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Flux-based ozone risk assessment for adult beech forests

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Abstract Tropospheric ozone (O_3) is a critical threat to forest ecosystems. A stomatal flux-based risk evaluation methodology at the leaf level was established recently in the context of the Convention on Long-Range Transboundary Air Pollution (LRTAP). This study demonstrates improvement and validation of the stomatal flux-effect approach for adult beech with results from the 8-year free-air O_3 enrichment experiment at "Kranzberger Forst" (Germany). The risk assessment module of the SVAT model FO_3REST, being under development for local scale O_3-risk assessment of adult beech stands, was parameterized according to the LRTAP Convention's Mapping Manual. Mean maximum stomatal conductance for water vapour of 245 mmol H_2O m^{-2} PLA s^{-1}, as suggested in the LRTAP Convention's Mapping Manual for beech, was affirmed by assessment at "Kranzberger Forst", resulting in 162 mmol O_3 m^{-2} PLA s^{-1} upon recommended adjustment of the O_3/water vapour diffusivity ratio to 0.663. Based on this ratio, a provisional corrected flux-effect function was deduced. Modelled Phytotoxic O_3 Doses (POD_1) and potential O_3-caused losses in biomass formation estimated with a site-specific stomatal conductance algorithm differed slightly only from estimates by the original LRTAP parameterisation. Analysis-derived POD_1 target value within the meaning of Article 2 of the European Council Directive 2008/50/EC of 10 mmol O_3 m^{-2} corresponded to potential loss in biomass formation of about 10 % in ambient air relative to "pre-industrial" conditions. However, exceedance occurred by about a factor of two during the study period, indicating high risk at "Kranzberger Forst" under ambient air. Assessment for doubled O_3 exposure indicated potential underestimation even of the O_3 risk because modelled losses in biomass formation are in the lower range of the standard deviation of the observed ones.

Keywords Ozone · Beech · Risk assessment · Model validation · LRTAP Convention

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Introduction

Tropospheric ozone (O_3) poses a critical hazard and problem to present and future forest ecosystem services such as fibre and timber production or carbon storage capacity. A recent data synthesis, published by the ICP Vegetation Coordination Centre (http://icpvegetation.ceh.ac.uk; Hayes et al. 2007), suggested widespread occurrence of O_3 effects on vegetation at ambient concentrations in Europe over the period of 1990 through 2006. In the recent years, a stomatal flux-based risk evaluation methodology at leaf level was established in the context of the Convention on Long-Range Transboundary Air Pollution for crops, forest trees and grasslands (LRTAP Convention 2010; Mills et al. 2011). According to the Directive 2008/50/EC of the European Union (EU 2008), the assessment of the O_3-related risk at local scale for relevant O_3-sensitive biological receptors has to be based on parameters routinely measured by the air quality monitoring networks and, where necessary, on micrometeorological parameters routinely measured by the national weather services. Furthermore, the ICP Forests level II monitoring site concept requires a methodology for the evaluation of O_3-related risk at forest stand level.

The LRTAP Convention's stomatal flux parameterisation, described in the so-called Mapping Manual (LRTAP Convention 2010), is based on O_3 concentrations at canopy top, which are not measured conventionally by the air quality monitoring networks or at the ICP Forests level II sites. Therefore, the O_3 concentrations measured at a nearby monitoring station at a reference height above ground must be transformed to that at the top of the forest canopy by applying an appropriate deposition model (SVAT, i.e. for soil–vegetation–atmosphere transfer).

The free-air O_3 fumigation experiment at "Kranzberger Forst" (Freising, southern Germany), conducted over 8 years (2000–2007) on adult trees of European beech (*Fagus sylvatica*, in mixture with Norway spruce, *Picea abies*; Nunn et al. 2002; Werner and Fabian 2002; Matyssek et al. 2010) offers the unique opportunity to improve and validate the LRTAP Convention's parameterisation of stomatal O_3 uptake for sunlit beech leaves. Further validated is the stomatal O_3 flux-based response function which together with the stomatal uptake parameterisation is the risk assessment module of the SVAT model FO_3REST, which is under development for the evaluation of O_3-related risk for adult beech stands at local scale. This paper highlights the proposed O_3 risk assessment approach for beech stands, presenting the outcome of the validation study.

The "Kranzberger Forst" study site

The mixed *Fagus sylvatica/Picea abies* forest is located near Freising/Germany at 48°25′8″N and 11°39′41″E at 485 m a.s.l. in the ecological region 12.8 "Tertiäres Hügelland. Oberbayerisches Tertiärhügelland". The research plot had a rectangular shape of 50 m × 100 m. Long-term mean annual temperature and precipitation (1971–2000) were 7.8 °C and 786 mm, respectively (Deutscher Wetterdienst, DWD, station "Weihenstephan"). Beech trees were 66 ± 4 years old in 2007 and mean tree height increased from 24.5 m in 1999 to 25.5 m in 2007 (Pretzsch et al. 2010). Mean leaf area index was 5.4 $m^2\ m^{-2}$.

The free-air canopy O_3-fumigation system consisted of 150 Teflon tubes vertically suspended at 0.5 m distances across the foliated canopy and releasing O_3 through pressure-calibrated capillary outlets at 0.3 m intervals (cf. Karnosky et al. 2007; Werner and Fabian 2002; Nunn et al. 2002). A volume of 2,000 m^3 was experimentally exposed to $2{\times}O_3$ relative to the unchanged ambient, i.e. $1{\times}O_3$ regime at the site comprising five adjacent beech and spruce trees each. The O_3 exposure was restricted to $<$150 nl l^{-1} in the $2{\times}O_3$ treatment to prevent acute injury (Matyssek and Sandermann 2003). Due to the prevailing westerly winds, the fumigated trees were located upwind. Ambient O_3 concentrations were recorded at $z = 28$ m, while the elevated O_3 concentrations were measured at $z = 20$ m inside the crown at the transition between shaded and sunlit crown area. Air temperature and air humidity were measured at $z = 24$ m and horizontal wind velocity at $z = 36$ m. Soil water potential was modelled with BROOK90 (Hammel and Kennel 2001).

The LRTAP convention's stomatal flux approach

As a consequence of the discussion about the reasons of the so-called Neuartige Waldschäden (forest dieback) since the mid-1980s ground-level O_3 and its impact on human health and vegetation have increasingly come into focus within the UNECE (United Nations Economic Commission for Europe) and the European Union. In the 1990s exposure response functions, mainly derived from experimental studies in open-top chambers, were used to establish concentration-based critical levels for O_3. Paoletti and Manning (2007) mentioned the following main reasons why the current concentration-based standards are inadequate and did not work well: (1) inadequacy of the scientific background, (2) insufficient database for the derivation of the standards, (3) insufficient database to select the effect to be evaluated in the field, (4) inappropriate grouping into categories, and (5) no field validation.

One of the basic rules of toxicology is that dose–response relationships can only be established if the effective dose (flux) at the target site (e.g. membranes) or at least the absorbed dose (flux) of the stressor is known (Dämmgen et al. 1993; Grünhage and Jäger 1996; Dämmgen and Grünhage 1998; Musselman and Massman

1999; Massman et al. 2000; Musselman et al. 2006; Matyssek et al. 2007, 2008). Intensive research over the last 10 years has led to significant developments in the methods for estimation of O_3 uptake by plants, i.e. the flux of O_3 from the atmosphere through the stomata (F_{st}; [nmol m^{-2} s^{-1}]; e.g. Wieser et al. 2003; Matyssek et al. 2004, 2008; Nunn et al. 2007). It has been shown that the cumulative O_3 uptake above a constant threshold flux of Y nmol m^{-2} PLA s^{-1} accumulated over a stated time period during daylight hours (global radiation >50 W m^{-2}; POD_Y, Phytotoxic Ozone Dose; [mmol m^{-2} PLA][1])

$$POD_Y = \sum_{i=1}^{n} [\max(F_{st} - Y, 0) \cdot \Delta t]_i$$

provides stronger relationships with effects than external concentration-based exposure indices. POD_Y is calculated from hourly values of F_{st}, n denotes the number of hours to be included in the calculation period and $\Delta t = 1$ h (LRTAP Convention 2010). The statistically derived constant flux threshold Y is interpreted as a provisional estimate of a detoxification threshold, below which it is assumed that any O_3 molecule absorbed by the plant will be detoxified in the apoplast before reaching a target site (e.g. membranes). To reflect this detoxification, for beech, Y is set to 1 nmol O_3 m^{-2} PLA s^{-1} due to expert judgement; flux–response relationships were strongest when either no or a small threshold above which flux was accumulated were applied (Mills et al. 2011). The definition of *COU* (cumulative O_3 uptake) as used in the papers of the "Kranzberger Forst" experiment (e.g. Wieser et al. 2003, 2012) is identical with the definition of POD_Y, if Y is set to zero.

The stomatal O_3 uptake–response function for beech is based on the assumption that the O_3 fumigation concentrations measured at canopy top in the respective exposure systems (free-air, open-top chambers, field chambers; see below) reflect the O_3 concentrations at the upper surface boundary of the leaves' laminar layer. Additionally, it is assumed that under ambient conditions the O_3 concentration at the top of the canopy is a reasonable estimate of the O_3 concentration at the upper surface boundary of the laminar layer of the sunlit upper canopy leaves, unless the roughness sublayer near the canopy is taken into account (LRTAP Convention 2010). Considering both assumptions, stomatal O_3 uptake of the sunlit upper canopy leaves can be approximated by

$$F_{\text{sunlit leaf, stom, }O_3} = c_{O_3}(z_h) \cdot g_{\text{sunlit leaf, stom, }O_3} \cdot \frac{R_{\text{sunlit leaf, total, }O_3}}{R_{\text{sunlit leaf, laminar layer, }O_3} + R_{\text{sunlit leaf, total, }O_3}}$$

with $F_{\text{sunlit leaf, stom, }O_3}$ the stomatal O_3 uptake by the sunlit upper canopy beech leaves [nmol m^{-2} s^{-1}], c_{O_3} (z_h) the O_3 concentration at canopy top h [nmol m^{-3}], $g_{\text{sunlit leaf, stom, }O_3}$ the stomatal conductance for O_3 [m s^{-1}], $R_{\text{sunlit leaf, total, }O_3}$ the total leaf resistance for O_3 [s m^{-1}] and $R_{\text{sunlit leaf, laminar layer, }O_3}$ the resistance of the sunlit beech leaf laminar layer for O_3 [s m^{-1}]. The total leaf resistance is calculated according to

$$R_{\text{sunlit leaf, total, }O_3} = \frac{1}{g_{\text{sunlit leaf, stom, }O_3} + g_{\text{sunlit leaf, external leaf surface, }O_3}}$$

where $g_{\text{sunlit leaf, external leaf surface, }O_3}$ is the conductance of the external leaf surface for O_3 [m s^{-1}] which currently is set constant to 0.0004 m s^{-1} according to LRTAP Convention (2010). The influence of the beech leaf laminar boundary layer on stomatal O_3 gas exchange is taken into account applying the parameterisation of McNaughton and van den Hurk (1995):

$$R_{\text{sunlit leaf, laminar layer, }O_3} = 1.3 \cdot 150 \cdot \sqrt{\frac{L_{\text{leaf}}}{u(z_h)}}$$

with the characteristic crosswind leaf dimension $L_{\text{leaf, beech}}$ = 0.07 m and $u(z_h)$ the horizontal wind velocity at canopy height h [m s^{-1}]. 150 as a constant exhibits the dimension $s^{0.5}$ m^{-1}, while the factor 1.3 accounts for the differences in diffusivity between sensible heat and ozone as given in Massman (1998, 1999).

Stomatal O_3 uptake is parameterized for a sunlit upper canopy beech leaf according to a multiplicative Jarvis-Stewart approach (Jarvis 1976; Stewart 1988):

$$g_{\text{sunlit leaf, stom, }O_3} = g_{\text{sunlit leaf, stom, max, }O_3} \times \left[\min(f_{\text{phen}}, f_{O_3})\right] \times f_{\text{light}} \times \max\left\{f_{\min}, (f_{\text{temp}} \times f_{\text{VPD}} \times f_{\text{SM}})\right\}$$

where $g_{\text{sunlit leaf, stom, max, }O_3}$ represents the maximum level of the stomatal conductance for O_3 $g_{\text{sunlit leaf, stom, }O_3}$ per unit projected leaf area [mmol O_3 m^{-2} PLA s^{-1}], i.e. the stomatal conductance under optimal environmental conditions, and f_i weighting factors expressed in relative terms. The weighting factors f_i take values between 0 and 1 as a proportion of $g_{\text{sunlit leaf, stom, max, }O_3}$ and incorporate the effects of plant phenology (f_{phen}), ozone load (f_{O_3}), radiation (f_{light}), air temperature (f_{temp}), water vapour pressure deficit of the air surrounding the leaves (f_{VPD}) and soil moisture (f_{SM}) on $g_{\text{sunlit leaf, stom, max, }O_3}$. $f_{\min}$ is based on the analysis of published data and set to 0.13 for beech in Continental Central Europe (LRTAP Convention 2010). Currently, no data are available to deduce a

[1] LRTAP Convention (2010): "The projected leaf area (PLA, m^2) is the total area of the sides of the leaves that are projected towards the sun. PLA is in contrast to the total leaf area, which considers both sides of the leaves. For horizontal leaves the total leaf area is simply 2*PLA."

parameterization for f_{O_3}, so this factor is set to unity. For beech specific data, sets of parameters for f_i were deduced for the three European climate regions: Atlantic Central Europe, Continental Central Europe and Mediterranean Coastal/Continental locations (LRTAP Convention 2010). Here, the values for Continental Central Europe are cited.

The influence of radiation on stomatal behaviour is given by:

$$f_{light} = 1 - e^{-light_a \times PPFD}$$

with $light_a = 0.006$ and $PPFD$ the photosynthetic photon flux density [μmol m^{-2} s^{-1}]. If $PPFD$ is not measured, it can be estimated from global radiation as described in Appendix L in Grünhage and Haenel (2008). f_{temp} is given by:

$$f_{temp} = \max\left\{f_{min}, \left(\frac{t_{air} - t_{min}}{t_{opt} - t_{min}}\right)\left(\frac{t_{max} - t_{air}}{t_{max} - t_{opt}}\right)^{\frac{t_{max} - t_{opt}}{t_{opt} - t_{min}}}\right\}$$

if $t_{min} < t_{air} < t_{max}$ and $f_{temp} = f_{min}$ if $t_{air} < t_{min}$ or $t_{air} > t_{max}$ with $t_{min} = 5$ °C, $t_{opt} = 16$ °C and $t_{max} = 33$ °C.

The stomatal response to VPD is considered by:

$$f_{VPD} = \min\left\{1, \max\left(f_{min}, \left[(1 - f_{min}) \times \frac{VPD_{min} - VPD}{VPD_{min} - VPD_{max}}\right] + f_{min}\right)\right\}$$

with $VPD_{min} = 3.1$ kPa and $VPD_{max} = 1.0$ kPa.

The function used to describe f_{SM} is similar to f_{VPD}:

$$f_{SM} = \min\left\{1, \max\left(f_{min}, \left[(1 - f_{min}) \times \frac{SWP_{min} - SWP}{SWP_{min} - SWP_{max}}\right] + f_{min}\right)\right\}$$

with SWP the soil water potential [MPa], $SWP_{min} = -1.25$ MPa and $SWP_{max} = -0.05$ MPa. The values for SWP_{min} and SWP_{max} are deduced from measurements at "Kranzberger Forst" for the first 20 cm soil layer (cf. Nunn et al. 2005).

In principle, the phenology function can be based on either an effective temperature sum accumulation or a fixed number of days, which is the option used for forest trees by LRTAP Convention (2010). For beech "the start of the growing season (SGS), which is defined as the date of budburst/leaf emergence is estimated using a simple latitude model where SGS occurs at year day 105 at latitude 50°N, SGS will alter by 1.5 days per degree latitude earlier on moving south and later on moving north. The end of the growing season (EGS), which is defined as the onset of dormancy, is estimated as occurring at year day 297 at latitude 50°N, EGS will alter by 2 days per degree latitude earlier on moving north and later on moving south. Leaf discolouration is assumed to occur 20 days prior to dormancy and is assumed to be the point at which f_{phen} will start to decrease from g_{max}. Between the onset of dormancy and leaf fall g_{sto} will be assumed to be zero. The effect of altitude on phenology is incorporated by assuming a later SGS and earlier EGS by 10 days for every 1,000 m a.s.l." (LRTAP Convention 2010). According to these definitions, f_{phen} is zero for days of year (DOY) before DOY_{SGS} and after DOY_{EGS}. For the time period of 20 days ($=f_{phen1}$) after DOY_{SGS} to 20 days ($= f_{phen2}$) before DOY_{EGS}, the phenology function takes 1. For yeardays between DOY_{SGS} and $DOY_{SGS} + f_{phen1}$, the influence of phenology on stomatal behaviour is approximated by

$$f_{phen} = \frac{DOY - DOY_{SGS}}{f_{phen1}}$$

and for yeardays between $DOY_{EGS} - f_{phen2}$ and DOY_{EGS} by:

$$f_{phen} = (1 - f_{phen3}) \cdot \frac{DOY_{EGS} - DOY}{f_{phen2}} + f_{phen3}$$

with $f_{phen3} = 0.4$.

As described above, stomatal O_3 uptake estimations depend on $g_{sunlit\ leaf,\ stom,\ max,\ O3}$ which cannot be measured directly. Generally, stomatal conductance for O_3 is related to that of water vapour or carbon dioxide by the ratio of the respective molecular diffusivities D:

$$g_{leaf,\,stom,\,O_3} = g_{leaf,\,stom,\,H_2O} \cdot \frac{D_{O_3}}{D_{H_2O}} \quad \text{or}$$

$$g_{leaf,\,stom,\,O_3} = g_{leaf,\,stom,\,CO_2} \cdot \frac{D_{O_3}}{D_{CO_2}}.$$

According to the values cited in LRTAP Convention (2010) for beech, the mean maximum stomatal conductance for water vapour is 245 mmol H_2O m^{-2} PLA s^{-1}, a value identical with the maximum stomatal water vapour conductance for beech leaves of the sun crown in "Kranzberger Forst" as assessed by Nunn et al. (2005). Since the molecular diffusivity of O_3 in air has never been measured, it must be derived from known molecular diffusivities of another gas or its characteristic properties. The diffusivity ratio $D_{O_3}/D_{H_2O} = 0.613$ often used by the flux/effect modelling community is based on the application of Graham's law of diffusion (cf. Mason and Kronstadt 1967). Massman (1998) stated in his review of molecular diffusivities that the derivation of D_{O_3} applying Grahams's law "is in opposition to all theoretical results". He recommends a molecular diffusivity for O_3 in air of 0.1444 cm^2 s^{-1} at standard temperature and pressure (273.15 K, 1013.25 hPa). Taking into account, the recommended molecular diffusivity for water vapour in air of 0.2178 cm^2 s^{-1} at standard temperature and pressure, the diffusivity ratio D_{O_3}/D_{H_2O} becomes 0.663 (cf. Grünhage et al. 2012). The application of this more sounded diffusivity ratio leads to a mean maximum stomatal O_3 conductance of 162 mmol O_3 PLA m^{-2} s^{-1}. While in Sect. 3.4.3 of LRTAP Convention (2010), the diffusivity ratio of 0.663 is described as the conversion

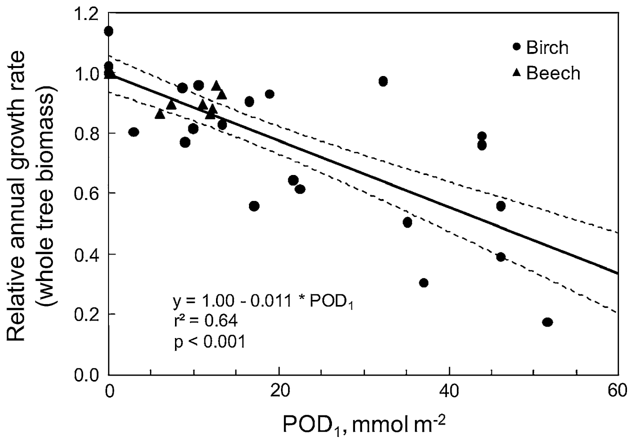

Fig. 1 Relationship between relative annual growth rates (whole-tree biomass) and *POD_1* for sunlit leaves of beech and birch (*POD_1* = Phytotoxic Ozone Dose [mmol m^{-2} PLA]) above a constant threshold flux of 1 nmol O_3 m^{-2} PLA s^{-1} accumulated over the growing season during daylight hours; *PLA* = projected leaf area; source: LRTAP Convention 2010)

factor for $g_{\text{sunlit leaf, stom, max, H}_2\text{O}} \rightarrow g_{\text{sunlit leaf, stom, max, O}_3}$, for beech the previously used ratio of 0.613 (see above) was applied in the LRTAP Convention (2010) forest sections which corresponds to a maximum stomatal O_3 conductance of 150 mmol O_3 PLA m^{-2} s^{-1} (cf. sections 3.6.2.3, A2.2 in LRTAP Convention 2010).

For the evaluation of O_3-related risk for reduction in annual growth rates (whole-tree biomass) of deciduous trees, a combined response function was derived from experiments with young beech and birch trees of up to 10 years of age. As illustrated in Fig. 1, this response function is dominated by results of experiments with birch (for experimental data set see Uddling et al. 2004). Because the results of CF/NF open-top chambers, experiments with seedlings of beech in Switzerland (cf. Braun and Flückiger 1995) fit quite well to this birch function, it was decided to derive a combined beech/birch function (Fig. 1):

$$\text{Relative total biomass (loss in annual growth rate)} = 1.00 - 0.011 \times POD_1.$$

While the accumulated stomatal O_3 fluxes are based on the diffusivity ratio of 0.613, a provisional corrected function considering the more sounded ratio of 0.663 will be used here:

$$\text{Relative total biomass (loss in annual growth rate)} = 1.00 - 0.0102 \times POD_1.$$

It must be emphasised (i) that the described stomatal O_3 flux approach should be viewed as an indication of potential risk for whole-tree biomass losses due to O_3 in the respective climate region (LRTAP Convention 2010) and (ii) that the estimated stomatal O_3 fluxes are not interpreted as the real, actual ones; the estimation of "real" losses requires site-specific parameterisations.

As mentioned before, O_3 concentrations are not measured at canopy height by the European air quality monitoring networks and the ICP Forest level II monitoring stations, the O_3 concentrations measured at a reference height (e.g. 3.5 m above ground) must be transformed to those at canopy top. Such a conversion has to be performed with an appropriate deposition model. While for short vegetation types such as crops, a downscaling is needed (cf. Grünhage et al. 2011), for forests an upscaling procedure is necessary. For the O_3-related risk evaluation for adult forests e.g. beech stands at local scale the Soil–Vegetation–Atmosphere-Transfer (SVAT) model FO_3REST is under development. The FO_3REST upscaling methodology and local scale risk assessment approach can be split in four steps: (1) upscaling of all the input parameters needed for the O_3 flux deposition module (e.g. air temperature, air humidity, wind velocity and O_3 concentration) from near ground measurement height (e.g. 3.5 m) to e.g. 50 m, (2) modelling total O_3 flux and calculation of O_3 concentration at assessment height taking into account the roughness sublayer near the forest canopy top, (3) calculation of sunlit-leaf stomatal uptake and Phytotoxic Ozone Dose (*POD_1*), and (4) risk evaluation. In this context, sap flow measurements are the appropriate tool for the improvement and validation of deposition models (Wieser et al. 2003; Matyssek et al. 2008; Nunn et al. 2007, 2010).

Improvement and validation of the LRTAP convention's stomatal conductance model

Due to the experimental setup of the "Kranzberger Forst" experiment, we assume (i) that the fumigation concentration measured at $z = 20$ m reflects the O_3 concentration at upper surface boundary of the laminar layer of the sunlit leaves in treatment $2\times O_3$, and (ii) that the ambient O_3 concentration measured at $z = 28$ m provides a reasonable estimate of the O_3 concentration at the upper surface boundary of the laminar layer of the sunlit leaves in treatment $1\times O_3$. Since, we assume that the horizontal wind velocity measured at $z = 36$ m is more or less identical to the wind velocity at canopy top, the actual leaf surface boundary layer resistance will be underestimated, i.e. stomatal O_3 uptake will be slightly overestimated.

The risk assessment module of FO_3REST is parameterized as described in the previous section of this paper. As mentioned before, the Jarvis-Stewart factor for O_3 is set to unity. Evidence exists from the experiment at "Kranzberger Forst" that there is an influence of O_3 on stomatal behaviour (Löw et al. 2006; Kitao et al. 2009). The

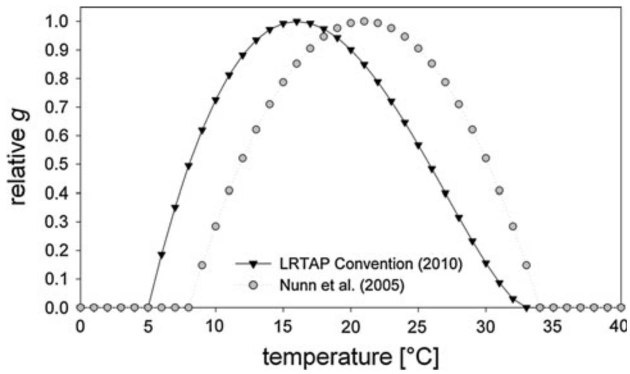

Fig. 2 Jarvis-Stewart function for temperature (f_{temp}) according to LRTAP Convention (2010) and Nunn et al. (2005)

assessments of stomatal conductance performed at "Kranzberger Forst" confirm the parameterisations of the Jarvis-Stewart functions for radiation, water vapour pressure deficit of the atmosphere and soil water potential. Modelled start ($DOY_{SGS} = 108$) and end ($DOY_{EGS} = 295$) of the stomatal O_3 flux accumulation period, i.e. the growing season, were compared with data for the mean date of incipient leaf unfolding ($DOY_{leaf\ unfolding} = 119 \pm 5$) and mean date of incipient leaf fall ($DOY_{leaf\ fall} = 297 \pm 6$) in 1991–2010, as observed at the phenological observation station "Weihenstephan" of the German Weather Service, which is located in the vicinity of the "Kranzberger Forst" study site. Taking into account, the small differences between modelled and observed data for start and end of the growing season, we recommend use of observed phenological data to calculate DOY_{SGS} and DOY_{EGS}. If leaf fall is not observed, DOY_{EGS} can be estimated from $DOY_{leaf\ colouring}$ ($DOY_{EGS} = DOY_{leaf\ colouring} + 20$).

The only site-specific parameterisation (cf. Nunn et al. 2005) which differs slightly from the above-mentioned ones is the weighting function for temperature with $t_{min} = 8$ °C, $t_{opt} = 21$ °C and $t_{max} = 34$ °C (Fig. 2).

The site-specific parameterisation of f_{temp} leads to POD_1 values which differ in the range of approx. −6.2 to 11.1 % in comparison to the ones calculated with f_{temp} according to LRTAP Convention (2010; Table 1). POD_1 values increase within the range of 8.2–11.0 %, if a $g_{sunlit\ leaf,\ stom,\ max,\ O_3}$ of 162 mmol O_3 m^{-2} s^{-1} is used. Due to the adapted flux-response relation, the potential losses in annual biomass growth rates differ only slightly (by 0.5–3.3 %).

During the hot and dry summer of 2003, there was a strong influence of soil moisture conditions on stomatal O_3 uptake (Table 1).

From the information given in LRTAP Convention (2010), it can be deduced that the estimated potential losses in biomass formation are expressed, in a strict sense of their meaning, relative to "pre-industrial" burden by O_3. The range of estimated potential loss in annual biomass growth rate at "Kranzberger Forst" is by 15.5–22.8 % under $1\times O_3$ and 28.8–38.5 % under $2\times O_3$, if $g_{sunlit\ leaf,\ stom,\ max,\ O_3} = 162$ mmol O_3 m^{-2} s^{-1} is used. The stomatal flux–response relationship was experimentally derived during the 1980s/1990s (cf. Braun and Flückiger

Table 1 Sensitivity of POD_1 estimations and potential loss in annual biomass growth rate to parameterisation of f_{temp} and $g_{sunlit\ leaf,\ stom,\ max,\ O_3}$

Year	$g_{sunlit\ leaf,\ stom,\ max,\ O_3} = 150$ mmol O_3 $m^{-2}s^{-1}$		$g_{sunlit\ leaf,\ stom,\ max,\ O_3} = 162$ mmol O_3 $m^{-2}s^{-1}$	
	LRTAP Convention (2010)	Nunn et al. (2005)	LRTAP Convention (2010)	Nunn et al. (2005)
Treatment "$1\times O_3$" POD_1 [mmol m^{-1}]/potential loss in annual biomass growth rate (%)				
2000	17.1/18.8	18.2/20.0	18.8/19.1	19.9/20.3
2001	18.8/20.6	18.5/20.3	20.6/21.0	20.3/20.7
2002	19.3/21.3	20.5/22.5	21.2/21.6	22.4/22.8
2003	13.7/15.0	15.2/16.7	15.2/15.5	16.7/17.1
2004	16.0/17.6	16.4/18.1	17.6/18.0	18.0/18.3
2005	13.8/15.2	13.8/15.1	15.3/15.6	15.2/15.5
2006	14.7/16.2	15.5/17.0	16.2/16.5	17.0/17.3
2007	16.5/18.2	15.6/17.2	18.1/18.5	17.2/17.5
Treatment "$2\times O_3$" POD_1 [mmol m^{-1}]/potential loss in annual biomass growth rate (%)				
2000	33.1/36.5	34.9/38.4	36.0/36.7	37.8/38.5
2001	32.0/35.1	31.7/34.8	34.8/35.5	34.4/35.1
2002	33.1/36.4	34.8/38.3	36.0/36.7	37.7/38.5
2003	23.6/26.0	25.8/28.4	25.9/26.4	28.2/28.8
2004	33.3/36.6	34.3/37.8	36.1/36.8	37.1/37.9
2005	32.1/35.3	31.6/34.7	34.8/36.5	34.3/35.0
2006	33.2/36.6	34.5/37.9	36.1/36.8	37.4/38.1
2007	29.9/32.9	28.0/30.8	32.5/33.1	30.5/31.1

1995; Uddling et al. 2004). As mentioned before, the beech data set was derived from open-top chamber experiments with unfiltered and filtered air. From the response function (cf. Fig. 1) and the observed losses in biomass formation for juvenile beech in unfiltered air (cf. position of triangles in Fig. 1), it can be concluded that the O_3 burden during the 1980s/1990s leads to an approx. 10 % reduction in annual whole plant biomass growth rates in ambient air relative to a "pre-industrial" situation. As shown in Table 1, the O_3 burden during the "Kranzberger Forst" experiment in $1\times O_3$ leads to an additional potential annual loss in biomass formation of about 5–13 % in comparison to the 1980s/1990s.

Validation of the LRTAP Convention's dose–response function

As described in detail in Pretzsch et al. (2010), the exposure of beech trees during 8 years to double ambient O_3 ($2\times O_3$) induced a shift in resource allocation into beech height growth at the expense of diameter growth, which leads to a neiloidal stem shape in comparison to ambient O_3 exposure ($1\times O_3$). Based on measurements of diameter increment at breast height, a statistically not significant mean annual loss in diameter increment at breast height by 11.5 % was observed under doubled ambient O_3 exposure relative to $1\times O_3$ (Fig. 3a). Therefore, validation studies performed as epidemiological studies under ambient air based on increment measurements at breast height seem to be questionable. Taking into account, the effect of O_3 on stem shape, the 8-year exposure of beech trees to $2\times O_3$ caused, on average, a decrease of 10.2 $m^3\ ha^{-1}$ in annual volume growth, i.e. a decrease by 43.5 % in relation to the annual growth rate occurring under ambient O_3 (Fig. 3b). The year-to-year variation in whole-stem productivity of beech under $2\times O_3$ exposure is illustrated in Fig. 3b. While the O_3 effect on stem productivity was significant, the year-to-year variation in losses in biomass formation was not.

Potential losses in biomass formation due to O_3—modelled as described before—for treatment $2\times O_3$ relative to the modelled potential losses under $1\times O_3$ (i.e. the differences of the modelled losses between the two treatments) are compared in Fig. 4 with the actually observed losses under $2\times O_3$ (i.e. differences between observed whole-stem productivity under $1\times O_3$ and $2\times O_3$). Since the year-to-year variation in observed losses in biomass formation are not significant, the modelled ones can be interpreted as losses in the lower range of the standard deviation of the observed ones. Thus, the LRTAP Convention's risk assessment for beech based on potential stomatal O_3 uptake of sunlit leaves must be seen as conservative, indicating (i) beech forests to be at risk and (ii) that risk may be underestimated.

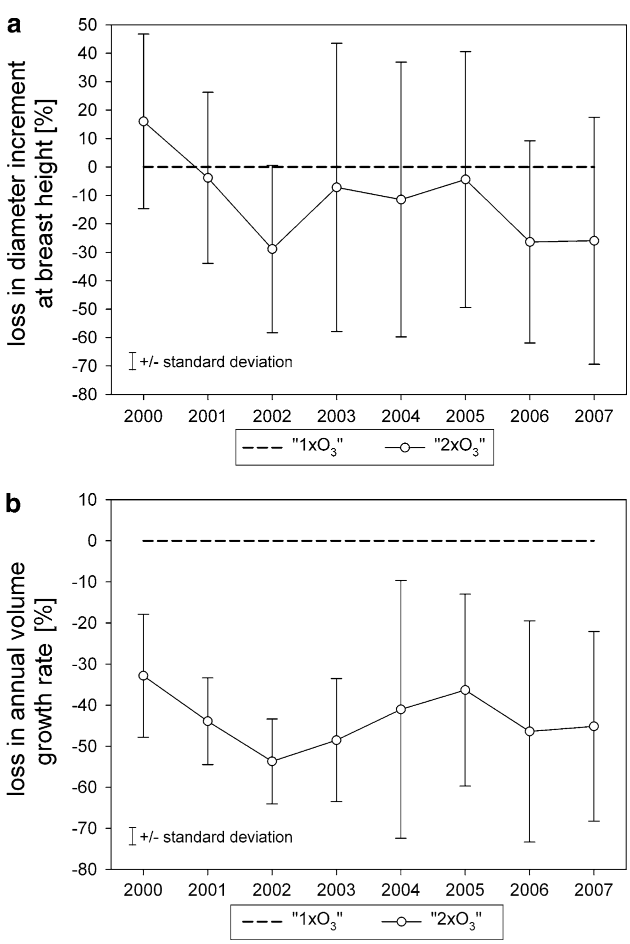

Fig. 3 Year-to-year deviation in **a** loss of diameter increment at breast height and in **b** whole-stem productivity, i.e. loss in annual volume growth rate, under doubled O_3 exposure ($2\times O_3$) relative to diameter increment and stem productivity under ambient O_3 (=0 %)

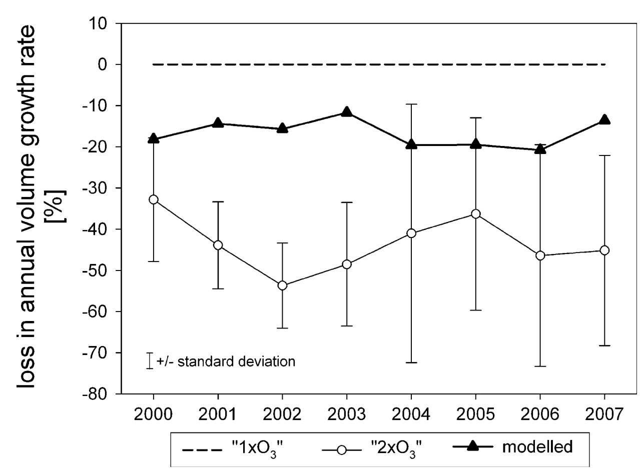

Fig. 4 Modelled and observed year-to-year deviation in loss of whole-stem productivity, i.e. loss in annual volume growth rate under doubled O_3 exposure ($2\times O_3$) relative to stem productivity under ambient O_3 (=0 %)

Conclusions and future perspectives

The LRTAP Convention's risk assessment approach for beech described in this paper is based on fumigation experiments with young trees of up to 10 years, while the "Kranzberger Forst" free-air O_3 fumigation study was performed with adult beech trees. Phytotoxic O_3 Doses (POD_1) and potential losses in biomass formation due to O_3 modelled with a site-specific stomatal conductance algorithm differ slightly only from the estimates derived with the original LRTAP Convention's parameterisation. While (i) the potential losses in biomass formation can be interpreted as relative to the "pre-industrial" O_3 burden, with (ii) the stomatal flux–effect relationship being experimentally derived during the 1980s/1990s along with observed productivity losses in the range of 10 % in unfiltered air (i.e. ambient O_3 exposure), it seems to be adequate to define a POD_1 target value within the meaning of Article 2 of the European Council Directive 2008/50/EC (EU 2008). Such target values are defined with the aim of at least reducing the harmful effects of air pollution and should be met at a specific date decided in Europe by the European Parliament and the Council of the European Union. We recommend a POD_1 target value of 10 mmol m^{-2} which corresponds to a potential productivity loss of approx. 10 % in ambient air relative to a "pre-industrial" situation. This target value can be interpreted as the upper margin of the O_3 burden before and during the 1980s/1990s. During the time of the "Kranzberger Forst" free-air O_3 enrichment study, this target value was extended by up to a factor of more than two under $1 \times O_3$ (i.e. ambient air; cf. Table 1). Obviously, the Kranzberger forest is at high risk. The estimates of potential losses in biomass formation due to doubling of the O_3 exposure indicate that $2 \times O_3$ even increases the risk and that this risk may be underestimated because estimated productivity losses are in the lower range of the standard deviation of the observed ones.

Next steps of our analysis will be the scaling of stomatal O_3 uptake between leaf, tree and stand level. Sap flow-based tree level stomatal O_3 uptake and conductance estimates from the "Kranzberger Forst" experiment will be used as validation parameters for the proposed scaling approach.

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