Dear Editor,

we implemented the changes suggest by the comments of the Referees. Major changes in the new manuscript version are that ozone damage is now calculated without a flux threshold, according to Referee F. Denteners comment 3e. Furthermore we removed redundancy from the Discussion section and extend the Discussion on requested issues. Please find below the answers to both Referees (section 1 and 2). Thereafter you can find the marked up version of the manuscript showing the implemented changes.

Yours Sincerely
Martina Franz

1 Answers to Referee # 2, F. Dentener

1.1 General remarks

One of the key-equations (derived from Wittig et al., 2007) is equation 16. There are number of issues with the use of this regression equation.

Q: 1) As the authors remark in their discussion, a conceptual problem of using equation is that even at cumulative O_3 uptake of zero, the equation still predicts a -6 % impact on photosynthesis. Also the slope of the equation -0.22 % per mmol m-2 is low compared to some other studies. I would suggest that refitting of the data, and forcing the values to go through zero is one option for a sensitivity study. Possibly another option is to re-fit these data to cumulative uptake above the threshold. On page 3/l. 28 the parameterization of Lombardozzi (2015) is mentioned, however without discussion on why this relationship is not used.
A: A refitting of the Wittig damage function would be desirable. However, a data request to V. Wittig remained unanswered. A refitting can not be done without repeating the work done by the meta-analysis.
There are several reasons for not using the Lombardozzi damage function. For tree species, Lombardozzi et al. (2015) assume a fixed reduction of net photosynthesis due to ozone independent of the actual ozone uptake. This fixed reduction is -12.5 % for broadleaved species and -16,1 % for needleleaved species. Only for crops and grasses ozone damage to net photosynthesis depends on ozone uptake. In other words, the atmospheric ozone

concentration and ozone uptake into the plants do not affect the damage estimate for tree species but only for grasses and crops. Due to the lack of impact of ozone uptake on ozone damage estimates, the offset implied by Lombardozzi et al. (2015) is actually higher. The effect of the step decrease in Lombardozzi et al. (2015) might be ameliorated by the fact that canopy conductance is affected in parallel. However, this results in a general decoupling of photosynthesis and canopy conductance. Our aim here was to investigate the effect of ozone damage to net photosynthesis under the assumption that photosynthesis and canopy conductance remain coupled. We have extended the discussion to make this point clearer.

Q: 2) I would appreciate some discussion with regard of the validity of the experimental relationship for leaf-level ozone, or whether it also suffers from some atmospheric diffusion effect? Ideally when using such parameterisations the experimental conditions should be reproduced. I propose the authors have a look at some of the data used in Wittig, to resolve this issue
A: The experiments used by Wittig et al. (2007) do not use the leaf-level ozone concentration to calculate ozone uptake but the atmospheric ozone concentrations. The ozone uptake calculation thus differs in this respect between our simulations and the experiments used to derive the damage relationship. However in the experiment, ozone uptake is not directly measured. Rather, it is calculated from mean ozone concentrations over the exposure period and the respective average stomatal conductances. Thus the estimated ozone uptake rates and hence the amount of accumulated ozone used to derive a damage relationship are a coarse approximation and underlie considerable uncertainty. Following this the error introduced by using leaf-level ozone concentrations instead of atmospheric concentrations seems small, especially since the use of the leaf-level ozone concentrations is the physiological more appropriate approach. We have extended the discussion to make this point clearer.

Q: 3) I have difficulties to understand equation (15) page 8. Several issues need clarificiation: a) Why is Fst, detox used? The cumulative ozone uptake is dependent on the overall flux, regardless whether it is detoxified or not.
A: The Wittig damage function bases on CUO which accumulates the ozone uptake without a threshold. We changed this equation and rerun all simulations. In the new version ozone damage is calculated on ozone uptake accumulated without a threshold. We note that this does not affect any of our conclusions, but agree with the reviewer that this is a cleaner way to address the issue.

Q: b) what is the rationale of using the factor fshed? Why would young leaves be less or not sensitive to ozone damage? What is the reference for this?
A: f_{shed} is the fraction of new developed leaves per time step and layer. In the revised version, this factor was renamed to (f_{new}) to facilitate the understanding. New grown leaves are assumed to be undamaged. For evergreen species the old damaged leaves still exist when new leaves are grown. In this condition, f_{new} causes the canopy layer CUO to be reduced when new leaves are grown, because they are health do not suffer ozone damage yet, i.e. if 10 % new leaves are grown ($f_{new} = 0.1$), the CUO is reduced by 10 %. Without this equation, newly grown leaves would be assumed to be similarly damaged to already existing foliar, which is not correct, and would cause the CUO for evergreen species would continuously increase over the years.

Q: c) Rearranging this equation 15 gives CUO=cFst,detoxdelta_t/fshed - I guess in times that fshed is close to zero, the values of CUO can get very large. I suspect something is not correct with this equation.
A: The equation was rewritten as:

$$\frac{dCUO_l}{dt} = (1 - f_{new})CUO_l + cF_{st,l} \tag{1}$$

As already mentioned in b). f_{new} (formerly f_{shed}) s the fraction of new developed leaves per time step and layer. f_{new} can take values between zero and one. $f_{new} = 0$ when no leaves are grown in the present time step, and $f_{new} = 1$ when newly grown leaves make up all of the present canopy. The CUO_l of the previous time step is reduced according to the fraction of new grown leaves $(1 - f_{shed})CUO_l$.

Q: d) I would expect that CUO is something integrated over the canopy, as mentioned in p. 8 l 18- but it would be good to have the equation already describing this.
A: An additional equation clarifying this was added (new eq. 15):

$$CUO = \sum_{l=1}^{n} CUO_l. \tag{2}$$

Q: e) see remark 2) but the equation 16 seems to be valid for cumulated ozone flux, not for fluxes corrected for detoxification, as suggested by equation 15.

A: Yes. Equation 15 was adapted to use F_{st} (without a threshold), and all simulations are rerun. Plots containing $CUO^1.6_5$ are substituted with CUO of skipped. Equation 14 was skipped.

Q: f) Somewhat related to the point above: even if plants can detoxify ozone, some costs will be associated with this mechanism. Where is the impact of this process accounted for
A: Costs for detoxification are not accounted for in the current model version. To our knowledge no suitable data are available to parametrise e.g. the increased respiration costs according to ozone uptake, since it is very hard to disentangle costs for ozone detoxification from other factors influencing leaf respiration under elevated ozone exposure.

Q: 4) Missing processes: there are several publications suggesting that ozone damage advances senescence (e.g. Gielen, 2007). Further ozone can damage of stomata- leading to sluggishness (e.g. Paoletti) To what extent are these processes included and how would they affect results?
A: Reduction of photosynthetic capacity is one feature of early senescence, others are not included. Omitting effects like early litter fall will underestimate ozone damage. Stomatal sluggishness is not included in the model version described here. Transpiration rates are thus underestimated compared to accounting for sluggishness. A model version of OCN exists where sluggishness can be accounted for however in this case it occurs permanently for all PFTs. This seems to overestimate the effect at least in regions where low ozone concentrations occur. Following this stomatal sluggishness is an important aspect of ozone damage however it seems not reasonable to generally include it in the base model version. The simulation of sluggishness might be very interesting in a sensitivity study where also other effects like detoxification (e.g. through various flux thresholds) are tested on their impact on ozone damage estimates. We have extended the discussion to clarify that the current model does not include all known ozone effects.

5) Coupled atmospheric dynamics-vegetation ozone models suggest rather strong at- mospheric responses and feedbacks. E.g. Super, Vilàâ RGuerau de Arellano, Krol, JGR, 2015 as well as some papers cited here. I think the virtue of this publication is an increased understanding of the vegetation dynamic response (still with a lot of uncertainties), but in addition coupled atmosphere-vegetation simulations are still in its infancy. This should be clearly mentioned in abstract and conclusions.

A: We add this point to discussion and abstract.

1.2 Minor remarks

Q: p. 1 l.6 free troposphere is the region above the boundary layer. I guess the authors mean near-surface ozone in the planetary boundary layer
A: Is changed to 45 m height.

Q: p.1 l.9 Although it probably doesn't matter: are the authors comparing the model with or without ozone effects
A: The model 'including O_3 damage' is used.

Q: p. 1 l. 17- outside the leaves: suggest to call this near-leaves, or leaf-level ozone.
A: Is called 'leaf-level ozone'.

Q: p. 2 l. 3 As raised in general comments, are the effects of anti-oxidant mechanisms somehow included?
A: No, since the flux threshold is omitted in the final version no detoxification occurs.

Q: p. 2 l. 4 Better to include a range: a factor 2 to 5. I personally do not think a factor of 5 is realistic.
A: Changed to 'a factor 2 to 5'.

Q: p. 2 l. 11 delete 'less polluted' transport is taking place regardless of pollution levels.
A: Done.

Q: p. 3 l. 18 'no damage' is observed. Detoxification: explain what consequences for productivity this can have
A: Detoxification causes increased respiration costs and following this reduces NPP what may reduce growth and biomass. Included: 'The production of defence compounds increases respiration costs and following this reduces net primary production what may result in reduced growth and biomass

(Ainsworth et al., 2012).'

Q: p. 3 l. 28. Explain why this parameterization was not used
A: Atmospheric ozone concentrations and cumulated O_3 uptake only impact net photosynthesis of one plant functional type directly. For the two other plant types net photosynthesis is reduced in a step function independent of the accumulated ozone uptake.

Q: p. 4 l. 6 sensitivity analysis towards selected critical parameters?
A: The aim of the sensitivity study is to test the functionality of the deposition model, because it is calculates leaf-level O_3 concentrations and hence has a large impact on O_3 uptake estimates. The variable R_b is also an important variable of the deposition model and was added to Fig. 3. The respective sentence was changed from "provide a sensitivity analysis of the model to evaluate the reliability of simulated values of O_3 uptake' ' to 'provide a sensitivity analysis of critical variables and parameters of the deposition model to evaluate the reliability of simulated values of O_3 uptake'.

Q: p. 4. L. 9 accumulation of what?
A: Accumulation of ozone. Changed to 'O_3 uptake and cumulated uptake'.

Q: p.4 l. 11-25 I would appreciate some more information on the models. How many canopy layers are in OCN?
A: There are maximum 20 layers. The number of actual simulated layers depends on the site and the PFT. Included in Methods section.

Q: Is there an interaction of N in leaves with ozone?
A: Yes. Photosynthetic capacity depends on leaf nitrogen concentration and leaf area, which are both affected by ecosystem available N. Increases in leaf nitrogen content enable higher net photosynthesis and higher stomatal conductance per unit leaf area. This in turn affects transpiration as well as ozone uptake and ozone damage estimates. Included in Methods section.

Q: What version of the EMEP model (output) was used, regional, global, resolution? Explain vertical structure of EMEP- can a constant mid-of-the gridbox of 45 meter be safely used, or are the regions (e.g. in the mountains) where this value is different (i.e. is the coordinate system fixed altitude, pressure or hybrid)?

A: We used version 4.4 of the EMEP MSC-W model, in essentially the same setup described in the nitrogen deposition study of Simpson et al. (2014b). The model was run for the regional RCA3 domain, driven by RCA3 meteorology. The vertical structure is the standard EMEP one (see Simpson et al., 2012) with a lower layer of about 90m thickness. The coordinates are terrain following (sigma coordinates) though, so the mid-point of ca. 45m is relative to the assumed ground surface in such a system. The main assumption of all EMEP deposition modelling is that this 45m height lies within the surface layer, so that standard similarity theory can be applied. This assumption is not always correct of course, but in general the EMEP model's predictions of near-surface ozone (and even fluxes, e.g. Klingberg et al, 2008) suggest that the methodology is reasonable.

Q: I think the model can also output near-surface ozone (diagnostic). Why was this not used- it would avoid additional uncertainty in the recalculation of the atmospheric resistance.
A: The leaf-level ozone concentrations computed by EMEP can not directly be used by OCN, since EMEP and OCN differ in a number of properties, as for instance in the number of simulated plant functional types, and importantly their ecophysiological process representation. Both models differ in the simulation of various ecosystem processes (e.g. phenology, canopy processes, biogeochemical cycles, and vegetation dynamics, which are more explicitly represented in OCN), which in sum impact stomatal and non-stomatal ozone deposition and through this the leaf-level ozone concentration. A possible further development of the new OCN is the coupling to a CTM, to allow for a consistent simulation of tropospheric O_3 burden and vegetation O_3 uptake. We added the explanation to the Methods section.

Q: p. 5 Ra is the resistance between the surface (near-canopy) and 45 meter (i.e. it is not at a level of 45m).
A: Changed to 'between 45 m height and the canopy'.

Q: p. 4 l. 18 Can something be said on how this conductance is distributed over the canopy layers- in general how vertical canopy structure is expected to influence ozone uptake
A: Leaf N is generally highest in the top canopy and monotonically decreases with increasing canopy depth. Following this stomatal conductance and O_3 uptake is highest in the upper canopy and lowest in the bottom of the canopy. Included in Methods section.

Q: p. 5 l. 19: was this calibration necessary for this study, or more general for OCN model results?
A: This calibration is generally necessary to yield reasonable conductance values in OCN.

Q: p. 6 mention which three PFTs were considered for this LAI+1 approach? Probably for the readers not wanting to go back to older papers, a table listing some characteristics of the PFTs (appendix) would be useful
A: The LAI+1 approach is applied for all tree PFTS (woody PFTs).

Q: p. 6/7 eq. 8,9,10 to what extent are these equations based on observations, or merely model assumptions (and what is the associated uncertainty)
A: These equations are largely the same as used in the EMEP model. As described in Simpson et al, 2012, Eqn (8), for F_T is taken from Zhang et al. (2012), Eqn (9), R_{inc} is from Erisman et al (1994), and Eqn (10), giving the effect of snow on R_{gs}, is also loosely based upon Zhang et al.
Although all such equations are uncertain (all depositions schemes are!), the EMEP model's deposition scheme (and associated DO_3SE module for O_3) has undergone extensive review and comparison measurements, see for example:

- Emberson, s. D.; Büker, P. & 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 Environ. Poll., 2007, 147, 454-466
- Emberson, L.; Ashmore, M.; Simpson, D.; Tuovinen, J.-P. & Cambridge, H. Modelling and mapping ozone deposition in Europe Water, Air and Soil Pollution, 2001, 130, 577-582
- Emberson, L.; Wieser, G. & Ashmore, M. Modelling of stomatal conductance and ozone flux of Norway spruce: comparison with field data Environ. Poll., 2000, 109, 393-402
- Klingberg, J.; Danielsson, H.; Simpson, D. & Pleijel, H Comparison of modelled and measured ozone concentrations and meteorology for a site in south-west Sweden: Implications for ozone uptake calculations Environ. Poll., 2008, 115, 99-111
- Simpson, D.; Tuovinen, J.-P.; Emberson, L. & Ashmore, M. Characteristics of an ozone deposition module II: sensitivity analysis Water, Air and Soil Pollution, 2003, 143, 123-137

- Simpson, D.; Tuovinen, J.-P.; Emberson, L. & Ashmore, M. Characteristics of an ozone deposition module Water, Air and Soil Pollution: Focus, 2001, 1, 253-262
- Tuovinen, J.-P.; Simpson, D.; Mikkelsen, T.; Emberson, L. D.; Ashmore, M. R.; Aurela, M.; Cambridge, H. M.; Hovmand, M. F.; Jensen, N. O.; Laurila, T.; Pilegaard, K. & Ro-Poulsen, H. Comparisons of measured and modelled ozone deposition to forests in Northern Europe Water, Air and Soil Pollution: Focus, 2001, 1, 263-274
- Tuovinen, J.-P.; Emberson, L. & Simpson, D. Modelling ozone fluxes to forests for risk assessment: status and prospects Annals of Forest Science, 2009, 66, 401
- Tuovinen, J.-P.; Ashmore, M.; Emberson, L. & Simpson, D. Testing and improving the EMEP ozone deposition module Atmos. Environ., 2004, 38, 2373-2385
- Tuovinen, J.-P.; Emberson, L. & Simpson, D. Modelling ozone fluxes to forests for risk assessment: status and prospects Annals of Forest Science, 2009, 66, 401

In any case, it can be noted that the low-temperature and snow terms given by Eqns (8) and (10) are only really important in conditions for which ozone uptake will be very small.

Q: p. 7 l.18 1 mmol/m2: is this referring to cumulative ozone uptake? Is this published (reference?). I am not sure if such sensitivity with an atmospheric model which would include chemistry feedbacks, can be translated into such small uncertainty for the vegetation O_3 uptake. Note that there is in general a quite large difference in PBL mixing in a variety of atmospheric models-which in itself already suggests a large uncertainty.
A: Yes, the 1 mmol/m2 is for CUO with threshold 1.6. This estimate comes from tests done for this paper, by running the EMEP model with different assumptions, but it only represented the uncertainty due to the OCN simplifications in resistance terms, not of course the overall uncertainty in the model system. In any case, since we now use a zero threshold, and have modified the OCN resistance terms, a new calculation was needed.
The respective paragraph was changed to: 'However, a series of calculations with the full EMEP model have shown that the uncertainties associated with these simplifications are small, typically 0.5 - 5 $\mathrm{mmol\,m^{-2}}$ As base-case values of POD0 are typically ca. 30-50 in EU regions, these approximations do not seem to be a major cause of error, at least in regions with substan-

tial ozone (and carbon) uptake. The full coupling of OCN to a CTM would be desirable to eliminate this bias and allow for a consistent calculation of tropospheric and surface near O3 burdens.'

Q: p. 8 l-1-20 See remarks above- need to get a better description if/how detoxification is included
A: The flux threshold simulating detoxification was skipped and all simulations were rerun.

Q: p. 8 l. 8: do I understand correct that in the rest of the text $\mathrm{CUO}_5^{1.6}$ would refer to equation 15; while CUO would refer to use Fst in equation 15. This needs to become clear- and the correct equations need to be given.
A: The flux threshold was skipped and following this also $\mathrm{CUO}_5^{1.6}$ was skipped. The cumulated O_3 uptake (CUO) derives from the accumulation of the ozone uptake without any flux threshold (F_{stC}).

Q: p. 9 l. 5. Would a sensible variation of dl (equation 16) also be a critical parameter?
A: The objective was to test functionality of the implemented deposition scheme. The validity of the implemented damage function is a very interesting topic however would have expanded the manuscript too much. We are currently working on evaluating different damage functions implemented in OCN in their ability to reproduce observe damage relationships.

Q: How was this subset of parameters selected.
A: Key parameters of the deposition scheme which determine leaf-level O_3 concentrations and hence O_3 uptake are investigated. The variable R_b is also a key variable of the deposition scheme and was added to Fig. 3.

Q: p. 9 l. 22 What is the La Thuille dataset?
A: The La Thuile Dataset contains the data of all sites and years of the FLUXNET network. The respective web link is included as a reference: 'http://fluxnet.fluxdata.org/data/la-thuile-dataset/'.

Q: p. 9 l. 25 How many years were need to reach equilibrium?
A: 1200 simulation years for the vegetation and 12000 years for the soil secure equilibrium.

Q: What was the criterium for equilibrium?

A: Equilibrium is reached when the carbon and nitrogen pools in vegetation and soil show no trend anymore as mentioned on p 10 l 7.

Q: p. 9 l 28: Which EMEP simulations were providing this 100 years transient concentrations? Is there a reference?
A: p 10 l 6 indicates that more details regarding EMEP are given in section 2.5, including also a reference.

Q: p. 9 l. 29 Appendix tab 1? I think just table 1
A: The appendix section was unintentionally included into the main part of the paper.

Q: p. 10 I understand that the purpose of section 2.4 is to derive trust in the model, when testing to observable parameters. I would however need some more insight in why morning/evening fluxes need to removed, and data with the different soil moisture. What would be the effect of not removing such data?
A: The morning and evening hours are removed since in this time dew condensation on the leaves causes a wet canopy. This causes an alteration in latent heat exchange (LE) such that FLUXNET observed canopy conductance, which is inferred from LE, is prone to a high uncertainty in these times. Soil moisture constraints directly impact the simulated net photosynthesis (see Θ in Eq. 5). It is hard for a global model, not tuned for the specific site, to properly model the drying of the soil and onset of soil moisture stress (which depends e.g. on soil type and texture as well as the degree of root penetration). By excluding data under soil moisture stress this bias is removed.

Q: p. 10 l. 29- brought into equilibrium. How done?
A: The model is run with the 1961-1970 forcing until equilibrium of the carbon and nitrogen pools in vegetation and soil is reached. The forcing for each year of the Spinup phase is randomly chosen from the period 1961-1970. Changed in the text from 'with 1961-1970 forcing' to 'by randomly iterating the forcing from the period 1961-1970'.

Q: p. 11 figure 1: obviously the largest discrepancy is found for LAI and in p. 12/l. 18 the authors suggest that this is not important. How is it possible to have realistic GPP etc; and such a spread in LAI? Please explain

A: The LAI measurements presented here are point measurements of years outside the simulation period. The actual LAI values at the FLUXNET sites during the simulation period might be different. Furthermore in OCN GPP depends on LAI in a non-linear relationship where GPP saturates with increasing values of LAI (saturation point at a LAI of approximately 4). When LAI increases further the lower canopy does not get sufficient light to increase GPP. GPP however is not only determined by LAI, but also e.g by temperature, radiation and soil moisture stress what might ameliorate differences in LAI. Added: 'Modelled GPP does not only depend on LAI, but also on light availability, temperature and soil moisture.'

Q: p. 12 l. 24. While it is facilitating the discussion to focus on only 3 stations, some words on how representative these stations were for others would be welcome.
A: The three sites were chosen to be examples of the 3 major categories. The respective sentence was reformulated to: 'For further evaluation of the modelled ozone uptake, we analysed the diurnal cycles at three sites, one of the three categories broadleaved, needle-leaved and C3 grass sites respectively.'

Q: p. 13 l. 5-25 I would advise to also see Hardacre, Atmos. Chem. Phys., 15, 6419-6436, 2015, for further opportunities to evaluate the ozone deposition velocities and fluxes.
A: Hardacre et al. 2015 was included into the evaluation of the deposition velocities. The fluxes given in Hardacre et al. 2015 are total dry deposition values. In the manuscript here we evaluate the stomatal fraction of the dry deposition (F_{stC}). A comparison of both is not possible.

Q: p. 13 l. 35 Can you confirm that sapflow measurements are not reliable for this study?
A: We can not judge which measurements (eddy covariance or sap flow) are reliable. However we observe that between both techniques the estimates of canopy conductance differ by a factor of more than 10 and that our estimates reported here are more similar to estimates done by measurements conducted by the eddy covariance technique. Since canopy conductance drives O_3 uptake, a 10 fold higher canopy conductance results in an approximately 10 fold higher O_3 uptake rate (disregarding in this approximation the feed back of O_3 uptake into the leaf on decreasing leaf-level O_3 concentrations).

Q: p. 14 repeat here that Fr is the ratio of stomatal to overall flux. It would

be interesting to give average values (per ecosystem/PFT) over the months. Perhaps for an appendix? I think this could be useful for comparison in future studies.
A: The explanation of Fr on p 14 l 6 was changed from 'The ratio between the vegetation ozone uptake and the total surface uptake (Fr)' to 'The ratio between the stomatal ozone uptake and the total surface uptake (Fr)'. A graph showing monthly mean values of key ozone metrics is added to the appendix (Appendix 11).

Q: p. 14 l. 34 I didn't quite understand the sentence not zero because accumulate over several years. Isn't it simply that there is already some photosynthesis activity?
A: Accumulated ozone is shed when leaves are shed. Deciduous PFT's shed all accumulated O_3 at the end of the growing season when the leaves are shed. Evergreen species only shed a fraction of their leaves and keep the leaves that have already taken up O_3 for several years. The CUO decreases in winter when the evergreens shed part of their leaves but since they do not shed all the CUO remains greater than zero.

Q: p. 15 l. 8-10. It is not clear to me whether OCN has croplands, and if so what crop? The authors mention C3 crops- I guess that would be mainly wheat?
A: OCN simulates 12 PFTs including 8 tree PFTs, 2 grass PFTs and 2 crop PFTs. The crop PFTs are a generic C3-crop and a generic C4-crop. As species are not explicitly simulated for the tree and grass PFTs this is also not done for the crops.

Q: p. 15 l. 17 Figure 6a is an EMEP model output?
A: Ozone concentration plotted in Fig. 6a is the forcing OCN uses for the simulations. This forcing is provided by EMEP.

Q: p. 15 Appendix 12 ab missing. Do you mean Figure 12? See=¿sea
A: The appendix section was unintentional included into the main part of the paper (Appendix 12 == Fig. 12).

Q: p. 15 l. 34 interesting dynamical/phonological feedback, but it also reminds that things like early senescence are probably not included.
A: Yes, early senescence is not included.

Q: p. 16 section 3.5 Please remind reader of what D-STO and ATM were? See section 2.6.
A: Changed to 'the D-STO model (non-stomatal depletion of ozone is zero) and 20-25% for the ATM model version (canopy O_3 concentration is equal to the atmospheric concentration) '.

Q: Appendix 11 and 13 are missing.
A: The appendix section was unintentional included into the main part of the paper.

Q:L. 13 uptake and accumulated: rephrase in:accumulated uptake.
A: Done.

Q: p. 16 spell out the meaning (remind the reader) of CUO1.5 and 5.
A: Due the omission of the flux threshold $\mathrm{CUO}_5^{1.6}$ does no longer occur.

Q: p. 17 section 4.1, l..10 Interactions with VOCs (as well as soil NOx emissions, see Ganzeveld's paper), are important. But I don't understand how they are implicitly included, especially in the OCN framework.
A: All ozone deposition models that we are aware of have terms for the stomatal uptake of O_3, and then for 'non-stomatal' terms in some form (G_{ns} in Eqn. 4). The stomatal terms can be estimated quite well, e.g. from water fluxes. Unfortunately, the values assigned to G_{ns} cannot be determined from first principles or even experiment because of the complexities of the surface characteristics (moisture films, chemical compounds on leaves, etc, Fowler 2009), and of interpreting flux measurements in the chemically-active conditions associated with vegetation canopies. Thus, the G_{ns} terms encompass both deposition and chemical processes - they are essentially tuned to give reasonable values for deposition velocities across diurnal cycles for example.

Q: p. 17 l. 20: was O_3 needed to reach this good agreement. Probably not-explain.
A: Given the uncertainty of the observations and model results the inclusion of ozone damage does not improve the fit of the model results to the observations. The comparison to FLUXNET data was mainly meant to show that the model in general produces realistic values especially for the canopy conductance (G_c), since G_c is a major factor determining ozone uptake and

hence estimated damage.

Q: p. 19 l 3. As explained above, I think this warrants some additional analysis.
A: The validity of the implemented damage function is a very interesting topic however would have expanded the manuscript too much. We are currently working on evaluating different damage functions implemented in OCN in their ability to reproduce observe damage relation ships. This is a topic of its own.

Q: p. 19 l. 22 impacted=¿determined?
A: Changed to determined.

1.3 Figures

Q: Figure 6: Why are the units of panels b and c different?
A: The units are different because different variables are plotted. In panel b the mean ozone uptake rate [$\mathrm{nmol\,m^{-2}\,s^{-1}}$] is plotted. In panel c the mean ozone accumulation [$\mathrm{mmol\,m^{-2}}$].

Q: The chosen range doesn't work well for panel b (all purple).
A: The color range is not the problem in the big purple area. The values of the mean uptake rate all lie between 1.9 and 2, which simply is a small range.

Q: Figure 7: it is hard to discriminate the colors in Figure 7.
A: The color palette is changed from rainbow to restricted color gradients (palettes from ColorBrewer 2.0).

Q: Figure 9: legenda describing a) can be improved.
A: Changed from 'no ozone deposition scheme (ATM),' to: 'canopy O_3 concentration is equal to the atmospheric concentration (ATM)'

Q: Figure 12: color scheme doesn't work (mostly red)- more resolution for low values is need (0-10%). For C4 crops- is irrigation considered?
A: Irrigation is not considered for crops. The graph is skipped due to it's

minor value in explaining observed results.

2 Answers to anonymous Referee # 2

2.1 General comments

Q: The authors use “ozone” and “O3” fairly randomly throughout the manuscript. I would suggest sticking with one or the other.
A: 'Ozone' and 'O_3' are not used randomly. 'O_3' is used when we refer to the chemical substance and 'ozone' is used when we refer to the damage O_3 causes or the included deposition scheme. In the cases where this was not consistent we changed it to the above mentioned rule. We would like to keep it that way if it is not distracting.

2.2 Abstract

Q: P1, L6 - This is the first use of the acronym OCN - please explain what it is.
A: Added: '(the OCN terrestrial biosphere model)'

Q: P1, L12 - “update” should read “uptake”
A: Done.

Q: P1, L15-6 - Please re-word, this is hard to follow. I think that you are saying: “When applied at the European scale, we find that including our new ozone deposition scheme substantially affects simulated ozone”
A: Changed to: 'When applied at the European scale, we find that the inclusion of the deposition scheme substantially affects simulated ozone ...'

2.3 Introduction

Q: P2, L22 - replace “consequence” with “result”
A: Done.

Q: P2, L24 - replace “extend” with “extent”
A: Done.

Q: P2, L27-29 - I suggest making the point here that AOT40 is currently used for regulatory assessment purposes in Europe.
A: Changed from ’A widely used example’ to ’The initial standard tool’ And furthermore is added: ’Observed ozone damage in the field seems to be better correlated to flux-based risk assessment compared to concentration based methods (Mills et al., 2011). Following this the LRTAP Convention recommends flux based methods as the preferred tool for risk assessment (LRTAP Convention, 2010).’

Q: P2, L32-33 - Please could the authors explain what they mean by “regional provenances”. Do they mean that the same species in different geographical locations differ? Or that different regions have different ecosystems?
A: It is meant that canopy conductance of the same species differs when grown in different geographical locations as well as differences exist between species. Changed to: ’A significant caveat of concentration-based assessments of ozone toxicity effects is that species differ vastly in their canopy conductance as well as regional provenances of one species.’

Q: P3, L8 - Up until this point the authors have referred to AOTX. As AOT40 is the regulatory metric and one that they use in subsequent analysis and discussion I suggest they clearly define AOT40 at this point.
A: ’(AOTX above a threshold of 40 ppb)’ is added.

Q: P3, L23 - I suggest the authors make the point that the threshold values are species-specific to account for plant sensitivity/tolerance to ozone.
A: ’, depending on the specific species sensitivity to ozone. ’ is added to the sentence.

2.4 Methods

Q: P4, L20 - The model acronym EMEP MSC-W should be defined here rather than at the end of the paragraph, e.g. “The ozone and N-deposition data used for this study are provided by the EMEP MSC-W (European

Monitoring and Evaluation Pro- gramme Meteorological Synthesising Centre - West) chemical transport model (CTM; Simpson et al., 2012a)."
A: Done as suggested.

Q: P4, L22 - insert "been" between "have" and "documented"
A: Done.

Q: P5, L1 - replace "in" with "at" and remove "height"
A: Done.

Q: P5, L7 - replace "in" with "at" and remove "height"
A: Changed to 'between 45 m height and the canopy' according to F. Dentener's comment.

Q: P5, L15-6 - replace "leafs internal" with "internal leaf"
A: Done.

Q: P5, L16 - parentheses should only be around "2005"
A: Done.

Q: P5, L17 - replace "ozone to water vapour" with "ozone from water vapour"
A: Done.

Q: P5, L19 - is this factor of 0.7 included in Zaehle and Friend or is this new for this current study?
A: It is new in this study. Yet this calibration is generally necessary to yield reasonable conductance values in OCN.

Q: P6, L11 - please explain more clearly what is meant by a low temperature correction factor and why it is needed.
A: According to Simpson et al. (2012) and Zhang et al. (2003) FT is needed since at temperatures below $-1\ ^{\circ}C$, non-stomatal resistances increase up to two times (hence also the boundary of $1 \leq F_T \leq 2$). Added: For temperatures below -1 °C non-stomatal resistances are increased up to two times (Simpson et al., 2012; Zhang et al., 2003)..

Q: P6, L11 - suggest rewording to: “is scaled by a low temperature correction factor, FT, such that”
A: Changed to: ’is scaled by a low temperature correction factor F_T and’

Q: P6, L13 - suggest rewording to: “where TS is the 2m air temperature (C; Simpson et al., 2012a, eq. 60) and 1¡FT¡2.”
A: The reference by Simpson et al. (2012) also refers to $1 \leq F_T \leq 2$, hence the proposed alteration would take away information.

Q: P6, L20 - replace “Like” with “As”
A: Done.

Q: P7, L1 - parentheses should only be around “2003”
A: Done.

Q: P7, L4 - suggest combining to give: “0.5, to prevent negative values in the first fraction of eq. 10”.
A: Done.

Q: P8, L4 - Why PODl? My understanding of PODY is that the Y stands for the threshold value not the canopy level.
A: Yes. The PODY usually refers to the top canopy layer and not the canopy integrated value contrary to CUO. The ’l’ was there to indicate the same canopy layer as in CUO, however I also see that it is misleading. I erased the ’l’.

Q: P8, L13 - What is the physical (real-world) interpretation of the parameters 0.22 and 6.16 in eq. 16?
A: The parameter 6.16 suggests that at zero ozone uptake net photosynthesis is damaged by 6.16 %. Per mmol accumulated ozone uptake the net photosynthesis is further damaged by 0.22 %.

Q: P8, L13-4 - Why not just divide by 100 in the equation itself?
A: The equation in the numerator is the original equation by Wittig et al. (2007) which gives the damage in percent. Since we needed the fraction [0,1] instead of the percentage it seemed the clearest way to indicate this.

Q: P8, L17 - Please explain to the general audience why a reduction in An results in reductions in Gst and (particularly) Ci. It is not intuitive why this would reduce internal concentrations.
A: The stated reduction of C_i was wrong. ' and C_i' was erased.

Q: P8, L23 - parentheses should only be around "2010"
A: Done.

Q: P9, L2-3 and throughout - I would suggest that the authors re-define or at least use a word description each time these parameters are re-introduced at the start of a new section; else provide a table listing the key parameters for the reader to refer back to.
A: Are reintroduces again.

Q: P9, L11 - Are the "summer months" defined here the same as what is then referred to as the "growing season"; if so, please make clear, if not, please define growing season separately.
A:Growing season is not equal to summer month. Growing season is defined: 'To derive average growing-season fluxes (bud break to litter fall), ...'

Q: P9, L21 - Please explain what is meant by "site levels". Is this "site-specific" i.e. OCN is run as a column model rather than a 3-D regional model?
A: site level means that the simulation is run only on a single set of coordinates and not for a region. Changed to: 'The site levels simulations (single point simulations) ...'

Q: P9, L22 - square parentheses are not required around CO2 as the text includes the word "concentrations".
A: Parentheses are erased.

Q: P9, L23 - parentheses should only be around "2015".
A: Done.

Q: P9, L23 - rearrange this to read: "Reduced and oxidised nitrogen deposition in wet and dry forms and hourly"
A: Done.

Q: P9, L27 - O3 should be subscript
A: Done.

Q: P9,L28-9 - Why not use GCM output or reanalyses data where there is a lack of observation data?
A: We have observation data for all stations but only for the observation period. The model however needs to be in equilibrium to yield sensible results hence a Spinup has to be run (approximately 1200 years for the vegetation). To be able to use the GCM climate it would have to be bias corrected for all climate variables to prevent a step change when changing to use the observed data at the FLUXNET stations for the observation period. This bias correction is much work besides the fact that bias correction except of temperature is not trivial. The use of the observed climate for the Spinup period constitutes a secure way to prevent step changes at the start of the observation period.

Q: P9, L30 - what do the authors mean by time-varying here? Surely the progressive simulations also used data that varied with time. Do the authors mean that here it is observations from the site in question for the years in question?
A: Meant is the year in question. Rephrased to: 'The observation years (see Appendix Tab. 1) are simulated with the climate and atmospheric conditions (N deposition, CO_2 and O_3 concentrations) of the respective years.'

Q: P10, L2 - Why have the authors chosen to base LAI on single point, time-specific observations rather than e.g. MODIS LAI data? It seems that this introduces a considerable source of uncertainty.
A: MODIS data are also subject to a considerable amount of uncertainty. Furthermore the resolution of MODIS data is an additional source of uncertainty. Using observation directly from the site in question seemed to be the most reliable source.

Q: P10, L5 - parentheses should only be around "2015"
A: Done.

Q: P10, L6-7 - suggest rewording to read: " are filtered prior to deriving average growing-season fluxes to reduce the effect of model biases on the

model-data comparison. Night-time and ”
A: Done.

Q: P10, L9 - please explain what a “modelled soil moisture constraint factor” is, and why a threshold of 0.8 has been chosen as a filter. Is this based on observations suggesting severe drought impacts alter fundamental plant functioning?
A: The soil moisture constraint factor is the Θ in Eq. 5. It constrains net photosynthesis when soil moisture decreases and takes values between zero and one. The threshold of 0.8 secures relative humid soils since site specific soil moisture constraints are hard to capture with a global model. The drying of soils is hard to capture for a model operating on 1 degree resolution since it depends e.g. on soil type and texture as well as the degree of root penetration). By excluding data under soil moisture stress this bias is removed.

Q: P10, L10-1 - suggest rewording to “Daily mean values are calculated from the remaining time steps only where both modelled ”
A: Done.

Q: P10, L14 - why only use July here when the rest of the analysis is conducted for JJA?
A: Only one month (July) was chosen since it is easier to compare means of one month to reported values in the literature than mean values over several months.

Q: P10, L14-15 - why not use the same light level to define daylight as you used to filter the data previously?
A: For the hourly mean values the threshold of 100 Wm^{-2} is used to have a sharp cut-off of values with small light intensities where photosynthesis is little active and dew might bias the estimated G_c of FLUXNET. To calculate daily mean values such a restrictive boundary is not necessary since the early morning hours are not investigated separately.

Q: P10, L16 - suggest rewording to “..FR and for both modelled and FLUXNET-observed GPP”
A: Done.

Q: P10, L22-3 - suggest rewording to “1999). Reduced and oxidised nitrogen

deposition in wet and dry forms and ozone"
A: Done.

Q: P10, L25 - parentheses should only be around "2014b"
A: Done.

Q: P10, L25 - insert "and are" before"scaled back"
A: Done.

Q: P10, L27 - parentheses should only be around "2011"
A: Done.

Q: P10, L28 - square parentheses are not needed around CO2.
A: Skipped.

Q: P10, L28 - parentheses should only be around "2015"
A: Done.

Q: P10, L29-30 - Please check dates. If 1961-1970 is used as a spin-up shouldn't the simulation then start at either 1961 (repeating the first 10 years) or from 1971?
A: The transient simulation starts at 1961 and ends in 2011 (since the MTE period extends to 2011). Changed to '1961-2011'.

Q: P10, L32 - Please explain what an MTE product is.
A: MTE is a machine learning technique. Changed to: 'An up-scaled FLUXNET-MTE-product of GPP (Jung et al., 2011), using the machine learning technique: model tree ensembles (MTE),'

Q: P11, L2 - replace "Different" by "In contrast"
A: Done.

Q: P11, L3 - O3 should be subscript
A: Done.

Q: P11, L3-4 - Please explain for the non-specialist audience why the resistances result in a lower canopy concentration.
A: Changed to: 'Due to these resistances, the deposition of ozone to leaf-level is reduced, and the canopy O_3 concentration is lower than the atmospheric O_3 concentration.'

2.5 Results

Q: P11, L11 - what do the authors mean that they agree "within the standard deviations"? Are they stating that the data overlap? It would be better to demonstrate this goodness of fit with robust statistical analysis.
A: 'within the standard deviation' is substituted by 'well'. A table reporting the: 'Coefficient of determination (R^2) and Root Mean Square Error (RMSE) for GPP, canopy conductance (G_c), and latent heat fluxes (LE) for all sites, sites dominated by broadleaved trees, needle-leaved trees, C3 grass, and C3 grass except of the AT-Neu site (outlier).' is added to the Appendix and cited in section 3.1 Evaluation against daily eddy-covariance data '(see Appendix Tab. 2 for R^2 and RMSE values)'. Given the observational uncertainty, the model performance appears to be acceptable.

Q: P11, L13 - should read "very close, with only slight under-"
A: Changed.

Q: P12, L3 - remove extra ")" after 10 a
A: Done.

Q: P12, L5-6 - please give an example of site management that might result in such variability
A: Fertilidsation might strongly increase GPP. Mowing can change LAI strongly and through this impact estimated GPP and G_c. '(e.g. mowing and fertilisation)' is added.

Q: P12, L8 - why should LE be overestimated and GPP underestimated by OCN at broadleaved forest sites?
A: We can only speculate that a bias in the estimation of the FLUXNET LE might be the cause for this. It might also be possible that the observed water use efficiency (WUE) is not properly captured by OCN, what however

seems unlikely to be the major reason since GPP and G_c do not show such a bias when compared to each other.

Q: P12, L13 - what do the authors mean by “vary more widely”? Do they mean that there is a greater difference between modelled and measured values or that there is greater variability in the differences?
A: There is greater difference between modelled and measured values compared to the needle-leaved tree sites mentioned in the preceding sentence.

Q: P12, L14 - Do the means still lie within one standard deviation or not? Is there a tendency for the model to consistently under- or overestimate?
A: Changed to 'The modelled G_c at sites dominated by C3 grasses is in very good agreement to FLUXNET G_c with slightly overestimating G_c at 2 out of 3 sites except for the DE-Meh site, where means differ outside the standard deviation (see Appendix Fig. 10 b).'

Q: P12, L15-22 - move to SI
A: We would like to keep this paragraph included (like Referee 1), however we can move it to SI if demanded.

Q: P12, L23 - general comment regarding section 3.2: Do the reported “biases” in the diurnal cycles reflect those of the means? i.e. is GPP underestimated at the broadleaf site.
A: The biases are partly reflected by the hourly value. For instance the fact that the needle-leaved trees site matches observed values best. For the broadleaved trees GPP shows a bias towards underestimation by the daily mean values, however is overestimated on the site shown for the hourly values. The Gc shows a slight bias towards overestimated by the mean and is also overestimated by hourly values. There seems to be little benefit for the readers gain of knowledge to compare the exemplary site to the bias of the category so much in detail. A sentence to compare the general pattern of daily means and hourly values is added: 'Similar to the daily mean values (see Fig. 1 a,b) the mean hourly values show the best match of GPP and G_c for the needle-leaved tree site and stronger deviations for the sites covered by broadleaved trees and C3 grasses.'

Q: P12, L24 - diurnal profiles of which variables? State here
A: Done.

Q: P12, L32 - remove unnecessary parentheses after m and n.
A: Done.

Q: P12, L32 - should read: “with particularly good agreement”
A: Changed.

Q: P12, L32 - surely it’s more relevant that it is an evergreen needle-leaf forest that it is Finnish?
A: Changed to ’needle-leaved site’.

Q: P12, L34 - again, state the type of landcover at this site
A: ’Italian’ substituted by ’grassland’.

Q: P13, L1 - Again please explain what is meant by the means being within the standard deviation.
A: Changed to: ’The modelled hourly values fall in the range of the observed values.’

Q: P13, L2 - The maximum variability at CH-Oe1 seems to occur during the middle of the day
A: Yes, this fact was erased and changed. Changed from ’where the observed values became highly variable. ’ to ’where the observed values increase again.’

Q: P13, L3 - “whereas” is all one word
A: Changed.

Q: P13, L4 - what about the peak GC at the CH-Oe1 site? Is it also overestimated by the model?
A: Yes. Respective sentence changed to: ’and overestimates peak G_c at the CH-Oe1 site.’

Q: P13, L5 - “simulate” rather than “simulated”
A: Changed.

Q: P13, L5-6 - is this not a serious short-coming of the model water response parameterisation? I thought the midday depression in GC was a well observed response to water stress. Please comment on the likely implications for your results and conclusions?
A: The midday depression of G_c is a well observed phenomena and ought to be captured better by the model. However how strong the midday depression is and if it occurs at all is species and site specific. It does not occur for instance at the FI-Hyy site. The IT-Ro1 site shows that the model is at least in some cases able to capture important patterns like the midday depression of G_c. OCN however is a global model and not especially tuned for the specific sites such that the features of some sites will be captured better than others. Furthermore the observations at the CH-Oe1 site show very wide error bars, which also indicates the uncertainty in the observations! In times when G_c is underestimated the ozone uptake will also be underestimated and will result in a lower estimated damage. However since it is not a general pattern that the midday dip is not reproduced, this seems not to have a strong impact on the reported results and conclusion. One has to keep in mind that the modelling of ozone damage underlies many uncertainties as well as the observations against which the modelling results are evaluated.

Q: P13, L7-¿ Please either change the order of the panels in Figure 2 or the order of the text so that you are presenting the results of the panels in the order in which they appear.
A: Order in the text is changed.

Q: P13, L9- 15 - How is RC measured? or is it back-calculated from observed ET and LE? Please comment on the reliability of the observations.
A: R_c can be inferred from measurements by the eddy covariance technique (Coyle et al., 2009; Gerosa et al., 2004; Mikkelsen et al., 2004). The total deposition of ozone is calculated from the ozone concentration at measurement height and the fluxes measured by the eddy covariance technique (total ozone deposition). R_c can be inferred from the total deposition as the residual when subtracting R_a and R_b. Eddy covariance measurements and derived flux and conductance estimates are subject to a diverse set of random and systematic errors (Richardson et al., 2012). A lack of energy balance closure can cause underestimation of sensible and latent heat as well as an overestimation of available energy, with a mean bias of 20 % where the imbalance is greatest during nocturnal periods (Wilson et al., 2002). Since R_c is inferred from measured fluxes the calculation of R_c underlies the uncertainties of the flux measurements.

Q: P13, L9-15 - what are the implications of the model deviations from observations?
A: The main purpose of this evaluation is to show that our model produces realistic values for key ozone variables. The modelled values are within the range of observed values and show the expected diurnal pattern. Deviations from the values reported in the literature are expected since we neither model the specific sites nor the species. That also means that also the climate and ozone concentrations of the observations can not be reproduced by OCN which both have a major impact on the modelled ozone variables. Since the modelled values are within the observed range reported in the literature it can be assumed that our model works fairly well.

Q: P13, L15 - should read " observed which is slightly lower"
A: Changed.

Q: P13, L16 - the minimum velocities appear to be lower than this value for crops
A: I am not sure what 'for crops' refers to, since we do not model crops here. In case it is meant that for the CH-Oe1 (grassland site) site minimum V_g is lower than 0.002: Yes, it is approximately 0.0015 for CH-Oe1, however I think it is 'approximately 0.002 $\mathrm{m\,s^{-1}}$' when stating the mean minimum V_g for all three sites.

Q: P13, L18 - "barely" should read "barley"
A: Changed.

Q: P13, L16-20 - The modelled velocities at your crop site are well below these.
A: We do not model a crop site, the CH-Oe1 site is a grassland site. The crop values only indicate the observed range, since trees might also not be the best choice to compare with. Besides our modelled peak values of V_g are approximately 0.0055 $\mathrm{m\,s^{-1}}$ which in our notion compares well to observed ranges of 0.003-0.009 $\mathrm{m\,s^{-1}}$ at noon (Gerosa et al. 2004) for a barley field and approximately 0.006 $\mathrm{m\,s^{-1}}$ at noon for a wheat field (Tuovinen et al., 2004).

Q: P13, L20 - please rephrase to "The estimates for Hyytiälä also agree"

A: Changed to: 'The estimates for FI-Hyy also agree'.

Q: P13, L16-23 - It would be helpful if you compared the data site by site as before
A: This is done here, only that we start with the CH-Oe1 site, followed by FI-Hyy and last IT-Ro1. The reason for evaluating IT-Ro1 last is that for broadleaved trees we found only daily mean values to compare with, such that the actual diurnal cycle can not be properly evaluated. Hence it seems better not to start with this site.

Q: P13, L23 - Why is Vg so noisy for IT-Ro1?
A: V_g is determined by the total ozone uptake which is composed of a stomatal and a non-stomatal fraction. The noise in the stomatal component of the total uptake (F_{stC}) causes the noise in V_g. F_{stC} is determined by G_c and leave-level ozone concentrations. Since G_c shows not much noise it can be assumed that the day to day variability of the leave-level ozone concentration is the cause of the noise in F_{stC} and V_g.

Q: P13, L24 - Perhaps it is worth making the point that Vg is not zero because of non-stomatal deposition.
A: The sentence on P13, L24 deals not anymore with V_g but with F_{stC}. And F_{stC} is not zero during the night since a minimum conductance occur also during the night even though photosynthesis is zero.

Q: P13, L27-28 - Why is there such large variability in the afternoon at IT-Ro1? Is that another sign of water stress?
A: As already mentioned above: F_{stC} is determined by G_c and leave-level ozone concentrations. Since G_c shows not much noise it can be assumed that the day to day variability of the leave-level ozone concentration is the cause of the noise in F_{stC}.

Q: P12-13 - general comments: For Rc, Vg, FR, FStC: what are typical/expected profiles of these variables? Do we really only have observations at 1 or 2 times per day with which to assess model skill? How do these output data compare with estimates from other models? I would strongly recommend that much of the content here is moved to SI and/or presented in a table, with this section only highlighting a few key or interesting features.
A: The expected diurnal profiles are as modelled by OCN, with peak value

during the day for all variables except of R_c where maximum values are expected during the night. Hence the diurnal pattern is modelled appropriately. Certainly there are observations that do report on a high temporal resolution (Mikkelsen et al., 2004; Gerosa et al., 2004, 2003). However, we do not model the sites where the observations are conducted, it does thus not seem appropriate to compare details of model and data, especially since differences between species are high (see ranges of cited values for different species). In our notion it is interesting to show the diurnal pattern including the hourly standard deviation. It seems important to show that the diurnal pattern of the variables can be reproduced by the model and how this varies between the sites. Information about when standard deviation is typically high or low and how and why it is high for some variables would be skipped when condensing Fig. 2 into a table. For instance the fact that the high noise level for F_{stC} at IT-Ro1 can not be explain by noise in G_c is information that would get missing.

Q: P14, L2 - add a reminder in the parentheses that GCO3=GC/1.51
A: Changed to: '$G_c^{O_3} = \frac{G_c}{1.51}$'

Q: P14, L3 - Is this ratio essentially the proportion of deposition that is stomatal?
A: Yes.

Q: P14, L3-9 - Why have the authors chosen to report the 24-hour average for this variable and not for the others? Section 3.3 This section and the accompanying figure should be moved to SI, with only a few key headline findings included in the main text.
A: The 24-hour average is given for F_R since for instance in Cieslik (2004) the reported flux ratios are mean values (for diverse sites listed in a table) and the daily mean value in our graph should facilitate the comparison with this table. If this 24-hour mean value is a distraction to the reader it can be removed, otherwise we would like to keep it.
The included ozone deposition module is the key component for simulating ozone uptake and damage. Since it is done the first time to include such a detailed deposition model into a global terrestrial biosphere model it seems to be very important to show that this inclusion worked properly. That means that the results are fairly robust against the exact parametrisation (Fig. 4) but also that perturbations in one variable cause expected effects in related/depending variables (Fig. 3). Furthermore it seems quite important

to show which variables of the deposition scheme mainly impact the estimated ozone uptake and hence damage (Fig. 3).

Q: P14, L12 - replace “constraint” with “constrained”
A: Changed.

Q: P14, L13 - “boreal” would be a more useful descriptor than “Finnish”
A: Changed.

Q: P14, L13 - replace “except of” with “except for”
A: Done.

Q: P14, L14 - replace “describing” with “which describes”
A: Done.

Q: P14, L17 - replace “compared” with “relative”
A: Done.

Q: P14, L22 - insert “canopy conductance” before “GC”
A: Done.

Q: P14, L23 - replace “what causes” with “resulting in”
A: Done.

Q: P14, L24 - replace “compared”with “relative”
A: Done.

Q: P14, L25 - remove “changed values for”
A: If changed values would be removed it would sound as if only r_{ext} and G_c are important for the fluxes however this is not the case. The message is that r_{ext} and G_c need to be properly modelled because changes in their values impact the modelled fluxes. Thus we would like to keep the sentence unchanged.

Q: P14, L26 - explain the units (%/%)
A: '0.1 (%/%)' is substituted by '0.1 % due to a 1% change in the vari-

ables/parameters of the deposition scheme.'

Q:P14, L27 - remove "very" and "varying"
A: 'very' is removed. Varying is not removed since the message is that perturbations (variations) of r_{ext} and G_c little effect F_R. It is not the case that F_R is little affected by r_{ext} and G_c!

Q: P15, L1-2 - has this phenomena (the effect of needle-shedding on CUO) been evaluated?
A: I am not sure what is meant by 'if the phenomena has been evaluated'. As in our response to reviewer one, we believe that the use of f_{shed} has caused some confusion, and therefore we have replaced this with f_{new}. The CUO itself is only representative of what actually happens in the plant. Ozone does not actually accumulate in the plants. However, CUO is a substitute to estimate potential damage to the leaves/plant. It can be assumed that new grown leaves are healthy. Deciduous plants grow a complete set of new leaves each year and shed all damaged leaf at the end of the growing season and hence also shed the CUO. Evergreen plants keep their leaves for several years but if they would keep accumulating the CUO they would die since damage keep increasing. Hence it is reasonable to assume that if old/damaged leaves are shed also the fraction of CUO they took up will be shed too.

Q: P15, L6-7- what percentage is 250 gC/m2/yr?
A: The range of $\pm$ 250 $\mathrm{g\,C\,m^{-2}\,yr^{-1}}$ is skipped and substituted by the European mean deviation of OCN from MTE, since this seems to be more informative. The respective sentence is rewritten to: 'Simulated mean annual GPP for the years 1982-2011 shows in general good agreement with an independent estimate of GPP based on up scaled eddy-covariance measurements (MTE, see Section 2.5), with OCN on average underestimating GPP by 16 % (European mean).'

Q: P15, L8 - remove "to this acceptable agreement"
A: Done.

Q: P15, L9 Again what percentage is 400 to 900 gC/m2/yr?
A: Added: '(58 % overestimation on average)'

Q: P15, L12-3 - It also makes it difficult to assess the reliability of the model!
A: Yes, because there might be no reliable source to compare with.

Q: P15, L16 - Please explain how N limitation can lead to overestimation of GPP
A: In the North OCN underestimates GPP compared to MTE not overestimates. Changed to : 'North of $60°N$ OCN has the tendency to produce lower estimates of GPP'. The underestimation might result from N limitation.

Q: P15, L20 - Fig. 6d does not show GPP. Should this read Fig. 5a?
A: Yes, changed to 5a.

Q: P15, L23-4 - Is it not to be expected that AOT40 closely follows absolute ozone concentrations?
A: Yes, it is expected and it is good to be able to compare the AOT40 pattern to the CUO pattern.

Q: P15, L26 - replace "averaged" with "ranged from 60 to 120"
A: Changed.

Q: P15, L27 - move "(Fig 7 a)" to between "Europe" and "and"
A: Done.

Q: P15, L28 - "larger" should read "large"
A: Changed.

Q: P15, L28 - does this refer to Fig. 7b?
A: Yes. '(Fig. 7b)' is inserted at the end of the sentence.

Q: P15, L29 - suggest rewording: "with high cover of C4 PFTs, e.g. Black Sea area (see Appendix 12 a,b)."
A: Done. The graph Appendix 12 is skipped due to it's minor value in explaining observed results.

Q: P15, L30-1 -suggest rewording: "where productivity is low and stomatal O3 uptake reduced by low O3 concentrations or drought control on stomatal fluxes respectively."
A: Changed to : 'where productivity is low and stomatal O_3 uptake is re-

duced by e.g. low O_3 concentrations or drought control on stomatal fluxes respectively.'

Q: P15, L31-2 - suggest removing the sentence beginning: "Slight increases or strong decreases"
A: We would like to keep the sentence since it puts the displayed outliers, the positive damage, and the strongest fractional damage into context.

Q: P15, L32 - "increases" should read "increase"
A: Changed.

Q: P16, L3 - replace "by" with "of"
A: Changed.

Q: P16,L4 - insert "Fig. " before "7 c"
A: Done.

Q: P16, L4 - insert "of transpiration" after "3-4%"
A: Done, and European changed to Europe.

Q: P16, L4 - remove "to" before "4-6%"
A: Done.

Q: Q: P16, L5 - insert "relative" before "reductions"
A: Done.

Q: P16, L7 - should read "Black Sea"
A: Changed.

Q: P16, L8 - insert "Fig." before "7 d" and replace "They are" with "These are"
A: Done.

Q: P16, L10 - please explain why a reduction in transpiration matters.
A: Changes in transpiration might impact run-off and surface cooling.

Q: P16, L15 - suggest rewording: "CUO1.6 increases more strongly by 35% "

A: This sentence has been removed since the flux threshold and hence $CUO_5^{1.6}$ has been removed.

Q: P16, L18-9 - It seems to me that in this study simulation D is effectively the base case and D-STO and ATM are sensitivity tests. It would therefore make more sense to swap panels a and c in Figure 9. Furthermore, it seems to me that this is the real headline message of this study - that the ozone deposition scheme substantially alters estimates of impacts. this needs far more emphasis (it is currently hidden by the wealth of detail in the rest of this discussion) and Figure 9 should include further panels showing how CUO changes (see below).
A: We put the ATM case first because this is the common approach if no deposition model is included (base case). The D-STO model here accounts for impacts of stomatal uptake on leaf-level O_3 concentration but still does not account for the non-stomatal fraction and can be seen as an intermediate approach. Our standard scheme accounting for both stomatal and non-stomatal uptake on leaf-level O_3 concentrations is the one that comes last such that complexity increases from panel a to c. We would like to keep the present order but can change it if it hampers the understanding of the graph. Furthermore in our notion the general pattern of a decrease in CUO from ATM to D-STO and D is easy to observe from the present graph. Additional panels showing the exact values seem to add little gain of knowledge. Thus we would like to not include them.
To highlight the importance of the deposition scheme more we changed in the Abstract: 'When applied at the European scale, we find that the added complexity of the ozone uptake simulation substantially affects simulated ozone uptake ' to 'When applied at the European scale, we find that accounting for stomatal and non-stomatal uptake substantially affects simulated ozone uptake, ...'
Furthermore we incorporate the importance of the deposition scheme into section 4.1 (Atmosphere-leaf transport of ozone).

2.6 Discussion

Q: This section seems redundant. Much of it is either already stated in the Results section or could be moved to form part of a more robust conclusion.

A: We would like to keep the conclusion short stating briefly the main insights from our work. We reduce redundancy between the results and discussion section and restructured the discussion section to have only 2 subsections, '4.1 Atmosphere-leaf transport of ozone, and '4.2 Estimating vegetation damage from ozone uptake'.

Q: P16, L24-5 - replace "with the aim" with "in order to"
A: Done.

Q: P16, L25 - replace "effect to net" to "effect on net"
A: Done.

Q: P16, L25 - remove "the" before "regional"
A: Done.

Q: P16, L28 - replace "assuming" with "the assumption"
A: Respective sentence is omitted.

Q: P16, L28 - replace "would be identical" with "is identical"
A: Respective sentence is omitted.

Q: P16, L29 - replace "in 45m" with "at 45m"
A: Respective sentence is omitted.

Q: P16, L30-1 - suggest rewording: "and deposition variables i.e. calculated ozone uptake"
A: Respective sentence is omitted.

Q: P16, L32 - P17, L2 - suggest rewriting: "Our sensitivity analysis does show that a correct estimate of canopy conductance is crucial for calculating plant ozone uptake. We find that the model produces reasonable estimates"
A: The respective sentences are omitted in this paragraph. The first sentence ('Our sensitivity analysis ...') is included in subsection 4.1.

Q: P17, L2 - replace "a range of" with "some"
A: Done (the respective half sentence is moved to subsection 4.2).

Q: P17, L7-8 - suggest rewriting: “Reliable estimates of surface ozone concentrations are also essential for calculating canopy ozone uptake FstC”
A: Changed to: ’Reliable estimates of surface ozone concentrations – besides a reliable estimate of G_c – are also essential for calculating canopy ozone uptake (F_{stC}).’

Q: LP17, 8-9 - suggest rewriting: “airspace due to biogenic volatile organic compounds (BVOCS) emitted by vegetation is (at least partly) implicitly included in the”
A: We would like to skip the respective sentence since after rewriting the discussion it does not fit anymore.

Q: P17, L9-10 - Does this mean there is a degree of double accounting?
A: No. EMEP accounts for BVOCS (to a certain extend) in the calculation of the O_3 concentration in 45 m height. OCN to a certain extend accounts for BVOCS in the calculation of the leaf-level O_3 concentration.

Q: P17, L11 - suggest “performance” or “efficacy” in place of “functionality”
A: Respective sentence is omitted.

Q: P17, L15 - suggest combining these to form a single sentence: “changes in GC emphasising the importance”
A: Respective sentences are omitted.

Q: P17, L15-16 - How can reliable estimates be obtained?
A: The respective sentence is omitted. It is of course not possible to simulate the true ozone uptake. However when variables determining ozone uptake are simulated in a reasonable range compared to observations one might call also the calculated uptake reliable (considering the uncertainty in both model simulations and observations). It will anyhow still be an estimate.

Q: P17, L18 - replace ”indicates”with “indicate”
A: Respective sentence is omitted.

Q: P17, L26 - replace “impose” with “introduce”
A: Done.

Q: P17, L29 - replace “suitable” with “well able”
A: Respective sentence is omitted.

Q: P17, L30 - remove first occurrence of “finding” and replace “encourages” with “supports”
A: The respective sentence does not anymore exist. “encourages” is replaced by “supports” in a similar sentence.

Q: P18, L2 - reword: “Estimates of the regional damage to annual average”
A: Done.

Q: P18, L2 - make clear this is transpiration rather than temperature (I assume)
A: Transpiration is spelled out.

Q: P18, L2-3 - remove “the period of the years”
A: Done.

Q: P18, L3 - replace “lower” with “low” and “previous” with “previously”
A: Changed to ’lower than previously reported’.

Q: P18, L3 - should read ”Meta-analyses” and “an 11¸%”
A: Changed.

Q: P18, L6 - should read “Land Model”
A: Changed.

Q: P18, L7 - reword: “..transpiration have been estimated as 5-20 % for Europe and 2.2% globally ”
A: Done.

Q: P18, L9 - reword: “plant types. Damage was only related to cumulative ozone uptake for one plant type with a very small slope”
A: Changed.

Q: P18, L9 - please explain the real-world meaning of a small slope.
A: The higher the slope the more damage occurs per accumulated ozone. The respective sentence is extended to: 'with a very small slope and hence little increase in damage due to increases in cumulative O_3 uptake.'

Q: P18, L14 - use "discrepancies" or "differences" rather than "deviations"
A: Changed to 'discrepancies'.

Q: P18, L14-15 - replace "the usage of very different" with "differences in" and then remove "different", "differing" and "non-identical"
A: Done.

Q: P18, L16 - replace "differences in simulating" with "simulation of"
A: Done.

Q: P18, L17 - reword: "The key difference from the previous study is our use of the ozone"
A: Changed to 'A key difference from the previous study is our use of the use of the ozone'.

Q: P18, L17 - remove "included in our study"
A: Done.

Q: P18, L21 - remove "the" before "non-stomatal"
A: Done.

Q: P18, L22 - should read "To obtain as accurate as possible an estimate "
A: Done.

Q: P18, L23 - replace "it's" with "their"
A: Done.

Q: P18, L24 - replace "considered" with "accounted for"
A: Done.

Q: P18, L25 - suggest moving "(possibly PFT specific)" to come before "flux

threshold”
A: Done.

Q: P18, L25 - “it’s” should read “its”
A: Changed.

Q: p18, L25 - should the “Y” in “CUOY” be a subscript?
A: No, similar to AOTX the Y is not a subscript.

Q: P18, L32 - insert “see” before “LRTAP”
A: The respective sentence is omitted.

Q: P18, L33 - replace “but only” with “there are” and “exists for” with “of”
A: Done.

Q: P19, L2-4 - What is the implication of this disadvantage to the findings reported here?
A: Two sentences explaining the implications are added: ’This might be an important factor explaining the lower ozone damage estimates of OCN compared to other terrestrial biosphere models. An evaluation of the different proposed damage functions implemented in terrestrial biosphere models (e.g. Wittig et al. (2007); Lombardozzi et al. (2015); Sitch et al. (2007)) is necessary to elucidate which are able to e.g. reproduce observed patterns of biomass damage and hence might be suitable to predict regional or global damage estimates.’

Q: P19, L5 - replace “damage estimates” with “relationships”
A: Done.

Q: P19, L6 - replace “estimates” with “metrics”
A: Done.

Q: P19, L13 - replace “should be regarded too” with “also requires further analysis”
A: Done.

2.7 Conclusion

Q: This section needs to be substantially expanded. The authors would also do well to identify (even using bullet points if necessary) the key findings of their study and the implications for the land surface and atmosphere research communities. Much of Section 4 could be distilled and included in the Conclusion section.
A: As mentioned above we would like to keep the conclusion short stating briefly the main insights from our work. The Discussion section was shortened and restructured to remove redundancy.

Q: P19, L20-1 - replace "to generally consider" with "that"
A: Done.

Q: P19, L21 - reword: "non-stomatal ozone uptake is routinely included in model assessments of ozone damage" and remove "estimate" after "better"
A: The rewording is done. The 'estimate' is not removed since it is an estimate.

Q: P19, L22 - remove "used"
A: Done.

Q: P19, L23 - insert "used here" after "scheme"
A: Done.

Q: P19, L23 - reword: "importance of reliable modelling of canopy conductances as well as realistic"
A: Done.

Q: P19, L24 - insert "as" before "accurate"
A: Done.

Q: P19, L26 - remove "Desirable are"
A: Done.

Q: P19, L27 - insert "are also desirable" after "types"

A: Done.

Q: P19, L29 - replace “regarded” with “considered”
A: Done.

Q: P19, L29 - insert “,” after “thresholds”
A: Done.

2.8 Appendix

Q: A P20, L1 - capitalise “Aerodynamic Resistance” and remove “(Appendix material)”
A: Done.

Q: P20, L3 - remove “,” after “heights” and replace “This data is” with “These data are”
A: Done.

Q: P20, L4 - replace “in 45m height” with “at 45m”
A: Done.

Q: P20, L7 - what does U10 mean? If at 10m, why is this an appropriate height at which to calculate u*?
A: 'u_{10}' is now explained as 'from the wind speed at 10 m height (u_{10})'. u_* is assumed to be constant within the surface near atmosphere layer. Since OCN is run offline (not coupled to a climate model) the necessary variables to calculate the friction velocity (e.g. wind speed and aerodynamic resistance) are only available in 10 m height.

Q: P20, L9 - replace “in 45m height” with “at 45m”
A: Done.

Q:Appendix B P20, L21 - Why not use ORCHIDEE to calculate biogenic emissions?
A: OCN was developed from a ORCHIDEE version where biogenic emissions

are not calculated. Modules of the current ORCHIDEE can not easily be included in OCN.

Q: P20, L22 - remove “NO from”
A: Done.

Q: P20, L24 - Volcanic emissions of what? Which compounds?
A: Volcanic emissions of SO_2 are meant. Respective sentence is changed to: 'Volcanic emissions of sulfur dioxide (SO_2) were set to a constant value from the year 2010.'

2.9 References

Please check references carefully.

Q: Tuovinen et al., 2004a and 2004b are the same paper Tuovinen et al., 2009a and 2009b are the same paper
A: This issue is resolved.

2.10 Figures

Q: Throughout - I would suggest that rainbow scale is not the most effective and that limited color graduated scales would be easier to interpret.
A: The color palettes are changes from rainbow to restricted color gradients (palettes from ColorBrewer 2.0).

Q: Fig. 1 Panel (d) - Again, why choose a non-varying measure of LAI (i.e. point samples) rather than MODIS or similar, particularly as you comment on the validity of these measurements for the specific time period modelled? Panel (d) - In its present form this is not a useful panel and I would suggest that it is removed or moved to SI. It distracts from the good fit the model shows to other (more important) variables. Caption - line 4 should read “which are based on point”
A: MODIS data are also subject to a considerable amount of uncertainty. Furthermore the resolution of MODIS data is an additional source of uncertainty. Using observation directly from the site in question seemed to be the most reliable source. We would like to keep panel d) however can remove it

or move it to SI when really requested. Caption is changed to “which are based on point”.

Q: Fig. 2 x-axis scale - Hours should have a 4-hour or 6-hour scale, not 5. Please state explicitly whether this is local time or UTC. y-axis scale - As the scale is the same across each row I would suggest only one axis scale is required. y-axis scale - for variables that can be negative please add a dashed horizontal line to indicate 0.0; otherwise the axes should cross at zero.
A: X-axis is changed to 3 hour scale (3h - 21h). The time is local time (added to figure caption). Y-axis: the separate scales for each plot secure the readability of the plot. Excluding all but the one in the left column would make it hard to see which values the variable in the other columns take. The minimum for the Y-axes is set to zero.

Q: Fig. 3 scales - please define the scales used in Fig 3 more carefully, either here in the caption or in the appropriate place in the main text. Fig. 4 This figure should be SI. In addition, it is virtually unreadable. I had to view at 600% zoom to make out the yellow and red lines
A: We would like to keep the figure in the main text since it illustrates the robustness of the included deposition module against the exact parameterisation. To make it better readable we skipped the interquartile-range (dark grey area) and stretched the plot. The red and yellow line lie on top of each other. The red line is dashed to show that the yellow line lies directly underneath. Furthermore we added a sentence in the text to explain this fact: 'For all four variables the unperturbed model and the ensemble mean lie on top of each other (see dashed red and yellow line in Fig. 4 a-d).'

Q: Fig. 5 scales - don't use the same colour scales for both absolute values and changes; changes are best shown on blue-red scales. Use e.g. green scale for crop cover.
A: Done.

Q: Fig. 7 scale - please improve the scales; I suggest using a graduated single or limited colour range. panel labels - please use more descriptive panel captions (not just “damage”)
A: The color palette is changed.
Regarding the panel label: Since there is only restricted space within the graph corner we choose to state only that damage is plotted and the respective unit which indicates which variable is plotted. In the figure caption it

is also stated what is plotted where. To us this seems quite explanatory however we can add also the plotted variable in the corner of the plot what however might overload it.

Q: Fig. 9 To me, this is the KEY figure in this paper. I suggest that you add panels showing changes in CUO from D to D-STO and ATM respectively (giving a 5 panel plot)
A: In our notion the general pattern of a decrease in CUO from ATM to D-STO and D is easy to observe from the present graph. Additional panels showing the exact values seem to add little gain of knowledge. Thus we would like to not include them.

Development and evaluation of an ozone deposition scheme for coupling to a terrestrial biosphere model

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Abstract.

Ozone (O_3) is a toxic air pollutant that can damage plant leaves and substantially affect the plant's gross primary production (GPP) and health. Realistic estimates of the effects of tropospheric anthropogenic ~~ozone~~ O_3 on GPP are thus potentially important to assess the strength of the terrestrial biosphere as a carbon sink. To better understand the impact of ozone damage on the terrestrial carbon cycle, we developed a module to estimate ~~ozone~~ O_3 uptake and damage of plants for ~~the state of the art~~ a state-of-the-art global terrestrial biosphere model called OCN. Our approach accounts for ozone damage by calculating (a) ~~ozone transport from the free troposphere~~ O_3 transport from 45 m height to leaf level, (b) ~~ozone~~ O_3 flux into the leaf, and (c) ozone damage of photosynthesis as a function of the accumulated ~~ozone~~ O_3 uptake over the life-time of a leaf.

A comparison of modelled canopy conductance, GPP, and latent heat to FLUXNET data across European forest and grassland sites shows a general good performance of OCN including ozone damage. In comparison to literature values, we demonstrate that the new model version produces realistic ~~stomatal flux ratios as well as ozone surface resistancesand deposition velocities~~O_3 surface resistances, O_3 deposition velocities, and stomatal to total O_3 flux ratios. A sensitivity study reveals that key metrics of the air-to-leaf ~~ozone transport and ozone~~ O_3 transport and O_3 deposition, in particular the stomatal ~~ozone update~~ O_3 uptake, are reasonably robust against uncertainty in the underlying parameterisation of the deposition scheme. ~~Correctly~~ Nevertheless, correctly estimating canopy conductance plays a pivotal role in the estimate of cumulative ~~ozone uptake.~~

~~When applied at the European scale, we find that the added complexity of the ozone uptake simulation~~ O_3 uptake. We further find that accounting for stomatal and non-stomatal uptake processes substantially affects simulated ~~ozone~~ plant O_3 uptake and accumulation, because aerodynamic resistance and non-stomatal ~~ozone~~ O_3 destruction reduce the predicted ~~ozone concentrationsoutside the leaves~~leaf-level O_3 concentrations. Ozone impacts on GPP and transpiration in a Europe-wide simulation indicate that tropospheric ~~ozone~~ O_3 impacts the regional carbon and water cycling less than expected from previous studies. This study presents a first step towards the integration of atmospheric chemistry and ecosystem dynamics modelling, which would allow to assess the wider feedbacks between vegetation ozone uptake and tropospheric ozone burden.

1 Introduction

Tropospheric ozone (O_3) is a highly reactive and toxic gas. It enters the plants mainly through the stomata of the leaf, where it forms reactive oxygen species (ROS) which have the potential to damage the leaf. While leaves possess physiological pathways to produce compounds like ascorbate and polyamines, which help to neutralise the oxidising power of ROS (Kronfuß et al., 1998; Kangasjärvi et al., 1994; Tausz et al., 2007), ozone injury may occur when the leaf's anti-oxidant system becomes overwhelmed (Wieser and Matyssek, 2007).

In Western Europe, tropospheric ~~ozone~~ O_3 levels have increased approximately by a factor ~~of~~ 2 to 5 from pre-industrial values to the 1990s (Cooper et al., 2014; Marenco et al., 1994; Staehelin et al., 1994) (although the low values at the start of this period are very uncertain) and approximately doubled between 1950 and 1990s in the northern hemisphere (Parrish et al., 2012; Cooper et al., 2014). The major causes for this increased O_3 formation is the increased emission of O_3 precursor trace gases such as NO_x and CO, primarily from combustion sources, and methane emissions from agriculture and industry (Fusco and Logan, 2003; Vingarzan, 2004). For instance, in Western Europe, NO_x emissions have risen by a factor of 4.5 between 1955 and 1985 (Staehelin et al., 1994). In addition, downward transport of O_3 from the stratosphere to the troposphere (Vingarzan, 2004; Young et al., 2013) and intercontinental transport ~~from polluted to less polluted areas~~ (Vingarzan, 2004; Jenkin, 2008; Fiore et al., 2009) can increase local and regional O_3 concentrations.

A commonly observed consequence of elevated levels of ~~ozone~~ O_3 exposure is a decline in net photosynthesis (Morgan et al., 2003; Wittig et al., 2007), which may result from the damage of the photosynthetic apparatus or increased respiration due to the production of defence compounds and investments in injury repair (Wieser and Matyssek, 2007; Ainsworth et al., 2012). The reduction in net photosynthesis results in reduced growth and hence a reduced leaf area, and plant biomass (Morgan et al., 2003; Lombardozzi et al., 2013; Wittig et al., 2009). The tight coupling between photosynthesis and stomatal conductance further affects canopy conductance, and thereby transpiration rates (Morgan et al., 2003; Wittig et al., 2009; Lombardozzi et al., 2013), likely affecting the ecosystem water balance.

Due to its phytotoxic effect, elevated O_3 levels as a consequence of anthropogenic air pollution may affect the land carbon cycle, and potentially reduce the net land carbon uptake capacity (Sitch et al., 2007; Arneth et al., 2010; Simpson et al., 2014a), which currently corresponds to about a quarter of the anthropogenic fossil fuel emissions as a ~~consequence~~ result of a sustained imbalance between photosynthetic carbon uptake and carbon loss through respiration and disturbance processes (Le Quéré et al., 2015). However, the ~~extend~~ extent to which O_3 affects plant health regionally and thereby alters terrestrial biogeochemistry and the terrestrial water balance is still subject of large uncertainty (Simpson et al., 2014a).

A number of O_3 exposure indices have been proposed to assess the potential detrimental effect of tropospheric O_3 on the plants (LRTAP-Convention, 2010; Mills et al., 2011b). ~~A widely used example~~ The initial standard tool of these indices is the concentration-based AOTX $[\mathrm{ppb\ h}]$ (accumulated O_3 concentration over a threshold of X ppb), which relates the free-air O_3 concentration to observed plant damage. Models assessing ozone damage to gross or net primary production based on AOTX have been used for many years and indicate that substantial reduction in plant growth and carbon sequestration occurs globally and may reach reductions of more than 40 % at O_3 hot spots (Felzer et al., 2004, 2005; Ren et al., 2011; Anav et al., 2011).

A significant caveat of concentration-based assessments of ozone toxicity effects is that species ~~and their regional provenances~~ differ vastly in their canopy conductance as well as regional provenances of one species. Stomatal control of the leaf gas exchange regulates photosynthesis, and varies inter alia with plant specific photosynthetic capacity and intrinsic water-use efficiency of photosynthesis, phenology, as well as environmental factors such as incident light, atmospheric vapour pressure deficit (VPD), air temperature. The consequent differences in stomatal conductance implies that the actual ~~ozone~~ O_3 dose, and thus the level of ozone-related damage, differs between species exposed to similar atmospheric O_3 concentrations (Wieser and Havranek, 1995). The ~~ozone~~ O_3 dose, that is the integral of the instantaneous O_3 stomatal flux over a given period of time, has been observed to strongly correlate with the amount of injury of a plant, suggesting that plants with higher stomatal conductance are subject to higher doses and hence more susceptible to injury (Reich, 1987; Wittig et al., 2009).

Accounting for the ~~ozone~~ O_3 dose rather than the O_3 exposure in assessments of ozone damage results in diverging regional patterns of ozone damage, as regions with the highest exposure (O_3 concentrations) do not always coincide with regions of high uptake (Emberson et al., 2000; Mills et al., 2011a; Simpson et al., 2007). Regions with low AOT40 (AOTX above a threshold of 40 ppb) values might show moderate to high values of O_3 uptake because the flux approach accounts for climatic conditions that enable high stomatal conductances and hence high values of O_3 uptake (Mills et al., 2011a). Observed ozone damage in the field seems to be better correlated to flux-based risk assessment compared to concentration based methods (Mills et al., 2011a) . Following this the LRTAP Convention recommends flux based methods as the preferred tool for risk assessment (LRTAP-Convention, 2010) .

When calculating the O_3 uptake into the plants, it is important to consider that stomatal uptake is not the only surface sink of ~~ozone. Ozone~~ O_3. O_3 destruction also occurs at non-stomatal surfaces such as the leafs cuticle and soil surface. The stomatal flux represents approximately half of the total O_3 flux to the surface (Gerosa et al., 2004; Fowler et al., 2009; Cieslik, 2004; Simpson et al., 2003). Accounting for this non-stomatal O_3 deposition reduces the amount of O_3 uptake into the plants by reducing the surface O_3 concentration (Tuovinen et al., 2009) and thus has the potential to affect flux-based ozone damage estimates.

A further challenge in estimating plant damage related to ~~ozone~~ O_3 uptake is that plants differ in their ability to remove any ROS from the leaf before damage of leaf cellular organs is incurred (Luwe and Heber, 1995). Conceptionally, one can describe the capacity as a plant-specific O_3 dose, with which the anti-oxidant system of the leaves can cope such that no damage is observed (Musselman et al., 2006). The production of defence compounds increases respiration costs and following this reduces net primary production what may result in reduced growth and biomass (Ainsworth et al., 2012) . Ozone damage is only incurred, once the O_3 flux into the leaf exceeds this dose. A commonly used index to assess flux-based damage to plants is the PODY [Phytotoxic Ozone Dose, $\mathrm{nmol\,m^{-2}\,s^{-1}}$], which gives the accumulated ~~ozone~~ O_3 flux above a threshold of Y $\mathrm{nmol\,m^{-2}\,s^{-1}}$ for all daylight hours and a given time period. Common threshold values for PODY range from 1-6 $\mathrm{nmol\,m^{-2}\,s^{-1}}$ (Pleijel et al., 2007; LRTAP-Convention, 2010; Mills et al., 2011b), depending on the specific species sensitivity to O_3.

Only a few terrestrial biosphere models have adopted the flux approach to relate ~~ozone~~ O_3 exposure to plant damage and thus estimate ~~ozone~~ O_3 induced reductions in terrestrial carbon sequestration in a process-based manner. Sitch et al. (2007)

developed a version of the JULES model in which stomatal ~~ozone~~ O_3 uptake directly affects net primary production (NPP), thereby ignoring the effect of reduced photosynthesis under elevated levels of O_3 on water fluxes. Lombardozzi et al. (2015) proposed a revised version of the CLM model, in which O_3 imposes fixed reductions to net photosynthesis ~~independent of~~ for two out of three modelled plant types. Atmospheric O_3 concentrations and the amount of cumulated O_3 uptake ~~for two out of three modelled plant types~~directly affect net photosynthesis only for one plant type.

In this paper, we present a new, globally applicable model to calculate O_3 uptake and damage in a process-oriented manner, coupled to the terrestrial energy, water, carbon and nitrogen budget of the OCN terrestrial biosphere model (Zaehle and Friend, 2010).

In this model, the canopy O_3 abundance is calculated using aerodynamic resistance and surface resistances to soil surface, vegetation surfaces and stomatal cavities to take account of non-stomatal O_3 destruction. Canopy O_3 abundance is used to simulate stomatal O_3 uptake given instantaneous values of net photosynthesis and stomatal conductance. ~~Ozone~~ O_3 uptake and its effect on net photosynthesis is then calculated based on an extensive meta-analysis across 28 tree species by Wittig et al. (2007) considering the ability of plants to detoxify a proportion of the O_3 dose (Sitch et al., 2007).

We first give a detailed overview of the ozone scheme (Section 2.1); evaluate modelled gross primary production (GPP), canopy conductance, latent heat fluxes and LAI against data from the FLUXNET database (Baldocchi et al., 2001) to test the ability of the model to simulate observed values of key components affecting calculate O_3 uptake (Section 3.1); evaluate the simulated ~~ozone~~ O_3 metrics against reported values in the literature (Section 3.2); provide a sensitivity analysis of ~~the~~ critical variables and parameters of the deposition model to evaluate the reliability of simulated values of O_3 uptake (Section 3.3); give an estimate of the effect of the present-day O_3 burden on European GPP and transpiration(Section 3.4); and estimate the impact of using the O_3 deposition scheme on ~~uptake and accumulation~~ O_3 uptake and cumulated uptake (Section 3.5).

2 Methods

We developed an ozone deposition and leaf-uptake module for the terrestrial biosphere model OCN (Zaehle and Friend, 2010). OCN is an extension of the land-surface-scheme ORCHIDEE (Krinner et al., 2005), ~~which~~ and simulates the terrestrial coupled carbon, nitrogen (N) and water cycles for twelve plant functional types driven by climate data, atmospheric composition (N deposition, as well as atmospheric CO_2 and O_3 burden), and land use information (land cover and fertiliser application).

In OCN net photosynthesis is calculated for shaded and sun-lit leaves in a multi-layer canopy with up to 20 layers (each with a thickness of up to 0.5 leaf area index) following a modified Farquhar-scheme and considering the light profiles of diffuse and direct radiation (Zaehle and Friend, 2010) . Photosynthetic capacity depends on leaf nitrogen concentration and leaf area, which are both affected by ecosystem available N. Increases in leaf nitrogen content enable higher net photosynthesis and higher stomatal conductance per unit leaf area. This in turn affects transpiration as well as ~~ozone~~ O_3 uptake and ozone damage estimates. Leaf N is highest in the top canopy and monotonically decreases with increasing canopy depth. Following this, stomatal conductance and O_3 uptake is generally highest in the upper canopy and lowest in the bottom of the canopy.

The ~~ozone~~ O_3 and N-deposition data used for this study are provided by the EMEP MSC-W (European Monitoring and Evaluation Programme Meteorological Synthesising Centre - West) chemical transport model (CTM) (Simpson et al., 2012). The ~~ozone~~ O_3 flux and deposition modules used in the EMEP model are ~~rather~~ advanced compared to most CTMs, and have been documented in a number of papers ~~(Emberson et al., 2001; ?; ?; ?; ?; Klingberg et al., 2008)~~ (Emberson et al., 2001; Tuovinen et al., 2 The ozone deposition scheme for OCN is adapted from the model used by ~~the Meteorological Synthesizing Centre - West of the European Monitoring and Evaluation Programme (~~EMEP MSC-W ~~)~~ (Simpson et al., 2012) to fit the land surface characteristics and process descriptions of the ORCHIDEE model. The leaf-level ozone concentrations computed by EMEP can not directly be used by OCN, since EMEP and OCN differ in a number of properties, as for instance in the number of simulated plant functional types, and importantly their ecophysiological process representation. Both models differ in the simulation of various ecosystem processes (e.g. phenology, canopy processes, biogeochemical cycles, and vegetation dynamics, which are more explicitly represented in OCN), which in sum impact stomatal and non-stomatal ozone deposition and through this the leaf-level ozone concentration. A possible further development of the new OCN is the coupling to a CTM, to allow for a consistent simulation of tropospheric O_3 burden and vegetation O_3 uptake.

2.1 Ozone module

The ozone deposition scheme calculates ~~ozone~~ O_3 deposition to the leaf surface from the free atmosphere, represented by the O_3 concentration at the lowest level of the atmospheric chemistry transport model (CTM), taken to be at 45 m above the surface. The total O_3 dry deposition flux (F_g) to the ground surface is calculated as

$$F_g = V_g \chi_{atm}^{O_3} \tag{1}$$

where $\chi_{atm}^{O_3}$ is the O_3 concentration ~~in~~ at 45 m ~~height~~ and V_g is the deposition velocity at that height. In OCN V_g is taken to be dependent on the aerodynamic resistance (R_a), canopy-scale quasi-laminar layer resistance (R_b) and the compound surface resistance (R_c) to ~~ozone~~ O_3 deposition.

$$V_g = \frac{1}{R_a + R_b + R_c}. \tag{2}$$

R_b is calculated from the friction velocity (u_*) as

$$R_b = \frac{6}{u_*}. \tag{3}$$

The R_a ~~in~~ between 45~~m height~~ m height and the canopy is not computed by OCN and is inferred from the logarithmic wind profile (for more details see Appendix A). R_c is calculated as the sum of the parallel resistances to stomatal/canopy $(1/G_c^{O_3})$ and non-stomatal ~~ozone~~ O_3 uptake $(1/G_{ns})$ (Simpson et al., 2012, eq. 55)

$$R_c = \frac{1}{G_c^{O_3} + G_{ns}}. \tag{4}$$

The stomatal conductance to ~~ozone~~ O_3 $G_{st}^{O_3}$ $(\mathrm{m\,s^{-1}})$ is computed by OCN (Zaehle and Friend, 2010) as:

$$G_{st}^{O_3} = g_1 \frac{f(\Theta) f(q_{air}) f(C_i) f(height) A_{n,sat}}{1.51} \tag{5}$$

where $G_{st}^{O_3}$ is calculated as a function of net photosynthesis at saturating C_i $(A_{n,sat})$ where g_1 is the intrinsic slope between A_n and G_{st}. It further depends on a number of scalars to account for the effect of soil moisture $(f(\Theta))$, water transport limitation with canopy height $(f(height))$, and atmospheric drought $(f_(q_{air}))$, as well as an empirical non-linear sensitivity to the ~~leafs internal~~ internal leaf CO_2 concentration $(f(C_i))$, all as described in ~~(Friend and Kiang, 2005)~~ Friend and Kiang (2005) . The factor 1.51 accounts for the different diffusivity of ~~ozone to~~ O_3 from water vapour (Massman, 1998). The canopy conductance to ~~ozone~~ O_3 $G_c^{O_3}$ is calculated by summing the $G_{st}^{O_3}$ of all canopy layers. To yield reasonable conductance values in OCN compared to FLUXNET data (see Sect. 3.1), the original intrinsic slope between A_n and G_c called α in Friend and Kiang (2005) is adapted such that $g_1 = 0.7\alpha$.

The non-stomatal conductance G_{ns} follows the EMEP approach (Simpson et al., 2012, eq. 60) and represents the O_3 fluxes between canopy air space and surfaces other than the stomatal cavities. The model accounts for ~~ozone~~ O_3 destruction on the leaf surface (r_{ext}), within-canopy resistance to ~~ozone~~ O_3 transport (R_{inc}), and ground surface resistance (R_{gs})

$$G_{ns} = \frac{SAI}{r_{ext}} + \frac{1}{R_{inc} + R_{gs}} \tag{6}$$

where the surface area index SAI is equal to the leaf area index LAI for herbaceous PFTs (grasses and crops), and $SAI = LAI + 1$ for tree PFTs according to Simpson et al. (2012), to account for ~~ozone~~ O_3 destruction on branches and stem. Unlike EMEP, we do not apply a day of the growing season constraint for crop exposure to O_3, which in OCN is accounted for by the simulated phenology and seasonality of photosynthesis. The external leaf-resistance (r_{ext}) per unit surface area is calculated as

$$r_{ext} = r_{ext,b} F_T \tag{7}$$

where the base external leaf-resistance ($r_{ext,b}$) of 2500 $\mathrm{m\,s^{-1}}$ is ~~altered by F_T a correction factor for low temperatures~~ scaled by a low temperature correction factor F_T and

$$F_T = e^{-0.2(1+T_s)} \tag{8}$$

with $1 \leq F_T \leq 2$ and T_s the 2 m air-temperature ($°C$ Simpson et al., 2012, eq. 60). For temperatures below -1 $°C$ non-stomatal resistances are increased up to two times (Simpson et al., 2012; Zhang et al., 2003) . The within-canopy resistance (R_{inc}) is calculated as

$$R_{inc} = bSAI\frac{h}{u_*} \tag{9}$$

where b is an empirical constant (set to 14 $\mathrm{s^{-1}}$) and h is the canopy height in m. The ground-surface resistance R_{gs} is calculated as

$$R_{gs} = \frac{1-2f_{snow}}{F_T\hat{R}_{gs}} + \frac{2f_{snow}}{R_{snow}} \tag{10}$$

(Simpson et al., 2012, eq. 59). $\hat{R}_{gs}$ represents base-values of R_{gs} and takes values of 2000 $\mathrm{s\,m^{-1}}$ for bare soil, 200 $\mathrm{s\,m^{-1}}$ for forests and crops and 1000 $\mathrm{s\,m^{-1}}$ for non-crop grasses (Simpson et al., 2012, Suppl.). ~~Like~~ As in EMEP, the ground-surface resistance of ~~ozone~~ O_3 to snow (R_{snow}) is set to a value of 2000 $\mathrm{s\,m^{-1}}$ according to ~~(Zhang et al., 2003)~~ Zhang et al. (2003) . f_{snow} is calculated from the actual snow depth (s_d) simulated by OCN, and the maximum possible snow depth ($s_{d,max}$):

$$f_{snow} = \frac{s_d}{s_{d,max}} \tag{11}$$

with the constraint of $0 \leq f_{snow} \leq 0.5$~~. The upper border prevents~~ , to prevent negative values in the first fraction of eq. 10. $s_{d,max}$ is taken to be 10 $\mathrm{kgm^{-2}}$ (Ducoudré et al., 1993).

Given these resistances, the canopy O_3 concentration (χ_c^{O3}, $\mathrm{nmol\,m^{-3}}$) is then calculated based on a constant flux assumption

$$\chi_c^{O3} = \chi_{atm}^{O3}(1 - \frac{R_a}{R_a + R_b + R_c}). \tag{12}$$

χ_c^{O3} and the stomatal conductance to ~~ozone~~ O_3 ($G_{st}^{O_3}$ in $\mathrm{m\,s^{-1}}$) are used to calculate the ~~ozone~~ O_3 flux into the leaf cavities (F_{st}, $\mathrm{nmol\,m^{-2}\,s^{-1}}$):

$$F_{st} = (\chi_c^{O3} - \chi_i^{O3}) G_{st}^{O_3}. \tag{13}$$

According to Laisk et al. (1989) the leaf internal O_3 concentration (χ_i^{O3}) is assumed to be zero.

~~It should be noted that the~~ The OCN implementation of deposition and flux described above is a simplification of the deposition system used by EMEP in order to fit the process representation of ORCHIDEE, from which OCN has inherited its biophysical modules. The external leaf resistance is not included in the calculation of F_{st} (Tuovinen et al., 2007, 2009) what results in an overestimation of stomatal O_3 uptake. Further, OCN's calculation of R_a is based upon neutral stability conditions (see Appendix), whereas the EMEP model makes use of rather detailed stability correction factors. However, a series of calculations with the full EMEP model have shown that the uncertainties associated with these simplifications are small, typically ~~less than 1~~0.5-5 $\mathrm{mmol\,m^{-2}}$. As base-case values of POD0 are typically ca. 30-50 in EU regions, these approximations do not seem to be a major cause of error, at least in regions with substantial ozone (and carbon) uptake. The full coupling of OCN to a CTM would be desirable to eliminate this bias and allow for a consistent calculation of tropospheric and surface near O_3 burdens.

2.2 Relating stomatal uptake to leaf damage

~~To estimate the ozone related damage due to stomatal uptake, a flux threshold (F_{detox}) is used to account for the plants ability to detoxify part of the ozone .~~

$$\cancel{F_{st,detox} = \mathrm{MAX}(F_{st} - F_{detox}, 0)}$$

~~where the detoxification threshold F_{detox} is set to 1.6 for forests and to 5 for grasses and crops (Sitch et al., 2007) . The function MAX prevents negative uptake values when $F_{st} < F_{detox}$.~~ An accumulation of ~~$F_{st,detox}$~~ F_{st} over time gives the accumulated uptake of ~~ozone~~ O_3 for a particular canopy layer (CUO_l, $\mathrm{mmol\,m^{-2}}$), or for $l = 1$ (top canopy layer) the phytotoxic ~~ozone~~ O_3 dose, (~~POD_l~~ POD, $\mathrm{mmol\,m^{-2}}$)

$$\cancel{CUO_l}\frac{dCUO_l}{dt} = \cancel{CUO_l}(1 - f_{\cancel{shed}new})CUO_l + cF_{\cancel{st,detox}\cancel{\Delta t}st,l} \tag{14}$$

where ~~$\Delta t = 1800$ seconds is the length of simulation time step and~~ c = 10^{-6} converts from nmol to mmol and the integration time step is 1800 seconds.

The phenology of leaves is accounted for by assuming that emerging leaves are undamaged, and by reducing the CUO_l by the fraction of new developed leaves per time step and layer ($\cancel{f_{shed}}\, f_{new}$). Furthermore deciduous PFTs shed all CUO at the end of the growing season and grow undamaged leaves the next spring. Evergreen PFTs shed proportionate amounts of CUO during the entire year always when new leaves are grown.

The full canopy cumulative uptake of O_3 is calculated by summing CUO_l over all present canopy layers (n)

$$CUO = \sum_{l=1}^{n} CUO_l. \tag{15}$$

The CUO_l is used to approximate the damage to net photosynthesis (A_n) by using the damage relationship of Wittig et al. (2007):

$$d_l^{O3} = \frac{0.22 CUO_l + 6.16}{100} \tag{16}$$

where the factor 100 scales the percentage values of damage to fractions. Net photosynthesis accounting for ozone damage (A_n^{O3}) is then calculated by subtracting the damage fraction from the undamaged value of A_n:

$$A_{n,l}^{O3} = A_{n,l}(1 - d_l^{O3}). \tag{17}$$

Since G_{st} and A_n are tightly coupled (see eq. 5), a damage of A_n results in a simultaneous reduction in G_{st}~~and C_i,~~. The canopy-scale ~~ozone~~ O_3 flux into the leaf cavities (F_{stC}) is calculated by summing F_{st} of all canopy layers, similar to the aggregation of $A_{n,l}$ and G_{st} and CUO_l. Canopy O_3 concentration, ~~ozone~~ O_3 uptake, canopy cumulative ~~ozone~~ O_3 uptake (CUO) and damage to net photosynthesis are solved iteratively to account for the feedbacks between ozone damage, canopy conductance and canopy-air ~~ozone~~ O_3 concentrations.

~~The CUO above a threshold for trees and grass/crop PFTs together is referred to as $CUO_5^{1.6}$ in the following.~~ Note that CUO and POD can be directly compared to estimates according to the ~~(LRTAP-Convention, 2010)~~ LRTAP-Convention (2010) notation, when analysing only the top canopy layer (Mills et al., 2011b).

2.3 Sensitivity analysis

A sensitivity analysis is conducted to estimate the sensitivity of the modelled plant ~~ozone~~ O_3 uptake to the parameterisation of the model, to establish the robustness of the model, and to identify the most influential parameters. Three parameters (ground-surface resistance ($\hat{R}_{gs}$~~,~~), external leaf-resistance (r_{ext}~~,~~), empirical constant (b), see eq. 10, 6, 9, respectively) and ~~two~~ three modelled quantities (canopy conductance (G_c ~~and R_a,~~), aerodynamic resistance (R_a), and canopy-scale quasi-laminar layer resistance (R_b), see eq. 5, 2~~, respectively~~), with considerable uncertainty due to the underlying parameters used to calculate these quantities, are perturbed within ~~±20%~~ ±20% of their central estimate.

A set of 100 parameter combinations is created with a latin hypercube sampling method (McKay et al., 1979), simultaneously perturbing all ~~five~~ six parameter values (R-package: FME, function: Latinhyper). For each parameter combination, a transient run (see Modelling protocol section) is performed creating an ensemble of estimates for the key prognostic variables F_{stC} (eq. 13), R_c (eq. 4), V_g (eq. 2) and the ~~ozone~~ O_3 flux ratio (F_R) calculated as the ratio of F_{stC} and the total ~~ozone~~ O_3 flux to the surface (F_g, eq. 1).

The summer months June, July, and August (JJA) are selected from the simulation output and used for further analysis. For each prognostic variable (F_{stC}, R_c, V_g, F_R), the sensitivity to changes in all ~~five~~ six perturbed parameters/variables is estimated by calculating partial correlation coefficients (PCC) and partial ranked correlation coefficients (PRCC) (Helton and Davis, 2002). PCC's record the linear relationship between two variables where the linear effects of all other variables in the analysis are removed (Helton and Davis, 2002). In case of nonlinear relationships, RPCC can be used, which implies a rank transformation to linearise any monotonic relationship, such that the regression and correlation procedures as in the PCC can follow (Helton and Davis, 2002). We estimate the magnitude of the parameter effect by creating mean summer values of the four prognostic variables for each sensitivity run, and regressing these values against the corresponding parameter/variable scaling values of the respective model run.

2.4 Modelling protocol and data for site-level simulations

The site levels simulations (single-point simulations) at the FLUXNET sites are run using observed metrological forcing, soil properties, and land cover from the La Thuile ~~data set~~ Dataset (http://fluxnet.fluxdata.org/data/la-thuile-dataset/) of the FLUXNET project (Baldocchi et al., 2001). Data on atmospheric CO_2 concentrations are obtained from ~~(Sitch et al., 2015) . Nitrogen deposition (reduced and oxidised~~ Sitch et al. (2015) . Reduced and oxidised nitrogen deposition in wet and dry forms ~~) and hourly ozone~~ and hourly O_3 concentrations at 45 m height are provided by the EMEP model (see Sect. 2.5).

OCN is brought into equilibrium in terms of the terrestrial vegetation and soil carbon and nitrogen pools in a first step with the forcing of the year 1900. In the next step, the model is run with a progressive simulation of the period 1900 up ~~untill~~ until the start year of the respective site. For this period atmospheric O_3 and CO_2 concentrations, as well as N deposition of the respective simulated years are used. Due to lack of observed climate for the sites for this period, the site-specific observed meteorology from recent years is iterated for these first two steps. The observation years (see Appendix ~~tab~~Tab. 1) are simulated with ~~time-varying~~ the climate and atmospheric conditions (N deposition, CO_2 and O_3 concentrations) of the respective years.

For the evaluation of the model output, net ecosystem exchange (NEE), and latent heat flux (LE), as well as meteorological observations are obtained for eleven evergreen needle-leaved forest sites, ten deciduous broadleaved forest sites and five C3 grassland sites in Europe (see Appendix ~~tab~~Tab. 1) from the La Thuile ~~data set~~ Dataset of the FLUXNET project (Baldocchi et al., 2001). Leaf area indices (LAI) based on discrete point measurements are obtained from the La ~~Thuille~~ Thuile ancillary data base.

NEE measurements are used to estimate gross primary production (GPP) by the flux-partitioning method according to (Reichstein et al., 2005). Canopy conductance (G_c) is derived by inverting the Penmen-Monteith equation given the observed LE and atmospheric conditions as described in ~~(Knauer et al., 2015)~~ Knauer et al. (2015) .

The half-hourly FLUXNET and model fluxes are filtered prior to deriving average growing-season fluxes (bud break to litter fall) to reduce the ~~effects~~ effect of model biases on the model-data comparison. ~~To derive average growing-season fluxes, night-time~~ Night-time and morning/evening hours are excluded by removing data with lower than 20 % of the daily maximum short-wave downward radiation. To avoid any biases associated with the soil moisture or atmospheric drought response of OCN, we further exclude data points with a modelled soil moisture constraint factor (range between 0-1) below 0.8 and an atmospheric vapour pressure deficit larger than 0.5 kPa.

Daily mean values are calculated ~~of~~ from the remaining time steps only where both modelled and observed values are present. The derived daily values are furthermore constrained to the main growing season by excluding days where the daily GPP is less than 20 % of the yearly maximum daily GPP.

To derive representative diurnal cycles, data for the month July are filtered for daylight hours (taken as incoming short-wave radiation $\geq 100\,\mathrm{W\,m^{-2}}$), and excluding periods of soil or atmospheric drought stress as above. This is done for modelled F_{stC}, R_c, V_g, F_R and ~~modelled as well as~~ for both modelled and FLUXNET observed GPP and G_c.

2.5 Modelling protocol and data for regional simulations

For the regional simulations, OCN is run at a spatial resolution of 0.5° x 0.5° on a spatial domain focused on Europe. Daily meteorological forcing (temperature, precipitation, short-wave and long-wave downward radiation, atmospheric specific humidity and wind speed) for the years 1961 to 2010 is obtained from RCA3 regional climate model (Samuelsson et al., 2011; Kjellstrom et al., 2011), nested to the ECHAM5 model (Roeckner et al., 2006), and has been bias corrected for temperatures and precipitation using the CRU climatology (New et al., 1999). ~~Nitrogen deposition (reduced and oxidised~~ Reduced and oxidised nitrogen deposition in wet and dry forms ~~) and ozone~~ and O_3 concentrations at 45 m height for the same years are obtained from the EMEP model, which is also run with RCA3 meteorology (as in Simpson et al., 2014b). Emissions for the EMEP runs in current years are as described in ~~(Simpson et al., 2014b) ,~~ Simpson et al. (2014b) , and are scaled back to 1900 using data from UN-ECE and van Aardenne et al. (2001) – see Appendix B. Further details of the EMEP model setup for this grid and meteorology can be found in Simpson et al. (2014b) and Engardt et al. (2016). For OCN, land cover, soil, and N fertiliser application are used as in ~~(Zaehle et al., 2011)~~ Zaehle et al. (2011) and kept at 2005 values throughout the simulation. Data on atmospheric CO_2 concentrations are obtained from ~~(Sitch et al., 2015)~~ Sitch et al. (2015) .

OCN is brought into equilibrium in terms of the terrestrial vegetation and soil carbon and nitrogen pools ~~with 1961-1970 forcing~~ by randomly iterating the forcing from the period 1961-1970. This is followed by a simulation for the years ~~1960-2010~~ 1961-2011 with time-varying climate and atmospheric conditions (N deposition, CO_2, and O_3 concentrations), but static land cover and land-use information (kept at year 2005 levels). An up-scaled FLUXNET-MTE-product of GPP (Jung et al., 2011), using the machine learning technique: model tree ensembles (MTE), is used to evaluate modelled GPP.

2.6 Impacts of using the ozone deposition scheme

~~Different~~ In contrast to other terrestrial biosphere models, the OCN ozone module accounts for the effects of aerodynamic, stomatal and non-stomatal resistance to ~~ozone~~ O_3 deposition. Due to these resistances~~the canopy~~ , the deposition of O_3 to leaf-level is reduced, and the canopy O_3 concentration is lower than the atmospheric O_3 concentration. Thus using such a deposition scheme reduces modelled ~~ozone~~ O_3 uptake into plants and accumulation. To get an estimate of the magnitude of this impact we compare simulations with the standard deposition scheme as described above (D) with a simulation where ~~ozone~~ O_3 surface resistance is only determined by stomatal resistance and the non-stomatal depletion of ~~ozone~~ O_3 is zero (D-STO), and a further simulation where no deposition scheme is used and the canopy O_3 concentration is equal to the atmospheric concentration (ATM).

3 Results

3.1 Evaluation against daily eddy-covariance data

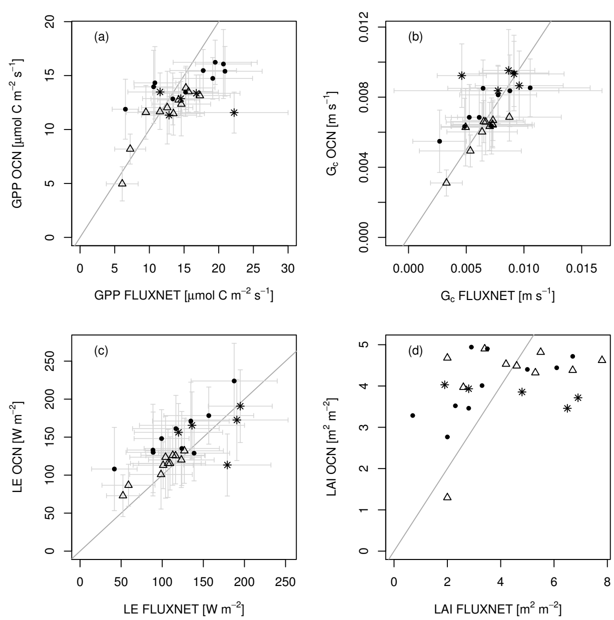

Figure 1. Comparison of measured a) GPP, b) canopy conductance (G_c), c) latent heat fluxes (LE) and d) LAI at 26 European FLUXNET sites and simulations by OCN. Displayed are means and standard deviations of daily means of the measuring/simulation period, with the exception of FLUXNET derived LAI, which ~~is~~ are based on point measurements. Dots symbolise sites dominated by broadleaved trees, triangles sites dominated by needle-leaved trees and asterisks sites dominated by C3 grasses. The grey line constitutes the 1:1 line.

Figure 1 a shows that, for most sites, modelled and observation-based GPP agree ~~within the standard deviation~~well (see Appendix Tab. 2 for R^2 and RMSE values). The standard deviation is larger for the observation-based estimates because of the high level of noise in the eddy-covariance data. For sites dominated by needle-leaved trees, the modelled and observation-based GPP values are very close, with ~~some slightly~~ only slight under- and overestimates by the model at some sites. At sites dominated by broadleaved trees, modelled GPP deviates more strongly from the observation-based GPP, underestimating the observations in ~~six~~ seven out of ten cases. However, the results are within the range of standard deviation except for the drought prone PT-Mi1 site (see Appendix Fig. 10 a ~~)~~ for an explicit site comparison). At C3 grassland sites, modelled GPP is in good agreement with the observation-based GPP except for AT-Neu, which has the highest mean GPP of all sites observed by FLUXNET with a ~~arge~~ large standard deviation, which may reflect the effect of site management ~~, not replicated by the model~~ (e.g. mowing and fertilisation), for which no data was readily available as model forcing.

When comparing modelled and observed latent heat fluxes (LE), the model fits the observations best at the needle-leaved forest sites (Fig. 1 c). However, LE is overestimated at nine out of ten broadleaved forest sites, but remains within the range of the large observational standard deviation. At sites dominated by C3 grasses the modelled LE differs considerably from observed value, at two sites overestimating and two underestimating the fluxes, again within the observational-standard deviation.

In agreement with the comparison of GPP and LE, the comparison of modelled to observation-based canopy conductance (G_c) shows the best agreement for sites dominated by needle-leaved trees (Fig. 1 b). At sites dominated by broadleaved trees, the modelled G_c varies more widely from the FLUXNET G_c. The modelled G_c at sites dominated by C3 grasses is in very good agreement to FLUXNET G_c with slightly overestimating G_c at 2 out of 3 sites except for the DE-Meh site, where means differ ~~widely~~outside the standard deviation (see Appendix Fig. 10b).

The comparison of the average modelled summertime LAI and point measurements at the FLUXNET illustrates that the variability in the measured LAI is much greater than that of OCN (Fig. 1 d). The modelled LAI values approach light-saturating, maximum LAI values and are not able to reproduce between-site differences in e.g. the growth stage, site-history, or maximum possible LAI values. Furthermore, it should be born in mind that the observed LAI values are averages of point measurements, which are not necessarily representative of the modelled time-period, and that the model had not been parameterised specifically for the sites. Modelled GPP does not only depend on LAI, but also on light availability, temperature and soil moisture. The much better represented values of GPP, G_c and LE compared to FLUXNET data (Fig. 1 a-c) indicate that OCN is able to adequately transform available energy into carbon uptake and water loss and thus to simulate key variables impacting ozone uptake within a reasonable range.

3.2 Mean diurnal cycles of key ~~ozone~~ O_3 parameters.

For further evaluation of the modelled ~~ozone~~ O_3 uptake, we analysed the diurnal cycles of four key O_3 variables (O_3 uptake (F_{stC}), O_3 surface resistance (R_c), O_3 deposition velocity (V_g), and flux ratio (F_R)) as well as GPP and G_c at three sites~~to represent typical ecosystems of Europe~~, one of the three categories broadleaved, needle-leaved and C3 grass sites respectively. The selection criteria are that modelled and FLUXNET GPP and LAI agree well and a minimum of five observation years is available to reduce possible biases from the inability of the model to simulate short-term variations from the mean. The selected sites are a temperate broadleaved summer green forest (IT-Ro1), a boreal needle-leaved evergreen forest (FI-Hyy), as well as a temperate C3 grass land (CH-Oe1). We evaluate modelled GPP and G_c against observations from the FLUXNET sites. ~~We~~

~~also present the~~ The modelled mean diurnal cycles of ~~ozone related variables for which we did not have access to site-specific observations~~ O_3 related variables (F_{stC}, R_c, V_g, F_R) ~~. Instead, we compare these quantities~~ are compared to reported values in the literature since we did not have access to site-specific observations.

Modelled and observed mean diurnal cycles of GPP and G_c are in general agreement at the three selected FLUXNET sites (see Fig. 2 a,g,m ~~)~~ and b,h,n) ~~) with a particular~~ with particularly good agreement for the mean diurnal cycle of GPP at the ~~Finnish~~ needle-leaved site FI-Hyy, where the hourly means are very close and the observational standard deviation is narrow (see Fig. 2 g). At the ~~Italian~~ grassland site IT-Ro1 the overall daytime magnitude of the fluxes is reproduced in general except for the observed afternoon reduction in GPP (see Fig. 2 a). The ~~hourly means are within the calculated standard deviation~~modelled hourly values fall in the range of the observed values. Modelled and observation-based hourly means of GPP at the site CH-Oe1 agree well except for the evening hours, where the observed values ~~became highly variable~~increase again. The mean diurnal cycles of G_c derived from the FLUXNET data are again best matched at the site FI-Hyy, ~~where as~~ whereas the model generally overestimates the diurnal cycle of G_c slightly at the site IT-Ro1, and overestimates peak G_c at the CH-Oe1 site. The fact that OCN does not always ~~simulated~~ simulate the observed midday depression of G_c, suggests that the response of stomata to atmospheric and soil drought in OCN requires further evaluation and improvement.

~~The modelled hourly mean ozone surface resistance R_c is highest with approximately 400 during night time and decreases during daytime to values of 100-180 , where the lowest surface resistance of approximately 100 is modelled at the grassland site CH-Oe1 (~~Similar to the daily mean values (see Fig. ~~2 f,l,r). These values are slightly higher than independent estimates (for grasses and crops obtained for other sites) of noon surface resistances ranging 50-100 (Padro, 1996; Coyle et al., 2009; Gerosa et al., 2004; T~~ ~~Tuovinen et al. (2004) reported noon values of approximately 140 for a Scots pine forest and 70-140 for a Norway spruce forest site (Tuovinen et al., 2001) , which compares well with the modelled R_c values at~~ 1 a,b) the mean hourly values show the best match of GPP and G_c for the needle-leaved ~~forest site (FI-Hyy; Fig. 2 l). Higher noon values of 250 are reported at a Danish Norway spruce site (Mikkelsen et al., 2004) . For a Mountain Birch forest noon values of 110-140 (Tuovinen et al., 2001) are observed what is slightly lower than the modelled value at the IT-Ro1 site (dominated by broadleaved tree PFT).~~

~~The modelled deposition velocities V_g are lowest during night time with values of approximately 0.002 (Fig. 2 e,k,q). These values increase to maximum hourly means of 0.006-0.007 during daytime. These values compare well to reported values of deposition velocities, which range from 0.003-0.009 at noon (Gerosa et al., 2004) for a barely field, approximately 0.006 at noon for a wheat field (Tuovinen et al., 2004) and approximately 0.009 at noon at a potato field (Coyle et al., 2009) . These estimates also agree well with maximum deposition velocities reported for Scots pine site of 0.006 (Keronen et al., 2003; Tuovinen et al., 20~~ ~~noon values from Danish Norway spruce sites of 0.006-0.010 (Mikkelsen et al., 2004; Tuovinen et al., 2001) . Mean daytime deposition velocities of 0.006 (range 0.003 - 0.008) are reported at a finish mountain birch site (Tuovinen et al., 2001)~~ tree site and stronger deviations for the sites covered by broadleaved trees and C3 grasses.

The stomatal ~~ozone~~ O_3 uptake F_{stC} (Fig. 2 c,i,o) is close to zero during night time when the stomata are assumed to be closed, because gross photosynthesis is zero. At FI-Hyy and CH-Oe1, peak uptake occurred at noon at values between 8-9 $\mathrm{nmol\,m^{-2}\,s^{-1}}$, when photosynthesis (Fig. 2 g,m) and stomatal conductance (Fig. 2 h,n) are highest. At the Italian site IT-Ro1, maximum uptake occurs in the afternoon hours around 15 h, with much larger standard deviation compared to the other two sites (Fig. 2 c)). The magnitude of stomatal ~~ozone~~ O_3 uptake corresponds well to some values reported e.g. for crops ~~(Gerosa et al., 2003, 2004, daily maxima of 4-9 $\mathrm{nmol\,m^{-2}\,s^{-1}}$)~~ (Gerosa et al., 2003, 2004, daily maxima of 4-9 $\mathrm{nmol\,m^{-2}\,s^{-1}}$) and holm oak ~~(Vitale et al., 2005, approx. 7-8 $\mathrm{nmol\,m^{-2}\,s^{-1}}$)~~ (Vitale et al., 2005, approx. 7-8 $\mathrm{nmol\,m^{-2}\,s^{-1}}$) . Lower daily maximum values have been reported for an evergreen Mediterranean Forest dominated by Holm Oak of 4 $\mathrm{nmol\,m^{-2}\,s^{-1}}$ under dry weather conditions (Gerosa et al., 2005) and ~~-~~1-6 $\mathrm{nmol\,m^{-2}\,s^{-1}}$ for diverse southern European vegetation types (Cieslik, 2004). Much higher values are reported for *Picea abies* (~~-~~50-90 $\mathrm{nmol\,m^{-2}\,s^{-1}}$), *Pinus cembra* (~~-~~10-50 $\mathrm{nmol\,m^{-2}\,s^{-1}}$) and *Larix decidua* (~~-~~10-40 $\mathrm{nmol\,m^{-2}\,s^{-1}}$) at a site near Innsbruck Austria (Wieser et al., 2003), where canopy ~~ozone~~ O_3 uptake was estimated by sapflow measurements in contrast to the studies mentioned before where the eddy covariance technique was applied. The much higher F_{stC} values in that study result from much higher canopy conductances to ~~ozone~~ O_3 ($G_c^{O_3}$), which are up to 12 times higher than the modelled $G_c^{O_3}$ values in our study (see Fig. 2, ~~$\frac{G_c}{1.51}$~~ $G_c^{O_3} = \frac{G_c}{1.51}$).

The ratio between the ~~vegetation ozone~~ stomatal O_3 uptake and the total surface uptake (F_R) is close to zero during night time hours and increases steeply in the morning hours (Fig. 2 d,j,p). The 24 h average is approximately 0.3 for IT-Ro1 and 0.4 for FI-Hyy and CH-Oe1 (Fig. 2 d,j,p). Peak hourly mean values are close to 0.6 at IT-Ro1, around 0.7 at FI-Hyy and close to 0.8 at CH-Oe1. These values are comparable to the ratios reported for crops ~~(0.5 - 0.6 Gerosa et al., 2004; Fowler et al., 2009)~~ (0.5-0.6 Gerosa et al., 2 Norway spruce ~~(Mikkelsen et al., 2004, 0.3 -0.33)~~ (Mikkelsen et al., 2004, 0.3-0.33) and diverse southern European vegetation types (Cieslik, 2004, 0.12 - 0.69). The modelled flux ratios here show slightly higher daily maximum flux ratios than reported in the listed studies. Daily mean flux ratios are well within the reported range.

The modelled deposition velocities V_g are lowest during night time with values of approximately 0.002 $\mathrm{m\,s^{-1}}$ (Fig. 2 e,k,q). These values increase to maximum hourly means of 0.006-0.007 $\mathrm{m\,s^{-1}}$ during daytime. These values compare well to reported values of deposition velocities, which range from 0.003-0.009 $\mathrm{m\,s^{-1}}$ at noon (Gerosa et al., 2004) for a barley field, approximately 0.006 $\mathrm{m\,s^{-1}}$ at noon for a wheat field (Tuovinen et al., 2004) and approximately 0.009 $\mathrm{m\,s^{-1}}$ at noon at a potato field (Coyle et al., 2009) . The estimates for FI-Hyy also agree well with maximum deposition velocities reported for Scots pine site of 0.006 $\mathrm{m\,s^{-1}}$ (Keronen et al., 2003; Tuovinen et al., 2004) and noon values from Danish Norway spruce sites of 0.006-0.010 $\mathrm{m\,s^{-1}}$ (Mikkelsen et al., 2004; Tuovinen et al., 2001) . Mean daytime deposition velocities of 0.006 $\mathrm{m\,s^{-1}}$ (range 0.003-0.008 $\mathrm{m\,s^{-1}}$) are reported at a finish mountain birch site (Tuovinen et al., 2001) . Simulated monthly mean values of V_g differ substantially between the sites (see Appendix 11). When comparing the monthly means over all sites (Appendix 11 dashed line) of a functional group (broadleaved, needle-leaved, C3 grasses) to the ensemble mean of 15 CTM's (Hardacre et al., 2015) the values simulated here are higher for needle-leaved tree sites. For broadleaved tree sites and and grassland sites higher value but still within the observed ensemble range are found for the summer months.

The modelled hourly mean O_3 surface resistance R_c is highest with approximately 400 $\mathrm{s\,m^{-1}}$ during night time and decreases during daytime to values of 100-180 $\mathrm{s\,m^{-1}}$, where the lowest surface resistance of approximately 100 $\mathrm{s\,m^{-1}}$ is modelled at the grassland site CH-Oe1 (Fig. 2 f,l,r). These values are slightly higher than independent estimates (for grasses and crops obtained for other sites) of noon surface resistances ranging 50-100 $\mathrm{s\,m^{-1}}$ (Padro, 1996; Coyle et al., 2009; Gerosa et al., 2004; Tuovinen et al., 2004 Tuovinen et al. (2004) reported noon values of approximately 140 $\mathrm{s\,m^{-1}}$ for a Scots pine forest and 70-140 $\mathrm{s\,m^{-1}}$ for a Norway spruce forest site (Tuovinen et al., 2001) , which compares well with the modelled R_c values at the needle-leaved forest site (FI-Hyy; Fig. 2 l). Higher noon values of approximately 250 $\mathrm{s\,m^{-1}}$ are reported at a Danish Norway spruce site (Mikkelsen et al., 2004) . For a Mountain Birch forest noon values of 110-140 $\mathrm{s\,m^{-1}}$ (Tuovinen et al., 2001) are observed which is slightly lower than the modelled value at the IT-Ro1 site (dominated by broadleaved tree PFT).

3.3 Sensitivity analysis

We assess the sensitivity of the modelled ~~ozone~~ O_3 uptake and deposition, represented by F_g, F_{stC}, V_g, and R_c to uncertainty in ~~five weakly constraint~~ six weakly constrained variables and parameters of the ~~ozone~~ O_3 deposition scheme (R_a, b, r_{ext}, $\hat{R}_{gs}$~~and~~, G_c, and R_b). Fig. 3 a shows for example the results for the ~~Finnish~~ boreal needle-leaved forest FI-Hyy. As expected, all uptake/deposition variables, except ~~of~~ for the flux ratio (F_R) are negatively correlated to the aerodynamic resistance R_a, ~~describing~~ which describes the level of decoupling of the atmosphere and land surface. Increasing R_a decreases the canopy internal O_3 concentration and hence stomatal (F_{stC}) and total (F_g) deposition as well as the deposition velocity (V_g). The flux ratio F_R is slightly positively correlated to changes in R_a due to the stronger negative correlation of F_{stC} ~~compared~~ relative to F_g.

In decreasing order, but as expected, the level of external leaf-resistance (r_{ext}), the scaling factor b (eq. 9), ~~and~~ the soil resistance ($\hat{R}_{gs}$), and the canopy-scale quasi-laminar layer resistance (R_b) increase R_c and consequently reduce F_g and V_g. Reducing the non-stomatal deposition by increasing r_{ext}, b, ~~and~~ $\hat{R}_{gs}$, and R_b increases the canopy internal O_3 concentration and thus stomatal ~~ozone~~ O_3 uptake (F_{stC}). The combined effects of a reduction of total deposition F_g and an increase of F_{stC} cause a positive correlation of F_R to r_{ext}, b, ~~and~~ $\hat{R}_{gs}$, and R_b.

Increasing canopy conductance (G_c~~increases stomatal ozone~~) increases stomatal O_3 uptake (F_{stC}) and thereby also increases V_g and F_g. The increased total ~~ozone~~ O_3 uptake (F_g) decreases the surface resistance to ~~ozone~~ O_3 uptake R_c ~~what causes~~ resulting in a negative correlation of R_c with G_c. The stronger increase in F_{stC} ~~compared~~ relative to F_g results in a positive correlation of F_R.

Despite these partial correlations, only changed values for r_{ext} and G_c have a notable effect on the predicted fluxes (Fig. 3 b), whereas for the other factors (R_a, b, and $\hat{R}_{gs}$), the impact on the simulated fluxes is less than 0.1 ~~(% / %).~~ % due to a 1 % change in the variables/parameters of the deposition scheme.

The flux ratio F_R is very little affected by varying r_{ext} and G_c.

Notwithstanding the perturbations, all four ~~ozone~~ O_3 related flux variables show a fairly narrow range of simulated values (Fig. 4). For all four variables the unperturbed model and the ensemble mean lie on top of each other (see dashed red and yellow line in Fig. 4 a-d). The seasonal course of the surface resistances and fluxes are maintained. The simulations show a strong day to day variability of F_{stC}, which is conserved with different parameter combinations, and which is largely driven by the day-to-day variations in G_c and the atmospheric ~~ozone concentration~~ O_3 concentration (see Fig. 4 f and e respectively). Ozone uptake by the leaves reduces the ~~ozone~~ O_3 surface resistance during the growing season such that R_c becomes lowest. The cumulative uptake of ~~ozone~~ O_3 (CUO) is lowest at the beginning of the growing season but not zero because the evergreen pine at the Hyytiälä site accumulates ~~ozone~~ O_3 over several years (Fig. 4 f). The CUO increases during the growing season and declines in autumn when a larger fraction of old needles are shed.

The little impact of the perturbations on the simulated O_3 uptake and deposition variables suggests that he calculated O_3 uptake is relatively robust against uncertainties in the parameterisation of some of the lesser known surface properties.

3.4 Regional simulations

We used the model to simulate the vegetation productivity, ~~ozone~~ O_3 uptake, and associated ozone damage of plant production over Europe for the period 2001-2010 (see Section 2.5 for modelling protocol).

Simulated mean annual GPP for the years 1982-2011 shows in general good agreement with an independent estimate of GPP based on up scaled eddy-covariance measurements (MTE, see Section 2.5), with ~~the estimates being within 250~~ OCN on average underestimating GPP by 16 % (European mean). A significant exception ~~to this acceptable agreement~~ are cropland dominated areas (Fig. 5) in parts of Eastern Europe, Southern Russia, Turkey and Northern Spain, which show consistent overestimation of GPP by OCN of ~~400 to 900~~ 400-900 $\mathrm{g\,C\,m^{-2}\,yr^{-1}}$ (58 % overestimation on average). Regions with a strong disagreement coincide with high simulated LAI values by OCN and a higher simulated GPP in summer compared to the summer GPP by MTE. In addition, OCN simulates a longer growing season for croplands since sowing and harvest dates are not considered. It is worth noting, nevertheless, that there are no FLUXNET stations present in the regions of disagreement hotspots, making it difficult to assess the reliability of the MTE product in this region.

North of $60°N$ OCN has the tendency to produce ~~larger~~ lower estimates of GPP than inferred from the observation-based product, which is particularly pronounced in low productivity mountain regions of Norway and Sweden. It is unclear whether this bias is indicative of a too strong N limitation in the OCN model.

Average decadal ~~ozone~~ O_3 concentrations generally increase from Northern to Southern Europe (Fig. 6 a) and with increasing altitude, with local deviations from this pattern in centres of substantial air pollution. The pattern of foliar ~~ozone~~ O_3 uptake differs distinctly from that of the O3 concentrations, showing highest uptake rates in Central, Eastern and parts of Southern Europe (Fig. 6 b), associated with centres of high rates of simulated gross primary production (Fig. ~~6 d~~5 a) and thus canopy conductance. The cumulative ~~ozone uptake beyond the PFT-specific detoxification threshold~~ O_3 uptake reaches values of ~~6-12~~ 40-60 $\mathrm{mmol\,m^{-2}}$ in large parts of Central Europe (Fig. 6 c). The highest accumulation rates of ~~up to 15-20~~ 80-110 $\mathrm{mmol\,m^{-2}}$ are found in ~~temperate Eastern Europe between $50°N$ and $60°N$,~~ Eastern Europe and parts of Scandinavia as well as in Italy, the Alps and the Bordeaux region. The concentration based exposure index AOT40 (Fig. 6 d) shows a strong north south gradient similar to the ~~ozone~~ O_3 concentration (Fig. 6 a) and is distinctly different to the flux based CUO ~~$\frac{1.6}{5}$~~ pattern (Fig. 6 c).

Simulated reduction of mean decadal GPP due to ~~ozone averaged 60-120~~ O_3 range from 80-160 $\mathrm{g\,C\,m^{-2}\,yr^{-1}}$ over large areas of Central, Eastern, and South-eastern Europe (Fig. 7 a) and is generally largest in regions of high productivity~~(fig 7 a)~~. The relative reduction of GPP is fairly consistent across ~~larger~~ large areas in Europe and averages ~~4-6%~~ 6-10 % (Fig. 7 b). Higher reductions in relative terms are found in regions with high cover ~~fraction~~ of C4 ~~PFT's (see Appendix ?? a, b) like in the Black see~~ PFTs, e.g. Black Sea area. Lower relative reductions are found in Northern and parts of Southern Europe where productivity is low and ~~probably either~~ stomatal O_3 uptake is reduced by e.g. low O_3 concentrations or drought control on stomatal fluxes respectively~~reduces stomatal uptake~~. Slight increases or strong decreases in relative terms are found in regions with very small productivity like in Northern Africa and the mountainous regions of Scandinavia. A slight ~~increases~~ increase in GPP might be caused by feedbacks of GPP damage on LAI, canopy conductance and soil moisture content such that e.g. water savings enable a prolonged growing season and thus a slightly higher GPP. Overall, simulated European productivity has been reduced from 10.6 $\mathrm{Pg\,C\,yr^{-1}}$ to ~~10.1~~ 9.8 $\mathrm{Pg\,C\,yr^{-1}}$ corresponding to a ~~4.7~~7.6 % reduction.

The ~~ozone~~ O_3 induced reductions in GPP are associated with a reduction in mean decadal transpiration rates ~~by 5-10~~ of 8-15 $\mathrm{mm\,yr^{-1}}$ over large parts of Central and Eastern Europe (Fig. 7 c). These reductions correspond to ~~3-4% in Central European and to 4-6~~3-6 % of transpiration in Central Europe and 6-10 % in Northern Europe. As expected, the relative reductions in transpiration rates are therefore slightly less than for GPP due to the role of aerodynamic resistance in controlling water fluxes in addition to canopy conductance. Very high reductions in transpiration are found in the Eastern Black ~~see~~ Sea area associated with strong reductions in GPP and in the mountainous regions of Scandinavia where absolute changes in transpiration are very small. Regionally (in particular in Eastern Spain, Northern Africa and around the Black Sea) lower reductions in transpiration or even slight increases are found (Fig. 7 d). ~~They~~ These are related to ~~ozone~~ O_3 induced soil moisture savings during the wet growing season, leading to lower water stress rates during the drier season. The very strong reduction in transpiration West of the Crimean Peninsula are related to the strong reductions in GPP mentioned above. Overall, simulated European mean transpiration has been reduced from 170.4 mm to ~~165.7~~ 163.3 mm corresponding to a ~~2.8~~4.2 % reduction.

3.5 Impacts of using the ozone deposition scheme

At the FI-Hyy site the canopy O3 concentration, uptake and ~~accumulation~~ accumulated uptake (CUO) increases approximately 10-15 % for the D-STO model (non-stomatal depletion of O_3 is zero) and 20-25 % for the ATM model version (canopy O_3 concentration is equal to the atmospheric concentration) compared to the standard deposition scheme (D) used here (Fig. 8a-c and Appendix 12). The ~~$CUO_{1.6}$ increases stronger and constitutes 35% and 65% for the D-STO and ATM model, respectively. The~~ exact values however are site and PFT specific (see Appendix 12 for the CH-Oe1 and IT-Ro1 site).

The regional impact of using the ozone deposition scheme on CUO is shown in Fig. 9~~(for $CUO^{1.6}_{5}$ see Appendix ??)~~. CUO substantially decreases for the D-STO (Fig. 9b) compared to the ATM model (Fig. 9a). Using the standard deposition model D (Fig. 9c) further reduces the CUO compared to the ATM version where the stomata respond directly to the atmospheric ~~ozone~~ O_3 concentration.

Calculating the canopy ~~ozone~~ O_3 concentration with the help of a deposition scheme that accounts for stomatal and non-stomatal ~~ozone~~ O_3 deposition thus reduces ~~ozone~~ O_3 accumulation in the vegetation.

4 Discussion

We extended the terrestrial biosphere model OCN by a scheme to account for the atmosphere–leaf transfer of ~~ozone with the aim~~ O_3 in order to better account for air pollution effects ~~to~~ on net photosynthesis and hence ~~the~~ regional to global water, carbon, and nitrogen cycling. This ozone deposition scheme calculates canopy O_3 concentrations and uptake into the leaves depending on surface conditions and vegetation carbon uptake. ~~We show that using the canopy concentration strongly impacts~~

~~the cumulative uptake of ozone (CUO) and CUO$_5^{1.6}$ compared to assuming that the concentration outside the leaf would be identical to the atmospheric concentration in 45 height as provided by the CTM. Perturbations of key variables and parameters of the implemented ozone deposition scheme show little impact on the simulated ozone uptake and deposition variables. In other words, the calculated ozone uptake is relatively robust against uncertainties in the parameterisation of some of the lesser known surface properties. Our sensitivity analysis shows that a further crucial part for calculating plant ozone uptake is a correct estimate of the canopy conductance. We provide an assessment of the modelled canopy conductance, and find that the model produces reasonable estimates of canopy conductance compared to FLUXNET data, with a range of caveats as discussed in Section **??**. We relate accumulated ozone uptake above a PFT-specific threshold to reductions in net photosynthesis and find that across large regions of Europe, ozone reduces production and transpiration by approximately 5% and 3%, respectively. This reflects the shape of the implemented damage function which is further discussed in Section **??**.~~

4.1 ~~Atmosphere-leaf transport~~

~~A crucial component for calculating canopy ozone uptake F_{stC} – besides a reliable estimate of G_c – is a reliable estimate of surface ozone concentrations. Ozone destruction above and within the canopy airspace due to compounds emitted by the plants (e.g. biological volatile organic compounds, BVOC's) is assumed to be (at least partly) implicitly included into the non-stomatal ozone destruction terms included in both the EMEP CTM and OCN deposition frameworks. To evaluate the functionality of the implemented ozone deposition scheme in OCN, mean simulated diurnal cycles of key ozone deposition and uptake variables are calculated and found to be within the range of values reported in the literature (see Section 3.2). The implemented deposition scheme is therefore assumed to produce realistic values for key variables.~~

~~Analysing partial correlation coefficients and the strength of the correlation calculated from the sensitivity runs shows that the F_{stC} is most sensitive to changes in G_c. This emphasise the importance of reliable estimates of canopy conductances to obtain reliable estimates of ozone uptake.~~

4.1 ~~Site-level evaluation~~

~~Our results indicates the importance of reliable estimates of the canopy conductance (G_c) for the calculation of ozone uptake. The site-level evaluation of simulated gross primary productivity (GPP), canopy conductance, and latent heat fluxes (LE) to FLUXNET observations at 26 European sites across diverse ecosystem types shows a general good model-data agreement.~~

~~Eddy covariance measurements and derived flux and conductance estimates are subject to a diverse set of random and systematic errors (Richardson et al., 2012) . A lack of energy balance closure can cause underestimation of sensible and latent heat as well as an overestimation of available energy, with a mean bias of 20 % where the imbalance is greatest during nocturnal periods (Wilson et al., 2002) . This imbalance propagates to estimates of canopy conductance, which is inferred from latent and sensible heat fluxes. The energy imbalance furthermore appears to affect estimates of uptake and respiration (Wilson et al., 2002) . Flux partitioning algorithms which extrapolate night-time ecosystem respiration estimates to daytime impose an additional potential for bias in the estimation of GPP (Reichstein et al., 2005) .~~

~~The good agreement of seasonal mean G_c at most of the 26 FLUXNET sites and the well reproduced diurnal cycles at the three selected sites indicates that the physiological processes simulated by OCN are suitable to replicate observed patterns of G_c. This finding, together with the finding that modelled values of V_g, R_c and F_R are within observed ranges, encourages the use of the extended OCN model for determining the effect of air pollution on terrestrial carbon, nitrogen, and water cycling.~~

4.1 ~~Regional damage estimates~~

~~The regional damage estimates of~~ Estimates of the regional damage to annual average GPP (- ~~4.7~~7.6 %) and ~~T~~ transpiration (- ~~2.8~~4.2 %) simulated by OCN for ~~the period of the years~~ 2001-2010 are lower ~~compared to previous~~ than previously reported estimates. ~~Meta-analysis~~ Meta-analyses suggest on average a 11 % (Wittig et al., 2007) and a 21 % (Lombardozzi et al., 2013) reduction of instantaneous photosynthetic rates. However because of carry-over effects this does not necessarily translate directly into reductions in annual GPP. Damage estimates using the Community ~~land model~~ Land Model (CLM) suggest GPP reductions of 10-25 % in Europe and 10.8 % globally (Lombardozzi et al., 2015). Reductions in transpiration ~~are estimated~~ have been estimated as 5-20 % for Europe and ~~globally~~ 2.2 % globally (Lombardozzi et al., 2015). Lombardozzi et al. (2015) however used fixed reductions of photosynthesis (12-20 %) independent of cumulative ~~ozone~~ O_3 uptake for 2 out of 3 simulated plant types. ~~Only for one plant type damage was~~ Damage was only related to cumulative ~~ozone uptake~~ O_3 uptake for one plant type with a very small slope and hence little increase in damage due to increases in cumulative O_3 uptake. Sitch et al. (2007) simulated global GPP reductions of 8-14 % (under elevated and fixed CO_2 respectively) for low plant ozone sensitivity and 15-23 % (under elevated and fixed CO_2 respectively) for high plant ozone sensitivity for the year 2100 compared to 1901. For the Euro-Mediterranean-region an average GPP reduction of 22 % was estimated by the ORCHIDEE-model for the year 2002 using an AOT40 based approach (Anav et al., 2011).

Possible causes for the ~~deviations are the usage of very different~~ discrepancies are differences in dose-response-relationships, ~~different~~ flux thresholds accounting for the detoxification ability of the plants, ~~differing atmospheric ozone concentrations, non-identical~~ atmospheric O_3 concentrations, simulation periods, and ~~differences in simulating~~ simulation of climate change (elevated CO_2) and air pollution (nitrogen deposition). We discuss the most important aspects below. To elucidate the reasons for the substantial differences in the damage estimates further studies are necessary to disentangle the combined effects of differing flux thresholds, damage relationships, climate change, and nitrogen deposition which acts as a O_3 precursor on the one hand and a growth enhancing nutrient on the other hand.

~~An importantfactor in the difference to the previous study is~~

4.1 Atmosphere-leaf transport of ozone

The sensitivity analysis in Section 3.3 demonstrates that the estimate of canopy conductance (G_c) is crucial for calculating plant ozone uptake, therefore reliable observations to constrain modelled canopy conductance are highly important. The site-level evaluation shows that OCN produces reasonable estimates of simulated gross primary productivity (GPP), canopy conductance, and latent heat fluxes (LE) compared to FLUXNET observations. This agreement has to be seen in the light of the a diverse set of random and systematic errors in the eddy covariance measurements, and derived flux and conductance estimates

(Richardson et al., 2012; Knauer et al., 2016) . Next to uncertainties about the strength of the aerodynamic coupling between atmosphere and canopy, problems exist at many sites with respect to the energy balance closure (Wilson et al., 2002) . Failure to close the energy balance can cause underestimation of sensible and latent heat, as well as an overestimation of available energy, with mean bias of 20 % where the imbalance is greatest during nocturnal periods (Wilson et al., 2002) . This imbalance propagates to estimates of canopy conductance, which is inferred from latent and sensible heat fluxes. The energy imbalance furthermore appears to affect estimates of CO_2 uptake and respiration (Wilson et al., 2002) . Flux partitioning algorithms which extrapolate night-time ecosystem respiration estimates to daytime introduce an additional potential for bias in the estimation of GPP (Reichstein et al., 2005) . Nevertheless, the general good agreement of G_c compared to FLUXNET estimates together with the finding that modelled values of key ozone variables are within observed ranges, supports the use of the extended OCN model for determining the effect of air pollution on terrestrial carbon, nitrogen, and water cycling.

A key difference from previous studies is our use of the use of the ozone deposition scheme~~included in our study~~, which reduces O_3 surface concentrations, and hence also the estimated ~~ozone~~ O_3 uptake and accumulation (see ~~fig~~Fig. 9). Accounting for stomatal and non-stomatal deposition in the calculation of the surface O_3 concentrations considerably impacts the estimated plant uptake of O_3. O_3 uptake and cumulated uptake are considerably overestimated when atmospheric ozone concentrations are used to calculate O_3 uptake or when in the calculation of leaf-level O_3 concentrations only stomatal destruction of O_3 is regarded (see section 3.5). Compared to the values that would have been obtained if the CTM O_3 concentrations of the atmosphere (from ca. 45 m height) had been used directly at the leaf surface, our simulations yield a decrease of CUO by 31 % (~~$CUO_5^{1.6}$ 65%) (~~European means for the years 2001-2010). A significant fraction of the decreases is associated with ~~the~~ non-stomatal ~~ozone~~ O_3 uptake and destruction at the surface, which decreased the simulated cumulative ~~ozone~~ O_3 uptake by 16 %~~($CUO_5^{1.6}$ 39%)~~. To obtain ~~an~~ as accurate as possible an estimate of CUO~~/ $CUO_5^{1.6}$~~, stomatal and non-stomatal destruction of ~~ozone and it's~~ O_3 and their impacts on canopy ~~ozone~~ O_3 concentrations should be ~~considered~~ accounted for in terrestrial biosphere models (Tuovinen et al., 2009). Flux-based ozone-damage assessment models may overestimate ozone-related damage unless they properly account for non-stomatal O_3 uptake at the surface.

~~The use of a flux threshold (possibly PFT specific) and it's magnitude naturally also impacts the CUOY (canopy cumulative ozone uptake above a threshold of Y) and possible damage estimates (Tuovinen et al., 2007) . The mean decadal CUO for Europe in the years 2001-2010 is 43.6 in our simulations whereas the mean $CUO_5^{1.6}$ is only 4.7 . Recent studies suggest flux thresholds of 6 for crops and 0 or 1 for trees and semi natural vegetation (LRTAP-Convention, 2010; Mills et al., 2011b) . The impacts of using different flux thresholds on regional estimates of ozone uptake , accumulation and damage are still poorly understood and need further research~~We note that vegetation type and dynamics also impact the stomatal and non-stomatal deposition of O_3, and hence the calculation of the leaf-level O_3 concentrations. This impedes the use of CTM-derived leaf-level O_3 concentration, as CTM and vegetation specifications may differ strongly. Using the O_3 from the lowest level of the atmosphere reduces this problem, but running a terrestrial biosphere with a fixed atmospheric boundary condition (and not coupled to a atmospheric chemistry-transport model) is still a simplification that prevents biosphere-atmosphere feedbacks and therefore to potential discrepancies between vegetation and CTM model. Not accounting for this feedback and stomatal and non-stomatal O_3 deposition might result in an overestimation of O_3 uptake and hence potential damage in the vegetation model. The deposition scheme in OCN offers the potential to couple vegetation and CTM modelling and is thus a step forward towards coupled atmosphere-vegetation simulations.

4.2 Estimating vegetation damage from ozone uptake

A key aspect of ozone damage estimates are the assumed dose-response-relationships, which relate ~~ozone~~ O_3 uptake to plant damage. The use of flux-based relationships is generally thought to improve damage estimates compared to concentration based metrics (e.g. AOT40), since stomatal constraints on O_3 uptake are taken into account, yielding very different spatial patterns of exposure hot spots (Simpson et al., 2007) . Similar to Simpson et al. (2007) , we find strongly differing patterns between cumulative O_3 uptake (CUO) and AOT40 in our simulations here (see Fig. 6), where highest exposure is not only found in southern Europe where the O_3 concentration is highest but also in eastern Europe.

Several dose-response-relationships exist for biomass or yield damage (LRTAP-Convention, 2010, for an overview), ~~but only few estimates exists for~~ there are few estimates of the likely cause of this damage, i.e. the reduction in net photosynthesis. In this study, the damage relationship to net photosynthesis proposed by Wittig et al. (2007) is used. The major advantage of this relationship is that it has been obtained by meta-analysis of many different tree species and thus might indicate an average response. This relationship is therefore used for all modelled plant functional types. However, a substantial disadvantage is that the meta-analysis implies a damage of 6.16 % at zero accumulated ~~ozone~~ O_3 uptake with a rather minor increase in damage with increasing ~~ozone uptake.~~

~~The use of flux-based damage estimates is generally thought to improve damage estimates compared to concentration based estimates~~ O_3 uptake. This might be an important factor explaining the lower ozone damage estimates of OCN compared to other terrestrial biosphere models. In Lombardozzi et al. (2015) also a damage relationship derived from a meta-analysis is used however the disadvantage of predicted ozone damage at zero accumulated O_3 uptake there is even stronger compared to Wittig et al. (2007) . Two out of three modelled plant functional types assume -12.5 % and -16,1 % ozone damage at zero accumulated O_3 uptake (broadleaved and needle-leaved species respectively) and the third plant functional type (grass and crop) assumes 19.8 % at zero accumulated O_3 uptake together with a small increase in damage with increasing O_3 uptake (Lombardozzi et al., 2015) . An evaluation of the different proposed damage functions implemented in terrestrial biosphere models (e.g. ~~AOT40) , since stomatal constraints on ozone uptake are taken into account, yielding very different spatial patterns of exposure hot spots (Simpson et al., 2007) . Similar to Simpson et al. (2007) , we find strongly differing patterns between cumulative ozone~~ Wittig et al. (2007); Lombardozzi et al. (2015); Sitch et al. (2007)) is necessary to elucidate which are able to e.g. reproduce observed patterns of biomass damage and hence might be suitable to predict regional or global damage estimates.

The use of a (possibly PFT specific) flux threshold and its magnitude naturally also impacts the CUOY (canopy cumulative O_3 uptake above a threshold ~~(CUO$^{1.6}_{5}$) and AOT40 in our simulations here (see Fig. 6), where highest exposure is not only found in southern Europe where the ozone concentration is highest but also in eastern Europe.~~ of Y $\mathrm{nmol\,m^{-2}\,s^{-1}}$) and possible damage estimates (Tuovinen et al., 2007) . The included damage function (Wittig et al., 2007) is designed for the CUO without a flux threshold (Y = 0). The impacts of using different flux thresholds on regional estimates of O_3 uptake, accumulation and damage are still poorly understood and need further research.

~~To elucidate the reasons for the substantial differences in the damage estimates further studies are necessary to disentangle the combined effects of differing flux thresholds, damage relationships, climate change, and nitrogen deposition which acts as a ozone precursor on the one hand and a growth enhancing nutrient on the other hand~~It should be noted that using plant O_3 uptake based on leaf-level O_3 concentrations, as done here, together with empirical ozone-damage functions, where O_3 uptake is calculated from atmospheric O_3 concentrations, introduces a discrepancy. The O_3 uptake rates of the experiments forming the damage relationship however are calculated from mean ozone concentrations e.g. over the exposure period and the respective average stomatal conductances (Wittig et al., 2007) such that the estimated O_3 uptake and cumulated uptake used to derive the damage relationship are coarse approximations and underlie considerable uncertainty. The error introduced in OCN by using leaf-level O_3 concentrations instead of atmospheric concentrations seems small, especially since the use of the leaf-level O_3 concentration is the physiological more appropriate approach.

In the current version of OCN only ozone damage to net photosynthesis is accounted for. Other processes like detoxification of O_3 and injury repair (Wieser and Matyssek, 2007; Ainsworth et al., 2012) , stomatal sluggishness (Paoletti and Grulke, 2010) and early senescence (Gielen et al., 2007; Ainsworth et al., 2012) are not accounted for. Decoupling of photosynthesis and stomatal conductance ~~might also~~ (e.g. through stomatal sluggishness) might impact GPP and transpiration damage estimates and ~~should be regarded too~~requires further analysis. Accounting for direct impairment of the stomata might reduce the reported reductions in transpiration or even cause an increase compared to simulations with no ozone damage. Reduced carbon gain due to early senescence might impact the growth and biomass accumulation of plants (Gielen et al., 2007; Ainsworth et al., 2012) and ought also be included in terrestrial biosphere models.

5 Conclusion

Estimates of ~~ozone~~ O_3 impacts on plant gross primary productivity vary substantially. This uncertainty in the magnitude of damage and hence the potential impact on the global carbon budget is related to different approaches to model ozone damage. The use of a comparatively detailed ozone deposition scheme that accounts for non-stomatal as well as stomatal deposition, when calculating surface O_3 concentrations substantially ~~impacts ozone~~ affects O_3 uptake in our model. We therefore recommend ~~to generally consider~~ that non-stomatal ~~ozone uptake in models estimating~~ O_3 uptake is routinely included in model assessments of ozone damage to obtain a better estimate of ozone uptake and accumulation. We show that ~~ozone~~ O_3 uptake into the stomata is mainly ~~impacted~~ determined by the canopy conductance in the ~~used~~ ozone deposition scheme used here. This highlights the importance of ~~modelling reliable canopy conductances besides~~ reliable modelling of canopy conductances as well as realistic surface O_3 concentrations to obtain as accurate as possible estimates of ~~ozone~~ O_3 uptake which are the basis for plant damage estimates. Suitable ozone damage relationships to net photosynthesis for different plant groups are essential to relate the accumulated ~~ozone~~ O_3 uptake to plant damage in a model. ~~Desirable are mean~~ Mean responses of plant groups similar to commonly modelled plant functional types are also desirable. Only few relationships exist which indicate mean responses of several species e.g. Wittig et al. (2007) and Lombardozzi et al. (2013) which however propose very different relationships. Furthermore, the impact of the plants ability to detoxify ~~ozone should be regarded~~ O_3 should be considered e.g. by using flux thresholds, as well as the combined effects of ~~ozone~~ O_3 with air pollution (nitrogen deposition) and climate change (elevated CO_2) on the plants carbon uptake.

Appendix A: ~~aerodynamic resistance(Appendix material)~~Aerodynamic Resistance

To calculate the ~~ozone~~ O_3 deposition of the free atmosphere at the lowest level of the CTM (approximately 45 m) to the vegetation canopy, it is necessary to know the aerodynamic resistance between these heights ~~.~~ ($R_{a,45}$). ~~This data is~~ These data are model and land-cover specific, and thus not provided by the CTM. Instead, we approximate $R_{a,45}$ from the wind speed ~~in~~ at 45 m height (u_{45}) and the friction velocity u_* according to

$$R_{a,45} = \frac{u_{45}}{{u_*}^2} \tag{A1}$$

where u_* is calculated from the wind speed at 10 m height (u_{10}) using the atmospheric resistance calculations of the ORCHIDEE model (Krinner et al., 2005). The wind at 45 m (u_{45}) is approximated by assuming the logarithmic wind profile for neutral atmospheric conditions (Monteith and Unsworth, 2007) due to the lack of information on any other relevant atmospheric properties ~~in~~ at 45 m height:

$$u_{45} = u_{10} \frac{log(\frac{45}{z_0})}{log(\frac{10}{z_0})} \tag{A2}$$

where z_0 is the roughness length.

Appendix B: Emissions inventory

Emissions for the EMEP model were derived by merging data from three main sources. Firstly, emissions for 2005 and 2010 were taken from the so-called ECLIPSE database produced by IIASA for various EU Projects and the Task Force on Hemispheric Transport of Air Pollution (Amann et al., 2013; Stohl et al., 2015), although with improved spatial resolution over Europe by making use of the 7 km resolution MACC-2 emissions produced by TNO (Kuenen et al., 2011). For 1990, emissions from land-based sources were taken directly from the EMEP database for that year, since 1990 had been the subject of recent review and quality-control (e.g. Mareckova et al., 2013). Emissions between 1990 and 2005 were estimated via linear interpolation between these 2005 and EMEP 1990 values. Emissions prior to 1990 were derived by scaling the EMEP 1990 emissions by the emissions ratios found in the historical data-series of Lamarque et al. (2010).

Emissions of the biogenic hydrocarbon isoprene from vegetation are calculated using the model's land cover and meteorological data ~~(?Simpson et al., 1999)~~ (Simpson et al., 2012, 1999) . Emissions of NO from biogenic sources (~~NO from~~ soils, forest-fires, etc) were set to zero given both their uncertainty and sporadic ~~occurance~~occurrence. Tests have shown that this approximation has only a small impact on annual deposition totals to the EU area, even for simulations at the start of the 20th century. Volcanic emissions of sulfur dioxide (SO_2) were set to a constant value from the year 2010.

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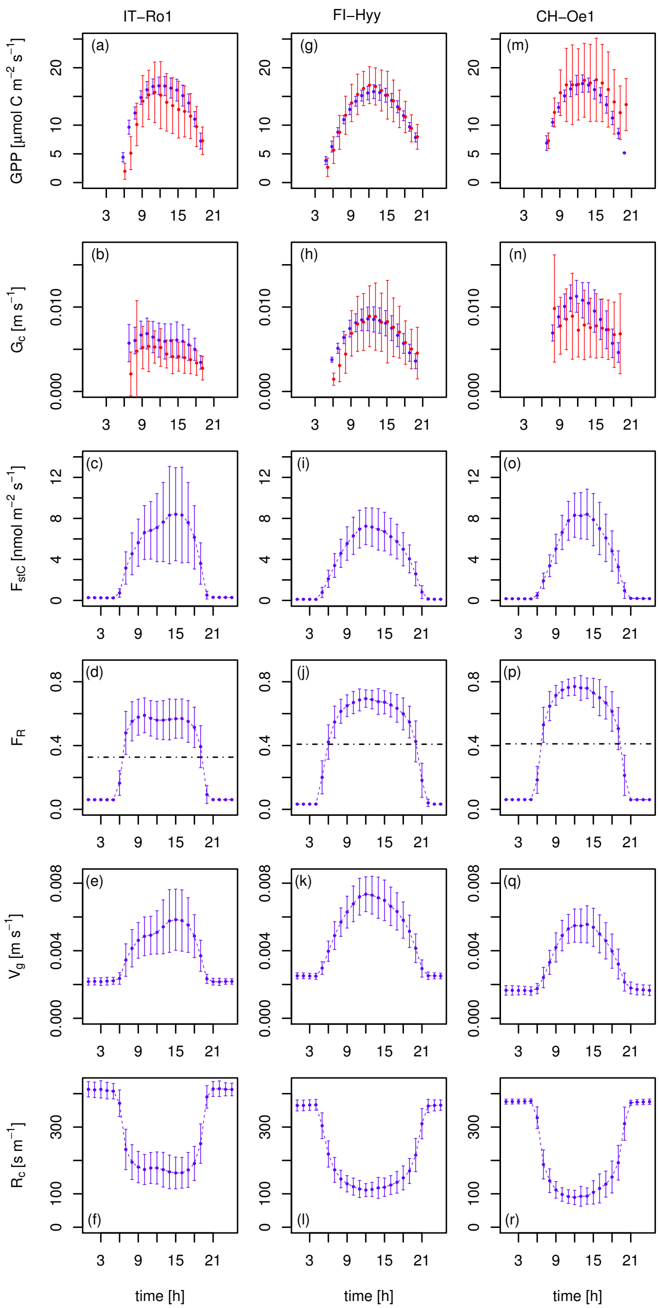

Figure 2. Simulated and observed hourly means over all days of the July months ~~2002 – 2006~~ 2002-2006 for CH-Oe1 and IT-Ro1, and for ~~2001 – 2006~~ 2001-2006 for FI-Hyy. Plotted are mean hourly values (local time) of a,g,m) GPP (blue: OCN, red: FLUXNET), b,h,n) canopy conductance (G_c) (blue: OCN, red: FLUXNET), c,i,o) ~~ozone~~ O_3 uptake (F_{stC}), d,j,p) the flux ratio (F_R), e,k,q) ~~ozone~~ O_3 deposition velocity (V_g) and f,l,r) ~~ozone~~ O_3 surface resistance (R_c). The error bars indicate the standard deviation from the hourly mean. The dotted line in d,j,p) indicates the daily mean value.

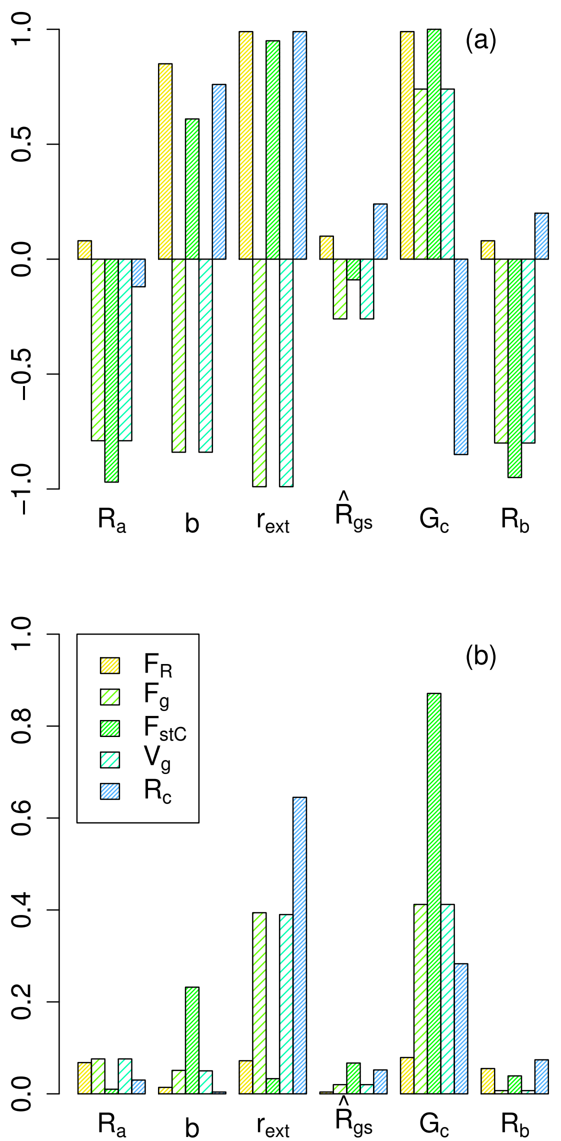

Figure 3. a) Mean partial correlation coefficients and b) strength of the correlation in % per %. R_a, b, r_{ext}, $\hat{R}_{gs}$ and G_c are perturbed within ~~±20%~~ ±20% of their central estimate. Results from simulations at the FLUXNET site FI-Hyy for the simulation period 2001-2006.

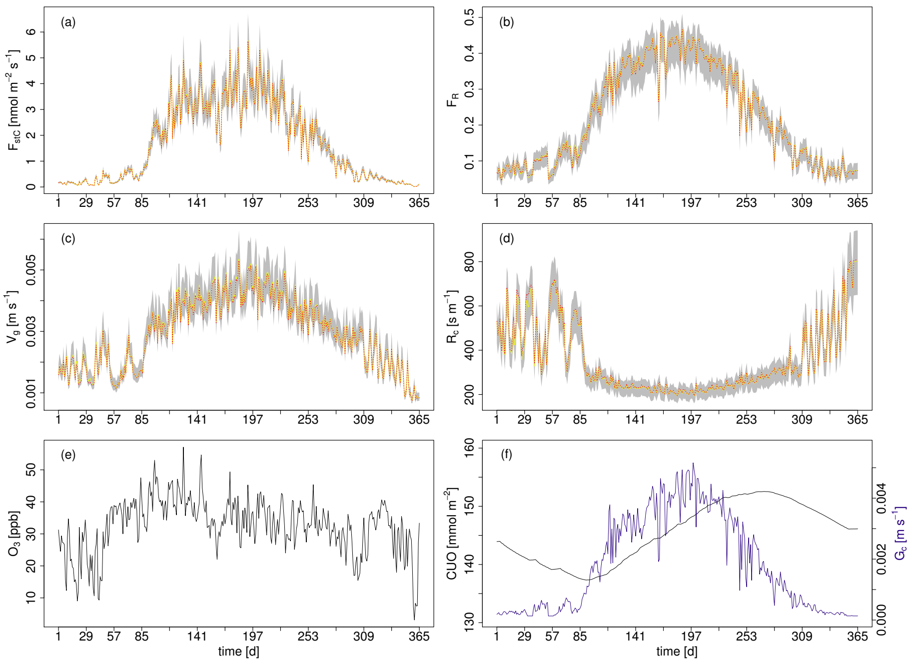

Figure 4. Ensemble range of key ~~ozone~~ O_3 uptake/deposition variables resulting from the perturbation of R_a, b, r_{ext}, $\hat{R}_{gs}$ and G_c within ~~±20%~~ ±20% of their central estimate. Shown are simulated daily mean values of a) ~~ozone~~ O_3 uptake (F_{stC}), b) the ~~ozone~~ O_3 flux ratio (F_R), c) ~~ozone~~ O_3 deposition velocity (v_g) and d) ~~ozone~~ O_3 surface resistance (R_c) for the boreal needle-leaved evergreen forest at the finish FLUXNET site FI-Hyy for the year 2001. Red dashed: unperturbed model; yellow: median of all sensitivity runs; ~~dark grey area: interquartile-range;~~ light grey area: min-max-range off all sensitivity runs. Simulated daily mean values for the respective site and year of e) atmospheric ~~ozone~~ O_3 concentrations O_3 and f) cumulative uptake of ~~ozone~~ O_3 (CUO) and canopy conductance G_c.

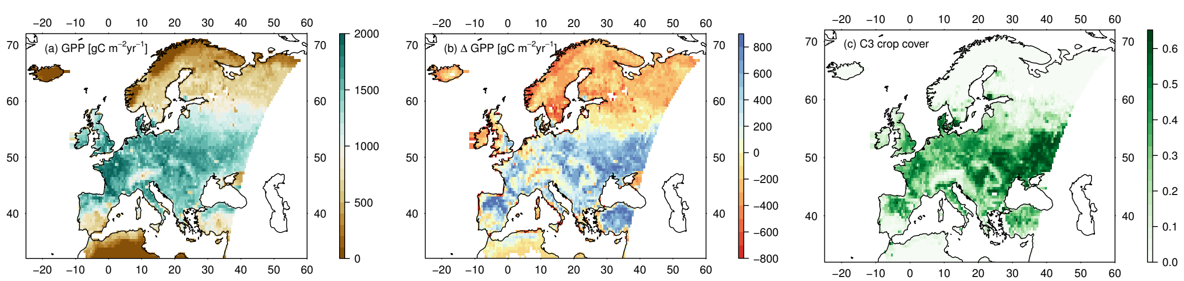

Figure 5. Europe-wide simulated GPP and difference between modelled GPP by OCN and a GPP estimate by a FLUXNET-MTE-product. Plotted are for the years 1982-2011 a) the simulated mean GPP accounting for ozone damage in $\mathrm{g\,C\,m^{-2}\,yr^{-1}}$, b) the mean differences for OCN - MTE GPP in $\mathrm{g\,C\,m^{-2}\,yr^{-1}}$ and c) the mean simulated grid cell cover of the C3-crop PFT in OCN, given as fractions of the total grid cell area.

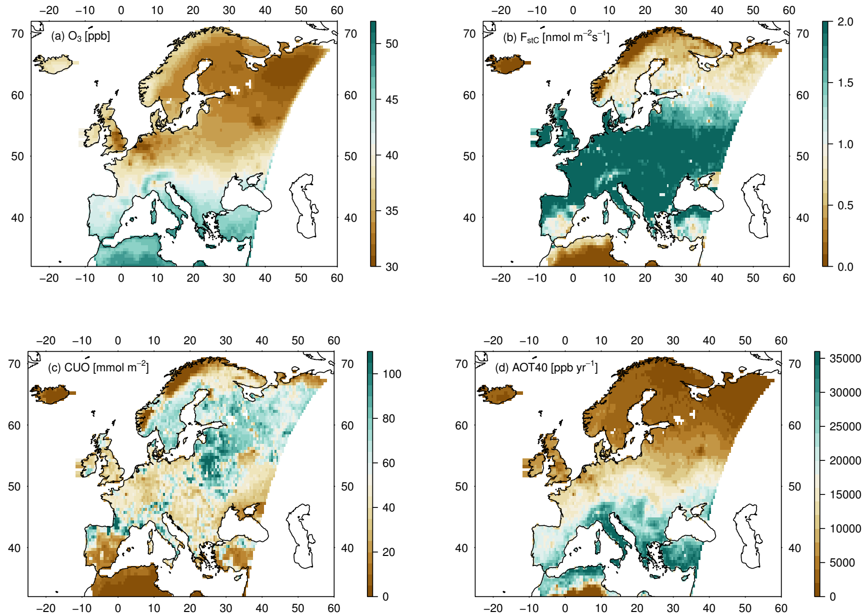

Figure 6. Mean decadal a) ~~ozone~~ $\mathrm{O_3}$ concentration [ppb], b) canopy integrated ~~ozone~~ $\mathrm{O_3}$ uptake into the leafs [$\mathrm{nmol\,m^{-2}\,s^{-1}}$], c) canopy integrated cumulative uptake of ~~ozone above a threshold~~ $\mathrm{O_3}$ ($\mathrm{CUO}_{1.6\,\cancel{5}}$) [$\mathrm{mmol\,m^{-2}}$] and d) AOT40 [$\mathrm{ppm\,yr^{-1}}$], for Europe of the years 2001-2010.

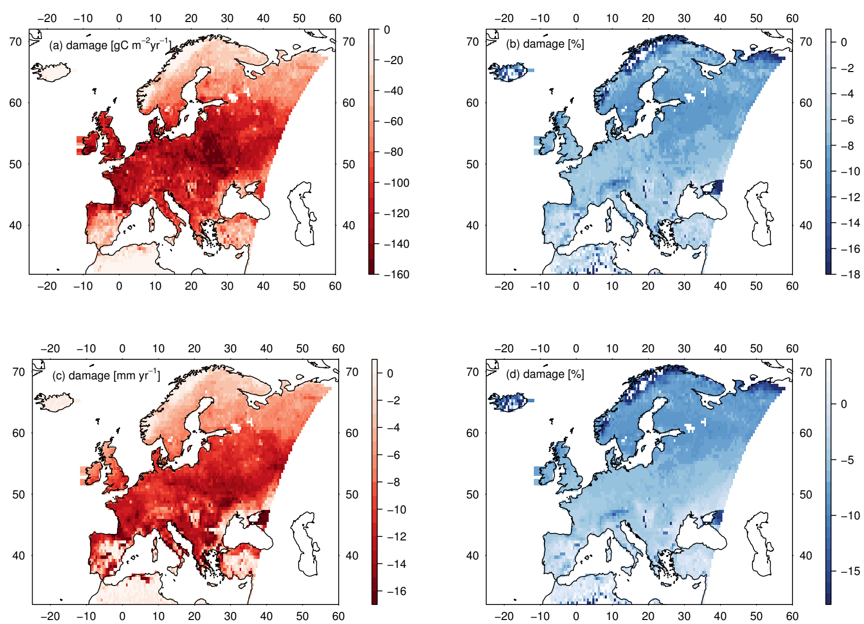

Figure 7. Mean decadal a) reduction in GPP [$\mathrm{g\,C\,m^{-2}\,yr^{-1}}$], b) percent reduction in GPP, c) reduction in transpiration [$\mathrm{mm\,yr^{-1}}$] and d) percent reduction in transpiration due to ozone damage averaged for the years 2001-2010.

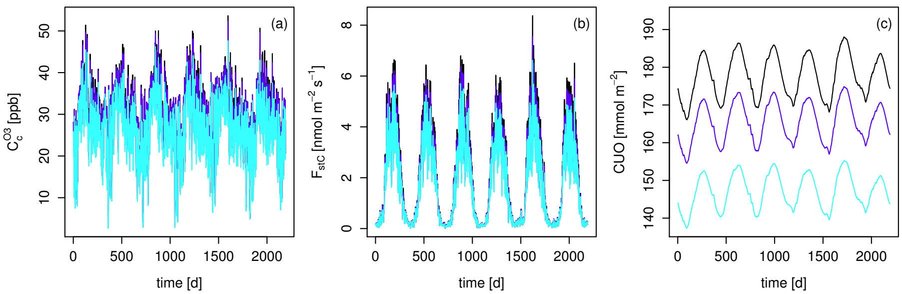

Figure 8. Mean daily values of the a) ~~ozone~~ O_3 surface concentration [ppb], b) canopy integrated ~~ozone~~ O_3 uptake into the leafs [$\mathrm{nmol\,m^{-2}\,s^{-1}}$], ~~c) canopy integrated cumulative uptake of ozone (CUO)~~ and ~~d~~c) canopy integrated cumulative uptake of ~~ozone above a threshold~~ O_3 (CUO~~1.6~~) [$\mathrm{mmol\,m^{-2}}$] at the FLUXNET site FI-Hyy. Black: ATM model, Dark blue: D-STO model, Light blue: standard deposition model (D).

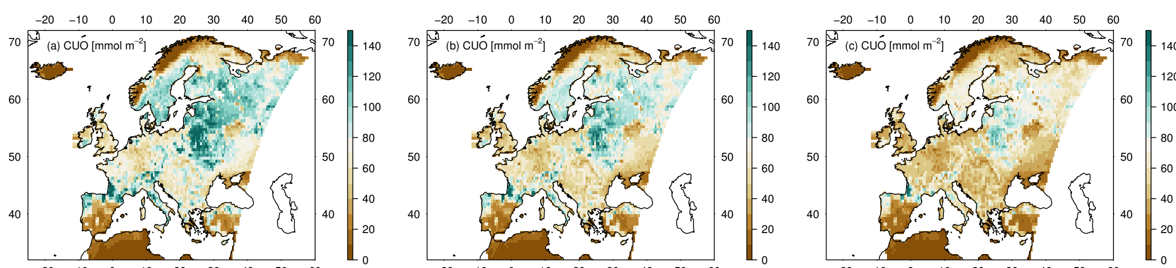

Figure 9. Mean decadal canopy integrated cumulative uptake of ~~ozone~~ O_3 (CUO) [$\mathrm{mmol\,m^{-2}}$] for Europe of the years 2001-2010. a) ~~no ozone deposition scheme~~ canopy O_3 concentration is equal to the atmospheric concentration (ATM), b) ~~ozone~~ O_3 surface resistance is only determined by stomatal resistance (D-STO) and c) standard ozone deposition scheme (D).

Table 1. Characteristics of the FLUXNET sites used in this study.

	Site	Latitude	Longitude	Climate [a]	PFT [b]	Years	Reference
1	AT-Neu	47.12	11.32	Cfb	TeH	2002- 2005	(Wohlfahrt et al., 2008b)
2	CH-Oe1	47.29	7.73	Cfb	TeH	2002- 2006	(Ammann et al., 2007)
3	DE-Bay	50.14	11.87	Cfb	CEF	1997- 1998	(Rebmann et al., 2004)
4	DE-Hai	51.08	10.45	Cfb	TeBDF	2000- 2006	(Kutsch et al., 2008)
5	DE-Meh	51.28	10.66	Cfb	TeH	2004- 2006	(Scherer-Lorenzen et al., 2007)
6	DE-Tha	50.96	13.57	Cfb	CEF	2004- 2006	(Grünwald and Bernhofer, 2007)
7	DK-Lva	55.68	12.08	Cfb	TeH	2005- 2006	(Gilmanov et al., 2007)
8	DK-Sor	55.49	11.65	Cfb	TeBDF	1997- 2006	(Lagergren et al., 2008)
9	ES-ES1	39.35	-0.32	Csa	CEF	1999- 2004	(Sanz et al., 2004)
10	FI-Hyy	61.85	24.29	Dfc	CEF	2001- 2006	(Suni et al., 2003)
11	FR-Hes	48.67	7.06	Cfb	TeBDF	2001- 2006	(Granier et al., 2000)
12	FR-LBr	44.72	-0.77	Cfb	CEF	2003- 2006	(Berbigier et al., 2001)
13	FR-Pue	43.74	3.60	Csa	TeBEF	2001- 2006	(Keenan et al., 2010)
14	IL-Yat	31.34	35.05	BSh	CEF	2001- 2002	(Grünzweig et al., 2003)
15	IT-Cpz	41.71	12.38	Csa	TeBEF	2001- 2006	(Tirone et al., 2003)
16	IT-Lav	45.96	11.28	Cfb	CEF	2006- 2006	(Marcolla et al., 2003)
17	IT-MBo	46.02	11.05	Cfb	TeH	2003- 2006	(Wohlfahrt et al., 2008a)
18	IT-PT1	45.20	9.06	Cfa	TeBDF	2003- 2004	(Migliavacca et al., 2009)
19	IT-Ro1	42.41	11.93	Csa	TeBDF	2002- 2006	(Rey et al., 2002)
20	IT-Ro2	42.39	11.92	Csa	TeBDF	2002- 2006	(Tedeschi et al., 2006)
21	IT-SRo	43.73	10.28	Csa	CEF	2003- 2006	(Chiesi et al., 2005)
22	NL-Loo	52.17	5.74	Cfb	CEF	1997- 2006	(Dolman et al., 2002)
23	PT-Esp	38.64	-8.60	Csa	TeBEF	2002- 2006	(Pereira et al., 2007)
24	PT-Mi1	38.54	-8.00	Csa	TeS	2003- 2005	(Pereira et al., 2007)
25	SE-Fla	64.11	19.46	Dfc	CEF	2000- 2002	(Lindroth et al., 2008)
26	SE-Nor	60.09	17.48	Dfb	CEF	1996- 1997	(Lagergren et al., 2008)

[a] Koeppen-Geiger climate zone (BSh = hot arid steppe; Cfa = humid, warm temperate, hot summer; Cfb = humid, warm temperate, warm summer; Csa = summer dry, warm temperate, hot summer; Dfb = Cold, humid, warm summer; Dfc = Cold, humid, cold summer).

[b] Plant functional type (TeBEF = Temperate broadleaf evergreen forest, TeBDF = Temperate broadleaf deciduous forest, CEF = Coniferous evergreen forest, TeS = Temperate open woodland with C3 grass, TeH = C3 grassland).

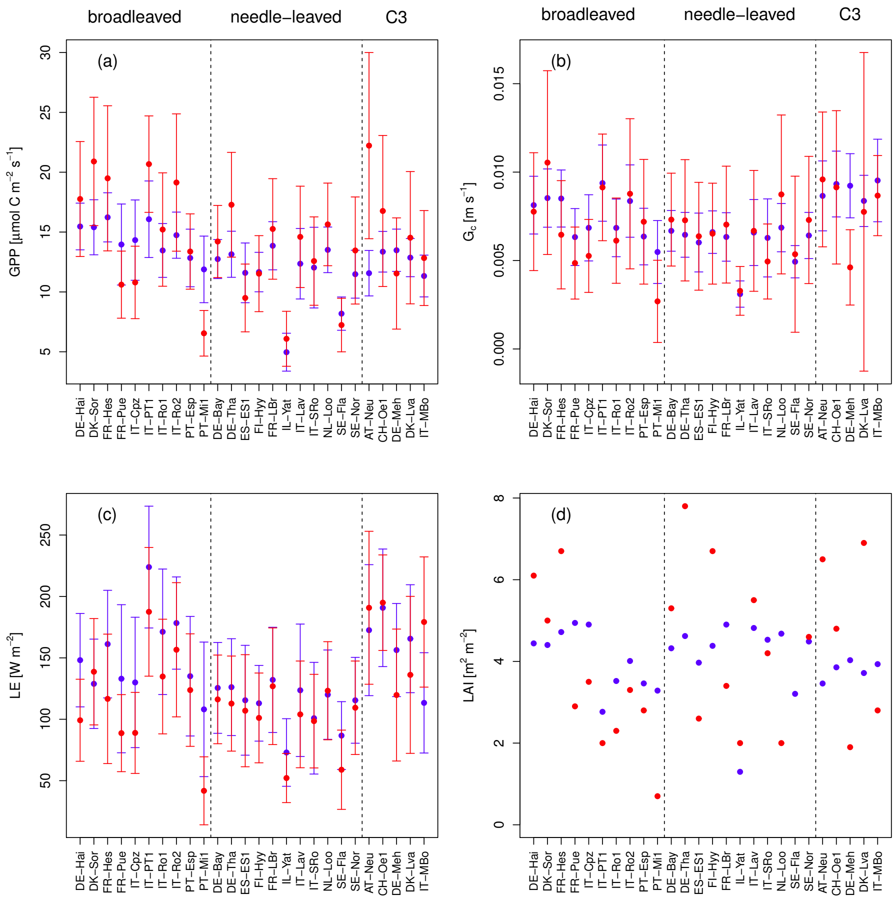

Figure 10. Comparison of measured a) GPP, b) G_c, c) latent heat fluxes (LE) and d) LAI at 26 European FLUXNET sites (red) and simulations by OCN (blue). Displayed are means and standard deviation of daily means of the measuring/simulation period, with the exceptions of FLUXNET derived LAI, which is based on point measurements.

Table 2. ~~Plotted are~~ Coefficient of determination (R^2) and Root Mean Square Error (RMSE) for GPP, canopy conductance (G_c), and latent heat fluxes (LE) for all sites, sites dominated by broadleaved trees, needle-leaved trees, C3 grass, and C3 grass except of the ~~differences between~~ AT-Neu site (outlier).

		all sites	broadleaved	needle-leaved	C3 grass	C3 grass (except AT-Neu)
1	R^2: GPP	0.465	0.714	0.8	0.139	0.058
2	$RMSE$: GPP	3.495	3.771	1.944	5.175	2.257
3	R^2: G_c	0.458	0.69	0.722	0.013	0.01
4	$RMSE$: G_c	0.001	0.002	0.001	0.002	0.002
5	R^2: LE	0.566	0.725	0.9	0.022	0.002
6	$RMSE$: LE	30.897	39.725	13.977	37.124	40.493

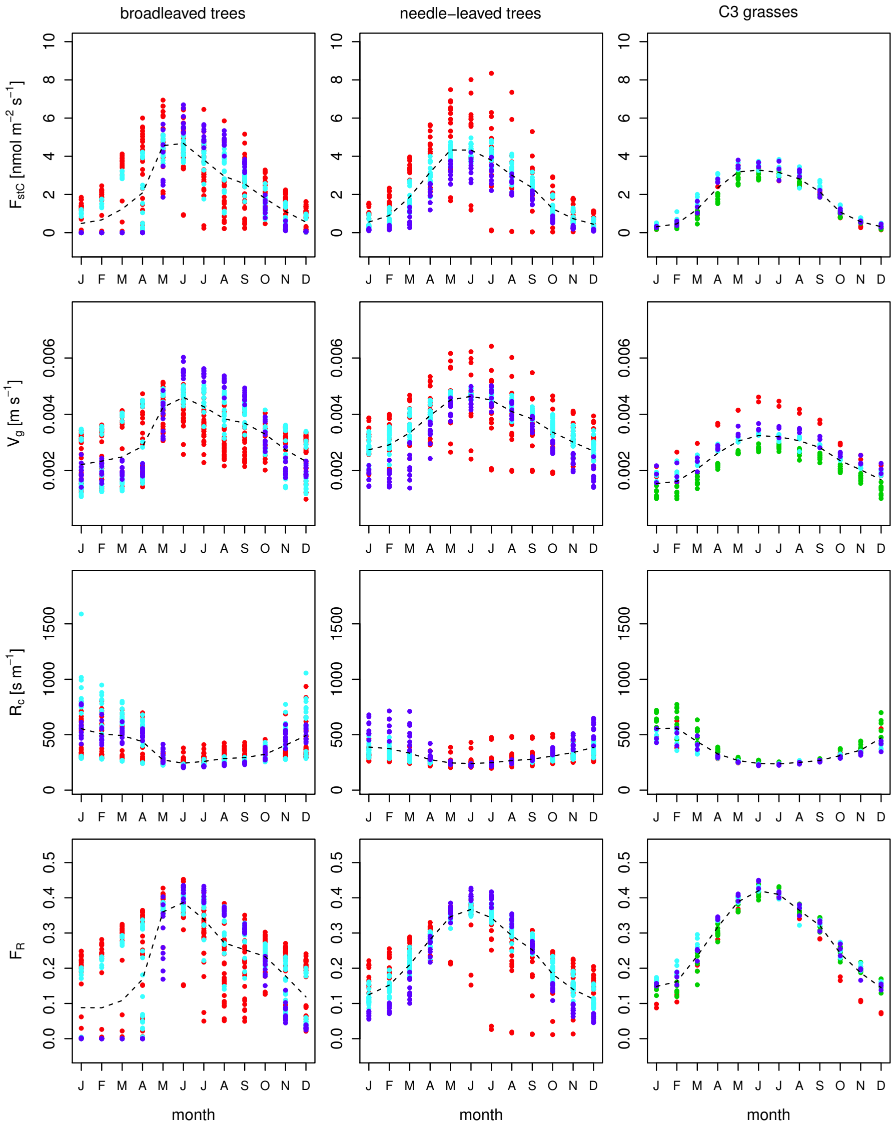

Figure 11. Simulated monthly mean values of O_3 uptake (F_{stC}), O_3 deposition velocity (V_g), O_3 surface resistance (R_c) and the ~~D~~flux ratio (F_R) for sites dominated by broadleaved trees (left column), ~~D-STO~~ needle-leaved trees (central column) and ~~ATM model version~~C3 grasses (right column). The colour indicates the location of the site: Denmark, Sweden, and Finnland (dark blue); Germany, France, and Netherlands (light blue); Austria, and Switzerland (green), and Italy, Portugal, Spain and Israel (red). Brocken line: Mean of all sites and years of the 12 months.

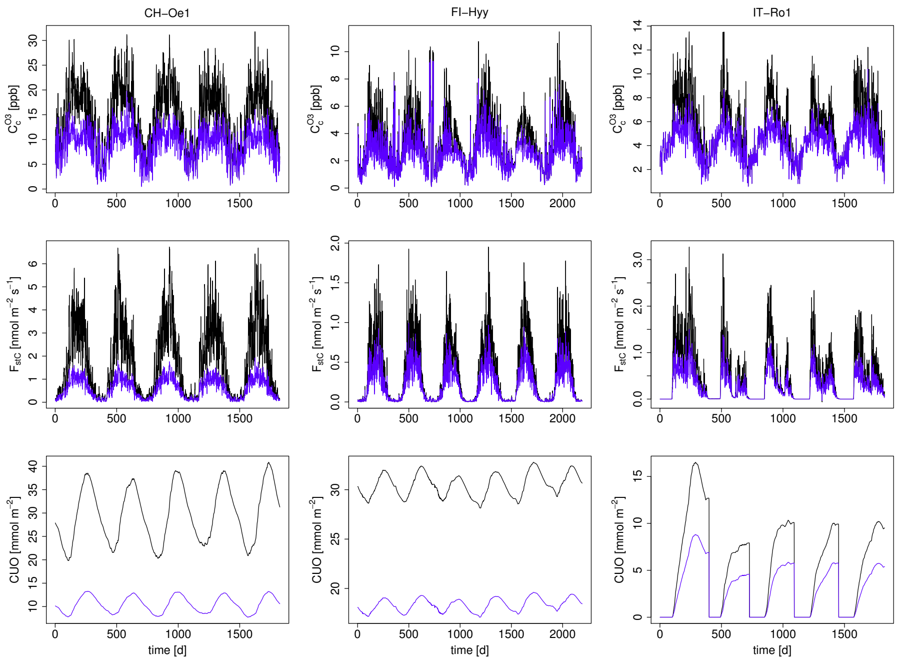

Figure 12. Differences in mean daily values of the a) ~~ozone~~ O_3 surface concentration [ppb], b) canopy integrated ~~ozone~~ O_3 uptake into the leafs [$\mathrm{nmol\,m^{-2}\,s^{-1}}$], ~~c) canopy integrated cumulative uptake of ozone (CUO)~~ and ~~d~~c) canopy integrated cumulative uptake of ~~ozone above a threshold~~ O_3 ($\mathrm{CUO}_{\cancel{1.6}\,5}$) [$\mathrm{mmol\,m^{-2}}$] for the three FLUXNET sites CH-Oe1, FI-Hyy and IT-Ro1. Blue: Difference between the D-STO model and the standard model (D), Black: Difference between the ATM model and the standard model (D).

~~Europe-wide simulated mean cover fractions of C4 plant functional types for the years 2001-2010. a) mean simulated grid cell cover of the C4-grass PFT in OCN, b) mean simulated grid cell cover of the C4-crop PFT, both given as fractions of the total grid cell area.~~

~~Mean decadal canopy integrated cumulative uptake of ozone above a threshold ($\mathrm{CUO}_{5}^{1.6}$) for Europe of the years 2001-2010. a) no ozone deposition scheme (ATM), b) ozone surface resistance is only determined by stomatal resistance (D-STO) and c) standard ozone deposition scheme (D).~~

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