.shtml); ORNL FACE, *Liquidambar styraciflua* in 2008, (Warren *et al.*, 2010); J. M. Warren, unpublished data; <http://public.ornl.gov/face/ORNL/>); Aspen FACE, pure *Populus tremuloides* stands in ambient ozone, 2005 (Uddling *et al.*, 2008, 2009).

stand structure, including increased tree size, LAI and fine-root biomass.

The response of transpiration to elevated CO_2 was similar to that of stomatal conductance in three of the six experiments shown in Fig. 5, but there were substantial differences in both magnitude and direction of response in the other three. The relationship between stomatal conductance and transpiration rate under elevated c_a is explained in part by changes in LAI, which were small in forests with high LAI (e.g. Web-FACE and ORNL FACE) but significantly greater in forests with lower LAI (Norby & Zak, 2011). Increased LAI under elevated CO_2 concentration in the POP-EuroFACE and Duke FACE experiments led to increased whole-tree water use (sap flux) despite reduced leaf-level stomatal conductance, but the large increase in LAI in the scrub oak ecosystem of the Florida OTC experiment did not increase water use. Li *et al.* (2003) argued that increased LAI in the scrub oak ecosystem indirectly reduced transpiration via self-shading of the lower canopy which reduced the amount of energy to drive transpiration.

Although partial stomatal closure may be a robust, primary response to elevated CO_2 , other biological and site factors can serve to modify its extent. Furthermore, the stomatal conductance response itself is not always an accurate predictor of stand-level transpiration and higher-order responses of water budgets to elevated CO_2 . Given the importance of water to ecosystem function, more formal ecohydrological modelling approaches (*sensu* Warren *et al.*, 2010) will be needed to evaluate the integrated hydrological response of forests to elevated CO_2 . The effect is likely to be different between regions depending on whether incoming energy or water is limiting (Farquhar, 1997; Roderick & Farquhar, 2011).

4. Drought mitigating effects of elevated CO_2

Some assessments of global trends in drought severity have concluded that, globally, droughts have become more intense and longer in duration (Dai *et al.*, 2004; IPCC, 2007a,b). These conclusions were based on the Palmer Drought Severity Index (PDSI), a supply-vs-demand model for soil moisture that typically uses air temperature to calculate potential evaporation (E_p). However, E_p , which characterises the 'demand' side of the PDSI, is more strongly affected by factors other than air temperature, such as changes in surface radiation, wind speed and humidity deficit (Roderick *et al.*, 2007). Therefore, the PDSI should be parameterised using information that more completely describes E_p , such as near-surface meteorological data or pan evaporation data (Hobbins *et al.*, 2008). Using a soil moisture-based drought index derived from a land surface model forced by precipitation and near-surface meteorological data, Sheffield & Wood (2008) concluded that there is a predominantly decreasing global trend in drought duration, severity and intensity, but noted also that statistically significant changes are limited to < 10% of continental areas. For the United States, using the Standardized Precipitation Index (SPI) calculated from precipitation records spanning years 1895–2007, Chen *et al.* (2012) concluded that there was no significant change in drought conditions overall, but that drought intensity had

increased in the eastern United States. Overall, it seems that there is insufficient evidence for a globally consistent trend in drought severity and duration.

The effects of elevated atmospheric CO_2 concentration on plants under drought are complex. As a defence against tissue water deficit, plants reduce transpirational water loss by closing stomata, but this substantially reduces net CO_2 assimilation rate. However, elevated c_a enhances net CO_2 assimilation rate in plants with C_3 photosynthesis, which includes most forest trees and food crops. On balance, if the photosynthesis-stimulating effect of elevated c_a is great enough it may cancel out the reduction in photosynthesis from drought-induced stomatal closure. When drought is severe enough to induce complete stomatal closure, then no direct benefit of elevated CO_2 on photosynthesis can be manifest.

Drought-stressed plants grown under elevated c_a maintained higher CO_2 assimilation rates than drought-stressed plants at ambient c_a (Fig. 6). In large-scale studies, the relative photosynthetic benefits of elevated CO_2 are generally greater in more arid environments compared to temperate, due at least in part to water savings through stomatal closure (Morgan *et al.*, 2004). Elevated c_a can affect stomatal conductance indirectly by altering the leaf or xylem hydraulic conductance, an important component of the stomatal feedback control system. In a stand of loblolly pine (*Pinus taeda*) grown under elevated c_a , Domec *et al.* (2009) reported lower leaf hydraulic conductance and greater diurnal loss of hydraulic conductance compared to ambient controls, which they concluded as contributing to a reduction in stomatal conductance.

In cases where leaf area increases in response to elevated CO_2 , there may be no soil water savings at the ecosystem scale; subsequently, drought impacts may be similar under ambient and elevated CO_2 . Further, increased leaf area per tree may result in lower stomatal conductance and photosynthesis during drought, and increase the risk of leaf-level hydraulic failure and subsequent canopy loss (Warren *et al.*, 2011). Prolonged suppression of CO_2 assimilation under drought may require plants to utilise stored

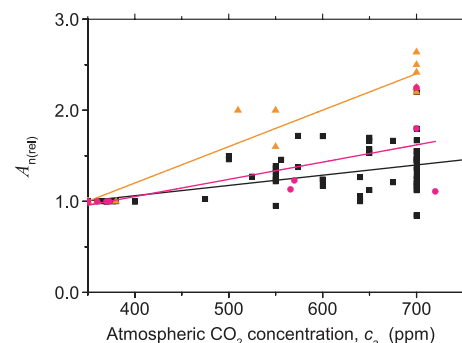


Fig. 6 Drought mitigation by elevated atmospheric CO_2 concentration, c_a (squares, C_3 plants, no drought; triangles, C_3 plants, drought; circles C_4 plants, drought). Drought reduces stomatal conductance and CO_2 assimilation rate, A_n , and elevated CO_2 concentration tends to increase A_n . At lower c_a , the effect of drought is more pronounced than at higher c_a . Therefore, the increase in relative net CO_2 assimilation rate, $A_{n(\text{rel})}$ (see Fig. 4 for definition of A_n) with c_a is higher in droughted plants than in nondroughted plants. Plants with C_4 photosynthesis are less affected by drought and elevated c_a , so the overall increase in $A_{n(\text{rel})}$ under the combined effects of drought and elevated c_a is less in C_4 plants than in C_3 plants. Data are from several independent studies (see Table S2).

carbohydrates to maintain metabolism, enhancing the chances of survival under prolonged drought (McDowell *et al.*, 2011a). Drought may also overwhelm the benefits of elevated c_a in certain biomes. In a study focusing on northern hemisphere mid- and high-latitude forests, tree ring records show that under increasing c_a growth has been enhanced in only 20% of forests, indicating that other factors, including drought, have counteracted the potential benefits of higher c_a (Gedalof & Berg, 2010). In Amazonian tropical forests tree growth has been stimulated by increasing c_a (Phillips *et al.*, 1998) but this could be reversed if drought in these forests becomes more prevalent this century, as some studies predict (Phillips *et al.*, 2009). If drought frequency and/or magnitude do not increase, then tropical forests are likely to continue as carbon sinks despite projected increases in leaf temperature (Lloyd & Farquhar, 2008) (but see section IV Plant sensitivity to CO₂ over the last 200 yr). In summary, it is likely that increasing c_a may benefit plant gas exchange and survival during droughts of moderate duration, but where droughts become more severe the negative effects may overwhelm the benefits of increasing c_a .

VI. Simple formulation of stomatal conductance in land surface models for simulating long-term CO₂ response

Global climate models (GCMs) and land surface models (LSMs) are used together to model the interactions of climate and landscape, for example the response of vegetation to perturbations in climate, or the response of climate to forcing factors such as elevated c_a . An important element in these exercises is the inclusion of climate–vegetation feedbacks. In LSMs the short-term response of vegetation to climate, or ‘physiological response’, is characterised principally by equations calculating net CO₂ assimilation rate (A_n) and stomatal conductance to CO₂ and water vapour (g_c , g_w , where g_w is *c.* 1.6 g_c). The equations used are based on leaf-level physiological observations, but scaled to the canopy level (Sellers *et al.*, 1992). Accurate estimates of canopy conductance are essential for realistic estimation of the carbon, hydrological and energy cycles in GCMs (Sellers *et al.*, 1997).

Over the last two decades, two leading community LSMs have emerged: (1) the National Center for Atmospheric Research (NCAR) model, which began as LSM1 (Bonan, 1996) and, via the Common Land Model (CLM1) (Dai *et al.*, 2003) is now the Community Land Model (CLM4; Oleson *et al.*, 2010); (2) The UK Meteorological Office MOSES model (Cox *et al.*, 1999), now the Joint UK Land Environment Simulator, JULES (Best *et al.*, 2011; Clark *et al.*, 2011). For the modelling of stomatal conductance, these and other LSMs (e.g. SDGVM (Woodward *et al.*, 1995); ORCHIDEE (Krinner *et al.*, 2005); JSBACH (Raddatz *et al.*, 2007)) draw an approach first enshrined within the Ball-Woodrow-Berry model (Ball *et al.*, 1987; Collatz *et al.*, 1991) where, contrary to earlier attempts that predicted g_w from environmental drivers such as light, temperature and humidity (Jarvis, 1976), g_w is predicted using the equations for photosynthesis (Berry *et al.*, 2010; Berry, 2012). In developing the Ball-Woodrow-Berry model advantage was taken of a recently established close and apparently almost inevitable covariance

between g_w and A_n (Wong *et al.*, 1979). This was combined with a second empirical result, namely that even when the ambient c_a varied, for any given leaf the relative gradient for CO₂ diffusion into the leaf, $1 - c_l/c_a$ (with c_l/c_a as in section IV Plant sensitivity to CO₂ over the last 200 yr) remains remarkably constant (Wong *et al.*, 1978; Wong *et al.*, 1979). Because $1 - c_l/c_a \approx 1.6 A_n/(g_w c_a)$ (Farquhar & Sharkey, 1982) this also implies that

$$g_w = \text{Constant} \times \frac{A_n}{c_a} \quad \text{Eqn 6}$$

Then, through taking into account that stomata also tend to close in response to variations in atmospheric humidity, the following ‘Ball-Woodrow-Berry’ equation was developed (Ball *et al.*, 1987; Berry *et al.*, 2010):

$$g_w = m \frac{A_n h_s}{c_s} + k \quad \text{Eqn 7}$$

where h_s is relative humidity at the leaf surface, c_s is CO₂ concentration at the leaf surface, and m and k are the slope and intercept which define the linear relationship between g_w and $A_n h_s/c_s$. A slightly modified version of Eqn 7 is used in CLM4, where A_n is replaced by the gross CO₂ assimilation rate, A_G (i.e. excluding foliar respiration). This modification was introduced to avoid numerical problems in the model simulations as it prevents k (effectively the minimum stomatal conductance) becoming negative at night when $A_n \leq 0$. Nevertheless, there are alternative ways to circumvent such problems (e.g. Lloyd *et al.*, 2002), and Bonan *et al.* (2011) have recently suggested that A_n (as in Eqn 7) rather than A_G should be used in the formulation of g_w in the NCAR model along with revised photosynthetic parameters.

Arguing that stomata respond to the leaf-to-air water vapour pressure difference rather than relative humidity *per se*, Leuning (1995) presented a slightly different formulation, written here in terms of g_w :

$$g_w = m' \frac{A_n}{(c_s - \Gamma)(1 + v_s/v_0)} + k' \quad \text{Eqn 8}$$

where m' and k' are again slope and intercept parameters, Γ is the CO₂ compensation point (including respiration in the light), v_s is the humidity deficit at the leaf surface or the water vapour pressure difference between the saturated air in the leaf intercellular air spaces and the leaf surface (section IV), and v_0 is an empirical coefficient which controls the sensitivity of stomata to humidity deficit (effectively defining the humidity deficit at which stomatal conductance assumes half its maximum value). Note that Γ is typically invariant at any given temperature and also typically much less than c_s (Farquhar, 1989), so when v_0 is equal to the water vapour pressure of the ambient air, Eqns 7 and 8 are virtually identical. The JULES land surface model adopts a simplified version of Eqn 8 which effectively solves for c_l/c_a using biome-specific values for v_0 (Best *et al.*, 2011).

Both stomatal models (Eqns 7, 8) are of similar form and their broad agreement with measurements has facilitated their wide

acceptance and incorporation into LSMs (Berry *et al.*, 1997). Therefore, we investigate how well these models simulate changes or adaptation in g_w in response to long-term manipulation of c_a . Taking Eqn 7, we assume $k \ll g_w$ and that average h_s is steady through multiple years or longer time periods (Allen & Ingram, 2002; Ingram, 2002). We also assume that c_s is approximately c_a due to boundary layer conductance being substantially higher than g_w in most instances. Further, for C_3 plants m is likely to remain constant as c_a varies over the long term, as suggested in a synthesis of long-term studies by Medlyn *et al.* (2001). With these simplifications Eqn 7 reduces to Eqn 6, where now the 'constant' comprises mb_s . To compare the qualitative effects of different c_a environments on g_w across a range of plants or vegetation systems, Eqn 6 may be reduced to

$$g_{w(\text{rel})} \approx \frac{A_{n(\text{rel})}}{c_{a(\text{rel})}} \quad \text{Eqn 9}$$

where $g_{w(\text{rel})}$, $A_{n(\text{rel})}$ and $c_{a(\text{rel})}$ are, respectively, g_w , A_n and c_a relative to the value in a similar system at constant current ambient c_a (noting that in this case the term mb_s is constant and cancels out of the expression).

This simple representation of $g_{w(\text{rel})}$ is plotted in Fig. 7(a) (dashed line) together with data from numerous experiments in which plants (including trees and crops) were grown at different c_a and measurements of g_w and A_n reported (see Table S1). The data agree closely with the model. In remarkable similarity to Wong's original formulation, the slope and intercept of a regression of measured $g_{w(\text{rel})}$ on $A_{n(\text{rel})}/c_{a(\text{rel})}$ (solid line) are not significantly different from the 1 : 1 line represented by Eqn 9 (dashed line). Long-term adaptation of A_n to c_a (Fig. 4) appears to follow a saturating form similar to the response to short-term manipulation of c_a (von Caemmerer & Farquhar, 1981), so there is an accompanying decline in $g_{w(\text{rel})}$ with increasing growth c_a (Fig. 7b). The variance in the data here is partially due to the wide range of species and slightly different protocols under which g_w and A_n were measured across the various independent studies. The high level of agreement between Eqn 9 and the data in Fig. 7(a) indicates that plants adapt to long-term changes in c_a in a manner that is consistent with their short-term responses, maintaining $1 - c_i/c_a$ relatively constant (i.e. within a narrow range).

Interestingly, a recent model of stomatal behaviour under short-term environmental changes (Medlyn *et al.*, 2011), which links back to the idea that stomata act to optimise the marginal water cost of daily carbon gain (Cowan & Farquhar, 1977), also reduces to Eqn 6 if changes in direct effects of atmospheric humidity are ignored. Consistent with Fig. 4, Medlyn *et al.* (2011) find also that stomata optimise for RuBP regeneration-limited carbon gain. Thus, it seems that g_w sensitivity to long-term changes in c_a can be parameterised simply in quantitative terms using Eqn 6 or qualitatively using Eqn 9. Although developed primarily to simulate diel and annual variations in g_w , these basic empirical models may constitute a convenient tool for use in LSMs to simulate the response of vegetation to c_a throughout Earth history and into the next century.

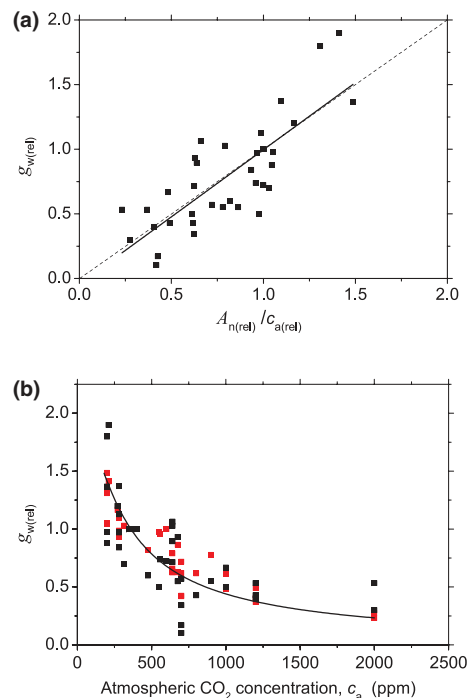


Fig. 7 Stomatal conductance predicted from photosynthesis. (a) Stomatal conductance to water vapour measured at any given growth c_a , relative to the conductance at the reference growth c_a , $g_{w(\text{rel})}$, correlates one-to-one with the ratio of relative net assimilation rate divided by relative growth c_a , $A_{n(\text{rel})}/c_{a(\text{rel})}$. The data are from numerous independent studies of plants grown at different c_a , from as low as 150 ppm (sub-ambient) to as high as 2000 ppm (super-elevated), and represent the normal operating conditions in those experimental systems (see Table S1). The slope and intercept of the unconstrained regression (solid line, $r^2 = 0.65$, $P = 1.0 \times 10^{-12}$) are not significantly different from the 1 : 1 relationship (dashed line) represented by Eqn 9. (b) For a range of plants grown under different atmospheric CO_2 concentrations the stomatal conductance under normal operating conditions at the growth c_a declines with increasing growth c_a . Black symbols are directly measured $g_{w(\text{rel})}$ data from numerous studies (as in panel a). From the same studies, the reported A_n and c_a values were converted to relative values of $A_{n(\text{rel})}$ and $c_{a(\text{rel})}$, then used to calculate $g_{w(\text{rel})}$ from Eqn 9 (red symbols). The line is $g_{w(\text{rel})}$ generated from Eqn 9 using prescribed values of c_a , where $c_{a(\text{rel})} = c_a/c_{a0}$, c_{a0} is 360 ppm, and $A_{n(\text{rel})}$ is obtained from Eqn 5 as per the line in Fig. 4.

VII. Conclusions

Across a range of timescales, leaf gas exchange responses to changing atmospheric CO_2 concentrations (c_a) have been surprisingly similar, even though the relative contributions of underlying mechanisms differ (such as changes in stomatal aperture vs changes in stomatal density and size). The observed response is typically consistent with negative feedback optimisation of RuBP regeneration-limited carbon gain with respect to water loss under limiting nitrogen availability. The feedback loops operate in different timeframes and with changeable or adaptable gain characteristics, so over developmental and evolutionary (geological) time the response to long-term changes in c_a is better described as adaptive negative feedback control. One example of this is found in the fossil record with good evidence of adaptations in maximum leaf

diffusive conductance in relation to changing c_a over the past *c.* 400 Ma.

Analysis of isotope chronologies in tree rings indicate a widespread increase in tree water-use efficiency over the last 200 yr, consistent with suggestions of increased continental river runoff in the twentieth century and with land surface simulations that incorporate the leaf-level feedback response to c_a . Vegetation systems, particularly those comprising C_3 plants, may exhibit a greater relative increase in net CO_2 assimilation rate in response to increasing c_a under drought compared to non-drought conditions. Long-term CO_2 enrichment experiments reveal potentially strong feedbacks on the initial response of CO_2 assimilation and growth rates, mediated by the rate of nutrient supply, particularly nitrogen. Over the full range of c_a considered here, the relationship between relative CO_2 assimilation rate (referenced to that in systems at ambient c_a) and the prevailing c_a at which plants grow is consistent with that predicted by optimal allocation of protein to the co-limiting biochemical components of carbon fixation in the chloroplast. This, in combination with the basic empirical models for stomatal response to c_a (Eqns 6–9) it is hoped may serve as a template for describing adaptation and evolution of plant photosynthetic capacity in response to, or under selection by, c_a .

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Supporting Information

Additional supporting information may be found in the online version of this article.

Table S1 Species, growth CO₂ concentration, relative CO₂ assimilation rate ($A_{n(\text{rel})}$), relative stomatal conductance to water vapour ($g_{w(\text{rel})}$), and source publication for data in Figs 4 and 7

Table S2 Species, growth CO₂ concentration, relative CO₂ assimilation rate ($A_{n(\text{rel})}$) and source publication for data in Fig. 6 (drought conditions)

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