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# Spatial variability and temporal trends in water-use efficiency of European forests

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## Abstract

The increasing carbon dioxide ( $CO_2$ ) concentration in the atmosphere in combination with climatic changes throughout the last century are likely to have had a profound effect on the physiology of trees: altering the carbon and water fluxes passing through the stomatal pores. However, the magnitude and spatial patterns of such changes in natural forests remain highly uncertain. Here, stable carbon isotope ratios from a network of 35 tree-ring sites located across Europe are investigated to determine the intrinsic water-use efficiency (iWUE), the ratio of photosynthesis to stomatal conductance from 1901 to 2000. The results were compared with simulations of a dynamic vegetation model (LPX-Bern 1.0) that integrates numerous ecosystem and land-atmosphere exchange processes in a theoretical framework. The spatial pattern of tree-ring derived iWUE of the investigated conferous and deciduous species and the model results agreed significantly with a clear south-to-north gradient, as well as a general increase in iWUE over the 20th century. The magnitude of the iWUE increase was not spatially uniform, with the strongest increase observed and modelled for temperate forests in Central Europe, a region where summer soil-water availability decreased over the last century. We were able to demonstrate that the combined effects of increasing  $CO_2$  and climate change leading to soil drying have resulted in an accelerated increase in iWUE. These findings will help to reduce uncertainties in the land surface schemes of global climate models, where vegetation–climate feedbacks are currently still poorly constrained by observational data.

Keywords: carbon isotope discrimination, climate change, dynamic vegetation model, tree rings

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#### Introduction

Interactions among direct  $CO_2$  (fertilization) effects on plants and climatic conditions such as drought are of

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particular interest for understanding past and for predicting future forest growth and carbon sequestration. The continually increasing atmospheric CO<sub>2</sub> concentration and concurrent climatic change are both likely to strongly affect the physiology of forests ecosystems and alter productivity, species distribution and vegetationclimate feedbacks. Yet, both the magnitude and mechanisms of forest response are unclear and furthermore are expected to be spatially very heterogeneous, depending at least upon both the local growth limitations (Babst et al., 2013) and the trajectory of future climate (IPCC, 2013). For many regions, ecosystem transitions are already underway (Parmesan & Yohe, 2003; Allen et al., 2010). The effects of climate change and increasing CO<sub>2</sub> concentrations could stimulate tree growth in some regions via enhanced photosynthesis through CO<sub>2</sub> fertilization (Ainsworth & Long, 2005). However, adverse effects from an increase in drought severity, for example, could result in reduced growth and increased stress and mortality (Zhao & Running, 2010). Such changes will inevitably also modify biospheric CO<sub>2</sub> and water fluxes and the relationships between them (Schimel et al., 2001; Keenan et al., 2013).

The slow adaption of late-successional forests, however, is difficult to assess either by observations or by experiments. Therefore, it is not well known how the physiology of natural forests has already changed due to the increase in atmospheric CO<sub>2</sub> concentration in the ca. 150 years since major global industrialisation. Increases in net ecosystem productivity inferred from small-scale CO<sub>2</sub> fumigation or depletion experiments may overestimate the CO<sub>2</sub>-response in natural forests (Norby et al., 2010). Down-regulation of photosynthesis under elevated CO<sub>2</sub> was observed and also the reduction in transpiration was found to be relatively small in a mature mixed deciduous forest (Leuzinger & Körner, 2007). Accurate quantification of changes in water and  $CO_2$  fluxes over the last century, which are spatially highly variable, would be important because of their relationship with the carbon sequestration potential of the forests (Schimel et al., 2001; Pan et al., 2011), and evaporation-temperature feedbacks, i.e. possibly additional or reduced warming due to changes in evapotranspiration (Betts et al., 1997).

Stable carbon isotope ratios ( $\delta^{13}$ C) of tree rings represent a valuable tool to improve understanding of forest response to the combined influence of climate and CO<sub>2</sub> over time (McCarroll & Loader, 2004).  $\delta^{13}$ C in plant organic matter is related to the ratio of net photosynthesis (A) to stomatal conductance to water (g), which is the intrinsic water-use efficiency (iWUE = A/g), and therefore provides a measure for the relative water loss per molecule carbon acquired at the leaf level (Farquhar *et al.*, 1982). Plant water-use efficiency (WUE) defined as the ratio of carbon uptake to actual water loss at the plant level is an essential element of the survival and productivity of plants. Actual WUE depends on the evaporative demand, as transpiration is determined by the product of stomatal conductance and vapour pressure deficit, while iWUE may be considered as a potential WUE and does not consider this variable environmental constraint and respiratory losses (Seibt et al., 2008). This limitation of the isotope approach may be overcome by using plant physiological models to assist data interpretation. For assessments that strive to consider both spatial and temporal variability in plant-climatic-CO<sub>2</sub> interactions, a dynamic vegetation model such as the Lund-Potsdam-Jena (LPJ) model that combines process-based vegetation dynamics with land-atmosphere carbon and water exchange is particularly useful (Sitch et al., 2003). Such models have been widely applied to study the influence of increasing CO<sub>2</sub> on net primary productivity changes (Hickler et al., 2008). Few studies have discussed possible changes in WUE under variable climate and CO<sub>2</sub> concentrations, and those that do generally find increasing WUE over time (Tian et al., 2010; De Kauwe et al., 2013); however, no long-term verification of model results with carbon isotope data of tree rings has been carried out. The uncertainty of this forest-climate feedback is therefore very high.

Previous tree-ring carbon isotope studies have identified an increase in iWUE over the 20th century at most forest sites so far investigated across the globe, including sites from the tropics (Hietz et al., 2005; Loader et al., 2011), northern high- and midlatitudes (Saurer et al., 2004; Waterhouse et al., 2004; Gagen et al., 2011) and the Mediterranean basin (Linares et al., 2009; Andreu-Hayles et al., 2011), but to varying degrees. Some studies indicated a passive response of the plants to increasing atmospheric  $CO_2$  concentration ( $c_a$ ), where the intercellular  $CO_2$  concentration ( $c_i$ ) increased by the same amount as  $c_a$  (implying  $c_a - c_i = \text{constant}$ ), which resulted in no iWUE improvement (Marshall & Monserud, 1996). However, more commonly an active response was observed, where  $c_i$  increased more slowly than  $c_a$  due to plant adaptation, resulting in higher iWUE (Feng, 1998). Several studies have reported that the response to increasing CO2 concentrations changed from being active  $(c_i/c_a \text{ constant})$  to passive  $(c_a - c_i \text{ con-}$ stant) around the 1970s (Waterhouse et al., 2004; Gagen et al., 2011) with an approximate levelling of iWUE, or even a decrease in polluted areas (Boettger et al., 2014). No clear spatial pattern of the iWUE increase has emerged from the published literature, partly due to the unsystematic and still sparse nature of the data collection.

In this investigation, we employ a network of tree-ring sites across Europe to study the spatial and temporal patterns in the carbon to water-use relationships. Thirty-five old-growth forest sites are investigated that cover a large climatic gradient from Mediterranean Southern Europe to the tree line Northern Fennoscandia, including both coniferous and deciduous species from five genera (*Abies, Larix, Pinus, Picea, Quercus*). The  $\delta^{13}$ C-derived changes in iWUE over the 20th century are investigated as a function of the climate change at each location (temperature, precipitation, soil-water content). The empirical data are compared to iWUE derived from a dynamic vegetation model, enabling a detailed assessment of functional relationships between physiology and climate for the entire European domain.

## Materials and methods

#### Sampling and isotope analysis

A network of 35 tree-ring sites across Europe was established within the two EU-funded projects ISONET (20 sites; Treydte *et al.*, 2007) and Millennium (15 additional sites, Table 1). The sampling design considered temperate sites, where tree growth is governed by a mixture of environmental factors, as well as ecologically extreme sites near the tree line, where a single climatic factor dominates tree growth (Treydte et al., 2007) (Table S1). The altitude range of study sites is from 5 to 2200 m, with the majority situated in two elevation bands: 0-500 m and 1500-2200 m. High-altitude sites are concentrated towards the south of the study region. We focused on oak as an important deciduous species together with various coniferous species, selecting the most abundant and long-lived trees at each location. At least four trees were sampled at all sites (usually 2 cores per tree), sufficient to develop an isotope site record representative of the population (Leavitt & Long, 1984). In general, whole rings (early- and latewood) were analysed for the conifers, whereas for oaks, latewood was separated from earlywood because a significant proportion of the carbon incorporated

**Table 1** Site information including site coordinates, site name, site code, country, species, altitude and iWUE-increase between the periods 1901–1910 and 1991–2000. Twenty chronologies indicated by a star (\*) were established during the ISONET-project (Treydte *et al.*, 2007) and 15 additional sites during the Millennium-project (see methods for details). The sites are listed from south-to-north

No.	°E	°N	Site name	Code	Country	Species	Altitude (m)	iWUE (%)
1	30.02	36.60	Elmali	Elm	Turkey	Juniperus excelsa	1937	20.5
2	-2.95	37.80	Cazorla	Caz*	Spain	Pinus nigra	1816	21.9
3	16.20	39.93	Serra di Cris.	Ser*	Italy	Pinus leucodermis	1900	20.3
4	1.70	42.23	Pedraforca	Ped*	Spain	Pinus uncinata	2100	21.5
5	-5.25	43.07	Pinar de Lillo	Lil*	Spain	Pinus sylvestris	1600	33.9
6	11.66	45.98	Trento	Tre	Italy	Picea abies	1150	29.6
7	8.60	46.35	Cavergno	Cav*	Switzerland	Quercus petraea	900	25.9
8	13.69	46.37	Veza	Vez	Slovenia	Larix decidua	1700	27.1
9	7.80	46.43	Lötschental	Loe	Switzerland	Larix decidua	2100	24.3
10	8.77	46.50	Vigera	Vig*	Switzerland	Pinus sylvestris	1400	29.5
11	13.36	47.28	Dachstein	Dac	Austria	Pinus cembra	1900	27.2
12	16.06	47.95	Poellau	Poe*	Austria	Pinus nigra	500	22.8
13	16.20	48.18	Lainzer Tierg.	Lai*	Austria	Quercus petraea	300	33.7
14	-1.70	48.25	Rennes	Ren*	France	Quercus robur	100	24.3
15	2.67	48.38	Fontainebleau	Fon*	France	Quercus petraea	100	28.3
16	11.00	49.15	Franconia	Fra	Germany	Abies alba	394	53.1
17	20.38	50.12	Niopol. Gibiel	Nie*	Poland	Quercus robur	190	30.3
18	-0.59	51.41	Windsor	Win*	UK	Pinus sylvestris	10	43.8
19	9.78	51.50	Dransfeld	Dra*	Germany	Quercus petraea	320	39.8
20	-0.59	51.98	Woburn	Wob*	UK	Quercus robur	50	29.3
21	0.50	52.83	Sandringham	San	UK	Quercus robur	56	30.2
22	22.93	54.10	Suwalki	Suw*	Poland	Pinus sylvestris	160	39.8
23	23.97	54.88	Pane. Silas	Pan*	Lithuania	Pinus sylvestris	45	42.9
24	-3.43	55.27	Lochwood	Loc	UK	Quercus robur	175	28.1
25	-5.43	57.10	Southern Upl.	Sup	UK	Pinus sylvestris	160	25.2
26	23.08	60.00	Bromarv	Bro*	Finland	Quercus robur	5	41.9
27	22.17	60.41	Turku	Tur	Finland	Quercus robur	20	44.3
28	12.18	62.00	Gutuli	Gut*	Norway	Pinus sylvestris	800	24.1
29	31.27	62.98	Ilomantsi	llo*	Finland	Pinus sylvestris	200	17.9
30	13.50	63.16	Furuberget	Fur	Sweden	Pinus sylvestris	650	10.4
31	33.15	67.41	Khibiny	Khi	Russia	Pinus sylvestris	300	19.8
32	19.72	68.22	Torneträsk	Tor	Sweden	Pinus sylvestris	400	15.7
33	27.50	68.50	Laanila	Laa	Finland	Pinus sylvestris	220	20.1
34	15.72	68.79	Forfjorddalen	For	Norway	Pinus sylvestris	110	1.0
35	28.32	68.93	Kessi, Inari	Ina*	Finland	Pinus sylvestris	150	24.1

in the earlywood is carried over in deciduous species from carbohydrate reserves established in the previous year (Treydte et al., 2007). Tree-ring samples were cross-dated to ensure the correct calendar year was assigned to each annual ring, and then individual rings separated with a razor-blade under a microscope. At some sites, tree rings from the same year were pooled prior to cellulose extraction, while at others individual trees were measured, but all analysis here is based on average site chronologies. Cellulose was extracted using standard techniques (Boettger et al., 2007). Carbon isotope analysis was conducted on CO2 obtained from combustion of the samples in an elemental-analyser and measurement in an isotope-ratio mass-spectrometer (McCarroll & Loader, 2004). Isotope values are given as δ-values calculated from the isotope ratios  ${}^{13}C/{}^{12}C$  (=R) as  $\delta^{13}$ C = ( $R_{\text{sample}}/R_{\text{standard}} - 1$ ) \* 1000% (referring to the international standard VPDB), and have a long-term estimated methodological error of <0.2% (Boettger et al., 2007). Most analyses were performed on the common period from AD 1901 to AD 2000, although some records extend up to 2006.

#### Data analysis

The isotope discrimination  $\Delta$  between plant material ( $\delta^{13}C_{plant}$ ) and atmospheric CO<sub>2</sub> ( $\delta^{13}C_{atm}$ ) is defined as  $\Delta = (\delta^{13}C_{atm} - \delta^{13}C_{atm})$  $\delta^{13}C_{\text{plant}}/(1 + \delta^{13}C_{\text{plant}}/1000)$ . According to Farquhar *et al.* (1982),  $\Delta$  can be related to plant physiological properties approximately via the linear relationship  $\Delta = a + (b - a) c_i/c_{a}$ where a is the fractionation associated with the diffusion of  $CO_2$ through the stomata ( $a = 4.4\%_{00}$ ) and b is the fractionation resulting from enzymatic C fixation (b = 27%). For the calculation of  $\Delta$  for tree rings,  $\delta^{13}$ C-values of the atmosphere ( $\delta^{13}$ C<sub>atm</sub>) through time need to be estimated from ice-core data and recent atmospheric measurements (Leuenberger, 2007). In addition, we corrected for the isotope offset between cellulose and total matter  $(\delta^{13}C_{\text{plant-corrected}} = \delta^{13}C_{\text{plant}}$ organic cellulose offset) because the leaf-level model for fractionation by Farquhar et al. (1982) is more representative for the bulk matter rather than a specific chemical compound such as cellulose. We used as offset the difference between cellulose and wood, which is  $1.1 \pm 0.2\%$  for oak (Borella *et al.*, 1998; Loader *et al.*, 2003) and  $1.3 \pm 0.2\%$  for conifers (Saurer *et al.*, 1997). This offset does, as a first approximation, consider different postphotosynthetic fractionations, including respiration, synthesis of secondary sugars, and fractionations during phloem loading and unloading (Seibt et al., 2008; Gessler et al., 2009). Based on the definition of the intrinsic water-use efficiency iWUE, the ratio of net photosynthesis  $A = g_{CO2}(c_a - c_i)$  to conductance for water vapour g<sub>H2O</sub>, we obtain  $iWUE = A/g_{H2O} = g_{CO2}$  $(c_a - c_i)/g_{H2O} = (c_a - c_i)/1.6$ , which can be calculated using  $c_i$ from above (Ehleringer & Cerling, 1995). Accordingly, the final equation for iWUE is:

$$iWUE = c_{a} \frac{b - \left[\frac{\delta^{13}C_{atm} - (\delta^{13}C_{cellulose} - offset)}{1 + \left(\frac{\delta^{13}C_{cellulose} - offset}{100}\right)}\right]}{1.6(b - a)}$$
(1)

The iWUE time series were investigated by high- and lowpass filtering using cubic smoothing splines with 50% frequency response cut-off at 20 years (Cook & Peters, 1981).

For climatological analysis, simulations and calculation of 1901–10 and 1991–2000 averages, we used the  $0.5^{\circ} \times 0.5^{\circ}$  monthly gridded temperature and precipitation CRU TS 3.1 data for 1901–2006 (Mitchell & Jones, 2005) and the Worldclim high-resolution dataset (1 km<sup>2</sup>) (Hijmans *et al.*, 2005). Climatological analyses were also carried out with soil-water content derived from the vegetation model (see below).

#### Vegetation model

For iWUE simulations, we used the Land surface Processes and eXchanges (LPX-Bern 1.0) model (Spahni et al., 2013; Stocker et al., 2013), an improved version of the Lund-Potsdam-Jena (LPJ) global dynamic vegetation model (Joos et al., 2001, 2004; Gerber et al., 2003; Sitch et al., 2003; Strassmann et al., 2008; Stocker et al., 2011). In addition to the standard LPJ version, the LPX-Bern (called LPX later in this article) model includes special modules and features important for the iWUE calculation: updated soil and plant hydrology using leaf interception, surface evaporation, snow parameterization and melting (Gerten et al., 2004; Wania et al., 2009; Murray et al., 2011), soil heat diffusion in eight soil layers regulating thawing and freezing in soil and related changes in the carbon pools (Wania et al., 2009), as well as dynamic interaction with the nitrogen cycle (Xu-Ri & Prentice, 2008; Xu-Ri et al., 2012; Stocker et al., 2013). The model is forced by monthly temperature, precipitation and cloud cover from the CRU data (CRU TS 3.1,  $0.5^{\circ}$ latitude  $\times$   $0.5^{\circ}$ longitude). The LPX uses a stochastic distribution of the monthly amount of precipitation for a prescribed number of rain days per month, as opposed to a constant amount of precipitation every day in LPJ (Sitch et al., 2003).

In LPX, the iWUE is calculated daily as the ratio of simulated net photosynthesis A to stomatal conductance g<sub>H2O</sub> for average tree individuals at the ecosystem level. Photosynthetic assimilation of CO2 is modelled using a form of the Farquhar scheme (Farquhar et al., 1980) with leaf-level optimized nitrogen allocation (Haxeltine & Prentice, 1996), and an empirical convective boundary layer parameterization to couple the carbon and water cycles (Gerten et al., 2004). The plant-available water is proportional to the soil-water content in the top 2 m and hydraulic constraints limiting tree height are not explicitly considered. Soils are subdivided in eight soil layers linked by percolation. Soil-water content is fed by precipitation (after interception by vegetation canopy) and snow melt. Beside plant transpiration, water is also removed from the soil by direct soil evaporation, percolation and runoff (Gerten et al., 2004). Transpiration is described as a function of Potential Evapotranspiration and stomatal conductance (g<sub>H2O</sub>), assuming water-saturated cell walls in the stomata. The CO<sub>2</sub> flux from the atmosphere to the stomatal cavity is proportional to the CO<sub>2</sub> difference between the atmosphere and the stomatal cavity  $(c_a - c_i)$ . Stomatal conductance for water, g<sub>H2O</sub> and carbon dioxide, g<sub>CO2</sub>, are linearly related by their molecular diffusivities  $(g_{H2O}/g_{CO2} = 1.6)$ . Stomatal conductance, and thus  $c_{i'}$  is varied to optimize assimilation for given environmental conditions, namely irradiance, temperature, water supply from soils and  $CO_2$  concentration. This optimization is performed with the constraints that the  $CO_2$  assimilation equals the  $CO_2$  flux from the atmosphere into the stomatal cavity and that transpiration does not exceed water supply. In summary, assimilation, canopy conductance and iWUE simulated by LPX depend in a nonlinear and complex way on environmental conditions.

The plant functional types used in simulations of iWUE were 'temperate and boreal needle evergreen', 'temperate broadleaf summergreen' and 'boreal needle summergreen' in LPX, from here on referred to as conifers, oak and larch, respectively. Simulations were performed using dynamic fractional plant cover with competing plant functional types. The iWUE average was weighted by the fractional tree cover (per year and grid cell) and its net primary productivity (per month and m<sup>2</sup>). For the site-by-site evaluation, the tree-ring based iWUE was compared to the modelled grid cell average iWUE at the tree site. If the model did not simulate the growth of the expected tree type, the fractional tree cover-weighted average of the eight neighbouring grid cells was used instead. The model results were further tested in sensitivity runs using constant atmospheric CO2, interactive nitrogen cycle [which used the deposition map by Lamarque et al. (2011)], prescribed vegetation type, constant climate or were forced with a high-resolution climate data set interpolated to the tree site with corrected altitude. In simulations with an interactive N cycle, there is no immediate feedback of e.g. a change in leaf N to iWUE on a daily time scale, but there is a long-term feedback by annual changes in vegetation structure and thus photosynthesis and C assimilation.

#### Results

#### Temporal trends

Tree-ring  $\delta^{13}$ C-derived iWUE calculated according to Eq. 1 shows a strongly increasing trend over the course of the 20th century when averaged over all sites (Fig. 1a). This average iWUE was 64.2  $\mu$ mol mol<sup>-1</sup> for the first decade of the 20th century and increased by 27.8% to 81.9  $\mu$ mol mol<sup>-1</sup> for the last decade of the last century (Table 2). iWUE increased between these two periods at all sites, but to a varying degree, ranging from +1.0% to +53.1% (Table 1). While starting from a lower level, oak iWUE increased more strongly (by 32.4%) compared to conifers (by 25.7%, Table 2); this difference is, however, not significantly different owing to large site-to-site variability. The average iWUE values and increase for the two larch sites was similar to the evergreen conifers. The spread of the data expressed as standard deviation between all sites increased with time from 10.7  $\mu$ mol mol<sup>-1</sup> in 1901–1910 to 13.8  $\mu$ mol mol<sup>-1</sup> in 1991–2000 (Table 2), also suggesting a nonuniform response, i.e. some sites responded more strongly than others.



**Fig. 1** (a) Temporal evolution of average tree-ring derived iWUE ('data';  $\pm$ SE of site variability as dotted lines), LPX standard model run iWUE averaged over all tree-ring sites ('model sites') and for all Europe with species represented by weight ('model Europe'), (b) the same data after high-pass filtering, and (c) low-pass-filtering with the variance scaled to 1.

The LPX standard model run derived iWUE averaged over all plant functional types increased by 21.7% between the first and last decade of the 20th century (Table 2), greater for oak (24.5%) than for conifers (20.6%) and for larch (18.7%). The values in the standard run were below the tree-ring values by about 20-25%, but showed many common features: particularly in the year-to-year, but also in the lower frequency variability, such as plateaus in the general increasing trend in the 1900s, 1950s and 1990s (Fig. 1a-c). This resulted in a highly significant correlation between model and data regarding the temporal variations ( $r^2 = 0.60$ ; P < 0.001). When averages for all Europe (instead of just grid cells with treering sites) are plotted, the agreement with the average tree-ring derived iWUE is even better ( $r^2 = 0.62$ ; P < 0.001) with values often overlapping with the tree-ring derived range of iWUE. The greater agreement with the European model runs may be fortuitous, but the similar trends of the two model curves suggest that the site selection is appropriately representative for all Europe. A correlation analysis between model and tree-ring derived iWUE was also conducted for all individual sites, yielding generally high correlations with only a few exceptions (Figure S1).

Table 2	Average iWUE values ( $\pm$ SD) for	1901-1910 and 1991-2000 and the change between these periods for individual species
and over	all sites for tree-ring derived data	[Eqn (1)], the standard LPX model run and a model run with constant CO <sub>2</sub> (the average
for 1901-	1910, i.e. 297 ppm)	

Data type	Species	iWUE 1901–1910 (µmol mol <sup>-1</sup> )	iWUE 1991–2000 (µmol mol <sup>-1</sup> )	iWUE change (%)
Tree-ring data	Conifers	67.6 ± 11.1	85.0 ± 15.5	25.7 ± 11.6
0	Larch	$65.5 \pm 0.4$	$82.4\pm1.9$	$25.7\pm2.0$
	Oak	$57.3 \pm 7.1$	$75.7\pm8.9$	$32.4\pm6.7$
	All	$64.2 \pm 10.7$	$81.9 \pm 13.8$	$27.8\pm10.3$
Model	Conifers	$54.0 \pm 15.4$	$64.6 \pm 17.0$	$20.6\pm9.3$
	Larch	$41.5 \pm 2.6$	$48.4\pm1.6$	$18.7\pm9.4$
	Oak	$55.8 \pm 12.6$	$69.2 \pm 14.7$	$24.5\pm6.7$
	All	$53.8 \pm 14.2$	$65.1 \pm 16.2$	$21.7\pm8.5$
Model constant CO <sub>2</sub>	Conifers	$53.8 \pm 15.3$	$55.2 \pm 14.8$	$3.1\pm8.0$
	Larch	$41.4\pm2.6$	$40.6 \pm 1.7$	$-1.4\pm10.4$
	Oak	$55.6 \pm 12.6$	$59.7 \pm 13.0$	$7.6\pm6.2$
	All	53.6 ± 14.2	55.8 ± 14.3	$4.3\pm7.8$

## Spatial patterns

Averages for the first and the last decade of the 20th century were also used to analyse the spatial iWUE patterns and their changes. Tree-ring derived iWUE shows a latitudinal gradient with increasing values from the North to the South for both decades, with the highest values observed in Turkey and southern Spain (Fig. 2a and b). This holds even though the results of different species are displayed together in this plot. The LPX model results (standard run without dynamic nitrogen cycle), too, show the North-South gradient and the general increase from 1901-1910 to 1991-2000 (Fig. 2c and d). Although model values are shifted to lower values as noted above, there is a qualitative agreement in the spatial patterns of tree ring and model data illustrated by these maps. The correlation between independent model and tree-ring data sets (Fig. 3a) is highly significant (1901–1910:  $r^2 = 0.26$ ; P <0.01; 1991–2000:  $r^2 = 0.30$ ; P < 0.001; combined:  $r^2 =$ 0.37, P < 0.001), which is mainly driven by a strong relationship for the evergreen conifers ( $r^2 = 0.67-0.69$ for individual and combined periods, P < 0.001), but not for oak ( $r^2 = 0.01 - 0.09$ ).

While iWUE increased at all sites, this increase, however, was not uniform (Table 1; Fig. 4). The tree-ring data show relatively low increases for the southern and northern sites, while the highest values are mostly observed at sites with relatively temperate conditions in Central Europe (Fig. 4a). The model also shows a hotspot of increased iWUE in Central Europe, and additionally in some Mediterranean subregions (Fig. 4b).

Different simulations were run to test the sensitivity of the modelled iWUE results. For example a simulation with dynamic N resulted in a better overall agreement between model and data (Fig. 3b;  $r^2 = 0.48$ , P < 0.001). We further investigated whether observed iWUE at the tree-ring sites was related to nitrogen deposition based on the map by Lamarque et al. (2011), which distinguishes between  $NO_{yy}$  NH<sub>x</sub> and total N ( $N_{tot}$ ). Current  $N_{tot}$  deposition averaged for 1991-2000 was not related to the spatial pattern of iWUE for the same period ( $r^2 = 0.05$ ). However, significant correlations were found for the increase in iWUE to NO<sub>v</sub>, NH<sub>x</sub> and  $N_{\text{tot}}$  ( $r^2 = 0.17$ , 0.18 and 0.19, respectively; P < 0.01). Using the high-spatial resolution climate data set rather than the  $0.5^{\circ} \times 0.5^{\circ}$  CRU data resulted only in marginal differences in iWUE results (not shown). Model runs with constant CO<sub>2</sub> fixed at the 1901-1910 level of 297 ppm and run with 20th century climate data show only a small iWUE-increase between 1901-1910 and 1991-2000 of 3.1% for conifers and 7.6% for oaks, and even a decrease for larch (-1.4%). Averaged for all species (weighted by representation in the model), the increase at constant CO<sub>2</sub> concentration (297 ppm) was 4.3% compared to 21.7% in the run with variable  $CO_2$  concentration (Table 2). This indicates that in the model 80.2% = [(21.7 - 4.3)/21.7] \* 100 of the increase between the investigated periods is due to the CO<sub>2</sub>-increase, and not due to climate or other factors. Also simulations with constant climate were performed: the resulting iWUE-increase when added to the results from the constant CO<sub>2</sub>-run was almost equal, but not identical (slightly higher) to the total iWUE-increase from the standard run, indicating some nonlinearity in the model. An investigation of the influence of summer temperature and precipitation amount on iWUE at interannual time scales in the model revealed that dry and warm conditions are



Fig. 2 Tree-ring derived iWUE for all sites in 1901–1910 (a) and in 1991–2000 in (b). LPX model-derived iWUE in 1901–1910 (c) and in 1991–2000 (d) combined for all tree species.

associated with high iWUE, while cold and wet summer conditions are associated with lower iWUE (not shown). The interannual analysis is consistent with the spatial patterns in WUE across the European climatic gradients (Fig. 2).

#### Climate relationships

To explain the observed spatial pattern of change in iWUE, we analysed corresponding climate patterns in Europe over the same 1901–1910 to 1991–2000 periods. The best agreement between climate and iWUE patterns was found for changes in summer (JJA) precipitation over the 20th century (Fig. 5b) as well as changes in summer soil-water content (Fig. 5c). Regions with a

summer drying trend are remarkably similar to the iWUE-change patterns of highly increasing values, both in model and empirical data (Fig. 4). In contrast, the change in the pattern for summer temperature is very different (Fig. 5a) and not congruent to the iWUE change, reflecting perhaps the indirect link between temperature and carbon isotope fractionation. This finding is further quantified by plotting the iWUE-changes vs. the soil-water changes (Fig. 6). There is a significant negative relationship between JJA soil-water changes and the tree-ring data (Fig. 6a,  $r^2 = 0.36$ , P < 0.01 for the conifers;  $r^2 = 0.27$ , P < 0.01 for all species combined; ns for oak), while no relationship was found with JJA temperature ( $r^2 = 0.00$ , all species combined). The model-derived iWUE-changes are also



**Fig. 3** Tree-ring derived iWUE (empirical data) plotted against LPX model iWUE, combining the data from 1901 to 1910 (closed symbols) and 1991 to 2000 (open symbols) by tree-species groups. Two model versions have been used: (a) standard simulation (b) and simulation with dynamical nitrogen (N). Regression lines for the correlation combining all data are shown.

significantly related to soil-water changes for all species (Fig. 6b–d;  $r^2 = 0.15$ –0.25; P < 0.001).

Our analysis of changes is based primarily on the comparison of the two periods 1901–1910 and 1991–2000, which could be biased by specific climatic conditions during these periods. As a further verification of the observed relationships, we additionally carried out analysis based on the linear temporal iWUE-trends (%-iWUE change per year). These results indicate that the iWUE-changes and trends are highly correlated (Figure S2) and that the relationship between iWUE and summer soil-water content does not depend



**Fig. 4** The change in iWUE between 1901–1910 and 1991–2000 derived from tree ring (a) and from model data (b). The colour code shows the change in iWUE expressed as the relative change compared to 1901–1910.

upon the particular metric for 20th century change (Figure S3).

#### Discussion

#### Observed iWUE patterns and trends

We applied two independent methods to investigate changes in iWUE of trees across Europe and through the 20th century, a period characterized by strongly increasing atmospheric  $CO_2$  concentrations and spatially inhomogeneous climatic changes. One method relies on carbon isotope variations in tree rings from a network of sites, and the other on calculations of a complex vegetation model. Results from the two methods show remarkable agreement regarding the spatial patterns, namely the strong latitudinal gradient with low iWUE in the north and high iWUE in the south of the study region (Figs 2 and 3), as well as the temporal evolution of changes (Fig. 1; Tables 1 and 2), characterized





**Fig. 5** Climate change in Europe shown as differences between 1991–2000 and 1901–1910 for summer (JJA) temperature (a), summer precipitation (b) and summer soil-water content (c). Temperature and precipitation data are from the CRU TS 3.1 data set and soil-water content is LPX model output.

**Fig. 6** The iWUE change from 1901–1910 to 1991–2000 as a function of the change in summer (JJA) soil-water content (SWC) for (a) data of all tree-ring sites, and (b) the modelled conifer, (c) modelled oak and (d) modelled larch results. In the model plots (b–d) there are data outside the displayed range which are not shown.

by an increasing trend in iWUE at almost all places. The general agreement between the two methods provides strong evidence for a wide-spread iWUE-increase in European forests over the 20th century, and suggests important changes in the biosphere–climate interactions such as reduced transpiration for a given carbon uptake.

## The influence of climate and CO<sub>2</sub>

The stationary spatial iWUE pattern and the corresponding latitudinal gradient is related to the summer temperature and precipitation variability in the model, showing higher iWUE for more water-stressed plants (growing in more arid conditions), consistent with studies of gas-exchange and concurrent isotope measurements over climate gradients or seasonal variations (e.g. Panek & Waring, 1997; Moore et al., 1999). It was observed for different species that when water stress resulted in stomatal closure, this was also reflected in decreasing carbon isotope discrimination as expected (Farquhar et al., 1982). However, surprisingly, we found the strongest increase in iWUE over the 20th century not at the most southern sites, but rather at more temperate latitudes (Fig. 4). This phenomenon was simulated by the model as well as observed in the tree-ring derived data. Analysing spatial variability in summer climate trends revealed that this region is generally characterized by a drying trend, reflected in decreasing summer soil-water content (Figs 5 and 6). Our results therefore suggest that the trees (mainly conifers) underwent an accelerated increase in iWUE in this region, as the sum of the effects of increasing CO<sub>2</sub> and soil drying, resulting in an effective reduction in stomatal conductance. A comparison of model simulations alternatively using observed and fixed (pre-industrial) CO<sub>2</sub> concentrations reveals significant differences in the magnitude of iWUE response. The differences between these simulations allow the relative importance of climatic vs. CO<sub>2</sub> drivers to be attributed in the model results. Accordingly, about 80% of the average iWUE increase in European forests is due to increased CO<sub>2</sub> (Table 2, constant CO<sub>2</sub> run) and the remainder attributable to climatic change.

## Assumptions and uncertainties

The application of dynamic vegetation model under a range of environmental conditions enables investigation of biosphere response to future environmental and climatic change scenarios, as well as the possibility to upscale these fluxes to larger spatial scales. Similarly, tree-ring stable isotopes provide an absolutely dated, replicable historical perspective on past carbon isotopic fractionation that may be coupled within modelling experiments to help evaluate and improve the model performance and resolve the nature/significance of key feedback mechanisms. But it is also important to stress that both the LPX model and the tree-ring isotopic approach to calculating iWUE rely on numerous assumptions, and should be interpreted cautiously. Regarding the isotope-derived iWUE, while Seibt et al. (2008) have pointed out that iWUE and actual wholeplant WUE do not necessarily need to be related, it was recently confirmed that  $\delta^{13}C$  in tree rings provides a good measure of transpiration to assimilation even on a seasonal scale (Michelot et al., 2011). Further, a largescale eddy-flux assessment showed indeed wide-spread forest WUE increase at least for the last 10-15 years (Keenan et al., 2013). The use of the LPX helps to circumvent some of the limitations of the isotope approach, because we do not need to draw conclusions about actual fluxes, such as transpiration, directly from our tree-ring derived iWUE. Modelled iWUE was on average about 20% lower than the tree-ring derived values and also the increase from the beginning to the end of the 20th century was somewhat lower in the model (Table 2). This suggests some uncertainties remain in the process and magnitude of iWUE responses both in the past and for the future. The agreement between model and data can vary depending on model assumptions, e.g. variable vs. fixed vegetation, but also the values for the isotope offset used in Eqn (1). We decided to use model settings from previous investigations (Spahni et al., 2013; Stocker et al., 2013), i.e. standard parameters, and not to optimize for best agreement between model and data, because this would involve some circular reasoning in testing modelled trends. Another implicit assumption of our study is that age-related trends are not significant. When trees grow in height, hydraulic resistance increases and turgor declines. This causes partial stomatal closure with an increase in iWUE, resulting in a reduction in height growth (Ryan & Yoder, 1997; Koch et al., 2004). Accordingly, clear changes of isotope discrimination with tree height have been reported (McDowell et al., 2011). However, measuring carbon isotopes in stem wood sampled at breast height where an average of carbon from the whole canopy is collected does not seem to reflect this process. Evidence from several studies has shown that trees generally do not show any age-related  $\delta^{13}$ C-trends after an initial juvenile phase of ca. 50 years (Francey & Farquhar, 1982), which can be particularly well seen for trees that were living before the industrial CO<sub>2</sub>-concentration rise (Gagen et al., 2007; Loader et al., 2013). Furthermore, the vast majority of analyses were conducted on trees that were long-lived (>250 years) additionally reducing the chances that the rapid juvenile increase in height or trends in  $\delta^{13}$ C ratios affect our 20th century analyses.

#### Physiological responses

The wide-spread iWUE increase was likely caused by a reduction in stomatal conductance at the leaf level (Gagen et al., 2011). The results from the LPX model indeed suggest that the cause of the iWUE increase was reduced stomatal conductance during recent decades rather than increased assimilation (Figure S4). However, the overall changes in canopy transpiration over the 20th century remain unclear. Other effects of the CO<sub>2</sub> fertilization like structural changes (such as increased leaf area) may compensate for the physiological response (reduced stomatal conductance) of plants to  $CO_2$  (Betts *et al.*, 1997). It is probably unlikely that runoff increases of rivers have occurred as a result of a reduced transpiration flux leading to wetter soils (Huntington, 2008), in contrast to earlier findings (Gedney et al., 2006). Tree responses in our study can be compared with elevated CO<sub>2</sub>/climate experiments with the caveat that such experimental studies have most often been conducted on juvenile trees (Wullschleger et al., 2002; Young et al., 2011) in CO<sub>2</sub> concentration ranges from 380 ppm to 500 ppm or more over short timescales. In contrast, our investigation was performed on samples of old-growth trees from intact ecosystems that have experienced CO<sub>2</sub> increases from 280 to 380 ppm over the course of a century. It has been observed in fumigation experiments that elevated CO<sub>2</sub> resulted in reduced stomatal conductance and therefore could ameliorate the negative effects of drought in many species through increased WUE (Ellsworth, 1999). A number of studies have reported such an increased plant-growth response to CO2 for trees growing under water-limiting conditions (Wullschleger et al., 2002; McMurtrie et al., 2008). Direct evidence for a sustained adaptation remains limited and it is likely that the magnitude and rate of this response will depend upon species as well as local site conditions (Leuzinger & Körner, 2007; Linares et al., 2009). A meta-analysis across different biomes did not detect a response in the radial growth, despite corresponding evidence for a significant and widespread increase in iWUE (Peñuelas et al., 2011).

#### Other factors than CO<sub>2</sub> and water

We have considered that factors other than  $CO_2$  and water could also be important for explaining the observed iWUE patterns and trends. Nitrogen is such a factor, representing one of the main nutrients for plant growth and a limiting factor in many ecosystems (Sun et al., 2010). The correlation between total N deposition and the tree-ring derived iWUE-increase in our network shows a significant relationship, and we can therefore not rule out the possibility that nitrogen deposition has partly contributed to the strong iWUEincrease in Central Europe. Furthermore, our simulations with dynamical N improved correlations to treering data (Fig. 3b). Other air pollutants, e.g. sulphur dioxide or ozone, may also influence tree physiological properties such as stomatal conductance and photosynthesis (Fairchild et al., 2009; Rinne et al., 2010). It was observed that long-term exposure of trees to background sulphur dioxide influenced carbon isotope signatures and iWUE of Abies alba Mill remote from industrial areas in western Germany (Boettger et al., 2014). However, through pollution reduction measures and legislation, the influence of nitrogen and especially sulphur is expected to further decrease in the future relative to climate change and CO<sub>2</sub> effect.

### Large-scale implications

Our finding of strongly increasing iWUE in regions of increasing summer drought is relevant in the context of refining projections of future climate development and carbon sequestration. It has been suggested that the drying trend in summer climate over Central and southern Europe in recent decades is consistent with modelderived changes in mean summer precipitation as well as changes in the large-scale circulation, and that this trend might continue into the future (Pal et al., 2004; IPCC, 2013). Therefore, it is likely that trees in Central Europe will be exposed to even stronger drought stress in the future to which some species may be poorly adapted. Accordingly, shifts in species distribution may occur, as already observed e.g. in the Valais (Switzerland), where the current drought-induced decline of Scots pine leads to a transition of these forests into mixed oak-deciduous at lower elevations and sprucefir forests at higher elevations (Bigler et al., 2006). Water has been identified as a major issue in a high-CO<sub>2</sub> world, with the question raised if the effect of elevated CO<sub>2</sub> on forest vegetation is 'more a water issue than a carbon issue' (Holtum & Winter, 2010), emphasizing the strong link between carbon and water cycles. Landatmosphere interactions, soil moisture-precipitation and temperature feedbacks play a crucial role in determining future summer climate variability (Seneviratne et al., 2006). Model and empirical studies clearly show the importance of biosphere/carbon-cycle feedbacks (Frank et al., 2010) and physiological forcing by CO<sub>2</sub> for predicting future climate (Cao et al., 2010). However, the magnitudes of such feedbacks are still highly uncertain and due to a lack of basic understanding of relevant

processes, terrestrial carbon models differ in their response to climate change (Plattner *et al.*, 2008; Frank *et al.*, 2010). Our study enables an assessment of a complex vegetation model with empirical data, which may ultimately lead to model improvements and better estimations of vegetation–climate feedbacks.

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## **Supporting Information**

Additional Supporting Information may be found in the online version of this article:

**Table S1.** Long-term annual temperature and precipitation for all study sites.

**Figure S1.** Correlation analysis between the 35 tree-ring derived iWUE time series from 1901 to 2000 and the output from the LPX model standard run for the nearest pixel.

**Figure S2.** Relationship between iWUE change in % (calculated as the relative difference between iWUE in the periods 1901–1910 and 1991–2000) and the iWUE trends (slope of the linear regression between iWUE and time for 1901–2000) for each tree-ring site.

**Figure S3.** The iWUE trends for the tree-ring sites as a function of the 20th century trends in summer (JJA) soil-water content (SWC).

Figure S4. LPX standard model calculated assimilation and stomatal conductance averaged for all Europe.