Large but decreasing effect of ozone on the European carbon sink

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26   **Abstract**

28   The capacity of the terrestrial biosphere to sequester carbon and mitigate climate change is governed by the ability

29   of vegetation to remove emissions of CO_2 through photosynthesis. Tropospheric O_3, a globally abundant and

30   potent greenhouse gas, is, however, known to damage plants, causing reductions in primary productivity, yet the

31   impact of this gas on European vegetation and the land carbon sink is largely unknown. Despite emission control

32   policies across Europe, background concentrations of tropospheric O_3 have risen significantly over the last

33   decades due to hemispheric-scale increases in O_3 and its precursors. Therefore, plants are exposed to increasing

34   background concentrations, at levels currently causing chronic damage. We use the JULES land-surface model

35   recalibrated for O_3 impacts on European vegetation, with an improved stomatal conductance parameterization, to

36   quantify the impact of tropospheric O_3, and its interaction with CO_2, on gross primary productivity (GPP) and

37   land carbon storage across Europe. A factorial set of model experiments showed that tropospheric O_3 can

38   significantly suppress terrestrial carbon uptake across Europe over the period 1901 to 2050. By 2050, simulated

39   GPP was reduced by 4 to 9% due to plant ozone damage, however, the combined effects of elevated future CO_2

40   (acting to reduce stomatal opening) and reductions in O_3 concentrations resulted in reduced O_3 damage in the

41   future, contrary to predictions from earlier studies. Reduced land carbon storage resulted from diminished soil

42   carbon stocks consistent with the reduction in GPP. Regional variations are identified with larger impacts shown

43   for temperate Europe compared to boreal regions. These results highlight that the effects of O_3 on plant physiology

44   add to the uncertainty of future trends in the land carbon sink and, as such, this should be incorporated into carbon

45   cycle assessments.

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1 Introduction

The terrestrial biosphere absorbs around 30% of anthropogenic CO_2 emissions and acts to mitigate climate change (Le Quéré et al., 2015). Early estimates of the European carbon balance suggest a terrestrial carbon sink of between 135 to 205 TgC yr^{-1} (Janssens et al., 2003). Schulze et al. (2009) determined a larger carbon sink of 274 TgC yr^{-1}, and more recent estimates suggest a European terrestrial sink of between 146 to 184 TgC yr^{-1} (Luyssaert et al., 2012). The carbon sink capacity of land ecosystems is dominated by the ability of vegetation to sequester carbon through photosynthesis and release it back to the atmosphere through respiration. Therefore, any change in the balance of these fluxes will alter ecosystem source-sink behaviour.

In recent decades much attention has focussed on the effects of rising atmospheric CO_2 on vegetation productivity (Ceulemans and Mousseau, 1994;Norby et al., 2005;Norby et al., 1999;Saxe et al., 1998). The Norby et al. (2005) synthesis of Free Air CO_2 Enrichment (FACE) experiments suggests a median stimulation ($23 \pm 2\%$) of forest NPP in response to a doubling of CO_2. Similar average increases (20%) were observed for C_3 crops, although this translated into smaller gains in biomass (17%) and crop yields (13%) (Long et al., 2006). The long-term effects of CO_2 fertilization on plant growth and carbon storage are nevertheless uncertain (Baig et al., 2015;Ciais et al., 2013). Little attention, however, has been given to tropospheric ozone (O_3), a globally abundant and increasing air pollutant recognised as one of the most damaging pollutants for forests (Karlsson et al., 2007;Royal-Society, 2008;Simpson et al., 2014b). Tropospheric O_3 is a secondary air pollutant formed by photochemical reactions involving carbon monoxide, volatile organic compounds, methane and nitrogen oxides from both man-made and natural sources, as well as downward transport from the stratosphere and lightning. The phytotoxic effects of O_3 exposure are shown to decrease vegetation productivity and biomass, with consequences for terrestrial carbon sequestration (Felzer et al., 2004;Loya et al., 2003;Mills et al., 2011b;Sitch et al., 2007). Few studies, however, consider the simultaneous effects of exposure to both gases, and few Earth-system models (ESMs) currently explicitly consider the role of tropospheric O_3 in terrestrial carbon dynamics (IPCC, 2013), both of which are key to understanding the carbon sequestration potential of the land-surface, and future carbon dynamics regionally and globally.

Due to increased anthropogenic precursor emissions over the industrial period, background concentrations of ground-level O_3 have risen (Parrish et al., 2012). As a result of controls on precursor emissions in Europe and North America, peak O_3 concentrations in these regions have decreased or stabilised over recent decades (Cooper et al., 2014;Logan et al., 2012;Parrish et al., 2012;Simpson et al., 2014b). Nevertheless, climate change may increase the frequency of weather events conducive to peak O_3 incidents in the future (e.g. summer droughts and heat-waves; e.g., (Sicard et al., 2013), and may increase biogenic emissions of the O_3-precursors isoprene and NO_x, although such impacts are subject to great uncertainty (Simpson et al., 2014b;Young et al., 2013;Young et al., 2009). Furthermore, intercontinental transport of air pollution means background O_3 concentrations have risen significantly over the last decades. Northern Hemisphere background concentrations of O_3 are now close to established levels for impacts on human health and the terrestrial environment (Royal-Society, 2008). Therefore, although peak O_3 concentrations are in decline across Europe, plants are exposed to increasing background levels, at levels currently causing chronic damage (Mills et al., 2011b). Intercontinental transport means future O_3

98   concentrations in Europe are dependent on how O_3 precursor emissions evolve globally, including regions such

99   as Asia that currently have poor emission controls (Cooper et al., 2010;Verstraeten et al., 2015).

100

101   Elevated O_3 concentrations impact agricultural yields and nutritional quality of major crops, with consequences

102   for global food security (Tai et al., 2014). As well as being a significant air pollutant, O_3 is a potent greenhouse

103   gas (Royal-Society, 2008). High levels of O_3 are damaging to ecosystem health and reduce the global land carbon

104   sink (Arneth et al., 2010;Sitch et al., 2007). Reduced uptake of carbon by plant photosynthesis due to O_3 damage

105   allows more CO_2 to remain in the atmosphere. This effect of O_3 on plant physiology represents an additional

106   climate warming to the direct radiative forcing of O_3 (Collins et al., 2010;Sitch et al., 2007), the magnitude of

107   which, however, remains highly uncertain (IPCC, 2013).

108

109   Dry deposition of O_3 to terrestrial surfaces, primarily uptake by stomata on plant foliage and deposition on external

110   surfaces of vegetation, is a significant sink for ground level O_3 (Fowler et al., 2009). On entry to sub-stomatal

111   spaces, O_3 reacts with other molecules to form reactive oxygen species (ROS). Plants can tolerate a certain level

112   of O_3 depending on their capacity to scavenge and detoxify the ROS (Ainsworth et al., 2012). Above this critical

113   level, long-term chronic O_3 exposure reduces plant photosynthesis and biomass accumulation (Ainsworth,

114   2008;Ainsworth et al., 2012;Matyssek et al., 2010a;Wittig et al., 2007;Wittig et al., 2009), either directly through

115   effects on photosynthetic machinery such as reduced Rubisco content (Ainsworth et al., 2012;Wittig et al., 2009)

116   and/or indirectly by reduced stomatal conductance (g_s) (Kitao et al., 2009;Wittig et al., 2007), alters carbon

117   allocation to different pools (Grantz et al., 2006;Wittig et al., 2009), accelerates leaf senescence (Ainsworth,

118   2008;Nunn et al., 2005;Wittig et al., 2009) and changes plant susceptibility to biotic stress factors (Karnosky et

119   al., 2002;Percy et al., 2002).

120

121   Understanding the response of plants to elevated tropospheric O_3 is challenged by the large variation in O_3

122   sensitivity both within and between species (Karnosky et al., 2007;Kubiske et al., 2007;Wittig et al., 2009).

123   Additionally, other environmental stresses that affect stomatal behaviour will affect the rate of O_3 uptake and

124   therefore the response to O_3 exposure, such as high temperature, drought and changing concentrations of

125   atmospheric CO_2 (Mills et al., 2016;Fagnano et al., 2009;Kitao et al., 2009;Löw et al., 2006), such that the

126   response of vegetation to O_3 is a balance between opposing drivers of stomatal behaviour.

127

128   Here we assess the impact of historical and projected changes in tropospheric O_3 concentration from 1901 to 2050

129   on the European land-carbon sink using the Joint UK land environment simulator (JULES) (Best et al., 2011;Clark

130   et al., 2011), the land-surface model of the UK Earth System Model (UKESM) (Collins et al., 2011). Plant O_3

131   sensitivity in JULES was re-calibrated (high and low plant O_3 sensitivity) using the latest observations for

132   European vegetation in order to capture a range of plant sensitivities to O_3 – this includes separate sensitivities for

133   Mediterranean regions, and for agricultural crops (wheat) versus natural grassland. We make a separate distinction

134   for the Mediterranean region where possible because the work of Büker et al. (2015) showed that different O_3

135   dose-response relationships are needed to describe the O_3 sensitivity of dominant Mediterranean trees. We modify

136   the representation of stomatal O_3 flux in JULES from Sitch *et al.*, (2007) by including a term for non-stomatal

137   deposition of O_3 to leaf surfaces which is recognised as an important sink for ground-level O_3. Given the critical

138 role g_s plays in the uptake of both CO_2 and O_3, we use an improved representation and parameterisation of g_s in

139 JULES by implementing the Medlyn *et al*. (2011) g_s formulation. Based on the optimal theory of stomatal

140 behaviour, Medlyn *et al*., (2011) has the following advantages over the current JULES g_s formulation: i) a single

141 parameter (g_1) which represents the marginal cost of water-use; ii) easy to parameterise with leaf or canopy level

142 observations, and (iii) values of g_1 are available for different plant functional types (PFTs) derived from a global

143 data set of measured leaf stomatal conductance, photosynthesis and vapour pressure deficit (VPD) (Lin et al.,

144 2015).

145

146 We use a factorial suite of model experiments to investigate the temporal and spatial evolution of O_3 impacts on

147 European vegetation from 1901 to 2050. We do not attempt to make a full assessment of the carbon cycle of

148 Europe, instead we target O_3 damage which is currently a missing component in earlier carbon cycle assessments.

149 Accounting for the well-known differences in plant sensitivity to O_3 is complex, therefore, here we provide a

150 sensitivity assessment by using two sets of simulations – a high and lower plant O_3 sensitivity parameterisation,

151 with O_3 sensitivities that vary by PFT and region. We investigate the interaction between CO_2 and O_3, the two

152 greenhouse gases that directly affect plant photosynthesis, and indirectly g_s. Our aim is to quantify the impact of

153 these two gases on GPP and land carbon storage across Europe. We go beyond the present-day carbon budget and

154 investigate the impact of state-of-art future scenarios up to year 2050.

155

156 **2 Methods**

157

158 **2.1 Representation of O_3 effects in JULES**

159

160 JULES calculates the land-atmosphere exchanges of heat, energy, mass, momentum and carbon on a sub-daily

161 time step, and includes a dynamic vegetation model (Best et al., 2011;Clark et al., 2011;Cox, 2001). This work

162 uses JULES version 3.3 (http://www.jchmr.org), which uses a multi-layer canopy radiation interception and

163 photosynthesis scheme (10 layers in this instance) that accounts for direct and diffuse radiation, sun fleck

164 penetration through the canopy, inhibition of leaf respiration in the light and change in photosynthetic capacity

165 with depth into the canopy (Clark et al., 2011;Mercado et al., 2009). Soil water content also affects the rate of

166 photosynthesis and g_s. It is modelled using a dimensionless soil water stress factor, β, which is related to the mean

167 soil water concentration in the root zone, and the soil water contents at the critical and wilting point (Best *et al*.,

168 2011).

169

170 To simulate the effects of O_3 deposition on vegetation productivity and water use, JULES uses the flux-gradient

171 approach of Sitch *et al*., (2007), modified to include non-stomatal deposition following Tuovinen et al. (2009).

172 JULES uses a coupled model of g_s and photosynthesis; because of the relationship between these two fluxes, the

173 direct effect of O_3 damage on photosynthetic rate also leads to a reduction in g_s. Changes in atmospheric CO_2

174 concentration also affect photosynthetic rate and g_s, consequently the interaction between changing concentrations

175 of both gases is allowed for. Specifically, the potential net photosynthetic rate (A_p, mol CO_2 m^{-2} s^{-1}) is modified

176 by an 'O_3 uptake' factor (F, the fractional reduction in photosynthesis), so that the actual net photosynthesis (A_{net},

177 mol CO_2 m^{-2} s^{-1}) is given by equation 1 (Clark *et al.,* 2011, Sitch *et al.,* 2007).

178

179     $A_{net} = A_P F$ (1)

180

181   The O_3 uptake factor (F) is defined as:

182

183     $F = 1 - a * max[F_{O3} - F_{O3crit}, 0.0]$ (2)

184

185   F_{O3} is the instantaneous leaf uptake of O_3 (nmol m^{-2} s^{-1}), F_{O3crit} is a PFT-specific threshold for O_3 damage (nmol

186   m^{-2} PLA s^{-1}, projected leaf area), and 'a' is a PFT-specific parameter representing the fractional reduction of

187   photosynthesis with O_3 uptake by leaves. Following Tuovinen et al. (2009), the flux of O_3 through stomata, F_{O3},

188   is represented as follows:

189

190     $F_{O3} = O_3 \left(\dfrac{g_b \left(\frac{g_l}{K_{O3}} \right)}{g_b + \left(\frac{g_l}{K_{O3}} \right) + g_{ext}} \right)$ (3a)

191

192   O_3 is the molar concentration of O_3 at reference (canopy) level (nmol m^{-3}), g_b is the leaf-scale boundary layer

193   conductance (m s^{-1}, eq 3b), g_l is the leaf conductance for water (m s^{-1}), K_{o3} is the ratio of leaf resistance for O_3 to

194   leaf resistance for water vapour and takes a value of 1.51 after Massman (1998), and g_{ext} is the leaf-scale non-

195   stomatal deposition to external plant surfaces (m s^{-1}) which takes a constant value of 0.0004 m s^{-1} after Tuovinen

196   et al. (2009). The leaf-level boundary layer conductance (g_b) is calculated as in Tuovinen *et al.* (2009)

197

198     $g_b = \alpha L d^{-1/2} U^{-1/2}$ (3b)

199

200   α is a constant (0.0051 m s$^{-1/2}$), Ld is the cross-wind leaf dimension (m) and U is wind speed at canopy height (m

201   s^{-1}). The rate of O_3 uptake is dependent on g_s, which is dependent on photosynthetic rate. Given g_s is a linear

202   function of photosynthetic rate in JULES (Clark et al., 2011), from eq 1 it follows that:

203

204     $g_s = g_l F$ (4)

205

206   The O_3 flux to stomata, F_{O3}, is calculated at leaf level and then scaled to each canopy layer differentiating sunlit

207   and shaded leaf photosynthesis, and finally summed up to the canopy level. Because the photosynthetic capacity,

208   photosynthesis and therefore g_s decline with depth into the canopy, this in turn affects O_3 uptake, with the top leaf

209   level contributing most to the total O_3 flux and the lowest level contributing least.

210

211   **2.2 Calibration of O_3 uptake model for European vegetation**

212

213   Each JULES PFT (broadleaf, needle leaf, C_3 and C_4 herbaceous, and shrub) was calibrated for a high and low

214   plant O_3 sensitivity to account for uncertainty in variation of species sensitivity to O_3, using the approach of Sitch

215   *et al.*, (2007). For the C_3 herbaceous PFT – the dominant land cover type across Europe in this study (Fig. S1) -

216   the O_3 sensitivity was calibrated against observations for wheat to give a representation of agricultural regions

217 (high plant O_3 sensitivity), versus natural grassland (low plant O_3 sensitivity), with a separate function for

218 Mediterranean grasslands (low plant O_3 sensitivity) (Table S1 and Figure S2).

219

220 To calibrate the JULES O_3 sensitivity (parameter 'a' in eq 2), JULES was run to be as directly comparable as

221 possible to the dose-based O_3 risk indicator used in CLRTAP (2017), using the O_3 flux per projected leaf area to

222 top canopy sunlit leaves. Hourly averaged F_{O3} in excess of F_{O3crit} were accumulated over a species specific

223 accumulation period. Values of F_{O3crit} came from observations, the parameter 'a' was modified until the modelled

224 change in response variable with cumulative uptake of O_3 above the specified threshold matched the observations

225 (see further method details in SI).

226

227 **2.3 Representation of stomatal conductance**

228

229 In JULES, g_s (m s^{-1}) is represented following the closure proposed by (Jacobs, 1994):

230

231 $$g_s = 1.6RT_l \frac{A_{net}\beta}{c_a - c_i} \quad (5)$$

232

233 Where in the default JULES model:

234

235 $$c_i = (c_a - c_*)f0\left(1 - \frac{dq}{dq_{crit}}\right) + c_* \quad (6)$$

236

237 β is a soil moisture stress factor, the factor 1.6 accounts for g_s being the conductance for water vapour rather than

238 CO_2, R is the universal gas constant (J K^{-1} mol^{-1}), T_l is the leaf surface temperature (K), c_a and c_i (both Pa) are the

239 leaf surface and internal CO_2 partial pressures, respectively, c_* (Pa) is the CO_2 photorespiration compensation

240 point, dq is the humidity deficit at the leaf surface (kg kg^{-1}), dq_{crit} (kg kg^{-1}) and f_0 are PFT specific parameters

241 representing the critical humidity deficit at the leaf surface, and the leaf internal to atmospheric CO_2 ratio (c_i/c_a)

242 at the leaf specific humidity deficit (Best *et al.* 2011), values are shown is Table S1.

243

244 In this work, we replace equation 6 with the Medlyn et al. (2011) closure using the key PFT specific model

245 parameter g_1 (kPa$^{0.5}$) and dq is expressed in kPa, shown in eq 7:

246

247 $$c_i = c_a\left(\frac{g_1}{g_1 + \sqrt{dq}}\right) \quad (7)$$

248

249 PFT specific values of the g_1 parameter were derived for European vegetation from the data base of Lin et al.

250 (2015) and are shown in Table S1. The g_1 parameter represents the sensitivity of the g_s to the assimilation rate,

251 i.e. plant water use efficiency. It was derived from fitting the Medlyn *et al.*, (2011) model to observations of g_s,

252 photosynthesis, and VPD (Lin et al., 2015). The study of Hoshika et al. (2013) found an effect of O_3 on g_1 for

253 beech trees (*Fagus crenata*) only at the start of the growing season (June), but not during the following months

254 (August and October). Quantifying an O_3 effect directly on g_1 would require a detailed meta-analysis of empirical

255 data on photosynthesis and g_s for different PFTs, which is currently lacking in the literature. As explained above,

256 here we take an empirical approach to modelling plant O_3 damage, essentially by applying a reduction factor to

257 the simulated plant photosynthesis based on observations of whole plant losses of biomass with O_3 exposure, for

258 which there is a lot more available data (e.g. CLRTAP, 2017). The impact of the g_s model formulation is shown

259 for two contrasting grid points (wet/dry) in central Europe (see SI).

260

261 **2.4 Model simulations for Europe**

262

263 **2.4.1 Forcing datasets**

264

265 We used the WATCH meteorological forcing data set (Weedon et al., 2010;Weedon et al., 2011) at 0.5º x 0.5º

266 spatial and three hour temporal resolution for our JULES simulations. JULES interpolates this down to an hourly

267 model time step. For this study, the climate was kept constant by recycling over the period 1901 to 1920, to allow

268 us to fully understand the impact O_3, CO_2 and their interaction.

269

270 Pre-industrial global CO_2 concentrations (1900 to 1960) were taken from Etheridge et al. (1996), 1960 to 2002

271 were from Mauna Loa (Keeling and Whorf, 2004), as calculated by the Global Carbon Project (Le Quéré et al.,

272 2016), and 2003-2050 were based on the IPCC SRES A1B scenario and were linearly interpolated to gap fill

273 missing years (Fig. 1).

274

275 JULES was run including dynamic vegetation with a land cover mask giving the fraction of agriculture in each

276 grid cell based on the Hurtt et al. (2011) land cover database for the year 2000. Within the agricultural mask, only

277 C_3/C_4 herbaceous PFTs are allowed to grow and all other PFTs are assumed absent. By including dynamic

278 vegetation, grid cell PFT coverage and Leaf Area Index (LAI) is a result of resource availability and simulated

279 competition. Following a full spin-up period (to ensure equilibrium vegetation, carbon and water states), the

280 fractional cover of each PFT changed little over the simulation period (1901 - 2050), the land cover for 2050 is

281 shown in Fig. S1. The model experiments in this study are run for both a high and low plant O_3 sensitivity: for the

282 high plant O_3 sensitivity, all herbaceous PFT fractional cover uses the O_3 sensitivity for wheat, and for the low

283 plant O_3 sensitivity, all herbaceous PFT fractional cover uses the O_3 sensitivity for natural grasslands.

284

285 Tropospheric O_3 concentration was produced by the EMEP MSC-W model (Simpson et al., 2012), driven with

286 meteorology from the regional climate model RCA3 (Kjellström et al., 2011;Samuelsson et al., 2011), which

287 provides a downscaling of the ECHAM A1B-r3 (simulation 11 of Kjellström *et al.*, 2011). This setup

288 (EMEP+RCA3) is also used by Langner et al. (2012), Simpson et al. (2014a), Tuovinen et al. (2013), Franz et al.

289 (2017) and Engardt et al. (2017), where further details and model evaluation can be found. Unfortunately, the 3-

290 dimensional RCA3 data needed by the EMEP model was not available prior to 1960, but as in Engardt et al.

291 (2017) the meteorology of 1900-1959 had to be approximated by assigning random years from 1960 to 1969. This

292 procedure introduces some uncertainty of course, but Langner et al. (2012) show that it is emissions change, rather

293 than meteorological change, that drives modelled ozone concentrations. The emissions scenarios for 1900-2050

294 merge data from the International Institute of Applied System Analysis (IIASA) for 2005-2050 (the so-called

295   ECLIPSE 4a scenario, Klimont *et al.* (2016)), recently revised EMEP data for 1990, and a scaling back from 1990

296   to 1900 using data from Lamarque et al. (2013). The EMEP model accounts for changes in BVOC emissions as a

297   result of predicted ambient temperature changes, however as with all uncoupled modelling studies, there is no

298   interaction between changes in leaf-level g_s, BVOCs and O_3 formation.

299

300   This study used daily mean values of tropospheric O_3 concentration from EMEP MSC-W disaggregated down to

301   the hourly JULES model time-step. Hourly O_3 values allow for variation in the diurnal response to O_3 exposure

302   resulting in more accurate estimation of O_3 uptake. O_3 concentrations from EMEP were calculated at canopy

303   height for two land-cover categories: forest and grassland (Fig. S3 and Fig. S4), which are taken as surrogates for

304   high and low vegetation, respectively. These canopy-height specific concentrations allow for the large gradients

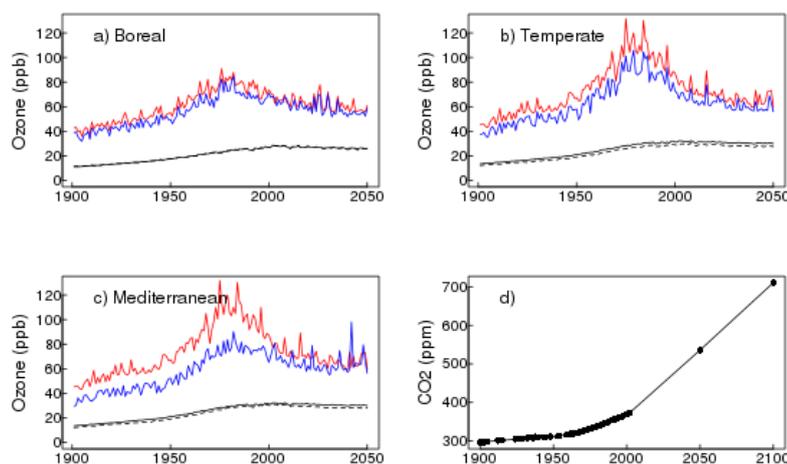
305   in O_3 concentration that can occur in the lowest 10s of metres, giving lower O_3 for grasslands than seen at e.g. 20

306   m in a forest canopy (Simpson et al., 2012;Tuovinen et al., 2009). Figure 1 shows the regional mean annual O_3

307   concentration (regions are depicted in Fig. S5) along with the annual maximum. Together these clearly show the

308   trend of increasing O_3 concentration on pre-industrial levels in all regions, although notably lower increases in the

309   Boreal region. Around the 1990's O_3 concentrations stabilise and then start to decrease into the future.

310

312   **Figure 1.** Regional time series of canopy height O_3 (ppb) forcing from EMEP a) to c), and d) global atmospheric

313   CO_2 (ppm) concentration (this does not vary regionally; black dots show data points, the black line shows

314   interpolated points). Each panel for the O_3 forcing shows the regional annual average (woody PFTs, black solid

315   line; herbaceous PFTs, black dashed line) and the annual maximum O_3 concentration above: woody PFTs (red)

316   and herbaceous PFTs (blue).

317

318   **2.4.2 Spin up and factorial experiments**

319

320   JULES was spun-up by recycling the climate from the early part of the twentieth century (1901 to 1920) using

321   atmospheric CO_2 (296.1 ppm) and O_3 concentrations from 1901 (Fig. S3 & Fig. S4). Model spin-up was 2000

322 years by which point the carbon pools and fluxes were in steady state with zero mean net land – atmosphere CO_2

323 flux. We performed the following transient simulations for the period 1901 to 2050 with continued recycling of

324 the climate as used in the spin-up, for both high and low plant O_3 sensitivities:

325

326 • **O3**   : Fixed 1901 CO_2, Varying O_3

327 • **CO2**   : Varying CO_2, Fixed 1901 O_3

328 • **CO2 + O3**   : Varying CO_2, Varying O_3

329

330 We use these simulations to investigate the direct effects of changing atmospheric CO_2 and O_3 concentrations,

331 individually and combined, on plant physiology through the twentieth century and into the future, specifically

332 over three time periods: historical (1901-2001), future (2001-2050) and over the full time series (1901-2050). See

333 more details in the SI.

334

335 **3 Results**

336

337 **3.1 Impact of g_s model formulation**

338

339 The impact of g_s model on simulated g_s is shown for the wet site (Fig. 2). For the broadleaf tree and C_3 herbaceous

340 PFT, the Medlyn g_s model simulates a significantly larger conductance compared to the Jacobs g_s model. In other

341 words, with the Medlyn g_s model these two PFTs are parameterised with a less conservative water use strategy,

342 which, for the grid point used in the simulation, increased the annual mean leaf-level water use by 22 % and 45

343 %, respectively. In contrast, the needle leaf tree, C_4 herbaceous and shrub PFTs are parameterised with a more

344 conservative water use strategy with the Medlyn g_s model, and the mean annual g_s was decreased by 16 %, 32 %

345 and 41 %, respectively, compared to the Jacobs g_s model. This comparison was also done for a dry site, and similar

346 results were found (Fig. S6), suggesting these results are representative across the domain. The effect of g_s model

347 on simulated photosynthesis was much smaller because of the low sensitivity of the limiting rates of

348 photosynthesis to changes in c_i (Fig. S7 & S8). Changes in leaf-level g_s impact the partitioning of simulated energy

349 fluxes. In general, increased g_s results in increased latent heat and thus decreased sensible heat flux, and vice versa

350 where g_s is decreased (Fig. S7 & S8). Also shown is the effect of the Medlyn g_s model on O_3 flux into the leaf

351 (Fig. 2 and Fig. S6, bottom panels). For the broadleaf tree and C_3 herbaceous PFT, the Medlyn model simulates a

352 larger conductance and therefore a greater flux of O_3 through stomata compared to Jacobs, and this is indicative

353 of the potential for greater reductions in photosynthesis (Fig. S7 & S8). The reverse is seen for the needle leaf

354 tree, C_4 herbaceous and shrub PFTs.

355

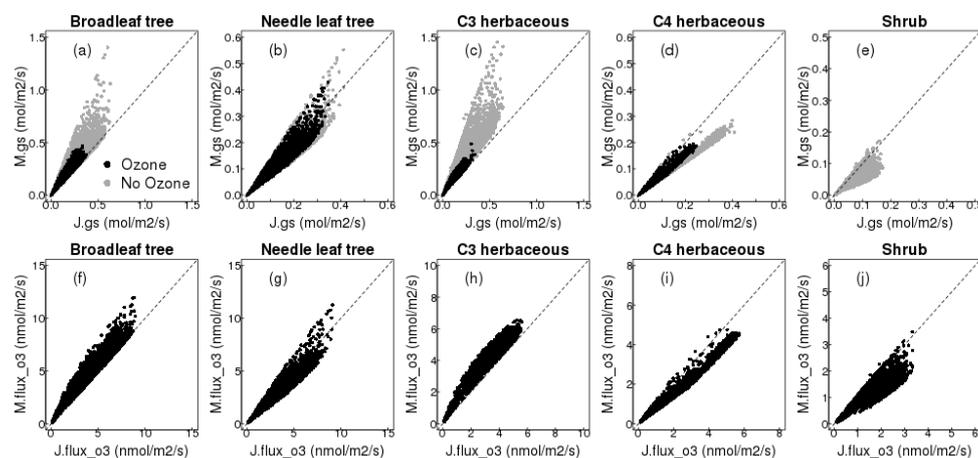



Figure 2. Comparison of simulated g_s with the Medlyn *et al.*, (2011) (y axis) versus the Jacobs (1994) formulation (x axis) currently used in JULES for all five JULES PFTs, for stomatal conductance (g_s, top row) and the flux of O_3 through the stomata (flux_o3, bottom row).

3.2 European simulations - Historical Period: 1901-2001

Over the historical period (1901-2001), the physiological effect of O_3 reduced GPP (-3% to -9%) for the low and high plant O_3 sensitivity parameterizations, respectively (Table 1). Figure 3 highlights regional variations, however, where simulated reductions in GPP are up to 20% across large areas of Europe, and up to 30% in some Mediterranean regions under the high plant O_3 sensitivity. Some Boreal and Mediterranean regions show increased GPP over this period, associated with O_3 induced stomatal closure enhancing water availability in these drier regions (Fig. 4), enhancing stomatal conductance later in the year (up to 10%, Fig. 5) and therefore GPP, but these regions comprise only a small area of the entire domain. Indeed, over much of the Europe, O_3-induced stomatal closure led to reduced g_s (up to 20%) across large areas of temperate Europe and the Mediterranean, and even greater reductions in some smaller regions of southern Mediterranean (Fig. 5), and these are not associated with significant increases in soil moisture availability (Fig. 4), resulting in depressed GPP over much of Europe as described above. Under the low plant O_3 sensitivity, similar spatial patterns occur, but the magnitude of GPP change (up to -10% across much of Europe) and g_s change (-5% to -10%) are lower compared to the high sensitivity. Over the twentieth century the land carbon sink is significantly suppressed (-2% to -6%, Table 1). Large regional variation is shown in Figure 3, with temperate and Mediterranean Europe seeing a large reduction in land carbon storage, particularly under the high plant O_3 sensitivity (up to -15%). Combined, the physiological response to changing CO_2 and O_3 concentrations results in a net loss of land carbon over the twentieth century under the high plant O_3 sensitivity (-2%, Table 1), dominated by loss of soil carbon (Table S2). This reflects the large increases in tropospheric O_3 concentration observed over this period (Fig. 1). Under the low plant O_3 sensitivity, the land carbon sink has started to recover by 2001 (+1.5%) owing to the recovery of the soil carbon pool beyond 1901 values over this period (Table S2).

384   To gain perspective on the magnitude of the O_3 induced flux of carbon from the land to the atmosphere we relate

385   changes in total land carbon to carbon emissions from fossil fuel combustion and cement production for the EU-

386   28-plus countries from the data of Boden et al. (2013). We recognise that our simulation domain is slightly larger

387   than the EU28-plus as it includes a small area of western Russia so direct comparisons cannot be made, but this

388   still provides a useful measure of the size of the carbon flux. For the period 1970 to 1979 the simulated loss of

389   carbon from the European terrestrial biosphere due to O_3 effects on vegetation physiology was on average 1.32

390   Pg C (high vegetation sensitivity) and 0.71 Pg C (low vegetation sensitivity) (Table 2). This O_3 induced reduced

391   C uptake of the land surface is equivalent to around 8% to 16% of the emissions of carbon from fossil fuel

392   combustion and cement production over the same period for the EU28-plus countries (Table 2). Currently the

393   emissions data availability goes up to 2011, so over the last observable decade (2002 to 2011) this land carbon

394   loss has declined but is still equivalent to 2% to 4% of the emissions of carbon from fossil fuels and cement

395   production for the EU28-plus countries (Table 2). Therefore, the indirect O_3 effect on the land carbon sink

396   potentially represents a significant additional source of anthropogenic carbon.

397

398   **3.3 European simulations - Future Period: 2001-2050**

399

400   Over the 2001 to 2050 period, region-wide GPP with O_3 only changing increased marginally (+0.1% to +0.2%,

401   high and low plant O_3 sensitivity, Table 1), although with large spatial variability (Fig. 3). This reflects changes

402   in tropospheric O_3 concentration as emission control policies reduce O_3 concentrations. Figures S4 and S5 show

403   that despite decreased tropospheric O_3 concentrations by 2050 in summer compared to 2001 levels, all regions are

404   exposed to an increase in O_3 over the wintertime, and some regions of Europe, particularly

405   temperate/Mediterranean experience increases in O_3 concentration in spring and autumn. Therefore, although

406   increased GPP (dominantly 10%, but up to 20% in some areas) on 2001 levels is simulated across large areas of

407   Europe, decreases of up to 21% are simulated in some areas of the Mediterranean, up to 15% in some areas of the

408   boreal region and up to 27% in the temperate zone. When O_3 and CO_2 effects are combined, simulated GPP

409   increases (+15% to +18%, high/low plant O_3 sensitivities respectively, Table 1). This increase is greater than the

410   enhancement simulated when CO_2 affects plant growth independently, because additional O_3 induced stomatal

411   closure increases soil water availability in some regions, which enhances growth more in the O_3 and CO_2

412   simulations, compared to the CO_2 only run. Nevertheless, although the percentage gain is larger, the absolute

413   value of GPP by 2050 remains lower compared to GPP with CO_2 only changing (Table S3).

414

415   Despite small increases in GPP in the O_3-only simulation, the land carbon sink continues to decline from 2001

416   levels (-0.7% to -1.6%, low and high plant O_3 sensitivity respectively, Table 1). This is because the soil and

417   vegetation carbon pools continue to lose carbon as they adjust slowly to small changes in input (GPP).

418   Nevertheless, the negative effect of O_3 on the future land sink is markedly reduced relative to the historical period.

419   Figure 3 however highlights regional differences. Boreal regions and parts of central Europe see minimal O_3

420   damage, whereas some areas of southern and northern Europe see further losses of up to 8% on 2001 levels. The

421   combined O_3 and CO_2 effects are dominated by the physiological effects of changing CO_2, with land carbon sink

422   increases of up to 7% (Table 1).

423

3.4 European simulations - Anthropocene: 1901-2050

Over the Anthropocene, O_3 reduces GPP (-4% to -9%) and land carbon storage (-3% to -7%, Table 1, Fig. S9). Regionally, O_3 damage is lowest in the boreal zone, GPP decreases are largely between 5% to 8% / 2% to 4% for the high/low plant O_3 sensitivity respectively, with large areas minimally affected by O_3 damage (Figure 6), consistent with lower g_s of needle leaf trees that dominate this region, and so lower O_3 uptake (Fig. S10 & S11). In the temperate region, O_3 damage is extensive with reductions in GPP dominantly from 10% to 15% for the low and high plant O_3 sensitivity respectively. Across significant areas of this region reductions in GPP are up to 20% under high plant O_3 sensitivity (Figure 6). In the Mediterranean region, O_3 damage reduces GPP by 5% to 15% / 3% to 6% for the high/low plant O_3 sensitivity respectively, with some areas seeing greater losses of up to 20% under the high plant O_3 sensitivity, but this is less extensive than that seen in the temperate zone (Figure 6). In these drier regions, O_3 induced stomatal closure can increase available soil moisture (Fig. S10 & S11).

Varying CO_2 and O_3 together shows that CO_2 induced stomatal closure can help alleviate O_3 damage by reducing the uptake of O_3 (Table S5). In these simulations, CO_2-induced stomatal closure was found to offset O_3-suppression of GPP, such that GPP by 2050 is 3% to 7% lower due to O_3 exposure, rather than 4% to 9% lower in the absence of increasing CO_2 (Table S5). Figure 6 shows this spatially, O_3 damage is reduced when the effect of atmospheric CO_2 on stomatal closure is accounted for, however despite this, the land carbon sink and GPP remain significantly reduced due to O_3 exposure.

Over the Anthropocene, changing O_3 and CO_2 in tandem results in an increase in European land carbon uptake (+5% to +9%), and an increase in GPP (+20% to +23%) by 2050 for the high and low plant O_3 sensitivity, respectively (Table 1). Nevertheless, despite this increase there remains a large negative impact of O_3 on the European land carbon sink (Fig. S9). By 2050 the simulated enhancement of land carbon and GPP in response to elevated CO_2 alone is reduced by 3% to 6% (land carbon) and 4% to 9% (GPP) for the low and high plant O_3 sensitivity respectively, when O_3 is also accounted for (Table 1). This is a significant reduction in the ability of the European terrestrial biosphere to sequester carbon.

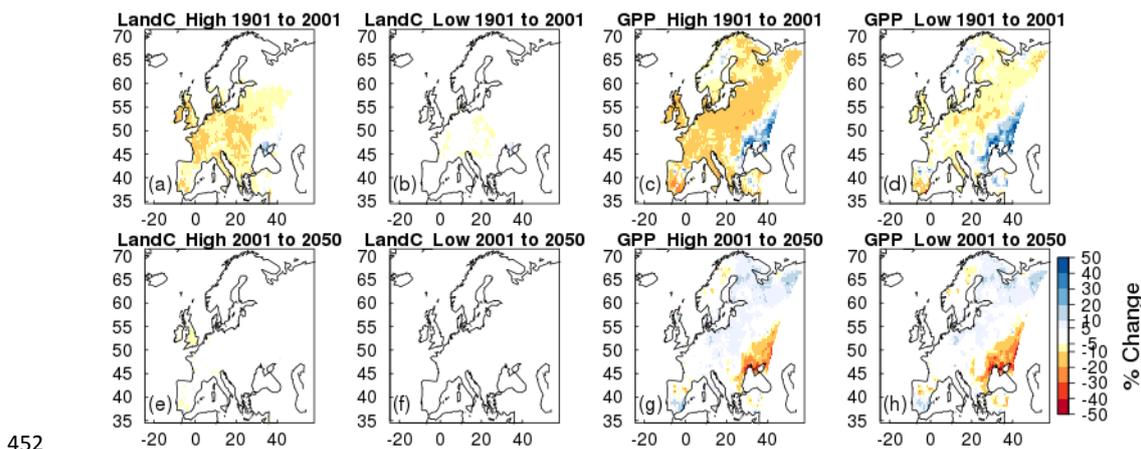



Figure 3. Simulated percentage change in total carbon stocks (Land C) and gross primary productivity (GPP) due to O_3 effects at fixed pre-industrial atmospheric CO_2 concentration. Changes are shown for the periods 1901 to 2001, and 2001 to 2050 for the high and low plant O_3 sensitivity.

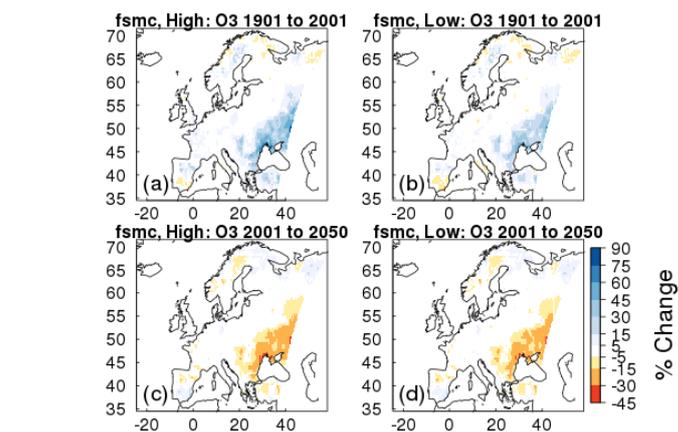



Figure 4. Simulated percentage change in plant available soil moisture (*fsmc*) due to O_3 effects at fixed pre-industrial atmospheric CO_2 concentration. Changes are shown for the periods 1901 to 2001, and 2001 to 2050 for the high and low plant O_3 sensitivity.

462

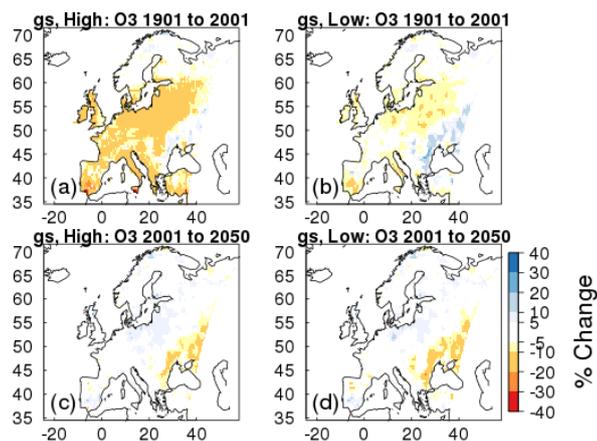



Figure 5. Simulated percentage change in stomatal conductance (g_s) due to O_3 effects at fixed pre-industrial

atmospheric CO_2 concentration. Changes are shown for the periods 1901 to 2001, and 2001 to 2050 for the high

and low plant O_3 sensitivity.

466

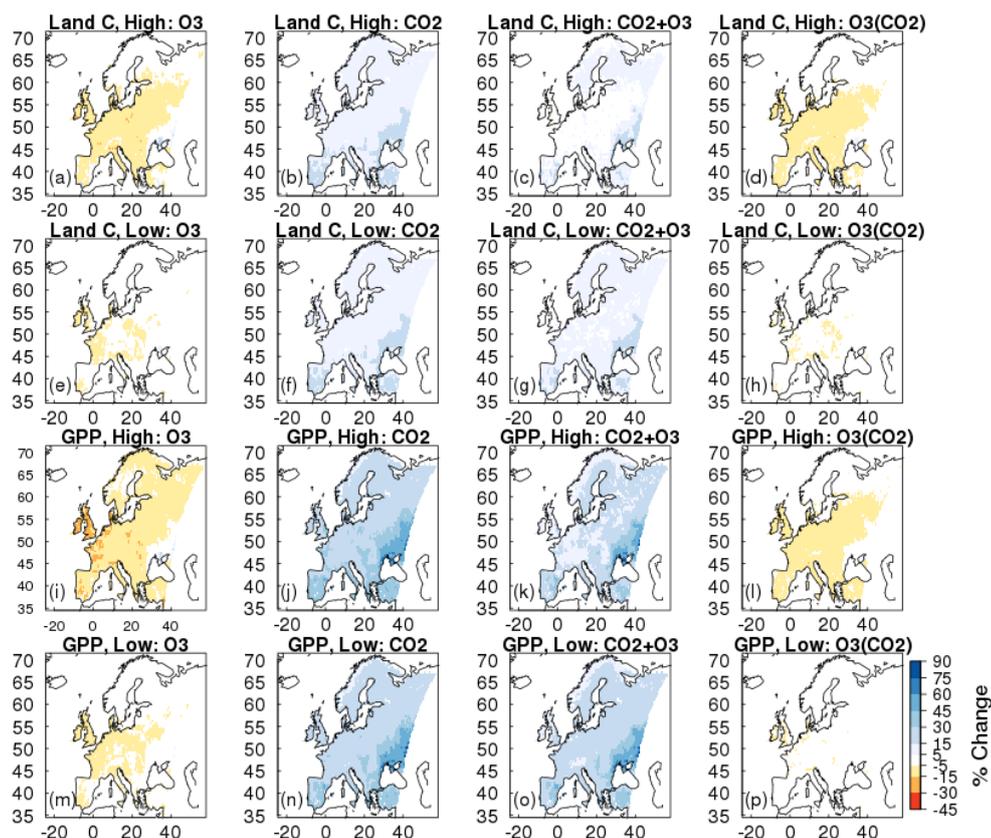



Figure 6. Simulated percentage change in total carbon stocks (Land C) and gross primary productivity (GPP) due to i) O_3 effects at fixed pre-industrial atmospheric CO_2 concentration (O_3), ii) CO_2 fertilisation at fixed pre-industrial O_3 concentration (CO_2), iii) the interaction between O_3 and CO_2 effects ($CO_2 + O_3$) iv) O_3 effects with changing atmospheric CO_2 concentration (i.e. O_3 damage accounting for the effect of CO_2 induced stomatal closure; $O_3(CO_2)$). Changes are depicted for the periods 1901 to 2050 for high and lower ozone plant sensitivity.

| | High Plant O_3 Sensitivity | | | | | |
| | 1901 - 2001 | | 2001 - 2050 | | 1901 - 2050 | |
	GPP (Pg C yr^{-1})	Land C (Pg C)	GPP (Pg C yr^{-1})	Land C (Pg C)	GPP (Pg C yr^{-1})	Land C (Pg C)
Value in 1901:	9.05	167	-	-	9.05	167
Absolute Change:						
O_3	-0.81	-9.21	0.01	-2.44	-0.80	-11.65
CO_2	1.16	4.24	1.42	12.98	2.58	17.22
$CO_2 + O_3$	0.13	-3.28	1.66	11.11	1.79	7.83
% Change:						
O_3	-8.95	-5.51	0.12	-1.55	-8.84	-6.98
CO_2	12.82	2.54	13.91	7.58	28.51	10.31
$CO_2 + O_3$	1.44	-1.96	18.08	6.79	19.78	4.69
	Low Plant O_3 Sensitivity					
	1901 - 2001		2001 - 2050		1901 - 2050	
	GPP (Pg C yr^{-1})	Land C (Pg C)	GPP (Pg C yr^{-1})	Land C (Pg C)	GPP (Pg C yr^{-1})	Land C (Pg C)
Value in 1901:	9.34	167.5	-	-	9.34	167.5
Absolute Change:						
O_3	-0.30	-3.59	0.02	-1.07	-0.40	-4.66
CO_2	1.15	6.43	1.35	13.14	2.50	19.57
$CO_2 + O_3$	0.65	2.50	1.50	12.35	2.15	14.85
% Change:						
O_3	-3.21	-2.14	0.22	-0.65	-4.28	-2.78
CO_2	12.31	3.84	12.87	7.55	26.77	11.68
$CO_2 + O_3$	6.96	1.49	15.02	7.26	23.02	8.87

480

481 **Table 1**. Simulated changes in the European land carbon cycle due to changing O_3 and CO_2 concentrations

482 (independently and together). Shown are changes in total carbon stocks (Land C) and gross primary productivity

483 (GPP), over three different periods (historical: 1901 to 2001, future: 2001 to 2050, and Anthropocene: 1901 to

484 2050). Absolute (top) and relative (bottom) differences are shown. For 2001 to 2050, please refer to Table S3 for

485 the initial value for each run. See the SI for details of the estimation of the O_3 and CO_2 effects and their interaction.

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	Mean (Pg C)				
	1970-1979	1980-1989	1990-1999	2000-2009	2002-2011
Modelled O₃ effect on land C sink :					
Higher sensitivity	-1.32	-1.01	-0.97	-0.53	-0.50
Low sensitivity	-0.71	-0.58	-0.50	-0.29	-0.26
Sum of C emissions from fossil fuel combustion and cement production (Pg C)	8.39	8.63	12.26	12.83	12.75
C lost from O₃ effect as a % of fossil fuel and cement emissions (%):					
Higher sensitivity	-15.73	-11.70	-7.91	-4.13	-3.92
Low sensitivity	-8.46	-6.72	-4.08	-2.26	-2.04

496

497 **Table 2.** Simulated change in total land carbon due to O_3 damage with changing atmospheric CO_2 concentration

498 for the two vegetation sensitivities. The sum of carbon emissions for each decade from fossil fuel combustion and

499 cement production for the EU-28 countries plus Albania, Bosnia and Herzegovina, Iceland, Belarus, Serbia,

500 Moldova, Norway, Turkey, Ukraine, Switzerland and Macedonia (EU28-plus) are shown, the data is from Boden

501 *et al.*, 2013. The simulated change in land carbon as a result of O_3 damage is depicted as a percentage of the EU28-

502 plus emissions to demonstrate the magnitude of the additional source of carbon to the atmosphere from plant O_3

503 damage.

504

505 **4 Discussion**

506

507 Comparison of the new g_s model implemented in this study (Medlyn *et al.*, 2011) with the g_s model currently used

508 as standard in JULES (Jacobs 1994) revealed large differences in leaf-level g_s for each PFT, principally as a result

509 of the data-based parameterisation of the new model. Leaf-level water use increased for the broadleaf tree and C_3

510 herbaceous PFTs using the Medlyn g_s model compared to Jacobs, but decreased for the needle leaf tree, C_4

511 herbaceous and shrub PFTs which displayed a more conservative water use strategy compared to the Jacobs

512 parameterisation. These changes are in line with the work of De Kauwe et al. (2015) who found a reduction in

513 annual transpiration for evergreen needle leaf, tundra and C_4 grass regions when implementing the Medlyn g_s

514 model into the Australian land surface scheme CABLE. Changes in leaf-level g_s in this study resulted in

515 differences in latent and sensible heat fluxes. Changes in the partitioning of energy fluxes at the land surface could

516 potentially have consequences for the intensity of heatwaves (Cruz et al., 2010;Kala et al., 2016), runoff (Betts et

517 al., 2007;Gedney et al., 2006) and rainfall patterns (de Arellano et al., 2012), although fully coupled simulations

518 would be necessary to detect these effects. The effect of the g_s model on simulated stomatal conductance led to

519 differences in the uptake of O_3 between the two g_s models because the leaf-level rate of g_s is the predominant

520 determinant of the flux of O_3 through stomata. Higher O_3 uptake is indicative of greater damage. Therefore, given

521 that C_3 herbaceous vegetation is the dominant land cover class across the European domain used in this study, this

522 suggests a greater O_3 impact for Europe could be simulated with the Medlyn g_s model, and that studies using the

523 Jacobs g_s formulation may underestimate the O_3 impact for Europe.

524

525   We compare results from the present study to values found in literature. The Wittig et al. (2007) meta-analysis of

526   temperate and boreal tree species showed future concentrations of O_3 predicted for 2050 significantly reduced leaf

527   level light saturated net photosynthetic uptake (-19%, range: -3% to -28%) and g_s (-10%, range: +5% to -23%) in

528   both broadleaf and needle leaf tree species. In the Feng et al. (2008) meta-analysis of wheat, projected O_3

529   concentrations for the future reduced aboveground biomass (-18%, CI -13% to -24%), photosynthetic rate (-20%)

530   and g_s (-22%). One of few long-term field based O_3 exposure studies (AspenFACE) showed that after 11 years of

531   exposing mature trees to elevated O_3 concentrations, O_3 decreased ecosystem carbon content (-9%), and decreased

532   NPP (-10%), although the O_3 effect decreased through time (Talhelm et al., 2014). GPP was reduced (-12% to -

533   19%) at two Mediterranean ecosystems (dominated by either *Pinus* species or *Citrus* species) studied by Fares et

534   al. (2013). Biomass of mature beech trees was reduced (-44%) after 8 years of exposure to elevated O_3 (Matyssek

535   et al., 2010a). After 5 years of O_3 exposure in a semi-natural grassland, annual biomass production was reduced

536   (-23%), and in a Mediterranean annual pasture O_3 exposure significantly reduced total aboveground biomass (up

537   to -25%) (Calvete-Sogo et al., 2014). Results from the present study suggest projected O_3 concentrations for 2050

538   will reduce mean GPP for Europe (-4% to -9%), NPP (-6% to -11%), total carbon content (-3% to -7%) and g_s (-

539   4% to -9%). Using GPP as a proxy for A_{sat} (these variables are not identical but they are related), our mean GPP

540   and g_s estimates fall within the range given by the meta-analysis of Wittig et al. (2007). The remaining studies are

541   not meta-analyses, so are site- and species- specific, our estimates appear to compare more conservatively with

542   these, however these are a mean value for Europe and spatially our estimates show greater variability.

543

544   The impact of O_3 on present day European GPP simulated in this study is slightly lower compared to previous

545   studies. Our estimates suggest present day O_3 reduced GPP by 3% to 9% on average across Europe and NPP by

546   5% to 11%. Anav et al. (2011) simulated a 22% reduction of GPP across Europe for 2002 using the ORCHIDEE

547   model. Present day O_3 exposure reduced GPP by 10% to 25% in Europe, and 10.8% globally in the study by

548   Lombardozzi et al. (2015) using the Community land model (CLM). O_3 reduced NPP by 11.2% in Europe from

549   1989 to 1995 using the Terrestrial Ecosystem Model (TEM) (Felzer et al., 2005). Globally, concentrations of O_3

550   predicted for 2100 reduced GPP by 14% to 23% using a former parameterisation of O_3 sensitivity in JULES (Sitch

551   et al., 2007). The recent study by Franz et al. (2017) showed mean GPP declined by 4.7% over the period 2001 to

552   2010 using the OCN model over the same European domain used in this study. These similarly 'lower than

553   expected' results are likely the result of using the same domain, and, more importantly, O_3 forcing produced by

554   the same model (EMEP MSC-W).

555

556   In this study, O_3 has a detrimental effect on the size of the land carbon sink for Europe. This is primarily through

557   a decrease in the size of the soil carbon pool as a result of reduced litter input to the soil, consistent with reduced

558   GPP/NPP. Field studies show that in some regions of Europe, soil carbon stocks are decreasing (Bellamy et al.,

559   2005;Capriel, 2013;Heikkinen et al., 2013;Sleutel et al., 2003). The study of Bellamy et al. (2005), for example,

560   showed that carbon was lost from soils across England and Wales between 1978 to 2003 at a mean rate of 0.6%

561   per year with little effect of land use on the rate of carbon loss, suggesting a possible link to climate change. It is

562   understood that climate change is likely to affect soil carbon turnover. Increased temperatures increase microbial

563   decomposition activity in the soil, and therefore increase carbon losses through higher rates of respiration (Cox et

564   al., 2000;Friedlingstein et al., 2006;Jones et al., 2003). However, some studies have found that O_3 can decrease

565   soil carbon content. Talhelm et al. (2014), for example, found O_3 reduced carbon content in near surface mineral

566   soil of forest soils exposed to 11 years of O_3 fumigation. Hofmockel et al. (2011) found elevated O_3 reduced the

567   carbon content in more stable soil organic matter pools, and Loya et al. (2003) showed that the fraction of soil

568   carbon formed in forest soils over a 4 year experimental period when fumigated with both CO_2 and O_3 was reduced

569   by 51% compared to the soil fumigated with CO_2 alone. It is agreed that amongst other factors that change with

570   O_3 exposure such as litter quality and composition, reduced litter quantity also has significant detrimental

571   consequences for soil carbon stocks (Andersen, 2003;Lindroth, 2010;Loya et al., 2003). Results from this study

572   therefore suggest that increasing tropospheric O_3 may be a contributing factor to the declining soil carbon stocks

573   observed across Europe as a result of reduced litter input to the soil carbon pool consistent with reduced NPP.

574

575   We acknowledge, however, that our model simulations do not include coupling of Nitrogen and Carbon cycles,

576   or land management practices. Although we include a representation of agricultural regions through the model

577   calibration against the wheat O_3 sensitivity function (CLRTAP, 2017), wheat is known to be one of the most O_3

578   sensitive crop species. As with all uncoupled modelling studies, a change in g_s and flux will impact the O_3

579   concentration itself. Therefore adopting the Medlyn formulation with a higher g_s and subsequently higher O_3 flux

580   for broadleaf and C_3 PFTs (Fig 2) would lead to reduced O_3 concentration, which in turn would act to dampen the

581   effect of higher g_s on O_3 flux. Essentially this study provides an 'upper bound' as in the high plant O_3 sensitivity

582   scenario, all C_3/C_4 fractional cover uses the wheat O_3 sensitivity. Additionally, this version of JULES does not

583   have a crop module; it has no land management practices such as harvesting, ploughing or crop rotation –

584   processes which may have counteracting effects on the land carbon sink. Further, without a coupled Carbon and

585   Nitrogen cycle, it is likely that the CO_2 fertilisation response of GPP and the land carbon sink is over estimated in

586   some regions of our simulations since nitrogen availability limits terrestrial carbon sequestration of natural

587   ecosystems in the temperate and boreal zone (Zaehle, 2013). This would have consequences for our modelled O_3

588   impact, particularly into the future where the large CO_2 fertilisation effect was responsible for partly offsetting

589   the negative impact of O_3. Although in our simulations a high fraction of land cover is agricultural which we

590   assume would be optimally fertilised. Nevertheless, we emphasise that this study provides a sensitivity assessment

591   of the impact of plant O_3 damage on GPP and the land carbon sink.

592

593   Another caveat we fully acknowledge is that at the leaf-level JULES is parameterised to reduce g_s with O_3

594   exposure. Whilst this response is commonly observed (Wittig et al., 2007;Ainsworth et al., 2012), there is evidence

595   to suggest that O_3 impairs stomata in some species, making them non-responsive to environmental stimuli (Hayes

596   et al., 2012;Hoshika et al., 2012;Mills et al., 2009;Paoletti and Grulke, 2010). In drought conditions the mechanism

597   is thought to involve O_3 stimulated ethylene production which interferes with the stomatal response to ABA

598   signalling (Wilkinson and Davies, 2009;Wilkinson and Davies, 2010). Such stomatal sluggishness can result in

599   higher O_3 uptake and injury, increased water-loss, and therefore greater vulnerability to environmental stresses

600   (Mills et al., 2016). McLaughlin (2007a;2007b) and Sun et al. (2012) provide evidence of increased transpiration

601   and reduced streamflow in forests at the regional scale in response to ambient levels of O_3, and suggest this could

602   increase the frequency and severity of droughts. To our knowledge, the study of Hoshika et al. (2015) is the first

603   to include an explicit representation of sluggish stomatal control in a land-atmosphere model, they show that

604   sluggish stomatal behaviour has significant implications for carbon and water cycling in ecosystems. However, it

605 is by no means a ubiquitous response, and it is not fully understood which species respond this way and under

606 what conditions (Mills et al., 2016;Wittig et al., 2007). Nevertheless, this remains an important area of future

607 work.

608

609 Comprehensive analyses of the European carbon balance suggest a significant biogenic carbon sink (Janssens et

610 al., 2003;Luyssaert et al., 2012;Schulze et al., 2009). However, estimates are hampered by large uncertainties in

611 key components of the land carbon balance, such as estimates of soil carbon gains and losses (Ciais et al.,

612 2010;Janssens et al., 2003;Schulze et al., 2009;Schulze et al., 2010). We suggest that the effect of O_3 on plant

613 physiology is a contributing factor to the decline in soil carbon stores observed across Europe, and as such this O_3

614 effect is a missing component of European carbon cycle assessments. Over the Anthropocene, our results show

615 elevated O_3 concentrations reduce the amount of carbon that can be stored in the soil by 3% to 9% (low and high

616 plant O_3 sensitivity, respectively), which almost completely offsets the beneficial effects of CO_2 fertilisation on

617 soil carbon storage under the high plant O_3 sensitivity . This would contribute to a significant change in the size

618 of a key carbon sink for Europe, and is particularly important when we consider the evolution of the land carbon

619 sink into the future given the impact of O_3 on soil carbon sequestration and the high uncertainty of future

620 tropospheric O_3 concentrations. Schulze et al. (2009) and Luyssaert et al. (2012) extended their analysis of the

621 European carbon balance to include additional non-CO_2 greenhouse gases (CH_4 and N_2O). Both studies found

622 that emissions of these offset the biogenic carbon sink, reducing the climate mitigation potential of European

623 ecosystems. This highlights the importance of accounting for all fluxes and stores in carbon/greenhouse gas

624 balance assessments, of which O_3 and its indirect effect on the CO_2 flux via direct effects on plant physiology is

625 currently missing.

626

627 We looked at the complex interaction between CO_2 and O_3 effects. Our results support the hypothesis that elevated

628 atmospheric CO_2 provides some protection against O_3 damage because of lower g_s that reduces uptake of O_3

629 through stomata (Harmens et al., 2007;Wittig et al., 2007). In the present study, reductions in GPP and the land

630 carbon store due to O_3 exposure were lower when simulated with concurrent changes in atmospheric CO_2. Despite

631 acclimation of photosynthesis after long-term exposure to elevated atmospheric CO_2 of field grown plants

632 (Ainsworth and Long, 2005;Medlyn et al., 1999), there is no evidence to suggest that g_s acclimates (Ainsworth et

633 al., 2003;Medlyn et al., 2001). This suggests the protective effect of elevated atmospheric CO_2 against O_3 damage

634 will be sustained in the long term. However, although meta-analysis suggest a general trend of reduced g_s with

635 elevated CO_2 (Ainsworth and Long, 2005;Medlyn et al., 1999), this is not a universal response. Stomatal responses

636 on exposure to elevated CO_2 with FACE treatment varied with genotype and growth stage in a fast-growing poplar

637 community (Bernacchi et al., 2003;Tricker et al., 2009). In other mature forest stands, limited stomatal response

638 to elevated CO_2 was observed after canopy closure (Ellsworth, 1999;Uddling et al., 2009). Also, some studies

639 found that stomatal responses to CO_2 were significant only under high atmospheric humidity (Cech et al.,

640 2003;Leuzinger and Körner, 2007;Wullschleger et al., 2002). These examples illustrate that stomatal responses to

641 elevated atmospheric CO_2 are not universal, and as such the protective effect of CO_2 against O_3 injury cannot be

642 assumed for all species, at all growth stages under wide ranging environmental conditions.

643

644 **5 Conclusion**

645

646   What is abundantly clear is that plant responses to both CO_2 and O_3 are complicated by a host of factors that are
647   only partly understood, and it remains difficult to identify general, global patterns given that effects of both gases
648   on plant communities and ecological interactions are highly context and species specific (Ainsworth and Long,
649   2005;Fuhrer et al., 2016;Matyssek et al., 2010b). This study quantifies the sensitivity of the land carbon sink for
650   Europe and GPP to changing concentrations of atmospheric CO_2 and O_3 from 1901 to 2050. We have used a state
651   of the art land surface model calibrated for European vegetation to give our best estimates of this sensitivity within
652   the limits of data availability to calibrate the model for O_3 sensitivity, current knowledge and model structure. In
653   summary, this study has shown that potential gains in terrestrial carbon sequestration over Europe resulting from
654   elevated CO_2 can be partially offset by concurrent rises in tropospheric O_3 over 1901-2050. Specifically, we have
655   shown that the negative effect of O_3 on the land carbon sink was greatest over the twentieth century, when O_3
656   concentrations increased rapidly from pre-industrial levels. Over this period soil carbon stocks were significantly
657   diminished over agricultural areas, consistent with reduced NPP and litter input. This loss of soil carbon was
658   largely responsible for the decrease in the size of the land carbon sink over Europe. The O_3 effect on the land
659   carbon store and flux was reduced into the future as CO_2 concentration rose considerably and changes in O_3
660   concentration were less pronounced. However, there remained a large cumulative negative impact on the land
661   carbon sink for Europe by 2050. The interaction between the two gases was found to reduce O_3 injury owing to
662   reduced stomatal opening in elevated atmospheric CO_2. However, primary productivity and land carbon storage
663   remained significantly suppressed by 2050 due to plant O_3 damage. Expressed as a percentage of the emissions
664   from fossil fuel and cement production for the EU28-plus countries, the additional carbon emissions from O_3-
665   induced plant injury is a potential significant additional source of anthropogenic carbon. Our results demonstrate
666   the sensitivity of modelled terrestrial carbon dynamics to tropospheric O_3 and its interaction with atmospheric
667   CO_2, highlighting that such effects of O_3 on plant physiology significantly add to the uncertainty of future trends
668   in the land carbon sink and climate-carbon feedbacks. Given the potential to limit the climate mitigation effect of
669   European terrestrial ecosystems, we suggest plant O_3 damage should be incorporated into carbon cycle
670   assessments.

671

672   **Data availability**

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674   The JULES model can be downloaded from the Met Office Science Repository Service
675   (https://code.metoffice.gov.uk/trac/jules - see here for a helpful how to http://jules.jchmr.org/content/getting-
676   started). Model output data presented in this paper and the exact version of JULES with namelists are available
677   upon request from the corresponding author.

678

679   **Supplementary Information**

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683   **Competing Interests**

684   The authors declare that they have no conflict of interest

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