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TELLUS

Ozone risk for vegetation in the future climate of Europe based on stomatal ozone uptake calculations

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ABSTRACT

The negative impacts of surface ozone (O_3) on vegetation are determined by external exposure, leaf gas exchange and plant antioxidant defence capacity, all dependent on climate and CO_2 concentrations. In this study the influence of climate change on simulated stomatal O_3 uptake of a generic crop and a generic deciduous tree at ten European sites was investigated, using the LRTAP Mapping Manual stomatal flux model. O_3 concentrations are calculated by a chemistry transport model (MATCH) for three 30-yr time-windows (1961–1990, 2021–2050, 2071–2100), with constant precursor emissions and meteorology from a regional climate model (RCA3). Despite substantially increased modelled future O_3 concentrations in central and southern Europe, the flux-based risk for O_3 damage to vegetation is predicted to remain unchanged or decrease at most sites, mainly as a result of projected reductions in stomatal conductance under rising CO_2 concentrations. Drier conditions in southern Europe are also important for this result. At northern latitudes, the current parameterisation of the stomatal conductance model suggest O_3 uptake to be mainly limited by temperature. This study demonstrates the importance of accounting for the influences by climate and CO_2 on stomatal O_3 uptake, and of developing their representation in models, for risk assessment involving climate change.

1. Introduction

Surface ozone (O_3) is an air pollutant of major concern, causing reduced crop yield and quality, impaired forest growth and negative effects on human health (The Royal Society, 2008). Due to its strong dependence on meteorological conditions, surface O_3 is sensitive to climate change (Jacob and Winner, 2009). A number of studies have reported increasing background O_3 concentrations over the mid-latitudes of the Northern Hemisphere during the last decades (Vingarzan, 2004; Derwent et al., 2007) and modelling studies indicate a significant rise in global mean O_3 concentration in the future unless large emission reductions are implemented (Prather et al., 2003; Dentener et al., 2006; Stevenson et al., 2006). For Europe, regional air quality models, simulating the conditions during future climate, generally show increasing O_3 concentrations despite constant anthropogenic precursor emissions (Langner et al., 2005; Meleux et al., 2007; Andersson and Engardt, 2010). The increase is mostly explained by increased temperature, decreased cloudiness (Meleux et al., 2007) and reduced dry deposition (Andersson and Engardt, 2010). Thus, climate change has the potential to counteract emission reductions aimed to limit surface O_3 concentrations. This is important to consider in future air quality and emission control policies (Meleux et al., 2007).

Current global yield losses has been estimated to be in the range of 3–16% for four major food crops due to exposure to surface O_3 concentrations, with additional reductions under possible higher O_3 concentrations in the future, posing a serious threat to food security (Van Dingenen et al., 2009). However, the negative effects of O_3 on vegetation are more closely related to the uptake of O_3 through the stomata than to the concentration in the ambient air (Emberson et al., 2000b; Ashmore et al., 2004; Pleijel et al., 2004; Uddling et al., 2004; Karlsson et al., 2007a). Factors influencing the stomatal uptake of O_3 therefore have to be considered in risk assessment. A flux-based index (PODY, phytotoxic O_3 dose above a flux threshold Y, earlier called $AF_{st}Y$, accumulated stomatal flux of O_3) has been developed, which takes into account the influence of temperature, solar radiation, water vapour pressure deficit (VPD), soil water potential (SWP), atmospheric O_3 concentration and plant development stage (phenology) on stomatal O_3 uptake (LRTAP Convention, 2004; Pleijel et al., 2007). For a given O_3

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concentration, the stomatal flux (O_3 uptake) will be greater under humid conditions since dry air and soil moisture deficit induce stomatal closure to minimize plant water loss through transpiration. Evaluations have shown the PODY index to be superior to the concentration-based index AOT40 in explaining yield reductions for wheat and potato (Pleijel et al., 2004) as well as biomass reductions and visible leaf injury for O_3 sensitive tree species (Uddling et al., 2004; Karlsson et al., 2007a). Unlike the concentration-based index AOT40, the flux-based approach allows modification of O_3 sensitivity by climatic conditions to be incorporated into the risk assessment.

Models of stomatal conductance (g_s) have been parametrized for a limited number of crop and tree species. Within the UNECE LRTAP Convention, a simplified multiplicative g_s model has been developed and employed to quantify stomatal O_3 flux and estimate the risk for O_3 damage to a generic crop and generic tree species across Europe (LRTAP Convention, 2004; Simpson and Emberson, 2006) to be used in large-scale modelling. Maps of modelled flux-based O_3 risk for a generic crop have been shown to better correspond with field-based evidence of adverse affects compared to AOT40 (Hayes et al., 2007).

In assessing future O_3 risk for vegetation it is important to consider the reduction of g_s in elevated CO_2 concentrations (Ainsworth and Rogers, 2007). Plants do not maximise the CO_2 uptake, but rather optimise the water use efficiency to loose as little water as possible per CO_2 taken up (Jones, 1992). In higher CO_2 concentrations, the optimum water use efficiency tends to be achieved with smaller stomatal opening. In a case study for winter wheat, Harmens et al. (2007) assumed a 35% reduction in g_s due to elevated CO_2 concentrations and modified current (1997) meteorology and O_3 concentrations input data to POD6 calculations for five grid squares in the EMEP model. The results showed that with a 3°C increase in temperature and constant absolute humidity, the absorbed O_3 dose decreased, despite an assumed 5 ppb increase in O_3 concentrations. The result is however based on simplified assumptions of the future climate. Sitch et al. (2007) estimated the impact of projected O_3 concentrations on the land-carbon sink, by including the effect of O_3 deposition on photosynthesis and the interactions between O_3 and CO_2 through stomatal closure in their global land-carbon cycle model. Their results suggest a significant suppression of the global land-carbon sink due to negative effects of O_3 on plant productivity. CO_2-induced stomatal closure was found to offset the O_3 suppression of gross primary production by more than one third. Hence, the combined effects of climatic conditions and elevated CO_2 concentrations on future O_3 risk for vegetation are important for future crop and forest production and should be accounted for in coupled biosphere-climate models.

In this study, O_3 concentrations from a regional chemistry transport model (MATCH), driven by meteorological data from a regional climate model (RCA3), was used to estimate reference (1961–1990), near future (2021–2050) and far future (2071–2100) flux-based O_3 risk (PODY) in Europe using the Mapping Manual stomatal flux model (LRTAP Convention, 2004). Anthropogenic precursor emissions and boundary concentrations were held constant in the chemistry transport model, in order to limit the investigation to the influence of climate change.

The aims of the study were:

- To assess the influence of climate change and elevated CO_2 concentrations on the flux-based risk for O_3 damage to a generic crop and a generic deciduous tree in Europe. The hypotheses were that even without changes in O_3 precursor emissions, climate change could significantly modify the flux-based O_3 risk and that the plant stomatal response to elevated CO_2 concentrations would have the potential to significantly reduce the O_3 risk.
- To describe the spatial differences in the modelled reference and future flux-based risk at ten sites along a transect from northeast to southwest in Europe. The hypothesis was that dry air and high soil moisture deficit would substantially limit stomatal flux of O_3 in the south, while low temperatures would be limiting in the north.
- To compare the flux-based risk assessment with that based on AOT40. The hypothesis was that AOT40 would predict a relatively smaller risk in the northern part of Europe and larger risk in the southern part of Europe in both reference and future climate, compared to the flux-based risk assessment.

2. Methods

2.1. *Stomatal O_3 flux*

The stomatal O_3 flux was calculated using a multiplicative algorithm (an extension of the concepts presented in Jarvis (1976), Emberson et al. (2000a) and Emberson et al. (2000b)), which includes functions accounting for the limiting effects of various abiotic factors on stomatal conductance, thereby regulating the O_3 flux into the plant leaf. The multiplicative algorithm is (LRTAP Convention, 2004)

$$g_s = g_{max} \times [\min(f_{phen}, f_{O_3})] \times f_{light} \times \max\{f_{min}, (f_{temp} \times f_{VPD} \times f_{SWP})\}, \quad (1)$$

where g_s is the stomatal conductance [mmol O_3 m^{-2} sunlit projected leaf area (PLA) s^{-1}] and g_{max} is the species-specific maximum g_s. The functions f_{phen}, f_{O_3}, f_{light}, f_{temp}, f_{VPD} and f_{SWP} are expressed in relative terms (take values between 0 and 1) as a proportion of g_{max}. These parameters allow for the influence of phenology and O_3, and the environmental variables (irradiance, temperature, water VPD and SWP) on g_s to be estimated. The part of eq. (1) related to f_{phen} and f_{O_3} is a most limiting factor approach; i.e. g_s is limited by either senescence due to normal aging or premature senescence induced by O_3.

The stomatal flux (F_{st}) of O_3 to a plant leaf is calculated using a resistance analogue

$$F_{st} = C(z) \times \frac{1}{r_b + r_c} \times \frac{g_s}{g_s + g_{ext}}. \quad (2)$$

The O_3 concentration at the top of the canopy [C(z)] is assumed to be a reasonable estimate of the concentration at the surface of the laminar leaf boundary layer near the sunlit upper canopy leaves. The $1/(r_b + r_c)$ term is the deposition rate to the leaf determined by the quasi-laminar resistance (r_b) and the leaf surface resistance (r_c) $g_s/(g_s + g_{ext})$ represents the fraction of O_3 taken up through the stomata, where $1/g_{ext}$ is the external leaf resistance.

The POD accumulated per unit projected sunlit leaf area above a threshold of Y nmol m^{-2} s^{-1} was calculated as

$$\text{PODY} = \int \max(F_{st} - Y, 0)\, dt. \quad (3)$$

The PODY is accumulated over a period of time corresponding to the part of the growing season when the plant is considered to be sensitive to O_3.

In this study a simplified flux-based method, recommended for large-scale modelling (LRTAP Convention, 2004; Simpson and Emberson, 2006) was used to indicate the risk for O_3 damage to a generic crop (POD3$_{crop}$) and a generic deciduous tree (POD1.6$_{tree}$). Important simplifications were that O_3-induced premature senescence was assumed to have no effect on g_s (i.e. $f_{O_3} = 1$) and that a lower threshold were used for the generic crop than recommended by the LRTAP Convention (2004) for specific crops ($Y = 3$ nmol m^{-2} PLA s^{-1} for a generic crop compared to $Y = 6$ nmol m^{-2} PLA s^{-1} for wheat and potato), making the method numerically more robust (Tuovinen et al., 2007). The threshold for trees is $Y = 1.6$ nmol m^{-2} PLA s^{-1}. For a generic crop a 3-month time-window for stomatal O_3 flux accumulation based on latitude (LRTAP Convention, 2004), was used to bypass the uncertainty in the timing of the relevant time-interval, which, especially for crops, is rather short. The start and length of the generic deciduous tree stomatal O_3 accumulation period was based on latitude and altitude (LRTAP Convention, 2004). Due to uncertainties caused by difficulties in modelling a plant-relevant SWP, the potentially large variations in soil moisture within a model grid and possible irrigation practices, soil moisture was assumed not to limit g_s (i.e. $f_{SWP} = 1$). In one PODY calculation for the generic deciduous tree, however, a SWP function was included to estimate the potential influence of this factor on g_s and the stomatal O_3 flux. It was assumed that water was freely available from 0 to –0.05 MPa SWP with a linear decrease in water availability (and g_s) below –0.05 MPa down to a minimum at –1.5 MPa SWP (Hall et al., 1977). The stomatal flux algorithm and the parameterisation of the simplified flux-based method for a generic crop and a generic deciduous tree are further described in the LRTAP Convention (2004).

2.2. The CO_2 function

Several studies have shown reduced g_s (on average 0.66 of that in 370 ppm CO_2) for wheat grown in 550–570 ppm CO_2 concentrations (Wall et al., 2000, 2006). For wheat grown in 700 ppm CO_2, the average g_s ratio at elevated to ambient (~350 ppm during daytime) CO_2 was 0.56 (Bunce, 2000). In a meta-analysis, Ainsworth and Rogers (2007) found the reduction in stomatal conductance of C_3 crops to be 25% in 567 ppm CO_2 compared to ambient (366 ppm) and Pleijel et al. (2002) found up to a 20% reduction in g_s for potato grown in 680 ppm CO_2 compared to 360 ppm. Ainsworth and Rogers (2007) also found a 19% reduction of g_s in trees grown in elevated CO_2 concentrations (567 ppm on average) compared to ambient CO_2 concentrations (366 ppm on average). Medlyn et al. (2001) reported a 21% decrease in g_s for trees in response to growth in 700 ppm CO_2 compared to 350 ppm CO_2. Based on these studies, CO_2 response functions (f_{CO_2}) for a generic crop and a generic deciduous tree were parametrized and added to eq. (1). The influence of increasing CO_2 on g_s was, in this study, assumed to linearly decrease between 360 and 560 ppm CO_2 concentration from 1 to 0.66 for a generic crop and to 0.8 for a generic deciduous tree, with no further reductions in g_s above 560 ppm CO_2 (Fig. 1). However, there is a large uncertainty with respect to the effect of elevated CO_2 on g_s in closed forest stands (Uddling et al., 2009 and references therein). Because of this uncertainty, the possible effect of CO_2 on PODY was explored as a sensitivity study, by performing the calculations with and without inclusion of f_{CO_2}. This was done also for the generic crop in order to separate effects of climatic changes from effects of rising CO_2 concentrations on stomatal O_3 flux.

2.3. MATCH and RCA3 model set-up and PODY input data

MATCH is a Eulerian, off-line, regional chemistry transport model. A detailed description of the model is available in Robertson et al. (1999). A number of previous studies have demonstrated the ability of MATCH to realistically simulate O_3 concentrations over Europe (e.g. Tilmes et al., 2002; Andersson et al., 2007; van Loon et al., 2007). In order to investigate the effect of future climate change on surface O_3, MATCH was driven by meteorology from the Rossby Centre's regional climate model (RCA3), described in Kjellström et al. (2005). In this study RCA3 was forced with climate data from the ECHAM4/OPYC3 global model on its boundaries simulating the IPCC SRES A2 and B2 emission scenarios (Nakicenovic et al., 2000). A2 is one of the more pessimistic greenhouse gas emission scenarios. The two emission scenarios yield similar climate change evolution for the first half of the 21st century, but in the second half of the 21st century there is an increasingly larger signal in the A2 scenario. The canopy-scale O_3 dry deposition to vegetation in MATCH is a function of soil moisture, air humidity, temperature and irradiance. Also, emissions

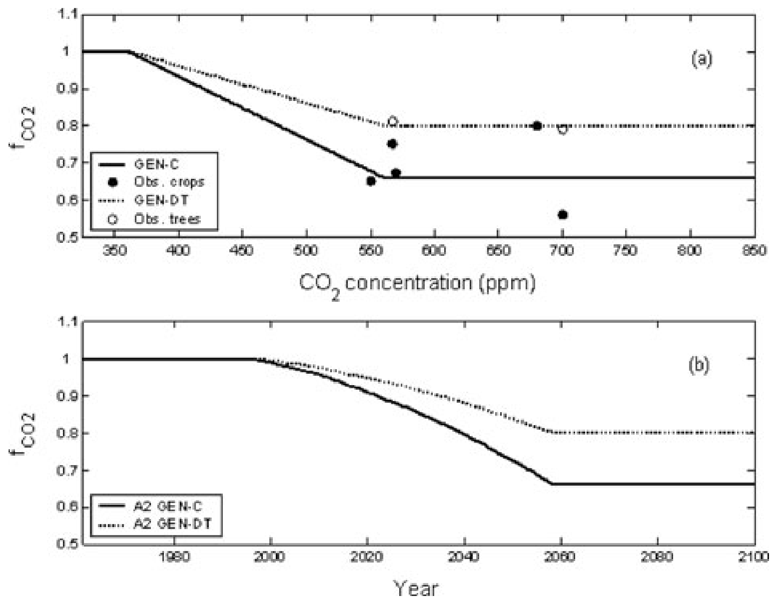

Fig. 1. The influence of atmospheric CO_2 concentrations on stomatal conductance for a generic crop (GEN-C) and for a generic deciduous tree (GEN-DT). Relative stomatal conductance (f_{CO_2}) in relation to (a) CO_2 concentration and (b) year using the A2 emission scenario. The data points in (a) are the observations on which the CO_2 functions were based, see Section 2.2 for references.

of biogenic isoprene are calculated online in MATCH. Natural emissions of other VOCs, sulphur or nitrogen containing compounds are not included in the present set-up. Hence, modelled future O_3 concentrations are affected by changes in photochemistry, transport patterns, emissions and uptake by vegetation. Anthropogenic emissions and tracer boundary concentrations were set constant in MATCH, representing the year 2000, in order to isolate the influence of climate change and not changes in emission patterns. In MATCH three 30-yr periods were simulated: reference (1961–1990), near future (2021–2050) and far future (2071–2100). The set-up of the model system is further described in Andersson and Engardt (2010). To test if average PODY for the far future (A2 emission scenario) was significantly different compared the reference period, a two-sided Student's *t*-test was applied, assuming unequal variances, with 58 degrees of freedom ($2 \times N-2$; $N = 30$ yr).

For risk assessment purposes it is the stomatal flux of O_3 to the sunlit leaf level of specified vegetation types and not the canopy-scale flux that is important. The PODY was, in this study, calculated off-line for a generic crop and a generic deciduous tree at 10 sites based on modelled data from the corresponding grid-cell in MATCH (O_3) and RCA3 (meteorology). The ten monitoring sites within the European Monitoring and Evaluation Programme (EMEP) within CLRTAP were selected in a transect from northeast to southwest to represent different climatic conditions in Europe. Site characteristics and estimated growing seasons can be found in Table 1 and site locations in Fig. 2 which also shows the modelled April to September average daily maximum O_3 in reference climate (1961–1990). PODY was calculated for both a generic crop and a generic deciduous tree at all sites although, for example, wheat is presently not a suitable crop at the northernmost site. However, the distribution of crops and trees are likely to change in response to climate changes.

O_3 and meteorological input data for the PODY calculations should be valid for the height of the canopy which is assumed to be 1 m above ground level for a generic crop and 20 m for a generic deciduous tree. In MATCH, the O_3 concentrations representing the lowest model layer were downscaled to 3 m at every time-step based on local stability and surface deposition velocity using similarity theory. To avoid systematic errors, a further correction was applied to adjust the 3 m O_3 concentrations from MATCH to canopy height. Since values of u^* and canopy-scale deposition to vegetation in MATCH were not available (not saved as output data), the simplified method in the Mapping Manual (LRTAP Convention, 2004) was used, assuming that the O_3 concentration at 1 m was 93% of the 3 m concentration and that the O_3 concentration at 20 m was 105% of the 3 m concentration). Modelled wind speed (3 h resolution, from RCA3) at 10 m was adjusted to canopy height using the logarithmic wind law, assuming neutral stability. Canopy height wind speed was used to estimate the leaf boundary layer resistance required in the flux calculation. Modelled temperature and relative humidity (from RCA3) corresponding to 2 m had 3 h temporal resolution. Linear interpolation was applied to receive the hourly temporal resolution needed for the PODY calculations. Incoming short-wave radiation data with 30 min resolution was recalculated to photosynthetic photon flux density (PPFD) by multiplying with a factor of 2 (Monteith and Unsworth, 2008). Soil water content summed over the top two soil layers in RCA3 (2.3 m) with six hour resolution was recalculated to SWP using the Groenevelt-Grant soil water retention model

Table 1. Site coordinates and characteristics according to the EMEP website (www.emep.int/) and soil texture class given by RCA3

Site	Country	Latitude Longitude	Altitude (m a.s.l.)	GEN-C growing season (day number)	GEN-DT growing season (day number)	Soil texture class (RCA3)
FI22	N Finland	66°19'N 29°24'E	310	165–254	133–261	Sandy loam
FI37	S Finland	62°33'N 24°13'E	162	154–243	125–271	Sandy loam
SE32	S Sweden	57°49'N 15°34'E	261	144–233	120–278	Sand
SE11	S Sweden	56°01'N 13°09'E	172	139–228	116–283	Loam
DE02	N Germany	52°48'N 10°45'E	74	131–220	111–290	Sand
DE03	S Germany	47°55'N 7°54'E	1205	118–207	115–288	Loam
FR10	C France	47°16'N 4°05'E	620	116–205	107–296	Silt loam
FR13	S France	43°37'N 0°11'E	236	106–195	97–308	Silt loam
ES09	N Spain	41°16'N 3°08'W	1360	100–189	106–301	Loam
ES07	S Spain	37°14'N 3°28'W	1230	90–179	98–310	Silt loam

Note: The growing season for a generic crop (GEN-C) is based on latitude and the growing season for a generic deciduous tree (GEN-DT) is based on latitude and altitude (LRTAP Convention, 2004).

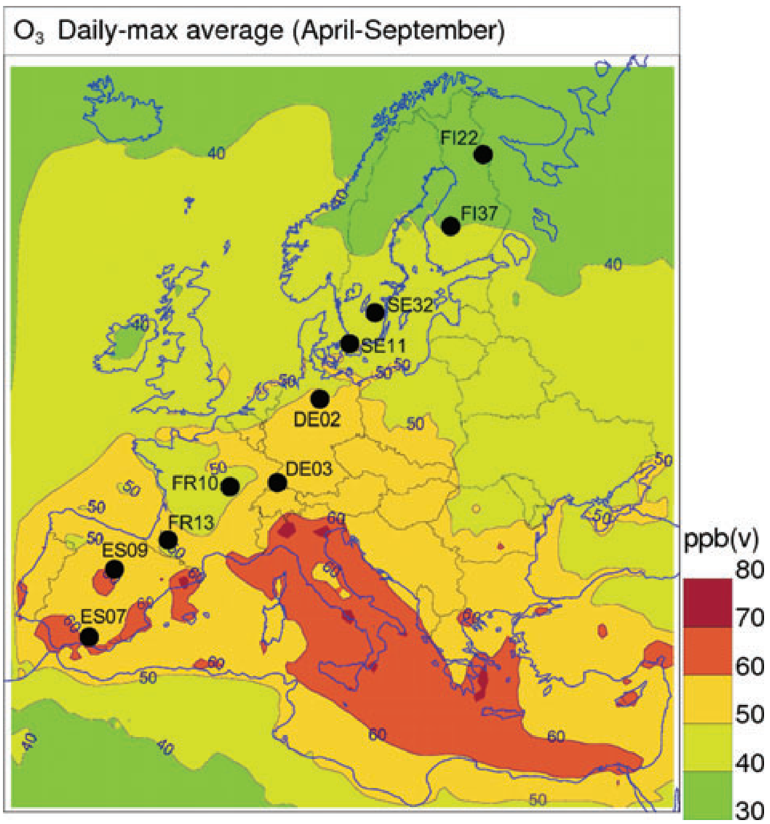

Fig. 2. Location of the ten EMEP sites considered in the PODY calculations. Also shown is the modelled average summer-time (April–September) daily maximum O_3 in reference climate (1961–1990).

(Groenevelt and Grant, 2004) anchored in the point of saturation (Grant et al., 2010)

$$\theta(h) = \theta_s - k_1 \left\{ \exp\left(\frac{k_0}{h}\right)^n \right\}, \quad (4)$$

where θ is the relative water content ($m^3\ m^{-3}$), θ_s is the relative water content at the point of saturation ($m^3\ m^{-3}$) and h is the matric suction (m). k_0, k_1 and n are freely adjustable fitting parameters. Values of the parameters used to fit 20 Dutch soils to the Groenevelt-Grant model (Grant et al., 2010) was used for the soil types most similar to those in RCA3 (see Table 1).

2.4. *Comparison of simulations and observations*

The MATCH-RCA3 performance have earlier been evaluated and shown good agreement with O_3 concentrations measured at EMEP stations in Europe (Engardt et al., 2009; Andersson and Engardt, 2010). The bias for summer and yearly average and daily maximum concentrations was within ±10% and correlations were 0.66–0.90. In this study, average and daily maximum O_3 concentrations during the generic crop O_3 accumulation period were used to estimate the performance of the MATCH-RCA3 modelling system for the ten selected EMEP monitoring sites. Modelled 3 m O_3 concentrations from the last 10 yr of the reference period (i.e.1981–1990) were compared to O_3 measurements during 1995–2006. The measurements were centred around the year of precursor emissions used in MATCH (year 2000). Only years with less than 10% missing data during the generic crop-growing season were included. The (spatial) correlations were high, 0.60 for average O_3 and 0.85 for daily maximum, and the average bias small (+1.5% bias of average O_3 and –3.7% bias of the daily maximum).

Hourly temperature and VPD from RCA3 (1981–1990) were compared to the surface analysis (1998–2005) from a high resolution (11 km) analysis system (MESAN, Häggmark et al., 2000). MESAN-data were only available for the five northernmost sites (FI22, FI37, SE32, SE11 and DE02). Comparison of data from the generic crop O_3 accumulation period showed a good agreement of average temperature from RCA3 (–2.5% bias in °C) but consistently smaller range and standard deviation. The 90-percentile temperature was 3.2°C lower in RCA3 compared to the surface analysis on average. Since high VPD occurs at high temperatures, the underestimation in temperature results in a large VPD underestimation (–43% bias on average). An explanation could be smaller diurnal variation in the regional scale RCA3 compared to the surface analysis with higher resolution and a better capture of local scale processes. The large underestimation of VPD may cause overestimation of g_s and stomatal O_3 flux. The sensitivity of the calculation of $POD3_{crop}$ due to underestimated VPD input data was analysed by the calculation of the percentage change in the $POD3_{crop}$ index caused by a 50% increase in the modelled VPD from RCA3. Modelled temperature, VPD and O_3 used for the PODY calculations are shown in Fig. 3.

3. Results

3.1. *Concentration-based versus flux-based risk for O_3 damage to a generic crop*

In general, model calculated O_3 concentrations increase towards southern Europe (Fig. 2). The concentration-based AOT40 index, calculated for daylight hours during the 3 months corresponding to the generic crop growing season (Fig. 4), indicates a higher O_3 risk in southern compared to northern Europe. Already in the reference climate the current AOT40 critical level of 3 ppm h (LRTAP Convention, 2004) is exceeded at many sites. Note, however, that the 3 month growing season depends on latitude in this study, which can differ from the May–July AOT40, used e.g. in EU directives for vegetation risk assessments. At 6 of the 10 sites investigated, there is a large increase in future AOT40, especially in the 2071–2100 A2 emission scenario, where five sites (DE03, FR10, FR13, ES09 and ES07) will experience more than a doubling of AOT40. The four Nordic sites (FI22, FI37, SE32 and SE11) exhibit small changes in AOT40. Since emissions were held constant during the simulations, the increase in O_3 concentration is entirely due to changes in the climate. The increase in southern Europe O_3 concentrations has been explained by reduced dry deposition caused by a changed climate affecting the uptake to vegetation, while increasing isoprene emissions played a minor role (Andersson and Engardt, 2010).

The geographical pattern of the generic crop $POD3_{crop}$ is considerably different compared to that of AOT40 (Fig. 5). Both in reference and future climate, the stomatal O_3 flux is largest in central Europe. The reduction in g_s is 34% in the 2071–2100 A2 period due to CO_2 concentrations above 550 ppm (Fig. 1). As a result, the sites with the largest increase in AOT40 by 2071–2100 in the A2 emission scenario show non-significant changes in $POD3_{crop}$ (DE03 and FR10) or a large decrease in $POD3_{crop}$ (statistically significant with $p < 0.0001$ for FR13, ES09 and ES07). In the same period, the two Swedish sites show a small but significant increase in $POD3_{crop}$ ($p = 0.0119$ for SE32 and $p = 0.0268$ for SE11). The same geographical pattern and direction of changes are found for $POD0_{crop}$ (data not shown), indicating that the result is not dependent on the choice of threshold.

At the northern sites the O_3 concentration is low and the temperature function significantly limits the g_s (average f_{temp} substantially below 1, see Table 2). With no inclusion of f_{CO_2}, $POD3_{crop}$ increase significantly at the Nordic sites in the 2071–2100 A2 period compared to the reference period ($p < 0.0001$ for FI22, FI37, SE32 and SE11). The increase can be explained by higher temperatures and, for SE11, to some extent higher O_3 concentrations. For example, at the northernmost

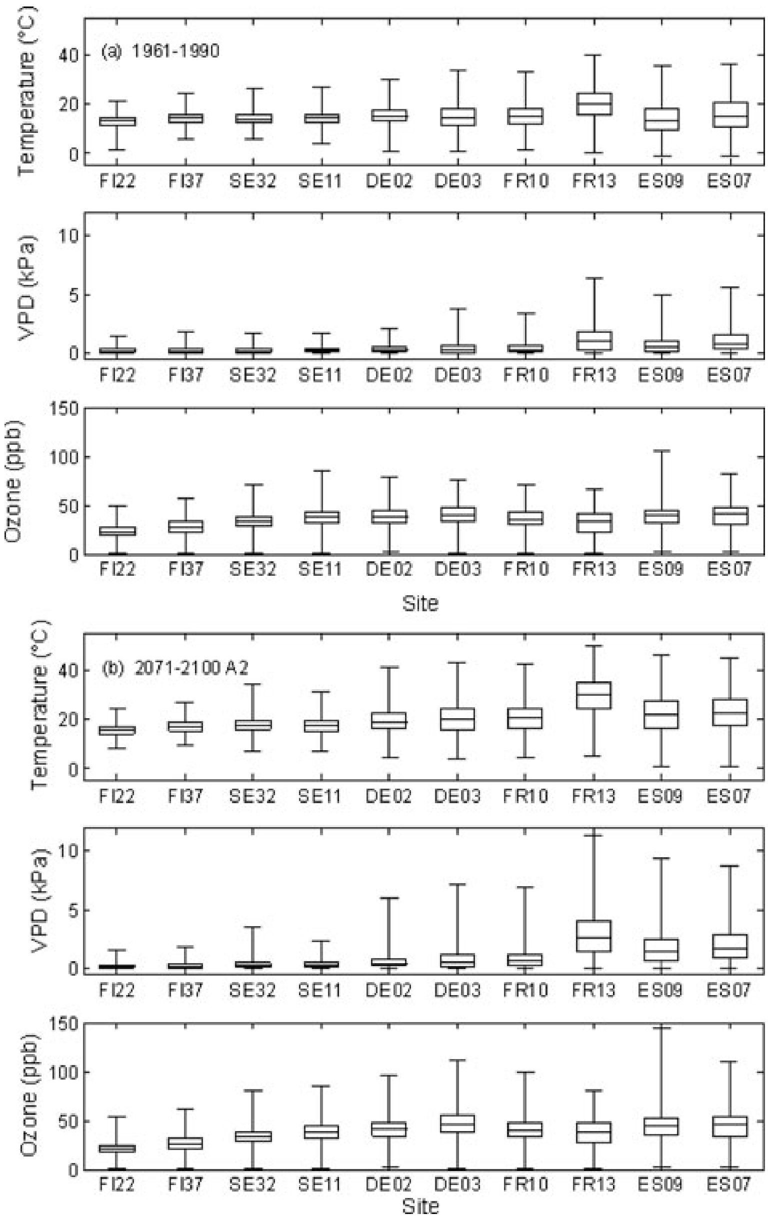

Fig. 3. Box- and whisker plots of the RCA3 and MATCH data for the accumulation period of the generic crop used as input in the stomatal conductance model during (a) 1961–1990 and (b) 2171–2100 using the SRES A2 emission scenario.

site (FI22) the average f_{temp} in reference climate is only 0.24 but increase to 0.49 in 2071–2100 A2, while the average VPD influence on stomatal conductance does not change (average f_{VPD} does not fall below 1.00 in reference or future climate, see Table 2). Without the influence of f_{CO_2}, $POD3_{crop}$ more than double at FI22 while the average O_3 concentration decrease by approximately 1 ppb.

In central Europe, the O_3 concentrations are high and climatic conditions favour stomatal O_3 uptake. The large increase in O_3 concentrations by 2071–2100 A2, together with considerably higher f_{temp}, but only slightly lower f_{VPD} (Table 2) result in a significantly increased stomatal O_3 flux compared to reference climate ($p < 0.0001$ for DE02, DE03 and FR10), when f_{CO_2} is not included in the calculation.

As shown in Table 2, f_{VPD} substantially limits the g_s at the three southernmost sites (FR13, ES09 and ES07) already in reference climate. For example, average f_{VPD} at the southernmost site (ES07) is 0.75 compared to 1.00 at the site with largest $POD3_{crop}$ (DE02). Even without the inclusion of f_{CO_2}, the stomatal O_3 flux does not increase in the future climate, despite a large increase in O_3 concentration (5 ppb on average) by 2071–2100 A2, due to increased VPD.

The pattern is similar for the B2 emission scenario (see Appendix S1 in the Supporting Information). The increase in O_3

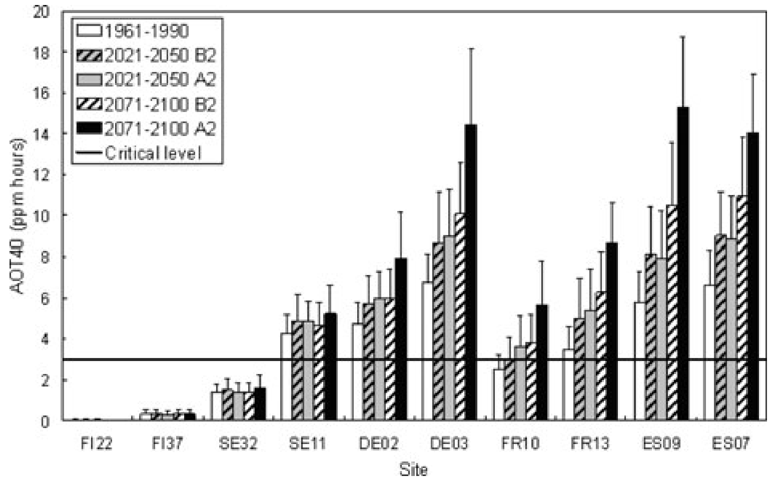

Fig. 4. Average 3-month daylight AOT40 (corresponding to the generic crop growing season) at the different sites, based on O_3 concentrations at 1 m. Error bars show standard deviation ($N = 30$ yr). Current critical level is 3 ppm h, which is indicated by the horizontal line.

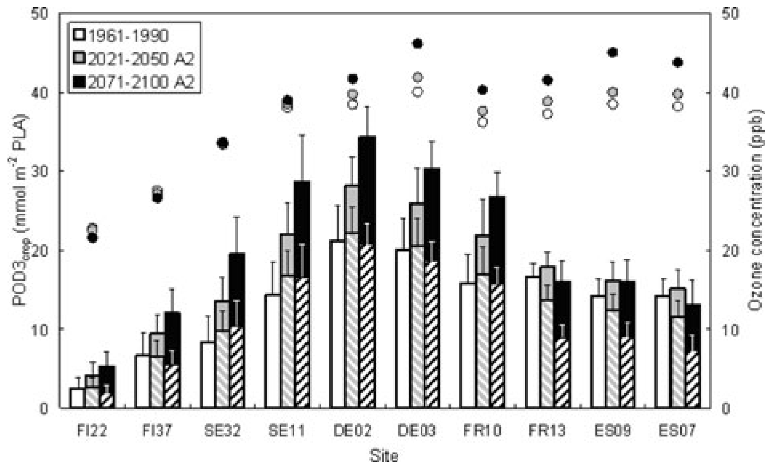

Fig. 5. Average phytotoxic O_3 dose for a generic crop ($POD3_{crop}$) during 1961–1990, 2021–2050 and 2071–2100 following the SRES A2 emission scenario. The striped part of the bars show average $POD3_{crop}$ when the g_s response function for CO_2 (f_{CO_2}) is included in the calculation. Error bars show standard deviation ($N = 30$ yr). Circles are average O_3 concentration (scale on the right) for the same time periods as $POD3_{crop}$ is accumulated.

Table 2. Average daylight f_{temp} and f_{VPD} for a generic crop during the 30-yr reference period and year 2071–2100 following the SRES A2 emission scenario

	1961–1990		2071–2100 A2	
Site	f_{temp}	f_{VPD}	f_{temp}	f_{VPD}
FI22	0.24	1.00	0.49	1.00
FI37	0.36	1.00	0.61	1.00
SE32	0.34	1.00	0.65	0.99
SE11	0.37	1.00	0.65	1.00
DE02	0.48	1.00	0.73	0.96
DE03	0.46	0.98	0.72	0.84
FR10	0.46	0.99	0.74	0.87
FR13	0.58	0.85	0.70	0.53
ES09	0.47	0.88	0.72	0.56
ES07	0.59	0.75	0.77	0.44

concentration at the five southernmost sites is smaller than in the A2 emission scenario (2.7 ppb difference between the 2071–2100 B2 and the reference period, compared to 5.3 ppb in A2). The f_{VPD} does not limit g_s at the southernmost sites to the same extent, while the increase in g_s due to increasing temperature at the northernmost sites is not as large.

The effect of increasing the calculated VPD from RCA3 with 50% on average daylight f_{VPD} and $POD3_{crop}$ is shown in Table 3. The g_s response to elevated CO_2 was not included in this calculation. The average f_{VPD} is mainly influenced at the southern sites in the reference period and the central and southern sites in the 2071–2100 A2 period. The decrease in f_{VPD} result in a considerable reduction of $POD3_{crop}$ compared to the base calculations at the southern sites. However, the temporal trend and geographical pattern is not affected.

3.2. *Concentration-based versus flux-based risk for O_3 damage to a generic deciduous tree*

The spatial pattern of AOT40 calculated over the generic deciduous tree growing season (data not shown) is very similar to that of the generic crop, with large increases in AOT40 at the six southernmost sites, especially for the 2071–2100 A2 period. The spatial pattern of the generic deciduous tree $POD1.6_{tree}$ following the A2 scenario (Fig. 6) is also similar to that for a generic crop ($POD3_{crop}$), with largest stomatal O_3 flux in central

Table 3. The effect of increasing the calculated VPD from RCA3 with 50% on the average daylight f_{VPD} and average $POD3_{crop}$ (f_{CO_2} not included) for a generic crop during the 30-yr reference period and year 2071–2100 following the SRES A2 emission scenario

	1961–1990 (VPD×1.5)			2071–2100 A2 (VPD×1.5)		
Site	f_{VPD}	$POD3_{crop}$	%change	f_{VPD}	$POD3_{crop}$	%change
FI22	0.99	2.2	−14	1.00	5.1	−3
FI37	0.99	6.2	−7	0.99	11.7	−2
SE32	0.99	7.9	−5	0.96	17.9	−8
SE11	0.99	14.0	−2	0.98	27.3	−5
DE02	0.98	19.7	−7	0.87	28.3	−18
DE03	0.92	16.0	−20	0.70	21.0	−31
FR10	0.96	13.6	−14	0.73	19.2	−28
FR13	0.70	9.5	−42	0.37	8.4	−47
ES09	0.73	7.2	−50	0.38	7.3	−54
ES07	0.55	5.6	−61	0.26	5.0	−62

Note: % change refers to the change in $POD3_{crop}$ compared to the calculation without the increase in VPD (shown in Fig. 5).

Table 4. Average daylight f_{temp}, f_{VPD} and f_{SWP} for a generic deciduous tree during the 30-yr reference period and year 2071–2100 following the SRES A2 emission scenario

	1961–1990			2071–2100 A2		
Site	f_{temp}	f_{VPD}	f_{SWP}	f_{temp}	f_{VPD}	f_{SWP}
FI22	0.77	1.00	0.99	0.88	1.00	0.99
FI37	0.82	1.00	0.99	0.91	1.00	0.99
SE32	0.83	1.00	1.00	0.92	0.99	1.00
SE11	0.84	1.00	1.00	0.94	1.00	0.98
DE02	0.87	1.00	1.00	0.92	0.95	0.97
DE03	0.88	0.97	0.96	0.84	0.78	0.83
FR10	0.88	0.99	0.99	0.85	0.81	0.79
FR13	0.86	0.81	0.45	0.66	0.53	0.22
ES09	0.85	0.80	0.80	0.71	0.55	0.60
ES07	0.84	0.64	0.38	0.64	0.44	0.20

Europe. At all sites $POD1.6_{tree}$ was significantly reduced in the 2071–2100 A2 period compared to the reference period ($p < 0.0001$) due to the assumed reduction of g_s by 20% with CO_2 concentrations above 550 ppm (Fig.1).

Low and decreasing O_3 concentrations as well as low average f_{temp} (see Table 4) result in small $POD1.6_{tree}$ at the two northernmost sites (FI22 and FI37), and no significant change by 2071–2100 A2 when the influence of f_{CO_2} is not included in the calculation. SE32, SE11 and DE02 experience increased stomatal O_3 flux in the 2071–2100 A2 period ($p < 0.0001$) compared to the reference period, due to climate changes, explained by increases in O_3 concentration and average f_{temp}.

The g_s of the generic deciduous tree has a lower temperature optimum compared to the generic crop ($T_{opt} = 21°C$ instead of 26°C) and at the southernmost sites (FR13, ES09 and ES07) high temperatures limit g_s in 2071–2100 A2. In addition to the dry air conditions (low average f_{VPD}, see Table 4) this result in a significantly reduced stomatal O_3 flux ($p < 0.0001$) despite a large increase in average O_3 concentration even without the inclusion of f_{CO_2} in the calculations. The result for the B2 emission scenario can be seen in Appendix S2 in the supporting information

To estimate the potential influence of soil moisture, a SWP function was included in one $POD1.6_{tree}$ calculations (Fig. 7). The g_s response to elevated CO_2 was not included. The results indicate that f_{SWP} mainly limit O_3 uptake at the southern sites (Table 4). The SWP function is especially limiting at FR13 and ES07.

4. Discussion

This study investigated how the changing climatic conditions and elevated atmospheric CO_2 concentrations may modify leaf uptake of O_3 and, hence, the risk for negative impacts on crops

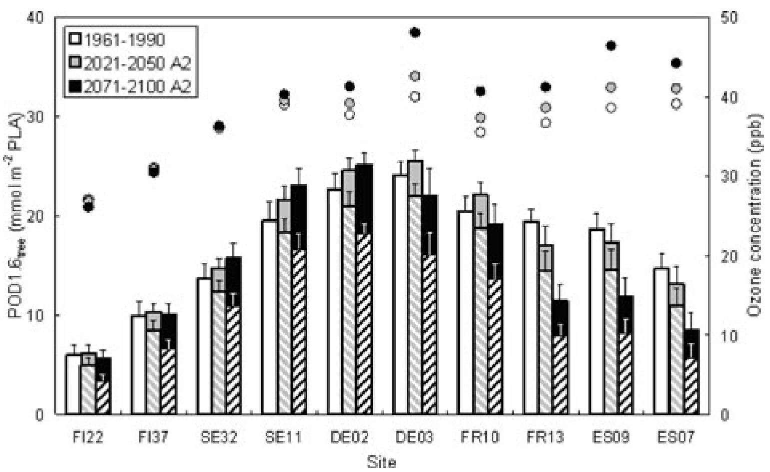

Fig. 6. Average phytotoxic O_3 dose for a generic deciduous tree ($POD1.6_{tree}$) during 1961–1990, 2021–2050 and 2071–2100 following the SRES A2 emission scenario. The striped part of the bars show average $POD1.6_{tree}$ when the g_s response function for CO_2 (f_{CO_2}) is included in the calculation. Error bars show standard deviation ($N =$ 30 yr). Circles are average O_3 concentration (scale on the right) for the same time periods as $POD1.6_{tree}$ is accumulated.

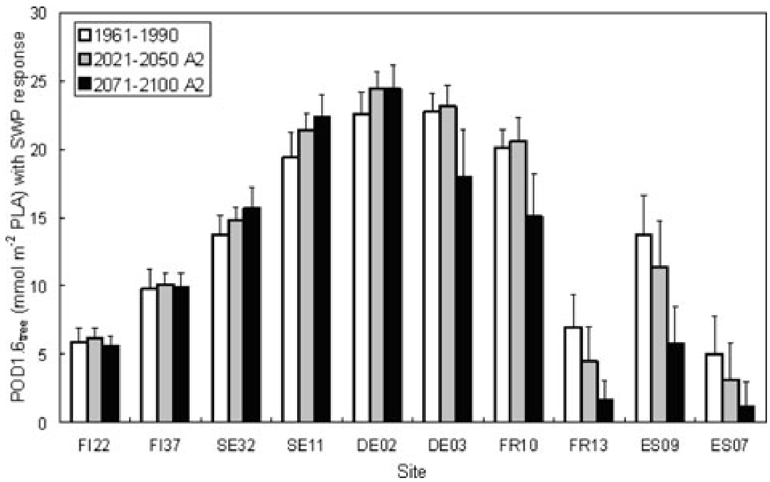

Fig. 7. Average phytotoxic O_3 dose for a generic deciduous tree (POD1.6_{tree}) during 1961–1990, 2021–2050 and 2071–2100 following the SRES A2 emission scenario. Included in the POD1.6_{tree} calculation is the g_s response function for SWP (f_{SWP}), but not for CO_2 (f_{CO_2}). Error bars show standard deviation ($N = 30$ yr).

and forest trees. In agreement with earlier studies (Simpson and Emberson, 2006; Emberson et al., 2007; Simpson et al., 2007) the spatial flux-based risk pattern for O_3 damage differed substantially from the AOT40-based risk, in the reference as well as future climate and irrespective of emission scenario (A2 or B2). It is important to take the influence of climate change into account in future risk assessment of surface O_3, since factors such as warming, changes in amount and distribution of precipitation, shifts in growing season and elevated CO_2 concentrations affect the stomatal uptake of O_3 into the leaves (Harmens et al., 2007). Flux-based models are generally considered to be more physiologically relevant compared to exposure-based indices such as AOT40. The AOT40 index only reflects changes in the ambient O_3 concentration, while the flux-based PODY index also allows climatic conditions and the CO_2 concentration to modify stomatal uptake rates of O_3, in line with important physiological mechanisms.

The results in this study clearly showed that the expected reduction in g_s with rising atmospheric CO_2 concentrations is of large importance for the decrease in projected future flux-based O_3 risk. Harmens et al. (2007) also showed that the reduced g_s under higher CO_2 concentrations has the potential to significantly reduce the risk for O_3 damage. For crops, there seem to be consensus among studies that plants growing under elevated CO_2 concentrations have reduced g_s (Bunce, 2000; Wall et al., 2000; Ainsworth and Rogers, 2007), but the magnitude of this effect under realistic agronomic conditions is less certain. For forests, g_s may not at all decrease with rising atmospheric CO_2 concentrations in many tree species. In four out of five free-air CO_2 enrichments experiments in closed forests, g_s was not significantly reduced under elevated CO_2 concentrations (Ellsworth, 1999; Bernacchi et al., 2003 (pre-coppice canopy closure); Keel et al., 2007; Maier et al., 2008; Uddling et al., 2009; but see Gunderson et al., 2002 and Domec et al., 2009). In the long term, cumulative effects of elevated CO_2 on plant growth and stand structure may be more important than the primary stomatal closure response to increased CO_2 in determining g_s in closed forests (Uddling et al., 2008; Uddling et al., 2009).

In a flux-based risk assessment for pine, beech and oak during one year at three sites in Europe, Emberson et al. (2007) found phenology (f_{phen}) and soil water (f_{SWP}) to be the key drivers limiting the seasonal profile of O_3 flux. Tuovinen et al. (2009) emphasised the urgent need to develop soil moisture status modelling and its effect on g_s for forests (crops may be irrigated). In the base case of our results, soil moisture is assumed to never limit g_s, mainly because of difficulties in estimating the plant-relevant soil water potential valid for the integrated rooting zone of vegetation inside an entire model grid. The simulation including f_{SWP} for the generic deciduous tree indicated, however, that soil water deficits have the potential to limit stomatal uptake of O_3 in southern Europe and that this limitation is likely to increase in the future (Fig. 7). The conversion (based on available data from RCA3) of mean soil water content to mean SWP across a 2.3 m soil profile may represent an oversimplification as it does not capture the vertical gradients in soil water status and its integrated, non-linear, effect on plant water availability. Therefore, the assessment of the impact of geographical differences and temporal changes in soil moisture on stomatal O_3 flux presented here indicates directions rather than magnitudes of the likely effects.

Several studies have reported an earlier onset of spring and a prolongation of the growing season in mid and high latitudes associated with global warming (Linderholm, 2006; Menzel et al., 2006). Changes in the timing of the growing season were omitted in this study, which assumed a fixed growth interval based on latitude. For specific crops, such as wheat, cumulative exposure may decline in a warmer climate due to accelerated plant development (Fuhrer, 2009). An earlier start of the growing season for wheat could result in a reduced overlap between the O_3 exposure accumulation period and the peak summer O_3 levels (Harmens et al., 2007; Fuhrer, 2009). Conversely, an earlier growing season may lead to more frequent co-occurrence of sensitive stages and spring-time peaks in O_3 in the northern part of the Nordic countries (Karlsson et al., 2007b; Klingberg et al., 2009). For the generic crop, the 3 month growing season, much longer than the approximately 55-d period of high O_3 sensitivity for wheat (LRTAP Convention, 2004), allows the $POD3_{crop}$ index to be less sensitive to changes in the timing of the growing season and can be interpreted as maximum potential risk. For trees, the growth period, and thus the period of O_3 uptake, is likely to be prolonged in a future warmer climate (Taylor et al., 2008), implying an underestimation of the future O_3 risk in this assessment.

There are large uncertainties in both climate projections and parameters in models for O_3 risk assessment, as pointed out by Fuhrer (2009). Leaf properties determining the rate of O_3 uptake and the biochemical defence capacity can be modified by changes in temperature, air humidity, soil moisture and increasing CO_2 concentration. O_3 uptake calculations may only be accurate when applied under conditions representative of those under which the parameterisation was performed, increasing the uncertainty when extrapolating to future climatic conditions (Fuhrer, 2009). Ashmore (2005) requested the development of new models, linking stomatal flux and detoxification processes to carbon assimilation and allocation, to include the capacity of species to adapt to changes in nutrient and water availability. Also, Musselman et al. (2006) emphasized the need to consider detoxification mechanisms in flux-based models, but noted the large uncertainty in quantifying the various defence mechanisms in plants.

In this study, the current LRTAP Convention (2004) parameterisation of the g_s model indicated that f_{temp} was considerably more important than f_{VPD} in limiting O_3 uptake for the generic crop (Table 2) as well as for the generic tree at the northern sites (Table 4). This large importance of f_{temp} compared to f_{VPD} in limiting g_s could be questioned, considering the fact that minimizing water loss is a main function of stomata. The bell shaped optimum curve of the temperature function for crops seems uncertain as it predicts complete stomatal closure at temperatures below 12°C and above 40°C, which is hardly realistic for the Nordic and Mediterranean regions, respectively. Most physiological responses are known to acclimate to the prevailing temperature (e.g. Larcher, 2003; Körner, 2006) and both spatial (cold north vs. warm south) and temporal (during climate change) acclimation of the stomatal temperature response are thus likely. A certain degree of stomatal closure in response to low temperatures can be expected in colder conditions, but further evaluation of the stomatal temperature response function is required to improve predictions of the effect of climate on stomatal O_3 flux. In addition, modelled stomatal O_3 flux strongly depends on the quality of the g_{max} value (Fuhrer, 2009), which differs significantly between species and is likely to depend on climatic region.

The PODY calculations are strongly dependent on the quality of the input data. The coarse spatial resolution of regional climate models (50×50 km^2) limits the details of important processes, such as diurnal range and extremes. Local scale characteristics, e.g. promoting nighttime air temperature inversions, may cause large diurnal O_3 range deviating from the average conditions of the model grid (Klingberg et al., 2008). The large underestimation of VPD in RCA3 (–43% bias on average for the five northernmost sites) is of special concern for the stomatal O_3 uptake calculations and may cause an overestimation of the flux-based O_3 risk (Table 3). The importance of representative diurnal dynamics in temperature and VPD is emphasized when regional scale modelled data are used for stomatal O_3 uptake calculations. Klingberg et al. (2008) estimated the sensitivity of POD0 and POD6 calculations for potato to small changes in input data in more detail for a site in southwest Sweden. A 10% change in O_3 and temperature resulted in approximately 20% and 34–40% change in POD6, respectively. Without the flux threshold (POD0), the change was smaller. The sensitivity to a 10% change in VPD was much smaller (3–5% change in POD), but it was concluded that this factor presumably plays a much greater role in drier conditions. Other important simplifications are that the land-cover does not change with changing climate in MATCH and that meteorology is only based on one set-up of the climate model.

The uncertainties discussed above result in considerably uncertainty in the exact value of the stomatal O_3 fluxes reported here. The most important uncertainties are considered to be the projections of future atmospheric changes and the parameterisation of the stomatal O_3 flux model, neglecting both spatial and temporal acclimation and adaptation. More research is required to reduce the uncertainties before projected $POD3_{crop}$ and $POD1.6_{tree}$ can be used to give reasonable estimates of European yield or growth loss due to O_3 damage. This study does therefore not intend to predict European yield or growth losses due to O_3 damage in the future. The flux-based approach is, however, an improvement compared to AOT40. Notwithstanding the mentioned uncertainties we believe that there is high confidence in the general pattern and directions of change obtained in this study of how future climatic conditions will influence the O_3 risk for vegetation in Europe.

5. Conclusions

- The increase in modelled future surface O_3 concentrations is large in southern Europe, despite constant precursor emissions in MATCH. However, the future stomatal O_3 uptake remain unchanged or decrease at most sites for a generic crop and decrease at all sites for a generic deciduous tree, mainly as a result of the modelled reduction in stomatal conductance with rising atmospheric CO_2. Drier air and high soil moisture deficit as well as high temperatures are also important in southern Europe. However, if the CO_2 g_s response function is excluded from the analysis, which may be realistic for forest trees (Uddling et al., 2009 and references therein), the future stomatal O_3 flux is reduced only in southern Europe for the generic deciduous tree.

- The largest flux-based O_3 risk for a generic crop and a generic deciduous tree is found at the central European sites, in both the reference and projected future climate. This pattern differs substantially from the concentration-based AOT40 index, indicating largest O_3 risk in southern Europe.
- According to the current g_s model parameterisation, the temperature function limits the stomatal O_3 flux in northern Europe considerably, while the VPD and SWP functions are important mainly in southern Europe.
- This study demonstrates the importance to account for the climatic and atmospheric CO_2 influences on stomatal O_3 uptake for improved risk assessment in a changing atmosphere and climate.

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Supporting Information

Additional supporting information may be found in the online version of this article:

Appendix S1: $POD3_{crop}$ following the B2 scenario.
Appendix S2: $POD1.6_{tree}$ following the B2 scenario.

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