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Are Bavarian Forests (southern Germany) at risk from ground-level ozone? Assessment using exposure and flux based ozone indices

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Exposure- and flux-based ozone indices suggest Bavarian forests to be at risk from ozone; the flux-based index offers a means of incorporating stand-specific and ecological variables that influence risk.

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A B S T R A C T

Exposure and flux-based indices of O_3 risk were compared, at 19 forest locations across Bavaria in southern Germany from 2002 to 2005; leaf symptoms on mature beech trees found at these locations were also examined for O_3 injury. O_3 flux modelling was performed using continuously recorded O_3 concentrations in combination with meteorological and soil moisture data collected from Level II forest sites. O_3 measurements at nearby rural open-field sites proved appropriate as surrogates in cases where O_3 data were lacking at forest sites (with altitude-dependent average differences of about 10% between O_3 concentrations). Operational thresholds of biomass loss for both O_3 indices were exceeded at the majority of the forest locations, suggesting similar risk under long-term average climate conditions. However, exposure-based indices estimated higher O_3 risk during dry years as compared to the flux-based approach. In comparison, minor O_3-like leaf injury symptoms were detected only at a few of the forest sites investigated. Relationships between flux-based risk thresholds and tree response need to be established for mature forest stands for validation of predicted growth reductions under the prevailing O_3 regimes.

1. Introduction

Throughout recent decades, surface ozone (O_3) has been considered as an air pollutant capable of causing substantial injury to vegetation (Sandermann et al., 1997; Krupa, 1998). High O_3 concentrations are common during growing seasons in forests of Europe and North America (Skärby et al., 1998; Emberson et al., 2000a; Coyle et al., 2003; Bytnerowicz et al., 2004; Vingarzan, 2004). Negative O_3 impacts on forest trees have been reported in numerous studies (Chappelka et al., 1997; Skärby et al., 1998; Matyssek and Innes, 1999; Skelly et al., 1999). Effects can vary with tree species and cultivars (Baumgarten et al., 2000; VanderHeyden et al., 2001; Matyssek et al., 2004; Wipfler et al., 2005; Nunn et al., 2006), region and growth conditions (Vollenweider et al., 2003a,b; Wieser et al., 2003a,b), stand structure and tree age (Matyssek et al., 2004; Herbinger et al., 2005; Nunn et al., 2005b), as O_3 can affect tree performance (Oksanen, 2001) directly or indirectly in combination with other stress factors (Percy et al., 2002; Bahnweg et al., 2005; Karnosky et al., 2005; Nunn et al., 2005a). Foliar response and growth reduction of forest trees have been reported in several studies (Chappelka and Samuelson, 1998; Braun et al., 1999; Dittmar et al., 2003; 2005; Karnosky et al., 2005; Vollenweider et al., 2003; Nunn et al., 2005a; Weinstein et al., 2005; Wipfler et al., 2005). However, the relation between O_3 concentration or dose and the extent of injury is not well understood. For real forest stands, acute or chronic O_3 effects are insufficiently investigated, and empirically based O_3 index thresholds are not well validated for mature forest stands.

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Different methods have been developed to assess the O_3 impact on forest trees. Currently, concentration-based indices quantifying plant exposure to O_3 above threshold levels over defined time periods are commonly used in Europe for risk assessment of forests and to inform protection policies. The exposure-based concept of the "Maximum Permissible Ozone Concentration" (MPOC) is one such concentration based O_3 index, application of which is mostly limited to Germany. This index is based on a descending ranking of hourly O_3 concentrations, aggregated for different time periods and classified into three risk categories (Grünhage et al., 2001; VDI, 2002; Krause et al., 2003). The latter were derived from O_3 effects reported in the literature and represent worst-case scenarios. The "Critical Level for Ozone" concept was introduced by the UNECE in the 1990s and resulted in the derivation of the AOT40 approach (accumulated hourly O_3 concentration over a threshold of 40 nl O_3 l^{-1}; Fuhrer, 1994; Skärby and Karlsson, 1996). This approach assumes the higher O_3 exposures to be strongly coupled with O_3 uptake, and aims to predict O_3 risk for vegetation by protecting the most sensitive species. A Critical Level AOT40 threshold (currently 5 µl O_3 l^{-1} h) was implemented for forest trees, exceedance of this threshold would indicate the potential for growth reductions of greater than 5% per growing season (Karlsson et al., 2004). More recently, O_3 flux-based concepts that are based on leaf O_3 uptake through the stomata have been favoured (Fuhrer and Achermann, 1999; Emberson et al., 2000a; Karlsson et al., 2003; 2004; Massman, 2004; Matyssek et al., 2004, 2007a, 2008; Uddling et al., 2004; Musselman et al., 2006; Nunn et al., 2007), as it is acknowledged that uptake rather than exposure drives injury. O_3 uptake, as the time integral of diffusive O_3 influx, is considered to represent a more physiologically relevant, i.e. internal O_3 dose (COU = cumulative O_3 uptake) O_3 index as compared to exposure based indices (i.e. time integrals of O_3 concentration in ambient air) as a kind of "external O_3 dose" (Musselman et al., 2006). A provisional flux threshold was established at $AF_{st>1.6} = 4$ mmol O_3 m^{-2} PLA (accumulated stomatal O_3 flux above a threshold of 1.6 nmol O_3 m^{-2} s^{-1} PLA (projected leaf area)) above which a 5% biomass loss is expected to occur in forest trees (Karlsson et al., 2004).

The assessment of leaf symptoms indicative of O_3 injury is believed to yield a practicable diagnosis of O_3 impact on forest tree species (Skelly et al., 1999; Günthardt-Goerg et al., 2000; Vander-Heyden et al., 2001; Novak et al., 2003; Smith et al., 2003; Vollenweider et al., 2003; Davis and Orendovici, 2006).O_3 can induce distinct foliar symptoms although genotypic differences and environmental interactions (e.g. radiation, humidity and precipitation) can have a significant influence in injury formation (Skelly et al., 1999; Günthardt-Goerg et al., 2000; Innes et al., 2001; Vander-Heyden et al., 2001; Manning et al., 2002; Bussotti et al., 2003; Matyssek and Sandermann, 2003; Novak et al., 2003; Vollenweider et al., 2003; Dalstein and Vas, 2004; Schaub et al., 2005; Karnosky et al., 2007; Kubisce et al., 2007).

The main objective of this study is to evaluate and compare the risk O_3 poses on Bavarian forests (southern Germany) using the different indices described above. Estimations of these O_3 indices ideally require data describing canopy height hourly O_3 concentrations. In Europe, O_3 is routinely monitored in urban and suburban regions, but information about the O_3 pollution regimes in rural, forest locations are, by comparison, rather rare (see also De Leeuw et al., 2001). At such sites O_3 monitoring is mostly limited to passive sampling which only provides weekly or monthly mean O_3 concentrations. As such, estimation of O_3 indices, which requires high resolution air quality data, could only be performed using continuously monitored O_3 data that were available from rural "open field sites" close to the forest locations. In addition, O_3 flux modelling was performed using meteorological and soil related data monitored from the nearest "Level II sites".

Risk assessments using exposure-based O_3 indices were conducted for 19 sites distributed over the most important forest regions in Bavaria for the period of 2002 through 2005, equivalent flux-based risk assessments were conducted at thirteen of these sites where necessary data were available, for the years 2002 and 2003. In addition, macroscopic leaf symptoms were assessed at selected sites and related to the O_3 indices. This study provides a unique O_3 risk assessment for forest trees at a regional level and a comparison of O_3 risk using a variety of O_3 indices. A critical comparison is made of the performance of these different risk assessment approaches.

2. Materials and methods

2.1. Concept, site characterisation, and input data

For this study data from 32 measuring sites (finally representing 19 investigation sites) were utilised. At these sites O_3 concentrations were recorded either by active continuous monitoring of O_3 concentration (AM), or using O_3 passive sampler which provided monthly integrated O_3 exposure values (PM). At a limited number of sites both methods of O_3 monitoring were performed. Sites also recorded one or more of the following: high resolution meteorological (MET) and soil/water related data (SW), phenological incidences (PHE), and O_3 leaf symptom assessment (OS). A summary of site characteristics, measurement details, responsible institutions, and geographical locations are provided in Table 1 and Fig. 1. The sites can be classified into the following three categories:

- "open field sites" ($n = 17$)—rural sites where background O_3 concentration is routinely measured continuously in 10 min to hourly intervals, (mostly comprising plots established for national air quality control), (AM, MET)
- "forest research sites" ($n = 2$)—intensively investigated forest sites with continuous O_3 recording above the forest canopy and O_3 passive sampling, with a variety of additional experimental instrumentation (Nunn et al., 2002; Beudert, 2005; Dieffenbach-Fries and Beudert, 2007; Matyssek et al. 2007b), (AM, PM, MET, SW, PHE, OS)
- "Level II sites" ($n = 13$)—Bavarian forest ecosystem monitoring sites which conform to a specific and continuous monitoring program me (BayLWF, 2002, 2008), (PM, MET, SW, PHE, OS)

This site selection was made for several reasons:

- To ensure that the sites are distributed across Bavaria encompassing the range of altitudes and climatic conditions of the important forested areas in this region.
- To compare the reliability of deriving O_3 indices using datasets of O_3 concentration from "open field" or "forest" plots which are of differing temporal and spatial monitoring refinement.
- To allow for the calculation of exposure based O_3 indices that required continuous O_3 data from "open field sites" and "forest research sites" available for 2002 through 2005 ($n = 19$).
- To allow for the calculation of the flux based O_3 index that required the identification of "plot pairs" for modelling providing the necessary combination of continuously recorded O_3 data from "open field sites" and "forest research sites", and meteorological, soil/water, and phenological data from the nearest suitable "Level II site" for the years 2002 and 2003 ($n = 13$).

2.2. Comparison of O_3 data at "open-field sites" and "forest sites"

Forest plots ("Level II sites", "forest research sites") with monthly integrated O_3 passive sampling (PM) were used to examine the comparability with O_3 data from active monitoring (AM) at the nearest available "open field sites" in a rural environment (Fig. 2). The resulting "plot pairs" (four in total: 1: *rot-AS*, 9: *fre-KF*, 18: *kre-HP*, 19: *ber-GW*) were distributed across Bavaria (Table 1), with each pair representing similar altitude and climate conditions. Within-pair comparisons (PM at forested "Level II site" vs. "open field site"/"forest research site" with AM at "open field") were conducted for the growing periods of 2002 to 2005 for each "plot pair".

2.3. Ozone measurements and calculation of ozone metrics

Continuous monitoring of O_3 (AM) was mostly carried out at 4 m aboveground at the "open field sites" exceptions include the sites of "Kranzberg Forest" (*KF*), "Hohenpeißenberg" (*HP*), and "Garmisch" (*GA*) where O_3 measurements were made at heights of 24 m, 20 m, and 15 m, respectively (Table 1). At "Forellenbach" (*FB*), recordings were conducted on a tower at 51 m height above a juvenile mixed

Table 1
Site and measuring characteristics of plots.

Plot -no.	Plot/ pair	Plot name	org	O_3	Position	alt	Situation	prec	tem	spec	stype	ssub	sbd	awc	cb	fb	met	mh	dis	a_dif
1	AS	Aschaffenburg	*LfU*	AM	9°7′7″E, 49°59′33″N	130	Open-field, suburban								x	x	a, r, w	4	50	−345
	rot	Rothenbuch	*LWF*	PM	9°28′E, 49°59′N	475	Forest clearing	945	7	oak/bee	be	Ls	f	198			t, h, p	4		
2	WB	Würzburg	*LfU*	AM	9°57′29″E, 49°48′22″N	230	Open-field, suburban								x	x	a, r, w	4	5	−100
	wue	Würzburg	*LWF*		9°53′E, 49°43′N	330	Forest clearing	651	8	oak	pb-pl	Tu	f-m	215			t, h, p	4		
3*	ER	Erlangen	*LfU*	AM	10°57′53″E, 49°36′25″N	284	Open-field, rural								x			4		
4	KU	Kulmbach	*LfU*		11°26′37″E, 50°6′15″N	306	Open-field, suburban								x			4		
5	NS	Neustadt	*LfU*	AM	11°46′45″E, 48°51′16″N	370	Open-field, slope, rural								x	x	a, r, w	4	100	−64
	rie	Riedenburg	*LWF*		11°46′E, 48°56′N	475	Forest clearing	656	7.5	oak	pb	Tu	f-m	243			t, h, p	4		
6	ME	Mehring	*LfU*	AM	12°10′58″E, 48°10′58″N	415	Open-field, basin, rural								x	x	a+r (*KF*), w	4	12	+9
	aoe	Altötting	*LWF*		12°45′E, 48°13′N	406	Forest clearing, plane	1001	7.5	spr	pb	Lt	f	197			t, h, p	4		
7	BR	Bad Reichenhall	*LfU*	AM	12°51′38″E, 47°43′28″N	465	Open-field, basin, suburban								x			4		
8	AR	Arzberg	*LfU*	AM	12°11′26″E, 50°3′34″N	480	Open-field, slope, suburban			spr					x			4		
9	KF	Kranzberger Forst	*TUM*	AM+PM	11°39′41″E, 45°25′08″N	485	Plane, rural, forest interior			bee	be/pb				x	x	a, r, w, t, h	24	2	−23
	fre	Freising	*LWF*	PM	11°39′E, 48°24′N	508	Forest clearing	826	7.5	bee	be/pb	Lu	f-m	215			p	4		
10	AB	Augsburg	*LfU*	AM	10°54′15″E, 48°19′38″N	500	Open-field, plane, suburban								x	x	a, r, w	4	25	−15
	zus	Zusmarshausen	*LWF*		10°32′E, 48°25′N	515	Forest clearing	782	7.5	spr	pg	Ut	f-m	220			t, h, p	4		
11	*RE*	*Regen*	*LfU*	AM	13°7′47″E, 48°58′25″N	536	Open-field, slope, urban								x	x	a, w (*TB*)	4	12	+140
	bod	Bodenwörth	*LWF*		12° 23′ E, 49° 17′ N	396	Forest clearing, plane	715	7.5	pin	be	St	m-c	159			t, h, p, r	4		
12	NA	Naila	*LfU*	AM	11°43′24″E, 50°19′28″N	540	Open-field, slope, suburban								x	x	a, r, w	4	50	−130
	rok	Rothenkirchen	*LWF*		11°21′E, 50°27′N	670	Forest clearing, plane	912	6	spr	be	Lt	f	150			t, h, p	4		
13	KE	Kempten	*LfU*	AM	10°18′28″E, 47°43′33″N	680	Open-field, valley, suburban								x			4		
14*	AN	Andechs	*LfU*	AM	11°13′18″E, 47°58′11″N	700	Open-field, slope, rural								x			4		
15	GA	Garmisch	*LfU*	AM	11°3′52″E, 47°28′40″N	735	Open-field, basin, rural			spr					x	x	a, r, w	15	50	−54
	sog	Schongau	*LWF*		10°48′E, 47°53′N	789	Forest clearing, plane	1253	6.5	bee	pb	Lt	f	188			t, h, p	4		
16	TB	Tiefenbach	*LfU*	AM	12°33′6″E, 49°26′22″N	750	Open-field, slope, rural								x	x	a, r, w	4	40	−90
	flo	Flossenbürg	*LWF*		12°24′E, 49°56′N	840	Forest clearing	820	6	spr	be-ps	Sl	f	223			t, h, p	4		
17	FB	Forellenbach	*UBA*	AM+PM	13°25′22″E, 48°56′54″N	807	Forest interior, valley			bee/spr	be				x	x	a, r, w, t, h	51	50	−218
	mit	Mitterfels	*LWF*		12°53′E, 48°59′N	1025	Forest clearing	1311	5.5	bee	be	Ls	f	226			p	4		
18	HP	Hohenpeißenberg	*DWD*	AM	11°0′38″E, 47°48′9″N	989	Open-field, hill, rural			spr					x	x	a, r, w	20	60	−86
	kre	Kreuth	*LWF*	PM	11°49′E, 47°44′N	1075	Forest clearing	1829	5	spr	be	Lt	c	205			t, h, p	4		
19*	GW	Garmisch/Wank	*LfU*	AM	11°8′37″E, 47°30′35″N	1776	Open-field, mountain, rural			spr					x	x	a, r, w	4	200	+301
	ber	Berchtesgaden	*LWF*	PM	12°57′E,47°35′N	1475	Forest clearing	1482	4	lar	re	Ls		218			t, h, p	4		

Plot no.: Plots are ranked and numbered according to rising altitude of sites with AM (active continuous (hourly) O_3 measurement); AM was conducted usually 2002–2005.
Plot no. with *: differing measuring period for AM. Plot no. 3: April 2004–December 2005; 14: April 2003–December 2005; 19: January 2002–March 2004.
Plots: upper case: open field background O_3 measuring stations “open field sites” or “forest research sites” (*KF, FB*); lower case: “Level II sites” (forest ecosystem monitoring stations).
Plot pairs: were used for O_3 flux modelling (conducted for 2002 and 2003) combining data sets with continuously recorded O_3, meteorological and phenological data from sites with AM (“open field” or “forest research sites”) and “Level II sites”.
org: responsible institution for meteorological and/or O_3 data (LfU: Bayerisches Landesamt für Umwelt, LWF: Bayerische Landesanstalt für Wald und Forstwirtschaft, TUM: Technische Universität München, UBA: Umweltbundesamt, DWD: Deutscher Wetterdienst).
O_3: type of O_3 measurement (AM: active continuous (hourly) O_3 measurement, PM: integrating (monthly) O_3 passive sampling).
alt, altitude (m a.s.l.); prec, precipitation (long term mean) (mm) (Hammel and Kennel, 2001); tem, air temperature (long term mean) (°C); spec, main tree species at the forested sites (bee: beech, spr: spruce, pin: pine, lar: larch); soilt, soil type (be: brownearth, pb: parabrownearth, pl: pelosol, pg: pseudogley, ps: podsol, re: rendzina); soils, soil substrate (Ls: sandy loam, Tu: silty clay, Lt: clay loam, Lu: silty loam, Ut: clay silt, St: clay sand, Sl: loamy sand); sbd, soil bulk density classified from mean layer-thickness-weighted data from organic layer—40 cm soil depth (f: fine, m: medium, c: coarse); awc, available water capacity (mm/soil depth), modelled with LWF BROOK90 (Federer et al., 2003; Hammel and Kennel, 2001) for 100 cm soil depth (range 60–140 mm m^{-1}: low awc, 140–220 mm m^{-1}: medium awc, 220–300 mm m^{-1}: high awc); cb, “concentration based”, calculations based on the external O_3 concentration (seasonal mean concentrations, SUM0, AOT40, MPOC); fb, “flux based”, calculations based on the modelling of the O_3 flux into the stomata (cumulative uptake of O_3 (AFst), accumulated O_3 flux into the stomata over threshold of 1.6 mmol O_3 m^{-2} PLA ($AF_{st>1.6}$)); met, continuous (hourly) meteorological measurements necessary for ozone flux modelling (a: air pressure, r: global radiation, w: wind speed, t: air temperature, h: relative humidity, p: precipitation, concerning plots in parentheses: *KF*: Kranzberg Forest, *TB*: Tiefenbach); mh, measuring height for O_3 and meteorological data (m) above ground; dis, distance between plot pairs (km); a_diff, altitude difference levels between plot pairs (m).

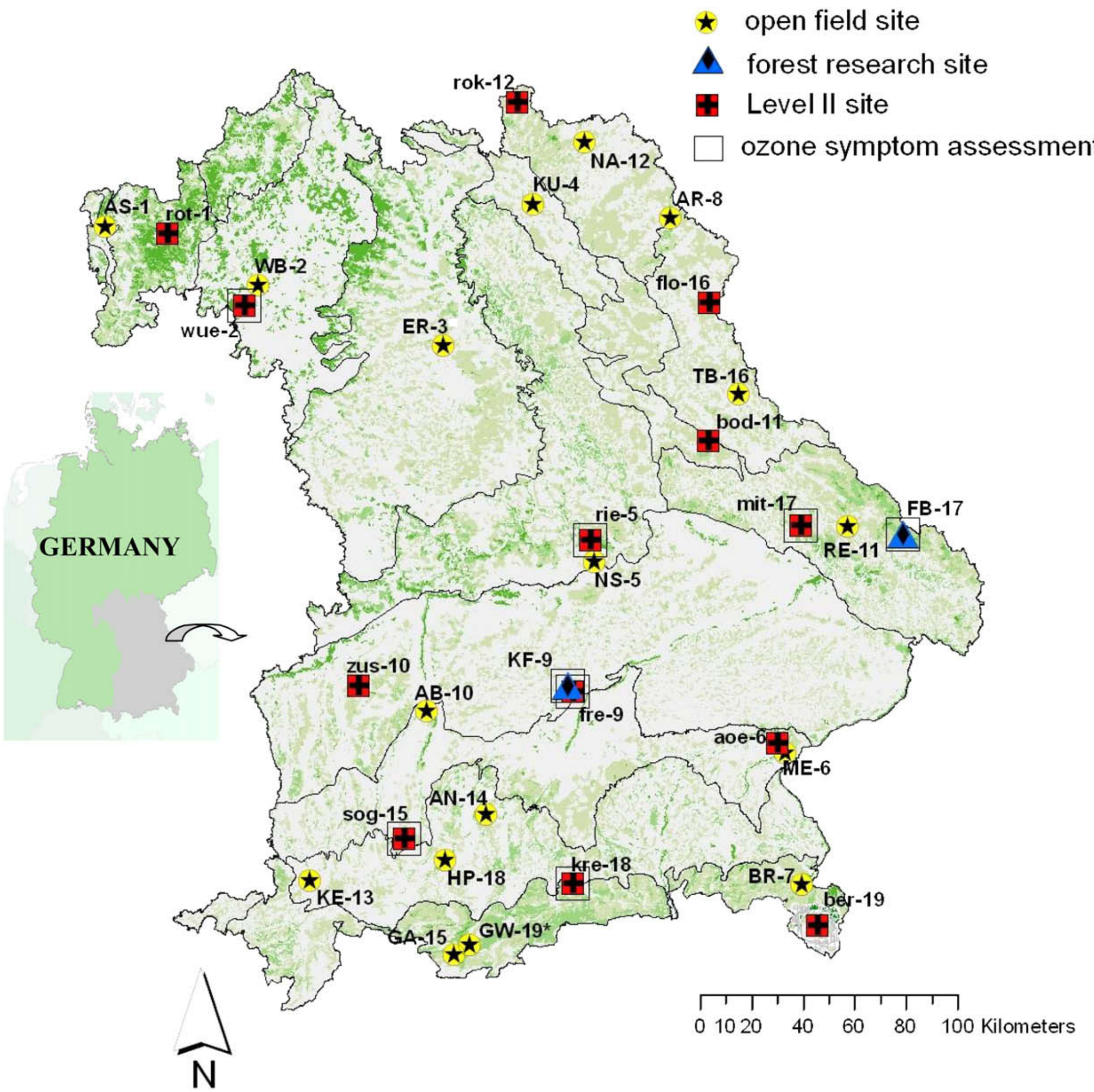


Fig. 1. Map of the investigation area: measuring programme, location, and type of forested area of the plots in Bavaria (Germany). Plot name: upper case: open field background O_3 measuring stations ($n = 17$: "open field sites", $n = 2$: "forest research sites"); lower case: $n = 13$: "Level II sites" (forest ecosystem monitoring stations). Measuring programme: *(red) quadrate with black cross*: "Level II site": O_3 passive sampling, meteorological measurements, soil characteristics; *(yellow) circle with black star*: "open field site": continuous O_3 monitoring at open field background O_3 measuring stations; *(blue) triangle with black rhombus*: "forest research site": continuous O_3 monitoring above forest canopy, O_3 passive sampling, meteorological measurements, soil characteristics; *framing square*: sites with assessment of ozone induced leaf injury symptoms; black lines: boundary of forest growth regions; shaded (green) fields: forested area. Plot abbreviations and numbers are used according to Table 1.

spruce-beech forest, which had originated from natural regeneration (Table 1). Passive samplers from the Swedish Environmental Research Institutes (IVL; recommended by ICP Forests, De Vries et al., 2003; Ferretti, 2004; UNECE, 2005a,b) were exposed at 4 m aboveground in forest clearings at the "Level II sites" and at 51 m height at the *FB* station, all for monthly intervals. At the *KF* site, passive samplers were exposed at canopy height (about 25 m aboveground) for 2-week intervals (Werner and Fabian, 2002).

O_3 data that were not collected at canopy height (i.e. data collected at 3–4 m above ground under "open field" conditions or 51 m above ground and hence above the canopy top (at the *FB* site)), were corrected for a reference canopy height of 25 m using a transfer function suggested by Krause et al. (2003) and recommendations of the UNECE Mapping Manual (UNECE, 2004a). Calculations of seasonal mean and maximum O_3 concentrations at the reference canopy height used AM and PM O_3 data collected from April–September from 2002 to 2005.

2.4. Calculation of ozone indices

SUM0 is defined as the daily sum of hourly O_3 concentrations from April to September (defined as the "growing season"), calculated for the years 2002 to 2005.

The **Maximum permissible O_3 concentrations** (MPOC) are based on hourly O_3 concentrations. Hourly mean O_3 concentrations occurring during the growing season are ranked in descending order; then maximum mean values are calculated for different time spans (e.g.: hourly maximum O_3 level, mean of the highest eight O_3 levels during 8 h). Usually the maximum O_3 means for 1 h, 8 h, 24 h, 7 days, 30 days, 90 days and the entire growing season are calculated (for details see the German Guideline VDI 2310 part 6, VDI, 2002). The maximum levels are compared with an O_3 exposure-response function based on current knowledge from peer-reviewed literature published before 2000. The 10% confidence interval of the function represents the range where compliance ensures protection with respect to growth, productivity, biodiversity and recreation. Below the confidence range, maximum protection-, and above, permanent injury of the plant (e.g. in photosynthesis, growth, reproduction; Grünhage et al., 2001; Krause et al., 2003) is likely to occur. MPOC indices were calculated for the growing seasons of 2002 to 2005.

The **AOT40** (accumulated hourly O_3 concentration over a threshold of 40 nl O_3 l^{-1}) was calculated as the sum of the differences between hourly mean O_3 concentrations at canopy height (see above) above a threshold of 40 nl O_3 l^{-1} h during daylight hours (hours with global radiation >50 W s^{-2}) throughout the growing seasons of 2002 to 2005 according to Fuhrer and Achermann (1994). The dates of exceedance of the

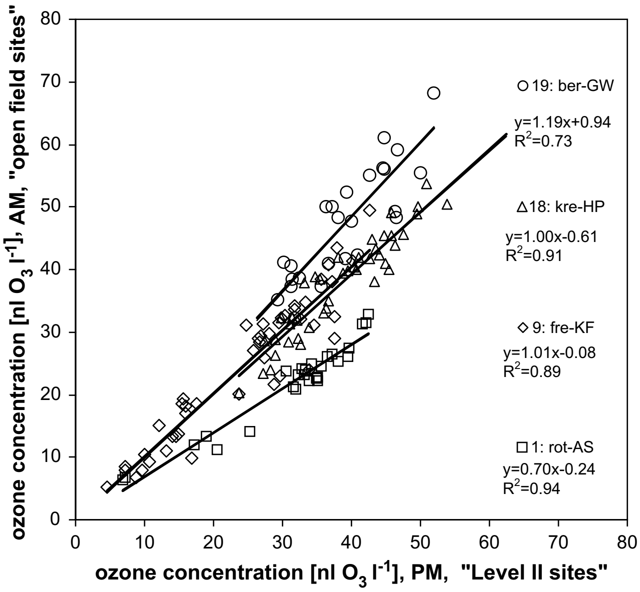


Fig. 2. Correlation of 4-week integrated O_3 concentrations (PM, O_3 passive sampling measurements) at forest ecosystem monitoring sites ("Level II sites") and the co-occurring, time congruent mean O_3 concentrations from hourly O_3 monitoring (AM, active continuous O_3 monitoring) for plot pairs in four altitude ranges (<500 m a.s.l.: plot pair no.1 (rot-AS), ∼500 m a.s.l.: 9 (fre-*KF*), ∼1000 m a.s.l.: 18 (kre-*HP*), >1000 m a.s.l.: 19 (ber-*GW*)) and altitude difference levels (<50 m: 9 (fre-*KF)* (+23 m), 50–100 m: 18 (kre-*HP)* (+86 m), ∼300 m: 19 (ber-*GW*) (−301 m), 1 (rot-AS) (+345 m)) for 2002–2005; measuring height at all plots 4 m above ground; for rot-*AS* data only from growing season (April–September); for plot pairs see Table 1.

current AOT40 Critical Level for trees of 5 µl O_3 l^{-1} h (Karlsson et al., 2004; UNECE, 2004a) and the former AOT40 Critical Level of 10 µl O_3 l^{-1} h (Ashmore and Davidson, 1996; UNECE, 1996; Fuhrer et al., 1997) were recorded. $AOT40_{phen}$ covers the effective site-specific growing season of a site and was calculated for the pairs of plots selected for each O_3 flux assessment in 2002 and 2003. Similar to MPOC, AOT40 represents an O_3 exposure-based concept of risk assessment.

Stomatal O_3 flux (Fst) was calculated on an hourly basis throughout the effective site-specific growing season to sun leaves of the upper canopy by multiplying the O_3 concentration at the leaf surface with the corresponding species-specific stomatal conductance for O_3 (g_{sto}). It is assumed that the intercellular O_3 concentration is close to zero (Laisk et al., 1989). g_{sto} was calculated using a multiplicative stomatal conductance model (as used within the DO3SE model, Emberson et al., 2000a, b, 2007) adapted from Jarvis (1976) as a function of species-specific maximum stomatal conductance (g_{max}, expressed on a projected leaf area (PLA) basis), phenology, and environmental conditions (photosynthetically effective photon flux density, air temperature, vapour pressure deficit and soil moisture deficit). Parameterisation of the model for beech was made according to Nunn et al. (2005b), implementing night-time stomatal conductance (Matyssek et al., 1995) and adjusting g_{max} for O_3 on the basis of diurnal gas exchange measurements from *KF.* Empirical coefficients describing the response of g_{sto} to soil moisture were derived from soil water potential measurements at *KF* (Table 2). Soil moisture was estimated using a simple water budget model further details of which are provided in Emberson et al. (2007), where the method was applied to estimate soil water potential and its influence on stomatal conductance and subsequent O_3 flux for key forest tree species across Europe. This model is based on the water balance model of Mintz and Walker (1993), which was developed and evaluated on consideration of important parameters (e.g. root-zone storage capacity, precipitation, air temperature and heat flux) that determine evapotranspiration at the regional and global scale. Information about type, substrate, texture, bulk density, depth and moisture status of soils were obtained for the selected plot pairs from the "Level II sites". Soil bulk density, soil matrix water potential and volumetric soil water content of different layers were measured and modelled at these sites with LWF- BROOK90 (Hammel and Kennel, 2001), a modification of Brook90 (Federer et al., 2003).

The flux (Fst) model was applied to selected pairs of plots (thirteen in total: 1: *rot-AS*, 2: *wue-WB*, 5: *rie-NS*, 6: *aoe-ME*, 9: *fre-KF*, 10: *zus-AB*, 11: *bod-RE*, 12: *rok-NA*, 15: *sog-GA*, 16: *flo-TB*, 17: *mit-FB*, 18: *kre-HP*, and 19: *ber-GW*; Table 1). Modelling was performed for the growing season of 2002, which represented a year with average meteorological conditions and O_3 pollution, and 2003, an extraordinarily warm and dry year (Ciais et al., 2005) with high O_3 concentrations and severe drought effects. Modelling was focused on beech (*Fagus sylvatica* L.), since this is both an ecologically and economically important climax tree species (Ellenberg, 1986), being increasingly propagated in Bavarian forestry (LWF (Bayerische Landesanstalt für Wald und Forstwirtschaft), 2003), and estimated to be the naturally dominant tree species for nearly all the study regions (Walentowski et al., 2004). The accumulated stomatal O_3 flux (AFst) was calculated as the hourly sum of stomatal O_3 flux through the stomata of sun-exposed leaves of the upper canopy, using methods described in the UNECE Mapping Manual (UNECE, 2004a). The Critical Flux (CF) for the seasonal accumulated O_3 flux ($AF_{st>1.6}$) was determined as the sum of the hourly stomatal O_3 flux above a threshold of 1.6 nmol O_3 m^{-2} PLA s^{-1} ($F_{st>1.6}$) during the site-specific growing season (Karlsson et al., 2004). The Critical Level $AF_{st>1.6}$ for deciduous and coniferous trees over one growing season was set provisionally to 4 mmol O_3 m^{-2} PLA (Karlsson et al., 2004; UNECE, 2004a). The date of exceedance of the Critical Level $AF_{st>1.6}$ during the growing season was recorded. In order to analyse the correlation of $AF_{st>1.6}$ vs. AOT40, $AOT40_{phen}$ (see above) was calculated for the effective growing season for each site.

2.5. *Site-specific environment data*

Monitoring of O_3 and meteorological factors (air temperature, relative humidity, precipitation, global radiation, wind speed, air pressure) was conducted continuously for 13 aggregated "plot-pairs" (Table 1: "open field sites" $n = 11$, and "forest research sites" $n = 2$ (*FB, KF)*). Assessments took into account altitude, distance between the "open field site" and the respective "forest site", forest growth region and climatic conditions. Air temperature, air humidity and precipitation data were obtained from the "Level II sites". However, global radiation and wind speed were taken from "open-field sites", as these "open-field sites" represent above canopy conditions far better than data measured at forest clearings (Mitscherlich, 1981). Air pressure data were available at the "open field sites" and the "forest research sites". Analysis was based on hourly means of O_3 concentration and meteorological factors, interpolating data gaps with regressions derived from data from the nearest available plot.

The phenology of beech (start of growing season: leaf emergence >50% of the foliation, end of growing season: autumnal leaf fall >50% of foliation) was observed directly at "Level II sites" or was estimated from air temperature data (assessed in clearings). Estimation made use of a thermal time model to define leaf emergence and beginning of growing season (Kramer, 1994), whereas autumnal leaf fall was defined to start on day 306 of the year (Kramer, 1995). The difference between the observed and estimated beginning of the growing season was always less than 7 days. Differences between the observed and the defined end of the growing season at day 306 were larger (data not shown), but without effect on O_3 flux modelling, because of decreasing photosynthetic activity and stomatal aperture at the end of the growing season. Direct observation could not detect premature leaf fall during the study period (data not shown).

2.6. *Assessment of O_3 induced leaf injury symptoms*

A manual for assessing macroscopic O_3-induced leaf injury was developed by the International Co-operative Programme on Assessment and Monitoring of Air Pollution Effects on Forests (ICP Forests) with the objective to estimate risk to European ecosystems from ambient O_3 concentrations. For our investigation eight sites were assessed, comprising six "Level II sites" and two "forest research sites". The assessment of macroscopic visible leaf injury at the "Level II sites" *wue*, *rie*, *mit*, *kre*, *fre* and *sog* was carried out annually from 2002 to 2005 in August on the sun exposed foliage in the upper third of the crown of five branches of five individuals of adult dominating beech trees according to the UNECE manual (UNECE, 2004b). The branches selected were as small as possible, but usually with all leaf age stages present. A representative number of leaves, usually between 20 and 40 leaves per branch, were examined under optimum light conditions and scored for occurrence of O_3 injury. Leaf injury was determined as the percentage of the leaves showing O_3 induced injury symptoms. Such symptoms were distinguished from other biotic or abiotic injury using a photo gallery (www.gva.es /ceam/ icp-forests), a flow chart for injury discrimination (Innes et al., 2001), microscopical differentiation (www.ozon.wsl.ch) and, if necessary, expertise diagnosis by the Ozone Validation Centre for Central Europe (WSL/FSL Birmensdorf/Switzerland). Assessment at the forest research site *FB* was carried out between the end of July and mid-August (five individual trees, two branches) as described above, but determination of O_3 induced leaf injury was assessed qualitatively. At *KF*, assessments on five sun exposed branches of five beech trees were conducted according to Hartmann et al. (1995) and Innes et al. (2001) by determining the percentage of symptomatic leaf area of total foliar area of assessed leaves at 2-week intervals from the end of May until leaf abscission (Nunn et al., 2002, 2005b). Injury occurred in sun-exposed leaves as chloroses and/or necroses in the intercostal fields between leaf veins.

3. Results

3.1. *Comparison of O_3 measurements at open-field and forested sites*

Both two-weekly and monthly O_3 averages from PM and AM showed methodological deviations of 5–10% from each other at the

Table 2
Details of the parameterisations used within the O_3 flux model simulations for Bavarian forest sites (DO_3SE) for deciduous species *(Fagus sylvatica* L.).

Parameter	Units	Parameterisation	
g_{max}	mmol O_3 m^{-2} (PLA) s^{-1}	148[a]	□
f_{min}	(fraction)	0.13	□
SGS[b]	year day	e.g. 110 (20 April)	▥
EGS[b]	year day	e.g. 273 (30 Sep)	▥
A_{start}[b]	year day	e.g. 110	▧
A_{end}[b]	year day	e.g. 273	▧
f_{phen_a}[b]	(fraction)	0.0	▧
f_{phen_b}[b]	(fraction)	0.0	▧
f_{phen_c}[b]	(fraction)	1.0	▧
f_{phen_d}[b]	(fraction)	0.0	▧
f_{phen_e}[b]	days	15	▧
f_{phen_f}[b]	days	20	▧
light_a	(co-efficient)	0.006	□
T_{min}	°C	0	▤
T_{opt}	°C	21	□
T_{max}	°C	35	▤
VPD_{max}	kPa	1.0	▤
VPD_{min}	kPa	3.25	▤
SWP_{max}	MPa	−0.05	□
SWP_{min}	MPa	−1.25	□
root depth	m	1.2	■
Y	nmol m^{-2} PLA s^{-1}	1.6	□
LAI_{min}	m^2 m^{-2}	0	■
LAI_{max}	m^2 m^{-2}	4	■
LAI_s	m^2 m^{-2}	30	■
LAI_e	m^2 m^{-2}	30	■
h	m	25	■
L	m	0.05	■

g_{max}, species-specific maximum stomatal conductance; f_{min}, minimum daytime stomatal conductance; SGS, day of the year at the start of the growing season; EGS, day of the year at the end of the growing season; f_{phen}, function for variation in stomatal conductance with leaf/needle age (subscripts a–f listed below); light_a, species-specific parameter co-efficient; $SWP_{max/min}$, maximum/minimum soil water potential; $VPD_{max/min}$, maximum/minimum water vapour pressure deficit; $T_{min/opt/max}$, minimum/optimum/maximum leaf temperature at which stomatal opening occurs; root-depth, species and soil texture related average rooting depth; *Y*, ozone stomatal flux rate threshold; $LAI_{min/max/s/e}$, minimum/maximum leaf area index; *h*, average canopy height; *L*, cross-wind leaf dimension for broadleaved trees.

Shading in column 4 as follows. □ no change to Nunn et al., 2005b parameterisation. ▤ parameterisation change to Nunn et al., 2005b because of new scientific knowledge, values taken from "generic" deciduous forest parameterisation based on *Fagus sylvatica* grown in central Europe (Emberson et al., 2007). ▥ new parameterisation based on site-specific observations from Bavarian sites. ▧ new parameters due to updated model code, i.e. parameters not used/listed in Nunn et al., 2005a,b and therefore taken from "generic" deciduous forest parameterisation based on *Fagus sylvatica* grown in central Europe (Emberson et al., 2007). ■ new parameterisation due to updated model code, i.e. parameters not used/listed in Nunn et al., 2005b and therefore taken from site-specific observations from Forellenbach site (please note that the findings from the Forellenbach (17: FB) site (var. LAI parameters, root depth and canopy height) will be used for all Bavarian sites).

[a] Value for sun crown, cf. Nunn et al., 2005b.

[b] It should be noted that the structure of the f_{phen} function has changed from that provided previously in the Mapping Manual (UNECE, 2004a) to allow for the "Mediterranean evergreen" f_{phen} profile. As such, the f_{phen} parameters for the generic species relate to the f_{phen} functions described below whilst the "Mapping Manual (UNECE, 2004a)" f_{phen} parameters are for use with functions detailed in the Mapping Manual (UNECE, 2004a). The new f_{phen} functions described here are consistent with the incorporation of the A_{start} and A_{end} terms used in the Mapping Manual (UNECE, 2004a) for crops:
$f_{phen} = 0$ when SGS $\geq$ dd $\geq$ EGS (SGS: start growing season, EGS: end growing season, dd: year day); $f_{phen} = f_{phen\_a}$ when SGS $<$ dd $< A_{start}$;$f_{phen} = f_{phen\_b} + (f_{phen\_c} - f_{phen\_b})$ AST! $(dd - A_{start})/f_{phen\_e}$ when $A_{start} \leq dd < (A_{start} + f_{phen\_e})$; $f_{phen} = f_{phen\_c}$ when $(A_{start} + f_{phen\_e}) \leq dd < (A_{end} - f_{phen\_f})$; $f_{phen} = (f_{phen\_d} + (f_{phen\_c} - f_{phen\_d})$ AST! $(A_{end} - dd)/f_{phen\_f}$ when $(A_{end} - f_{phen\_f}) \leq dd < A_{end}$;$f_{phen} = f_{phen\_d}$ when $A_{end} \leq dd \leq$ EGS.

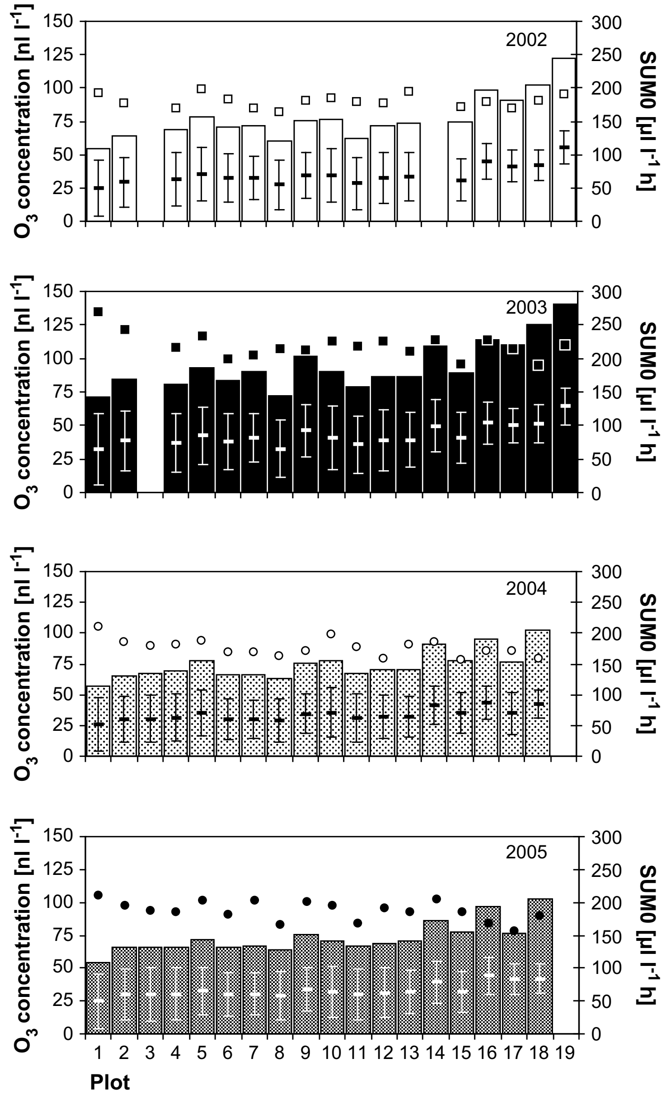


Fig. 3. Seasonal mean O_3 concentrations with standard deviation (bars), maximum O_3 concentrations (symbols) and the cumulative O_3 exposure SUM0 (columns, second ordinate) at canopy height on all plots with AM (active continuous O_3 monitoring; "forest research sites", "open field sites") during the growing season (April–September) for 2002–2005.

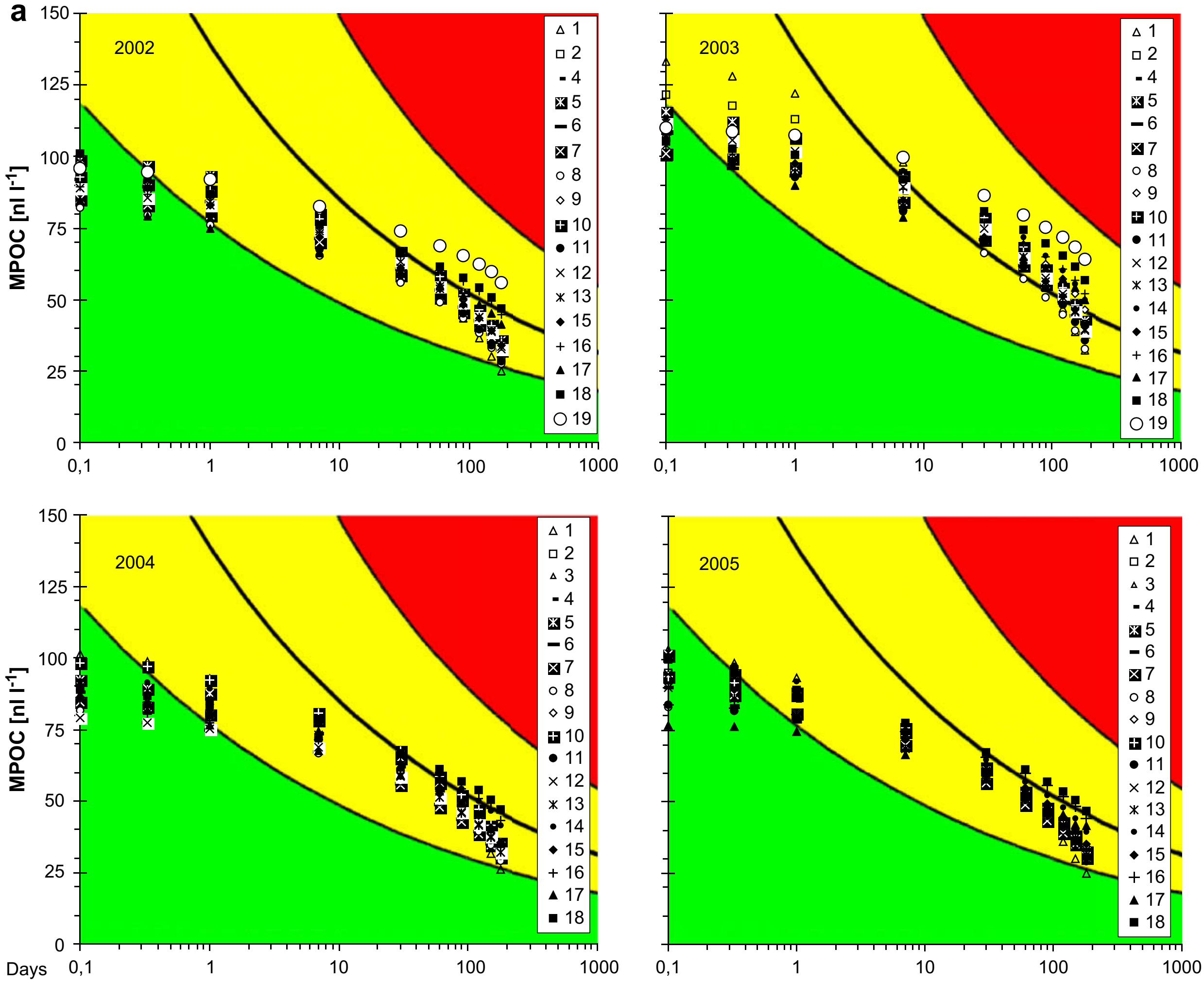
same site (data not shown). Differences between O_3 measured by AM at "open field sites" and PM at "Level II sites" mainly depended on the altitude, whereas distances between plots were less important (Fig. 2, Table 1). For example, the two plots of pair 9 (*fre-KF*), situated close (2 km) to each other and only differing in altitude by about 20 m, hardly differed in O_3 concentration. Similarly, the two plots of pair 18 (*kre-HP*), differing in altitude only by 86 m, but in distance by approximately 60 km, nonetheless resembled each other in O_3 concentration. In contrast, larger altitudinal differences between sites resulted in an overestimation of 19% for Berchtesgaden (*ber*, 1475 m a.s.l) when using surrogate data from substantially higher elevations (*GW*, 1776 m a.s.l.), while an underestimation of ca. 30% was observed for Rothenbuch (*rot*, 475 m a.s.l.) when using data from Aschaffenburg (*AS*, 130 m a.s.l). The variation of 30% can be assumed as the maximum in this region, as Rothenbuch (*rot*) represents the higher and Aschaffenburg (*AS*) the lower elevation in this area. Across the 13 plots selected for flux modelling, the *rot-AS* comparison also demarcated the maximum variation in this analysis, as differences in altitude between the other plot pairs were much smaller (Table 1).

3.2. O_3 exposure

3.2.1. Seasonal means, hourly maximum O_3 concentrations, and SUM0

Seasonal mean and hourly maximum O_3 concentrations as well as SUM0 were highest at all plots investigated in 2003 as compared to the other years (Fig. 3), with a seasonal mean across the sites of 44±8 nl O_3 l^{-1} and SUM0 of 190±36 µl O_3 l^{-1} h. Conversely, these two indices only slightly differed across the sites in the humid years of 2002, 2004 and 2005, ranging between 35 and 36 nl O_3 l^{-1}, and 150 and 155 µl O_3 l^{-1} h, respectively. Seasonal mean O_3 concentrations were

a



b

Exposure (Apr.–Sep.)	MPOC [nl $O_3 l^{-1}$] at the plant surface		
	compliance ensures maximum possible protection	compliance ensures substantial protection	exceedance leads to permanent damage
8 h	92	164	295
24 h	74	130	229
7 days	50	86	148
30 days	37	63	108
90 days	29	50	86
Apr. - Sep.	25	43	74

Fig. 4. (a) MPOC (maximum permissible O_3 concentrations) at the top of a forest canopy at all plots with AM (active continuous O_3 monitoring; "forest research sites", "open field sites") during different time spans in the growing season (April–September) for 2002–2005. (b) MPOC values to protect European conifer and deciduous tree species according to Grünhage et al., 2001; VDI 2310 part 6, 2002.

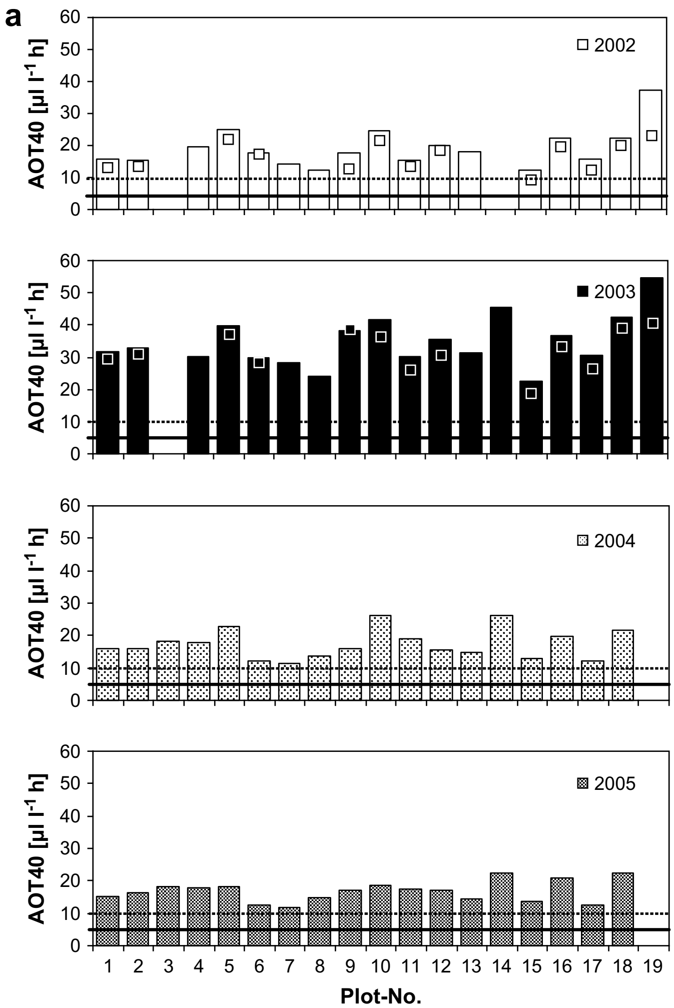


Fig. 5. (a) AOT40 (accumulated hourly ozone concentration over a threshold of 40 nl O_3 l^{-1}) at canopy height on all plots with AM (active continuous O_3 monitoring; "forest research sites", "open field sites") for the growing season (April–September, in 2005 for Plot 17 (*FB*) May–September) for 2002–2005. Solid line: current AOT40-Critical Level of 5 µl O_3 l^{-1} h. Dotted line: former AOT40-Critical Level of 10 µl O_3 l^{-1} h. (b) Date of exceedance of AOT40-Critical Level for forests for all plots with AM (active continuous O_3 monitoring; "forest research sites", "open field sites") during the growing season for 2002–2005 (April–September, in 2005 for Plot 17 (*FB*) May–September). Current AOT40 critical level >5 µl O_3 l^{-1} h, former AOT40 Critical Level >10 µl O_3 l^{-1} h.

highest at the most elevated plot 19 (GW) at 1776 m a.s.l. and at plot 18 (HP) at 989 m a.s.l.. In 2003, the seasonal mean O_3 concentration of plot 19 was 64 nl O_3 l^{-1}, with maximum hourly concentrations of more than 90 nl O_3 l^{-1} frequently occurring in August. Concentrations were also high at plots between 750 and 1000 m a.s.l (plot 16 (TB), 17 (FB), 18 (HP)).

3.2.2. MPOC

MPOC indices, calculated across the plots for time spans between 1 week and the whole growing season (April–September) for the years 2002 to 2005, fell into in the "yellow risk category", where moderate O_3 susceptibility of forest trees is indicated, with a low probability of direct O_3 effects (Fig. 4a,b). Hence, minor O_3 effects on growth may occur in this category, which nonetheless does not exclude O_3-induced macroscopic leaf injury across the study sites and years (Fig. 4). Highest MPOC values were determined in 2003, indicating a tendency towards the "red risk category" (reflecting permanent damage to forest trees). Highest MPOC levels occurred at the high-altitudinal plot 19 (*GW*, 1776 m a.s.l.), followed by the plots situated above 700 m a.s.l. (plot no. 14–18). MPOC values also tended to increase with increasing altitude of the study sites during the humid years. Lowest MPOC levels were found at the low-elevation plot 1 (*AS*, 130 m a.s.l.) throughout all study years.

3.2.3. AOT 40

The current and the former Critical Levels (CL) of AOT40 at 5 and 10 µl O_3 l^{-1} h were exceeded across the plots and study years (Fig. 5a). The current CL was exceeded on average by a factor of 2 to 4 in 2002, 2004 and 2005, but by 4–10-fold in 2003, when AOT40 reached or exceeded 30 µl O_3 l^{-1} h regardless of site location. In 2003, AOT40 was even greater at high altitudes, reaching more than 50 µl O_3 l^{-1} h at plot 19 (*GW*, 1776 m a.s.l.), about 40 µl O_3 l^{-1} h at plot 18 (*HP*, 989 m a.s.l.) and more than 45 µl O_3 l^{-1} h at plot 14 (*AN*, 700 m a.s.l.). In 2002, AOT40 was highest at plot 19 (37 µl O_3 l^{-1} h), whereas the levels at the other plots ranged between 13 and 25 µl O_3 l^{-1} h. In 2004 and 2005, AOT40 values ranged from 11 to 26 µl O_3 l^{-1} h (no measurement at plot 19). The current CL of 5 µl O_3 l^{-1} h was exceeded already at the beginning of the growing season (in general, by the end of April through mid-May) across the plots and study years (Fig. 5b). In 2003, the CL was exceeded rather early (end of April) at the high-altitudinal plot 19 (*GW*, 1776 m a.s.l.). The former CL of 10 µl O_3 l^{-1} h was almost always exceeded by June and July, with the exception of 2003 at plot 19 where exceedance occurred as early as May.

In addition, $AOT40_{phen}$ of the effective growing season of 2002 and 2003 was calculated for the 13 plots which were used for O_3 flux assessment (Fig. 6a,b). Although being on average lower by 20%, $AOT40_{phen}$ resembled AOT40 at each plot. Deviations increased with altitude. The date of CL exceedances (Fig. 6a, 5b) of $AOT40_{phen}$ and AOT40 was similar at low-altitude plots, whereas discrepancy increased towards high elevation. Regarding the plot pair of Garmisch/Wank-Berchtesgaden (19: *GW-ber*), the CL of AOT40 was exceeded, for example, in 2002 on April 22, whereas the CL of $AOT40_{phen}$ was exceeded on June 30, due to the late beginning of the growing season on June 11.

3.3. Cumulative stomatal O_3 uptake

Consistent with the CL of $AOT40_{phen}$ (see above), the CL of $AF_{st>1.6} = 4$ mmol O_3 m^{-2} for forest trees was exceeded across the plots in 2002 and 2003 (Fig. 6b). The percentage of CL exceedance of $AF_{st>1.6}$ and AOT40 were similar in 2002, but in 2003 the exceedance of the CL of AOT40 was clearly increased compared to the CL $AF_{st>1.6}$ (Fig. 6b).

In 2003, $AF_{st>1.6}$ was nearly unchanged or lower than in 2002, irrespective of the plot (Fig. 6b), even though seasonal mean O_3 concentrations were on average 20% higher in 2003 (Fig. 3). The correlation between the mean seasonal O_3 concentration and plot altitude was high both in 2002 and 2003, whereas in the case of $AF_{st>1.6}$ the correlation with altitude was poor in 2003 and absent in 2002 (Fig. 7a,b).

The exceedance of the CL of $AF_{st>1.6}$ was high across all plots, on average by about 300% in 2002 and 2003. Conversely, in the case of $AOT40_{phen}$ CL was exceeded on average by about 250% in 2002 and more than 500% in 2003 (Fig. 6 b). CL exceedances of $AOT40_{phen}$ and $AF_{st>1.6}$ occurred, in general, approximately 1 month after the beginning of the growing season, and about 10 days earlier at the lower compared to the higher altitude plots (Fig. 6a). The CL of $AF_{st>1.6}$ was usually exceeded at about the same date or at least within 14 days after the CL of $AOT40_{phen}$ had been reached.

Tentatively, the CL of $AF_{st>1.6}$ was doubled to 8 mmol O_3 m^{-2} for reasons of comparison with the former CL of AOT40 of 10 µl O_3 l^{-1} h (Fig. 6b). In 2003, the higher CL of $AF_{st>1.6}$ led only to slight exceedances, if any, rather late in the growing season (end of June through end of July) at the plots 1, 2, 5, 11, 12, 15, whereas the former CL of AOT40 was still exceeded by some margin at all plots (on average by about 200%).

Contrasting with $AOT40_{phen}$, plots could be differentiated in terms of $AF_{st>1.6}$ into two "O_3 uptake response groups" during the dry year of 2003 (Fig. 8):

(1) enhanced O_3 uptake during the growing season in 2003 compared to 2002 at plot 6, 9, 10, 15, 17, 18 and 19;
(2) reduced O_3 uptake during the growing season in 2003 compared to 2002 at plot 2, 5, 11, 12, 16.

Regarding the relationship between $AOT40_{phen}$ and $AF_{st>1.6}$ for each individual plot (Fig. 8), an approximately linear correlation prevailed during the humid year 2002 which represented the climatic long-term average. A similar relationship existed in 2003 by early July, whereas thereafter, $AOT40_{phen}$ shifted towards high levels by early August under the prolonged, extraordinarily sunny and warm summer conditions (Ciais et al., 2005), while cumulative O_3 uptake stagnated upon stomatal closure because of the developing drought (group 2 plots, see above). Conversely, an unchanged, more or less linear relationship persisted at group 1 plots (Fig. 8). The precipitation sum (Fig. 9) during the entire growing season was clearly responsible for the two response groups of plots: decreased O_3 uptake in 2003 was associated with low precipitation (~200 mm during the growing season), and increased uptake in 2002 with moderate or high precipitation (>300 mm). The two "O_3 uptake response groups" are consistent with the increased CL exceedances in 2003 (see above): group 2 plots displayed only slight exceedance (if any) of the increased $AF_{st>1.6}$ of 8 mmol O_3 m^{-2} (Fig. 6b).

3.4. O_3 induced leaf injury symptoms

O_3 induced leaf injury symptoms were found on beech trees throughout the study years, although not across all study plots and always only to a small extent (<3% of total leaf area, < 1–5% of leaves, respectively, Table 3). At the "Level II sites" wue, rie, mit, and kre macroscopic O_3 induced leaf injury was absent during all years, while at fre and sog O_3 injury was found in 2005, although to a limited extent. At KF, leaf injury symptoms were also low and occurred in June in 2002, 2004 and 2005. Under the drought conditions in 2003 the injury extend at KF was nearly negligible. At FB O_3 injury could be detected in 2003 and 2004.

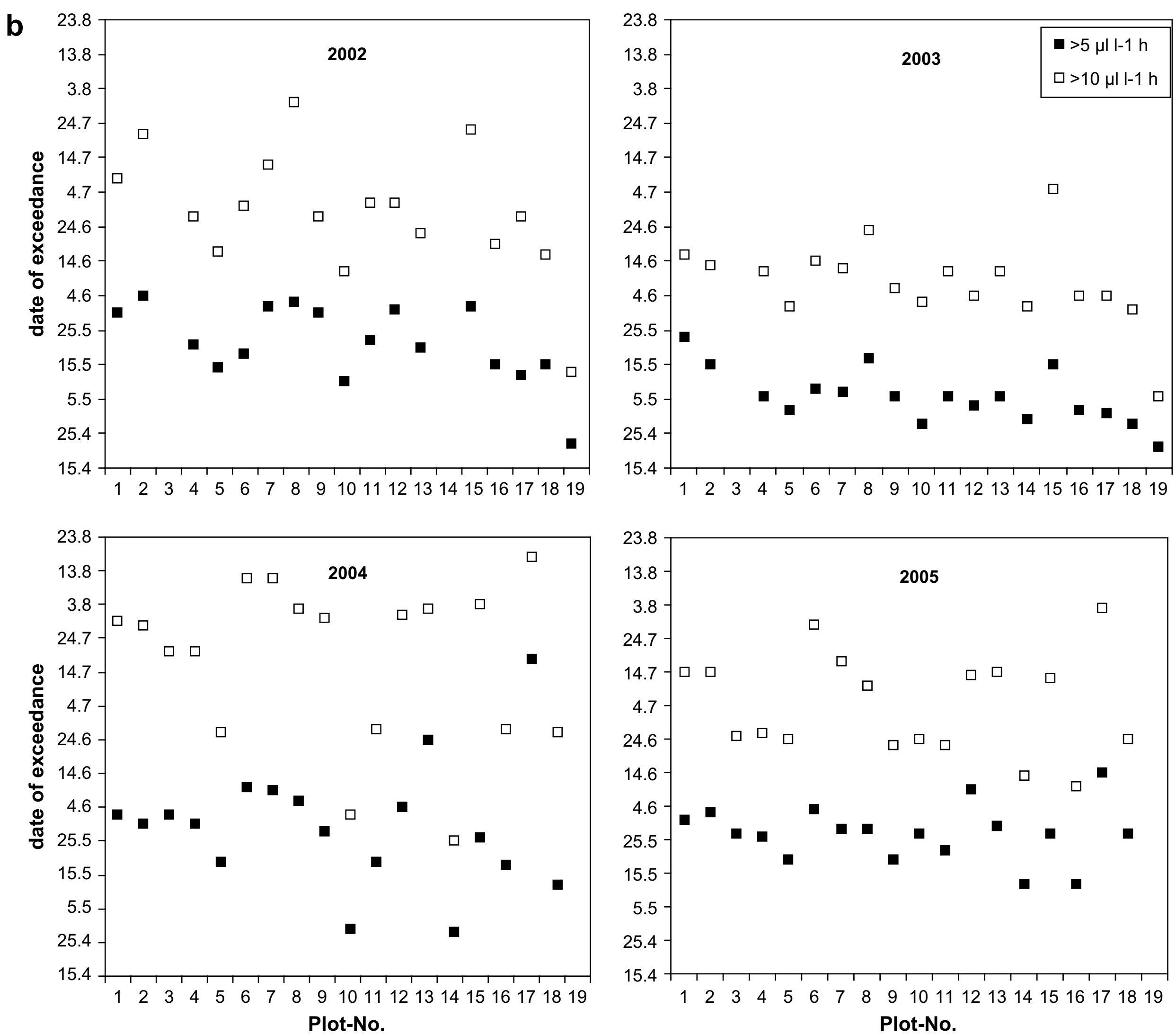


Fig. 5. (*continued*).

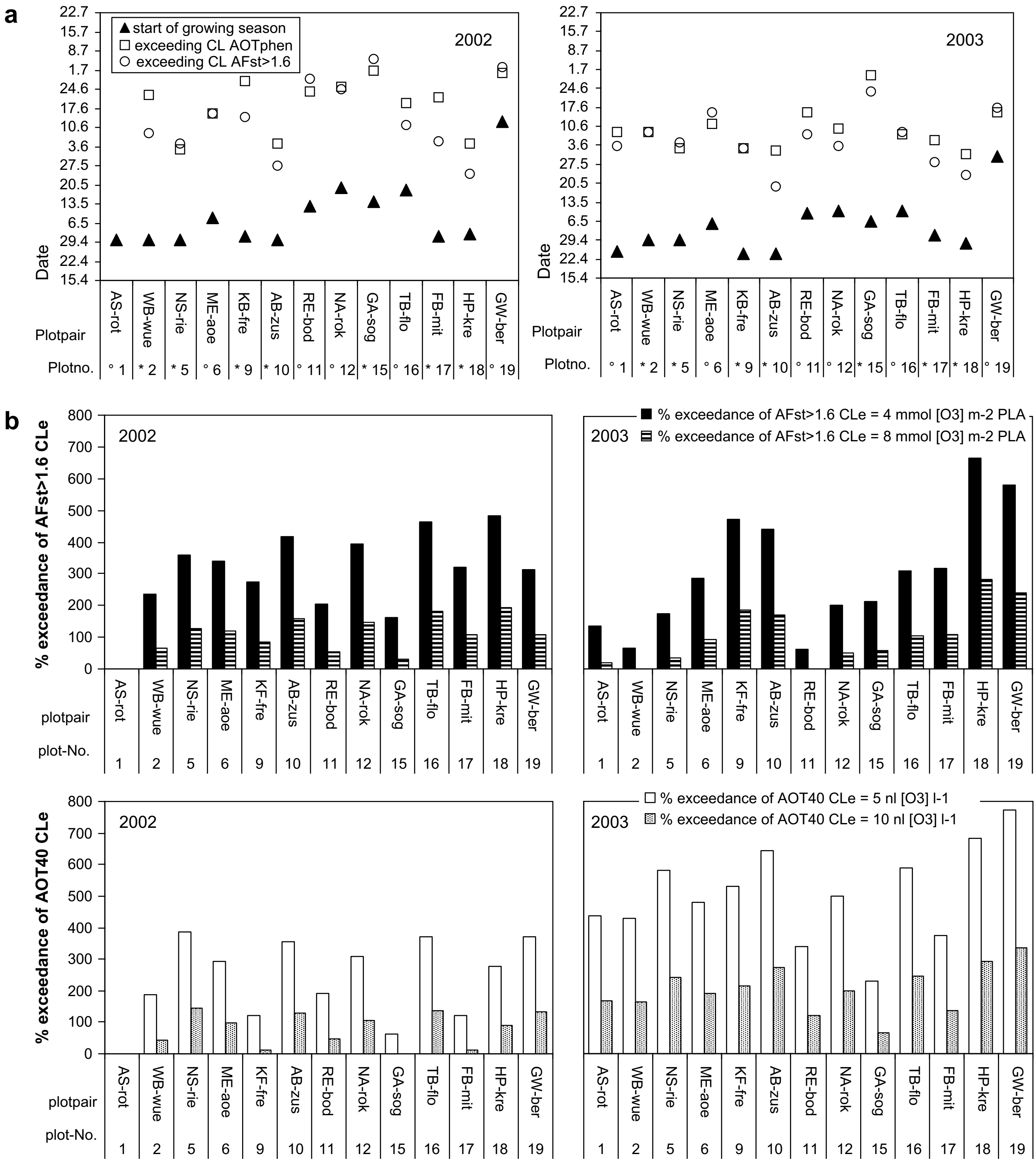


Fig. 6. (a) Date of exceedance of the current $AOT40_{phen}$ and $AF_{st>1.6}$, Critical Levels for forests for the plot pairs (Table 1) for the effective phenological growing season for 2002 and 2003. Start of growing season: leaf emergence >50% of foliation; CL AOT 40: AOT40 Critical Level >5 µl O_3 l^{-1} h; CL $AF_{st>1.6}$: (provisional) Critical Level for forest trees $AF_{st>1.6} = 4$ mmol O_3 m^{-2} s^{-1}; PLA*: beginning of growing season for beech, directly observed at plot (>50% leaf emergence);°: beginning of growing season for beech, calculated from "Level II sites" data (according to Kramer 1994, 1995). (b) Percentage of exceedance of the $AF_{st>1.6}$ (accumulated flux based stomatal ozone uptake above a threshold of 1.6 nmol O_3 m^{-2} s^{-1} PLA) and $AOT40_{phen}$ (accumulated hourly ozone concentration over a threshold of 40 nl O_3 l^{-1} for the effective phenological season) Critical Levels for forest trees for the plot pairs (Table 1) for the effective phenological growing seasons 2002 and 2003. Current (provisional) Critical Level $AF_{st>1.6} = 4$ mmol O_3 m^{-2} PLA, and tentatively doubled Critical Level $AF_{st>1.6} = 8$ mmol O_3 m^{-2} PLA. Current $AOT40_{phen}$ Critical Level AOT40 = 5 µl O_3 l^{-1} h, and formerly twofold high Critical Level AOT40 = 10 µl O_3 l^{-1} h.

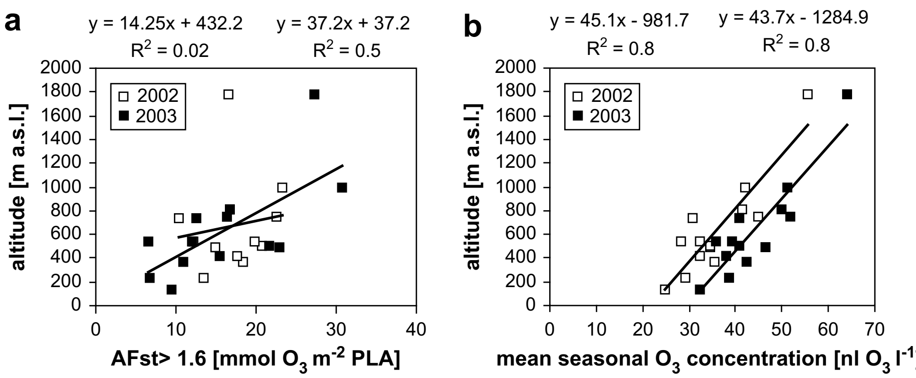


Fig. 7. (a) Correlation of the $AF_{st>1.6}$ (accumulated flux based stomatal O_3 uptake above a threshold of 1.6 nmol O_3 m^{-2} s^{-1} PLA) with the altitude, and b) correlation of altitude with mean seasonal O_3 concentration for the plot pairs (Table 1) for 2002 and 2003.

4. Discussion

4.1. O_3 data from open field sites as surrogate for forest sites

We demonstrated in this study that continuously monitored O_3 concentrations from open field sites can be used as surrogates for proximal forest sites at similar altitudes. Variation was influenced by altitude rather than distance between plots. An increase of O_3 concentration with altitude is well known in literature (e.g., Zaveri et al., 1995; Brönnimann et al., 2000; Chevalier et al., 2007). The differences in O_3 concentrations were mostly below 10% and increased up to 20–30% towards altitude differences of about 300 m between the plots being compared. It is assumed that deviations are negligible for regional forest risk assessments.

4.2. Exposure in comparison with flux based approaches

The exposure-based indices AOT, $AOT40_{phen}$ and MPOC as well as the flux-based index $AF_{st>1.6}$ suggest that Bavarian forests are at risk from O_3. According to MPOC, forest sites were within the risk category "where compliance ensures substantial protection for forest trees" throughout the study period (2002–2005), i.e. adverse O_3 effects including leaf injury might occur (Grünhage et al., 2001). Across the study sites, both the current and former CL of AOT40 (i.e. 5 µl O_3 l^{-1} h and 10 µl O_3 l^{-1} h, respectively; Karlsson et al., 2004) were exceeded often for several times during each growing season investigated, indicating the risk of growth reduction at all forest sites. Exceedance of the current CL mostly occurred at the beginning of the growing season before the completion of shoot and leaf formation, i.e. on average around the end of April to mid-May. The hot, sunny conditions that caused the drought in 2003 also caused the exposure-based exceedance to occur somewhat earlier. The AOT40 definition requires risk assessment to be restricted to a mean growing season of April to September. However, the "effective growing season" (as observed at individual sites) showed a delay in springtime by 2–3 weeks at sites above approximately 700 m a.s.l. and an additional delay of about 2 weeks at the high altitude site GW. Hence, $AOT40_{phen}$ was in agreement with AOT40 at low altitudes, whereas substantial discrepancy occurred at high altitudes between these two indices. Also, the CL of $AOT40_{phen}$ was exceeded at almost all sites in 2002 and 2003, irrespective of the current or former determination. Exposure based indices indicated a substantially increased risk in the dry year of 2003 (also Löw et al., 2006), especially in altitudes above 1000 m a.s.l..

The current provisional CL of $AF_{st>1.6}$ of 4 mmol O_3 m^{-2} for forest trees was substantially exceeded in our study, both during the humid year of 2002 (representing average climate conditions for the region) and the extremely dry year of 2003. However, the doubled CL of $AF_{st>1.6}$ was only moderately exceeded at some of the sites.

The relationship between $AF_{st>1.6}$ and $AOT40_{phen}$ was approximately linear under humid conditions. Schaub et al. (2007) also found high correlation between O_3 flux and AOT40 (e.g. Karlsson et al., 2004). However, in our study the relationship between $AF_{st>1.6}$ and $AOT40_{phen}$ was largely dependent on the prevailing climatic conditions (Löw et al., 2006). During the drought period in 2003, stomatal closure and hence O_3 uptake decreased (Wieser and Havranek, 1993) so that $AF_{st>1.6}$ stagnated at several sites, whereas $AOT40_{phen}$ continued to increase (Fig. 8). At such sites, water limitation (<200 mm precipitation within the growing season) and soil properties (high percentage of sand fraction or high proportion of coarse textured soil) led to an increase in soil moisture deficit, which limited stomatal conductance and hence O_3 uptake (Retzlaff et al., 2000; Panek et al., 2002). Other plots were characterised by high water availability (at least in one soil layer) during the 2003 growing season, due to non-limiting precipitation and favourable soil properties. Thus, the $AOT40_{phen}$ index tends to overestimate the O_3 impact on trees under water limitation, as opposed to the AFst index (Panek et al., 2002; Panek, 2004; Löw et al., 2006).

4.3. Relationship between O_3 threshold exceedance and leaf symptoms

Macroscopic O_3-induced leaf injury was detected at some of the "Level II sites", as well as at the "forest research sites" *KF* and *FB*; however, the affected leaf area was always small. This kind of leaf injury was hardly found during the dry summer of 2003. At *KF*, symptoms were reported in 2002, 2004 and 2005 but were negligible in 2003, whereas at *FB*, 2003 and 2004 were the only years when they occurred to some minor degree. The low extent of injury hardly indicates impairment of leaf photosynthesis, although minor adverse effects cannot be ruled out in the absence of leaf symptoms, as reported by Nunn et al. (2006) in spruce for photosynthesis. Such minor limitations of photosynthesis appear to conflict with the distinct growth reduction suggested by the risk assessment approaches (MPOC, AOT40 and $AF_{st}Y$ analysis). Conversely, even minor O_3 effects on photosynthesis (temporary limitation by up to 15%) in adult beech, as demonstrated in a free-air O_3 fumigation experiment, did not significantly reduce annual stem production (Matyssek et al., 2007a). However, it is difficult to evaluate the predicted O_3-induced 5% growth reduction after exceedance of the CL, because this percentage might well be attributed to intra-annual differences of and multi-factorial

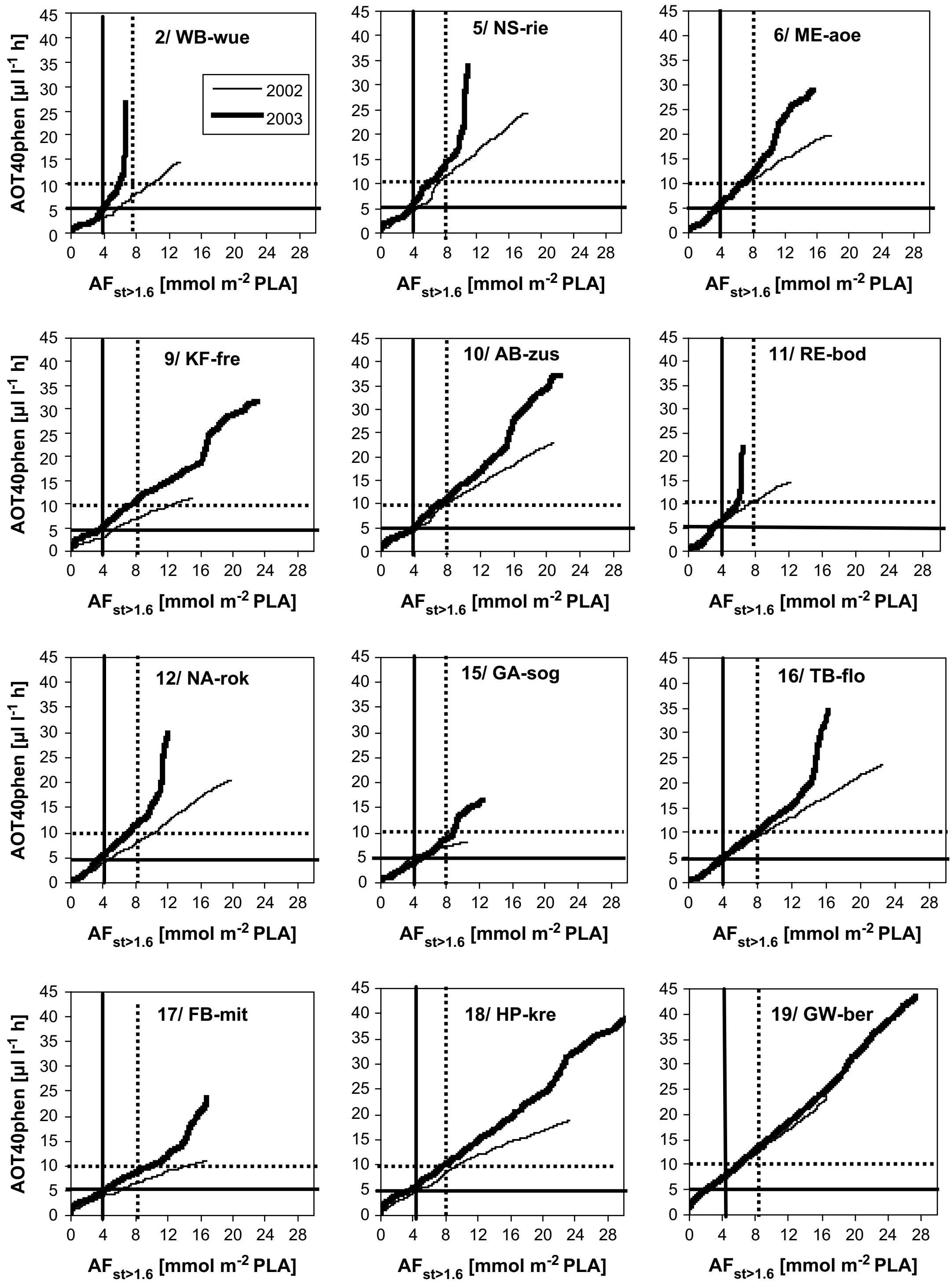


Fig. 8. Flux modelled cumulative hourly $AF_{st>1.6}$ (accumulated flux based stomatal O_3 uptake above a threshold of 1.6 nmol O_3 m^{-2} s^{-1} PLA) and $AOT40_{phen}$ (accumulated hourly ozone concentration over threshold of 40 nl O_3 l^{-1} for the real phenological season) for the plot pairs (Table 1, plot pair rot-AS (no. 1) not shown because O_3 measurements only for 2003) during the growing seasons 2002 and 2003. Bold lines: current (provisional) Critical Level for forest trees $AF_{st>1.6}$ = 4 mmol O_3 m^{-2} PLA, and current AOT40 = 5 µl O_3 l^{-1} h; bold dotted lines: tentatively doubled Critical Level for forest trees $AF_{st>1.6}$ = 8 mmol O_3 m^{-2} PLA, and formerly twofold high Critical Level AOT40 = 10 µl O_3 l^{-1} h.

influences on productivity (Spiecker, 1999; Pretzsch and Dursky, 2002; Dittmar et al., 2003; Huber et al., 2004). Since the O_3 dose was already high at the beginning of the growing season for most sites, i.e. in parallel to leaf emergence and tissue differentiation, influences on subsequent leaf performance cannot be ruled out. Numerous studies have reported that O_3 impacts during leaf growth result in structural modifications at the cellular level and macroscopic leaf injury in a variety of forest tree species (Skelly et al., 1999; Günthardt-Goerg et al., 2000; VanderHeyden et al., 2001; Vollenweider et al., 2003a,b; Kivimäenpää et al., 2005; Schaub et al., 2005). Modifications or damage at the cellular level due to high O_3 concentrations during this early stage of development may potentially lead to enhanced detoxification and subsequently to improved adaptation, but are causing structural costs

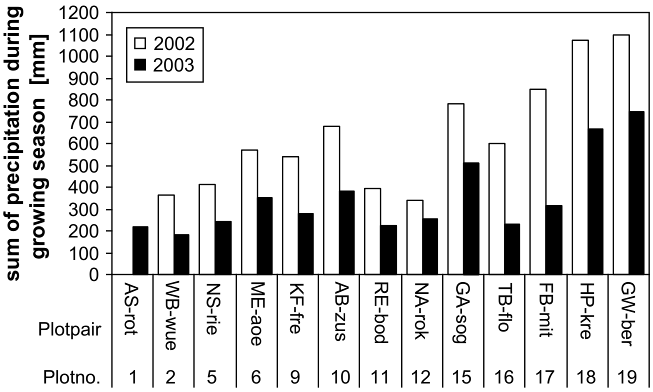


Fig. 9. Sum of precipitation during the growing seasons (April–September) 2002 and 2003 for the plot pairs (Table 1); data from "Level II sites".

(cf. Matyssek and Sandermann, 2003). Thus one can speculate that early exposure to O_3 could be one reason for the minor or absent occurrences of visible leaf injury, in spite of the CL exceedances estimated. However, it should be reconsidered whether the method conducted to assess visible injury is adequate for detecting effects of O_3 on mature forest trees. Baumgarten et al. (2000) demonstrated consistency between occurrence of macroscopic leaf injury and AF_{st} of about 3 mmol O_3 m^{-2}, both for adult beech under stand conditions and young beech in phytotrons. However, no clear relation between leaf injury and both AOT40 and AF_{st} was found at *KF* (Matyssek et al., 2004). Visible leaf injury might be suitable for indicating acute O_3 impact on trees (e.g. under controlled conditions or after short episodes of very high O_3 concentrations, cf. Matyssek and Sandermann, 2003), rather than under prevalent stand conditions where chronic effects are likely.

4.4. Limitation of tested approaches and thresholds

The MPOC approach, which is not able to reflect the influence of climatic conditions, tree species or phenology, suggested the probability of leaf injury and additional effects. As leaf symptoms are small, detoxification reactions may potentially cause a decrease in productivity. Although both AOT40 and AF_{st} indices predicted growth reductions of 5%, a CL exceedance generally indicates the probability only of statistically significant damage, and it is important to be aware that exposure based concepts are not phytomedically relevant (as not being related to O_3 uptake) and, hence, not mechanistically founded (Skärby et al., 2004; Matyssek et al., 2008). Also, the threshold applied is questionable, given its derivation from only a limited number of studies investigating juvenile beech trees cultivated in controlled environments (Matyssek and Innes, 1999; Baumgarten et al., 2000; Kolb and Matyssek, 2001; Wieser et al., 2002a, b; Herbinger et al., 2005; Nunn et al., 2005b). The CL approach was created to assess risk for the most sensitive species or genotype without differentiating between tree species, forest types or environmental conditions (Karenlämpi and Skärby, 1996; Fuhrer et al., 1997; Matyssek and Innes, 1999; Uddling et al., 2004). In addition, it is debateable whether the occurrence of O_3 induced leaf injury symptoms are reflected at all by the exceedance of the thresholds. More recently, Ferretti et al. (2007) reported the relationship between O_3 induced leaf injury symptoms and AOT40 in SW-Europe to be limited, perhaps due to varying ecological,

Table 3
Visible O_3 induced leaf injury symptoms in the upper sun crown of mature beech at "Level II sites" (forest ecosystem monitoring sites) and "forest research sites" for the years 2002–2005.

Plot	Plot -no.	2002	2003	2004	2005	References
wue	2	No	No	No	No	Dietrich and Preuhsler (2003)
rie	5	No	No	No	No	Dietrich and Preuhsler (2003)
KF	9	Intercostal necrotic symptoms during vegetation period, first ozone induced symptoms end of May-mid of June, clear ozone induced symptoms July-mid of September, leaf injury of sun leaves in the upper crown, 0.8 ± 0.8% of total foliar area[a]	Negligible macroscopic leaf injury	Intercostal necrotic symptoms during vegetation period, leaf injury of sun leaves inthe upper crown, 2.2% oftotal foliar area (no standard deviation given)	Chlorotic and necrotic leaf injury during vegetation period, first ozone inducedsymptoms mid of June slightly increasing until September, leaf injuryof sun leaves in the upper crown, 3 ± 1% of total foliar area	Nunn et al. (2005a); for 2002 Löw, pers. comm., Löw et al. (2006); Klotsche (2005) for 2004; Metzger, pers. comm. for 2005
fre		No	No	No	Intercostal but "untypical" chlorotic symptoms on sun leaves in the upper crown, lower than 1–5% of the leaves show ozone symptoms[a]	Dietrich and Preuhsler (2003)
sog	15	No	No	No	Intercostal but "untypical" chlorotic symptoms on sun leaves in the upper crown, lower than 1–5% of the leaves show ozone symptoms[a]	Dietrich and Preuhsler (2003)
FB	17	No	Chlorotic and necrotic symptoms on sun leaves in the upper crown end of July	Chlorotic and necrotic symptoms on sun leaves in the upper crown end of July	No	Beudert (2005)
mit		No	No	No	No	Dietrich and Preuhsler (2003)
kre	18	No	No	No	No	Dietrich and Preuhsler (2003)

At most plots assessment was conducted by the Bavarian Forest Institute (LWF), otherwise references are given; lower case: "Level II sites" (forest ecosystem monitoring stations); in capital letters: "forest research sites", continuous ozone monitoring, for plot information Table 1.
[a] Validation for ozone induced injury effects by "Ozone Validation center (WSL/FSL, Birmensdorf, Switzerland).

biological and methodological scenarios. For instance, Paoletti (2006) reported that application of current AOT40 in Mediterranean regions led to the exceedance of CLs for forests in past, present and projected future O_3 concentrations, while at the moment direct effects remain unclear, largely due to the difficulty in monitoring growth reductions of mature forest trees. It is concluded that current exposure-based CL calculations are not adequate for Mediterranean evergreen forests mainly due to a different growing season and subsequently altered periods of photosynthetic activity and stomatal conductance (apart from not being phytomedically relevant).

In general, both exposure and flux based approaches still lack ecologically meaningful validation for adult trees under actual forest conditions. Thresholds are a technical construction usually established for political or legal application, and do not reflect the biological meaningful transitional range in which a displacement of homeostasis, acute damage or chronic injury may take place (Matyssek et al., 2007a).

5. Conclusions and outlook

The main findings of this regional O_3 risk assessment were:

- Regularly collected O_3 monitoring data for national pollution control from rural sites measuring background concentrations outside forests can be used as surrogates to characterise the O_3 regime above forest canopies at similar altitudes with only minor shortcomings. This provides the chance to calculate O_3 indices for representative regional forest risk assessments in a very practicable way without further measurement costs. In fact, the exposure-based indices and even the flux-based index can be applied for many forested sites through a combination of O_3 data with meteorological and soil/water related data derived from the regional "Level II sites".
- According to the most common O_3 indices and thresholds in Europe, trees growing in the Bavarian forests appear to be at risk under the prevailing O_3 regimes and climatic conditions across the region with thresholds for damage being exceeded mostly in all years. According to the different approaches used in our study, O_3-induced productivity and growth reduction (Critical Level approach (AOT40, $AF_{st>1.6}$, MPOC approach) and leaf injury (MPOC approach) are predicted to be likely effects after threshold exceedance. Data describing the potential O_3-induced growth reductions for forest trees are still not available and severe visible O_3-induced leaf injury was not observed in our study.
- Regarding exposure-based indices, the highest degree of threshold exceedance occurred in the dry year of 2003, whereas the flux-based approach indicated the highest risk at moist sites or during humid years. This underlines the erratic potential of exposure-based approaches (cf. Matyssek et al., 2007b), as they are not related to the phytomedically relevant O_3 dose, and hence, are not mechanistic (Matyssek et al., 2008).

Further, continuative approaches for advanced O_3 risk assessment are postulated:

- The ultimate aim is the replacement of exposure- by flux-based concepts in O_3 risk assessment. As well as providing a mechanistic understanding of the effective O_3 dose (i.e. the trees' sensitivity per unit of O_3 uptake), it also represents a ecologically meaningful risk assessments (Matyssek et al., 2008). This latter aspect appears to be driven by tree phenology and ontogeny, as part of the overall biotic and abiotic scenario at forest sites, predominantly including soil moisture. The calculation of $AF_{st>1.6}$ has proved to be the most suitable approach here, especially under water-limited conditions as predicted to occur more frequently in Central Europe under "climate change".
- Our study also indicates that attention should be directed to forest regions with non-limited water supply, because O_3 uptake will be far less restricted under such conditions, although many sites may be prone to some kind of drought in the future (Rebetez et al., 2006; Sitch et al., 2007). O_3 flux modelling can be refined by further consideration of soil/water budget characteristics and improved estimations of crown transpiration of trees and whole forest stands (Nunn et al., 2007; Matyssek et al., 2008). It is advisable to introduce threshold ranges for forest trees in view of their specific regional climatic conditions for developing more meaningful risk predictions.
- Risk validation is problematic, as there is no reference scenario with reduced O_3 exposure available in the field. This dilemma may be overcome through free-air O_3 fumigation experiments in forests as reported by Matyssek et al. (2007a), however, such experimental approaches and mechanistic case studies (*sensu* Level III concepts) are largely missing. In particular, growth analysis of tree and forest stands needs to be fostered in relation to the actual site-specific O_3 uptake.
- It appears to be reasonable to establish ecologically adopted thresholds for forest trees in the different climatic regions, especially regarding soil water availability, in Europe. It is important to foster assessment and analysis of productivity parameters (e.g. stem increment) in combination with O_3 impact and climatic conditions. In addition, the negative O_3 effect on productivity needs further examination during long-term observations in mature forests. Considering the fluctuating environmental conditions, the flux based approach should be preferred for the assessment of the O_3 impact, and input data concerning the soil/ water budget should be further refined.

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