Suggestions for revision or reasons for rejection **(will be published if the paper is accepted for final publication)**

I find the manuscript improved in the second round of submission. However, there are still major revisions necessary. But the only new analysis that I suggest is a supplementary table or figure showing changes in major ozone precursor emissions throughout the time period analyzed. The other major revisions I suggest involve word-choice, organization, and clarity of the manuscript, which I think need substantial improvement before publication. I would like to see more discussion of the calibration of the ozone damage parameterization discussed and model evaluation of the high vs. low ozone sensitivity simulations in the main text (especially in regards to the implications of the model evaluation for the investigation here), as they are central to the novelty of the study and thus its findings. I also think the authors need to describe supplemental material and findings in the main text; not only refer the readers to them without context.

We would like to thank the reviewer for their detailed reading of the manuscript and suggestions for edits to improve the manuscript. We hope that the reviewer will find the manuscript much improved again. We have provided the additional figure in the SI (figure S5) showing the trend in precursor emissions of CH_4, NO_X, NMVOC and Isoprene from 1900 to 2050 over Europe. The remaining revisions were largely to do with word-choice, organisation and clarity, and we hope we have sufficiently improved this. We have moved a lot of information from the SI to the main text to help with clarity, and to ensure some of the key points, such as the model evaluation, are discussed in the main text instead of the SI. We address the comments line by line below.

To help readers, I encourage the authors to name their sensitivity simulations and refer to their sensitivity simulations by these names, as opposed to saying things like "the CO2 only run", or "O3 and CO2 simulations", "varying CO2 and O3 together", as these phrases are rather ambiguous.

We have hopefully clarified this. We call our simulations O3, CO2 and CO2+O3 and we now refer to our simulations using these names.

Line by line comments:

Line 31: I disagree that the "impact of the gas on European vegetation and the land carbon sink is largely unknown" - the authors show in their discussion of the literature that there is a substantial amount of work on this. I urge the authors to motivate their work in a way that complements the previous work.

We have removed this sentence and replaced it with the following to clarify the motivation behind our work and its novelty (Lines 33 to 40):

"Studying the impact of O_3 on European vegetation at the regional scale is important for gaining greater understanding of the impact of O_3 on the land carbon sink at large spatial scales. In this work we take a regional approach and update the JULES land-surface model using new measurements specifically for European vegetation. Given the importance of stomatal conductance in determining the flux of O_3 into plants, we implement an alternative stomatal closure parameterization and account for diurnal variations in O_3 concentration in our simulations. We conduct our analysis specifically for the European region to quantify the impact of tropospheric O_3, and its interaction with CO_2, on gross primary productivity (GPP) and land carbon storage across Europe."

Line 35: I don't think the authors can call their new stomatal conductance parameterization "an improved" one. I would suggest finding another way to describe it.

We have changed this to describe it as the following (Line 37):

"an alternative stomatal closure parameterization".

Line 41: Where is this discussed in the paper?

We have removed this sentence.

Line 82: Please cite papers showing that ozone damage is "key" take into account

We have changed key to important, and have added two references here (Line 83/84):

(Le Quéré et al., 2016;Sitch et al., 2015).

Line 98-101: Please clarify the relevance of this text to the analysis

The text in this paragraph is discussing the observed changes in O_3 concentration through the 20th century. The text in these two lines is discussing future O_3 concentrations - they will depend on emissions of O_3 precursors, of which intercontinental transport is an important factor, and climate change which may increase the occurrence of peak O_3 episodes, and the emission of O_3 precursors isoprene and NO_X.

Lines 107-108: The authors should revise their phrasing here.

This has been amended to read (Line 108):

" Intercontinental transport means future O_3 concentrations in Europe will be partly dependent on how O_3 precursor emissions evolve globally."

Line 118: A paper that examines the ozone budget and source/sink terms would be more appropriate to reference here. Fowler et al. (2009) can be the citation for "primarily uptake by … vegetation", but these papers don't show that it's an important sink. Admittedly it's hard to find papers with the surface ozone budget, but there are many papers that examine the parts of the tropospheric ozone budget.

We moved the Fowler et al., references and added the Young et al., 2013 and Wild 2007 reference (Line 120 - 121):

"Dry deposition of O_3 to terrestrial surfaces, primarily uptake by stomata on plant foliage and deposition on external surfaces of vegetation (Fowler et al., 2001;Fowler et al., 2009), is a large sink for ground level O_3 (Wild, 2007; Young et al., 2013)."

Line 131-132: Instead of saying "future concentrations of ozone predicted for 2050", the authors should give the concentration, as these numbers are highly dependent on emission scenario and not necessarily generalizable to a given year.

This paragraph has been updated to add specific O_3 concentrations (Line 131 - 148):

"The response of plants to O_3 is very wide ranging as reported in the literature from different field studies. The Wittig et al. (2007) meta-analysis of temperate and boreal tree species showed future concentrations of O_3 predicted for 2050 significantly reduced leaf level light saturated net photosynthetic uptake (-19%, range: -3% to -28% at a mean O_3 concentration of 85 ppb) and g_s (-10%, range: +5% to -23% at a mean O_3 concentration of 91 ppb) in both broadleaf and needle leaf tree species. In the Feng et al. (2008) meta-analysis of wheat, projected O_3 concentrations for the future reduced aboveground biomass (-18% at a mean O_3 concentration of 70 ppb) photosynthetic rate (-20% at a mean O_3 concentration of 73 ppb) and g_s (-22% at a mean O_3 concentration of 79 ppb). One of few long-term field based O_3 exposure studies (AspenFACE) showed that after 11 years of exposing mature trees to elevated O_3 concentrations (mean O_3 concentration of 46 ppb), O_3 decreased ecosystem carbon content (-9%), and decreased NPP (-10%), although the O_3 effect decreased through time (Talhelm et al., 2014). Zak et al. (2011) showed this was partly due to a shift in community structure as O_3-tolerant species, competitively inferior in low O_3 environments, out competed O_3-sensitivie species. GPP was reduced (-12% to -19%) at two Mediterranean ecosystems exposed to high ambient O_3 concentrations (ranging between 20 to 72 ppb across sites and through the year) studied by Fares et al. (2013). Biomass of mature beech trees was reduced (-44%) after 8 years of exposure to elevated O_3 (~150 ppb) (Matyssek et al., 2010a). After 5 years of O_3 exposure (ambient +20 to +40 ppb) in a semi-natural grassland, annual biomass production was reduced (-23%), and in a Mediterranean annual pasture O_3 exposure significantly reduced total aboveground biomass (up to -25%) (Calvete-Sogo et al., 2014)."

Line 150: What are the "new measurements"? The g1 parameter? g1 can't be measured, only inferred. Please revise.

Here we are referring to the data we use to calibrate JULES for plant sensitivity to O_3 uptake. This wasn't clear in the text so we have revised accordingly (Line 154 to 157):

“Here we take a regional approach and take advantage of the latest measurements showing changes in plant productivity with exposure to O_3 specifically for a range of European vegetation from different regions (CLRTAP 2017) with which to calibrate the JULES model for plant sensitivity to O_3 , and conduct a dedicated analysis for the European region.”

Line 151: “conduct a dedicated analysis” has little meaning. Please revise.

This has been changed to (Line 157):

“and conduct our analysis specifically for the European region.”

Line 157-158: I would cut everything in this sentence starting with “such that” because I think that it implies independent responses.

This has been removed.

Line 169-170: Here the phrase about not including stomatal sluggishness is a bit awkward.

This phrase has been removed (Line 173):

“This model is based on the optimal theory of stomatal behaviour and has the following advantages over the current JULES g_s formulation of Jacobs (1994):…..”

Line 167-176: This is a rather technical paragraph for the introduction. I wonder if the authors could illustrate the novelty of the study without as much jargon.

We have revised this paragraph to remove the jargon (Line 172 - 178):

“Given the critical role g_s plays in the uptake of both CO_2 and O_3, we use an alternative representation and parameterisation of g_s in JULES by implementing the Medlyn *et al*. (2011) g_s formulation. This model is based on the optimal theory of stomatal behaviour and has advantages over the current JULES g_s formulation of Jacobs (1994) including i) a single parameter (g_1) compared to two parameters in Jacobs (1994), ii) the g_1 parameter is related to the water-use strategy of vegetation and is easier to parameterise with commonly measured leaf or canopy level observations of photosynthesis, g_s and humidity, and (iii) values of g_1 are available for many different plant functional types (PFTs) derived from a global data set of leaf-level measurements (Lin et al., 2015).“

Line 183: “look at the interaction between O3 and CO2” is ambiguous. Same thing for Lines 196-197, and other points in the text. Please revise.

This has been revised to the following (Line 185):

“……to investigate the impact of both O_3 and CO_2 on plant water-use and carbon uptake.”

Line 183-185: As discussed below, I don’t think this is a reason for why this study is novel, and urge the authors to cut this from the introduction.

This has been removed.

Line 189: I’m not certain how the high and low ozone sensitivity simulations represent the large variation within and between species specifically, rather than just the large uncertainty in the ozone response generally. Please clarify that for both the high and low sensitivity simulations, there is a distinction between the sensitivities of crops vs. grasslands. What about forests?

This paragraph has been clarified (Line 186 - 193):

“In this work, the JULES model is re-calibrated using the latest observations of vegetation sensitivity to O_3, with the addition of a separate parameterisation for temperate/boreal regions versus the Mediterranean. The O_3 sensitivity of each PFT in JULES was re-calibrated for both a high and low sensitivity to account for uncertainty in the O_3 response, in part due to the observed variation in O_3 sensitivity between species. This includes O_3 sensitivities for agricultural crops (wheat – high sensitivity) versus natural grassland (low sensitivity), with separate sensitivities for Mediterranean grasslands. For forests JULES is parameterised with O_3 sensitivities for broadleaf and needle leaf trees (both high and low O_3 sensitivity), with separate sensitivities (high and low) for Mediterranean broadleaf species."

Line 193-6: Please clarify here that the authors are forcing with daily ozone concentrations that are scaled to a diurnal cycle. The authors' phrasing implies that hourly concentrations are archived and used to force the model.

The phrasing has been amended here (Line 196 - 197):

"JULES is forced with spatially varying daily O_3 concentrations from a high resolution atmospheric chemistry model for Europe that are disaggregated to hourly concentrations,……"

Line 199-201: The authors should also note here that not using coupled chemistry and climate also creates additional uncertainty.

A sentence has been added (Line 204 -205):

"In addition, using uncoupled chemistry and climate is a further source of uncertainty."

Line 216: The order of the supplemental figures should reflect the order that they are mentioned in the main text

This has been changed.

Line 226: Lombardozzi and colleagues's work shows that there are separate impacts of ozone on stomatal vs. photosynthesis. This merits mention, as this is how ozone damage is configured in one of the only other land surface models with ozone damage. Ozone damage is also a function of cumulative ozone exposure, rather than an instantaneous effect. Please comment on this.

We have amended this paragraph as follows to mention other models that include O_3 damage and discuss O_3 damage as a function of cumulative O_3 exposure (Line 227 - 240):

"To simulate the effects of O_3 deposition on vegetation productivity and water use, JULES uses the flux-gradient approach of Sitch *et al*., (2007), modified to include non-stomatal deposition following Tuovinen et al. (2009). A similar approach is taken by Franz et al. (2017) in the OCN model, however plant O_3 damage is a function of accumulated O_3 exposure over time. In JULES, plant O_3 damage is instantaneous, the degree to which photosynthesis and g_s are modified at each time step with O_3 exposure having already been calibrated against observations of the change in plant productivity with cumulative O_3 exposure for each PFT (i.e. O_3 dose-response functions described later). JULES uses a coupled model of g_s and photosynthesis, the potential net photosynthetic rate (A_p, mol CO_2 m^{-2} s^{-1}) is modified by an 'O_3 uptake' factor (*F,* the fractional reduction in photosynthesis), so that the actual net photosynthesis (A_{net}, mol CO_2 m^{-2} s^{-1}) is given by equation 1 (Clark *et al.,* 2011, Sitch *et al.,* 2007). Because of the relationship between these two fluxes, the direct effect of O_3 damage on photosynthetic rate also leads to a reduction in g_s. An alternative approach was taken by Lombardozzi et al. (2012) in the CLM model where photosynthesis and g_s are decoupled, so that O_3 exposure affects carbon assimilation and transpiration independently. In JULES, changes in atmospheric CO_2 concentration also affect photosynthetic rate and g_s, consequently the interaction between changing concentrations of both CO_2 and O_3 is allowed for."

Line 247-249: Nonstomatal conductances are highly variable and substantially larger than this single prescribed value on average across sites. Including this term as 0.04 cm/s should only decrease stomatal uptake by a very small amount. I disagree that this adds any value to the authors' study.

Non-stomatal resistances are highly variable and uncertain. This was discussed extensively for the original EMEP formulation in Emberson et al. (2000), where the choice of g_{ext} = 0.04 cm/s was explained, and EMEP has so far retained the same choice due to the uncertainties of alternative formulations (e.g. Tuovinen et al., 2009). As a first approach in JULES, for this work we followed the same approach to add a term for non-stomatal deposition of O_3. We appreciate there are more complex processes involved, and this is an area for development within JULES. However, for this work it represents a significant step on from previous studies with JULES where non-stomatal conductance wasn't considered, and as such it requires mentioning here. The O_3 forcing we use is at canopy height from the EMEP model which includes many of the complex processes in the resistance network already.

Emberson, L., Simpson, D., Tuovinen, J.-P., Ashmore, M. & Cambridge, H. Towards a model of ozone deposition and stomatal uptake over Europe The Norwegian Meteorological Institute, Oslo, Norway, The Norwegian Meteorological Institute, Oslo, Norway, 2000

Tuovinen, J.-P., Emberson, L., and Simpson, D.: Modelling ozone fluxes to forests for risk assessment: status and prospects, Annals of Forest Science, 66, 1-14, 2009.

Line 253: How are leaf dimensions defined?

Leaf dimensions are defined per PFT, this has been added to the text as follows (Line 263 - 264):

"*Ld* is the cross-wind leaf dimension (m) defined per PFT as 0.05 for trees, 0.02 for grasses (C_3 and C_4) and 0.04 for shrubs,"

Line 262: What about the resistance to turbulence in the canopy? This is highly uncertain, but can be substantial. Could it be that too much ozone is getting deep into the canopy?

JULES uses an O_3 concentration at reference level, this does not change with depth into the canopy due to changes in turbulence and mixing. This is something that needs consideration for development within JULES as it is possible that leaves at the bottom of the canopy will see too much O_3. But currently, because this process is highly uncertain, in JULES the simplest approach is taken whereby each layer of the canopy has the same O_3 concentration and the uptake of O_3 is dependent on the rate of stomatal conductance at that canopy layer, which does vary with depth into the canopy depending on light and Nitrogen availability. This would not be an issue for short vegetation such as grasslands and crops which are the dominant land cover in our study, but may be more significant for forests. However, some studies show minimal vertical O_3 concentration gradients within forest canopies. For example, Karlsson et al (2006) found daytime mean O_3 of 34.5 ppb at 13m, and 33.1 ppb at 3m, in an 18-20m tall Norway Spruce forest. The 13m values were just 4% lower than measurements made at 13m in clearing, suggesting rather uniform conditions. The same study reported a 6% difference in O_3 between 10m and 20m observations in a separate 20-25m Norway spruce site. Reactions with biogenic VOC emissions can also reduce O_3 in the canopy, but even at a chemically very reactive oak forest in the USA, O_3 concentrations gradients within the upper canopy were small (Fuentes et al, 2007).

Karlsson, P., Hansson, M., Hoglund, H.-O. & Pleijel, H. Ozone concentration gradients and wind conditions in Norway spruce (Picea abies) forests in Sweden Atmos. Environ., 2006, 40, 1610-1618.

Fuentes, J. D., Wang, D., Bowling, D. R., Potosnak, M., Monson, R. K., Goliff, W. S. & Stockwell, W. R. Biogenic hydrocarbon chemistry within and above a mixed deciduous forest J. Atmos. Chem., 2007, 56, 165-185.

Line 270-273: I'm finding this hard to follow, especially because how the authors refer to the sensitivity simulations is "high/low plant ozone sensitivity". I understand that within each sensitivity simulation there are variations among land cover types in terms of the degree of the sensitivity to ozone applied. Further, ozone "dose-response functions" is never defined. Again, it seems like this calibration is a fundamental part of the authors' analysis. I would suggest that some supplemental material is moved to the main text and cleaned up so the methods are very clear.
Line 286-9: What observations? I am certain FO3crit cannot be measured, only inferred. Cumulative ozone uptake over what time period?

To address both points above (Line 270 to 289), we have amended section 2.2 (Calibration of O_3 uptake model) to improve clarity and have moved information for the SI into the main text (Line 277 - 352):

“Here we use the latest literature on flux based O_3 dose-response relationships derived from observed field data across Europe (CLRTAP, 2017) to determine the key PFT-specific O_3 sensitivity parameters in JULES (*a* and Fo_{3crit}). Synthesis of information expressed as O_3 flux based dose-response relationships derived from field experiments is carried out by The United Nations Convention on Long-Range Transboundary Air Pollution (CLRTAP Convention), this information is then used as a policy tool to inform emission reduction strategies in Europe to improve air quality (CLRTAP, 2017;Mills et al., 2011a). Derivation of O_3 flux based dose-response relationships for different vegetation types uses the accumulated stomatal O_3 flux above a threshold (often referred to as the phytotoxic O_3 dose above a threshold of ‘y’ i.e. POD_y) as the dose metric, and the percentage change in biomass as the response metric (Emberson et al., 2007;Karlsson et al., 2007). We use these observation based O_3 dose-response relationships to calibrate each JULES PFT for sensitivity to O_3 using available relationships for the closest matching vegetation type. For JULES, Fo_{3crit} is the threshold for O_3 damage, and values for this parameter are taken from the O_3 dose-response relationships as the POD_y value. The actual sensitivity to O_3 is determined by the slope of the O_3 dose-response relationship, i.e. how much biomass changes with accumulated stomatal uptake of O_3 above the damage threshold, this relates to the parameter *a* in JULES. The parameter ‘*a*’ is a PFT-specific parameter representing the fractional reduction of photosynthesis with O3 uptake by leaves. Values for this parameter are found for each PFT by running JULES with different values of ‘*a*’, which alter the instantaneous photosynthetic rate, but then calculating the accumulated stomatal flux of O_3 and resulting change in productivity over the same period, until the slope of this relationship produced by the JULES simulations matches that of the O_3 dose-response relationships derived from observations. Essentially we calibrate each JULES PFT for sensitivity to O_3 by reproducing the observed O_3 dose-response relationships.

Each PFT was calibrated for a high and low plant O_3 sensitivity to account for uncertainty in the sensitivity of different plant species to O_3, using the approach of Sitch *et al*., (2007). Therefore, when using our results to assess the impact of O_3 at the land surface, we are able to provide a range in our estimates to help address some of the uncertainty in the O_3 response of different vegetation types. In addition, where possible owing to available data, a distinction was made for Mediterranean regions. This was because the work of Büker et al. (2015) showed that different O_3 dose-response relationships are needed to describe the O_3 sensitivity of dominant Mediterranean trees. For the C_3 herbaceous PFT, the dominant land cover type across the European domain in this study (Fig. S1), the high plant O_3 sensitivity was calibrated against observations for wheat to give a representation of agricultural regions and wheat is one of the most sensitive grasses to O_3 (Fig. S2, Table S1). For the low plant O_3 sensitivity JULES was calibrated against the dose-response function for natural grassland to give a representation of natural grassland and this vegetation has a much lower sensitivity to O_3 damage, for the Mediterranean region we used a function for Mediterranean natural grasslands, all taken from CLRTAP (2017) (Fig. S2, Table S1). Tree/shrub PFTs were calibrated against observed O_3 dose-response functions for the high plant O_3 sensitivity: broadleaf trees (temperate/boreal) = Birch/Beech dose-response relationship, broadleaf trees (Mediterranean) = deciduous oaks dose-response relationship, needle leaf trees = Norway spruce dose-response relationship, shrubs = Birch/Beech dose-response relationship, all from CLRTAP (2017) (Fig. S2, Table S1). Data on O_3 dose-response relationships for different vegetation types is very limited, therefore for the low plant O_3 sensitivity calibration for trees/shrubs we assumed a 20% decrease in sensitivity to O_3 based on the difference in sensitivity between high and low sensitive tree species in the Karlsson et al. (2007) study. Due to limitations in data availability, the shrub parameterisation uses the observed dose-response functions for broadleaf trees. Similarly, the parameterisation for C_4 herbaceous uses the observed dose-responses for C_3 herbaceous, however the fractional cover of C_4 herbs across Europe is low (Fig. S1), so this assumption affects a very small percentage of land cover.

To calibrate the JULES O_3 uptake model, JULES was run across Europe forced using the WFDEI observational climate dataset (Weedon, 2013) at 0.5^o X 0.5^o spatial and three hour temporal resolution. JULES uses interpolation to disaggregate the forcing data down from 3 hours to an hourly model time step. The model was spun-up over the period 1979 to 1999 with a fixed atmospheric CO_2 concentration of 368.33 ppm (1999 value from Mauna Loa observations, (Tans and Keeling)). Zero tropospheric ozone concentration was assumed for the control simulation, for the simulations with O_3, spin-up used spatially explicit fields of present day O_3

concentration produced using the UK Chemistry and Aerosol (UKCA) model with standard chemistry from the run evaluated by O'Connor et al. (2014). A fixed land cover map was used based on IGBP (International Geosphere-Biosphere Programme) land cover classes (IGBP-DIS), therefore as the vegetation distribution was fixed and the calibration was not looking at carbon stores, a short spin-up was adequate to equilibrate soil temperature and soil moisture. JULES was then run for the year 2000 with a corresponding CO_2 concentration of 369.52 ppm (from Mauna Loa observations, (Tans and Keeling)) and monthly fields of spatially explicit tropospheric O_3 (O'Connor et al., 2014) as necessary.

Calibration was performed using four simulations: with i) zero tropospheric O_3 concentration, this was the control simulation (control), ii) tropospheric O_3 at current ambient concentration (O3), iii) ambient +20 ppb (O3+20) and iv) ambient +40 ppb (O3+40). The different O_3 simulations (i.e. O3, O3+20 and O3+40) were used to capture the range of O_3 conditions in the data used to derive the observed O_3 dose-response relationships used here for calibration, often data were used from experiments using artificially manipulated conditions of ambient + 40 ppb O_3 for example. For each JULES O_3 simulation, the value of F_{O3crit} was taken from the vegetation specific O_3 dose-response relationship as the threshold O_3 concentration above which damage to vegetation occurs. An initial estimate of the parameter '*a*' was used, then for each PFT and each simulation, hourly estimates of NPP (our proxy for biomass – although not identical they are related) and O_3 uptake in excess of F_{O3crit} were accumulated over a PFT dependent accumulation period. The accumulation periods were ~6 months for broadleaf trees and shrubs, all year for needle leaf trees, and ~3 months for herbaceous species, through the growing season, following guidelines in CLRTAP (2017). Additionally, in accordance with the methods used in the CLRTAP (2017) that describe how the O_3 dose-response relationships are derived from observations, we use the stomatal O_3 flux per projected leaf area to top canopy sunlit leaves. The percentage change in total NPP was calculated for each O_3 simulation and plotted against the cumulative uptake of O_3 over the PFT-specific accumulation period. The linear regression of this relationship was calculated, and slope and intercept compared against the slope and intercept of the observed dose-response relationships. Values of the parameter 'a' were adjusted, and the procedure repeated until the linear regression through the simulation points matched that of the observations (Fig. S2, Table S1)."

Line 317-324: This discussion seems out-of-place here. It might be more appropriate in the conclusions w.r.t. the "next steps", please revise.

This has been moved, and is a new paragraph in section 4.3 (Line 879 - 887):

"In this work we implement the stomatal closure proposed in Medlyn et al., (2011), this uses the parameter g_1. Hoshika et al. (2013) show a significant difference in the g_1 parameter (higher in elevated O_3 compared to ambient) in Siebold's beech in June of their experiment. However, this is only at the start of the growing season, further measurements show no difference in this parameter between O_3 treatments. Quantifying an O_3 effect directly on g_1 would require a detailed meta-analysis of empirical data on photosynthesis and g_s for different PFTs, which is currently lacking in the literature. With such information, here we take an empirical approach to modelling plant O_3 damage, essentially by applying a reduction factor to the simulated plant photosynthesis based on observations of whole plant losses of biomass with O_3 exposure, for which there is a lot more available data (e.g. CLRTAP, 2017)."

Line 324-325. Please include more details in the main text as to why the author would go to the supplemental for this analysis.

We have amended the main text as follows, and moved information from the SI into the main text (Line 382 - 401):

"The impact of gs model formulation (JAC versus MED) on simulated water, O3, carbon and energy fluxes is compared for two contrasting grid points - wet (low soil moisture stress) and dry (high soil moisture stress) in the European domain. JULES was spun-up for 20 years (1979-1999) at two grid points in central Europe representing a wet (low soil moisture stress, lat: 48.25; lon:, 5.25) and a dry site (high soil moisture stress, lat:

38.25; lon:, -7.75). The modelled soil moisture stress factor (fsmc) at the wet site ranged from 0.8 to 1.0 over the year 2000 (1.0 indicates no soil moisture stress), and at the dry site fsmc steadily declined from 0.8 at the start of the year to 0.25 by the end of the summer. The WFDEI meteorological forcing dataset was used (Weedon, 2013), along with atmospheric CO2 concentration for the year 1999 (368.33 ppm), and either no O3 (i.e. the O3 damage model was switched off) for the control simulations, or spatially explicit fields of present day O3 concentration produced using the UK Chemistry and Aerosol (UKCA) model from the run evaluated by O'Connor et al. (2014) for the simulations with O3. Following the spin-up period, JULES was run for one year (2000) with corresponding atmospheric CO2 concentration, and tropospheric O3 concentrations as described above. The control and ozone simulations were performed for both JAC and MED model formulations. Land cover for the spin-up and main run was fixed at 20% for each PFT. For the simulations including O3 damage, the high plant O3 sensitivity parameterisation was used. The difference between these simulations was used to assess the impact of gs model formulation on the leaf level fluxes of carbon and water. We calculate and report in the main manuscript (results section 3.1), the difference in mean annual water-use that results from the above simulations using the different gs models. For each day of the simulation we calculate the percentage difference in water-use between the two simulations, we then calculate the mean and standard deviation over the year to give the annual mean leaf-level water-use."

Line 326: Please briefly state the results of the FLUXNET model evaluation in the main text. Also, the text in the supplemental says that there are large improvements in the seasonal cycle. Large seems like a stretch - instead I would quantify the changes in the RMSE.

We have moved the results of the Fluxnet model evaluation from the SI to the main text (results section 3.1). We quantify the changes in RMSE (Line 554 - 572).

"Site level evaluation of the seasonal cycles of latent and sensible heat with both JAC and MED models compared to FLUXNET observations showed in general, the MED model improved the seasonal cycle of both fluxes (lower RMSE), but the magnitude of this varied from site to site (Fig. S12). At the deciduous broadleaf site, US-UMB, MED resulted in improvements of the simulated seasonal cycle particularly in the summer months for both fluxes (RMSE decreased from 42.7/31.5 to 38.5/28.0 W/m^2 for latent/sensible heat respectively). At the second deciduous broadleaf site IT-CA1 however, there was almost no difference between the two g_s models. Both evergreen needle leaf forest sites (FI-Hyy and DE-Tha) saw improvements in the simulated seasonal cycles of latent and sensible heat with the MED model, primarily as a result of lower latent heat flux in the spring and summer months, and higher sensible heat flux over the same period. At FI-Hyy, RMSE decreased from 10.1/7.4 to 6.7/6.7 W/m^2 for latent/sensible heat respectively, and at DE-Tha, RMSE decreased from 16.0/11.9 to 10.5/10.6 W/m^2 for latent/sensible heat respectively. With the MED model the monthly mean latent heat flux was improved at the C_3 grass site (CH-Cha) as a result of increased flux in the summer months (RMSE decreased from 15.7 to 13.8 W/m^2), however there was no improvement in the sensible heat flux and RMSE with MED was increased (from 3.9 to 4.9 W/m^2). At the C_4 grass site (US-SRG), small improvements were made in the seasonal cycle of both latent and sensible heat with the MED model. At the deciduous savannah site (CG-Tch) which included a high proportion of shrub PFT in the land cover type used in the site simulation, large improvements in the seasonal cycle of both fluxes were simulated with the MED model, as a result of a decrease in the latent heat flux and an increase in the sensible heat flux (RMSE decreased from 39.5/31.6 to 30.4/24.4 W/m^2 for latent/sensible heat respectively)."

Line 335: I would urge the authors to stay away from suggesting that their analysis will allow a "full understanding"

This has been changed to "focus on" (Line 417).

Line 345: By "no form of land management", do the authors mean that there is no harvesting of crops or grazing of grasses? If so, how "big" do crops and grasses get, and what does this mean for their results?

We have modified this paragraph to clarify it (Line 424 - 433):

"JULES was run including dynamic vegetation with a land cover mask giving the fraction of agriculture in each 0.5^o x 0.5^o grid cell based on the Hurtt et al. (2011) land cover database for the year 2000. This means that whilst the model is allowed to evolve its own vegetation cover, within the agricultural mask only C_3/C_4 herbaceous PFTs are allowed to grow, with no competition from other PFTs. Therefore, through the simulation period, regions of agriculture are maintained as such and not out-competed by forests for example, allowing for a more accurate representation of the land cover of Europe in the model. No form of land management is simulated (i.e. no crop harvesting, ploughing, rotation or grazing), growth and leaf area index (LAI) are determined by resource availability and phenology. Outside of the agricultural mask, dynamic vegetation means that grid cell PFT coverage and LAI are the result of resource availability, phenology and simulated competition. Across the model domain, simulated mean annual LAI was dominantly within the range of 2 to 5 m^2/m^2 (Fig. S3 and S4)."

We mention the implications of no land management in the model in the discussion section 4.3 (Line 848 - 850):

"Additionally, this version of JULES does not have a crop module; it has no land management practices such as harvesting, ploughing or crop rotation – processes which may have counteracting effects on the land carbon sink."

Line 347: Does the change follow Hurtt et al. 2011? I would refrain from using the term "little" as this gives the reader little understanding of what is going on. Showing only 2050 is not too helpful.

We have modified this sentence to give more detail on the change in fractional land cover over the simulation period. This change does not follow Hurrtt et al., 2011 as we clearly state in the preceding sentences that the model is run with dynamic vegetation so is allowed to evolve its own vegetation cover, but that we apply an agricultural mask to maintain the extent of agricultural regions and these are based on the Hurrtt et al., 2011 data for the year 2000 (Line 433 - 437).

"Following a full spin-up period (to ensure equilibrium vegetation, carbon and water states), there was no significant change in the fractional cover of each PFT over the simulation period (1901 - 2050). By 2050, increases in boreal forest cover occurred, but this was less than 2% and limited to very small areas, given this small change we show just the land cover for 2050 in Fig. S1."

Lines 356-8: Whether emissions or meteorology matters more is going to depend on the emissions and climate variability. Langner et al. (2012b) only examine 1990 onwards and so I don't think the authors can use this work to comment on emissions vs. meteorology in 1900-1959.

We have amended this sentence as follows (Line 446 - 447):

"This procedure introduces some uncertainty of course, although Langner et al. (2012b) show that for the period 1990 to 2100 it is emissions change, rather than meteorological change, that drives modelled ozone concentrations."

The period 1990-2100 covers a period of 111 years, and a period in which climate-change is likely to show most effects, it therefore seems unlikely that things would be very different over the 60 year period of 1900-1959.

Lines 358-361: I don't really know what this means. The authors should show the trend in emissions of NOx, methane, and isoprene from 1900-2050 over Europe, which is standard practice in atmospheric chemistry papers, so that readers can fully understand the emission scenarios used, as this is central to the findings.

We didn't show such trends since there are many papers dealing with such emissions, and the focus of this paper is on the carbon sink and impacts at the land surface rather than on atmospheric chemistry. However, we have now added a new Figure, and text (Line 450 - 455):

"The trend in emissions of the major O_3 precursors NO_x, NMVOC and Isoprene are shown from 1900 to 2050 over Europe in Fig. S5. Isoprene emissions are not inputs to the EMEP model, but rather calculated at each time-step using temperature, radiation, and land-cover specific emission factors (Simpson et al., 2012). Changes in the assumed background concentration of CH_4 (from RCP6.0) (van Vuuren et al., 2011) are also shown in Fig. S5. Engardt et al. (2017) show the trend in emissions of SO_2 and NH_3 from 1900 to 2050 over Europe."

Line 362-3: This is confusing. I would cut this everything after "however"

This has been removed.

Line 367-369: Please clarify how the authors map the ozone concentrations from the land cover categories to the model. What do the differences in ozone concentrations over the different land cover types represent? Differences in dry deposition, BVOC emissions, or just turbulent mixing? Instead of saying "more accurate", the authors should just say something like ozone concentrations peak during the day so it's important to take the diurnal cycle into account.

This paragraph has been clarified (Line 458 - 469):

"O_3 concentrations from EMEP MSC-W were calculated at canopy height for two land-cover categories: forest and grassland (Fig. S6 and Fig. S7), which are taken as surrogates for high and low vegetation, respectively. These canopy-height specific concentrations allow for the large gradients 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 m in a forest canopy (Gerosa et al., 2017;Simpson et al., 2012;Tuovinen et al., 2009). These canopy level O_3 concentrations are used as input to JULES, using the EMEP O_3 concentrations for forest for the forest JULES PFTs (broadleaf/needle leaf tree and shrub), and the EMEP O_3 concentrations for grassland for the grass/herbaceous JULES PFTs (C_3 and C_4). This study used daily mean values of tropospheric O_3 concentration from EMEP disaggregated down to the hourly JULES model time-step. The daily mean O_3 forcing was disaggregated to follow a mean diurnal profile of O_3, this was generated from hourly O_3 output from EMEP MSC-W for the two land cover categories (forest and grassland as described above) across the same model domain. O_3 concentrations follow a diurnal cycle and peak during the day, therefore accounting for the diurnal variation in O_3 concentrations allows for a more realistic estimation of O_3 uptake."

Line 377: Typo

This has been amended.

Lines 381-394: Some of this is incorrect and the discussion is lengthy. I simply wanted the authors to note changes in the seasonal cycle of ozone depend strongly on anthropogenic Nox (not because the timing of emissions during the year, rather nonlinear ozone chemistry), the emissions scenario matters for the results regarding uptake of ozone to vegetation, which it seems like they are getting at eventually. I would cut most of this.

This paragraph has been modified as follows (Line 471 - 483):

"Figure 1 shows large increases in tropospheric O_3 from pre-industrial to present day (2001), this is in line with modelling studies (Young et al., 2013) and site observations (Derwent et al., 2008;Logan et al., 2012;Parrish et al., 2012), and is predominantly a result of increasing anthropogenic emissions (Young et al., 2013). Figures S6 and S7 show this large increase in ground-level O_3 concentrations from 1901 to 2001 occurs in all seasons. Present day O_3 concentration show a strong seasonal cycle, with a spring/summer peak in concentrations in the mid-latitudes of the Northern Hemisphere (Derwent et al., 2008;Parrish et al., 2012;Vingarzan, 2004). Seasonal cycles have been changing over the past decades however, attributed to changes in NO_x and other emissions, as well as changes in transport patterns (Parrish et al., 2013). These changes will likely continue in future as emissions and meteorological factors impact photo-chemical ozone production and transport patterns. Indeed, the O_3 concentrations used in the simulations in this study show increased O_3 levels in winter and in some regions in autumn and spring in 2050 compared to present day, this may be due to reduced titration of O_3 by NO as a result of reduced NO_X emissions in the future (Royal Society, 2008). Summer O_3 concentrations are lower in 2050 however, compared to 2001. "

Lines 394-397: Jumping from surface ozone seasonality to plant phenology seems erratic. I would suggest moving this discussion elsewhere.

This has been deleted and moved into the discussion section 4.3 (Line 855 - 858).

Line 420: I see that the authors examine ozone impacts on stomatal conductance, which could be referred to as "plant physiology", but it doesn't seem to me like GPP and C sink are "plant physiology" entities.

We have changed this to (Line 505 - 507):

"We use these simulations to investigate the direct effects of changing atmospheric CO_2 and O_3 concentrations, individually and combined, on plant water-use, GPP and the land C sink through the twentieth century and into the future, specifically over three time periods:…"

Line 421-423: The authors should tell the reader why we should go to the supplemental. "for calculation of the effects due to" is vague.

This has been changed to move information from the SI into the main text (Line 508 – 525):

"For each time period we calculate the difference between the decadal means calculated at the start and end of the analysis period for each variable of interest. Therefore our results report the change in GPP, for example, over the analysis period. For each variable analysed (GPP, NPP, vegetation carbon, soil carbon, total land carbon and *gs*), we use the mean over 10 years to represent each time period, e.g. the mean over 2040 to 2050 is what we call 2050, 1901 to 1910 is what we refer to as 1901. The difference between the simulations gives the effect of O_3 and CO_2 either separately or in combination over the different time periods. We look at the percentage change due to either O_3 at pre-industrial CO_2 concentration (i.e. without the additional effect of atmospheric CO_2 on stomatal behaviour - O3 simulation), CO_2 (at fixed pre-industrial O_3 concentration, CO2 simulation) or the combined effect of both gases (CO2+O3 simulation), which is calculated as:

$$100 * (var[y_1] – var[y_2]) / var[y_2] \qquad (8)$$

Where $var[y_x]$ represents the variable in time period y, e.g. $100 * (varO_3[2050] – varO_3[1901]) / varO_3[1901]$ gives the O_3 effect (at fixed CO_2) over the full experimental period. The meteorological forcing is prescribed in these simulations and is therefore the same between the model runs. Other climate factors, such as VPD, temperature and soil moisture availability are accounted for in our simulations, but our analysis isolates the effects of O_3, CO_2 and $O_3 + CO_2$. We also use paired t-test to determine statistically significant differences between the different (high and low) plant O_3 sensitivities."

Line 435: What is a wet site? Specify in the main text.

This is now specified in the main text (Line 537):

"The impact of g_s model on simulated g_s is shown for the site with low soil moisture stress (wet site, Fig. 2)."

Line 441: Same for dry site.

This has been changed (Line 544):

"This comparison was also done for a dry site (high soil moisture stress)…"

Line 442-5: Why should one wet and one dry site represent the entire domain?

This has been removed.

Line 445-447: It's not clear what the authors' point here is. Since the authors' simulations are uncoupled, it's an added uncertainty that changing stomatal conductance is going to impact energy partitioning and thus meteorology. Is that all they are trying to get at here?

We simply show here that changes in the stomatal conductance of the model alter the partitioning between the energy fluxes in these uncoupled simulations. We discuss later that potentially this could have impacts on meteorology, but that fully coupled simulations would be necessary to detect these effects (see discussion section 4.1). This is an interesting discussion point and area of future work worth noting.

Line 455-457: Why do the authors show the bottom row? Is it giving more information then the top row? I would understand if the stomatal uptake and ozone damage fed back onto the ozone concentrations the authors would need the bottom row. As this is not the case, this bottom row should be cut; but I agree that the authors should make this point in the text, which they do. Please clarify in the text what further details are in the supplemental.

The bottom row was simply to show that the different gs models simulate different rates of gs for each of the PFTs, and that consequently this affects the flux of O_3 into stomata. We have moved the bottom row of this figure in the SI (Fig. S11).

Line 473: What do the authors conclude about the comparison between the simulations with high and low ozone sensitivity vs the MTE-GPP product?

We have added this paragraph to the discussion section 4.1 (Line 780 - 794):

"We evaluated the JULES O_3 model by comparing modelled GPP against the Jung et al (2011) MTE product. Similar spatial patterns of GPP were simulated by JULES compared to MTE. Zonal means also showed similar patterns of GPP, although JULES under predicted GPP compared to MTE at latitudes >45ºN (temperate and boreal regions; all simulations) and over predicted GPP at latitudes <45ºN (Mediterranean region; all simulations). The simulations with transient O_3 (i.e. O_3 only and CO_2 + O_3) showed large differences in GPP between the high and low plant O_3 sensitivity simulations, this is to be expected given that the high plant O_3 sensitivity simulations were parameterised to be 'damaged' more by O_3, i.e. greater reduction of photosynthesis/g_s with O_3 exposure compared to the low plant O_3 sensitivity simulations. This difference was largest in the temperate zone, largely because of C_3 grass cover being the dominant land cover here and the difference in the sensitivity to O_3 between the high and low calibrations is significantly larger for C_3 grasses compared to the needle leaf trees that dominate in the boreal region. Additionally, a longer growing season in the temperate region may allow for greater uptake of O_3 into vegetation. C_3 grass is also the dominant land cover in the Mediterranean region with a different calibration used for Mediterranean grasses for the low plant O_3 sensitivity which is less sensitive to O_3 than the temperate C_3 grasses, but high soil moisture stress is common throughout the growing season in the Mediterranean limiting the uptake of O_3 through stomata, which likely diminishes the difference between the high and low calibrations."

Line 484-5: Please re-phrase so that it is clear that the GPP simulated by the low vs. high ozone sensitivity is significantly different

This has been changed to (Line 601 - 603):

"Over the historical period (1901-2001), O_3 reduced GPP under both the low and high plant O_3 sensitivity parameterizations by -3% to -9% respectively (Table 1), and this difference in simulated GPP was significant (t=102.2, $d.f.$=6270, p<2.2e^{-16})."

Lines 483-503: It's confusing in the text whether the authors are discussing changes in the trend from 1901-2001, or changes in the average, due to ozone. Please revise the text accordingly.

In response to a comment above, in section 2.4.2 we have clarified this, and we state what we report in the results, we move details from the SI to the main text of how we calculate this (Line 505 - 525):

"We use these simulations to investigate the direct effects of changing atmospheric CO_2 and O_3 concentrations, individually and combined, on plant water-use, GPP and the land C sink through the twentieth century and into the future, specifically over three time periods: historical (1901-2001), future (2001-2050) and over the full time series (1901-2050). For each time period we calculate the difference between the decadal means calculated at the start and end of the analysis period for each variable of interest. Therefore our results report the change in GPP, for example, over the analysis period. For each variable analysed (GPP, NPP, vegetation carbon, soil carbon, total land carbon and gs), we use the mean over 10 years to represent each time period, e.g. the mean over 2040 to 2050 is what we call 2050, 1901 to 1910 is what we refer to as 1901. The difference between the simulations gives the effect of O_3 and CO_2 either separately or in combination over the different time periods. We look at the percentage change due to either O_3 at pre-industrial CO_2 concentration (i.e. without the additional effect of atmospheric CO_2 on stomatal behaviour – O3 simulation), CO_2 (at fixed pre-industrial O_3 concentration, CO2 simulation) or the combined effect of both gases (CO2+O3 simulation), which is calculated as:

$$100 * (var[y_1] – var[y_2]) / var[y_2] \quad (8)$$

Where $var[y_x]$ represents the variable in time period y, e.g. $100 * (varO_3[2050] – varO_3[1901]) / varO_3[1901]$ gives the O_3 effect (at fixed CO_2) over the full experimental period. The meteorological forcing is prescribed in these simulations and is therefore the same between the model runs. Other climate factors, such as VPD, temperature and soil moisture availability are accounted for in our simulations, but our analysis isolates the effects of O_3, CO_2 and O_3 + CO_2. We also use paired t-test to determine statistically significant differences between the different (high and low) plant O_3 sensitivities."

Line 516-7: Again, suggesting that the O3 impact on the land carbon sink is a source of carbon is not really appropriate; re-phrasing would allow for the same take-away

This has been changed to (Line 636 – 638):

"By comparison with one of the largest anthropogenic emissions of carbon for Europe, we show here the effect of O_3 on reducing the size of the European land carbon sink is notable."

Line 523: Please quantify the "large" spatial variability
Line 527-529: With "therefore", are the authors suggesting that the decreases in GPP are from springtime increases in temperate/Mediterranean regions are because springtime ozone is increases? Please clarify in the text. What is going on in the boreal region?
Line 529: Ok, so the previous sentences are 13nalysing the simulations without CO2 fertilization? It would be best to make this clear before this point.

The changes is this paragraph address all three points above (Line 523 to 529).

We have added "as discussed below" in the first sentence because we go on to describe the large spatial variability in the next few sentences. We clarify that the results are referring to the simulations with O_3 only, and that the variability is due to the variability in the O_3 concentration:

"Over the 2001 to 2050 period, region-wide GPP with O3 only changing (O3 simulation) increased marginally (+0.1% to +0.2%, high and low plant O3 sensitivity, Table 1, with a significant difference between the two plant O3 sensitivities ($t=57$, d.f.=6270 $p<2.2e-16$)), although with large spatial variability as discussed below (Fig. 4g & h). Figures S6 and S7 show that despite decreased tropospheric O3 concentrations by 2050 in summer compared to 2001 levels, all regions are exposed to an increase in O3 over the wintertime, and some regions of Europe, particularly temperate/Mediterranean experience increases in O3 concentration in spring and autumn. Therefore, although in the O3 simulation, overall simulated GPP for Europe shows a small increase, large spatial variability is shown in Fig's 4g &h because of the variability in O3 concentration with region and season. Increased GPP (dominantly 10%, but up to 20% in some areas) on 2001 levels is simulated across areas of Europe, however, decreases of up to 21% are simulated in some areas of the Mediterranean, up to 15% in some areas of the boreal region and up to 27% in the temperate zone (Fig. 4g & h). "

Line 533-534: What are the implications of this?

We have changed this sentence as below (Line 658 - 660):

"Nevertheless, although the percentage gain is larger, the absolute value of GPP by 2050 remains lower compared to GPP with CO_2 only changing, highlighting the negative impact of O_3 at the land surface (Table S4)."

Line 567: "Over the Anthropocene" is ambiguous

We have removed the use of the term Anthropocene and refer to it as the full experimental period or give the years 1901 to 2050.

Line 634: The authors' use of "leaf-level" stomatal conductance in this paragraph is confusing; earlier they define leaf-level stomatal conductance as non-canopy integrated stomatal conductance; is this what they are examining here?

Apologies, we have removed use of the term leaf-level to stop confusion.

Lines 633-648: I would like to see some discussion of the model evaluation of the stomatal conductance models (e.g., FLUXNET). Regarding the last sentence of this paragraph, I would make this statement specific to the uncoupled approach. Higher deposition would reduce ozone concentrations in a coupled chemistry-land study.

We add discussion of the sites-level evaluation of the gs models here (Line 766 - 769):

"Site-level evaluation of the models against Fluxnet observations showed that in general the MED model improved simulated seasonal cycles of latent and sensible heat. The magnitude of the improvement varied with site, large improvements were seen at the deciduous savanna site, and at the NT sites and BT site (US_UMB) in the spring and summer. However, much smaller improvements were seen at the grass sites."

We changes the last sentence accordingly (Line 775 - 778):

"Therefore, given that C_3 herbaceous vegetation is the dominant land cover class across the European domain used in this study, this suggests a greater O_3 impact for Europe would be simulated with MED model compared to JAC in our simulations where chemistry is uncoupled from the land surface."

Lines 649-661: Do the authors have any hypotheses for why their study shows lower impact on GPP, or do the authors think their results are reasonable in comparison to the other work? On that note, I do not see any support for the last sentence of the paragraph. I would encourage the authors to change the phrasing to be more speculative (instead of saying that this is "likely" the result of).

We have amended this paragraph. It is difficult to hypothesis as to why estimates differ between the models and as such we have removed the last sentence (Line 806 - 812):

"Our estimates of changes in current day GPP and NPP are at the lower end of previously modelled estimates. Simulated O_3 impacts will depend in a large part on the scenario of O_3 concentrations used as forcing, meteorological forcing and how sensitive vegetation is parameterised to be to O_3 damage, in addition to the different process representation of O_3 damage in each model. It is therefore difficult to hypothesise as to exactly why modelled estimates differ, but suggests that an ensemble approach to modelling O_3 impacts on the terrestrial biosphere would be beneficial to understand some of these differences and provide estimates of O_3 damage with uncertainties."

Line 687-691: Using a stomatal conductance parameterization that simulates higher gs will certainly lead to higher uptake. The higher uptake may decrease ozone concentrations, but the stronger ozone damage may increase ozone concentrations. It's hard to say which will dominate in the authors' uncoupled simulations, especially because ozone is fairly well-buffered in models (one sink reduces, another sink kicks in), how the high vs. low ozone sensitivity simulations will be different, and if this high sensitivity study is indeed an "upper bound".

We have modified this paragraph accordingly and remove the sentences referring to this study as an upper bound (Line 836 - 848):

"We include a representation of agricultural regions through the model calibration against the wheat O_3 sensitivity function (CLRTAP, 2017), and in our simulations the high plant O_3 sensitivity scenario uses this calibration against wheat for all C_3/C_4 land cover which dominates our model domain. Wheat is known to be one of the most O_3 sensitive crop species however, so it is possible that our simulations over-estimate the O_3 impact at the land surface. However, the low plant O_3 sensitivity calibration against natural grasslands provides a counter estimate of the impact of O_3 at the land surface, therefore it is important to consider the range our results provide (i.e. both the high and low plant O_3 sensitivity) as an indicator of the impact of O_3 on the land surface. As with all uncoupled modelling studies, a change in g_s and flux will impact the O_3 concentration itself. Therefore adopting the Medlyn formulation with a higher g_s and subsequently higher O_3 flux for broadleaf and C_3 PFTs (Fig 2) would lead to reduced O_3 concentration, which in turn would act to dampen the effect of higher g_s on O_3 flux, although the higher uptake of O_3 by vegetation may lead to more damage and increase O_3 concentrations, in an uncoupled chemistry-land modelling system such as this it is not possible to predict which process would dominate."

Line 711: Typo

This has been changed.

Line 718: Here it is relevant to discuss the findings of Lombardozzi that there are separate impacts of ozone on photosynthesis and stomatal conductance

We discuss the modelling approach of Lombardozzi and the results at other points in the manuscript, so we prefer not to discuss again here. This paragraph is also discussing O_3 induced sluggish stomatal behaviour observed in plants, whilst Lombardozzi et al separate the impacts of O_3 on photosynthesis and stomatal conductance, it is not a representation of sluggish stomatal control.

Line 724: By gmax are the authors referring to input, or output of the model? If input, I don't think the authors' reasoning makes sense. The parameterizations are different and act to scale stomatal conductance by very different entities.

We have clarified this point below (Line 892 - 896):

"We acknowledge this inconsistency as a caveat of our study, however comparison of *gmax* (maximum g_s) values from both models (EMEP (*gmax* is an input parameter determining the maximum g_s) and JULES (*gmax* is not used as an input parameter in JULES, instead we calculated *gmax* for each PFT taking the mean across the model domain for the year 2001) suggests the differences are small for deciduous forest……".

Line 728-730: Ozone deposition can have a substantial impact on surface ozone concentrations (Val Martin et al., 2014). I would not argue this.

Line 730-732: Where is there evidence that stomatal conductance does not influence ozone concentrations above the canopy? I would not argue this. Further, strong vertical mixing above trees means that this is not the limiting factor for deposition – rather stomatal deposition and nonstomatal deposition are.

Line 732-733: Again, where in the literature is there evidence of this?

Line 733-736: I'm not sure what the authors are getting at here.

The points raised concerning lines 728-736 are related, and so we address them as one here. We have re-phrased the text of lines 728-736 to make a clearer distinction between the role of deposition in regional and above-canopy O_3 (Line 898 - 909):

"It should be noted that the role of EMEP in this study is not to provide g_s, but to provide O_3 at the top of the vegetation canopy. This firstly entails a calculation of the large-scale ozone concentrations for Europe, which are represented by the gridded values of grid-cell average concentration, and secondly to calculate the vertical gradients between these grid-cell centres (at ca. 45m) and the top of the vegetation canopy. O_3 deposition is important for both steps; it is known to have a substantial impact on the lifetime and concentrations of O_3 in the planetary boundary layer (Garland and Derwent, 1979;Val Martin et al., 2014), and also in determining the local vertical gradients above different land-covers (CLRTAP, 2017;Gerosa et al., 2017;Tuovinen et al., 2009). Vertical gradients between the 45m level and the top of forest canopies tend to be limited (Fuentes et al., 2007;Karlsson et al., 2006) due to the good mixing normally induced by forest roughness. Vertical gradients between 45m and the top of shorter vegetation such as grasslands or crops can be larger however (CLRTAP, 2017;Gerosa et al., 2017). Accounting for such land-cover specific gradient effects has been shown to have large impacts on estimates of O_3 metrics (Simpson et al., 2007)."

Large but decreasing effect of ozone on the European carbon sink

Rebecca J Oliver[1], Lina M Mercado[1,2], Stephen Sitch[2], David Simpson[3,4], Belinda E Medlyn[5], Yan-Shih Lin[5], Gerd A Folberth[6]

[1] Centre for Ecology and Hydrology, Benson Lane, Wallingford, OX10 8BB, UK

[2] College of Life and Environmental Sciences, University of Exeter, EX4 4RJ, Exeter, UK

[3] EMEP MSC-W Norwegian Meteorological Institute, PB 43, NO-0313, Oslo, Norway

[4] Dept. Space, Earth & Environment, Chalmers University of Technology, Gothenburg, SE-41296 Sweden

[5] Hawkesbury Institute for the Environment, Western Sydney University, Locked Bag 1797, Penrith NSW 2751 Australia

[6] Met Office Hadley Centre, Exeter, UK.

Correspondence to: Rebecca Oliver (rfu@ceh.ac.uk)

Abstract

The capacity of the terrestrial biosphere to sequester carbon and mitigate climate change is governed by the ability of vegetation to remove emissions of CO_2 through photosynthesis. Tropospheric O_3, a globally abundant and potent greenhouse gas, is, however, known to damage plants, causing reductions in primary productivity~~, yet the impact of this gas on European vegetation and the land carbon sink is largely unknown~~. Despite emission control policies across Europe, background concentrations of tropospheric O_3 have risen significantly over the last decades due to hemispheric-scale increases in O_3 and its precursors. Therefore, plants are exposed to increasing background concentrations, at levels currently causing chronic damage. Studying the impact of O_3 on European vegetation at the regional scale is important for gaining greater understanding of the impact of O_3 on the land carbon sink at large spatial scales. In this work we take a regional approach and update the JULES land-surface model using new measurements specifically for European vegetation. Given the importance of stomatal conductance in determining the flux of O_3 into plants, we implement an alternative stomatal closure parameterization and account for diurnal variations in O_3 concentration in our simulations. We conduct our analysis specifically for the European region to quantify the impact of tropospheric O_3, and its interaction with CO_2, on gross primary productivity (GPP) and land carbon storage across Europe. ~~We use the JULES land-surface model recalibrated for O_3 impacts on European vegetation, with an improved stomatal conductance parameterization, to quantify the impact of tropospheric O_3, and its interaction with CO_2, on gross primary productivity (GPP) and land carbon storage across Europe.~~ A factorial set of model experiments showed that tropospheric O_3 can suppress terrestrial carbon uptake across Europe over the period 1901 to 2050. By 2050, simulated GPP was reduced by 4 to 9% due to plant O_3~~ozone~~ damage and land carbon storage by 3 to 7%. The combined physiological effects of elevated future CO_2 (acting to reduce stomatal opening) and reductions in O_3 concentrations resulted in reduced O_3 damage in the future~~, contrary to predictions from earlier studies~~. This alleviation of O_3 damage by CO_2 induced stomatal closure was around 1 to 2% for low and high sensitivity respectively (on both land carbon and GPP). Reduced land carbon storage resulted from diminished soil carbon stocks consistent with the reduction in GPP. Regional variations are identified with larger impacts shown for temperate Europe (GPP reduced by 10 to 20%) compared to boreal regions (GPP reduced by 2 to 8%). These results highlight that O_3 damage needs to be considered when predicting GPP and land carbon, and that the effects of O_3 on plant physiology need to be considered in regional land carbon cycle assessments.

Commented [OT1]: Line 35

Commented [OT2]: Line 31

Formatted: Subscript

Commented [ORJ3]: Line 41

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 ± 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). Little attention, however, has been given to tropospheric ozone (O_3), a globally abundant 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 (CO), volatile organic compounds (VOCs), methane (CH_4) and nitrogen oxides (NO_x) from both man-made and natural sources, as well as downward transport from the stratosphere and lightning which is a source of NO_x. 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 important~~key~~ to understanding the carbon sequestration potential of the land-surface, and future carbon dynamics regionally and globally (Le Quéré et al., 2016;Sitch et al., 2015).

Commented [ORJ4]: Line 82

Due to increased anthropogenic precursor emissions over the industrial period, background concentrations of ground-level O_3 have risen (Vingarzan, 2004). O_3 levels at the start of the 20^{th} century are estimated to be around 10 ppb for the site Montsouris Observatory near Paris, data for Arkona on the Baltic coast increased from ca. 15 ppb in the 1950s to 20-27 ppb by the early 1980s, and the Irish coast site Mace Head shows around 40 ppb by the year 2000 (Logan et al., 2012;Parrish et al., 2012). Present day annual average background O_3 concentrations reported in the review of Vingarzan (2004) show O_3 concentrations range between approximately 20 and 45 ppb, with the greatest increase occurring since the 1950s. Trends vary from site to site though, even on a decadal basis (Logan et al., 2012;Simpson et al., 2014b), depending, for example, on local/regional trends in precursor (especially NOx) emissions, elevation, and exposure to long-range transport. Nevertheless, there is some indication that background O_3 levels over the mid-latitudes of the Northern Hemisphere have continued to rise at a rate of approximately 0.5–2% per year, although not uniform (Vingarzan, 2004). 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). Intercontinental transport of air pollution from regions such as Asia that currently have poor emission controls are thought to contribute substantially to rising background O_3 concentrations over the last decades (Cooper et al., 2010;Verstraeten et al., 2015). 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 concentrations in Europe will be partly~~are~~ dependent on how O_3 precursor emissions evolve globally.

Elevated O_3 concentrations impact agricultural yields and nutritional quality of major crops (Ainsworth et al., 2012;Avnery et al., 2011), with consequences for global food security (Tai et al., 2014). As well as being a significant air pollutant, O_3 is a potent greenhouse gas (Royal-Society, 2008). High levels of O_3 are damaging to ecosystem health and reduce the global land carbon sink (Arneth et al., 2010;Sitch et al., 2007). Reduced uptake of carbon by plant photosynthesis due to O_3 damage allows more CO_2 to remain in the atmosphere. This effect of O_3 on plant physiology represents an additional climate warming to the direct radiative forcing of O_3 (Collins et al., 2010;Sitch et al., 2007), the magnitude of which, however, remains highly uncertain (IPCC, 2013).

Dry deposition of O_3 to terrestrial surfaces, primarily uptake by stomata on plant foliage and deposition on external surfaces of vegetation (Fowler et al., 2001;Fowler et al., 2009), is a large sink for ground level O_3 (Wild, 2007;Young et al., 2013)~~(Wild, 2007) (Fowler et al., 2009;Fowler et al., 2001;Wild, 2007)~~. On entry to sub-stomatal spaces, O_3 reacts with other molecules to form reactive oxygen species (ROS). Plants can tolerate a certain level of O_3 depending on their capacity to scavenge and detoxify the ROS (Ainsworth et al., 2012). Above this critical level, long-term chronic O_3 exposure reduces plant photosynthesis and biomass accumulation (Ainsworth, 2008;Ainsworth et al., 2012;Matyssek et al., 2010a;Wittig et al., 2007;Wittig et al., 2009), either directly through effects on photosynthetic machinery such as reduced Rubisco content (Ainsworth et al., 2012;Wittig et al., 2009) and/or indirectly by reduced stomatal conductance (g_s) (Kitao et al., 2009;Wittig et al., 2007), alters carbon allocation to different pools (Grantz et al., 2006;Wittig et al., 2009), accelerates leaf senescence (Ainsworth, 2008;Nunn et al., 2005;Wittig et al., 2009) and changes plant susceptibility to biotic stress factors (Karnosky et al., 2002;Percy et al., 2002).

The response of plants to O_3 is very wide ranging as reported in the literature from different field studies. The Wittig et al. (2007) meta-analysis of temperate and boreal tree species showed future concentrations of O_3 predicted for 2050 significantly reduced leaf level light saturated net photosynthetic uptake (-19%, range: -3% to

Commented [OT5]: Line 107

-28% at a mean O_3 concentration of 85 ppb) and g_s (-10%, range: +5% to -23% at a mean O_3 concentration of 91 ppb) in both broadleaf and needle leaf tree species. In the Feng et al. (2008) meta-analysis of wheat, projected O_3 concentrations for the future reduced aboveground biomass (-18% at a mean O_3 concentration of 70 ppb) photosynthetic rate (-20% at a mean O_3 concentration of 73 ppb) and g_s (-22% at a mean O_3 concentration of 79 ppb). One of few long-term field based O_3 exposure studies (AspenFACE) showed that after 11 years of exposing mature trees to elevated O_3 concentrations (mean O_3 concentration of 46 ppb), O_3 decreased ecosystem carbon content (-9%), and decreased NPP (-10%), although the O_3 effect decreased through time (Talhelm et al., 2014). Zak et al. (2011) showed this was partly due to a shift in community structure as O_3-tolerant species, competitively inferior in low O_3 environments, out competed O_3-sensitivie species. GPP was reduced (-12% to -19%) at two Mediterranean ecosystems exposed to high~~elevated~~ ambient O_3 concentrations (ranging between 20 to 72 ppb across sites and through the year) studied by Fares et al. (2013). Biomass of mature beech trees was reduced (-44%) after 8 years of exposure to elevated O_3 (~150 ppb) (Matyssek et al., 2010a). After 5 years of O_3 exposure (ambient +20 to +40 ppb) in a semi-natural grassland, annual biomass production was reduced (-23%), and in a Mediterranean annual pasture O_3 exposure significantly reduced total aboveground biomass (up to -25%) (Calvete-Sogo et al., 2014). However, these were empirical studies at individual sites, and these focus on O_3 effects on plant physiology and productivity, but do not quantify the impact on the land carbon sink. Modelling studies are needed to scale site observations to the regional and global scales. Models generally suggest that plant productivity and carbon sequestration will decrease with O_3 pollution, though the magnitudes vary. For example, based on a limited dataset to parameterise plant O_3 damage for a global set of plant functional types, Sitch et al. (2007) predicted a decline in global GPP of 14 to 23% by 2100. A second study by Lombardozzi et al. (2015) similarly predicted a 10.8% decrease of global GPP. Here we take a regional approach and take advantage of the latest~~new~~ measurements showing changes in plant productivity with accumulated exposure to O_3 specifically for a range of European vegetation from different regions (CLRTAP 2017) with which to calibrate the JULES model for plant sensitivity to O_3, and conduct our~~a dedicated~~ analysis specifically for the European region.

Understanding the response of plants to elevated tropospheric O_3 is challenged by the large variation in O_3 sensitivity both within and between species (Karnosky et al., 2007;Kubiske et al., 2007;Wittig et al., 2009). Additionally, other environmental stresses that affect stomatal behaviour will affect the rate of O_3 uptake and therefore the response to O_3 exposure, such as high temperature, drought and changing concentrations of atmospheric CO_2 (Mills et al., 2016;Fagnano et al., 2009;Kitao et al., 2009;Löw et al., 2006)~~such that the response of vegetation to O_3 is a balance between opposing drivers of stomatal behaviour~~. Increasing concentrations of atmospheric CO_2, for example, are suggested to provide some protection against O_3 damage by causing stomata to close (Harmens et al., 2007;Wittig et al., 2007), however the long-term effects of CO_2 fertilisation on plant growth and carbon storage remain uncertain (Baig et al., 2015;Ciais et al., 2013). Further, in some studies, stomata have been shown to respond sluggishly, losing their responsiveness to environmental stimuli with exposure to O_3 which can lead to higher O_3 uptake, increased water-loss and therefore greater vulnerability to environmental stresses such as drought (Mills et al., 2016;Mills et al., 2009;Paoletti and Grulke, 2010;Wilkinson and Davies, 2009).

Commented [ORJ6]: Line 131-132

Commented [OT7]: Line 150

Commented [OT8]: Line 151

Commented [OT9]: Line 157-158

Given the critical role g_s plays in the uptake of both CO_2 and O_3, we use an alternative representation and parameterisation of g_s in JULES by implementing the Medlyn *et al*. (2011) g_s formulation. This model is based on the optimal theory of stomatal behaviour~~, it does not currently include a representation of sluggish stomatal control, but it~~ and has ~~the following~~ advantages over the current JULES g_s formulation of Jacobs (1994)~~:~~ including i) a single parameter (g_1) ~~which represents the marginal cost of water-use,~~ compared to two parameters in Jacobs (1994) ~~representing the the critical humidity deficit at the leaf surface (*dqcrit*) and the *ci*/*ca* ratio at the leaf critical humidity deficit (*f0*) (Clark et al., 2011);~~, ii) the g_1 parameter is related to the water-use strategy of vegetation and is easier to parameterise with commonly measured leaf or canopy level observations of photosynthesis, g_s and humidity, ~~– all variables that are commonly measured,~~ and (iii) values of g_1 are available for many different plant functional types (PFTs) derived from a global data set of leaf-level measurements (Lin et al., 2015).

The main objective of this work is to assess the impact of historical and projected (1901 to 2050) changes in tropospheric O_3 and atmospheric CO_2 concentration on predicted GPP and the land-carbon sink for Europe. These are the two greenhouse gases that directly affect plant photosynthesis and g_s. We use a factorial suite of model experiments, using the Joint UK land environment simulator (JULES) (Best et al., 2011;Clark et al., 2011), the land-surface model of the UK Earth System Model (UKESM) (Collins et al., 2011) to simulate plant O_3 uptake and damage, and to investigate ~~look at~~ the impact of both ~~interaction between~~ O_3 and CO_2 on plant water-use and carbon uptake. In this work, ~~plant O_3 damage in JULES is developed further by introducing a term for dry deposition of O_3 to external plant surfaces, an important sink for tropospheric O_3 that was previously absent from the model. Further,~~ the JULES model is re-calibrated using the latest observations of vegetation sensitivity to O_3, with the addition of a separate parameterisation for temperate/boreal regions versus the Mediterranean. The ~~plant~~ O_3 sensitivity of each PFT in JULES was re-calibrated for both a high and low ~~plant O_3~~ sensitivity to account for uncertainty in the O_3 response, in part due to the ~~the large~~ observed variation in O_3 sensitivity ~~within and~~ between species. This includes O_3 ~~separate~~ sensitivities for ~~Mediterranean regions, and for~~ agricultural crops (wheat – high sensitivity) versus natural grassland (low sensitivity), with separate sensitivities for Mediterranean grasslands. For forests JULES is parameterised with O_3 sensitivities for broadleaf and needle leaf trees (with a high and low O_3 sensitivity for both), with separate sensitivities (high and low) for Mediterranean broadleaf species. We make a separate distinction for the Mediterranean region where possible because the work of Büker et al. (2015) showed that different O_3 dose-response relationships are needed to describe the O_3 sensitivity of dominant Mediterranean trees. In addition, we introduce an alternative g_s scheme into JULES as described above. JULES is forced with spatially varying daily~~hourly~~ O_3 concentrations from a high resolution atmospheric chemistry model for Europe that are disaggregated to hourly concentrations, therefore our simulations account for diurnal variations in O_3 concentration and O_3 responses allowing for improved ~~more accurate~~ estimates~~ions~~ of O_3 uptake by vegetation. We do not attempt to make a full assessment of the carbon cycle of Europe, instead we target O_3 damage, ~~and its interaction with CO_2,~~ which is currently a missing component in earlier carbon cycle assessments (Le Quéré et al., 2017;Sitch et al., 2015). To this end, we prescribe changing O_3 and CO_2 concentrations from 1901 to 2050, but use a fixed pre-industrial climate. We acknowledge the use of a 'fixed' pre-industrial climate omits the additional uncertainty of the interaction between climate change and g_s which will affect the rate of O_3 uptake and therefore O_3 concentrations. In addition, using uncoupled chemistry and climate is a further source of uncertainty. To understand the impact of

Commented [OT10]: Line 169-170

Commented [ORJ11]: Line 167-176

Commented [ORJ12]: Line 183

Commented [ORJ13]: Line 183 to 185

Commented [ORJ14]: Line 189

Commented [ORJ15]: Line 193-196

Commented [ORJ16]: Line 196-197

Commented [ORJ17]: Line 199 to 201

these complex feedback mechanisms is an important area for future work, but in the current study our aim is to isolate the physiological response of plants to both O_3 and CO_2, and determine the sensitivity of predicted GPP and the land carbon sink to this process, as the impact of O_3 on ~~European vegetation and~~ the land carbon sink currently remains largely unknown at large spatial scales for Europe.

2 Methods

2.1 Representation of O_3 effects in JULES

JULES calculates the land-atmosphere exchanges of heat, energy, mass, momentum and carbon on a sub-daily time step, and includes a dynamic vegetation model (Best et al., 2011;Clark et al., 2011;Cox, 2001). This work uses JULES version 3.3 (http://www.jchmr.org) at 0.5° x 0.5° spatial resolution and hourly model time step, the spatial domain is shown in Fig. S5. JULES has a multi-layer canopy radiation interception and photosynthesis scheme (10 layers in this instance) that accounts for direct and diffuse radiation, sun fleck penetration through the canopy, inhibition of leaf respiration in the light and change in photosynthetic capacity with depth into the canopy (Clark et al., 2011;Mercado et al., 2009). Soil water content also affects the rate of photosynthesis and g_s. It is modelled using a dimensionless soil water stress factor, β, which is related to the mean soil water concentration in the root zone, and the soil water contents at the critical and wilting point (Best *et al*., 2011).

To simulate the effects of O_3 deposition on vegetation productivity and water use, JULES uses the flux-gradient approach of Sitch *et al*., (2007), modified to include non-stomatal deposition following Tuovinen et al. (2009). A similar approach is taken by Franz et al. (2017) in the OCN model, however plant O_3 damage is a function of accumulated O_3 exposure over time. In JULES, plant O_3 damage is instantaneous, the degree to which photosynthesis and g_s are modified at each time step with O_3 exposure having already been calibrated against observations of the change in plant productivity with cumulative O_3 exposure for each PFT (i.e. O_3 dose-response functions described later). JULES uses a coupled model of g_s and photosynthesis, the potential net photosynthetic rate (A_p, mol CO_2 m^{-2} s^{-1}) is modified by an 'O_3 uptake' factor (*F,* the fractional reduction in photosynthesis), so that the actual net photosynthesis (A_{net}, mol CO_2 m^{-2} s^{-1}) is given by equation 1 (Clark *et al.,* 2011, Sitch *et al.,* 2007). Because of the relationship between these two fluxes, the direct effect of O_3 damage on photosynthetic rate also leads to a reduction in g_s. An alternative approach was taken by Lombardozzi et al. (2012) in the CLM model where photosynthesis and g_s are decoupled, so that O_3 exposure affects carbon assimilation and transpiration independently. In JULES, changes in atmospheric CO_2 concentration also affect photosynthetic rate and g_s, consequently the interaction between changing concentrations of both CO_2 and O_3 is allowed for.

Commented [ORJ18]: Line 226

$$A_{net} = A_P F \quad (1)$$

The O_3 uptake factor (F) is defined as:

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

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 m^{-2} PLA s^{-1}, projected leaf area), and '*a*' is a PFT-specific parameter representing the fractional reduction of photosynthesis with O_3 uptake by leaves. Following Tuovinen et al. (2009), the flux of O_3 through stomata, F_{O3}, is represented as follows:

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

O_3 is the molar concentration of O_3 at reference (canopy) level (nmol m^{-3}), g_b is the leaf-scale boundary layer conductance (m s^{-1}, eq 3b), g_l is the leaf conductance for water (m s^{-1}), K_{o3} accounts for the different diffusivity of ozone to water vapour and takes a value of 1.51 after Massman (1998), and g_{ext} is the leaf-scale non-stomatal deposition to external plant surfaces (m s^{-1}) which takes a constant value of 0.0004 m s^{-1} after Tuovinen et al. (2009). The leaf-level boundary layer conductance (g_b) is calculated as in Tuovinen *et al*. (2009)

$$g_b = \alpha Ld^{-1/2} U^{-1/2} \quad (3b)$$

α is a constant (0.0051 m s$^{-1/2}$), *Ld* is the cross-wind leaf dimension (m) defined per PFT as 0.05 for trees, 0.02 for grasses (C_3 and C_4) and 0.04 for shrubs, ~~and~~ *U* is wind speed at canopy height (m 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 function of photosynthetic rate in JULES (Clark et al., 2011), from eq 1 it follows that:

Commented [ORJ19]: Line 253

$$g_s = g_l F \quad (4)$$

The O_3 flux to stomata, F_{O3}, is calculated at leaf level and then scaled to each canopy layer differentiating sunlit and shaded leaf photosynthesis, and finally summed up to the canopy level. Because the photosynthetic capacity, photosynthesis and therefore g_s decline with depth into the canopy, this in turn affects O_3 uptake, with the top leaf level contributing most to the total O_3 flux and the lowest level contributing least.

2.2 Calibration of O_3 uptake model

Here we use the latest literature on flux based O_3 dose-response relationships derived from observed field data across Europe (CLRTAP, 2017) to determine the key PFT-specific O_3 sensitivity parameters in JULES (*a* and F_{O3crit}). Synthesis of information expressed as O_3 flux based dose-response relationships derived from field experiments is carried out by The United Nations Convention on Long-Range Transboundary Air Pollution (CLRTAP Convention), this information is then used as a policy tool to inform emission reduction strategies in Europe to improve air quality (CLRTAP, 2017;Mills et al., 2011a). Derivation of O_3 flux based dose-response relationships for different vegetation types uses the accumulated stomatal O_3 flux above a threshold (often referred to as the phytotoxic O_3 dose above a threshold of 'y' i.e. POD_y) as the dose metric, and the percentage change in biomass as the response metric (Emberson et al., 2007;Karlsson et al., 2007). We use these observation based O_3 dose-response relationships to calibrate each JULES PFT for sensitivity to O_3 using available relationships for the closest matching vegetation type. For JULES, Fo_{3crit} is the threshold for O_3 damage, and values for this parameter are taken from the O_3 dose-response relationships as the POD_y value. The actual sensitivity to O_3 is determined by the slope of the O_3 dose-response relationship, i.e. how much biomass changes with accumulated stomatal uptake of O_3 above the damage threshold, this relates to the parameter *a* in JULES. The parameter '*a*' is a PFT-specific parameter representing the fractional reduction of photosynthesis with O_3 uptake by leaves. Values for this parameter are found for each PFT by running JULES with different values of '*a*', which alter the instantaneous photosynthetic rate, but then calculating the accumulated stomatal flux of O_3 and the change in productivity, until the slope of this relationship produced by the JULES simulations matches that of the O_3 dose-response relationships derived from observations. Essentially we calibrate each JULES PFT for sensitivity to O_3 by reproducing the observation-based O_3 dose-response relationships.

Each PFT was calibrated for a high and low plant O_3 sensitivity to account for uncertainty in the sensitivity of different plant species to O_3, using the approach of Sitch *et al.*, (2007). Therefore, when using our results to assess the impact of O_3 at the land surface, we are able to provide a range in our estimates to help address some of the uncertainty in the O_3 response of different vegetation types. In addition, where possible owing to available data, a distinction was made for Mediterranean regions. This was because the work of Büker et al. (2015) showed that different O_3 dose-response relationships are needed to describe the O_3 sensitivity of dominant Mediterranean trees. For the C_3 herbaceous PFT, the dominant land cover type across the European domain in this study (Fig. S1), the high plant O_3 sensitivity was calibrated against observations for wheat to give a representation of agricultural regions and wheat is one of the most sensitive grasses to O_3 (Fig. S2, Table S1). For the low plant O_3 sensitivity JULES was calibrated against the dose-response function for natural grassland to give a representation of natural grassland and this vegetation has a much lower sensitivity to O_3 damage, for the Mediterranean region we used a function for Mediterranean natural grasslands, all taken from CLRTAP (2017) (Fig. S2, Table S1). Tree/shrub PFTs were calibrated against observed O_3 dose-response functions for the high plant O_3 sensitivity: broadleaf trees (temperate/boreal) = Birch/Beech dose-response relationship, broadleaf trees (Mediterranean) = deciduous oaks dose-response relationship, needle leaf trees = Norway spruce dose-response relationship, shrubs = Birch/Beech dose-response relationship, all from CLRTAP (2017) (Fig. S2, Table S1). Data on O_3 dose-response relationships for different vegetation types is very limited, therefore for the low plant O_3 sensitivity calibration for trees/shrubs we assumed a 20% decrease in sensitivity to O_3 based on the difference in sensitivity between high and low sensitive tree species in the Karlsson et al. (2007) study. Due to limitations in data availability, the shrub parameterisation uses the observed dose-response functions for broadleaf trees. Similarly, the parameterisation for C_4 herbaceous uses the observed dose-responses for C_3 herbaceous, however the fractional cover of C_4 herbs across Europe is low (Fig. S1), so this assumption affects a very small percentage of land cover.

To calibrate the JULES O_3 uptake model, JULES was run across Europe forced using the WFDEI observational climate dataset (Weedon, 2013) at 0.5° X 0.5° spatial and three hour temporal resolution. JULES uses interpolation to disaggregate the forcing data down from 3 hours to an hourly model time step. The model was spun-up over the period 1979 to 1999 with a fixed atmospheric CO_2 concentration of 368.33 ppm (1999 value from Mauna Loa observations, (Tans and Keeling)). Zero tropospheric ozone concentration was assumed for the control simulation, for the simulations with O_3, spin-up used spatially explicit fields of present day O_3 concentration produced using the UK Chemistry and Aerosol (UKCA) model with standard chemistry from the run evaluated by O'Connor et al. (2014). A fixed land cover map was used based on IGBP (International Geosphere-Biosphere Programme) land cover classes (IGBP-DIS), therefore as the vegetation distribution was fixed and the calibration was not looking at carbon stores, a short spin-up was adequate to equilibrate soil temperature and soil moisture. JULES was then run for the year 2000 with a corresponding CO_2 concentration of 369.52 ppm (from Mauna Loa observations, (Tans and Keeling)) and monthly fields of spatially explicit tropospheric O_3 (O'Connor et al., 2014) as necessary.

Calibration was performed using four simulations: with i) zero tropospheric O_3 concentration, this was the control simulation (control), ii) tropospheric O_3 at current ambient concentration (O3), iii) ambient +20 ppb (O3+20) and iv) ambient +40 ppb (O3+40). The different O_3 simulations (i.e. O3, O3+20 and O3+40) were used to capture the range of O_3 conditions in the data used in the observation-based O_3 dose-response relationships used in this study for calibration, often data were from experiments using artificially manipulated conditions of ambient + 40 ppb O_3 for example. For each JULES O_3 simulation, the value of F_{O3crit} was taken from the vegetation specific O_3 dose-response relationship as the threshold O_3 concentration above which damage to vegetation occurs. An initial estimate of the parameter '*a*' was used, then for each PFT and each simulation, hourly estimates of NPP (our proxy for biomass – although not identical they are related) and O_3 uptake in excess of F_{O3crit} were accumulated over a PFT dependent accumulation period. The accumulation periods were ~6 months for broadleaf trees and shrubs, all year for needle leaf trees, and ~3 months for herbaceous species, through the growing season, following guidelines in CLRTAP (2017). Additionally, in accordance with the methods used in the CLRTAP (2017) that describe how the O_3 dose-response relationships are derived from observations, we use the stomatal O_3 flux per projected leaf area to top canopy sunlit leaves. The percentage change in total NPP was calculated for each O_3 simulation and plotted against the cumulative uptake of O_3 over the PFT-specific accumulation period. The linear regression of this relationship was calculated, and slope and intercept compared against the slope and intercept of the observed dose-response relationships. Values of the parameter 'a' were adjusted, and the procedure repeated until the linear regression through the simulation points matched that of the observations (Fig. S2, Table S1).

~~Here we use the latest literature on O_3 dose-response relationships derived from observed field data across Europe (CLRTAP, 2017) to determine the key PFT-specific O_3 sensitivity parameters in JULES (*a* and Fo_{3crit}). Each JULES PFT (broadleaf, needle leaf, C_3 and C_4 herbaceous, and shrub) was calibrated for a high and low plant O_3 sensitivity to account for uncertainty in variation of species sensitivity to O_3, using the approach of Sitch *et al*., (2007). For the C_3 herbaceous PFT – the dominant land cover type across Europe in this study (Fig. S1) - the O_3 sensitivity was calibrated against observations for wheat to give a representation of agricultural regions (high plant O_3 sensitivity), versus natural grassland (low plant O_3 sensitivity), with a separate function for Mediterranean grasslands (low plant O_3 sensitivity) (Table S1 and Figure S2). Broadleaf tree and shrub PFTs were calibrated against the birch/beech observed O_3 dose-response functions for the high plant O_3 sensitivity, with a separate function for Mediterranean broadleaf trees (deciduous oaks), needle leaf trees were calibrated against the function for Norway spruce, all data for dose-response functions were from CLRTAP (2017). The low plant O_3 sensitivity~~

functions for trees/shrubs were calibrated as being 20% less sensitive based on the difference in sensitivity between high and low sensitive tree species in the Karlsson et al. (2007) study. Due to limitations in data availability, the parameterisation for C_4 herbaceous uses the observed dose-responses for C_3 herbaceous, however the fractional cover of C_4 herbs across Europe is low (Fig. S1), so this assumption affects a very small percentage of land cover.

To calibrate each JULES PFT for sensitivity to O_3, JULES was run, varying the value of parameter *a*, until model output of change in NPP with cumulative O_3 exposure matched the observed O_3 dose-response functions in CLRTAP (2017). JULES was run to be as directly comparable as possible to the dose-based O_3 risk indicator used in CLRTAP (2017), using the O_3 flux per projected leaf area to top canopy sunlit leaves. Values of F_{O3crit} came from the observations, the parameter '*a*' was modified until the modelled change in response variable with cumulative uptake of O_3 above the specified threshold matched the observations (see further method details in SI section S2).

Commented [ORJ20]: Line 286 to 289

2.3 Representation of stomatal conductance and site level evaluation

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

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

In this parameterisation, c_i is unknown and in the default JULES model is calculated as in equation 6, hereafter called JAC:

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

β is a soil moisture stress factor, the factor 1.6 accounts for g_s being the conductance for water vapour rather than 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 leaf surface and internal CO_2 partial pressures, respectively, c_* (Pa) is the CO_2 photorespiration compensation 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 representing the critical humidity deficit at the leaf surface, and the leaf internal to atmospheric CO_2 ratio (c_i/c_a) at the leaf specific humidity deficit (Best *et al.* 2011), values are shown is Table S1.

In this work, we replace equation 6 with the closure described in Medlyn et al. (2011), using the key PFT specific model parameter g_1 (kPa$^{0.5}$), and dq is expressed in kPa, shown in eq 7, hereafter called MED:

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

PFT specific values of the g_1 parameter were derived for European vegetation from the data base of Lin et al. (2015) and are shown in Table S1. The g_1 parameter represents the sensitivity of g_s to the assimilation rate, i.e.

plant water use efficiency, and was derived as in Lin et al. (2015) by fitting the Medlyn *et al*., (2011) model to observations of g_s, photosynthesis, and VPD, with no g_0 term.

The impact of g_s model formulation (JAC versus MED) on simulated water, O_3, carbon and energy fluxes is compared for two contrasting grid points - wet (low soil moisture stress) and dry (high soil moisture stress) in the European domain. JULES was spun-up for 20 years (1979-1999) at two grid points in central Europe representing a wet (low soil moisture stress, lat: 48.25; lon:, 5.25) and a dry site (high soil moisture stress, lat: 38.25; lon:, -7.75). The modelled soil moisture stress factor (*fsmc*) at the wet site ranged from 0.8 to 1.0 over the year 2000 (1.0 indicates no soil moisture stress), and at the dry site *fsmc* steadily declined from 0.8 at the start of the year to 0.25 by the end of the summer. The WFDEI meteorological forcing dataset was used (Weedon, 2013), along with atmospheric CO_2 concentration for the year 1999 (368.33 ppm), and either no O_3 (i.e. the O_3 damage model was switched off) for the control simulations, or spatially explicit fields of present day O_3 concentration produced using the UK Chemistry and Aerosol (UKCA) model from the run evaluated by O'Connor et al. (2014) for the simulations with O_3. Following the spin-up period, JULES was run for one year (2000) with corresponding atmospheric CO_2 concentration, and tropospheric O_3 concentrations as described above. The control and O_3 simulations were performed for both JAC and MED model formulations. Land cover for the spin-up and main run was fixed at 20% for each PFT. For the simulations including O_3 damage, the high plant O_3 sensitivity parameterisation was used. The difference between these simulations was used to assess the impact of g_s model formulation on the leaf level fluxes of carbon and water. We calculate and report (results section 3.1) the difference in mean annual water-use that results from the above simulations using the different g_s models. For each day of the simulation we calculate the percentage difference in water-use between the two simulations, we then calculate the mean and standard deviation over the year to give the annual mean leaf-level water-use.

Site level evaluation of the two g_s models compared to FLUXNET observations was carried out to evaluate the seasonal cycles of latent and sensible heat using the two g_s models JAC and MED compared to observations. Seven Fluxnet towers were selected to represent a range of land cover types as shown in Table S2. JULES was setup for each site using observed site-level hourly meteorology, and the vegetation cover was prescribed according to the fractional covers of the different JULES surface types shown in Table S2. Following a spin-up period, simulations were run at each site for the years shown in Table S2.

2.4 Model simulations for Europe

2.4.1 Forcing datasets

We used the WATCH meteorological forcing data set (Weedon et al., 2010;Weedon et al., 2011) at 0.5º x 0.5º spatial and three hour temporal resolution for our JULES simulations. JULES interpolates this down to an hourly model time step. For this study, the climate was kept constant by recycling over the period 1901 to 1920, to allow us to focus on ~~fully understand~~ the impact O_3, CO_2 and their interaction.

Formatted: Space After: 8 pt, Adjust space between Latin and Asian text, Adjust space between Asian text and numbers

Formatted: Font: Italic

Formatted: Font: Italic

Commented [ORJ21]: Line 335

JULES was run with prescribed annual mean atmospheric CO_2 concentrations. Pre-industrial global CO_2 concentrations (1900 to 1960) were taken from Etheridge et al. (1996), 1960 to 2002 were from Mauna Loa (Keeling and Whorf, 2004), as calculated by the Global Carbon Project (Le Quéré et al., 2016), and 2003-2050 were based on the IPCC SRES A1B scenario and were linearly interpolated to gap fill missing years (Fig. 1).

JULES was run including dynamic vegetation with a land cover mask giving the fraction of agriculture in each 0.5° x 0.5° grid cell based on the Hurtt et al. (2011) land cover database for the year 2000. This means that whilst the model is allowed to evolve its own vegetation cover, within tThe agricultural mask means that only C_3/C_4 herbaceous PFTs are allowed to grow, with no competition from other PFTs. Therefore, through the simulation period, regions of agriculture are maintained as such and not out-competed by forests for example, allowing for a more accurate representation of the land cover of Europe in the model., Nno form of land management is simulated (i.e. no crop harvesting, ploughing, rotation or grazing), growth and leaf area index (LAI) are determined by resource availability and phenology.. Outside of the agricultural mask, By including dynamic vegetation means that, grid cell PFT coverage and Leaf Area Index (LAI) is are thea result of resource availability, and penology and simulated competitionn. Across the model domain, simulated mean annual LAI was dominantly within the range of 2 to 5 m^2/m^2 (Fig. S3 and S4). Following a full spin-up period (to ensure equilibrium vegetation, carbon and water states), there was no significant change in the fractional cover of each PFT changed little oover the simulation period (1901 - 2050)., By 2050, increases in boreal forest cover occurred, but this was less than 2% and limited to very small areas, given this small change we show just the land cover for 2050 is shown in Fig. S1.

Tropospheric O_3 concentration was produced by the EMEP MSC-W model at 0.5° x 0.5° (Simpson et al., 2012), driven with meteorology from the regional climate model RCA3 (Kjellström et al., 2011;Samuelsson et al., 2011), which provides a downscaling of the ECHAM A1B-r3 (simulation 11 of Kjellström *et al.*, 2011). This setup (EMEP+RCA3) is also used by Langner et al. (2012a), Simpson et al. (2014a), Tuovinen et al. (2013), Franz et al. (2017) and Engardt et al. (2017), where further details and model evaluation can be found. Unfortunately, the 3-dimensional RCA3 data needed by the EMEP model was not available prior to 1960, but as in Engardt et al. (2017) the meteorology of 1900-1959 had to be approximated by assigning random years from 1960 to 1969. This procedure introduces some uncertainty of course, althoughbut Langner et al. (2012b) show that for the period 1990 to 2100 it is emissions change, rather than meteorological change, that drives modelled O_3ozone concentrations. The emissions scenarios for 1900-2050 merge data from the International Institute of Applied System Analysis (IIASA) for 2005-2050 (the so-called ECLIPSE 4a scenario), recently revised EMEP data for 1990, and a scaling back from 1990 to 1900 using data from Lamarque et al. (2013). The trend in emissions of the major O_3 precursors NO_x, NMVOC and Isoprene are shown from 1900 to 2050 over Europe in Fig. S5. Isoprene emissions are not inputs to the EMEP model, but rather calculated at each time-step using temperature, radiation, and land-cover specific emission factors (Simpson et al., 2012). Changes in the assumed background concentration of CH_4 (from RCP6.0) (van Vuuren et al., 2011) are also shown in Fig. S5. Engardt et al. (2017) show the trend in emissions of SO_2 and NH_3 from 1900 to 2050 over Europe. The EMEP model accounts for changes in BVOC emissions as a result of predicted ambient temperature changes, however as with all uncoupled modelling studies, there is no interaction between changes in leaf-level g_s, BVOCs and O_3 formation.

Commented [ORJ22]: Line 345
Formatted: Not Highlight
Commented [ORJ23]: Line 347
Formatted: Not Highlight
Formatted: Subscript
Commented [ORJ24]: Lines 356 to 358
Formatted: Not Highlight
Formatted: Font: (Default) Times New Roman, 10 pt
Formatted: Font: (Default) Times New Roman, 10 pt
Formatted: Font: (Default) Times New Roman, 10 pt
Formatted: Not Highlight
Formatted: Font: (Default) Times New Roman, 10 pt
Commented [ORJ25]: Line 358 to 361

O_3 concentrations from EMEP MSC-W were calculated at canopy height for two land-cover categories: forest and grassland (Fig. S6~~3~~ and Fig. S7~~4~~), which are taken as surrogates for high and low vegetation, respectively. These canopy-height specific concentrations allow for the large gradients 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 m in a forest canopy (Gerosa et al., 2017;Simpson et al., 2012;Tuovinen et al., 2009) ~~(Simpson et al., 2012;Tuovinen et al., 2009).~~ These canopy level O_3 concentrations are used as input to JULES, using the EMEP O_3 concentrations for forest for the forest JULES PFTs (broadleaf/needle leaf tree and shrub), and the EMEP O_3 concentrations for grassland for the grass/herbaceous JULES PFTs (C_3 and C_4). This study used daily mean values of tropospheric O_3 concentration from EMEP disaggregated down to the hourly JULES model time-step. The daily mean O_3 forcing was disaggregated to follow a mean diurnal profile of O_3, this was generated from hourly O_3 output from EMEP MSC-W for the two land cover categories (forest and grassland as described above) across the same model domain. ~~Hourly~~ O_3 concentrations follow a diurnal cycle and peak during the day, therefore accounting for the diurnal variation in O_3 concentrations ~~values~~ allows for a ~~variation in the diurnal response to O_3 exposure resulting in~~ more realistic~~accurate~~ estimation of O_3 uptake.

Figure 1 shows large increases in tropospheric O_3 from pre-industrial to present day (2001), this is in line with modelling studies (Young et al., 2013) and site observations (Derwent et al., 2008;Logan et al., 2012;Parrish et al., 2012), and is predominantly a result of increasing anthropogenic emissions (Young et al., 2013). Figure~~'~~s S6~~3~~ and S7~~4~~ show this large increase in ground-level O_3 concentrations from 1901 to 2001 occurs in all seasons. Present day O_3 concentration show a strong seasonal cycle, with a spring/summer peak in concentrations in the mid-latitudes of the Northern Hemisphere (Derwent et al., 2008;Parrish et al., 2012;Vingarzan, 2004). Seasonal cycles have been changing over the past decades however, attributed to changes in NO_x and other emissions, as well as changes in transport patterns (Parrish et al., 2013). These changes will likely continue in future as emissions and meteorological factors impact photo-chemical O_3 production and transport patterns. Indeed, ~~This is largely related to(Lin et al., 1988) he seasonal cycle ofphotochemical O_3 production which is highest during periods of high radiation and temperature (Young et al., 2013), although increased stratospheric input is also thought to contribute (Vingarzan, 2004). Anthropogenic emissions, especially NO_X, contribute to the seasonal cycle of O_3 through traffic, energy production and residential heating and cooling demands (Royal-Society, 2008). Bioegenic emissions are also seasonal which contributes to the seasonal change in O_3 concentrations (Pacifico et al., 2012;Young et al., 2009), and dry deposition, driven by plant productivity also has a strong seasonal component. How the seasonality of ground level O_3 changes in the future will depend on how these multiple different drivers change and interact. Modelling studies such as Dentener et al. (2006) and Young et al. (2013) suggest that anthropogenic emissions will be the main factor controlling the evolution of future O_3 concentrations, and in the recent study of Young et al., (2013) most scenarios suggest reduced O_3 burden in the future as a result predominantly of reduced precursor emissions. Seasonally, t~~the O_3 concentrations used in the simulations in this study show increased O_3 levels in winter and in some regions in autumn and spring in 2050 compared to present day, this may be due to reduced titration of O_3 by NO as a result of reduced NO_X emissions in the future (Royal Society, 2008). Summer O_3 concentrations are lower in 2050 however, compared to 2001. ~~Our simulations use a fixed climate, so we do not include the effect of climate change on shifting plant phenology. Therefore, our results~~

Commented [OT26]: Line 367-369

Formatted: Font: (Default) Times New Roman, 10 pt

Formatted: Font: (Default) Times New Roman, 10 pt, Subscript

Formatted: Font: (Default) Times New Roman, 10 pt

Formatted: Font: (Default) Times New Roman, 10 pt

Formatted: Font: (Default) Times New Roman, 10 pt

Commented [ORJ27]: Line 381 to 394

~~may underestimate plant O_3 damage, since if the growing season started earlier or finished later, plants in some regions would be exposed to higher O_3 concentrations.~~

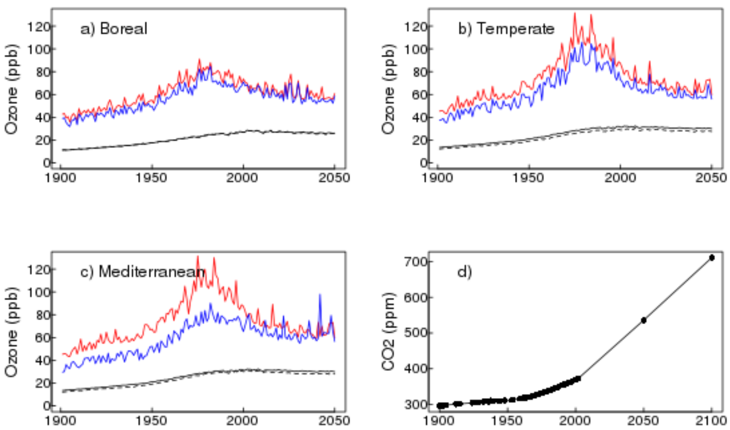

Figure 1. Regional time series of canopy height O_3 (ppb) forcing from EMEP a) to c), and d) global atmospheric CO_2 (ppm) concentration (this does not vary regionally; black dots show data points, the black line shows interpolated points). Each panel for the O_3 forcing shows the regional annual average (woody PFTs, black solid line; herbaceous PFTs, black dashed line) and the annual maximum O_3 concentration above: woody PFTs (red) and herbaceous PFTs (blue).

2.4.2 Spin up and factorial experiments

JULES was spun-up by recycling the climate from the early part of the twentieth century (1901 to 1920) using atmospheric CO_2 (296.1 ppm) and O_3 concentrations from 1901 (Fig. S3 & Fig. S4). Model spin-up was 2000 years by which point the carbon pools and fluxes were in steady state with zero mean net land – atmosphere CO_2 flux. We performed the following transient simulations for the period 1901 to 2050 with continued recycling of the climate as used in the spin-up, for both high and low plant O_3 sensitivities:

- **O3** : Fixed 1901 CO_2, Varying O_3
- **CO2** : Varying CO_2, Fixed 1901 O_3
- **CO2~~+~~O3** : Varying CO_2, Varying O_3

We use these simulations to investigate the direct effects of changing atmospheric CO_2 and O_3 concentrations, individually and combined, on plant water-use, GPP and the land C sink~~physiology~~ through the twentieth century and into the future, specifically over three time periods: historical (1901-2001), future (2001-2050) and over the full time series (1901-2050). For each time period we calculate the difference between the decadal means

Formatted: Normal

Commented [ORJ28]: Line 420

calculated at the start and end of the analysis period for each variable of interest. Therefore our results report the change in GPP, for example, over the analysis period. ~~.~~ For each variable analysed (GPP, NPP, vegetation carbon, soil carbon, total land carbon and *gs*), we use the mean over 10 years to represent each time period, e.g. the mean over 2040 to 2050 is what we call 2050, 1901 to 1910 is what we refer to as 1901. The difference between the simulations gives the effect of O_3 and CO_2 either separately or in combination over the different time periods. We look at the percentage change due to either O_3 at pre-industrial CO_2 concentration (i.e. without the additional effect of atmospheric CO_2 on stomatal behaviour - O3 simulation), CO_2 (at fixed pre-industrial O_3 concentration, CO2 simulation) or the combined effect of both gases (CO2+O3 simulation), which is calculated as:

$$100 * (var[y_1] - var[y_2]) / var[y_2] \quad (8)$$

Where $var[y_x]$ represents the variable in time period y, e.g. $100 * (varO_3[2050] - varO_3[1901]) / varO_3[1901]$ gives the O_3 effect (at fixed CO_2) over the full experimental period. The meteorological forcing is prescribed in these simulations and is therefore the same between the model runs. Other climate factors, such as VPD, temperature and soil moisture availability are accounted for in our simulations, but our analysis isolates the effects of O_3, CO_2 and O_3 + CO_2. We also use paired t-test to determine statistically significant differences between the different (high and low) plant O_3 sensitivities.

2.4.3 Evaluation

To evaluate our JULES simulations we compare mean GPP from 1991 to 2001 for each of the JULES scenarios and both high and low plant O_3 sensitivities against the observation based globally extrapolated Flux Network model tree ensemble (MTE) (Jung et al., 2011). We use paired t-test to determine statistically significant differences in the mean responses.

3 Results

3.1 Impact of g_s model formulation and site level evaluation

The impact of g_s model on simulated g_s is shown for the site with low soil moisture stress (wet site, Fig. 2). For the broadleaf tree and C_3 herbaceous PFT, the MED model simulates a larger conductance compared to the JAC model. In other words, with the MED model these two PFTs are parameterised with a less conservative water use strategy, which, for the grid point shown in Fig. 2, increased the annual mean ~~leaf level~~ water use by 35% (±29%) and 45% (±32%), respectively. In contrast, the needle leaf tree, C_4 herbaceous and shrub PFTs are parameterised with a more conservative water use strategy with the MED model, and the mean annual g_s was decreased by 13% (±12%), 27% (±10%) and 36% (±13%), respectively, compared to the JAC model. This comparison was also done for a dry site (high soil moisture stress), and similar results were found (Fig. S8~~6~~)~~), suggesting these results are representative across the domain~~. The effect of g_s formulation on simulated photosynthesis was much smaller because of the lower sensitivity of the limiting rates of photosynthesis to changes in c_i in the model compared to

Commented [ORJ29]: Line 435

Commented [ORJ30]: Line 441

Formatted: Not Highlight

Formatted: Highlight

Commented [ORJ31]: Line 442 to 445

the effect of the same change in c_i on modelled g_s (Fig. S9~~7~~ & S10~~8~~). Changes in~~ leaf level~~ g_s impact the partitioning of simulated energy fluxes. In general, increased g_s results in increased latent heat and thus decreased sensible heat flux, and vice versa where g_s is decreased (Fig. S9~~7~~ & S10~~8~~). Also shown is the effect of the MED model on O_3 flux into the leaf (Fig. S11~~2~~ and Fig. S~~86,~~ bottom panel~~s~~). For the broadleaf tree and C_3 herbaceous PFT, the MED model simulates a larger conductance and therefore a greater flux of O_3 through stomata compared to JAC, and this is indicative of the potential for greater reductions in photosynthesis (Fig. S9~~7~~ & S10~~8~~ top row). The reverse is seen for the needle leaf tree, C_4 herbaceous and shrub PFTs.

Site level evaluation of the seasonal cycles of latent and sensible heat with both JAC and MED models compared to FLUXNET observations showed in general, the MED model improved the seasonal cycle of both fluxes (lower RMSE), but the magnitude of this varied from site to site (Fig. S12). At the deciduous broadleaf site, US-UMB, MED resulted in improvements of the simulated seasonal cycle particularly in the summer months for both fluxes (RMSE decreased from 42.7/31.5 to 38.5/28.0 W/m^2 for latent/sensible heat respectively). At the second deciduous broadleaf site IT-CA1 however, there was almost no difference between the two g_s models. Both evergreen needle leaf forest sites (FI-Hyy and DE-Tha) saw improvements in the simulated seasonal cycles of latent and sensible heat with the MED model, primarily as a result of lower latent heat flux in the spring and summer months, and higher sensible heat flux over the same period. At FI-Hyy, RMSE decreased from 10.1/7.4 to 6.7/6.7 W/m^2 for latent/sensible heat respectively, and at DE-Tha, RMSE decreased from 16.0/11.9 to 10.5/10.6 W/m^2 for latent/sensible heat respectively. With the MED model the monthly mean latent heat flux was improved at the C_3 grass site (CH-Cha) as a result of increased flux in the summer months (RMSE decreased from 15.7 to 13.8 W/m^2), however there was no improvement in the sensible heat flux and RMSE with MED was increased (from 3.9 to 4.9 W/m^2). At the C_4 grass site (US-SRG), small improvements were made in the seasonal cycle of both latent and sensible heat with the MED model. At the deciduous savannah site (CG-Tch) which included a high proportion of shrub PFT in the land cover type used in the site simulation, large improvements in the seasonal cycle of both fluxes were simulated with the MED model, as a result of a decrease in the latent heat flux and an increase in the sensible heat flux (RMSE decreased from 39.5/31.6 to 30.4/24.4 W/m^2 for latent/sensible heat respectively).

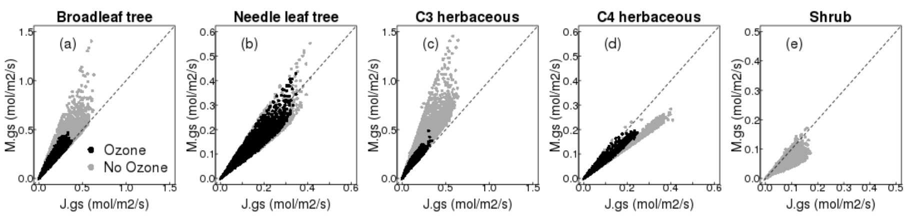

Figure 2. Comparison of simulated g_s with MED (y axis) versus JAC (x axis) for all five JULES PFTs at one grid point (lat: 48.25; lon:, 5.25) shown are hourly values for the year 2000 (see SI section S3 for further details).

Formatted: Not Highlight

Formatted: Not Highlight

Formatted: Not Highlight

Commented [ORJ32]: Line 326

3.2 Evaluation of the JULES O_3 model

For all JULES scenarios similar spatial patterns of GPP are simulated compared to MTE (Fig. 3 and Fig. S130). MTE estimates a mean GPP for present day in Europe of 938 gC m^2 yr^{-1} (Fig. 3). JULES tends to under-predict GPP relative to the MTE product, estimates of GPP from JULES with both transient CO_2 and O_3 (CO2+O3 simulation) gives a mean across Europe of 813 gC m^2 yr^{-1} (high plant O_3 sensitivity) to 881 gC m^2 yr^{-1} (low plant O_3 sensitivity), both of which are significantly different to the MTE product (t=27, $d.f.$=5750, p<2.2e^{-16} (high); t=4.3, $d.f.$=5750, p<1.5e^{-05} (low); Fig. 3). Forcing with CO_2 alone (CO2 simulation~~fixed 1901 O_3~~) gives a mean GPP across Europe of 900 to 923 gC m^2 yr^{-1} (high and low plant O_3 sensitivity respectively), and O_3 alone (O3 simulation - without the protective effect of CO_2) reduces estimated GPP to 732 to 799 gC m^2 yr^{-1} (Fig. S130). At latitudes >45°N JULES has a tendency to under-predict MTE-GPP, and at latitudes <45 °N JULES tends to over-predict MTE-GPP (Fig. S141). These regional differences are highlighted in Fig. S152, where in the Mediterranean region, JULES tends to over-predict compared to MTE-GPP, so simulations with O_3 reduce the simulated GPP bringing it closer to MTE. In the temperate region however, JULES tends to under-estimate MTE-GPP, so the addition of O_3 reduces simulated GPP further (Fig. S152). In the boreal region, JULES under-predicts GPP, but to a lesser extent than in the temperate region, and the addition of O_3 has less impact on reducing the GPP further (Fig. S152).

Formatted: Not Highlight

Formatted: Not Highlight

Formatted: Not Highlight

Formatted: Not Highlight

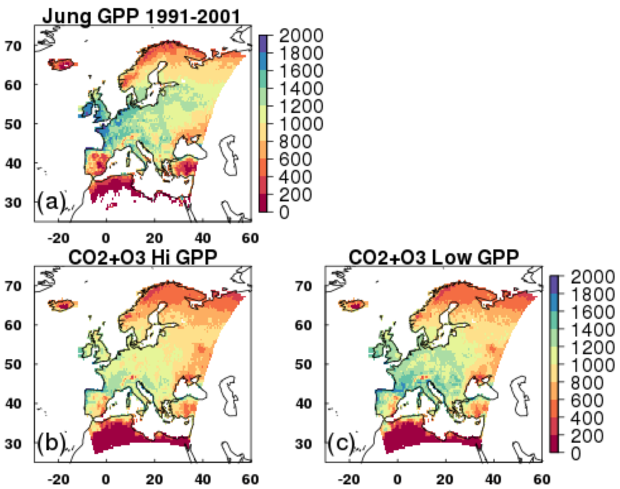

Figure 3. Mean GPP (g C m^2 yr^{-1}) from 1991 to 2001 for a) the observationally based globally extrapolated Flux Network model tree ensemble (MTE) (Jung *et al*., 2011); b, c) model simulations with transient CO_2 and transient O_3 (CO2+O3), high and low plant O_3 sensitivity respectively.

3.3 European simulations - Historical Period: 1901-2001

Over the historical period (1901-2001), ~~the physiological effect of~~ O_3 (O3 simulation) reduced GPP under both the low and high plant O_3 sensitivity parameterizations by ~~(~~-3% to -9% respectively (Table 1)~~, for the low and high plant O_3 sensitivity parameterizations, respectively (Table 1). The~~ and this difference in simulated GPP ~~plant O_3 sensitivity~~ was significant (t=102.2, $d.f.$=6270, p<2.2e^{-16}). Figure 4 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 small increases in GPP over this period, associated with O_3 induced stomatal closure enhancing water availability in these drier regions (Fig. 5). This allows for greater stomatal conductance later in the year when soil moisture may otherwise have been limiting to growth (up to 10%, Fig. 5), and therefore higher 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. 6), and these are not associated with notable increases in soil moisture availability (Fig. 5), 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 suppressed (-2% to -6%, Table 1). Large regional variation is shown in Figure 4, with temperate and Mediterranean Europe seeing a large reduction in land carbon storage, particularly under the high plant O_3 sensitivity (up to -15%).

Commented [ORJ33]: Line 484 to 485

Combined, the physiological response to changing CO_2 and O_3 concentrations (CO2+O3 simulation) 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 S3). 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 S3).

To gain perspective on the magnitude of the O_3 induced flux of carbon from the land to the atmosphere we relate changes in total land carbon to carbon emissions from fossil fuel combustion and cement production for the EU-28-plus countries from the data of Boden et al. (2013). We recognise that our simulation domain is slightly larger than the EU28-plus as it includes a small area of western Russia so direct comparisons cannot be made, but this still provides a useful measure of the size of the carbon flux. For the period 1970 to 1979 the simulated loss of carbon from the European terrestrial biosphere due to O_3 effects on vegetation physiology was on average 1.32 Pg C (high vegetation sensitivity) and 0.71 Pg C (low vegetation sensitivity) (Table 2). This O_3 induced reduced C uptake of the land surface is equivalent to around 8% to 16% of the emissions of carbon from fossil fuel combustion and cement production over the same period for the EU28-plus countries (Table 2). Currently the emissions data availability goes up to 2011, over the last observable decade (2002 to 2011) the simulated reduction in~~is~~ land carbon due to O_3 ~~loss~~ has declined, but is still equivalent to 2% to 4% of the emissions of carbon from fossil fuels and cement production for the EU28-plus countries (Table 2). By comparison with one of the largest anthropogenic emissions of carbon for Europe, we show here~~Therefore,~~ the potential effect of ~~indirect~~ O_3 ~~effect~~

on reducing the size of the European land carbon sink is notable. ~~potentially represents a significant source of anthropogenic carbon.~~

3.4 European simulations - Future Period: 2001-2050

Over the 2001 to 2050 period, region-wide GPP with O_3 only changing (O3 simulation) increased marginally (+0.1% to +0.2%, high and low plant O_3 sensitivity, Table 1, with a significant difference between the two plant O_3 sensitivities (t=57, $d.f.$=6270 p<2.2e^{-16})), although with large spatial variability as discussed below (Fig. 4g & h). Figures S6~~3~~ and S7~~4~~ show that despite decreased tropospheric O_3 concentrations by 2050 in summer compared to 2001 levels, all regions are exposed to an increase in O_3 over the wintertime, and some regions of Europe, particularly temperate/Mediterranean experience increases in O_3 concentration in spring and autumn. Therefore, although in the O3 simulation, overall simulated GPP for Europe shows a small increase, large spatial variability is shown in Fig's 4g &h because of the variability in O_3 concentration with region and season. I~~i~~ncreased GPP (dominantly 10%, but up to 20% in some areas) on 2001 levels is simulated across areas of Europe, however, decreases of up to 21% are simulated in some areas of the Mediterranean, up to 15% in some areas of the boreal region and up to 27% in the temperate zone (Fig. 4g & h).

When O_3 and CO_2 effects are combined (CO2+O3 simulation), simulated GPP increases (+15% to +18%, high/low plant O_3 sensitivities respectively, Table 1). This increase is greater than the enhancement simulated when CO_2 affects plant growth independently (CO2 simulation), because additional O_3 induced stomatal closure increases soil water availability in some regions, which enhances growth more in the CO2+O3~~O_3, and CO_2~~ simulation~~s~~, compared to the CO2~~$_2$~~ simulation~~only run~~. Nevertheless, although the percentage gain is larger, the absolute value of GPP by 2050 remains lower ~~compared~~ in CO2+O3 compared to GPP in the CO2 simulations~~with CO_2 only changing~~, highlighting the negative impact of O_3 at the land surface (Table S4).

Despite small increases in GPP in the O3 ~~O_3-only~~ simulation, the land carbon sink continues to decline from 2001 levels (-0.7% to -1.6%, ~~-~~low and high plant O_3 sensitivity respectively, Table 1). This is because the soil and vegetation carbon pools continue to lose carbon as they adjust slowly to small changes in input (GPP), i.e. the soil carbon pool is not in equilibrium in 2001, and is declining in response to reduced litter input as a result of 20th C O_3 impacts on GPP. Nevertheless, the negative effect of O_3 on the future land sink is markedly reduced relative to the historical period. Figure 4e & f however highlights regional differences. Boreal regions and parts of central Europe see minimal O_3 damage, whereas some areas of southern and northern Europe see further losses of up to 8% on 2001 levels. The CO2+O3 simulation ~~combined O_3 and CO_2 effects~~ are dominated by the physiological effects of changing CO_2, with land carbon sink increases of up to 7% (Table 1).

3.5 European simulations ~~–~~ – Full experimental period~~Anthropocene~~: 1901-2050

From ~~Over the Anthropocene~~1901 to 2050, the O3 simulation~~O_3~~ reduces GPP (-4% to -9%, with a significant difference between the low and high plant O_3 sensitivity (t=95, $d.f.$=6270 p<2.2e^{-16})) and land carbon storage (-3% to -7%, Table 1~~, Fig. S13~~). Regionally, O_3 damage is lowest in the boreal zone, GPP decreases are largely

Commented [ORJ34]: Line 516 to 517

Commented [ORJ35]: Line 523
Line 527 to 529
Line 529

Commented [ORJ36]: Line 533 to 534

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 7), consistent with lower g_s of needle leaf trees that dominate this region, and so lower O_3 uptake (Fig. S16~~4~~ & S17~~5~~). 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 7). 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 7). In these drier regions, O_3 induced stomatal closure can increase available soil moisture (Fig. S16~~4~~ & S17~~5~~).

The CO2+O3 simulation ~~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 S6). 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 (CO2+O3), rather than 4% to 9% lower in the absence of increasing CO_2 (O3 simulation, Table S6). 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.

From ~~Over the Anthropocene~~1901 to 2050, the CO2+O3 simulation~~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. S18~~3~~). By 2050 the simulated enhancement of land carbon and GPP in response to elevated CO_2 alone (CO2 simulation) 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 (CO2+O3 simulation, Table 1). This is a large reduction in the ability of the European terrestrial biosphere to sequester carbon.

Commented [ORJ37]: Line 567

Formatted: Not Highlight

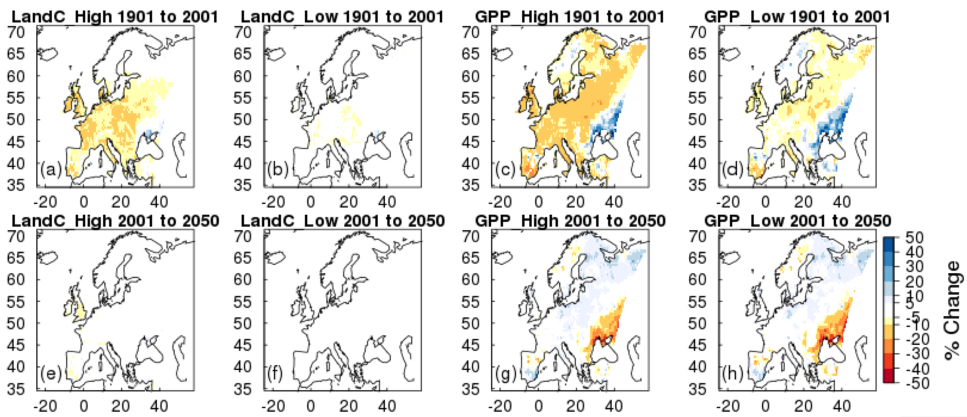

Figure 4. Simulated percentage change in total carbon stocks (Land C) and gross primary productivity (GPP) due to O_3 effects at fixed pre-industrial atmospheric CO_2 concentration (O3 simulation). Changes are shown for the periods 1901 to 2001, and 2001 to 2050 for the high and low plant O_3 sensitivity.

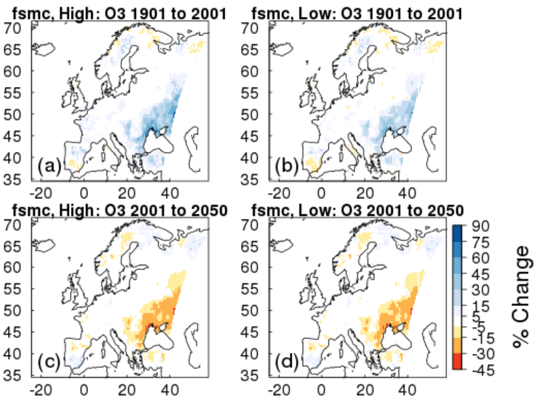

Figure 5. Simulated percentage change in plant available soil moisture (*fsmc*) due to O_3 effects at fixed pre-industrial atmospheric CO_2 concentration (O3 simulation). Changes are shown for the periods 1901 to 2001, and 2001 to 2050 for the high and low plant O_3 sensitivity.

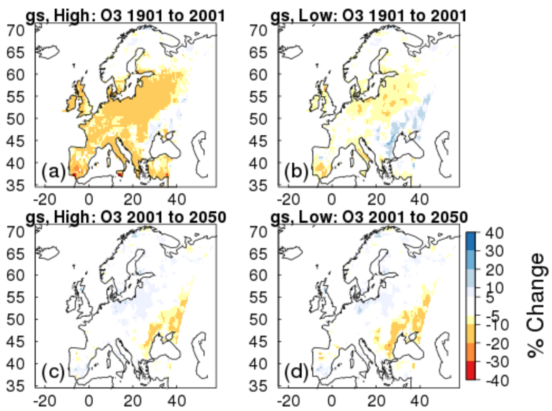

Figure 6. Simulated percentage change in stomatal conductance (g_s) due to O_3 effects at fixed pre-industrial atmospheric CO_2 concentration (O3 simulation). Changes are shown for the periods 1901 to 2001, and 2001 to 2050 for the high and low plant O_3 sensitivity.

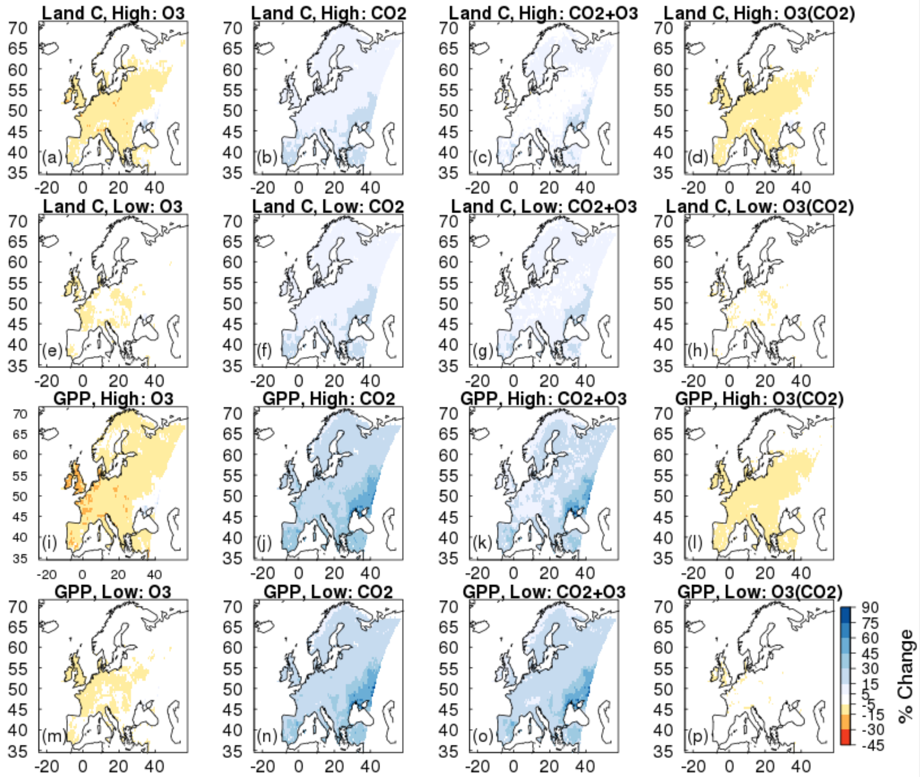

Figure 7. Simulated percentage change in total carbon stocks (Land C) and gross primary productivity (GPP) due to i) (a, e, i, m) O_3 effects at fixed pre-industrial atmospheric CO_2 concentration (O3 simulation~~$_3$~~), ii) (b, f, j, n) CO_2 fertilisation at fixed pre-industrial O_3 concentration (CO2 simulation~~$_2$~~), iii) (c, g, k, o) the interaction between O_3 and CO_2 effects (CO2+~~$_2$ +~~ O3 simulation~~$_3$~~) iv) (d, h, l, p) O_3 effects with changing atmospheric CO_2 concentration (i.e. O_3 damage accounting for the effect of CO_2 induced stomatal closure; CO2+O3 – CO2~~$O_3(CO_2)$~~). 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_2	-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

Table 1. Simulated changes in the European land carbon cycle due to changing O_3 and CO_2 concentrations (independently and together). Shown are changes in total carbon stocks (Land C) and gross primary productivity (GPP), over three different periods (historical: 1901 to 2001, future: 2001 to 2050, and full time series~~Anthropocene~~: 1901 to 2050). Absolute (top) and relative (bottom) differences are shown. For 2001 to 2050, please refer to Table S4 for 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.

	Mean (Pg C)				
	1970-1979	1980-1989	1990-1999	2000-2009	2002-2011
Modelled O_3 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 O_3 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

Table 2. Simulated change in total land carbon due to O_3 damage with changing atmospheric CO_2 concentration for the two vegetation sensitivities. The sum of carbon emissions for each decade from fossil fuel combustion and cement production for the EU-28 countries plus Albania, Bosnia and Herzegovina, Iceland, Belarus, Serbia, Moldova, Norway, Turkey, Ukraine, Switzerland and Macedonia (EU28-plus) are shown, the data is from Boden *et al.*, 2013. The simulated change in land carbon as a result of O_3 damage is depicted as a percentage of the EU28-plus emissions to demonstrate the magnitude of the additional source of carbon to the atmosphere from plant O_3 damage.

4 Discussion

4.1 Evaluation~~Comparison~~ of g_s models and JULES O_3 model

Comparison of the new g_s model implemented in this study (MED) with the g_s model currently used as standard in JULES (JAC) revealed large differences in ~~leaf-level~~ g_s for each PFT, principally as a result of the data-based parameterisation of the new model. W~~Leaf-level w~~ater use increased for the broadleaf tree and C_3 herbaceous PFTs using the MED model compared to JAC, but decreased for the needle leaf tree, C_4 herbaceous and shrub PFTs which displayed a more conservative water use strategy compared to JAC~~the Jacobs parameterisation~~. These changes are in line with the work of De Kauwe et al. (2015) who found a reduction in annual transpiration for evergreen needle leaf, tundra and C_4 grass regions when implementing the Medlyn g_s model into the Australian land surface scheme CABLE. Site-level evaluation of the models against Fluxnet observations showed that in general the MED model improved simulated seasonal cycles of latent and sensible heat. The magnitude of the improvement varied with site, large improvements were seen at the deciduous savanna site, and at the NT sites and BT site (US_UMB) in the spring and summer. However, much smaller improvements were seen at the grass sites. Changes in ~~leaf-level~~ g_s in this study resulted in differences in latent and sensible heat fluxes. Changes in the partitioning of energy fluxes at the land surface could have consequences for the intensity of heatwaves (Cruz et al., 2010;Kala et al., 2016), runoff (Betts et al., 2007;Gedney et al., 2006) and rainfall patterns (de Arellano et al., 2012), although fully coupled simulations would be necessary to detect these effects. The differences in simulated g_s led to differences in uptake of O_3 between the two models because the ~~leaf-level~~ rate of g_s is the predominant determinant of the flux of O_3 through stomata. Higher O_3 uptake is indicative of greater damage. Therefore, given that C_3 herbaceous vegetation is the dominant land cover class across the European domain used in this study, this suggests a greater O_3 impact for Europe would be simulated with MED model compared to JAC in our simulations where chemistry is uncoupled from the land surface.

We evaluated the JULES O_3 model by comparing modelled GPP against the Jung et al (2011) MTE product. Similar spatial patterns of GPP were simulated by JULES compared to MTE. Zonal means also showed similar patterns of GPP, although JULES under predicted GPP compared to MTE at latitudes >45 °N (temperate and boreal regions; all simulations) and over predicted GPP at latitudes <45 °N (Mediterranean region; all simulations). The simulations with transient O_3 (i.e. O3 and CO2+O3) showed large differences in GPP between the high and low plant O_3 sensitivity simulations, this is to be expected given that the high plant O_3 sensitivity simulations were parameterised to be 'damaged' more by O_3, i.e. greater reduction of photosynthesis/g_s with O_3 exposure compared to the low plant O_3 sensitivity simulations. This difference was largest in the temperate zone, largely because of C_3 grass cover being the dominant land cover here and the difference in the sensitivity to O_3 between the high and low calibrations is significantly larger for C_3 grasses compared to the needle leaf trees that dominate in the boreal region. Additionally, a longer growing season in the temperate region may allow for greater uptake of O_3 into vegetation. C_3 grass is also the dominant land cover in the Mediterranean region with a different calibration used for Mediterranean grasses for the low plant O_3 sensitivity which is less sensitive to O_3 than the temperate C_3 grasses, but high soil moisture stress is common throughout the growing season in the Mediterranean limiting the uptake of O_3 through stomata, which likely diminishes the difference between the high and low calibrations.

4.2 Lower than expected O_3 damage?

~~The impact of O_3 on present day European GPP simulated in this study is slightly lower compared to previous modelled estimates.~~ Our estimates suggest present day O_3 reduced GPP by 3% to 9% on average across Europe and NPP by 5% to 11% (Table S3). Anav et al. (2011) simulated a 22% reduction of GPP across Europe for 2002 using the ORCHIDEE model. Present day O_3 exposure reduced GPP by 10% to 25% in Europe, and 10.8% globally in the study by Lombardozzi et al. (2015) using the Community land model (CLM). O_3 reduced NPP by 11.2% in Europe from 1989 to 1995 using the Terrestrial Ecosystem Model (TEM) (Felzer et al., 2005). Globally, concentrations of O_3 predicted for 2100 reduced GPP by 14% to 23% using a former parameterisation of O_3 sensitivity in JULES (Sitch et al., 2007). The recent study by Franz et al. (2017) showed mean GPP declined by 4.7% over the period 2001 to 2010 using the OCN model over the same European domain ~~used in this study~~and using the same O_3 forcing produced by EMEP MSC-W as used in this study. Our estimates of changes in current day GPP and NPP are at the lower end of previously modelled estimates. Simulated O_3 impacts will depend in a large part on the scenario of O_3 concentrations used as forcing, meteorological forcing and how sensitive vegetation is parameterised to be to O_3 damage, in addition to the different process representation of O_3 damage in each model. It is therefore difficult to hypothesise as to exactly why modelled estimates differ, but suggests

Commented [ORJ38]: Line 633 to 648

Commented [ORJ39]: Line 473

Formatted: Justified, Line spacing: 1.5 lines

Field Code Changed

Field Code Changed

Field Code Changed

Field Code Changed

Field Code Changed

Field Code Changed

Field Code Changed

Formatted: Font: (Default) Times New Roman, 10 pt

Formatted: Font: (Default) Times New Roman, 10 pt

Formatted: Font: (Default) Times New Roman, 10 pt

Formatted: Font: (Default) Times New Roman, 10 pt

Formatted: Font: (Default) Times New Roman, 10 pt

Formatted: Font: (Default) Times New Roman, 10 pt that an ensemble approach to modelling O_3 impacts on the terrestrial biosphere would be beneficial to understand some of these differences and provide estimates of O_3 damage with uncertainties.

Formatted: Font: (Default) Times New Roman, 10 pt

~~These similar results are likely the result of using the same domain, and, more importantly, O_3 forcing produced by the same model (EMEP MSC-W).~~

4.3 Impacts of O_3 at the land surface

In this study, O_3 has a detrimental effect on the size of the land carbon sink for Europe. This is primarily through a decrease in the size of the soil carbon pool as a result of reduced litter input to the soil, consistent with reduced GPP/NPP. Field studies show that in some regions of Europe, soil carbon stocks are decreasing (Bellamy et al., 2005;Capriel, 2013;Heikkinen et al., 2013;Sleutel et al., 2003). The study of Bellamy et al. (2005), for example, showed that carbon was lost from soils across England and Wales between 1978 to 2003 at a mean rate of 0.6% per year with little effect of land use on the rate of carbon loss, suggesting a possible link to climate change. It is understood that climate change is likely to affect soil carbon turnover. Increased temperatures increase microbial decomposition activity in the soil, and therefore increase carbon losses through higher rates of respiration (Cox et al., 2000;Friedlingstein et al., 2006;Jones et al., 2003). However, some studies have found that O_3 can decrease soil carbon content. Talhelm et al. (2014), for example, found O_3 reduced carbon content in near surface mineral soil of forest soils exposed to 11 years of O_3 fumigation. Hofmockel et al. (2011) found elevated O_3 reduced the carbon content in more stable soil organic matter pools, and Loya et al. (2003) showed that the fraction of soil carbon formed in forest soils over a 4 year experimental period when fumigated with both CO_2 and O_3 was reduced by 51% compared to the soil fumigated with CO_2 alone. It is agreed that amongst other factors that change with O_3 exposure such as litter quality and composition, reduced litter quantity also has significant detrimental consequences for soil carbon stocks (Andersen, 2003;Lindroth, 2010;Loya et al., 2003). Results from this study therefore suggest that increasing tropospheric O_3 may be a contributing factor to the declining soil carbon stocks observed across Europe as a result of reduced litter input to the soil carbon pool consistent with reduced NPP.

We acknowledge, however, that our model simulations do not include coupling of Nitrogen and Carbon cycles, or land management practices. ~~Although~~ W~~w~~e include a representation of agricultural regions through the model calibration against the wheat O_3 sensitivity function (CLRTAP, 2017), and in our simulations the high plant O_3 sensitivity scenario uses this calibration against wheat for all C_3/C_4 land cover which dominates our model domain. W~~w~~heat is known to be one of the most O_3 sensitive crop species however, so it is possible that our simulations over-estimate the O_3 impact at the land surface. However, the low plant O_3 sensitivity calibration against natural grasslands provides a counter estimate of the impact of O_3 at the land surface, therefore it is important to consider the range our results provide (i.e. both the high and low plant O_3 sensitivity) as an indicator of the impact of O_3 on the land surface. ~~.~~As with all uncoupled modelling studies, a change in g_s and flux will impact the O_3 concentration itself. Therefore adopting the Medlyn formulation with a higher g_s and subsequently higher O_3 flux for broadleaf and C_3 PFTs (Fig 2) would lead to reduced O_3 concentration, which in turn would act to dampen the effect of higher g_s on O_3 flux, although the higher uptake of O_3 by vegetation may lead to more damage and increase O_3 concentrations, in an uncoupled chemistry-land modelling system such as this it is not possible to predict which process would dominate.~~. Essentially this study provides an 'upper bound' as in the high plant O_3~~

Commented [ORJ40]: Line 687 to 691

sensitivity scenario, all C_3/C_4 fractional cover uses the wheat O_3 sensitivity. Additionally, this version of JULES does not have a crop module; it has no land management practices such as harvesting, ploughing or crop rotation – processes which may have counteracting effects on the land carbon sink. Further, without a coupled Carbon and Nitrogen cycle, it is likely that the CO_2 fertilisation response of GPP and the land carbon sink is over estimated in some regions of our simulations since nitrogen availability limits terrestrial carbon sequestration of natural ecosystems in the temperate and boreal zone (Zaehle, 2013). This would have consequences for our modelled O_3 impact, particularly into the future where the large CO_2 fertilisation effect was responsible for partly offsetting the negative impact of O_3. Although in our simulations a high fraction of land cover is agricultural which we assume would be optimally fertilised. Our simulations also use a fixed climate, so we do not include the effect of climate change on shifting plant phenology. Therefore, our results may underestimate plant O_3 damage, since if the growing season started earlier or finished later, plants in some regions would be exposed to higher O_3 concentrations. Nevertheless, we emphasise that this study provides a sensitivity assessment of the impact of plant O_3 damage on GPP and the land carbon sink.

Another caveat we fully acknowledge is that at the leaf-level JULES is parameterised to reduce g_s with O_3 exposure. Whilst this response is commonly observed (Wittig et al., 2007;Ainsworth et al., 2012), there is evidence to suggest that O_3 impairs stomata in some species, making them non-responsive to environmental stimuli (Hayes et al., 2012;Hoshika et al., 2012a;Mills et al., 2009;Paoletti and Grulke, 2010). In drought conditions the mechanism is thought to involve O_3 stimulated ethylene production which interferes with the stomatal response to ABA signalling (Wilkinson and Davies, 2009;Wilkinson and Davies, 2010). Such stomatal sluggishness can result in higher O_3 uptake and injury, increased water-loss, and therefore greater vulnerability to environmental stresses (Mills et al., 2016). McLaughlin (2007a;2007b) and Sun et al. (2012) provide evidence of increased transpiration and reduced streamflow in forests at the regional scale in response to ambient levels of O_3, and suggest this could increase the frequency and severity of droughts. Hoshika et al. (2012b)Hoshika et al (2012) however found that despite sluggish stomatal control in O_3 exposed trees, whole tree water use was lower in these trees because of lower gas exchange and premature leaf shedding of injured leaves. To our knowledge, the study of Hoshika et al. (2015) is the first to include an explicit representation of sluggish stomatal control in a land-atmosphere model, they show that sluggish stomatal behaviour has implications for carbon and water cycling in ecosystems. However, it is by no means a ubiquitous response, and it is not fully understood which species respond this way and under what conditions (Mills et al., 2016;Wittig et al., 2007). Nevertheless, this remains an important area of future work.

In this work we implement the stomatal closure proposed in Medlyn et al., (2011), this uses the parameter g_1. Hoshika et al. (2013) show a significant difference in the g_1 parameter (higher in elevated O_3 compared to ambient) in Siebold's beech in June of their experiment. However, this is only at the start of the growing season, further measurements show no difference in this parameter between O_3 treatments. Quantifying an O_3 effect directly on g_1 would require a detailed meta-analysis of empirical data on photosynthesis and g_s for different PFTs, which is currently lacking in the literature. With such information lacking, here we take an empirical approach to modelling plant O_3 damage, essentially by applying a reduction factor to the simulated plant photosynthesis based on

Commented [ORJ41]: Line 394 to 397

Commented [ORJ42]: Line 711

observations of whole plant losses of biomass with accumulated O_3 exposure, for which there is a lot more available data (e.g. CLRTAP, 2017).

The calculation of O_3 deposition in the EMEP model uses the stomatal conductance formulation presented in Emberson et al. (2000;2001), which depends on temperature, light, humidity and soil moisture (commonly referred to as DO_3SE). Because we link two different model systems, the g_s values in the EMEP model differ from those obtained using the Medlyn formulation. We acknowledge this inconsistency as a caveat of our study, however comparison of *gmax* (maximum g_s) values from both models (EMEP (*gmax* is an input parameter determining the maximum g_s) and JULES (*gmax* is not used as an input parameter in JULES, instead we calculated *gmax* for each PFT taking the mean across the model domain for the year 2001) suggests the differences are small for deciduous forest (EMEP 150-200, JULES ~180, all units in mmole O_3/m^2 (PLA)/s), and C_3/C_4 crops (EMEP 270-300, JULES ~260-390~~ — the dominant land cover in our simulations~~), but are larger for coniferous forest (EMEP 140-200, JULES ~60-70) and shrubs (EMEP 60-200, JULES 360-390). ~~The role of EMEP in this study is not to provide g_s, however, but to provide O_3 at the top of the vegetation canopy. The main driver of such O_3 levels is the regional-scale production and transport of O_3, and the main impact of g_s is in affecting the vertical O_3 gradients just above the plant canopy. Differences in g_s are known to have minimal impact on canopy-top O_3 for trees, mainly due to the efficient turbulent mixing above tall canopies, but also due to non-stomatal sink processes. For shorter vegetation, substantial O_3 gradients, driven by deposition, occur in the lowest 10s of metres of the atmosphere, and stomatal sinks (as given by g_s) can have a significant role. However, calculations of such gradients made with the EMEP model for CLRTAP (2017) showed that differences amounted to only ca. 10% when comparing O_3 concentrations at 1m height above high-g_s crops compared to moderate-g_s (gmax = 450 and 270 mmole O_3/m^2 (PLA)/s respectively), therefore this uncertainty is small.~~ It should be noted that the role of EMEP in this study is not to provide g_s, but to provide O_3 at the top of the vegetation canopy. This firstly entails a calculation of the large-scale ozone concentrations for Europe, which are represented by the gridded values of grid-cell average concentration, and secondly to calculate the vertical gradients between these grid-cell centres (at ca. 45m) and the top of the vegetation canopy. O_3 deposition is important for both steps; it is known to have a substantial impact on the lifetime and concentrations of O_3 in the planetary boundary layer (Garland and Derwent, 1979;Val Martin et al., 2014)~~(Garland and Derwent, 1979;M. et al., 2014)~~, and also in determining the local vertical gradients above different land-covers (CLRTAP, 2017;Gerosa et al., 2017;Tuovinen et al., 2009). Vertical gradients between the 45m level and the top of forest canopies tend to be limited (Fuentes et al., 2007;Karlsson et al., 2006) due to the good mixing normally induced by forest roughness. Vertical gradients between 45m and the top of shorter vegetation such as grasslands or crops can be larger however (CLRTAP, 2017;Gerosa et al., 2017). Accounting for such land-cover specific gradient effects has been shown to have large impacts on estimates of O_3 metrics (Simpson et al., 2007).

These offline simulations show the sensitivity of GPP and the land carbon sink to tropospheric O_3, suggesting that O_3 is an important predictor of future GPP and the land carbon store across Europe. There are uncertainties in our estimates however from the use of uncoupled tropospheric chemistry, meteorology and stomatal function. For example, increased frequency of drought in the future would reduce stomatal conductance (assuming no sluggish

Commented [ORJ43]: Line 317 -324

Formatted: Justified, Line spacing: 1.5 lines

Field Code Changed

Commented [ORJ44]: Line 724

Formatted: Font: (Default) Times New Roman, 10 pt

Commented [ORJ45]: Line 728 to 730

Formatted: Font: (Default) Times New Roman, 10 pt

Field Code Changed

Formatted: Font: (Default) Times New Roman, 10 pt

Formatted: Font: (Default) Times New Roman, 10 pt, Italic

Formatted: Font: (Default) Times New Roman, 10 pt, Italic, Subscript

Formatted: Font: (Default) Times New Roman, 10 pt

Formatted: Font: (Default) Times New Roman, 10 pt, Subscript

Formatted: Font: (Default) Times New Roman, 10 pt

Formatted: Font: (Default) Times New Roman, 10 pt

Formatted: Font: (Default) Times New Roman, 10 pt

Formatted: Font: (Default) Times New Roman, 10 pt

Formatted: Font: (Default) Times New Roman, 10 pt

Formatted: Font: (Default) Times New Roman, 10 pt

Formatted: Font: (Default) Times New Roman, 10 pt

Formatted: Font: (Default) Times New Roman, 10 pt stomatal response) and thus O_3 uptake. Since our offline simulations do not include this feedback it is possible the O_3 effect is over estimated here. Given the complexity of potential interactions and feedbacks it remains difficult to diagnose the importance of individual factors (e.g. the direct physiological response) in a fully coupled simulation. Once the importance of a process is demonstrated offline, it provides evidence of the need to incorporate such process in coupled regional and global simulations.

4.4 O_3 as a missing component of carbon cycle assessments?

Comprehensive analyses of the European carbon balance suggest a large biogenic carbon sink (Janssens et al., 2003;Luyssaert et al., 2012;Schulze et al., 2009). However, estimates are hampered by large uncertainties in key components of the land carbon balance, such as estimates of soil carbon gains and losses (Ciais et al., 2010;Janssens et al., 2003;Schulze et al., 2009;Schulze et al., 2010). We suggest that the effect of O_3 on plant physiology is a contributing factor to the decline in soil carbon stores observed across Europe, and as such this O_3 effect is a missing component of European carbon cycle assessments. Over the full experimental period (1901 to 2050)~~Anthropocene~~, our results show elevated O_3 concentrations reduce the amount of carbon that can be stored in the soil by 3% to 9% (low and high plant O_3 sensitivity, respectively), which almost completely offsets the beneficial effects of CO_2 fertilisation on soil carbon storage under the high plant O_3 sensitivity . This would contribute to a change in the size of a key carbon sink for Europe, and is particularly important when we consider the evolution of the land carbon sink into the future given the impact of O_3 on soil carbon sequestration and the high uncertainty of future tropospheric O_3 concentrations. Schulze et al. (2009) and Luyssaert et al. (2012) extended their analysis of the European carbon balance to include additional non-CO_2 greenhouse gases (CH_4 and N_2O). Both studies found that emissions of these offset the biogenic carbon sink, reducing the climate mitigation potential of European ecosystems. This highlights the importance of accounting for all fluxes and stores in carbon/greenhouse gas balance assessments, of which O_3 and its indirect effect on the CO_2 flux via direct effects on plant physiology is currently missing.

4.5 The interaction between O_3 and CO_2

We looked at the interaction between CO_2 and O_3 effects. Our results support the hypothesis that elevated atmospheric CO_2 provides some protection against O_3 damage because of lower g_s that reduces uptake of O_3 through stomata (Harmens et al., 2007;Wittig et al., 2007). In the present study, reductions in GPP and the land carbon store due to O_3 exposure were lower when simulated with concurrent changes in atmospheric CO_2. Despite acclimation of photosynthesis after long-term exposure to elevated atmospheric CO_2 of field grown plants (Ainsworth and Long, 2005;Medlyn et al., 1999), there is no evidence to suggest that g_s acclimates (Ainsworth et al., 2003;Medlyn et al., 2001). This suggests the protective effect of elevated atmospheric CO_2 against O_3 damage will be sustained in the long term. However, although meta-analysis suggest a general trend of reduced g_s with elevated CO_2 (Ainsworth and Long, 2005;Medlyn et al., 1999), this is not a universal response. Stomatal responses on exposure to elevated CO_2 with FACE treatment varied with genotype and growth stage in a fast-growing poplar community (Bernacchi et al., 2003;Tricker et al., 2009). In other mature forest stands, limited stomatal response to elevated CO_2 was observed after canopy closure (Ellsworth, 1999;Uddling et al., 2009). Also, some studies found that stomatal responses to CO_2 were significant only under high atmospheric humidity (Cech et al., 2003;Leuzinger and Körner, 2007;Wullschleger et al., 2002). These examples illustrate that stomatal responses to elevated atmospheric CO_2 are not universal, and as such the protective effect of CO_2 against O_3 injury cannot be assumed for all species, at all growth stages under wide ranging environmental conditions.

5 Conclusion

What is abundantly clear is that plant responses to both CO_2 and O_3 are complicated by a host of factors that are only partly understood, and it remains difficult to identify general, global patterns given that effects of both gases on plant communities and ecological interactions are highly context and species specific (Ainsworth and Long, 2005;Fuhrer et al., 2016;Matyssek et al., 2010b). This study quantifies the sensitivity of the land carbon sink for Europe and GPP to changing concentrations of atmospheric CO_2 and O_3 from 1901 to 2050. We have used a state of the art land surface model calibrated for European vegetation to give our best estimates of this sensitivity within the limits of data availability to calibrate the model for O_3 sensitivity, current knowledge and model structure. In summary, this study has shown that potential gains in terrestrial carbon sequestration over Europe resulting from elevated CO_2 can be partially offset by concurrent rises in tropospheric O_3 over 1901-2050. Specifically, we have shown that the negative effect of O_3 on the land carbon sink was greatest over the twentieth century, when O_3 concentrations increased rapidly from pre-industrial levels. Over this period soil carbon stocks were diminished over agricultural areas, consistent with reduced NPP and litter input. This loss of soil carbon was largely responsible for the decrease in the size of the land carbon sink over Europe. The O_3 effect on the land carbon store and flux was reduced into the future as CO_2 concentration rose considerably and changes in O_3 concentration were less pronounced. However, there remained a large cumulative negative impact on the land carbon sink for Europe by 2050. The interaction between the two gases was found to reduce O_3 injury owing to reduced stomatal opening in elevated atmospheric CO_2. However, primary productivity and land carbon storage remained suppressed by 2050 due to plant O_3 damage. Expressed as a percentage of the emissions from fossil fuel and cement production for the EU28-plus countries, the carbon emissions from O_3-induced plant injury are a source of anthropogenic carbon previously not accounted for in carbon cycle assessments. Our results demonstrate the sensitivity of modelled terrestrial carbon dynamics to the direct effect of tropospheric O_3 and its interaction with atmospheric CO_2 on plant physiology, demonstrating this process is an important predictor of future GPP and trends in the land-carbon sink. Nevertheless, this process remains largely unconsidered in regional and global climate model simulations that are used to model carbon sources and sinks and carbon-climate feedbacks.

Data availability

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

Supplementary Information

Supplementary_Information_Oliver_et_al.docx

Competing Interests

The authors declare that they have no conflict of interest

Acknowledgements

RJO and LMM were supported by the EU FP7 (ECLAIRE, 282910) and JWCRP (UKESM, NEC05816). This work was also supported by EMEP under UNECE. SS and LMM acknowledge the support of the NERC SAMBBA project (NE/J010057/1). The UK Met Office contribution was funded by BEIS under the Hadley Centre Climate Programme (GA01101). GAF also acknowledges funding from the EU's Horizon 2020 research and innovation programme (CRESCENDO, 641816). We also thank Magnuz Engardt of SMHI for providing the RCA3 climate dataset. This work used eddy covariance data acquired and shared by the FLUXNET community, including these networks: AmeriFlux, AfriFlux, AsiaFlux, CarboAfrica, CarboEuropeIP, CarboItaly, CarboMont, ChinaFlux, Fluxnet-Canada, GreenGrass, ICOS, KoFlux, LBA, NECC, OzFlux-TERN, TCOS-Siberia, and USCCC. The ERA-Interim reanalysis data are provided by ECMWF and processed by LSCE. The FLUXNET eddy covariance data processing and harmonization was carried out by the European Fluxes Database Cluster, AmeriFlux Management Project, and Fluxdata project of FLUXNET, with the support of CDIAC and ICOS Ecosystem Thematic Center, and the OzFlux, ChinaFlux and AsiaFlux offices.

References

Ainsworth, E., and Long, S.: What have we learned from 15 years of free-air CO_2 enrichment (FACE)? A meta-analytic review of the responses of photosynthesis, canopy properties and plant production to rising CO2, New Phytologist, 165, 351-372, 2005.

Ainsworth, E. A., Davey, P. A., Hymus, G. J., Osborne, C. P., Rogers, A., Blum, H., Nosberger, J., and Long, S. P.: Is stimulation of leaf photosynthesis by elevated carbon dioxide concentration maintained in the long term? A test with *Lolium perenne* grown for 10 years at two nitrogen fertilization levels under Free Air CO_2 Enrichment (FACE), Plant, Cell and Environment, 26, 705-714, 2003.

Ainsworth, E. A.: Rice production in a changing climate: a meta-analysis of responses to elevated carbon dioxide and elevated ozone concentration, Global Change Biology, 14, 1642-1650, 10.1111/j.1365-2486.2008.01594.x, 2008.

Ainsworth, E. A., Yendrek, C. R., Sitch, S., Collins, W. J., and Emberson, L. D.: The Effects of Tropospheric Ozone on Net Primary Productivity and Implications for Climate Change, Annual Review of Plant Biology, 63, 637-661, doi:10.1146/annurev-arplant-042110-103829, 2012.

Anav, A., Menut, L., Khvorostyanov, D., and Viovy, N.: Impact of tropospheric ozone on the Euro-Mediterranean vegetation, Global change biology, 17, 2342-2359, 2011.

Andersen, C. P.: Source–sink balance and carbon allocation below ground in plants exposed to ozone, New Phytologist, 157, 213-228, 10.1046/j.1469-8137.2003.00674.x, 2003.

Arneth, A., Harrison, S. P., Zaehle, S., Tsigaridis, K., Menon, S., Bartlein, P. J., Feichter, J., Korhola, A., Kulmala, M., O'Donnell, D., Schurgers, G., Sorvari, S., and Vesala, T.: Terrestrial biogeochemical feedbacks in the climate system, Nature Geosci, 3, 525-532, http://www.nature.com/ngeo/journal/v3/n8/suppinfo/ngeo905_S1.html, 2010.

Avnery, S., Mauzerall, D. L., Liu, J., and Horowitz, L. W.: Global crop yield reductions due to surface ozone exposure: 1. Year 2000 crop production losses and economic damage, Atmospheric Environment, 45, 2284-2296, https://doi.org/10.1016/j.atmosenv.2010.11.045, 2011.

Baig, S., Medlyn, B. E., Mercado, L. M., and Zaehle, S.: Does the growth response of woody plants to elevated CO2 increase with temperature? A model-oriented meta-analysis, Global Change Biology, 21, 4303-4319, 10.1111/gcb.12962, 2015.

Bellamy, P. H., Loveland, P. J., Bradley, R. I., Lark, R. M., and Kirk, G. J.: Carbon losses from all soils across England and Wales 1978–2003, Nature, 437, 245-248, 2005.

Bernacchi, C. J., Calfapietra, C., Davey, P. A., Wittig, V. E., Scarascia-Mugnozza, G. E., Raines, C. A., and Long, S. P.: Photosynthesis and stomatal conductance responses of poplars to free-air CO_2 enrichment (PopFACE) during the first growth cycle and immediately following coppice., New Phytologist, 159, 609-621, 2003.

Best, M. J., Pryor, M., Clark, D. B., Rooney, G. G., Essery, R. L. H., Menard, C. B., Edwards, J. M., Hendry, M. A., Porson, N., Gedney, N., Mercado, L. M., Sitch, S., Blyth, E., Boucher, O., Cox, P. M., Grimmond, C. S. B., and Harding, R. J.: The Joint UK Land Environment Simulator (JULES), Model description - Part 1: Energy and water fluxes, Geoscientific Model Development Discussions, 4, 595-640, 10.5194/GMDD-4-595-2011, 2011.

Betts, R. A., Boucher, O., Collins, M., Cox, P. M., Falloon, P. D., Gedney, N., Hemming, D. L., Huntingford, C., Jones, C. D., and Sexton, D. M.: Projected increase in continental runoff due to plant responses to increasing carbon dioxide, Nature, 448, 1037-1041, 2007.

Boden, T. A., Marland, G., and Andres, R. J.: Global, Regional, and National Fossil-Fuel CO2 Emissions, Oak Ridge National Laboratory, U.S. Department of Energy, Oak Ridge, Tenn., USA, 2013.

Büker, P., Feng, Z., Uddling, J., Briolat, A., Alonso, R., Braun, S., Elvira, S., Gerosa, G., Karlsson, P. E., Le Thiec, D., Marzuoli, R., Mills, G., Oksanen, E., Wieser, G., Wilkinson, M., and Emberson, L. D.: New flux based dose-response relationships for ozone for European forest tree species, Environmental Pollution, 163-174, 2015.

Calvete-Sogo, H., Elvira, S., Sanz, J., González-Fernández, I., García-Gómez, H., Sánchez-Martín, L., Alonso, R., and Bermejo-Bermejo, V.: Current ozone levels threaten gross primary production and yield of Mediterranean annual pastures and nitrogen modulates the response, Atmospheric Environment, 95, 197-206, http://dx.doi.org/10.1016/j.atmosenv.2014.05.073, 2014.

Capriel, P.: Trends in organic carbon and nitrogen contents in agricultural soils in Bavaria (south Germany) between 1986 and 2007, European Journal of Soil Science, 64, 445-454, 2013.

Cech, P. G., Pepin, S., and Korner, C.: Elevated CO_2 reduces sap flux in mature deciduous forest trees, Oecologia, 137, 258-268, 2003.

Ceulemans, R., and Mousseau, M.: Effects of elevated atmospheric CO_2 on woody plants, New Phytologist, 127, 1994.

Ciais, P., Wattenbach, M., Vuichard, N., Smith, P., Piao, S., Don, A., Luyssaert, S., Janssens, I., Bondeau, A., and Dechow, R.: The European carbon balance. Part 2: croplands, Global Change Biology, 16, 1409-1428, 2010.

Ciais, P., Sabine, C., Bala, G., Bopp, L., Brovkin, V., Canadell, J., Chhabra, A., DeFries, R., Galloway, J., Heimann, M., Jones, C., Le Quéré, C., Myneni, R. B., Piao, S., and Thornton, P.: Carbon and Other Biogeochemical Cycles. In: Climate Change 2013: The Physical Science Basis. Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change [Stocker, T.F., D. Qin, G.-K. Plattner, M. Tignor, S.K. Allen, J. Boschung, A. Nauels, Y. Xia, V. Bex and P.M.

Midgley (eds.)]. Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA., 2013.

Clark, D. B., Mercado, L. M., Sitch, S., Jones, C. D., Gedney, N., Best, M. J., Pryor, M., Rooney, G. G., Essery, R. L. H., Blyth, E., Boucher, O., Harding, R. J., and Cox, P. M.: The Joint UK Land Environment Simulator (JULES), Model description - Part 2: Carbon fluxes and vegetation, Geoscientific Model Development Discussions, 4, 641-688, 10.5194/gmdd-4-641-2011, 2011.

CLRTAP: The UNECE Convention on Long-range Transboundary Air Pollution. Manual on Methodologies and Criteria for Modelling and Mapping Critical Loads and Levels and Air Pollution Effects, Risks and Trends: Chapter III Mapping Critical Levels for Vegetation, accessed via, http://icpvegetation.ceh.ac.uk/publications/documents/Chapter3-Mappingcriticallevelsforvegetation_000.pdf, 2017.

Collins, W. J., Sitch, S., and Boucher, O.: How vegetation impacts affect climate metrics for ozone precursors, Journal of Geophysical Research: Atmospheres, 115, D23308, 10.1029/2010JD014187, 2010.

Collins, W. J., Bellouin, N., Doutriaux-Boucher, M., Gedney, N., Halloran, P., Hinton, T., Hughes, J., Jones, C. D., Joshi, M., Liddicoat, S., Martin, G., O'Connor, F., Rae, J., Senior, C., Sitch, S., Totterdell, I., Wiltshire, A., and Woodward, S.: Development and evaluation of an Earth-System model – HadGEM2, Geosci. Model Dev., 4, 1051-1075, 10.5194/gmd-4-1051-2011, 2011.

Cooper, O. R., Parrish, D. D., Stohl, A., Trainer, M., Nedelec, P., Thouret, V., Cammas, J. P., Oltmans, S. J., Johnson, B. J., Tarasick, D., Leblanc, T., McDermid, I. S., Jaffe, D., Gao, R., Stith, J., Ryerson, T., Aikin, K., Campos, T., Weinheimer, A., and Avery, M. A.: Increasing springtime ozone mixing ratios in the free troposphere over western North America, Nature, 463, 344-348, http://www.nature.com/nature/journal/v463/n7279/suppinfo/nature08708_S1.html, 2010.

Cooper, O. R., Parrish, D., Ziemke, J., Balashov, N., Cupeiro, M., Galbally, I., Gilge, S., Horowitz, L., Jensen, N., and Lamarque, J.-F.: Global distribution and trends of tropospheric ozone: An observation-based review, Elementa: Science of the Anthropocene, 2, 000029, 2014.

Cox, P. M., Betts, R. A., Jones, C. D., Spall, S. A., and Totterdell, I. J.: Acceleration of global warming due to carbon-cycle feedbacks in a coupled climate model, Nature, 408, 184-187, 2000.

Cox, P. M.: Description of the TRIFFID dynamic global vegetation model, Hadley Centre technical note 24, 2001.

Cruz, F. T., Pitman, A. J., and Wang, Y. P.: Can the stomatal response to higher atmospheric carbon dioxide explain the unusual temperatures during the 2002 Murray-Darling Basin drought?, Journal of Geophysical Research: Atmospheres, 115, 2010.

de Arellano, J. V.-G., van Heerwaarden, C. C., and Lelieveld, J.: Modelled suppression of boundary-layer clouds by plants in a CO2-rich atmosphere, Nature geoscience, 5, 701-704, 2012.

De Kauwe, M., Kala, J., Lin, Y.-S., Pitman, A., Medlyn, B., Duursma, R., Abramowitz, G., Wang, Y.-P., and Miralles, D.: A test of an optimal stomatal conductance scheme within the CABLE land surface model, 8, 431-452, 2015.

Derwent, R. G., Stevenson, D. S., Doherty, R. M., Collins, W. J., Sanderson, M. G., and Johnson, C. E.: Radiative forcing from surface NO x emissions: spatial and seasonal variations, Climatic Change, 88, 385-401, 10.1007/s10584-007-9383-8, 2008.

Ellsworth, D. S.: CO_2 enrichment in a maturing pine forest: are CO_2 exchange and water status in the canopy affected?, Plant, Cell and Environment, 22, 461-472, 1999.

Emberson, L. D., Ashmore, M. R., Cambridge, H. M., Simpson, D., and Tuovinen, J.-P.: Modelling stomatal ozone flux across Europe, Environmental Pollution, 109, 403–413, 2000.

Emberson, L. D., Simpson, D., Tuovinen, J.-P., Ashmore, M. R., and Cambridge, H. M.: Modelling and mapping ozone deposition in Europe, Water Air Soil Pollution, 130, 577–582, 2001.

Emberson, L. D., Büker, P., and Ashmore, M. R.: Assessing the risk caused by ground level ozone to European forest trees: A case study in pine, beech and oak across different climate regions, Environmental Pollution, 147, 454–466, 2007.

Engardt, M., Simpson, D., Schwikowski, M., and Granat, L.: Deposition of sulphur and nitrogen in Europe 1900-2050. Model calculations and comparison to historical observations, *Tellus B: Chem. Phys. Meteor.*, 69, 2017.

Etheridge, D. M., Steele, L. P., Langenfelds, R. L., Francey, R. J., M., B., and Morgan, V. I.: Natural and anthropogenic changes in atmospheric CO2 over the last 1000 years from air in Antarctic ice and firn, Journal of Geophysical Research, 101(D2), 4115–4128, doi:10.1029/95JD03410, 1996.

Fagnano, M., Maggio, A., and Fumagalli, I.: Crops' responses to ozone in Mediterranean environments, Environmental Pollution, 157, 1438-1444, 2009.

Fares, S., Vargas, R., Detto, M., Goldstein, A. H., Karlik, J., Paoletti, E., and Vitale, M.: Tropospheric ozone reduces carbon assimilation in trees: estimates from analysis of continuous flux measurements, Global change biology, 19, 2427-2443, 2013.

Felzer, B., Reilly, J., Melillo, J., Kicklighter, D., Sarofim, M., Wang, C., Prinn, R., and Zhuang, Q.: Future Effects of Ozone on Carbon Sequestration and Climate Change Policy Using a Global Biogeochemical Model, Climatic Change, 73, 345-373, 10.1007/s10584-005-6776-4, 2005.

Felzer, B. S. F., Kicklighter, D. W., Melillo, J. M., Wang, C., Zhuang, Q., and Prinn, R. G.: Ozone effects on net primary productivity and carbon sequestration in the conterminous United States using a biogeochemistry model, Tellus, 56B, 230-248, 2004.

Feng, Z., Kobayashi, K., and Ainsworth, E. A.: Impact of elevated ozone concentration on growth, physiology, and yield of wheat (Triticum aestivum L.): a meta-analysis, Global Change Biology, 14, 2696-2708, 10.1111/j.1365-2486.2008.01673.x, 2008.

Fowler, D., Flechard, C., Cape, J. N., Storeton-West, R. L., and Coyle, M.: Measurements of Ozone Deposition to Vegetation Quantifying the Flux, the Stomatal and Non-Stomatal Components, Water, Air, and Soil Pollution, 130, 63-74, 10.1023/a:1012243317471, 2001.

Fowler, D., Pilegaard, K., Sutton, M., Ambus, P., Raivonen, M., Duyzer, J., Simpson, D., Fagerli, H., Fuzzi, S., and Schjørring, J. K.: Atmospheric composition change: ecosystems–atmosphere interactions, Atmospheric Environment, 43, 5193-5267, 2009.

Franz, M., Simpson, D., Arneth, A., and Zaehle, S.: Development and evaluation of an ozone deposition scheme for coupling to a terrestrial biosphere model, Biogeosciences, 14, 45-71, doi:10.5194/bg-14-45-2017, 2017.

Friedlingstein, P., Cox, P., Betts, R., Bopp, L., von Bloh, W., Brovkin, V., Cadule, P., Doney, S., Eby, M., Fung, I., Bala, G., John, J., Jones, C., Joos, F., Kato, T., Kawamiya, M., Knorr, W., Lindsay, K., Matthews, H. D., Raddatz, T., Rayner, P., Reick, C., Roeckner, E., Schnitzler, K. G., Schnur, R., Strassmann, K., Weaver, A. J., Yoshikawa, C., and Zeng, N.: Climate–Carbon Cycle Feedback Analysis: Results from the C4MIP Model Intercomparison, Journal of Climate, 19, 3337-3353, 10.1175/jcli3800.1, 2006.

Fuentes, J. D., Wang, D., Bowling, D. R., Potosnak, M., Monson, R. K., Goliff, W. S., and Stockwell, W. R.: Biogenic hydrocarbon chemistry within and above a mixed deciduous forest, Atmopsheric Chemistry, 56, 165-185, 2007.

Fuhrer, J., Val Martin, M., Mills, G., Heald, C. L., Harmens, H., Hayes, F., Sharps, K., Bender, J., and Ashmore, M. R.: Current and future ozone risks to global terrestrial biodiversity and ecosystem processes, Ecology and Evolution, 6, 8785-8799, 10.1002/ece3.2568, 2016.

Garland, J. A., and Derwent, R. G.: Destruction at the ground and the diurnal cycle of concentration of ozone and other gases, Quarterly Journal of the Royal Meteorological Society, 105, 169-183, doi:10.1002/qj.49710544311, 1979.

Gedney, N., Cox, P. M., Bett, R. A., Boucher, O., Huntingford, C., and Stott, P. A.: Detection of a direct carbon dioxide effect in continental river runoff records, Nature, 439, 835-838, 2006.

Gerosa, G., Marzuoli, R., Monteleone, B., Chiesa, M., and Finco, A.: Vertical Ozone Gradients above Forests. Comparison of Different Calculation Options with Direct Ozone Measurements above a Mature Forest and Consequences for Ozone Risk Assessment, Forests, 8, 337, 2017.

Grantz, D., Gunn, S., and VU, H. B.: O3 impacts on plant development: a meta-analysis of root/shoot allocation and growth, Plant, cell & environment, 29, 1193-1209, 2006.

Harmens, H., Mills, G., Emberson, L. D., and Ashmore, M. R.: Implications of climate change for the stomatal flux of ozone: A case study for winter wheat, Environmental Pollution, 146, 763-770, http://dx.doi.org/10.1016/j.envpol.2006.05.018, 2007.

Hayes, F., Wagg, S., Mills, G., Wilkinson, S., and Davies, W.: Ozone effects in a drier climate: implications for stomatal fluxes of reduced stomatal sensitivity to soil drying in a typical grassland species, Global Change Biology, 18, 948-959, 2012.

Heikkinen, J., Ketoja, E., Nuutinen, V., and Regina, K.: Declining trend of carbon in Finnish cropland soils in 1974–2009, Global Change Biology, 19, 1456-1469, 10.1111/gcb.12137, 2013.

Hofmockel, K. S., Zak, D. R., Moran, K. K., and Jastrow, J. D.: Changes in forest soil organic matter pools after a decade of elevated CO2 and O3, Soil Biology and Biochemistry, 43, 1518-1527, http://dx.doi.org/10.1016/j.soilbio.2011.03.030, 2011.

Hoshika, Y., Watanabe, M., Inada, N., and Koike, T.: Ozone-induced stomatal sluggishness develops progressively in Siebold's beech (Fagus crenata), Environmental Pollution, 166, 152-156, 2012a.

Hoshika, Y., Omasa, K., and Paoletti, E.: Whole-Tree Water Use Efficiency Is Decreased by Ambient Ozone and Not Affected by O3-Induced Stomatal Sluggishness, PLOS ONE, 7, e39270, 10.1371/journal.pone.0039270, 2012b.

Hoshika, Y., Watanabe, M., Inada, N., and Koike, T.: Model-based analysis of avoidance of ozone stress by stomatal closure in Siebold's beech (*Fagus crenata*), Annals of Botany, 112, 1149-1158, 2013.

Hoshika, Y., Katata, G., Deushi, M., Watanabe, M., Koike, T., and Paoletti, E.: Ozone-induced stomatal sluggishness changes carbon and water balance of temperate deciduous forests., Scientific Reports, doi:10.1038/srep09871, 2015.

Hurtt, G., Chini, L. P., Frolking, S., Betts, R., Feddema, J., Fischer, G., Fisk, J., Hibbard, K., Houghton, R., Janetos, A., and Jones, C. D.: Harmonization of land-use scenarios for the period 1500–2100: 600 years of global gridded annual land-use transitions, wood harvest, and resulting secondary lands, Climatic Change, 109, 117-161, 2011.

IGBP-DIS: International Geosphere-Biosphere Programme, Data and Information System, Potsdam, Germany. Available from Oak Ridge National Laboratory Distributed Active Archive Center, Oak Ridge, TN, available at: http://www.daac.ornl.gov,

IPCC: Climate change 2013: The Physical Science Basis, IPCC Working Group I Contribution to AR5, 2013.

Jacobs, C. M. J.: Direct impact of atmospheric CO_2 enrichment on regional transpiration, Wageningen Agricultural University, 1994.

Janssens, I. A., Freibauer, A., Ciais, P., Smith, P., Nabuurs, G.-J., Folberth, G., Schlamadinger, B., Hutjes, R. W. A., Ceulemans, R., Schulze, E.-D., Valentini, R., and Dolman, A. J.: Europe's Terrestrial Biosphere Absorbs 7 to 12% of European Anthropogenic CO2 Emissions, Science, 300, 1538-1542, 10.1126/science.1083592, 2003.

Jones, C. D., Cox, P., and Huntingford, C.: Uncertainty in climate–carbon-cycle projections associated with the sensitivity of soil respiration to temperature, Tellus B, 55, 642-648, 10.1034/j.1600-0889.2003.01440.x, 2003.

Jung, M., Reichstein, M., Margolis, H. A., Cescatti, A., Richardson, A. D., Arain, M. A., Arneth, A., Bernhofer, C., Bonal, D., Chen, J., Gianelle, D., Gobron, N., Kiely, G., Kutsch, W., Lasslop, G., Law, B. E., Lindroth, A., Merbold, L., Montagnani, L., Moors, E. J., Papale, D., Sottocornola, M., Vaccari, F., and Williams, C.: Global patterns of land-atmosphere fluxes of carbon dioxide, latent heat, and sensible heat derived from eddy covariance, satellite, and meteorological observations, Journal of Geophysical Research: Biogeosciences, 116, n/a-n/a, 10.1029/2010JG001566, 2011.

Kala, J., De Kauwe, M. G., Pitman, A. J., Medlyn, B. E., Wang, Y. P., Lorenz, R., and Perkins-Kirkpatrick, S. E.: Impact of the representation of stomatal conductance on model projections of heatwave intensity., Scientific Reports, 1-7, 10.1038/srep23418, 2016.

Karlsson, P., Hansson, M., Höglund, H. O., and Pleijel, H.: Ozone concentration gradients and wind conditions in Norway spruce (Picea abies) forests in Sweden, Atmospheric Environment, 1610-1618 pp., 2006.

Karlsson, P. E., Braun, S., Broadmeadow, M., Elvira, S., Emberson, L., Gimeno, B. S., Le Thiec, D., Novak, K., Oksanen, E., Schaub, M., Uddling, J., and Wilkinson, M.: Risk assessments for forest trees: The performance of the ozone flux versus the AOT concepts, Environmental Pollution, 146, 608-616, http://dx.doi.org/10.1016/j.envpol.2006.06.012, 2007.

Karnosky, D., Percy, K. E., Xiang, B., Callan, B., Noormets, A., Mankovska, B., Hopkin, A., Sober, J., Jones, W., and Dickson, R.: Interacting elevated CO2 and tropospheric O3 predisposes aspen (Populus tremuloides Michx.) to infection by rust (Melampsora medusae f. sp. tremuloidae), Global Change Biology, 8, 329-338, 2002.

Karnosky, D. F., Skelly, J. M., Percy, K. E., and Chappelka, A. H.: Perspectives regarding 50years of research on effects of tropospheric ozone air pollution on US forests, Environmental Pollution, 147, 489-506, 2007.

Keeling, C. D., and Whorf, T. P.: Atmospheric CO2 records from sites in the SIO air sampling network. In Trends: A Compendium of Data on Global Change, Carbon Dioxide Information Analysis Center, Oak Ridge National Laboratory, Oak Ridge, Tenn., U.S.A. , 2004.

Kitao, M., Löw, M., Heerdt, C., Grams, T. E., Häberle, K.-H., and Matyssek, R.: Effects of chronic elevated ozone exposure on gas exchange responses of adult beech trees (Fagus sylvatica) as related to the within-canopy light gradient, Environmental Pollution, 157, 537-544, 2009.

Kjellström, E., Nikulin, G., Hansson, U., Strandberg, G., and Ullerstig, A.: 21st century changes in the European climate: uncertainties derived from an ensemble of regional climate model simulations, Tellus A, 63, 24-40, 2011.

Kubiske, M., Quinn, V., Marquardt, P., and Karnosky, D.: Effects of Elevated Atmospheric CO2 and/or O3 on Intra-and Interspecific Competitive Ability of Aspen, Plant biology, 9, 342-355, 2007.

Lamarque, J., Shindell, D. T., Josse, B., Young, P., Cionni, I., Eyring, V., Bergmann, D., Cameron-Smith, P., Collins, W. J., and Doherty, R.: The Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP): overview and description of models, simulations and climate diagnostics, Geoscientific Model Development, 6, 179-206, 2013.

Langner, J., Engardt, M., Baklanov, A., Christensen, J. H., Gauss, M., Geels, C., Hedegaard, G. B., Nuterman, R., Simpson, D., and Soares, J.: A multi-model study of impacts of climate change on surface ozone in Europe, Atmospheric Chemistry and Physics, 12, 10423-10440, 2012a.

Langner, J., Engardt, M., and Andersson, C.: European summer surface ozone 1990–2100, Atmospheric Chemistry and Physics, 12, 10097-10105, 2012b.

Le Quéré, C., Moriarty, R., Andrew, R. M., Peters, G. P., Ciais, P., Friedlingstein, P., Jones, S. D., Sitch, S., Tans, P., Arneth, A., Boden, T. A., Bopp, L., Bozec, Y., Canadell, J. G., Chini, L. P., Chevallier, F., Cosca, C. E., Harris, I., Hoppema, M., Houghton, R. A., House, J. I., Jain, A. K., Johannessen, T., Kato, E., Keeling, R. F., Kitidis, V., Klein Goldewijk, K., Koven, C., Landa, C. S., Landschützer, P., Lenton, A., Lima, I. D., Marland, G., Mathis, J. T., Metzl, N., Nojiri, Y., Olsen, A., Ono, T., Peng, S., Peters, W., Pfeil, B., Poulter, B., Raupach, M. R., Regnier, P., Rödenbeck, C., Saito, S., Salisbury, J. E., Schuster, U., Schwinger, J., Séférian, R., Segschneider, J., Steinhoff, T., Stocker, B. D., Sutton, A. J., Takahashi, T., Tilbrook, B., van der Werf, G. R., Viovy, N., Wang, Y. P., Wanninkhof, R., Wiltshire, A., and Zeng, N.: Global carbon budget 2014, Earth Syst. Sci. Data, 7, 47-85, 10.5194/essd-7-47-2015, 2015.

Le Quéré, C., Andrew, R. M., Canadell, J. G., Sitch, S., Korsbakken, J. I., Peters, G. P., Manning, A. C., Boden, T. A., Tans, P. P., Houghton, R. A., Keeling, R. F., Alin, S., Andrews, O. D., Anthoni, P., Barbero, L., Bopp, L., Chevallier, F., Chini, L. P., Ciais, P., Currie, K., Delire, C., Doney, S. C., Friedlingstein, P., Gkritzalis, T., Harris, I., Hauck, J., Haverd, V., Hoppema, M., Klein Goldewijk, K., Jain, A. K., Kato, E., Körtzinger, A., Landschützer, P., Lefèvre, N., Lenton, A., Lienert, S., Lombardozzi, D., Melton, J. R., Metzl, N., Millero, F., Monteiro, P. M. S., Munro, D. R., Nabel, J. E. M. S., Nakaoka, S. I., O'Brien, K., Olsen, A., Omar, A. M., Ono, T., Pierrot, D., Poulter, B., Rödenbeck, C., Salisbury, J., Schuster, U., Schwinger, J., Séférian, R., Skjelvan, I., Stocker, B. D., Sutton, A. J., Takahashi, T., Tian, H., Tilbrook, B., van der Laan-Luijkx, I. T., van der Werf, G. R., Viovy, N., Walker, A. P., Wiltshire, A. J., and Zaehle, S.: Global Carbon Budget 2016, Earth Syst. Sci. Data, 8, 605-649, 10.5194/essd-8-605-2016, 2016.

Le Quéré, C., Andrew, R. M., Friedlingstein, P., Sitch, S., Pongratz, J., Manning, A. C., Korsbakken, J. I., Peters, G. P., Canadell, J. G., Jackson, R. B., Boden, T. A., Tans, P. P., Andrews, O. D., Arora, V. K., Bakker, D. C. E., Barbero, L., Becker, M., Betts, R. A., Bopp, L., Chevallier, F., Chini, L. P., Ciais, P., Cosca, C. E., Cross, J., Currie, K., Gasser, T., Harris, I., Hauck, J., Haverd, V., Houghton, R. A., Hunt, C. W., Hurtt, G., Ilyina, T., Jain, A. K., Kato, E., Kautz, M., Keeling, R. F., Klein Goldewijk, K., Körtzinger, A., Landschützer, P., Lefèvre, N., Lenton, A., Lienert, S., Lima, I., Lombardozzi, D., Metzl, N., Millero, F., Monteiro, P. M. S., Munro, D. R., Nabel, J. E. M. S., Nakaoka, S.-I., Nojiri, Y., Padín, X. A., Peregon, A., Pfeil, B., Pierrot, D., Poulter, B., Rehder, G., Reimer, J., Rödenbeck, C., Schwinger, J., Séférian, R., Skjelvan, I., Stocker, B. D., Tian, H., Tilbrook, B., van der Laan-Luijkx, I. T., van der Werf, G. R., van Heuven, S., Viovy, N., Vuichard, N., Walker, A. P., Watson, A. J., Wiltshire, A. J., Zaehle, S., and Zhu, D.: Global Carbon Budget 2017, Earth Syst. Sci. Data Discuss, in review, 2017.

Leuzinger, S., and Körner, C.: Water savings in mature deciduous forest trees under elevated CO_2, Global Change Biology, 13, 2498-2508, doi:10.1111/j.1365-2486.2007.01467.x, 2007.

Lin, Y.-S., Medlyn, B. E., Duursma, R. A., Prentice, I. C., Wang, H., Baig, S., Eamus, D., de Dios, V. R., Mitchell, P., and Ellsworth, D. S.: Optimal stomatal behaviour around the world, Nature Climate Change, 5, 459-464, 2015.

Lindroth, R. L.: Impacts of Elevated Atmospheric CO2 and O3 on Forests: Phytochemistry, Trophic Interactions, and Ecosystem Dynamics, Journal of Chemical Ecology, 36, 2-21, 10.1007/s10886-009-9731-4, 2010.

Logan, J. A., Staehelin, J., Megretskaia, I. A., Cammas, J. P., Thouret, V., Claude, H., De Backer, H., Steinbacher, M., Scheel, H. E., Stübi, R., Fröhlich, M., and Derwent, R.: Changes in ozone over Europe: Analysis of ozone measurements from sondes, regular aircraft (MOZAIC) and alpine surface sites, Journal of Geophysical Research, 117, 1-23, 2012.

Lombardozzi, D., Levis, S., Bonan, G., and Sparks, J. P.: Predicting photosynthesis and transpiration responses to ozone: decoupling modeled photosynthesis and stomatal conductance, Biogeosciences, 3113-3130, 2012.

Lombardozzi, D., Levis, S., Bonan, G., Hess, P. G., and Sparks, J. P.: The Influence of Chronic Ozone Exposure on Global Carbon and Water Cycles, Journal of Climate, 28, 292-305, 10.1175/jcli-d-14-00223.1, 2015.

Long, S. P., Ainsworth, E. A., Leakey, A. D. B., Nosberger, J., and Ort, D. R.: Food for Thought: Lower-Than-Expected Crop Yield Stimulation with Rising CO_2 Concentrations, Science, 312, 1918-1921, 10.1126/science.1114722, 2006.

Löw, M., Herbinger, K., Nunn, A., Häberle, K.-H., Leuchner, M., Heerdt, C., Werner, H., Wipfler, P., Pretzsch, H., and Tausz, M.: Extraordinary drought of 2003 overrules ozone impact on adult beech trees (Fagus sylvatica), Trees, 20, 539-548, 2006.

Loya, W. M., Pregitzer, K. S., Karberg, N. J., King, J. S., and Giardina, C. P.: Reduction of soil carbon formation by tropospheric ozone under increased carbon dioxide levels., Nature, 425, 705-707, 2003.

Luyssaert, S., Abril, G., Andres, R., Bastviken, D., Bellassen, V., Bergamaschi, P., Bousquet, P., Chevallier, F., Ciais, P., Corazza, M., Dechow, R., Erb, K. H., Etiope, G., Fortems-Cheiney, A., Grassi, G., Hartmann, J., Jung, M., Lathière, J., Lohila, A., Mayorga, E., Moosdorf, N., Njakou, D. S., Otto, J., Papale, D., Peters, W., Peylin, P., Raymond, P., Rödenbeck, C., Saarnio, S., Schulze, E. D., Szopa, S., Thompson, R., Verkerk, P. J., Vuichard, N., Wang, R., Wattenbach, M., and Zaehle, S.: The European land and inland water CO2, CO, CH4 and N2O balance between 2001 and 2005, Biogeosciences, 9, 3357-3380, 10.5194/bg-9-3357-2012, 2012.

Massman, W. J.: A review of the molecular diffusivities of H2O, CO2, CH4, CO, O3, SO2, NH3, N2O, NO, and NO2 in air, O2 and N2 near STP, Atmospheric Environment, 32, 1111-1127, http://dx.doi.org/10.1016/S1352-2310(97)00391-9, 1998.

Matyssek, R., Wieser, G., Ceulemans, R., Rennenberg, H., Pretzsch, H., Haberer, K., Löw, M., Nunn, A., Werner, H., and Wipfler, P.: Enhanced ozone strongly reduces carbon sink strength of adult beech (Fagus sylvatica)–Resume from the free-air fumigation study at Kranzberg Forest, Environmental Pollution, 158, 2527-2532, 2010a.

Matyssek, R., Karnosky, D., Wieser, G., Percy, K., Oksanen, E., Grams, T., Kubiske, M., Hanke, D., and Pretzsch, H.: Advances in understanding ozone impact on forest trees: messages from novel phytotron and free-air fumigation studies, Environmental Pollution, 158, 1990-2006, 2010b.

McLaughlin, S. B., Nosal, M., Wullschleger, S. D., and Sun, G.: Interactive effects of ozone and climate on tree growth and water use in a southern Appalachian forest in the USA, New Phytologist, 174, 109-124, 10.1111/j.1469-8137.2007.02018.x, 2007a.

McLaughlin, S. B., Wullschleger, S. D., Sun, G., and Nosal, M.: Interactive effects of ozone and climate on water use, soil moisture content and streamflow in a southern Appalachian forest in the USA, New Phytologist, 174, 125-136, 10.1111/j.1469-8137.2007.01970.x, 2007b.

Medlyn, B. E., Badeck, F. W., De Pury, D. G. G., Barton, C. V. M., Broadmeadow, M., Ceulemans, R., De Angelis, P., Forstreuter, M., Jach, M. E., Kellomaki, S., Laitat, E., Marek, M., Philippot, S., Rey, A., Strassemeyer, J., Laitinen, K., Liozon, R., Portier, B., Roberntz, P., Wang, K., and Jstbid, P. G.: Effects of elevated [CO_2] on photosynthesis in European forest species: a meta-analysis of model parameters, Plant, Cell & Environment, 22, 1475-1495, doi:10.1046/j.1365-3040.1999.00523.x, 1999.

Medlyn, B. E., Barton, C. V. M., Broadmeadow, M. S. J., Ceulemans, R., De Angelis, P., Forstreuter, M., Freeman, M., Jackson, S. B., Kellomaki, S., Laitat, E., Rey, A., Roberntz, P., Sigurdsson, B. D., Strassemeyer, J., Wang, K., Curtis, P. S., and Jarvis, P. G.: Stomatal conductance of forest species after long-term exposure to elevated CO_2 concentration: a synthesis, New Phytologist, 149, 247-264, 2001.

Medlyn, B. E., Duursma, R. A., Eamus, D., Ellsworth, D. S., Prentice, I. C., Barton, C. V., Crous, K. Y., de Angelis, P., Freeman, M., and Wingate, L.: Reconciling the optimal and empirical approaches to modelling stomatal conductance, Global Change Biology, 17, 2134-2144, 2011.

Mercado, L. M., Bellouin, N., Sitch, S., Boucher, O., Huntingford, C., Wild, M., and Cox, P. M.: Impact of changes in diffuse radiation on the global land carbon sink, Nature, 458, 1014-1017, http://www.nature.com/nature/journal/v458/n7241/suppinfo/nature07949_S1.html, 2009.

Mills, G., Hayes, F., Wilkinson, S., and Davies, W. J.: Chronic exposure to increasing background ozone impairs stomatal functioning in grassland species, Global Change Biology, 15, 1522-1533, 2009.

Mills, G., Pleijel, H., Braun, S., Büker, P., Bermejo, V., Calvo, E., Danielsson, H., Emberson, L., Grünhage, L., Fernández, I. G., Harmens, H., Hayes, F., Karlsson, P.-E., and Simpson, D.: New stomatal flux-based critical levels for ozone effects on vegetation, Atmospheric Environment, 5064-5068, 2011a.

Mills, G., Hayes, F., Simpson, D., Emberson, L., Norris, D., Harmens, H., and BÜKer, P.: Evidence of widespread effects of ozone on crops and (semi-)natural vegetation in Europe (1990–2006) in relation to AOT40- and flux-based risk maps, Global Change Biology, 17, 592-613, 10.1111/j.1365-2486.2010.02217.x, 2011b.

Mills, G., Harmens, H., Wagg, S., Sharps, K., Hayes, F., Fowler, D., Sutton, M., and Davies, B.: Ozone impacts on vegetation in a nitrogen enriched and changing climate, Environmental Pollution, 208, 898-908, 2016.

Norby, R. J., Wullschleger, S. D., Gunderson, C. A., Johnson, D. W., and Ceulemans, R.: Tree responses to rising CO_2 in field experiments: implications for the future forest, Plant, Cell and Environment, 22, 683-714, 1999.

Norby, R. J., DeLucia, E. H., Gielen, B., Calfapietra, C., Giardina, C. P., King, J. S., Ledford, J., McCarthy, H. R., Moore, D. J. P., Ceulemans, R., De Angelis, P., Finzi, A. C., Karnosky, D. F., Kubiske, M. E., Lukac, M., Pregitzer, K. S., Scarascia-Mugnozza, G. E., Schlesinger, W. H., and Oren, R.: Forest response to elevated CO2 is conserved across a broad range of productivity, Proc. Natl. Acad. Sci. U. S. A., 102, 18052-18056, 10.1073/pnas.0509478102, 2005.

Nunn, A. J., Reiter, I. M., Häberle, K.-H., Langebartels, C., Bahnweg, G., Pretzsch, H., Sandermann, H., and Matyssek, R.: Response patterns in adult forest trees to chronic ozone stress: identification of variations and consistencies, Environmental Pollution, 136, 365-369, 2005.

O'Connor, F. M., Johnson, C. E., Morgenstern, O., Abraham, N. L., Braesicke, P., Dalvi, M., Folberth, G. A., Sanderson, M. G., Telford, P. J., Voulgarakis, A., Young, P. J., Zeng, G., Collins, W. J., and Pyle, J. A.: Evaluation of the new UKCA climate-composition model – Part 2: The Troposphere, Geosci. Model Dev., 7, 41-91, 10.5194/gmd-7-41-2014, 2014.

Paoletti, E., and Grulke, N. E.: Ozone exposure and stomatal sluggishness in different plant physiognomic classes, Environmental Pollution, 158, 2664-2671, 2010.

Parrish, D. D., Law, K. S., Staehelin, J., Derwent, R., Cooper, O. R., Tanimoto, H., Volz-Thomas, A., Gilge, S., Scheel, H. E., Steinbacher, M., and Chan, E.: Long-term changes in lower tropospheric baseline ozone concentrations at northern mid-latitudes, Atmos. Chem. Phys., 12, 11485-11504, 10.5194/acp-12-11485-2012, 2012.

Parrish, D. D., Law, K. S., Staehelin, J., Derwent, R., Cooper, O. R., Tanimoto, H., Volz-Thomas, A., Gilge, S., Scheel, H. E., Steinbacher, M., and Chan, E.: Lower tropospheric ozone at northern midlatitudes: Changing seasonal cycle, Geophysical Research Letters, 40, 1631-1636, 2013.

Percy, K. E., Awmack, C. S., Lindroth, R. L., Kubiske, M. E., Kopper, B. J., Isebrands, J., Pregitzer, K. S., Hendrey, G. R., Dickson, R. E., and Zak, D. R.: Altered performance of forest pests under atmospheres enriched by CO2 and O3, Nature, 420, 403-407, 2002.

Royal-Society: Ground-level ozone in the 21st century: future trends, impacts and policy implications, Science Policy Report 15/08, 2008.

Samuelsson, P., Jones, C. G., Willén, U., Ullerstig, A., Gollvik, S., Hansson, U., Jansson, C., Kjellström, E., Nikulin, G., and Wyser, K.: The Rossby Centre Regional Climate model RCA3: model description and performance, Tellus A, 63, 4-23, 2011.

Saxe, H., Ellsworth, D. S., and Heath, J.: Tree and forest functioning in an enriched CO_2 atmosphere, New Phytologist, 139, 395-436, doi:10.1046/j.1469-8137.1998.00221.x, 1998.

Schulze, E.-D., Ciais, P., Luyssaert, S., Schrumpf, M., Janssens, I. A., Thiruchittampalam, B., Theloke, J., Saurat, M., Bringezu, S., and Lelieveld, J.: The European carbon balance. Part 4: integration of carbon and other trace-gas fluxes, Global Change Biology, 16, 1451-1469, 2010.

Schulze, E. D., Luyssaert, S., Ciais, P., Freibauer, A., Janssens, I. A., and et al.: Importance of methane and nitrous oxide for Europe's terrestrial greenhouse-gas balance, Nature Geosci, 2, 842-850, http://www.nature.com/ngeo/journal/v2/n12/suppinfo/ngeo686_S1.html, 2009.

Sicard, P., De Marco, A., Troussier, F., Renoua, C., Vas, N., and Paoletti, E.: Decrease in surface ozone concentrations at Mediterranean remote sites and increase in the cities, Atmospheric Environment, 79, 705-715, 2013.

Simpson, D., Emberson, L., Ashmore, M., and Tuovinen, J.: A comparison of two different approaches for mapping potential ozone damage to vegetation. A model study Environmnetal Pollution, 146, 715-725, 2007.

Simpson, D., Benedictow, A., Berge, H., Bergström, R., Emberson, L. D., Fagerli, H., Flechard, C. R., Hayman, G. D., Gauss, M., and Jonson, J. E.: The EMEP MSC-W chemical transport model–technical description, Atmospheric Chemistry and Physics, 12, 7825-7865, 2012.

Simpson, D., Andersson, C., Christensen, J. H., Engardt, M., Geels, C., Nyiri, A., Posch, M., Soares, J., Sofiev, M., and Wind, P.: Impacts of climate and emission changes on nitrogen deposition in Europe: a multi-model study, Atmospheric Chemistry and Physics, 14, 6995-7017, 2014a.

Simpson, D., Arneth, A., Mills, G., Solberg, S., and Uddling, J.: Ozone—the persistent menace: interactions with the N cycle and climate change, Current Opinion in Environmental Sustainability, 9, 9-19, 2014b.

Sitch, S., Cox, P. M., Collins, W. J., and Huntingford, C.: Indirect radiative forcing of climate change through ozone effects on the land-carbon sink, Nature, 448, 791-794, http://www.nature.com/nature/journal/v448/n7155/suppinfo/nature06059_S1.html, 2007.

Sitch, S., Friedlingstein, P., Gruber, N., Jones, S. D., Murray-Tortarolo, G., Ahlström, A., Doney, S. C., Graven, H., Heinze, C., Huntingford, C., Levis, S., Levy, P. E., Lomas, M., Poulter, B., Viovy, N., Zaehle, S., Zeng, N., Arneth, A., Bonan, G., Bopp, L., Canadell, J. G., Chevallier, F., Ciais, P., Ellis, R., Gloor, M., Peylin, P., Piao, S. L., Le Quéré, C., Smith, B., Zhu, Z., and Myneni, R.: Recent trends and drivers of regional sources and sinks of carbon dioxide, Biogeosciences, 12, 653-679, 10.5194/bg-12-653-2015, 2015.

Sleutel, S., De Neve, S., and Hofman, G.: Estimates of carbon stock changes in Belgian cropland., Soil Use and Management, 19, 166-171, 10.1079/SUM2003187, 2003.

Sun, G. E., McLaughlin, S. B., Porter, J. H., Uddling, J., Mulholland, P. J., Adams, M. B., and Pederson, N.: Interactive influences of ozone and climate on streamflow of forested watersheds, Global Change Biology, 18, 3395-3409, 10.1111/j.1365-2486.2012.02787.x, 2012.

Tai, P. K. A., Val Martin, M., and Heald, C. L.: Threat to future global food security from climate change and ozone air pollution, Nature Climate Change, 4, 817 - 821, 2014.

Talhelm, A. F., Pregitzer, K. S., Kubiske, M. E., Zak, D. R., Campany, C. E., Burton, A. J., Dickson, R. E., Hendrey, G. R., Isebrands, J. G., Lewin, K. F., Nagy, J., and Karnosky, D. F.: Elevated carbon dioxide and ozone alter productivity and ecosystem carbon content in northern temperate forests, Global Change Biology, 20, 2492-2504, 10.1111/gcb.12564, 2014.

Tans, P., and Keeling, R.: Dr. Pieter Tans, NOAA/ESRL (www.esrl.noaa.gov/gmd/ccgg/trends/) and Dr. Ralph Keeling, Scripps Institution of Oceanography (scrippsco2.ucsd.edu/).

Tricker, P. J., Pecchiari, M., Bunn, S. M., Vaccari, F. P., Peressotti, A., Miglietta, F., and Taylor, G.: Water use of a bioenergy plantation increases in a future high CO_2 world, Biomass and Bioenergy, 33, 200-208, 2009.

Tuovinen, J.-P., Emberson, L., and Simpson, D.: Modelling ozone fluxes to forests for risk assessment: status and prospects, Annals of Forest Science, 66, 1-14, 2009.

Tuovinen, J., Hakola, H., Karlsson, P., and Simpson, D.: Air pollution risks to Northern European forests in a changing climate, Climate Change, Air Pollution and Global Challenges Understanding and Perspectives from Forest Research, 2013.

Uddling, J., Teclaw, R. M., Pregitzer, K. S., and Ellsworth, D. S.: Leaf and canopy conductance in aspen and aspen-birch forests under free-air enrichment of carbon dioxide and ozone, Tree Physiology, 29, 1367-1380, 2009.

Val Martin, M., Heald, C. L., and Arnold, S. R.: Coupling dry deposition to vegetation phenology in the Community Earth System Model: Implications for the simulation of surface O3, Geophysical Research Letters, 41, 2988-2996, doi:10.1002/2014GL059651, 2014.

van Vuuren, D. P., Edmonds, J., Kainuma, M., Riahi, K., Thomson, A., Hibbard, K., Hurtt, G. C., Kram, T., Krey, V., Lamarque, J.-F., Masui, T., Meinshausen, M., Nakicenovic, N., Smith, S. J., and Rose, S. K.: The representative concentration pathways: an overview, Climatic Change, 109, 5, 10.1007/s10584-011-0148-z, 2011.

Verstraeten, W. W., Neu, J. L., Williams, J. E., Bowman, K. W., Worden, J. R., and Boersma, K. F.: Rapid increases in tropospheric ozone production and export from China, Nature Geoscience 8, 690-695, 2015.

Vingarzan, R.: A review of surface ozone background levels and trends, Atmospheric Environment, 38, 3431-3442, https://doi.org/10.1016/j.atmosenv.2004.03.030, 2004.

Weedon, G. P., Gomes, S., Viterbo, P., Österle, H., Adam, J. C., Bellouin, N., Boucher, O., and Best, M. J.: The WATCH Forcing Data 1958-2001: a meteorological forcing dataset for land surface- and hydrological models. , WATCH Tech. Rep. 22, 41p (available at www.eu-watch.org/publications). 2010.

Weedon, G. P., Gomes, S., Viterbo, P., Shuttleworth, W. J., Blyth, E., Österle, H., Adam, J. C., Bellouin, N., Boucher, O., and Best, M.: Creation of the WATCH Forcing data and its use to assess global and regional reference crop evaporation over land during the twentieth century, Journal of Hydrometerology, 12, 823-848, doi: 10.1175/2011JHM1369.1., 2011.

Weedon, G. P.: Readme file for the "WFDEI" dataset.available at: http://www.eu-watch.org/gfx_content/documents/README-WFDEI.pdf, 2013.
Wild, O.: Modelling the global tropospheric ozone budget: exploring the
variability in current models, Atmopsheric Chemistry and Physics, 2643–2660, 2007.
Wilkinson, S., and Davies, W. J.: Ozone suppresses soil drying-and abscisic acid (ABA)-induced stomatal closure via an ethylene-dependent mechanism, Plant, Cell & Environment, 32, 949-959, 2009.
Wilkinson, S., and Davies, W. J.: Drought, ozone, ABA and ethylene: new insights from cell to plant to community, Plant, Cell & Environment, 33, 510-525, 10.1111/j.1365-3040.2009.02052.x, 2010.
Wittig, V. E., Ainsworth, E. A., and Long, S. P.: To what extent do current and projected increases in surface ozone affect photosynthesis and stomatal conductance of trees? A meta-analytic review of the last 3 decades of experiments, Plant, Cell & Environment, 30, 1150-1162, 10.1111/j.1365-3040.2007.01717.x, 2007.
Wittig, V. E., Ainsworth, E. A., Naidu, S. L., Karnosky, D. F., and Long, S. P.: Quantifying the impact of current and future tropospheric ozone on tree biomass, growth, physiology and biochemistry: a quantitative meta-analysis, Global Change Biology, 15, 396-424, 10.1111/j.1365-2486.2008.01774.x, 2009.
Wullschleger, S. D., Gunderson, C. A., Hanson, P. J., Wilson, K. B., and Norby, R. J.: Sensitivity of stomatal and canopy conductance to elevated CO_2 concentration; interacting variables and perspectives of scale, New Phytologist, 153, 485-496, doi:10.1046/j.0028-646X.2001.00333.x, 2002.
Young, P., Arneth, A., Schurgers, G., Zeng, G., and Pyle, J. A.: The CO_2 inhibition of terrestrial isoprene emission significantly affects future ozone projections, Atmospheric Chemistry and Physics, 9, 2793-2803, 2009.
Young, P., Archibald, A., Bowman, K., Lamarque, J.-F., Naik, V., Stevenson, D., Tilmes, S., Voulgarakis, A., Wild, O., and Bergmann, D.: Pre-industrial to end 21st century projections of tropospheric ozone from the Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP), Atmospheric Chemistry and Physics, 13, 2063-2090, 2013.
Zaehle, S.: Terrestrial nitrogen–carbon cycle interactions at the global scale, Philosophical Transactions of the Royal Society B: Biological Sciences, 368, 20130125, 10.1098/rstb.2013.0125, 2013.
Zak, D. R., Pregitzer, K. S., Kubiske, M. E., and Burton, A. J.: Forest productivity under elevated CO2 and O3: positive feedbacks to soil N cycling sustain decade-long net primary productivity enhancement by CO2, Ecology Letters, 14, 1220-1226, 10.1111/j.1461-0248.2011.01692.x, 2011.