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Effects of climate change on air pollution impacts
and response strategies for European ecosystems



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ÉCLAIRE

Effects of Climate Change on Air Pollution Impacts and Response Strategies for European Ecosystems

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1. Executive Summary

- Current Dose-Response Relationships (DRRs) are not suitable for use in assessing the influence of O₃ on C sequestration in living forest biomass across Europe. This report identifies this short-coming as the most important ecosystem service for which there is uncertainty in how O₃ influence is assessed and one that can most readily be addressed with new data and model developments collected and undertaken as part of the ECLAIRE project.
- The main limitations with the current DRRs for forests are that they only provide an assessment of biomass loss as a fraction of total biomass. As such, they cannot be used for ‘snap-shot’ style annual assessments where the total forest biomass is highly variable (as is the case for forests across Europe with a mix of trees of very different age classes).
- We describe the development of a means of re-analysing ‘total biomass’ DRRs to give ‘net annual increment (NAIs)’ DRRs. These allow an assessment of the influence of O₃ on the biomass of forest trees laid down annually (irrespective of tree age).
- These re-analysis methods have been applied to existing forest tree O₃ fumigation datasets and have produced NAI DRRs for Beech/birch, Norway spruce/Scots pine, temperate oak and Aleppo pine. All PODy values are calculated using the multiplicative DO₃SE model.
- The development of the An-gsto DO₃SE model (conducted as part of ECLAIRE) allows the estimate of PODy values that can also take into account varying levels of leaf N through parameterisation of a species-specific maximum carboxylation term (VCmax).
- A small study was undertaken to see what effect likely ‘extreme’ values of VCmax (as derived from the literature and assumed to be representative of different leaf N levels) might have on PODy values and hence the slopes of the DRRs for selected forest tree species (Norway spruce/Scots pine and Beech/birch). These datasets were selected as they represented the largest (and hence most robust) O₃ fumigation datasets; new Birch data were also available from ECLAIRE experimental work.
- For Beech/birch the multiplicative and An-gtso models gave very similar DRs for standard (medium leaf N) conditions. This gives confidence to the An-gsto DO₃SE model parameterisation for these species’ (in that it is comparable to the results derived from the parameterisation of the DO₃SE multiplicative models). This is also supported by the ECLAIRE experimental data for birch.
- For Norway spruce the multiplicative and An-gsto models gave rather different DRRs for standard (medium leaf N) conditions. This needs further investigation, the parameterisation of the Norway spruce model seems questionable since values derived from the literature seem to be inconsistent with parameterisation suggested by the UK JULES land-surface model (for broad vegetation categories).
- The An-gtso model was used to see what effect varying leaf N (through variations in Vcmax) would have on PODy values and hence DRRs. Three different Vcmax values were chosen – intended to represent low, medium and high leaf N conditions. For

Beech/Birch the influence of leaf N was fairly substantial, less so, but still significant for Norway spruce.

- A simple method by which the %NAI loss DRRs of Beech/Birch and Norway spruce could be modified for different levels of leaf N levels is suggested by relating leaf N to N deposition using data provided from ICP Forests. This could be used to provide a provisional indication of how N deposition may influence forest tree sensitivity to O₃ across Europe.
- Methods and datasets that can be used to apply the new %NAI DRRs to assess the influence of O₃ on living C biomass across Europe are briefly described.
- Collation and re-analysis of O₃ fumigation datasets for forest trees using the 'total biomass' response parameter has been submitted as a journal paper (Bueker et al, sub). A first application of the NAI C sequestration method is currently being written up in a paper that will shortly be submitted by Bueker et al. The description of the new An-gsto model is being written up as a paper and will be submitted in the coming months by Emberson et al.

Objectives:

Delivery of novel thresholds for key dose-response relationships for use in regional scale modelling and mapping relevant for ecosystem service assessment

1. Identification of major knowledge gaps in our ability to perform ecosystem service assessments.
2. Development of novel methods to fill these knowledge gaps – re-analysis of flux response relationships for forest trees.
3. Application of methods to provide novel dose-response relationships (DDRs) for application at the regional scale.

2. Activities:

- Assessment and critique (in terms of knowledge gaps) of existing dose-response relationships (DRRs) for use in regional scale modelling and mapping.
- Identification of datasets that can be used to develop and apply new dose-response relationships (DRRs) across Europe.
- Development and trialling of new methods to further the use and application of available datasets.
- Delivery of resulting novel dose-response relationships to ECLAIRE C5 modellers and mappers.

3. Results:

- Identification of new methods to develop new dose-response relationships (DRRs) to allow annual assessments of the effects on ozone on C sequestration in the living biomass of European forest trees
- Identification of a method to incorporate the influence of nitrogen availability on O₃ dose response relationships (DRRs) for C sequestration – involving a comparison of the multiplicative and photosynthetic based gsto modelling approaches.

4. Milestones achieved:

MS 54: Identification of key response variables for ozone and nitrogen pollution.

MS 55: Application of DO3SE_C to develop dose-response relationships

5. Deviations and reasons:

This deliverable was delayed to allow re-analysis of the DRs for derivation of novel thresholds for those responses that will be used by C5.

6. Publications:

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

Büker,P., Harmens, H., Mills, M., Cambridge, H., Karlsson, P.E., Simpson, D., Norris, D., Falk, R., Briolat, A., Cinderby, S., Langer, J., Engardt, M., Emberson L.D. Current day and future tropospheric ozone burdens substantially reduce the carbon sequestration potential of living tree biomass across Europe (in prep).

Emberson, L.D., Bueker, P., Briolat, A., Mills, G., Harmens, H., Hayes, F., Gerosa, G. Double trouble: Modelling the combined effects of ozone and nitrogen pollution on vegetation health. (in prep).

7. Meetings:

Presentation of results at ECLAIRE Annual General Meetings (Zagreb 2013; Budapest 2014).

Presentation of results at C5 ECLAIRE Meeting (Austria 2015).

8. List of Documents/Annexes:

See Annex: Delivery of novel thresholds for key dose-response relationships, below.

Delivery of novel thresholds for key dose-response relationships

1. Identification of major knowledge gaps in our ability to perform assessments of the O₃ effect on ecosystem services.

A recent paper by Mills et al. (2011) reviewed progress in establishing ozone critical levels according to the methodologies outlined by the UNECE LRTAP Convention in the 'Mapping Manual' (LRTAP Convention, 2010). For crops, critical levels have been derived for effects on wheat (grain yield, grain mass, and protein yield), potato (tuber yield) and tomato (fruit yield). For forest trees, critical levels were derived for effects on changes in annual increment in whole tree biomass for beech and birch, and Norway spruce. Finally, for (semi-) natural vegetation, the critical level for effects on productive and high conservation value perennial grasslands was based on effects on important component species of the genus *Trifolium* (clover species). These critical levels are all based on the estimate of Phytotoxic Ozone Dose above a threshold 'y' which nominally represents a detoxification threshold (POD_y). Values of POD_y have been calculated using the DO₃SE model according to methods described in the Mapping Manual' (LRTAP Convention, 2010). This allows the development of POD_y dose-response (DR) relationships from which critical levels are derived.

The paper states that these critical levels can be used to assess protection against the damaging effects of ozone on food security, important ecosystem services provided by forest trees (roundwood production, C sequestration, soil stability and flood prevention) and the vitality of pasture. This is of course true however, for forest trees there are problems in the application of the DRRs for forest trees since the response parameter is the annual % reduction in **total** living biomass; in contrast, readily available data describing annual changes in forest biomass are presented as Net Annual Increment (NAI) i.e. the additional biomass laid down each year.

This is seen as a serious gap in the current risk assessment methodologies, especially given the damaging role played by O₃ to forestry across Europe. This influence has long been acknowledged with a large body of observational and experimental evidence documenting the means by which O₃ affects European forests; including foliar injury, accelerated leaf senescence, reduced photosynthesis, altered carbon (C) allocation, and reduced growth and productivity (Skärby et al., 1998; Matyssek & Innes, 1999; Matyssek et al., 2010). Many of these impacts will have implications for C sequestration (Matyssek et al., 2010). The size of O₃ effects on forest trees can be inferred from recent meta-analyses conducted by Wittig et al. (2007, 2009), which suggest that current ambient O₃ concentrations averaging 40 ppb can reduce the total biomass of trees by 7% compared with trees grown under pre-industrial O₃ concentrations. Reductions are likely to increase up to 11% were future O₃ concentrations to reach 64 ppb (Wittig et al., 2007).

The potential impact of O₃ on C sequestration on European forests could be of substantial global significance. Over recent decades European forests have acted as C sinks largely driven by forest expansion due to the abandonment of agricultural land (Kaplan et al., 2012). European, along with other global non-tropical forests, act as C sinks playing an important role in helping to off-set C loss from tropical deforestation (Pan et al., 2011). Therefore, any factors that might contrive to reduce the sink capacity of these forests could have serious

implications for atmospheric CO₂ concentrations. The importance of European forests in this capacity was clearly demonstrated by (Janssens et al., 2003) who estimated that European forests are likely to account for ~10 % of all European anthropogenic C emissions. Pan et al. (2011) quantified the annual flux of C to European (excluding Russia) boreal and temperate forests to be between ~0.27 to 0.3 Petagrams of C per year (Pg C year⁻¹) over 1990 to 2007. European forests (excluding Russia) have seen an increase in the sink strength from ~0.17 to 0.2 PgC year⁻¹ between the 1990 to 1999 and 2000 to 2007 periods (Pan et al., 2011). This has been attributed to a number of different factors including the large proportion of relatively young fast-growing trees (Nabuurs et al., 1998), changes in forest management that influence the standing stock (Ciais et al., 2008; Nabuurs et al., 2001), as well as increases in atmospheric CO₂ concentration (e.g. Friedlingstein et al., 1995), temperature (e.g. Myneni et al., 1997), and nitrogen (N) deposition (e.g. Magnani et al., 2007).

Previous modelling studies that have assessed the influence of O₃ on C sequestration (e.g. Felzer et al., 2005; Felzer et al., 2009; Sitch et al., 2007; Ren et al., 2007) have used approaches that, to varying extents, have been based on risk assessment methods adopted by the United Nations Economic Commission for Europe (UNECE) Convention on Long-Range Transboundary Air Pollution (LRTAP) (LRTAP Convention, 2010). Felzer et al. (2004; 2005; 2007 and 2009) and Ren et al. (2007) applied a method that used concentration-based AOT40 (Accumulated hourly mean Ozone concentration above a Threshold of 40 ppb, during daylight hours) response relationships for conifers (based on Reich, 1987), hardwoods (Ollinger et al., 2012) and crops (based on Reich, 1987). These AOT40 values were weighted according to photosynthetic based estimates of stomatal conductance (g_{sto}) and the ratio of potential to actual evapotranspiration to allow plant physiology and soil moisture stress to modify O₃ exposure. As such, this approach attempts to incorporate the environmental conditions that are likely to limit O₃ damage by reducing O₃ uptake and thereby acknowledges the need to move towards flux-based risk assessments for O₃ that incorporate environmental factors known to modify species sensitivity to O₃ (Emberson et al., 2007a). However, this modification of AOT40 omits O₃ concentrations below the 40 ppb cut-off which are known as being capable of causing damage to plants. Unequivocal evidence for this comes from the fact that AOT30 was as statistically robust as AOT40 in terms of estimating plant damage (Kärenlampi & Skärby, 1996). The main reason that a 40 ppb, rather than 30 ppb, cut-off concentration was selected was due to the fact that 30 ppb was considered too close to background European O₃ concentration levels. As such, achieved air quality goals based on AOT30 would have required implementation of emission control strategies outside of Europe (Kärenlampi & Skärby, 1996). Therefore, the application of methods that modify AOT40 may substantially underestimate the role of O₃ in reducing C sinks.

In contrast, Sitch et al. (2007) modified modelled estimates of photosynthesis by a factor that accounted for O₃ uptake and detoxification based on LRTAP flux-based POD_Y (Phytotoxic Ozone Dose over a threshold Y) response-relationships (LRTAP Convention, 2004) allowing a consistent solution to be obtained for O₃ uptake, g_{sto} , and O₃ effects on photosynthesis. This approach is more consistent with the empirical derivation of flux-based response-relationships but still has some major disadvantages: a) it is not able to account for diurnal co-variation of O₃ concentrations with environmental factors that modify O₃ uptake since O₃ and meteorological data are only available at monthly mean temporal resolutions; b) it only estimates the extremes of known plant sensitivity to O₃ with no indication of how the distribution and O₃ sensitivity of individual species might influence C loss, and finally, c) it

was applied for five broad plant functional types at a rather coarse $0.5^\circ \times 0.5^\circ$ spatial resolution.

These studies, although rather limited in number, suggest that O_3 may have substantial impacts on C sequestration with estimates of losses of between 18 to 60 Pg C year⁻¹ over periods lasting from ~1950s to 2000 in global regions with high O_3 concentrations such as the south-eastern US, eastern Europe and eastern China. Therefore, improving our means of assessing the role O_3 plays in altering C fluxes of European forests is an important focus for future research. This has also been highlighted by modelling studies that have focussed on the importance of atmospheric pollution in determining C fluxes. For example, a study by de Vries & Posch (2011) suggested that past increases in growth by N deposition may have been largely off-set by past increases in O_3 concentrations. An improved understanding of the role of trace gases in C sequestration will be important for international negotiations on climate mitigation actions to limit atmospheric concentrations of greenhouse gases (UNFCCC, 1997).

Therefore, we have focussed efforts in this ECLAIRE Work Package 12 to develop new methods that are capable of applying existing knowledge of O_3 effects on living forest biomass using readily available data describing changes in forest biomass presented as Net Annual Increment (NAI). To achieve this we have re-analysed the existing forest datasets that have been used to establish forest DRRs (Bueker et al., submitted) to convert the response parameter from annual % reduction in *total* living biomass to % reduction in NAI. For European forests this metric is particularly relevant in terms of C sinks since the living forest biomass and harvestable wood products of European systems comprise between 55 to 75% of the total C stock change per year (Pan et al., 2011).

2. Development of novel methods to fill knowledge gaps i. re-analysis of experimental data to derive Net Annual Increment (NAI) DRs for forest trees.

Ozone flux based DRRs derived from field-experiments have been used to derive critical levels (CLs) which have identified levels below which O_3 damage would not be expected to occur (LRTAP Convention, 2010)(Mills et al., 2011). The UNECE LRTAP Convention has used such CLs as a policy tool to identify areas of CL exceedance across Europe and subsequently to formulate European emission control strategies to improve air quality.

The DRR relationships for forest trees that have been presented previously use the accumulated stomatal O_3 flux above a threshold 'y' as the dose metric (Emberson et al., 2007b), often referred to as PODy (Phytotoxic Ozone Dose above a threshold 'y'), and the relative change in annual whole tree biomass production as the response metric (Karlsson et al., 2007). Current CLs for forest trees are based on an analysis performed by (Karlsson et al., 2007) on Norway spruce (27 data points from 3 countries and 8 experiments) and beech/birch (38 data points from 3 countries and 14 different experiments) and were set to values for which there was a > 95% confidence of finding a significant effect at the percentage loss chosen (LRTAP Convention, 2010). Since the publication of (Karlsson et al., 2007) new methods to assess the influence of soil moisture on stomatal O_3 flux (Büker et al., 2012) and investigate a new response parameter (Net Annual Increment -NAI) have been developed. These new methods are used in the derivation of the DRRs provided in this report.

Description of experimental datasets

O₃ experimental fumigation or filtration datasets collected for eight different species: birch (*Betula pendula*), beech (*Fagus sylvatica*), temperate oak (*Quercus robur* or *petraea*), Populus species, Norway spruce (*Picea abies*), Scots pine (*Pinus sylvestris*), Holm oak (*Quercus ilex*) and Aleppo pine (*Pinus halepensis*). These datasets were from ten different sites in seven countries (Sweden, Finland, U.K., France, Switzerland, Italy and Spain), thereby representing all major European climatic zones defined in the UNECE Mapping Manual (LRTAP Convention, 2010). Five experiments from France, Spain and Sweden contained reduced water supply treatments created either by i. reduced irrigation for up to 10 weeks, ii. reduced precipitation (with the help of an OTC roof) for up to 20 weeks or iii. no irrigation but only precipitation for the entire growing season. Further details of these treatments by dataset are provided in Table 1.

In comparison to the previous analysis by (Karlsson et al., 2007), all but one dataset (which was excluded due to limited replication) have been used again in this analysis. Apart from an open-field system in Kuopio (Oksanen, 2003), all other sites were equipped with Open Top Chambers (OTC) for the exposure of young trees to O₃. Details about the individual experiments can be found in Table 1 and the papers referenced therein. The control treatment in these experiments was either charcoal-filtered (CF) or ambient (non-filtered (NF)) air, and the O₃ treatments were ambient air, charcoal filtered plus additional O₃ or ambient air plus additional O₃. The response parameter used for the derivation of the DRR relationship was wherever possible total biomass; where root biomass was not available, above-ground biomass was used.

, The growing season is estimated using the latitude model for forests that do not have a year-round growth habit (LRTAP Convention, 2010). However, the experiments included in this analysis were predominantly performed in OTCs and the fumigation or filtration periods were sometimes outside the latitude function-derived growth period. Hence, only experiments that captured 60% or more of this period were included here to ensure that O₃ exposures covered the majority of the estimated growing season.

The calculation of POD_y required each experimental site to provide a full complement of hourly meteorological data from which *T*, *D* and *PPFD* could be determined according to standard methods (Jones, 1992). Hourly canopy height O₃ concentration data as well as a seasonal profile of soil water status was also required.

Table 1. Description of experiments. Those shaded in grey are new datasets as compared to Karlsson et al. (2007). AA = ambient air; CF = Charcoal-filtered; NF = Non-filtered (ambient); CF/NF+/++ = Charcoal-filtered or Non-filtered plus addition of O₃. OTC = Open top chamber.

Species	Site	Exposure system	Control treatment	Ozone treatments	Experiment duration (yrs)	Red. H ₂ O treatment	Tree age at start of experiment (yrs)	Response parameter	Reference
<i>Betula pendula</i>	Birmensdorf (CH)	OTC	CF	CF+, CF++, CF+++	1	No	Cuttings	Total biomass	(Uddling, Pleijel, & Karlsson, 2004)
	Kuopio (FIN)	Open-field	AA	AA+	2, 2, 5	No	1	Perennial biomass	(Oksanen, 2003)
	Östad (S)	OTC	NF	NF+, NF++	1, 2	No	< 1	Perennial biomass	(P.E Karlsson, Uddling, Skärby, Wallin, & Selldén, 2003)
<i>Fagus sylvatica</i>	Schönenbuch (CH)	OTC	CF	NF	2	No	< 1	Total biomass	(Braun & Flückiger, 1995)
	Zugerberg (CH)	OTC	CF	NF	2, 3	No	< 1	Total biomass	(Braun & Flückiger, 1995)
<i>Picea abies</i>	Östad (S)	OTC	CF	NF, NF+	1, 2, 3, 4	Yes	3	Total biomass	(Ottosson, Wallin, Skärby, Medin, & Räntfors, 2003), (P.E. Karlsson et al., 2004)
	Schönenbuch (CH)	OTC	CF	NF	2	No	< 1	Total biomass	(Braun & Flückiger, 1995)
	Zugerberg (CH)	OTC	CF	NF	1, 2, 3	No	< 1	Total biomass	(Braun & Flückiger, 1995)
<i>Pinus halepensis</i>	Ebro Delta (SP)	OTC	CF	NF, NF+	3	Yes	2	Total biomass	(Alonso et al., 2003), (Elvira, Alonso, & Gimeno, 2007)
<i>Pinus sylvestris</i>	Headley (U.K.)	OTC	NF	NF+	2	No	1	Above-ground biomass	(Medlyn et al., 1999)
<i>Populus spec.*</i>	Curno (I)	OTC	CF	NF	1	Yes	1 - 2	Above-ground biomass	(Marzuoli, Gerosa, Desotgiu, Bussotti, & Ballarin-Denti, 2009), (Pollastrini et al., 2010)
	Grignon (F)	Open-field	NF	NF+, NF++	1	No	≤ 1	Above-ground biomass	Le Thiec, pers. comm., (Ahmad et al., 2012)
<i>Quercus ilex</i>	Ebro Delta (SP)	OTC	CF	NF, NF+	2	Yes	2	Above-ground biomass	(Alonso et al., 2014)
<i>Quercus robur or petraea</i>	Col-du-Donon (F)	OTC	CF	NF, NF+	2	Yes	2	Total biomass	(Dixon, Le Thiec, & Garrec, 1998)
	Headley (U.K.)	OTC	NF	NF+	2	No	1	Above-ground biomass	(Medlyn et al., 1999)

* either *Populus maximowicz* × *Populus berolinensis* (Curno) or *Populus deltoides* × *Populus nigra* (Grignon)

Estimating accumulated stomatal O₃ flux (POD_y).

The DO₃SE (Deposition of Ozone for Stomatal Exchange) dry deposition model has been described in detail elsewhere (Emberson et al., 2000; 2007; B ker et al., 2012). It has been specifically designed to estimate the total and stomatal deposition of O₃ to European vegetation. Here, the DO₃SE model was used to calculate the stomatal O₃ flux (F_{st}) component of this deposition, representative for a leaf of the upper canopy. The estimate of leaf/needle stomatal conductance (g_{sto}), which is a key component of this F_{st} calculation, was made using the DO₃SE models multiplicative algorithm (Jarvis et al., 1976; Emberson et al., 2000; (LRTAP Convention, 2010)). This model employs a multiplicative algorithm, based on that first developed by Jarvis (Jarvis, 1976), modified for O₃ flux estimates (Emberson et al., 2000a; 2000b; Emberson et al., 2001; Emberson et al., 2007b); B ker et al., 2012) to estimate leaf/needle g_{sto} as given in eq. [1]

$$g_{sto} = g_{max} f_{phen} f_{PPFD} \max\{f_{min}, f_T f_D f_{SW}\} \quad [1]$$

where the species-specific maximum g_{sto} (g_{max}) is modified by relative response functions (scaled from 0 to 1) to account for g_{sto} variation with leaf/needle age over the course of the growing season (f_{phen}) and the functions f_{PPFD}, f_T, f_D and f_{SW} relating g_{sto} to irradiance (described as photosynthetic photon flux density, PPFD, $\mu\text{mol}/\text{m}^2/\text{s}$), temperature (T, degrees Celcius), vapour pressure deficit of the air (D, kPa) and soil water, respectively. f_{SW} can either be related to soil water potential (SWP in MPa) or plant available soil water expressed in volumetric terms (PAW in % vol/vol). f_{min} is the minimum daylight g_{sto} under field conditions, expressed as a fraction of g_{max}. The parameterisations for this model for different forest tree species are given in Table 2.

The accumulated F_{st} above a threshold of ‘y’ nmol m⁻² s⁻¹ (POD_y) was calculated by summing modelled hourly F_{st} values (F_{sti}) over the species-specific growing season, estimated according to latitude as described in (LRTAP Convention, 2010) and eq. [2].

$$POD_y = \sum_{i=1}^n [F_{sti} - y] \text{ for } F_{sti} \geq y \text{ nmol m}^{-2} \text{ projected leaf area (PLA) s}^{-1} \quad [2]$$

POD_y values (POD_y) were derived for each receptor species of interest. The threshold ‘y’ value was set to 1 nmol O₃ m⁻² PLA s⁻¹ and is intended to represent the ability of plants to detoxify a certain amount of O₃ (Karlsson et al., 2007; LRTAP Convention, 2010; Mills et al., 2011).

Table 2. Parameterisation of multiplicative DO₃SE model for forest trees.

Parameter	Units	Beech	Birch	Temperate oak	Norway spruce	Norway spruce	Holm oak	Aleppo pine	Scots pine	Poplar
Region*****		CCE	NE	ACE	NE	CCE	ME	ME	ACE	***
g_{max}	mmol O ₃ m ⁻² PLA s ⁻¹	150	240	255	125	130	195	230	190	<i>575/540</i>
f_{min}	fraction	0.13	0.1	0.06	0.1	0.16	0.02	0.15	0.1	0.1
SGS*****	year day	LF or DP	LF	LF or DP	DP	DP	1 - 365	1 - 365	DP	DP
EGS*****	year day	LF or DP	LF	LF or DP	DP	DP	1 - 365	1 - 365	DP	DP
$f_{phen\ lim}^A$	year day	-	-	-	-	-	80	80	-	-
$f_{phen\ lim}^B$	year day	-	-	-	-	-	320	320	-	-
$f_{phen\ a}$	fraction	0.0	0.0	0.0	0.0	0.0	1.0	1.0	0.8	0.0
$f_{phen\ b}$	fraction	(1.0)	(1.0)	(1.0)	(1.0)	(1.0)	1.0	1.0	(1.0)	(1.0)
$f_{phen\ c}$	fraction	1.0	1.0	1.0	1.0	1.0	0.3	0.1	1.0	1.0
$f_{phen\ d}$	fraction	(1.0)	(1.0)	(1.0)	(1.0)	(1.0)	1.0	1.0	(1.0)	(1.0)
$f_{phen\ e}$	fraction	0.4	0.0	0.0	0.0	0.0	1.0	1.0	0.8	0.0
$f_{phen\ f}$	days	20	20	20	20	-	130	130	40	20
$f_{phen\ g}$	days	20	30	30	30	-	60	60	40	20
light_a		0.006	0.0042	0.003	0.006	0.01	0.012	0.013	0.006	0.006
T _{min}	°C	5	5	0**	0	0	1	10	0	11
T _{opt}	°C	16	20	20**	20	14	23	27	20	27
T _{max}	°C	33	200*	35**	200*	35	39	38	36	36
VPD _{max}	kPa	1.0	0.5	1.0**	0.8	0.5	2.2	1.0	0.6	2.1
VPD _{min}	kPa	3.1	2.7	3.25**	2.8	3.0	4.0	3.2	2.8	3.7
SWP/PAW	MPa / % vol vol ⁻¹	If SWP, distinction between boreal/temperate and Mediterranean forest tree species; details of SWP and PAW parameterisation in footnote****								
Leaf dimension	cm	7.0	5.0	5.0	0.8	0.8	5.5	0.8	0.8	10

* The T_{max} value is set at 200 °C to simulate the weak response to high temperatures of Norway spruce and birch trees growing under Northern European conditions (the stomatal response is instead mediated by high VPD values). Hence, the T_{max} value should be viewed as a forcing rather than descriptive parameter.

** “generic” deciduous parameterisation used as surrogate.

*** Parameterisation as given by Marzuoli et al. (Marzuoli et al., 2009) for young poplar trees, apart from g_{max} , which is based on site-specific measurements ($575\text{ mmol O}_3\text{ m}^{-2}\text{ PLA s}^{-1}$ = Curno, $540\text{ mmol O}_3\text{ m}^{-2}\text{ PLA s}^{-1}$ = Grignon; italics represent values taken from beech ACE parameterisation;

**** boreal/temperate $f_{SWP} = \min\{1, \max\{f_{min}, 0.355 (-LWP_{pd})^{-0.706}\}\}$; Mediterranean $f_{SWP} = \min\{1, \max\{f_{min}, 0.619 (-LWP_{pd})^{-1.024}\}\}$, where LWP_{pd} is the predawn leaf water potential (MPa); $f_{PAW} = \min\{1, \max\{f_{min}, f_{min} + (1 - f_{min}) ((100 PAW/FC) - PAW_{min}) / (PAW_{crit} - PAW_{min})\}\}$, where FC is the field capacity, PAW_{min} is the soil texture dependent soil water content at minimum soil water potential and PAW_{crit} is 50% of PAW. For details see (Büker et al., 2012)

*****LF = Latitude function according to LRTAP Convention (2010); DP = Data provided by data holders of experiments presented in Table 3. See also Table S1.

*****NE = Northern Europe; ACE = Atlantic Central Europe; CCE = Continental Central Europe; ME = Mediterranean Europe

Estimating the O₃ effect on total biomass.

The fumigation datasets have certain limitations when it comes to being used to estimate NAI. A key limitation is that the biomass of the plants is only determined at the end of the fumigation experiment. In many instances the fumigation experiment was started after the trees had already been growing for a number of years, therefore the ozone effect (difference between fumigated and control plants) is only provided as % total biomass reduction of the biomass accumulated over the entire life span of the tree (i.e not only the fumigation period). This is converted into an annual effect over the fumigation period using eq [3].

$$RB_{year} = RB_{total}^{1/year} \quad [3]$$

where RB_{year} is the relative biomass per growing season (a value between 0 and 1, relative to the control at zero O₃ exposure); RB_{total} is the relative biomass at the time of harvest; $year$ is the number of growing seasons from the start of the experiment until the time of harvest. Thus, both exposure and effects are expressed on the basis of a single growing season.

N.B. The '1/year' assumes that relative growth is equal across all years. The power function assumes exponential growth which is valid for a young tree but not older trees.

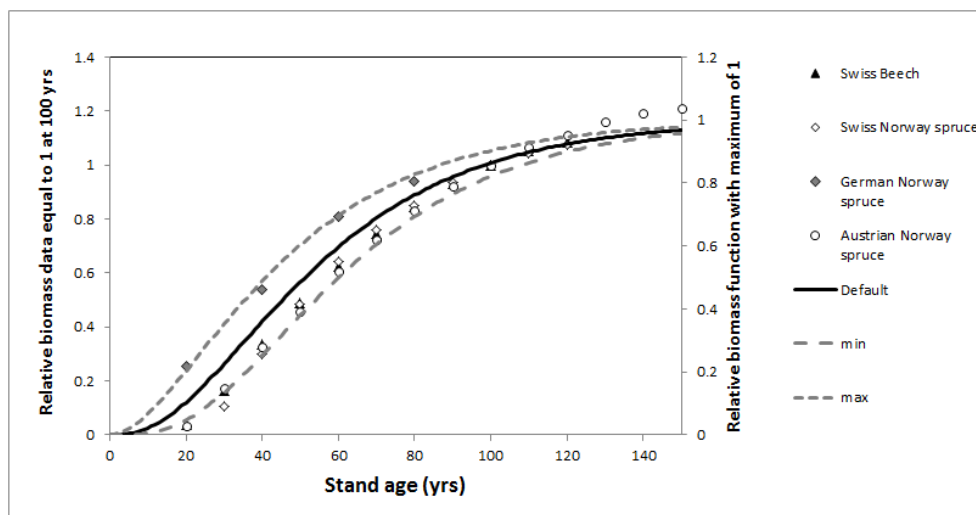
Estimating O₃ effect on Net Annual Increment (NAI).

To estimate the O₃ effect on NAI requires an estimate of the amount of biomass laid down each year. Forest tree growth is often defined by a standard sigmoidal growth function based on the Richards equation shown in eqn. [4] (Richards, 1959; Nishizono, 2010). This has been parameterised to provide a 'default' and 'upper' and 'lower' limit to growth (maximum and minimum respectively).

$$\text{Tree biomass} = y_0 \left(1 - \text{EXP}(-y_1 \cdot \text{tree age}) \right)^{\left(\frac{1}{1-y_2} \right)} \quad [4]$$

Where y_0 is 1, y_1 is 0.03 and y_2 is 0.65 for the 'standard' growth function; y_0 is 1, y_1 is 0.03 and y_2 is 0.5 for the max growth function and y_0 is 1, y_1 is 0.03 and y_2 is 0.74 for the min growth function. Figure 1 shows these curves along with standardised data describing growth of beech and Norway Spruce from different countries in Europe (Mund et al., 2002; Cabanettes et al., 1998; Teobaldelli, 2009....).

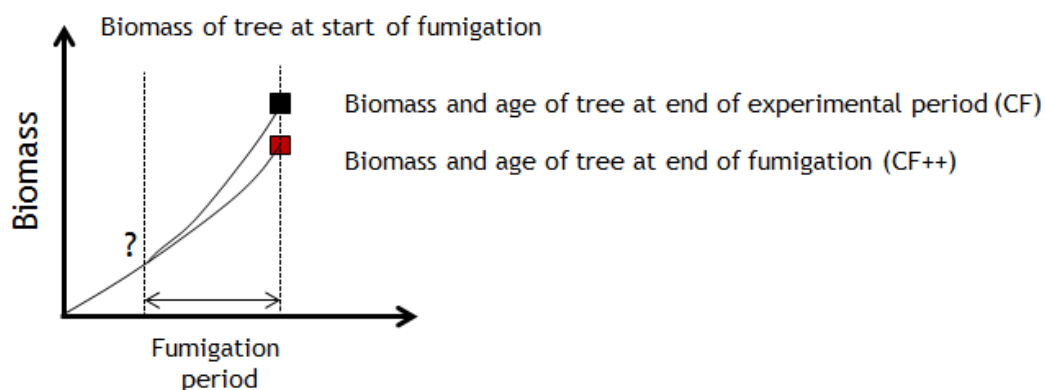
Figure 1. 'Default', 'upper' and 'lower' Richard's eqn. growth curves plotted with data (standardised for final biomass) for different European tree species.



These growth curves are used to estimate the effect of O₃ on NAI through re-analysis of the existing dose-response data (see method below). This provides a ‘standard’, ‘min’ and ‘max’ limit for O₃ effects on NAI. This assumes that the O₃ effect on NAI is a constant % irrespective of age of tree or NAI size.

This re-analysis used empirical O₃ dose-response data (here we use the same as in LRTAP Convention, (2010) and Karlsson et al., (2007) with the exception described above of all but one Norway Spruce data set omitted due to too few replicates) describing absolute biomass under control and treatment (e.g. NF, CF, NF+, CF+, etc....) conditions. To ensure all data are compared with an equivalent control value, a regression is made through all treatment data points to give the control biomass at zero O₃ (according to the metric used). This follows the Fuhrer (1994) method commonly applied in O₃ DRR analysis. This value then represents the biomass, under zero O₃ exposure of the O₃ metric used, at the end of the experiment. Details of the start and end of the fumigation period and the age of the trees are also provided in the datasets. The ‘standard’ growth curve is used to estimate how tree biomass changes over the entire age of the tree, scaled to the actual final total standing tree biomass in O₃ free air value (in kg) (the black square in Figure 2), this is performed in half-yearly steps as many of the experiments start or end mid-year.

Figure 2. A schematic providing an example of how the effect of O₃ on NAI is estimated from O₃ DRR fumigation data and assumed growth curves. The origin of the graph represents zero for both the tree age and the biomass.



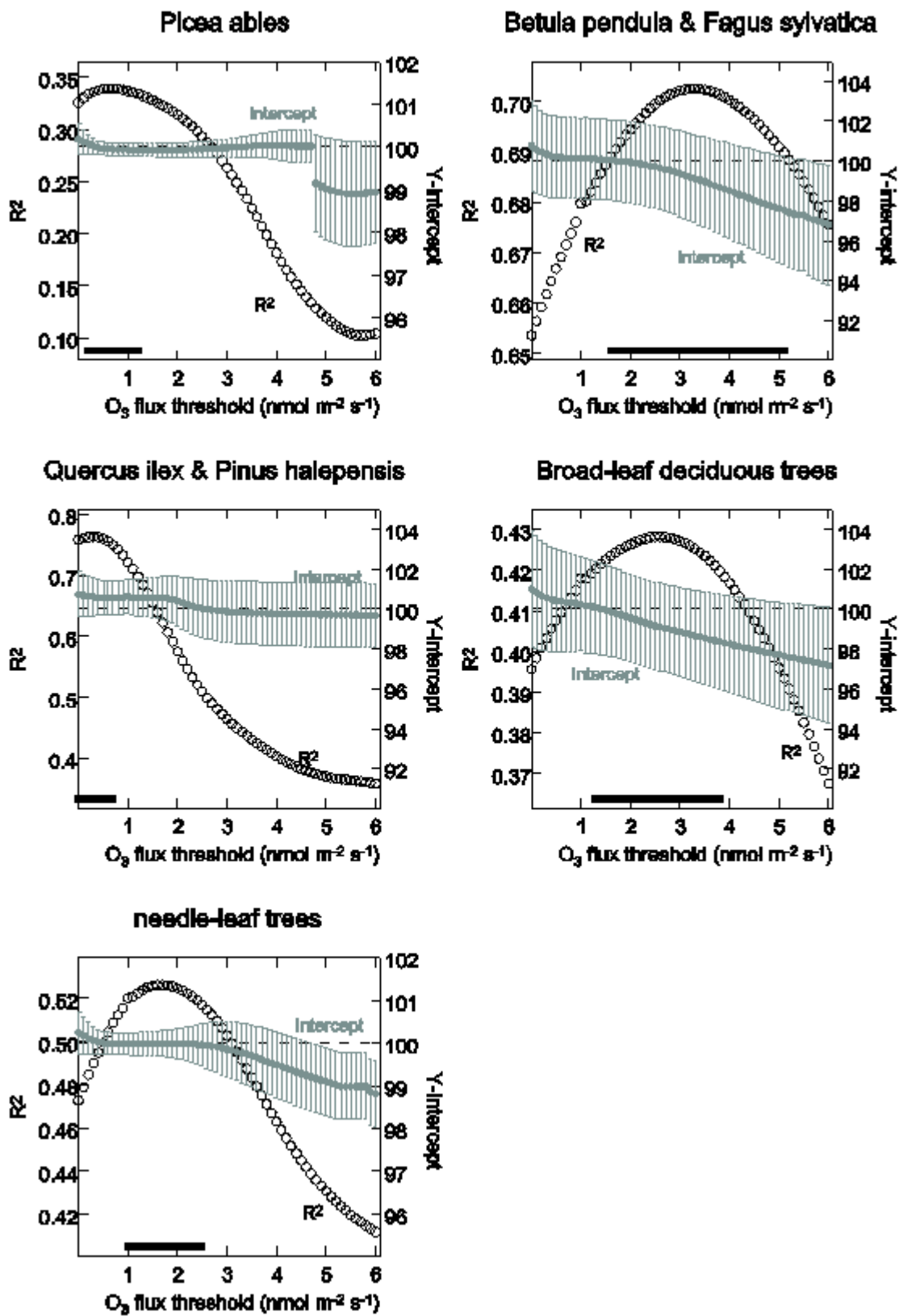
This then allows the incremental biomass (in kg) each half year under zero O₃ metric air to be estimated. The final total standing tree biomass under each different O₃ treatment (e.g. NF+, NF++, CF+, CF++ etc....) is available (red square in Figure 2) and allows the calculation of the total standing tree biomass ‘not laid down’ due to the O₃ treatment over the fumigation period. The biomass gained during the entire experiment under O₃ can then be estimated by subtracting the estimated biomass at the start of the fumigation period estimated using the Richards eqn. (marked as ‘?’ in Figure 2) from the final total standing tree biomass under O₃. The fractional biomass laid down each half-year is calculated as a fraction of total biomass laid down over the experimental period under zero O₃ metric conditions. The actual biomass ‘laid down’ each half-year under O₃ is then estimated by multiplying the total biomass ‘laid down’ over the entire fumigation period under O₃ by the respective half year fractions. The total standing biomass gained each half year under O₃ is then estimated by adding the actual biomass ‘laid down’ each half-year to the estimated total standing biomass at the start of the experiment (i.e. before any O₃ fumigation occurs).

The % change in annual increment due to the O₃ treatment is then estimated (and is assumed to be constant with tree age). This gives the % NAI reduction under O₃ for that treatment. The % NAI reductions for each experiment and treatment are pooled on a single figure and a regression drawn

through the data without forcing the regression through 100% on the Y-axis. Each data point included in the regression analysis represented a single value for each species, treatment and harvest of each experiment. This then gives the % NAI reduction DRR. To estimate the 'max' and 'min' % NAI reduction values the same process is repeated using the 'upper' or 'lower' defined growth curves in place of the 'standard' growth curve.

The selection of the optimal value to use as the y threshold ($\text{nmol m}^{-2} \text{s}^{-1}$) for each tree species or combination of tree species can be informed by plotting intercepts and R^2 values of the flux-response regression analysis against y . The two criteria for the threshold selection were a) an intercept close to 100 ($\pm 3\%$) and b) an R^2 value within 2% of the maximum R^2 value. The results of this exercise are shown in Figure 3 for the species and species groups. These indicate that a ' y ' threshold > 1 might be more appropriate for broadleaved species whereas a ' y ' of 1 seems appropriate for evergreen species. However, the UNECE Mapping Manual (LRTAP Convention, 2010) currently recommends a ' y ' value of $1 \text{ nmol O}_3 \text{ m}^{-2} \text{ PLA s}^{-1}$ and the ECLAIRE source-receptor matrices that will be used for emission control policy derivation in the ECLAIRE project have both used a ' y ' value of $1 \text{ nmol O}_3 \text{ m}^{-2} \text{ PLA s}^{-1}$ we continue to use that value in the analysis presented in this report.

Figure 3. Intercept and R^2 for various O_3 flux thresholds (y) of each species or species group. The black bars represent the y range where the confidence interval of the intercept includes 100 % and where the R^2 is within 2% of the maximum R^2 .



Comparison of total biomass and NAI DRs for O₃.

The resulting % reduction in NAI DRRs can be compared to those for % reduction in total living biomass as shown in the Figures 4 and 5 for Beech/Birch and Norway Spruce/Scots Pine respectively.

Figure 4. Comparison between relative 'NAI' and 'total biomass' for Beech/Birch using a POD₁ metric estimated using the multiplicative stomatal conductance (g_{sto}) DO₃SE model.

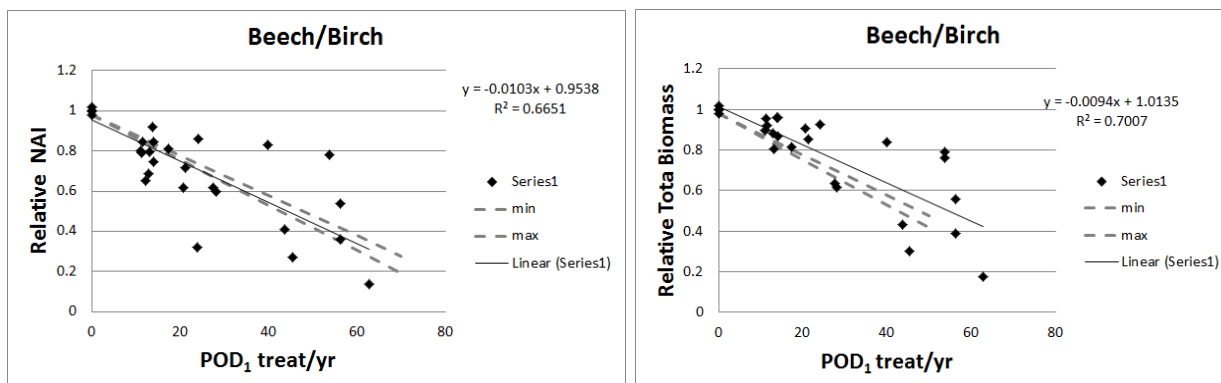
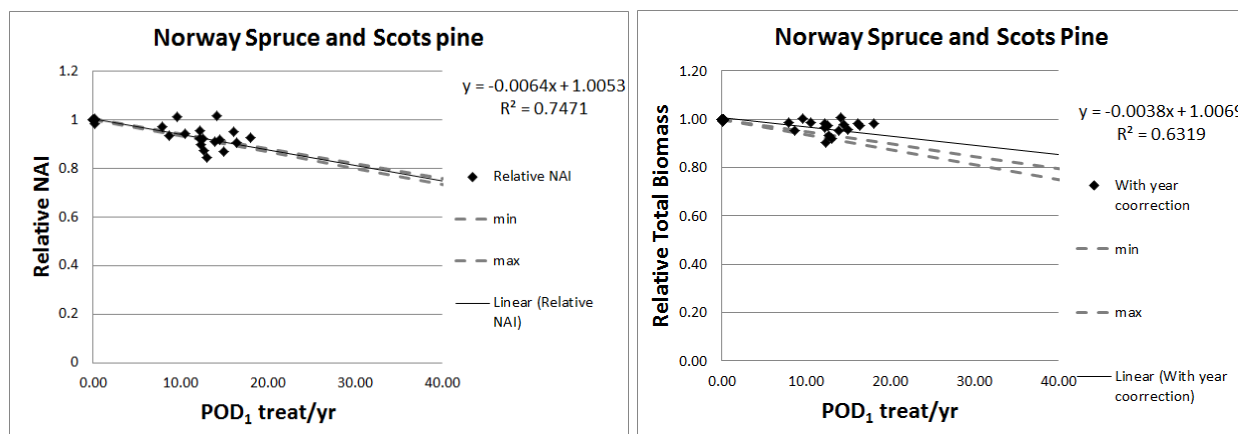


Figure 5. Comparison between relative 'NAI' and 'total biomass' for Norway Spruce/Scots Pine using a POD₁ metric estimated using the multiplicative stomatal conductance (g_{sto}) DO₃SE model.



A key issue is that the Norway Spruce and Scots Pine NAI dose-response relationship is more sensitive than the total biomass relationship. When the single Scots Pine dataset is excluded this sensitivity becomes even less extreme. This means that re-analysis for NAI response parameter suggests a more similar ranking of sensitivity of Norway Spruce and Beech/Birch than he previously been thought.

The Norway spruce and Beech/Birch data sets are made up of data that differ in terms of the age of trees at the end of the fumigation. For Norway spruce (31 data points), 23 data points come from trees ≥ 3 yrs old at the end of the experiment. For Beech/Birch (40 data points) only 8 data points are from trees ≥ 3 yrs old. This means that the Richard's growth curve (or any other year correction method (e.g. eqn. [4]) is required to simulate conditions for longer periods for Norway spruce than Beech/Birch.

This re-analysis was also performed for two other tree species – temperate Oak and Aleppo Pine. The results are shown in Figure 6. There were far fewer datasets for these tree species and therefore these dose-response relationships are considered less robust.

Figure 6. Relative ‘NAI’ dose-response relationships for Oak and Aleppo pine using a POD₁ metric estimated using the multiplicative stomatal conductance (g_{sto}) DO₃SE model.

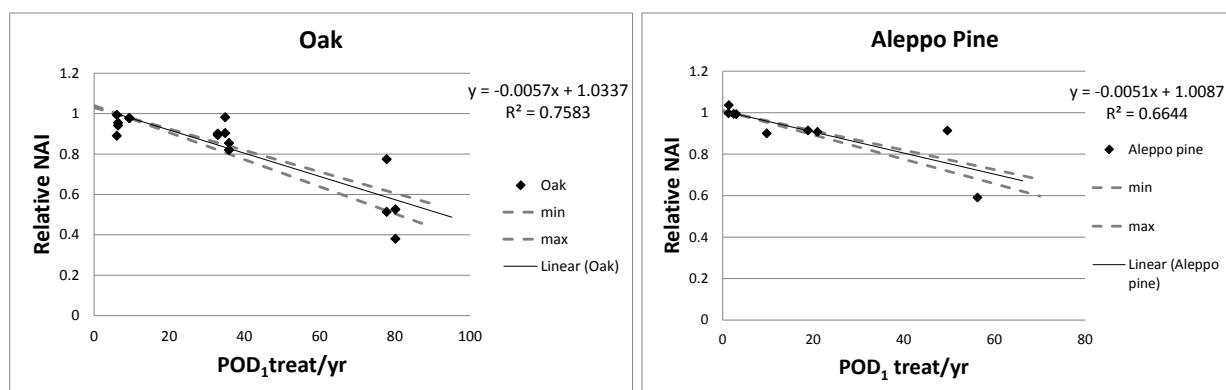


Table 3 summarises the details of the NAI DRs that have been developed using these re-analysis methods using a ‘y’ threshold of 1.

Table 3. Statistical details (regression equation and R²) of the Net Annual Increment (NAI) DRs developed on re-analysis of fumigation data for a variety of European forest tree species. Results for the ‘standard’, ‘min’ and ‘max’ growth curves are shown.

	POD	Standard	R2	min	R2	max	R2
Norway spruce & Scots pine	1	$y = -0.0064x + 1.0053$	0.56	$y = -0.0062x + 1.0066$	0.75	$y = -0.0067x + 1.0029$	0.73
Norway spruce	1	$y = -0.0054x + 1.0002$	0.58	$y = -0.0051x + 1.0003$	0.55	$y = -0.0062x + 1$	0.56
Birch & Beech	1	$y = -0.0103x + 0.9538$	0.67	$y = -0.0098x + 0.9475$	0.64	$y = -0.0109x + 0.9464$	0.65
Oak	1	$y = -0.0057x + 1.0337$	0.76	$y = -0.0053x + 1.0301$	0.76	$y = -0.0067x + 1.0406$	0.76
Aleppo Pine	1	$y = -0.0051x + 1.0087$	0.66	$y = -0.0047x + 1.0073$	0.66	$y = -0.0059x + 1.0112$	0.67

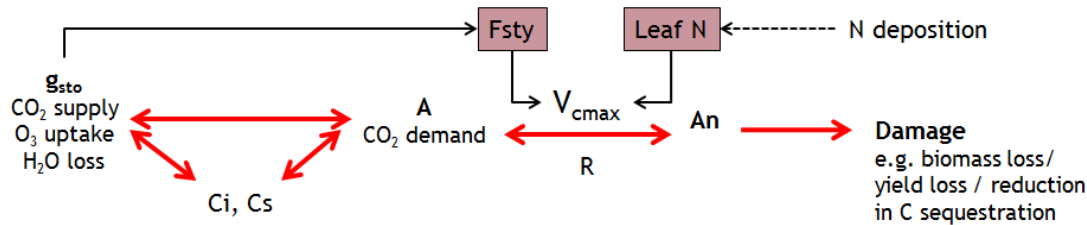
4. Incorporating the effect of Nitrogen in modifying biomass response to O₃.

It is well known that the levels of N in the ecosystem will affect sensitivity to O₃. This can be considered from two different viewpoints, either O₃ can reduce the N fertilization effect or the addition of N can reduce the damaging effect of O₃. Whichever point of view is considered, there is a need to understand how O₃ and N interact to affect plant responses such as biomass. This is not straightforward as N affects many different aspects of plant and ecosystem productivity and functioning. Similarly, many of these aspects will also be influenced by O₃ and climate. Therefore, the understanding of whole ecosystem responses will require more complex, ecosystem scale, process models to effectively simulate the most important interactions between O₃ and N. However, the development of the photosynthesis based stomatal conductance model (see WP4 and 12) provides a means of starting to understand the interaction of O₃ and N at the leaf level on plant physiology. Since photosynthesis is fundamental to plant productivity this provides an interesting way of developing an understanding of the potential interacting effects of leaf N and O₃ on C assimilation and hence biomass. This focus on C assimilation also incorporates the effects of climate (meteorology) on fundamental aspects of plant productivity since this determines the rate of biochemical processes and extent of gas exchange.

The *An-gsto* DO₃SE model has been described in previous ECLAIRE reports (i.e. D 4.3) and will not be repeated in detail here. However, it is useful to describe the core leaf level physiological interactions that are simulated by the *An-gsto* DO₃SE model as shown in Figure 7. Photosynthesis (A) and stomatal conductance (g_{sto}) govern the balance between CO₂ supply and demand (and

thereby determine internal and supply CO₂ concentrations (C_i & C_s). The rate of respiration (R) will determine the rate of net photosynthesis (A_n) and the ultimate rate of C fixation. A reduced C fixation can be assumed to have caused damage and may result in biomass losses, yield reductions and reductions in C sequestration.

Figure 7. Connections between the different leaf level physiological processes that are affected by ozone and N deposition.



From Figure 7 it is clear that the maximum carboxylation capacity (V_{cmax}) is central, influencing both pollutant uptake and hence O₃ deposition, as well as those processes that will determine damage. This is directly related to leaf N which will in turn be influenced by a variety of factors, one of which will be N deposition. Therefore, by altering V_{cmax} to simulate variations in leaf N it is possible to assess the effect of N deposition on O₃ uptake. This provides the opportunity to assess how leaf N might influence PODy and DRs (assuming there is no influence of leaf N on the toxicity of a given PODy). The key here is to alter leaf N and V_{cmax} by values that can simulate the influence of N deposition likely to be experienced across Europe within different tree species. In this analysis we use a top-down approach whereby we search the literature to identify the range of V_{cmax} values found within particular species (focussing on Beech & Birch and Norway spruce). We assume these values are indicative of the full range of leaf N values (though we acknowledge that V_{cmax} will also vary with phenology, tree age, tree height and will acclimate to environmental conditions (Niinemets, 2002). We then assume that these leaf N values can be related to N deposition using empirical data collected from ICP Forests (see section 5). An alternative approach would have been to estimate V_{cmax} from values of leaf N (as described in Wilson et al., 2000), however, given the uncertainty in estimating the variability in leaf N across Europe we decided against this approach.

The aim of the following exercise is to provide an indication of the extent that variable leaf N might be expected to affect PODy and subsequent DRs through alteration of stomatal O₃ uptake. In practical terms it *could* be used to identify European regions that might be more or less sensitive to prevailing O₃ profiles according to levels of N deposition and N availability. This could provide very crude assessments of the spatial location of combined risk from O₃ and N deposition. In theoretical terms it provides a modelling and mapping framework that can be used to explore these interactions further in the future through design of new experiments (both controlled chamber and epidemiological studies) as well as modelling experiments (possibly linking with the larger scale regional models that incorporate N dynamics).

The following describes the use of the new An-g_{sto} model to construct DRs (with NAI as the response parameter) which explore the effect of leaf N on PODy, the aim is to understand the likely size of the influence of variable leaf N.

The range of V_{cmax} values used here come from a literature review (results provided in Table 4) which also provides details of J_{max} and the parameter m that is necessary for An-g_{sto} modelling. There is a well recognised relationship between V_{cmax} and J_{max} (Medlyn et al., 2002) which we define and use here so that we focus on defining V_{cmax} only.

N.B. The ration of J_{max}/V_{Cmax} indicates the relative limitations of electron transport vs. carboxylase activity. A higher J_{max}/V_{Cmax} indicates that A_n is significantly co-limited by lower carboxylation rather than by lower RuBP regeneration associated with electron transport.

Table 4a. Parameterisation of the DO₃SE_C model – maximum carboxylation (V_{Cmax} , $\mu\text{mol}/\text{m}^2/\text{s}$) and maximum photosynthetic electron transport rate (J_{max} , $\mu\text{mol}/\text{m}^2/\text{s}$) (J_{max}) for forest species.

Species / cover type	V_{Cmax} , $\mu\text{mol}/\text{m}^2/\text{s}$ (mean, median, range)	Ref	J_{max} , $\mu\text{mol}/\text{m}^2/\text{s}$ (mean, median, range)	Ref
Deciduous forest	56	N.B. JULES value is 36.8	112	
Birch	51, 57, [42-71]	Hayes, 2014 Pers. Comm. [57], Rey & Jarvis, 1998 [42], Dreyer, et al., 2001 [71]	102, 92, [84-125]	Hayes, 2014 Pers. Comm. [84], Rey & Jarvis, 1998 [92], Dreyer, et al., 2001 [125]
Beech	52, 52, [35-62]	Bader et al., 2010 [44], Löw et al., 2007 [62], Parelle et al., 2006 [50], Dreyer, et al., 2001 [66], Balandier et al., 2007 [35], Fleck, 2001 [55]	107, 107, [83-128]	Bader et al., 2010 [120], Löw et al., 2007 [113], Parelle et al., 2006 [98], Dreyer, et al., 2001 [128], Balandier et al., 2007 [83], Fleck, 2001 [100]
Coniferous forest	65	N.B. JULES value is 26.4 (Clark et al., 2011)	190	
Norway spruce	44, 39, [24-, 81]	Zheng et al., 2002 [81], Niinemets, 2002 [60], cf Bergh et al., 2003 [24*], Marek et al., 2002 [40], Tarvainen et al., 2013 [69], Thum et al., 2008 [33], Urban et al., 2007 [37], Urban et al., 2003 [45], Sptova & Marek, 1999 [25]	[120, 110, [76-180]	Zheng et al., 2002 [180], cf Bergh et al., 2003 [76*], Marek et al., 2002 [144], Tarvainen et al., 2013 [100], Urban et al., 2007 [37], Urban et al., 2003 [120], Sptova & Marek, 1999 [100]
Scots pine	87, 83, [38-144]	Niinemets et al., 2001 [48], Niinemets, 2002 [83], Warren et al., 2003 [123], Jach & Ceulemans, 2000 [144], Wang, 1996 [38]	219, 237, [110-345]	Niinemets et al., 2001 [110], Niinemets, 2002 [237], Warren et al., 2003 [259], Jach & Ceulemans, 2000 [345], Wang, 1996 [146]

*Based on 6 references cited in Bergh et al., 2003 representing V_{Cmax} values for upper canopy needles from Scandinavian climates.

Table 4b cont.... Parameterisation of the DO₃SE_C model –minimum stomatal conductance (g_0 , $\text{mol H}_2\text{O m}^{-2} \text{s}^{-1}$), the ratio of $V_{Cmax}:J_{max}$ (where red denotes a ratio based on the mean values of J_{max} and V_{Cmax} found in the literature as in Table 3a) and species-specific composite sensitivity of g_s to A_n (m),

Species / cover type	g_0	Ref	$V_{Cmax}:J_{max}$	Ref	m	Ref
Deciduous forest	0.03	0.03	2			
Birch	0.03	Büker	2	Hayes (2014) Pers. Comm. [1.47],	8.6	Hayes

		et al., 2007	1.81, 1.76 [1.47-2.19]	Rey & Jarvis, 1998 [2.19], Dreyer, et al., 2001 [1.76]		(2014) Pers. Comm. [8.6]
Beech	0.03	Büker et al., 2007	2.05 2.11, 1.95, [1.82-2.37]	Bader et al., 2010 [2.73], Löw et al., 2007 [1.82], Parelle et al., 2006 [1.96], Dreyer, et al., 2001 [1.94], Balandier et al., 2007 [2.37], Fleck, 2001 [1.83]	8.6	Birch value used as surrogate
Beech/Birch						
Temperate oak	0.03	Use Beech value	1.98 2, 2.7, [1.73-3.0]	Marzuoli & Gerosa (2014) Pers. Comm. [1.82], Dreyer et al., 2001 [1.75] & [1.731], Bader et al., 2010 [3.00]	8.6	Birch value used as surrogate
Coniferous Forest			2.7		9.2	
Norway spruce	0.03	Use Beech value	2.7 2.84, 2.95, [2.22-2.38]	Zheng et al., 2002 [2.22], Niinemets, 2002 [2.38]	9.2	Nikolov & Zeller, 2003 [9.2]
Scots pine	0.03	Use Beech value	2.7 2.65, 2.40, [2.11-3.84]	Niinemets et al., 2001 [2.32], Niinemets, 2002 [2.86], Warren et al., 2003 [2.11], Jach & Ceulemans, 2000 [2.40], Wang, 1996 [3.84]	9.2	Nikolov & Zeller, 2003 [9.2]

This dataset was used to define the Vcmax range described in Table 5 that was used in the model runs.

Table 5. Values used in the An-gsto DO3SE model runs to assess the effect of Vcmax on PODy and subsequent DRs.

Species	Vcmax			Jmax			Jmax/Vcmax	m
	Low N	Medium N	High N	Low N	Medium N	High N		
Birch	35	55	75	70	110	150	2	8.6
Beech	35	55	75	70	110	150	2	8.6
Norway spruce	30	45	60	75	120	165	2.7	9.2
Scots pine	50	75	100	120	180	240	2.7	9.2

The Vcmax range described in Table 5 can be usefully compared with the range in Vcmax found in ECLAIRE experiments conducted at Bangor and Curno on young Birch, oak and Hornbeam trees. These experiments were performed in solardomes and involved fumigation of trees with different O₃ levels (from 35 to 70 ppb as a 24 hour mean) under a variety of different N addition treatments. The additions of N ranged from 10 to 70 kg N ha⁻¹ y⁻¹). The variation in Vcmax is described in Figure 8; this shows that the VCmax values varied by species with the highest VCmax values following the order of birch > oak > hornbeam. Figure 9 shows that values also differ by season and treatment; in almost all species and treatments VCmax declined over the season (the exception being Bangor birch which showed an increase in VCmax between July and September in all but the low N, high O₃). Relative to the low O₃, low N treatment treatments with higher N increase the VCmax value, this increase is tempered by high O₃; in contrast, treatments with low N and high O₃ consistently show the lowest VCmax values. Based on this, our values in Table 3 defined from the literature *look likely to represent a maximum range and in reality leaf N may have a less extreme effect on Vcmax.*

Figure 8. Variation in V_{cmax} by N and O_3 treatment over the course of the year for birch grown in Bangor and oak and hornbeam grown in Curno. Low and high N treatments are 10 and 70 kg/ha/yr respectively; low and high O_3 treatments are 35 to 70 ppb as a 24 hour mean respectively. The whiskers represent standard error.

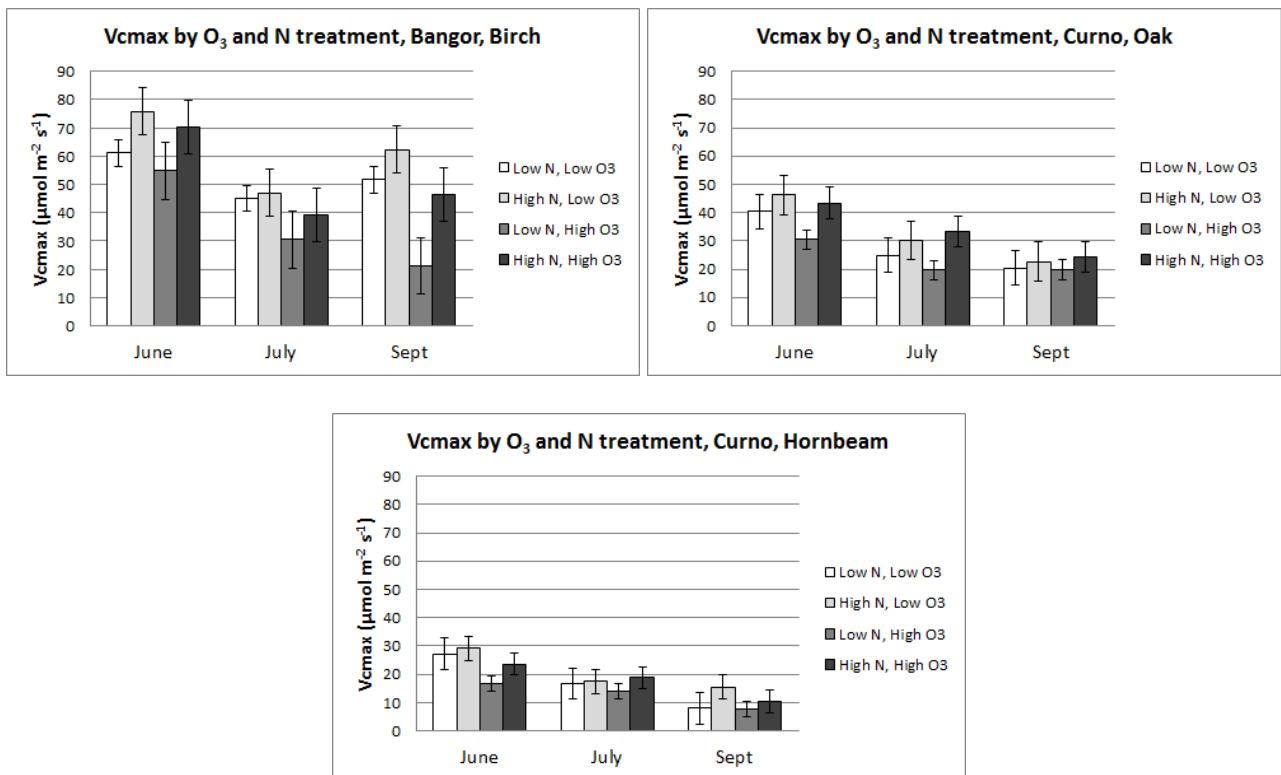
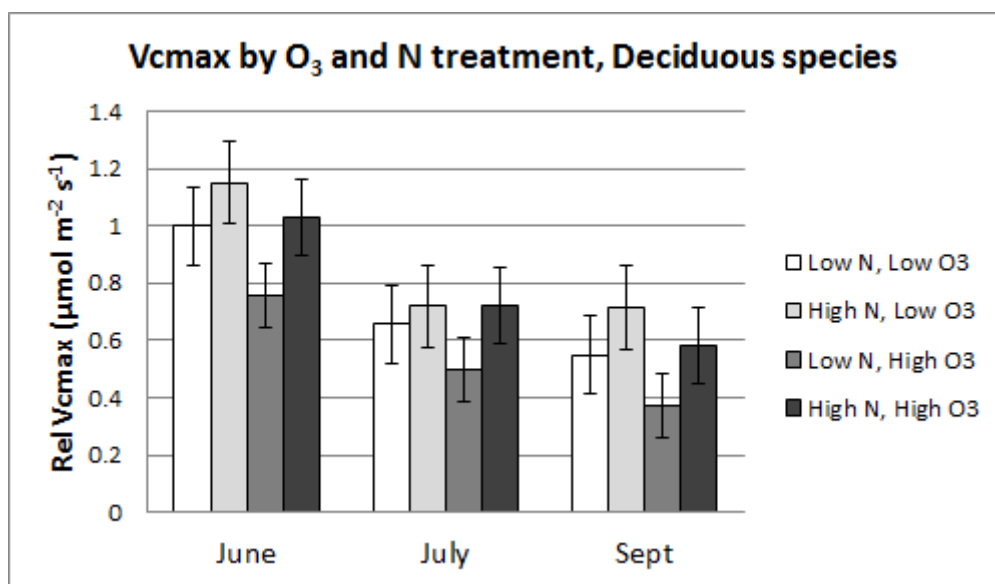
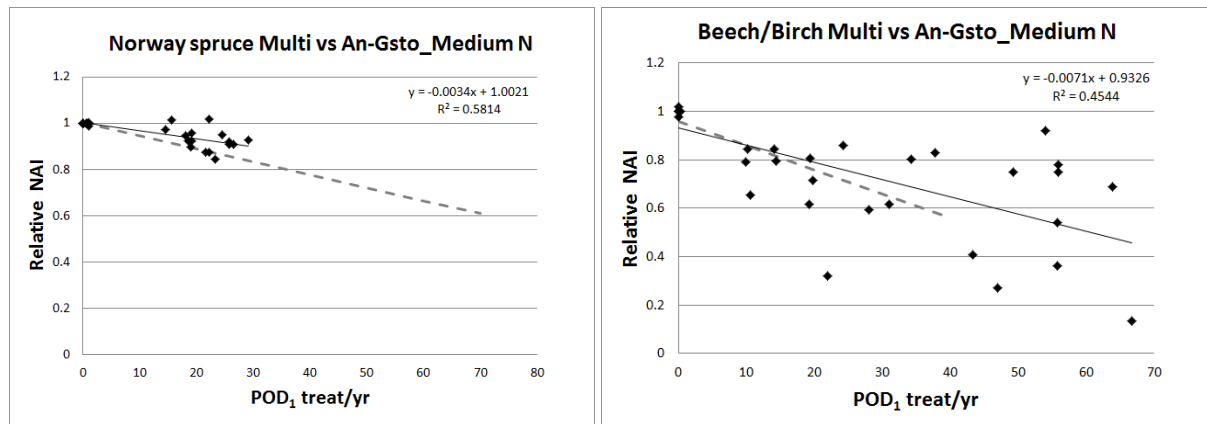


Figure 9. Relative variation in V_{cmax} by N and O_3 treatment over the course of the year for deciduous species (birch grown in Bangor and oak and hornbeam grown in Curno). Low and high N treatments are 10 and 70 kg/ha/yr respectively; low and high O_3 treatments are 35 to 70 ppb as a 24 hour mean respectively. The whiskers represent standard error.



There are a number of results that are useful to consider from these model runs. Firstly, it is interesting to see how the multiplicative and An-gsto DO3SE models compare in terms of PODy values and subsequent DRs. Figure 9 shows these comparisons for both Beech/birch and Norway Spruce.

Figure 9. Comparison between DRRs derived using the DO₃SE multiplicative model and the DO₃SE An-gsto model for Norway spruce and beech/birch. The dotted line in both plots represents the regression using the multiplicative model. The data and solid regression line represent the An-gsto model, assuming a medium level of leaf N (and hence V_{cmax} value for that species).



N.B. The multiplicative regressions in Fig 8 are slightly different to those in Fig 4 & 5 as experiments that were conducted under some form of drought stress have been excluded from the analysis since the An-gsto model is not currently set up to deal with soil deficits.

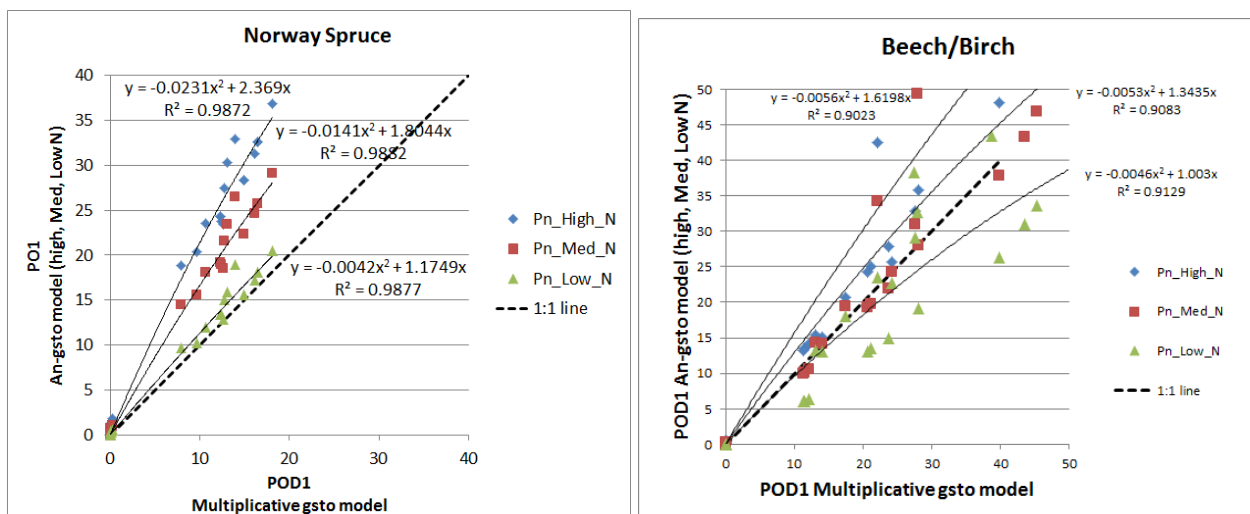
Figure 9 shows that for both Norway spruce and beech/birch there is some difference between the POD_y values that are produced by the two DO₃SE modelling approaches. For Beech/birch the An-gsto model predicts a slightly less sensitive DRR than had been produced using the multiplicative model. For Norway spruce the opposite is true. This is likely due to the parameterisation of the models, with the g_{max} of the multiplicative model not being representative of the V_{cmax} selected for the An-gsto model. This could be resolved through further review of literature data and testing modelled g_{sto} predictions against observed data.

In order to use the information provided by the An-gsto modelling framework within there are two different approaches that could be taken in order to use the information provided by this An-gsto modelling framework to inform European scale modelling and mapping of the combined influences of O₃ pollution and N deposition. The first involves using the model to inform how N deposition may influence POD_y values, this approach assumes that N will only affect the uptake of ozone and uses existing O₃ DRRs. The second approach involves using the model to derive new new DRRs for key forest tree species (e.g. Norway spruce and Beech/birch) that allow for the influence of N on biomass, this approach has the ability to assess how N affects both the uptake as well as plant biomass.

i. Influence of N on POD_y values.

This approach uses the An-gsto model to estimate POD₁ values for high, medium and low N (according to the V_{Cmax} values given in Table 5). These are plotted against the POD₁ values derived from the multiplicative g_{sto} model. From these plots it is possible to develop regressions that could be used to weight the multiplicative derived POD₁ values for high and low N conditions (i.e. under high and low N deposition). These could then be used to modify the POD₁ data provided by EMEP for the emission control optimisation runs to account for the influence of N deposition. The results for Norway spruce and Beech/birch are shown in Figure 10.

Figure 10. Comparison of POD_1 values derived using the multiplicative DO₃SE model against those derived using the An-gsto DO₃SE model for Norway spruce and Beech/Birch using VCmax values selected to represent low, medium and high leaf N availability.



It also possible to use a version of the An-gsto model that allows for the influence of O₃ on photosynthetic capacity through a damage function that reduces VCmax when stomatal O₃ flux exceeds a certain threshold (this model has been described in ECLAIRE deliverable 4.3). The results from applying this model are presented in Figure 11 for both Norway spruce and Beech/Birch. Both Figures 10 and 11 have polynomial trendlines applied to the data to indicate whether the relationship is linear or curve-linear.

Figure 11. Comparison of POD_1 values derived using the multiplicative DO₃SE model against those derived using the An-gsto DO₃SE model which includes a VCmax damage function for O₃ for Norway spruce and Beech/Birch using starting VCmax values selected to represent low, medium and high leaf N availability.

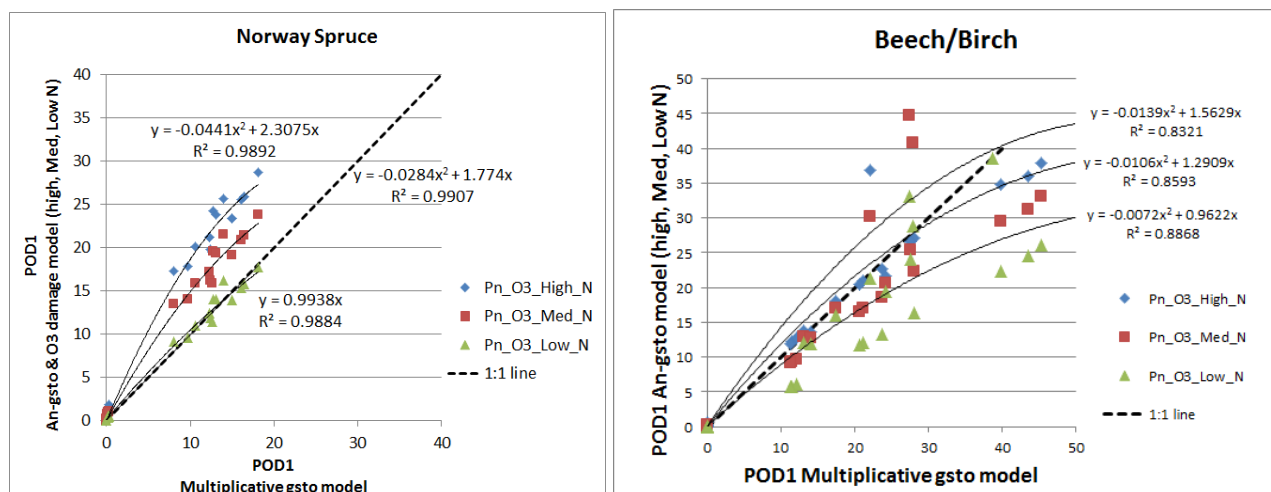


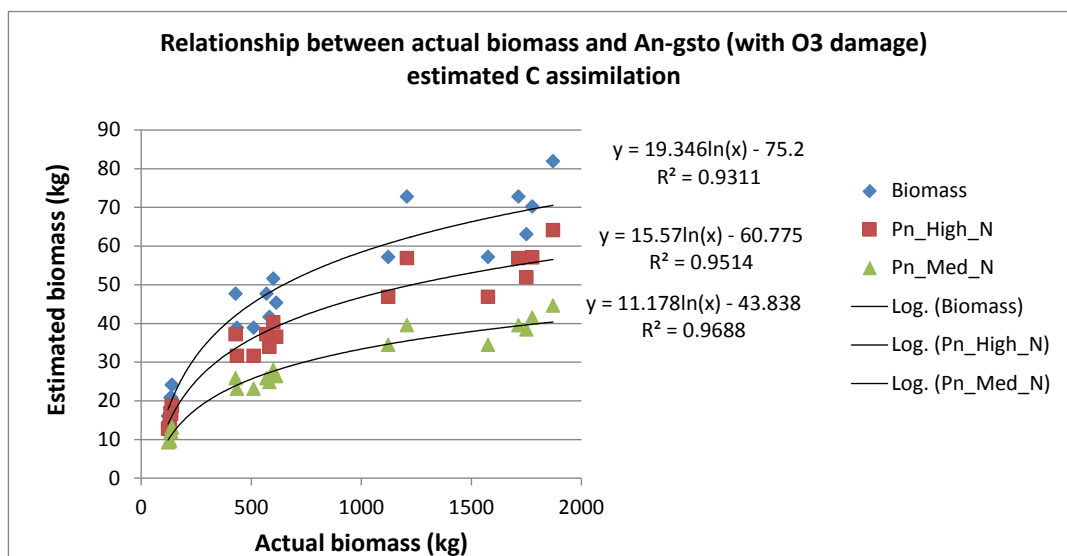
Figure 11 shows that using the An-gsto model which includes the O₃ damage function suggests that the rate of accumulation of POD_1 value will start decline with increasing POD_1 values. This is due to the accumulation of O₃ flux causing a decline in photosynthesis (as VCmax is damaged) which in turn reduces stomatal conductance and hence POD_1 accumulation over time.

ii. *Influence of N on DRRs.*

This approach uses the estimate of C assimilation obtained from application of the An-gsto and O₃ damage function model to modify the biomass data of the existing DRRs so that conditions of higher N will produce higher biomass but will also induce higher O₃ uptake. Ultimately this is likely to lead to a faster reduction V_cmax and hence photosynthesis and consequently an eventual decline in stomatal O₃ flux.

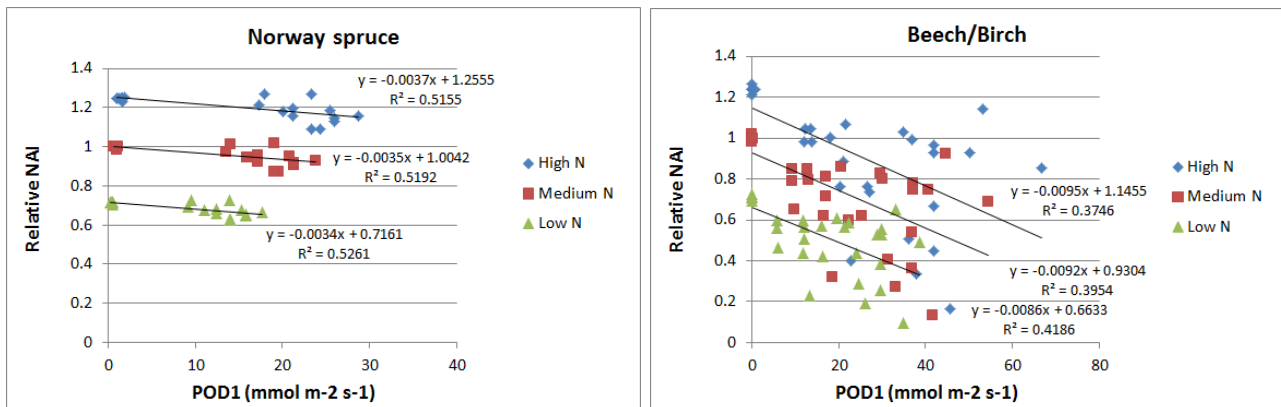
To develop these DRRs first requires an estimate of the relationship between the actual biomass (obtained from the DRR biomass data) and the simulated C assimilation produced by the An-gsto model; Figure 12 shows this relationship for Norway spruce. N.B. the An-gsto modelled biomass will be lower than the actual biomass as the model is only applied during the fumigation period (rather than over the entire lifetime of the tree. Since these data are only used to scale the actual biomass data to account for the influence of N this discrepancy should be largely irrelevant.

Figure 12 Relationship between Actual biomass (from the existing DRR relationships) and An-gsto (with O₃ effect on V_cmax) simulated C assimilation.



From the regressions developed in Figure 12 we can estimate the influence of N on biomass as resulting in a relative increase of 1.25 under high N conditions and decrease of 0.71 under low N conditions as compared to the medium N condition (which is assumed to be represented by the actual biomass data). These weightings can then be used to convert the actual biomass data, to data representative of high and low N. These data can then be plotted against the POD₁ data, simulated using the An-gsto model (including the method that accounts for O₃ damage on V_cmax); this method therefore integrates the effect of N on biomass and the influence of N and O₃ on uptake and therefore the rate of damage within the resulting DRRs are shown in Figure 13.

Figure 13. DRRs estimated using the An-gsto model to estimate relative biomass for high and low N treatments (in comparison to actual DRR data assumed equivalent to the medium N treatment) and POD1 values estimated by the An-gsto model that accounts for O₃ influence on Vcmax.



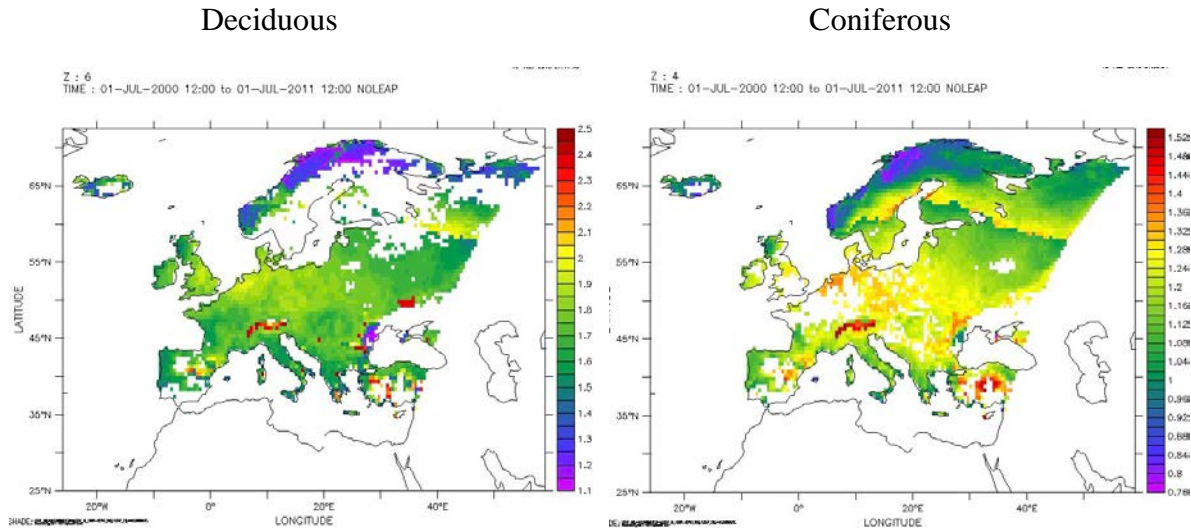
N.B. The Vcmax range in these model applications are likely to be rather extreme in terms of representing a range in leaf N. Further investigation of the role of leaf N on Vcmax for these species would be useful to parameterise the range more realistically for European conditions

5. How could this information be used in C5 modelling and mapping?

PODy estimates used in DRs need to be compatible with EMEP model outputs (e.g. PODy values) used in C5 (i.e. that have been used to develop source-receptor matrices). The PODy values for use in GAINS in C5 have been derived from EMEP runs that have used the multiplicative DO₃SE model. Therefore, any DRR relationships that might be provided to C5 will need to be based on the multiplicative DO₃SE model. This means that the newly developed An-gsto DO₃SE model (to understand O₃-N interactions) can only be brought into C5 working by using the An-gsto methods to *modify* DRs derived from the multiplicative DO₃SE model.

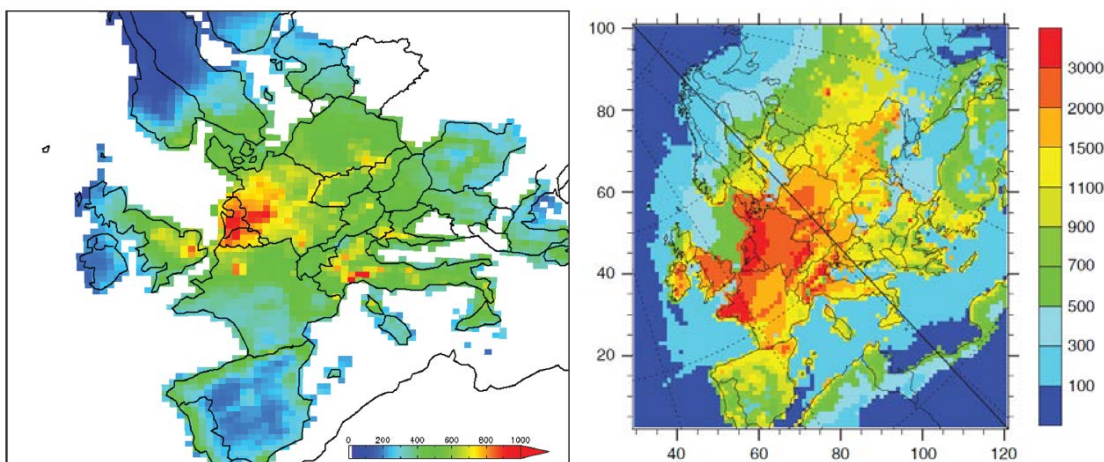
A key issue here in terms of application of this method is the ability to define leaf N (and hence VCmax) for forest trees across Europe and relate this to variable levels of N deposition and N saturation of ecosystems. One option is to use modelled data; Fig 11 below describes provisional plots made using the OCN model as part of the ECLAIRE project. These investigated the transient effects of climate, CO₂, N deposition and O₃ damage, the plots show provisional mass based foliar N concentrations as percentages for deciduous (Z:6) and coniferous (Z:4) trees. These maps suggest that N saturation is unlikely to have been reached though anything above 2% for deciduous and 1.28% for coniferous forests is getting close to saturation. The relationship between foliar N and leaf N is suggested with the area close to the Netherlands with the highest N deposition rates also showing the highest foliar N values; the Alps may also be showing high foliar N values due to a combination of high N input and low temperatures.

Figure 11. Provisional plots showing the transient effects of climate, CO₂, N deposition and O₃ damage on mass based foliar N concentration (as %) for deciduous (Z:6) and coniferous (Z:4) trees. These plots were made using the OCN model as part of the C4 component of ECLAIRE. N.B. the distribution of land cover types has been done using a simple overlay and therefore does not show the smooth transition between these land cover types that actually exists.



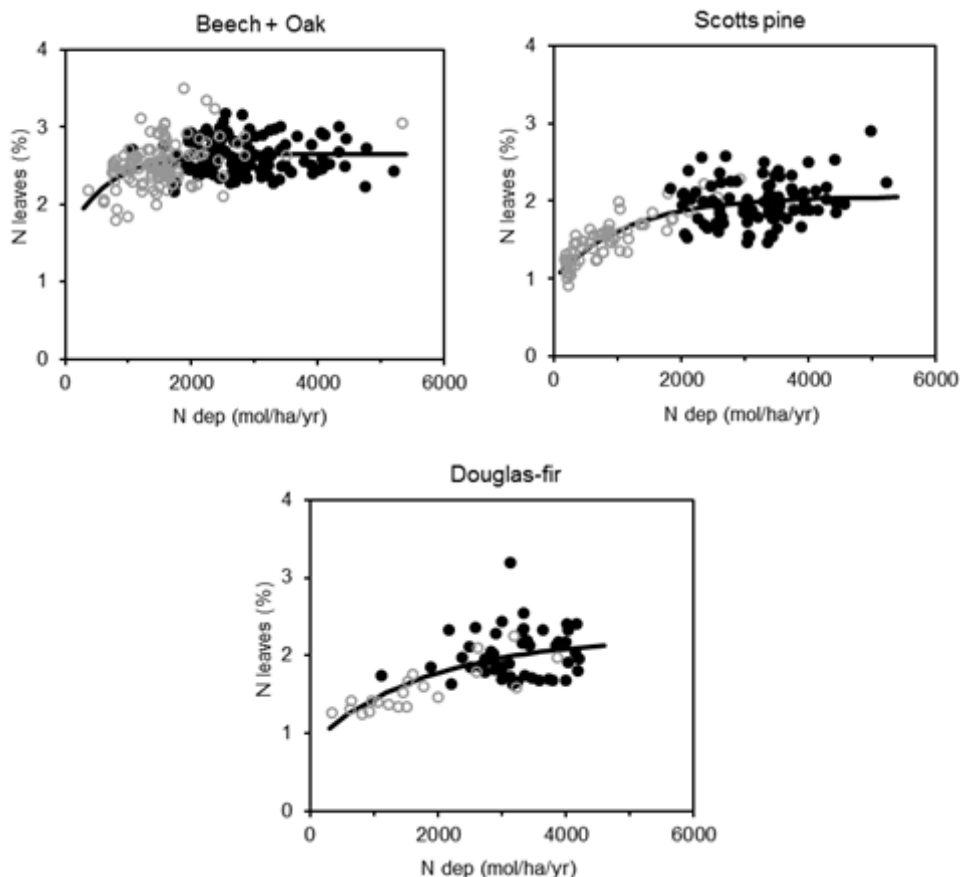
This type of modelling approach can be compared with a more empirical approach whereby N deposition estimates can be compared against observations of foliar leaf N. Fig 12 shows estimates of modelled N deposition for Europe and specifically for coniferous ecosystems from Simpson et al. (2011). These show broad consistency with those areas with higher leaf N concentrations (Fig 11) being those tending towards having higher N deposition (Fig 12).

Figure 12. Calculated nitrogen deposition from an ensemble of seven models for 2001. Units: mg(N)m⁻² (100mg(N)m⁻² = 1 kg(N)ha⁻²) – left figure. Calculated N-deposition densities (mg(N) per m² of ecosystem per year) to coniferous forest ecosystems (year 2000) – right figure. Source: EMEP MSC-W. (Simpson et al., 2011).



Data collected from several hundreds of ICP Forests plots across Europe has allowed the foliar N status to be related to N deposition allowing an estimate of N concentration in the foliage. Fig 13 shows that the % content of leaves tends to saturate at a lower value (~ 1, 500 mol/ha/yr) for beech and oak (deciduous species) compared to Scots pine (~ 2, 500 mol/ha/yr) (coniferous species).

Figure 13. Relationship between N concentrations of N in leaves in beech and oak (left), Scots Pine (middle) and Douglas fir (right) at ICP forest plots from DeVries et al (2000).



How could this information be used – either now or in the future? The Beech/birch DRR data seem relatively robust since the multiplicative and An-gsto (medium N level Vcmax parameterisation) models give very consistent DRRs indicating that the POD_1 values derived from both are similar. For Norway spruce the multiplicative DRR is consistent with the low N An-gsto DRR. One option for using these new DRRs would be to alter the slope of the DRR according to the N availability (or N deposition – see below) at the location at which the DRR is being applied. Using the ICP forest data (DeVries et al., 2000) we could identify low, medium and high N availabilities related to N deposition rates for both Deciduous and Coniferous trees as described in Table 6.

Table 6. Low, Medium and high N availability categories related to N deposition rates (expressed as mol/ha/yr).

	Coniferous (Norway spruce)			Deciduous (Beech/Birch)		
	Low N	Med N	High N	Low N	Med N	High N
N deposition (mol/ha/yr)	< 2, 000	2, 000 – 3, 000	> 3, 000	< 1, 500	1, 500 – 2, 500	> 2, 500
Vcmax ($\mu\text{mol}/\text{m}^2/\text{s}$)	30	45	60	35	55	75

These categories could be used to define European regions over which the O_3 DRR could be modified to allow for differential sensitivity to O_3 under different levels of N availability.

Additional runs could also be performed using the ESX model which now incorporates the An-gsto algorithm. Data collected from the ICP Forests database could be used to determine a spatially relevant variation in VCmax across Europe for forest trees. Results from these runs could be compared with results derived using the simple modification of the % NAI loss_{multi} values (see eq 5) to estimate the influence of low, medium and high leaf N on % NAI loss. This would allow a comparison of the risk estimates coming from the assuming 3 broad levels of leaf N across Europe compared to a more sophisticated assessment that would use actual leaf N data to drive the POD1 estimate. For Beech/birch POD1 estimates derived from both the multiplicative and An-gsto DO3SE models would use the same DRR given in eq [6] consistent with the data assessed in Fig 4 and Table 3.

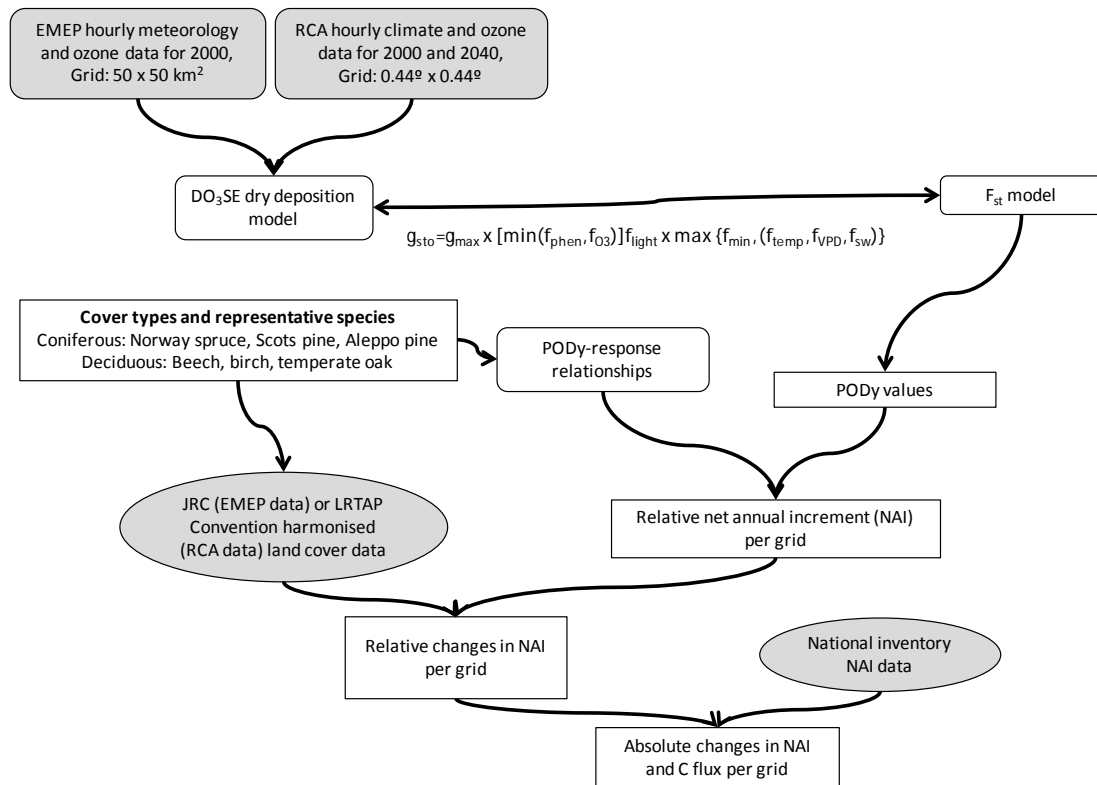
Further work that could be considered includes: i). re-analysis of the data for crops (using responses for both yield and quality where available) for wheat, potato and tomato; ii). investigation of the compatibility of the modelling of O₃-N interactions used in DO3SE with those algorithms used in the regional scale models used in the C4 component of ECLAIRE. Together, such information could inform the ECLAIRE C5 work based on the DO3SE multiplicative model in an 'ex poste' type analysis.

6. Application of methods to provide novel dose-response relationships for application at the regional scale

The above analysis has concentrated in the derivation of the DRs that could be applied across Europe to i. estimate the influence of O₃ on C sequestration of forest trees (using NAI response parameter) and ii. a suggestion of how the influence of N availability may be incorporated, using an extremely provisional methodology.

To apply these methods requires the collation of a number of different datasets and methods. Figure 14 describes the integration of the models and European datasets required to apply this flux-based NAI O₃ risk assessment approach (LRTAP Convention, 2010) to estimate the influence of O₃ on changes in annual C flux (Mt CO₂e yr⁻¹) of living forest biomass. The datasets are described in brief in the following text.

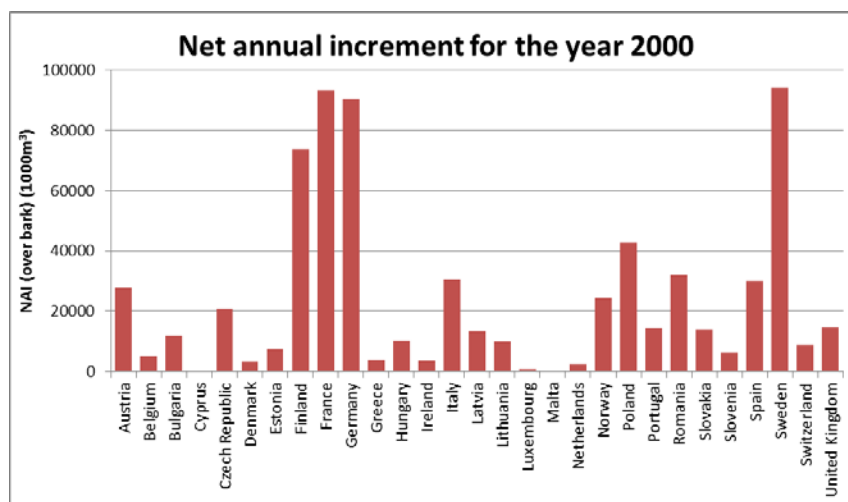
Figure 14. The datasets and methods that would be required to estimate the influence of O₃ on C sequestration, via net annual increment (NAI), on living forest biomass across Europe.



National inventory Net Annual Increment (NAI) data

Country-specific NAI data (m³ over bark) for the year 2000 (NAI_c) for European coniferous and broad-leaved forests were derived from the State of Europe's Forest report (Forest Europe, UNECE and FAO, 2011). Fig 15 gives an indication of the data derived from this dataset.

Figure 15. Country-specific NAI data (m³ over bark) for the year 2000 (NAI_c) for European coniferous and broad-leaved forests. Source: State of Europe's Forest report (Forest Europe, UNECE and FAO, 2011).



The % reduction in NAI of species and species groups (all forest trees should be assigned a representative NAI DRR from those described in Table 2) were then used to estimate the NAI that would have occurred in the absence of O₃ (i.e. pre-industrial Europe). To achieve this, values for NAI should be area-weighted according to the distribution of each forest tree receptor in the grids of the modelling domain. The species distributions can be defined by the land cover datasets described in Fig 11. This would result in a grid averaged relative NAI (NAI_{rel}). In cases where a grid square extends over more than one climate region, the NAI_{rel} can be estimated using the appropriate ecotype specific parameterisation of the majority climate region. It would be necessary to assume that the forest per grid was of uniform age, i.e. that O₃ had no differential effect due to tree/stand age.

National values for absolute current NAI were obtained from national inventory data provided by Forest Europe, UNECE and FAO (2011; see Fig 11) which describe the NAI (over bark) data collected for each EU country. To calculate the absolute values of NAI associated with each grid of the modelling domain (NAI_c), these data can be dis-aggregated to individual grids using the same forest distribution data as used to define the NAI_{rel} values. Pre-industrial NAI (NAI_{pi}) values that represent absolute NAI under O₃ concentrations that would not be expected to have any damaging effect on forest productivity were calculated through division of the NAI_c by NAI_{rel}.

For each country of the modelling domain, the annual change in the C sequestration to living tree biomass ΔC (tonnes C yr⁻¹) as based on changes in either NAI_c or NAI_{pi} can be calculated according to Penman et al. (2003) and von Arnold et al. (2005) as described in eq. [9]

$$\Delta C = NAI_{c \text{ or } pi} \cdot BEF \cdot D \cdot CF \quad [9]$$

Where *BEF* is the biomass expansion factor which converts between stem biomass and total living biomass including branches, leaves and roots; *D* is the density stem (tonnes dry weight m⁻³) and *CF* is the “C fraction” of dry matter (tonnes tonnes⁻¹).

The value of ΔC can be converted to CO₂-equivalents (CO₂e) by multiplying by 3.67.

The effect of O₃ on ΔC can then be quantified by calculating the difference between applying NAI_c or NAI_{pi} in eq [7]; this difference represents the reduction in annual C sequestration due to O₃ (Mt CO₂e yr⁻¹).

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