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Article type : Essay Review

Handling Editor: Kun-Fang Cao

## **Ozone effects on European forest growth – towards an integrative approach**

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Running head: Ozone effects on European forest growth

Keywords: ozone, forest, growth, monitoring, modeling, experiments, global change ecology

### **Abstract**

1. The phytotoxic impacts of tropospheric ozone on cellular and molecular processes are increasingly understood and quantified. They generally lead to a decrease in plant carbon uptake that is mainly detectable at leaf scale. At larger scales, most of the empirical and modeling studies reported negative ozone effects on productivity, but

This article has been accepted for publication and undergone full peer review but has not been through the copyediting, typesetting, pagination and proofreading process, which may lead to differences between this version and the Version of Record. Please cite this article as doi:

10.1111/1365-2745.12941

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the latest empirical studies and model developments temper these observations and simulations.

2. Ozone impacts on European forest growth are controversial and seem negligible because of (i) differences among tree sizes and species, sites, data sources, and methodological approaches, (ii) the moderate link between adult tree growth and carbon uptake, (iii) the presence of several confounding factors such as drought or nitrogen fertilization, and of legacy effects of past land-use and management on stand development, and (iv) the presence of compensatory processes such as tree acclimation, population adaptation, and changes in competition intensity and species composition.
3. We provide a comprehensive review of the empirical and modeling approaches available for ozone risk assessment by detailing their respective main outputs, advantages and limits, and examine research gaps.
4. *Synthesis*. To disentangle between the forcing factors and to better understand ozone impacts at each ecosystem level and feedbacks across levels, and to reinforce the strength of ozone-impact predictions, we recommend to combine ozone-controlled experiments and long-term monitoring data with physiological and forest succession process-based models.

## **Introduction**

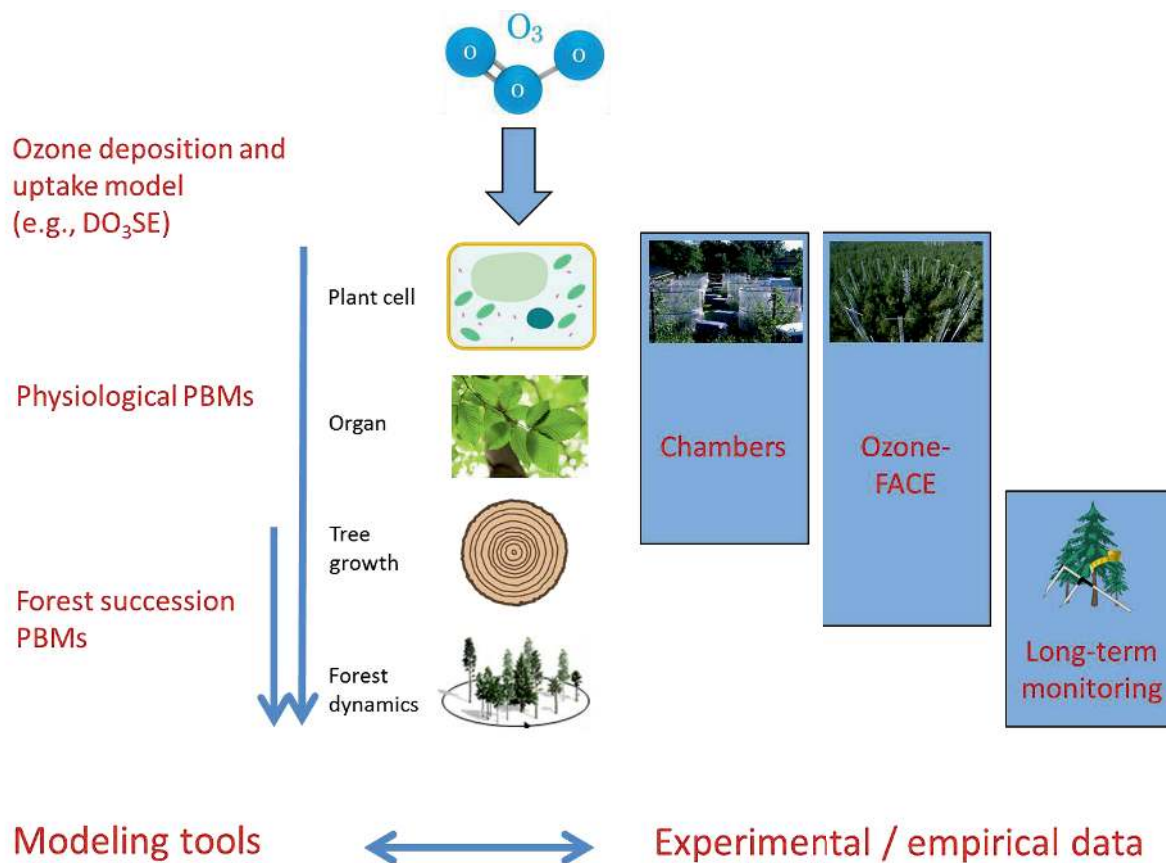
Tropospheric ozone is considered to be currently more damaging to vegetation than any other air pollutant, impacting air quality and various ecosystem functions and services (Ainsworth et al., 2012; Sitch et al., 2007; Matyssek et al., 2010a; Fuhrer et al., 2016). As a strong oxidant, ozone has a negative impact on many cellular and molecular processes (see Jolivet et al., 2016). Among others, ozone induces an alteration of Rubisco activity and

content through the formation of reactive oxygen species (Saxe, 2002), a reduction in stomatal conductance (Ainsworth et al., 2012), leaf chlorosis and early senescence (Novak et al., 2003; Waldner et al., 2007; Sicard et al., 2016b; but see Ferretti et al., 2018). All these effects combined may decrease carbon gain at leaf- (Novak et al., 2007), tree- (Wittig et al., 2009), and stand-scales (Matyssek et al., 2010b; but see Verryckt et al., 2017) if ozone concentration levels are high enough (see Fares et al., 2013b). Based on remote sensing data, Proietti et al. (2016) estimated the negative impact of ozone on Gross Primary Production of European forests from 2000 to 2010 to range between 0.4 and 30%.

However, considering that tree growth is mainly sink-driven (Körner, 2015), i.e., that reductions in tree carbon uptake and increases in respiration through stress- or pest-induced defense and repair systems may not necessarily directly reduce its growth (Fatichi et al., 2014; Delpierre et al., 2016a), ozone effects on annual and decadal individual tree radial growth may be negligible. In addition, ozone is involved in ecosystem nitrogen (N)- and carbon (C)-cycles in a complex way, and its impacts on vegetation often interact or counteract with the effects of climate, atmospheric deposition of N or other pollutants, and biotic stressors. This interdependency among environmental drivers ultimately leads to complex synergistic and antagonistic relationships that are difficult to disentangle and predict (Simpson et al., 2014; De Marco et al., 2016; Mills et al., 2016; Etzold et al., in prep.). For instance, ozone concentration is usually positively correlated with air temperature and N deposition (De Vries et al., 2014a), which both can be related to increased tree growth in various geographical regions (e.g., de Vries et al., 2014b; Pretzsch et al., 2014). Also, depending on species-specific stomatal regulation strategy, drought-induced stomatal closure may reduce ozone uptake (Jolivet et al., 2016). In the long-term, O<sub>3</sub> effects on forest growth may also be potentially reduced by acclimation/adaptation processes (Zak et al., 2011), and compensated by changes in competition intensity and species composition (Wang et al., 2016).

Recent projections by regional chemistry models highlight a likely decrease in ozone concentrations in Europe by the end of the century (Watson et al., 2016), which was already identified by measurements over the last decade (Schaub et al., 2015). Yet, ozone levels are, and should be, still high enough to impact forest ecosystems (Simpson et al., 2014; Karlsson et al., 2017), which, in addition, might add to the adverse impacts of the increased frequency of heat and summer drought events in future (IPCC, 2014). In consequence, there is an urgent need to (i) better quantify ozone uptake by forests and its impacts on their growth by deriving robust and realistic species-specific dose-response relationships (DRRs) based on experimental and monitoring data; and (ii) develop modeling tools that can upscale these functions at longer temporal and larger spatial scales, and at a higher level of complexity integrating feedbacks across scales. These information and tools should necessarily respect interactions with confounding factors such as drought or nitrogen deposition, and would be highly useful to derive robust projections of ozone effects under varying environmental conditions, and to adapt forest management strategies in the face of global change.

On the basis of reviews and opinion papers on ozone-impacts on tree physiology and forest ecosystems (e.g., Andersen, 2003; Matyssek and Sandermann, 2003; Ashmore, 2005; Paoletti, 2006; Ferretti et al., 2007; Ainsworth et al., 2012; De Vries et al., 2014a; Simpson et al., 2014; Fuhrer et al., 2016; Jolivet et al., 2016; Sicard et al., 2016a) and on up-scaling considerations (e.g., Kolb and Matyssek, 2001; Samuelson and Kelly, 2001; Karnosky et al., 2005; Matyssek et al., 2008), we suggest research directions from a ‘forest growth’ rather than from an ‘air pollution’ perspective. Based on the recent advances in our understanding of tree physiology and on the latest modeling developments, we provide a roadmap to quantify ozone effects on forest growth, from stand to European scale (Fig. 1).



**Figure 1.** Summary of the approaches and data sources available to assess ozone impacts on forest productivity

### Ozone concentration and uptake assessment

The first step to understand and quantify ozone effects is an adequate assessment of ozone concentration, fluxes, and uptake by the canopy.

Different sources of ozone concentration measurements are available, from regional and national (e.g., NABEL network in Switzerland; Brönnimann et al., 2000), to European (e.g., ICP Forests: Schaub et al., 2016; or AirBase: <http://acm.eionet.europa.eu/databases/airbase/>) and global monitoring networks (Tropospheric Ozone Assessment Report database: <https://join.fz-juelich.de>). Note that, in general, ozone concentration is not measured at the top of the canopy layer. Ozone data with the highest quality and temporal resolution ( $\leq$  hourly) usually derive from active analysers, but are available at few forest sites only as maintaining

such monitoring stations is costly and technically challenging. In consequence, to detect spatial and temporal trends in ozone concentration, several studies highlighted the interest of using passive samplers that measure ozone integratively on a bi-weekly or monthly basis (Sanz et al., 2007; Calatayud et al., 2016; Schaub et al., 2016), or using simulated data from atmospheric chemistry-transport models such as EMEP (Simpson et al., 2012), CHIMERE (Bessagnet et al., 2004), and others (see Sharma et al., 2017).

Together with the traditional concentration-based approach that reflects external ozone *exposure* of a given forest entity (leaf, tree, stand) over a growing season, the ozone-flux approach is recommended by the Convention on Transboundary Air Pollution (CLRTAP, 2017) for the assessment of the risk posed by ozone on forest vegetation (Karlsson et al., 2007; Matyssek et al., 2010a; Anav et al., 2016). This method quantifies leaf ozone *uptake*, i.e., the dose that actually enters the plant tissues via stomata, and considers the environmental constraints (i.e., climate, phenology, or soil properties) that may limit optimal stomatal conductance over the growing season. Specifically, the receptor- and species-specific critical phytotoxic ozone dose ( $POD_ySPEC$  in  $\text{mmol m}^{-2}$ ; accumulated ozone stomatal fluxes above a species-specific threshold  $y$ ) is mainly used.  $POD_ySPEC$  values are calculated by estimating ozone deposition and stomatal ozone fluxes based on multiplying the hourly ozone concentrations by the species-specific stomatal diffuse conductance ( $G_{sto}$ ) and assuming null ozone concentration in the sub-stomatal chamber, as does the  $DO_3SE$  (Deposition of  $O_3$  for Stomatal Exchange) model for various vegetation types in Europe (Emberson et al., 2000; Simpson et al., 2012):

$$POD_ySPEC = \sum_{i=1}^N \max \left( G_{sto,i} * \frac{r_{c,i}}{r_{b,i} + r_{c,i}} * [O_3]_i - y, 0 \right) \quad (\text{eq.1})$$

where  $N$  is the number of daylight hours ( $i$ ) of the growing season,  $[O_3]$  is ozone concentration,  $r_c$  and  $r_b$  are the total leaf surface and boundary-layer resistances, respectively, and depend on Leaf Area Index (LAI). Based on an optimization of dose-response relationships (Büker et al., 2015), a uniform  $y$  threshold of  $1 \text{ nmol m}^{-2} \text{ s}^{-1}$  is commonly used. However, considering the lack of information on the detoxification processes and their temporal variability (e.g., Haberer et al., 2007), the use of a constant threshold is still under debate (Anav et al., 2016).

DO<sub>3</sub>SE was continuously re-parameterized and tested during the last decades with a specific focus on the impact of drought on stomatal conductance (e.g., Büker et al., 2012; De Marco et al., 2016). However, some developments are still required to improve both its accuracy and generality, and to extend its applicability from Northern to Southern Europe:

1. DO<sub>3</sub>SE is currently parameterized for a few tree species and functional groups, including *Picea abies*, *Pinus sylvestris*, *Fagus sylvatica*, *Betula* sp., *Populus* sp., Mediterranean and temperate deciduous *Quercus* species, and Mediterranean evergreen species (Büker et al., 2015; CLRTAP, 2017). A further calibration effort is strongly needed to increase the number of species considered, which could benefit from the growing availability of functional trait databases (e.g., Kattge et al., 2011; Lin et al., 2015). Moreover, the intra-specific variability in parameter estimates can be derived from such databases, and can be considered to improve the predictions in ozone uptake and assess their uncertainty by running an ‘ensemble’ of DO<sub>3</sub>SE simulations with different parameter combinations when site-specific estimates are not available.
2. The duration of the period used to accumulate  $POD_ySPEC$  values also needs to be better defined. When leaf phenology data are not available, the start and end of the growing season are simply estimated based on site latitude (CLRTAP, 2017). This latitude model may agree well with field observations (e.g., Zhang et al., 2004), but cannot allow for

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climate change effects on species phenology, and should be thus replaced by species-specific climate-sensitive functions fitted to pan-European phenological databases (e.g., <http://www.pep725.eu/>; see Delpierre et al., 2016b). Similarly, night-time ozone fluxes may be considered (Matyssek et al., 1995; Anav et al., 2016), especially at sites with high night-time ozone like mountainous or coastal sites (Simpson, 2016). This seems especially important since night-time stomatal conductance can make plants in the night to transpire up to 23% of the day-time integrated transpiration (Resco de Dios et al., 2015) and a comparable ratio might be assumed between day and night ozone uptake.

3. The multiplicative Jarvis' stomatal model (1976) modified by Emberson et al. (2000) is usually preferred to (semi-) mechanistic algorithms, mainly because of the lower number of parameters required and because its modular structure makes it easy to implement in larger models (Buckley and Mott 2013; B ker et al., 2007; Anav et al., 2016). Their performance may be similar for ozone-risk assessment at selected sites (e.g., Fares et al., 2013a; Hoshika et al., 2017), but only the photosynthesis-based models take into account effects of CO<sub>2</sub> concentration, surface air temperature and humidity on stomatal conductance. The Ball-Berry model (Ball et al., 1987) – widely used in physiological process-based forest models (Egea et al., 2011) – should be preferred, especially if it is modified to consider for both ozone-induced decoupling between photosynthesis and stomatal conductance (Lombardozzi et al., 2012), and ozone-induced stomatal sluggishness (Keller and H sler, 1984).
4. Considering that many European forest stands are mixed and uneven-aged, and that both stomatal conductance and ozone concentration change within the canopy because of variations in light intensity, canopy wetness, and in the turbulence intensity, the so-called ‘big-leaf’ approach of DO<sub>3</sub>SE may not be explicit enough. Multiple leaf layers should be simulated either explicitly (e.g., Launianen et al., 2013; Zhou et al., 2017) or implicitly



using simple empirically-based canopy ozone gradients based on Leaf Area Index (Ollinger et al., 1997).

### **Empirical approach based on experimental and observational data sources**

Most of the experimental studies reported a negative impact of ozone on tree growth, while the evidence from observational studies is much less clear. Based on a meta-analysis, Wittig et al. (2009) estimated the reduction in biomass production of northern hemisphere forests to amount up to ca. 7% at current ozone levels (in comparison to charcoal-filtered air). However, there is a large variety in the magnitude of this effect among studies, which mainly arises from differences in inter- and intra-specific sensitivity to ozone (De Vries et al., 2014a; Büker et al., 2015). For instance, deciduous species tend to be more sensitive than evergreen ones, mainly because of higher carbon allocation to defense, and of the higher stomatal activity in leaves with low mass per area (Wieser et al., 2013). Another reason for such diversity in growth responses to ozone relies in the type and properties of the data source. Although Wittig et al. (2009) did not find any effect of the fumigation method and of the experiment duration (in days), results of correlative field studies based on long-term monitoring results tend to show less significant impacts than experimental ones (Bussotti and Ferretti, 2009; Ferretti *et al.*, 2014; Karlsson *et al.*, 2006). For instance, *Fagus sylvatica* showed growth reductions by 19.5% and 43.5% at particular Swiss sites (Braun et al., 2014), and at the long-term Kranzberg ozone fumigation experiment (Pretzsch et al., 2010), respectively; while no clear evidence emerged from both studies for *Picea abies*. Ozone effects on stand growth could even become statistically non-significant when considering for the synergistic or antagonistic roles of climate, CO<sub>2</sub> fertilization, nitrogen deposition, soil properties, pathogens, and stand structure (Ferretti et al., 2018; Etzold et al., in prep.).

Clearly, dose-response relationships that derive from short-term continuously-stirred tank reactor (CSTR) or open-top chamber (OTC) experiments where tree individuals – mainly seedlings growing in a non-competitive environment – experienced different ozone levels for a short period, cannot be extrapolated for predicting the response of adult trees growing under real conditions (Kolb and Matyssek, 2001; Ferretti et al., 2007; Ainsworth et al., 2012; see Table 1). The environmental conditions within the canopy of seedlings differ from adult stands, and sensitivity to ozone may change during the ontogenetic development (e.g., Schaub et al., 2005; but see Matyssek et al., 2010a). The ability to buffer phases of reduced C gain is high in adult trees (e.g., Hentschel et al., 2016) due to the huge storage pools, which comprise up to four times the carbon of the leaved canopy (Hoch et al., 2003). However, experiments with seedlings allow for easy replication and are key to improve our understanding of the physiological mechanisms involved with ozone impacts in interaction with other stressors (Table 1), and of the genetic variation in response to ozone (Resco de Dios et al., 2016). Physiological responses of adult trees can be estimated from free-air canopy fumigation experiment sites (FACE; Matyssek et al., 2010a). As maintaining such sites is costly, this technique is implemented only at a few sites and for a few species, but it has also the advantage of assessing ozone impacts at stand-scale for the entire ecosystem. Even so, their results (and those emerging from other observational studies lacking a formal statistical design) can be up-scaled in space and time only if the investigated trees/sites were selected from an explicitly defined and representative population. Indeed, the magnitude of the responses obtained in global change experiments tends to be smaller at larger spatial scales, at longer time periods, and with increasing interactions complexity because of both acclimation and complementarity processes, and because of the presence of other limiting factors (Leuzinger et al., 2011; but see King et al., 2005).

**Table 1:** Data sources available to assess ozone effects on tree individual and stand-scale forest growth.

Main approach	Type	Key papers	Advantages	Limits
Ozone controlled experiment	Individual chambers (CSTR, OTC, branch chamber)	Büker et al., 2015 Resco de Dios et al., 2016 Matyssek et al., 2010 Reich, 1987 Wittig et al., 2009	Possibility to analyze other traits and physiological processes to investigate the mechanisms. No confounding factor. Can be easily replicated. All ozone levels can be tested from zero to unnatural, very high levels. Other treatments can be tested (e.g., drought, elevated CO <sub>2</sub> , nutrients).	Mainly on saplings. Individual scale only. Limited set of species and individuals. Short-term data (a couple of years).
	Stand-scale Free-Air fumigation experiment (Ozone FACE) or greenhouses	Matyssek et al., 2010 Pretzsch et al., 2010 Oksanen, 2003	Possibility to analyze other traits and physiological processes to investigate mechanisms and interactions. No confounding factor. Stand-scale. Both juvenile and adult trees.	Costly and requires long-term monitoring to detect robust results. Qualitative approach: few O <sub>3</sub> levels (usually ambient vs. elevated). Few replicates.
Field observations	Growth data from intensive forest monitoring (ICP Forests Levels I and II) or from forest inventories (NFI; forest reserves) with or without a probabilistic design	Bussotti and Ferretti, 2009 Braun et al., 2014, 2017b Ferretti et al., 2014 Etzold et al., in prep.	Realistic in terms of tree population and ozone exposure. Large environmental gradient covered. Generally higher number of trees/sites.	Limited past temporal (e.g., ICP Forests Level II) or spatial (e.g., NFI) coverage of measured O <sub>3</sub> data. Confounding abiotic and biotic (natural and anthropic) factors have to be considered in the statistical analysis. Tree-rings and dendrometers: only the dominant trees are usually sampled. O <sub>3</sub> extremes (higher than current conditions) are missing. Results may be biased when trees/sites were haphazardly selected and not representative of the overall population (ICP Forests Level II or similar).
	Annual growth data from ring-widths or shoot elongation measurements	Novak et al., 2007 Braun et al., 2007	Long-term growth data (except for dendrometers).	
	Intra-annual growth data from dendrometers.	Kuhn et al., 2015	Dendrometer data can indicate intra-annual changes in cambial activity in response to ozone	

In this aspect, analyzing growth data under field conditions seems highly promising as it is usually available for a high number of individuals, sites, and species. It allows for the coverage of large environmental gradients in terms of climate, soil, and ozone levels, and for the detection of differences related to stand properties. The temporal resolution of available growth data varies from continuous (point dendrometers) to annual (tree-ring) and multi-annual (forest inventory) diameter or height measurements (Table 1). However, to substitute the general limitation of ozone data in terms of temporal extent (usually < 20 years), sample size and spatial distribution should be as large as possible, for instance, by combining growth datasets from aerial surveys (McRoberts and Tomppo, 2007) with pan European- (ICP Forests Level I and Level II networks) and national-scale forest inventories (see Vidal et al., 2016) carried out at both unmanaged and managed plots, and intensive, fully equipped single experimental sites (e.g., Percy and Ferretti, 2004). This will also help in quantifying the intra-specific variability in ozone sensitivity related to differences among size classes (Ferretti et al., 2018). Beyond the recommendations of Braun et al. (2017a) for the development of statistical correlative models, especially regarding the treatment of the confounding factors, we identified five additional suggestions.

First, sampling design – through the selection of the sites and of the trees within each site – is essential for the outcome of the study, and for possible inference at larger scale and across population. For instance, it is well established in forest science that allometric equations must be developed formally taking into account tree size distribution or site fertility (Chave et al., 2004). This implies that the target population of a given study must be clearly identified and representative of the entire population.

Second, different ozone metrics should be tested, as errors accumulate while calculating AOT40 and POD<sub>y</sub>SPEC from [O<sub>3</sub>] (e.g., Ferretti et al., 2018), and as AOT40 estimates may perform better than POD ones (see Büker et al., 2015).

Third, to establish DRRs on the basis of field studies, it will be important to rely as much as possible on measured predictor data (ozone, climate, soil), rather than on modeled ones. Large-scale models are not designed to provide site-relevant estimates for the variable of interest, and thus DRRs calibrated on their outputs may be considerably biased (Ferretti et al., 2007). In case of limited measured data in terms of spatial and temporal extent, modeled data are valuable, but only if the resulting bias is quantified and considered in the interpretation of the results (e.g., Baker et al., 2016).

Fourth, all predictor variables that reflect the abiotic, biotic, and ontogenic characteristics of the trees/stands should be considered as omitting one of them may change the sign of the final relationship (e.g., Ferretti et al., 2014). For example, tree species richness is an emerging important driver of forest productivity (Liang et al., 2016), but was almost ignored in previous studies. We particularly highlight the need to include metrics that reflect historic land use and management, such as past forest cover (Munteanu et al., 2015) or management intensity indices (Kahl and Bauhus, 2014) as their legacy effects on stand regeneration, structure, and composition strongly modulate trajectories of ecosystem change (Perring et al., 2016).

Fifth, as it may not be possible to identify a single most-appropriate correlative model, and to accommodate and quantify uncertainty about model structure and parameterization, we recommend the use and joint interpretation of multiple correlative modeling approaches, from multivariable fixed or mixed effects models to Structural Equation Models, including regressions by RandomForest (see Kühn and Johnson, 2013).

All different approaches spanning from the experimental assessments to the analysis of data from field observations are valuable and complementary as they provide different insights while having their own limits (Table 1). However, theoretically, empirical relationships that emerge from field data are not valuable outside of the calibration domain,

and can barely be scaled up in time and space, especially under varying environmental conditions.

### **Process-based modeling approach**

Process-based models (PBMs) are essential scientific tools to encapsulate disparate pieces of information and knowledge (Mäkela et al., 2000), and consequently to attribute patterns that emerge from field data to the underlying processes and to up-scale impacts of different stressors and disturbances at higher resolution levels (Seidl, 2017). Their explanatory predictions tend to be considered as less reliable in practice than those of empirical models, as they embody many uncertainties and assumptions and require a high number of parameters that are hardly available or measurable in the field (Mäkela et al., 2000; Dormann et al., 2012). However, as they explicitly simulate impacts of multiple stressors in a mechanistic and interactive way, they are less restricted to their calibration domain and can be used to simulate scenarios over large environmental gradients, including extreme values that are not observable in the field (yet). Moreover, as they can consider for feedbacks between ozone uptake and impacts on tree and stand development, and for immediate, lag-, and integrated (i.e., additive, multiplicative, or compensatory) responses, they are more reliable for long-term anticipatory predictions (Mouquet et al., 2015). Although they are rare in Europe (Reyer, 2015), several physiological PBMs have been developed in the last years to predict ozone effects on forest productivity, from the individual to the global scale, while none stand- or landscape-scale hybrid forest succession PBMs have been set up yet (Table 2).

**Table 2:** Examples of process-based models that simulate ozone effects on forest growth.

Spatial resolution	Organization level of the processes impacted by ozone	Model and reference	Which effects due to O <sub>3</sub> uptake are considered?	Tree species / plant functional types (PFTs) considered
Individual	Physiology	ECOPHYS* (Martin et al., 2001)	Reduction in $V_{Cmax}$ and $J_{max}$ based on CUO.	<i>Populus tremuloides</i>
Stand	Physiology	ANAFORÉ-CASIROZ (Deckmyn et al., 2007)	Reduction in $V_{Cmax}$ based on [O <sub>3</sub> ]. Carbon necessary for defense (and the additional respiration it induced) against ozone and for repair of damage to $V_{Cmax}$ .	<i>Fagus sylvatica</i> L.
Stand	Physiology	PnET-II* (Ollinger et al., 1997)	Reduction in $A_n$ and $g$ based on AOT40 (DRR from Reich, 1987 and Tjoelker et al., 1995)	Mature hardwood
Stand	Demography	UVAFME* (Wang et al., 2016)	Reduction in growth by 0%, 10%, and 20% for resistant, intermediate and sensitive species, respectively, irrespective of [O <sub>3</sub> ]	32 species
Landscape	Demography	LANDIS-II* (Gustafson et al., 2013)	Reduction in $maxANPP$ under elevated O <sub>3</sub> (in comparison to control; measured in the field [Aspen-FACE experiment])	<i>Populus tremuloides</i> ; <i>Betula papyrifera</i> ; <i>Acer saccharum</i>
Global	Physiology	CLM (Lombardozzi et al., 2015)	Reduction in $A_n$ and $g$ based on CUO. DRR from Lombardozzi et al. (2013)	4 PFTs including broadleaves and conifers
Global	Physiology	ORCHIDEE-CHIMERE (Anav et al., 2011)	Reduction in annual $GPP$ based on AOT40 (DRR from Reich, 1987 and Ollinger et al., 1997)	4 PFTs including broadleaves and conifers
Global	Physiology	OCN (Franz et al., 2017)	Reduction in $A_n$ based on CUO (DRR from Wittig et al., 2007). Ability of plants to detoxify a proportion of O <sub>3</sub>	12 PFTs including evergreen broadleaves, deciduous broadleaves, conifers, temperate open woodland with C3 grass
Global	Physiology	JULES (Sitch et al., 2007)	Reduction in $A_n$ and $g$ based on CUO (DRR from Karlsson et al., 2004)	4 PFTs including broadleaves and conifers

$A_n$ : net photosynthesis; [O<sub>3</sub>]: Ozone concentration;  $V_{Cmax}$ : Maximal carboxylation rate;  $g$ : stomatal conductance;  $J_{max}$ : light-saturated rate of electron transport;  $GPP$ : Gross Primary Productivity;  $maxANPP$ : highest above-ground Net Primary Productivity that a species can biologically attain;  $DRR$ : Dose-Response Relationship;  $CUO$ : Cumulative Uptake of Ozone.

\* PBMs from North-America as only few stand- or landscape-scale models are able to simulate ozone effects in Europe (Reyer, 2015).

On the one hand, physiological PBMs explicitly and mechanistically simulate ozone effects on stomatal conductance, photosynthesis and carbon allocation within the tree (e.g., Ollinger et al., 1997; Deckmyn et al., 2007; Franz et al., 2017; Lombardozzi et al., 2015). At European scale, they generally predict a negative impact of ozone on annual average GPP, ranging from -4.7% (period 2001-2010; Franz et al., 2017) to -20% (period 2002-2009; Lombardozzi et al., 2015) or -22% (year 2002; Anav et al., 2011), depending on how ozone impacts and plant sensitivity are considered in the model (Sitch et al., 2007; see Table 2). In terms of biomass yield, Deckmyn et al. (2007) simulated a significant decrease in growth at the Kranzberg beech site at long time scales, especially at younger or less dense stands (reduction from 5% to 15% after 5 years). However, there are still a lot of uncertainties regarding the implementation of ozone response functions for most of the physiological processes due to the complexity of the mechanisms that are involved in ozone impacts (Jolivet et al., 2016), and due to the general lack of high-resolution data at both temporal and spatial scales on this topic, which reduces the possibility of model calibration for multiple species, especially for adult trees. Indeed, most of the PBMs use dose response relationships (DRRs) that derived from seedlings growing under controlled experiments (see Table 2). Moreover, the PBMs previously mentioned comprise limitations that are inherent to most current PBMs. A key drawback is related to the simulation of plant growth control, which is mainly assumed to be source-limited (i.e., photosynthesis; see Fatichi et al., 2014) and this may probably cause an overestimation of ozone effects on forest productivity (as they most likely also do for CO<sub>2</sub> fertilization; Reyer, 2015).

On the other hand, climate-sensitive forest succession models are more appropriate to simulate ozone impacts on long-term forest productivity, stand structure and composition as they explicitly model tree regeneration, growth, and mortality for multiple species and subsequent changes in competition (Bugmann, 2001). Indeed, linking ecophysiological



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responses to demographic processes is key to avoid erroneous inference at the ecosystem level as negative effects of stressors can be offset by an increase in tree longevity (Bugmann and Bigler, 2011), decrease in competition intensity, and changes in species competition towards more tolerant species (e.g., Wang et al., 2016). For instance, drought tolerant *Quercus spp.* and *Pinus spp.* that are predicted to gradually replace *Fagus sylvatica* because of climate change (Hickler et al., 2012) are also less sensitive to ozone (Büker et al., 2015). However, similarly as for physiological PBMs, long-term forest demographic data at sites with ozone concentration monitoring are scarce, which limits model validation and calibration. Also, upscaling the leaf-level physiological ozone response to annual changes in tree growth, reproduction capacity, or mortality probability is quite challenging (e.g., Samuelson and Kelly, 2001) as it requires size-, site- and species-specific ozone DRRs. As most PBMs use arbitrary DRRs, or the ones that derived from chamber experiments, ozone effects on forest ecosystems may be overestimated a second time (e.g., Wang et al., 2016; Franz et al., 2017; Table 2).

### **Towards a better assessment of short- and long-term ozone effects on forest growth**

Despite the difference in explaining ozone effects on tree growth can be comparatively small between exposure-based and flux-based approach (e.g., Büker et al., 2015), we acknowledge the large efforts undertaken to further develop ozone deposition and uptake models such as DO<sub>3</sub>SE. Improvements are still required (see above): our understanding and ability to predict the impact of ozone on forest productivity is still unsatisfactory, and we believe that the following scientific objectives may advance our understanding of ozone effects on forest ecosystems:

- Derive robust species-specific DRRs (i) for different variables such as biomass increment or net photosynthesis (ii) for trees with different sizes with a particular focus on adult trees,

and (iii) for an extended number of tree species, not only for the ones currently dominating the forest ecosystem. In this context, studies that focused on European tree species which should be favored by climate change like Mediterranean *Pinus* and *Quercus spp.* are highly valuable (e.g., Ribas et al., 2005; Vitale et al., 2005; Paoletti, 2006; Gerosa et al., 2015).

- Disentangle the respective negative effects of ozone and other stressors such as drought or pathogens to the positive effects of nitrogen deposition and CO<sub>2</sub> fertilization, and quantify their interactive effects. As individual tree responses to these environmental drivers change among stands with different structure and composition, differences in land use and management histories need to be accounted for.
- Better understand the plant physiology, especially how the alteration in the sink-source growth control by ozone (Andersen, 2003) change with tree size and under different stressors limiting carbon uptake for a long period (e.g., under shade or drought). Indeed, differences between seedling and mature-tree allocation patterns can cause differences in ozone response at the plant level (Ollinger et al., 1997), and short-term effects of changes in carbon supply on tree growth can differ from long-term ones (Norby et al., 2016).
- Quantify the contribution of ozone to forest dynamics using an integrated approach. For that (i) studies should also focus on other demographic processes such as tree reproduction (e.g., Leisner and Ainsworth, 2012) and mortality (e.g., Diaz-de-Quijano et al., 2016), which may be more linked to the tree carbon budget than growth (carbon starvation hypothesis; see Hartmann, 2015). Moreover, (ii) the different feedback across scales, from the physiological processes (e.g., change in ozone uptake due to drought-induced stomatal closure) to the demographic ones (e.g., changes in species mixture and composition towards more tolerant species) need to be considered.

To reach these objectives, the combined use of (i) experimental, (ii) long-term and large-scale monitoring, and (iii) process-based modeling approaches will allow us to profit from their respective advantages and lessen their restrictions (Fig. 1). This comprehensive, overarching concept links different investigation levels - from the physiological processes to the ecosystem dynamics - into a consistent framework (Ferretti et al., 2007). We especially identified the following research needs and frontiers:

- Collection of long-term and multidimensional data with different spatial and temporal resolution at numerous sites covering broad gradients in environmental conditions, species, and forest types. As recommended by Matyssek et al. (2012), integration and conception of new highly instrumented ‘supersites’ (i.e., new ozone-FACE experiments), monitoring networks of sites intensively measured (e.g., ICP Forests; De Vries et al., 2014a) and large-scale surveys would be highly needed, especially if maintained for a long period of time and if the data is analyzed with adequate statistical tools (see above). Integration in this concept of additional chamber experiments would also be valuable if they comprise supplementary manipulations of precipitations, temperature, CO<sub>2</sub>, and nutrients concentrations.
- Better link between empirical and modeling studies (Fig. 1) that can benefit from each other. On the one hand, the collection of new data and the better use of existing databases (e.g., data from the TRY and TOAR databases; from the ICOS infrastructure, AnaEE- and LTER networks, or from the Global Forest Biodiversity Initiative) are needed to (re)calibrate and validate PBMs (e.g., Medlyn et al., 2015). On the other hand, outputs from correlative models can be corroborated or contradicted by predictions from PBMs. For instance, comparing stand growth simulated by PBMs and estimated in the field based on dendrometric measurements can highlight ozone-induced species-specific changes in carbon allocation to above- and below-ground organs (e.g., Mäkelä et al., 2016). Finally,

sensitivity analyses from PBMs can be useful to prioritize research objectives by detecting the parameters and processes with the highest importance or the highest uncertainty, i.e., for which we need better information.

- Further development of PBMs via implementation of new equations or modules simulating ozone effects on different physiological and demographic processes. It would be especially useful to develop European-scale forest succession models that could simultaneously predict effects of ozone, climate change, natural disturbances and management on long-term forest dynamics. In addition to the developments that are inherent to most PBMs to better simulate the direct- and lag-effects of chronic and acute stress-induced changes in carbon uptake (e.g., Anderegg et al., 2015), concerning the representation of the hydraulic pathway and its dysfunctions (Mackay et al., 2015), the simulation of the temporal dynamics of carbon allocation to above- and below-ground organs and of the growth control via a combined source-sink-driven approach (Fatichi et al., 2014), and the coupling of the energy, water, carbon, and nutrient cycles (see Hickler et al., 2015), ozone effects should be fully included into this modeling scheme (Andersen, 2003). Amongst the challenges for modeling are (i) the parameterization for a diversity of species and forest types, (ii) the improvement of the ‘ozone uptake module’ to better consider the dependencies with temperature and drought (see above), (iii) the allocation of carbohydrates to build detoxifying compounds and antioxidant enzymes (Castagna and Ranieri, 2009), or (iv) the decoupling of photosynthesis and stomatal conductance induced by ozone that might impact carbon assimilation and transpiration damage estimates (Franz et al., 2017).
- Run PBMs with a multifactorial simulation design dedicated to quantify the importance of ozone on changes in simulated forest productivity (via sensitivity analysis), and to assess model uncertainty and quantify the respective contribution of each uncertainty source (via

variance decomposition analysis). Among others, we highlight the need to consider for the intra-specific variability in parameter estimates (e.g., Reyer et al., 2016), and to run PBMs under multiple environmental scenarios as projections in future ozone concentration and climate will depend on the global- and regional-scale socio-economic development.

In any case, studies should consider as much as possible the complexity of the forest ecosystem in terms of structure and composition and in the responses of individual trees, and should be carried out with an objective of extrapolation in a climate change context, which is predicted to have a positive impact on forest productivity at high latitude/altitude and a negative one under Mediterranean conditions.

## **Conclusion**

Despite our better understanding of the mechanisms of ozone impacts on physiological processes, its long-term effects on tree growth and forest composition and structure remains controversial because of the large variability among sites, species, data sources and methodological approaches. Ozone impacts on European forest productivity are probably negative, but slightly, and were most likely overestimated by previous empirical and modeling studies mainly because too much emphasis has been placed on outputs from experimental chamber studies. Compared with climate, site history, management or nitrogen deposition, ambient ozone remains as an unknown factor explaining spatial and temporal changes in forest growth, and their respective and interactive effects need to be quantified. Combining bottom-up and top-down approaches, based on ozone-controlled experiments and long-term monitoring, field-based correlative and process-based models, would be adequate for an integrated and comprehensive coverage of the entire gradient from the direct phytotoxic impacts of ozone on the biochemical processes at cellular level to multi-annual effects on stand-scale growth and dynamics.

## Authors contribution

M.C. led the writing, with input from M.F., A.G., A.R., and M.S. regarding paper structure and content.

## Data accessibility

This paper does not use data.

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