# Background Document

**Joint ICP Vegetation/EMEP**

**Ad-hoc Expert Panel Meeting on Modelling and Mapping of Ozone Flux and Deposition to Vegetation**

*to be held under the*

**UN/ECE Convention on Long-range Transboundary Air Pollution**

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# An $O_3$ flux-based risk assessment for spring wheat

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As a consequence of the discussions about the reasons of the so-called 'Neuartige Waldschäden' (forest die-back) ground-level ozone ($O_3$) and its impact on human health and vegetation has come into focus more and more within the UNECE (United Nations Economic Commission for Europe) and the European Union since mid eighties of the last century. The first European workshop on critical levels for $O_3$ to protect vegetation was held 1988 in Bad Harzburg, Germany, (UN-ECE 1988), followed by a second one 1992 in Egham, UK (Ashmore & Wilson 1992). While the 1988 long-term critical level for $O_3$ was defined as a 7-hour mean of 25 ppb over the vegetation/growing period, at the Egham workshop a change to a cumulative exposure index over a certain threshold was recommended. The basis for the current European Convention on Long-Range Transboundary Air Pollution to abate Acidification, Eutrophication and Ground-level $O_3$ (UNECE 1999) and the European Directive on Ground-level $O_3$ (EU 2002) was initiated at the UNECE workshop in Bern, Switzerland, 1993. This was followed by a discussion on the suitability of the concept in the scientific literature in the following years. Meanwhile a reorientation from cumulative exposure index-based critical levels to flux-based limiting values took place (cf Grünhage & Jäger 2002; Fig. 1).

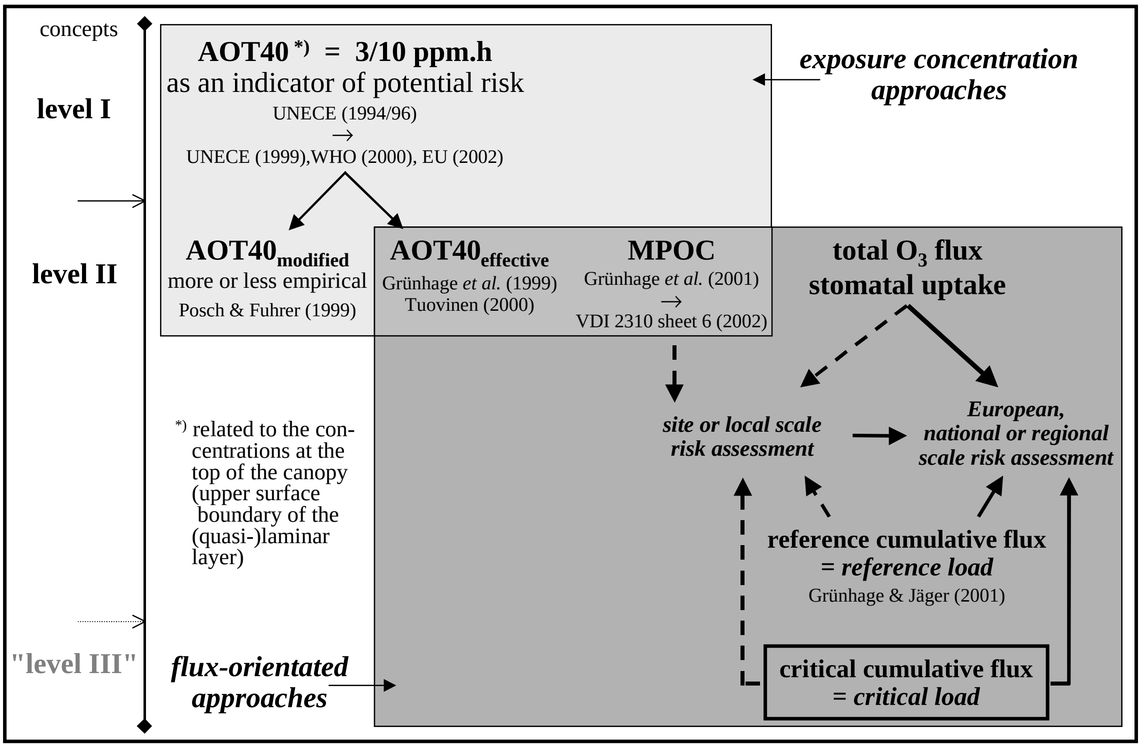


Fig. 1. From critical levels to critical loads for ozone (Grünhage & Jäger 2002)
The light grey rectangle contains approaches based on exposure concentrations, whereas the dark grey rectangle summarizes flux-orientated concepts. Concepts listed in the overlay are based on canopy concentrations, which have to be estimated applying a resistance model for ambient conditions.
(AOT40: accumulative exposure over a threshold of 40 ppb; MPOC: maximum permissible $O_3$ concentration)

The only adequate tool to ensure effective protection against adverse effects of $O_3$ on vegetation is the derivation of critical cumulative fluxes/stomatal uptake (critical loads) for sensitive vegetation types similar to the critical loads for acidification and eutrophication as determined in accordance with the Convention's Manual on Methodologies and Criteria for Mapping Critical Levels/Loads (UBA 1996). It seems advisable to differentiate between approaches for site or local (km) scale risk assessments and for risk assessments on a European, national or regional scale as indicated in Figure 1. While European, national and regional risk assessments are based on more or less generalizing concepts, site and local scale risk assessments require a higher degree of precision. In addition, generalizing concepts have to be based on approaches validated on representative flux measurement sites distributed over Europe as indicated by the arrow in Figure 1 between the two levels of risk assessments proposed.

At present, the data base for the derivation of critical loads for $O_3$ is extremely insufficient. For spring wheat, a flux (stomatal uptake) - response (relative yield) relationship was deduced by Pleijel et al. (2000) from 5 open-top chamber experiments with two wheat varieties only (Fig. 2).

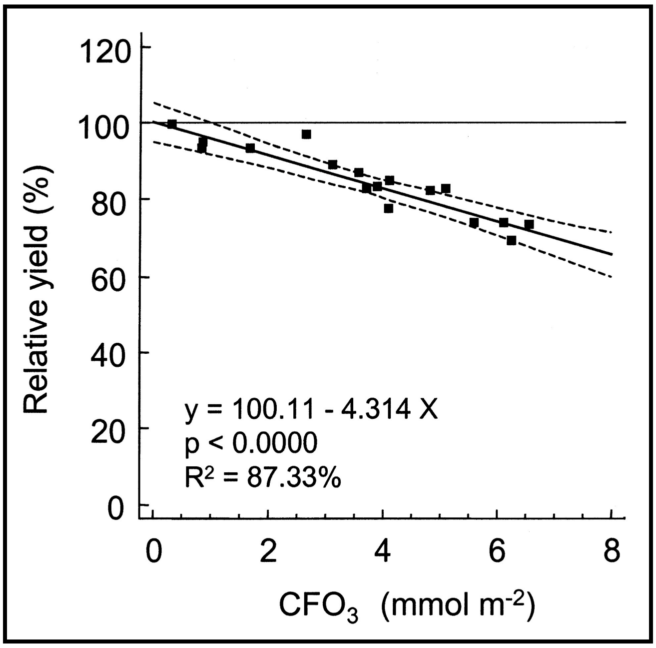


Fig. 2. Relative yield of spring wheat vs cumulative stomatal uptake of $O_3$ ($CFO_3$) by the flag leaf during grain filling (Pleijel et al. 2000, modified)

This relation was applied for a representative agricultural site in Hesse, one of the federal states of Germany, using the SVAT model WINDEP (Worksheet-Integrated Deposition Estimation Programme; Grünhage & Haenel 2000).

Stomatal uptake by the flag leaf was parameterized as described in Pleijel et al. (2000), the development of spring wheat canopy during the grain filling period (phenological stage codes 61 to 87; after Zadoks et al. 1974 and Tottman 1987) as described in Grünhage et al. (1999) in addition with an up-scaling from leaf to canopy according to eq. (36) in Grünhage et al. (2000). The WINDEP model version used can be downloaded from:

http://www.uni-giessen.de/~gf1034/ENGLISH/WINDEP.htm

Taking into account the statistical uncertainties indicated by the confidence interval in Figure 2, stomatal uptake above 1 $mmol \cdot m^{-2}$ $O_3$ is linked with a yield loss deviating significantly from a

100 % yield. To avoid an overestimation of risk i.e. yield loss, it seems to be reasonable to subtract this threshold from the modelled $O_3$ absorbed dose, $PAD(O_3)$, which then results in:

$$\textit{relative yield loss} = 100 - \left\{100.11 - \left[\left(4.314\,\mathrm{m^2\cdot mmol^{-1}}\right)\cdot\left(PAD(\mathrm{O_3}) - 1\,\mathrm{mmol\cdot m^{-2}}\right)\right]\right\}$$

with $PAD(O_3)$ in $\mathrm{mmol\cdot m^{-2}}$

As shown in Figure 3 more than 10 % yield loss due to $O_3$ stomatal uptake could be estimated for 1994, only. According to the experimental conditions optimal water supply, i.e. soil moisture at field capacity, was assumed. Moderate water stress reduce the impact of $O_3$ significantly due to reduced aperture of the stomata.

This example demonstrates the applicability of the flux approach for site and local scale risk assessments in principle. On the other hand the application of Pleijel's flux-response relation can be criticized due to the small number of experiments with two "old" wheat varieties from the late eighties and mid nineties only at one site in Sweden and due to the fact that the model parameterization was not validated and therefore is more or less empirical.

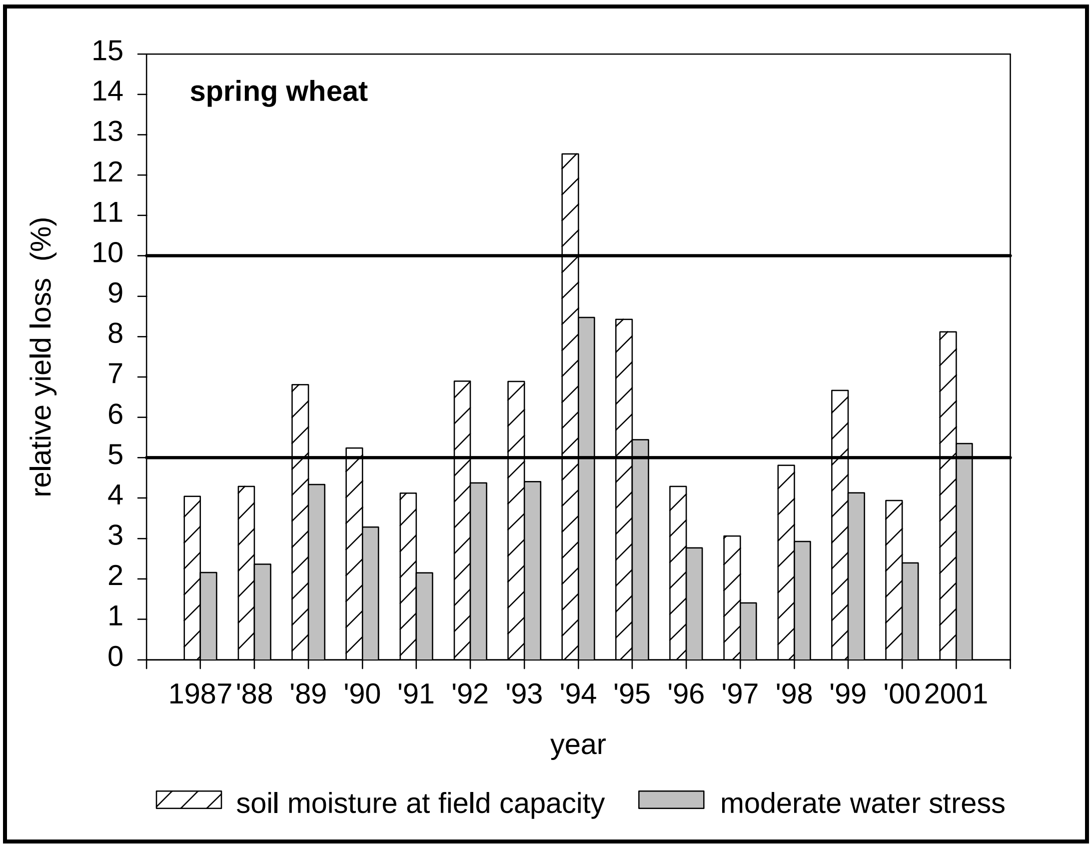


Fig. 3. Variation in time of potential relative yield loss (%) due to $O_3$ stomatal uptake under optimal water supply (Jarvis factor for soil moisture = 1) and moderate water stress (Jarvis factor for soil moisture = 0.7) for a representative Hessian agricultural site (fixed growing season; Grünhage & Jäger 2002)

## Model description

The *big leaf* model WINDEP is a resistance model (Fig. 4), based on the soil-vegetation-atmosphere-transfer (SVAT) model PLATIN (PLant-ATmosphere INteraction; Grünhage & Haenel 1997). WINDEP can be used in computer spreadsheets for Windows Lotus and Excel.

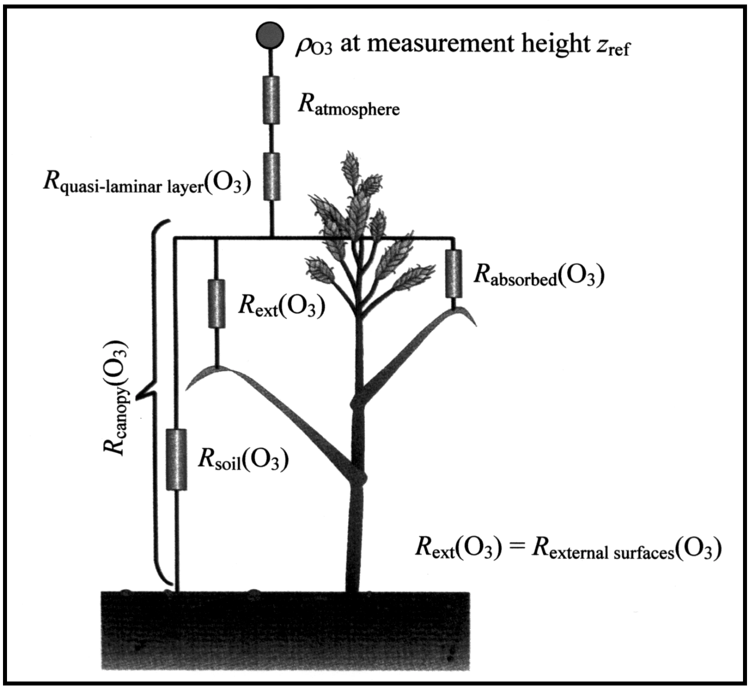


**Fig. 4:** A deposition resistance analogy for ozone (modified from PORG 1997)

The exchange of $O_3$ between the phytosphere and the atmosphere near the ground, $F_{total}(O_3)$ [$\mu g \cdot m^{-2} \cdot s^{-1}$], can be modelled by:

$$F_{total}(O_3) = - \frac{\rho_{O3}(z_{ref})}{R_{ah}(d + z_{0m}, z_{ref,O3}) + R_{b,O3} + R_{c,O3}} \tag{1}$$

with $\rho_{O3}(z_{ref})$ — $O_3$ concentration measured at reference height $z_{ref}$ [$\mu g \cdot m^{-3}$]

$R_{ah}(d+z_{0m}, z_{ref, O3})$ — turbulent atmospheric resistance [$s \cdot m^{-1}$] describing the atmospheric transport properties between a reference height $z_{ref, O3}$ above the canopy and the conceptual height $z = d + z_{0m}$ which represents the sink for momentum ($d$ = displacement height, $z_{0m}$ = roughness length for momentum)

$R_{b, O3}$ — quasi-laminar layer resistance [$s \cdot m^{-1}$] between momentum sink height $z = d + z_{0m}$ and the $O_3$ sink height $z = d + z_{0,O3}$

$R_{c, O3}$ — bulk canopy or surface resistance [$s \cdot m^{-1}$] describing the influences of the plant/soil system on the vertical exchange of $O_3$

The resistance network (Fig. 4) allows to partition the total atmosphere-canopy flux $F_{total}(O_3)$ into the fluxes reaching the stomatal caves ($F_{absorbed}$), the external plant surfaces ($F_{external\ plant\ surfaces}$) and the soil beneath the canopy ($F_{soil}$)

$$F_{total}(O_3) = F_{absorbed} + F_{external\ plant\ surfaces} + F_{soil} \tag{2}$$

(see eqs. (38) - (40)). The integral of $F_{\mathrm{absorbed}}$ over time $t$ is the ***p**ollutant **a**bsorbed **d**ose*, $PAD(\mathrm{O_3})$ [$\mu g \cdot m^{-2}$], (Fowler & Cape 1982):

$$PAD(\mathrm{O}_3) = \int_{t_1}^{t_2} \left| F_{\mathrm{absorbed}}(\mathrm{O}_3) \right| \cdot \mathrm{d}t \tag{3}$$

According to the Monin-Obukhov theory (Monin & Obukhov 1954), the atmospheric resistance $R_{\mathrm{ah}}$ between the heights $z_1$ und $z_2$ can be expressed by

$$R_{\mathrm{ah}}(z_1, z_2) = \frac{\ln\left(\frac{z_2 - d}{z_{0\mathrm{m}}}\right) - \Psi_{\mathrm{h}}\left(\frac{z_2 - d}{L}\right) + \Psi_{\mathrm{h}}\left(\frac{z_1 - d}{L}\right)}{\kappa \cdot u_*} \tag{4}$$

with $z_2 = z_{\mathrm{ref,\,O3}}$ and $z_1 = d + z_{0\mathrm{m}}$

and $L$ is the Monin-Obukhov length [m], $\kappa$ is the dimensionless von Kármán constant (= 0.41; cf Dyer 1974), $u_*$ is the friction velocity [$m \cdot s^{-1}$] and $\Psi_{\mathrm{h}}$ is the integrated atmospheric stability function for sensible heat (see chapter "spreadsheet stratification"). The friction velocity is given by:

$$u_* = \frac{\kappa \cdot u(z_2')}{\ln\left(\frac{z_2' - d}{z_{0\mathrm{m}}}\right) - \Psi_{\mathrm{m}}\left(\frac{z_2' - d}{L}\right) + \Psi_{\mathrm{m}}\left(\frac{z_1 - d}{L}\right)} \tag{5}$$

with $z_2' = z_{\mathrm{ref,}\,u}$ and $z_1 = d + z_{0\mathrm{m}}$

and $\Psi_{\mathrm{m}}$ the atmospheric stability function for momentum.

The quasi-laminar layer resistance for ozone $R_{\mathrm{b,\,O3}}$ is estimated according to a simple approach by Hicks et al. (1987) taking into account the empirical results for permeable rough canopies described by Brutsaert (1984); for details see Grünhage et al. (2000):

$$\begin{aligned} R_{\mathrm{b,\,O3}} &= R_{\mathrm{b,\,heat}} \cdot \left(\frac{\mathrm{Sc}}{\mathrm{Pr}}\right)^{\frac{2}{3}} = \frac{\ln\left(\frac{z_{0\mathrm{m}}}{z_{0\mathrm{h}}}\right) - \Psi_{\mathrm{h}}\left(\frac{z_{0\mathrm{m}}}{L}\right) + \Psi_{\mathrm{h}}\left(\frac{z_{0\mathrm{h}}}{L}\right)}{\kappa \cdot u_*} \cdot \left(\frac{\mathrm{Sc}}{\mathrm{Pr}}\right)^{\frac{2}{3}} \\ &= \frac{2 - \Psi_{\mathrm{h}}\left(\frac{z_{0\mathrm{m}}}{L}\right) + \Psi_{\mathrm{h}}\left(\frac{z_{0\mathrm{h}}}{L}\right)}{\kappa \cdot u_*} \cdot 1.18 \end{aligned} \tag{6}$$

with $\ln(z_{0\mathrm{m}} / z_{0\mathrm{h}}) = 2$, i.e. roughness length for sensible heat $z_{0\mathrm{h}} = z_{0\mathrm{m}} / \exp(2)$

where $R_{\mathrm{b,\,heat}}$ is the quasi-laminar layer resistance for sensible heat, Sc is the Schmidt number (the ratio of the kinematic viscosity of dry air and the molecular diffusivity of the respective trace gas) und Pr is the Prandtl number (the ratio of the kinematic viscosity of dry air and the molecular diffusivity of heat). For water vapor $(\mathrm{Sc/Pr})^{2/3}$ is 0,90.

The bulk canopy resistance $R_{\mathrm{c,\,O3}}$ is a composite resistance describing the transfer through the leaf stomata $R_{\mathrm{leaf,\,stom,\,O3}}$ and into the mesophyll tissue $R_{\mathrm{leaf,\,mes,\,O3}}$, the transfer through the cuticle of the leaves $R_{\mathrm{leaf,\,cut,\,O3}}$ and the deposition on external plant surfaces $R_{\mathrm{leaf,\,ext,\,O3}}$ and on the soil $R_{\mathrm{soil,\,O3}}$. By upscaling from leaf to canopy, these resistances are combined as follows:

$$\frac{1}{R_c(O_3)} = \left[ LAI_{green} \cdot \left( \frac{1}{R_{leaf, stom, O3} + R_{leaf, mes, O3}} + \frac{1}{R_{leaf, cut, O3}} \right) + \frac{LAI_{total}}{R_{leaf, ext, O3}} + \frac{\beta}{R_{soil, O3}} \right] \qquad (7)$$

According to the discussion in Grünhage et al. (2000), the in-canopy aerodynamic transfer resistance $R_{in\text{-}canopy}$ is replaced by the use of a weighted $R_{soil}$: with $\beta$ the actual canopy development stage is taken into account (cf Grünhage & Haenel (1997; eq. (12)).

The calculation of the aforementioned resistances, i.e. the exchange of $O_3$ between phytosphere and atmosphere near the ground requires the following measured input parameters:

- ozone concentration $\rho_{O3}$ [$\mu g \cdot m^{-3}$] at a reference height $z_{ref, O3}$
- horizontal wind velocity $u$ [$m \cdot s^{-1}$] at a reference height $z_{ref, u}$
- global radiation $S_t$ [$W \cdot m^{-2}$]
- air temperature $t_a$ [°C] at a reference height $z_{ref, T}$
- air humidity $rH$ [%] at a reference height $z_{ref, rH}$
- air pressure $p$ [hPa] at a reference height $z_{ref, p}$

Spring wheat canopy architecture and development is characterized by

- roughness length for momentum $z_{0m}$ [m]
- displacement height $d$ [m]
- leaf area index of non-senescent leaves $LAI_{green}$ [$m^2 \cdot m^{-2}$]
- leaf area index of the whole canopy $LAI_{total}$ [$m^2 \cdot m^{-2}$]
- shortwave albedo $\alpha$ [= 0.22 for $S_t > 0$ $W \cdot m^{-2}$]

with $d = 0.67 \cdot h$, $z_{0m} = 0.13 \cdot h$ and $h$ the height of the canopy according to Grünhage et al. (1999):

**Table 1:** Encoding of typical phenological stages of spring and winter wheat in the AMBAV model (Grünhage et al. 1999)

| | **spring wheat** | **winter wheat** |
|---|---|---|
| $DOY_{start}$ | first leaf through coleoptile (code [*)] 10) | 60 |
| $DOY_{code\ 31}$ | stem elongation, first node detectable (code 31) | |
| $DOY_{between}$ | $DOY_{code\ 31}$ - 10 | $DOY_{code\ 31}$ - 5 |
| $DOY_{code\ 51}$ | first spikelet of inflorescence just visible (code 51) | |
| $DOY_{max}$ | $DOY_{code\ 51}$ + 5 | $DOY_{code\ 51}$ + 5 |
| $DOY_{code\ 61}$ | beginning of anthesis (code 61) = $DOY_{max}$ | |
| $DOY_{code\ 87}$ | hard dough (code 87) | |
| $DOY_{harvest}$ | harvest (caryopsis hard, code 92) | |

DOY, day of the year

[*)] after Zadoks et al. (1974) and Tottmann (1987)

**Table 2:** Approximation of canopy height $h$ of wheat (Grünhage et al. 1999)

| | |
|---|---|
| $\mathrm{DOY} < \mathrm{DOY}_{start}$ | $: \; h = h_1$ |
| $\mathrm{DOY}_{start} \le \mathrm{DOY} < \mathrm{DOY}_{between}$ | $: \; h = h' = \frac{h_2}{8} \cdot \frac{(\mathrm{DOY} - \mathrm{DOY}_{start})}{(\mathrm{DOY}_{between} - \mathrm{DOY}_{start})}$ if $h' > h_1$ else $h = h_1$ |
| $\mathrm{DOY}_{between} \le \mathrm{DOY} < \mathrm{DOY}_{max}$ | $: \; h = \frac{h_2}{7} + \left(h_2 - \frac{h_2}{7}\right) \cdot \frac{(\mathrm{DOY} - \mathrm{DOY}_{between})}{(\mathrm{DOY}_{max} - \mathrm{DOY}_{between})}$ |
| $\mathrm{DOY}_{max} \le \mathrm{DOY} \le \mathrm{DOY}_{harvest}$ | $: \; h = h_2$ |
| with $h_1 = 0$ and $h_2 = 0.8$ m for spring wheat, | |
| or $h_1 = 0.05$ m and $h_2 = 0.8$ m for winter wheat. | |

**Table 3:** Approximation of total leaf area index (non-senescent and senescent leaves) $LAI_{total}$ of wheat (Grünhage et al. 1999)

| | |
|---|---|
| $\mathrm{DOY} \le \mathrm{DOY}_{start}$ | $: LAI_{total} = LAI_{start}$ |
| $\mathrm{DOY}_{start} < \mathrm{DOY} < \mathrm{DOY}_{between}$ | $: LAI_{total} = (LAI_{between} - LAI_{start}) \cdot \frac{(\mathrm{DOY} - \mathrm{DOY}_{start})}{(\mathrm{DOY}_{between} - \mathrm{DOY}_{start})} + LAI_{start}$ |
| $\mathrm{DOY}_{between} \le \mathrm{DOY} < \mathrm{DOY}_{max}$ | $: LAI_{total} = (LAI_{max} - LAI_{between}) \cdot \frac{(\mathrm{DOY} - \mathrm{DOY}_{between})}{(\mathrm{DOY}_{max} - \mathrm{DOY}_{between})} + LAI_{between}$ |
| $\mathrm{DOY}_{max} \le \mathrm{DOY} \le \mathrm{DOY}_{max} + 5$ | $: LAI_{total} = LAI_{max}$ |
| $\mathrm{DOY}_{max} + 5 < \mathrm{DOY} \le \mathrm{DOY}_{harvest}$ | $: LAI_{total} = LAI_{max} - \frac{(\mathrm{DOY} - [\mathrm{DOY}_{max} + 5])}{(\mathrm{DOY}_{harvest} - [\mathrm{DOY}_{max} + 5])} \cdot \frac{LAI_{max}}{1.7}$ |
| with $LAI_{start} = 0$, $LAI_{between} = 0.3\ m^2 \cdot m^{-2}$ and $LAI_{max} = 4.3\ m^2 \cdot m^{-2}$ for spring wheat, | |
| or $LAI_{start} = 0.4\ m^2 \cdot m^{-2}$, $LAI_{between} = 0.7\ m^2 \cdot m^{-2}$ and $LAI_{max} = 6.5\ m^2 \cdot m^{-2}$ for winter wheat. | |

**Table 4:** Approximation of leaf area index of non-senescent leaves, $LAI_{non\text{-}senescent}$, of wheat (Grünhage et al. 1999)

| | |
|---|---|
| $\mathrm{DOY} \le \mathrm{DOY}_{between}$ | $: LAI_{non\text{-}senescent} = LAI_{total}$ |
| $\mathrm{DOY} = \mathrm{DOY}_{code\ 31}$ | $: LAI_{non\text{-}senescent} = 0.95 \cdot LAI_{total}$ |
| $\mathrm{DOY} = \mathrm{DOY}_{code\ 51}$ | $: LAI_{non\text{-}senescent} = 0.9 \cdot LAI_{total}$ |
| $\mathrm{DOY} = \mathrm{DOY}_{code\ 61}$ | $: LAI_{non\text{-}senescent} = 0.8 \cdot LAI_{total}$ |
| $\mathrm{DOY} \ge \mathrm{DOY}_{code\ 87}$ | $: LAI_{non\text{-}senescent} = 0$ |
| Between these characteristic stages $LAI$ values are derived by linear interpolation. | |

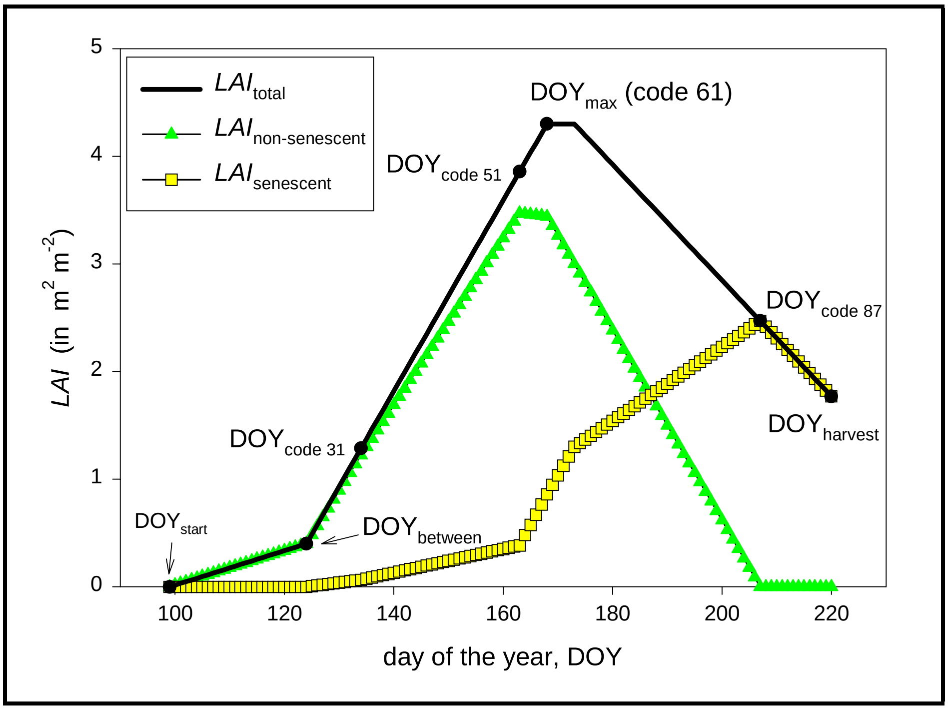


**Fig. 5:** Mean development of leaf area index (*LAI*) of spring wheat at Braunschweig, Germany (Grünhage et al. 1999)

($DOY_{start}$ = 99; $DOY_{code\ 31}$ = 134; $DOY_{code\ 51}$ = 163; $DOY_{code\ 87}$ = 207; $DOY_{harvest}$ = 220)

Minimum value of flag leaf stomatal resistance for $H_2O$ is calculated from the maximum stomatal conductance $g_{max}$ for $O_3$ of 0.154 mol·m$^{-2}$·s$^{-1}$ (cf Pleijel et al. 2000) according to:

$$R_{stom,min,\,H20} = \left[ g_{max,\,O3} \cdot \left( 44.6 \cdot \frac{273.15}{T} \cdot \frac{p}{1013.25} \right)^{-1} \cdot \frac{D_{H2O}}{D_{O3}} \right]^{-1} = 179 \text{ s·m}^{-1} \qquad (8)$$

with $T$ = 293.15 K

$p$ = 1013.25 hPa

$D$ molecular diffusivity ($10^{-6}$·m$^2$·s$^{-1}$)

and $D_{H2O}/D_{O3}$ = 1.51 according to Grünhage & Haenel (1997).

WINDEP consists of 7 spreadsheets. In the spreadsheet "information" the following data must be provided by the user:

- measurement height of horizontal wind velocity $z_{ref,\,u}$ [m]
- measurement height of air temperature $z_{ref,\,T}$ [m]
- measurement height of air humidity $z_{ref,\,rH}$ [m]
- measurement height of air pressure $z_{ref,\,p}$ [m]
- measurement height of ozone concentration $z_{ref,\,O3}$ [m]
- latitude [degree]
- longitude [degree]
- number of days in the respective year (365 or 366)
- difference between Local Standard Time and Greenwich Mean Time

[h; MET – GMT = +1 h] *)

- length of measurement interval [h]
- units of ozone concentration (1 = ppb; 2  $\mu g \cdot m^{-3}$)
- minimum value of flag leaf stomatal resistance for $H_2O$ [= 179 $s \cdot m^{-1}$]

*) GMT = Greenwich Mean Time; MET = Mean European Time

The other spreadsheets must be worked off in the following order:

1. : spreadsheet "input"
2. : spreadsheet "R_canopy"
3. : spreadsheet "radiation"
4. : spreadsheet "MO length"
5. : spreadsheet "stratification"
6. : spreadsheet "output"

***Spreadsheet "input"***

The aforementioned input parameters must be entered. Additionally, daylight hours and data sets with $LAI_{\mathrm{green}} > 0$ are indicated. If air pressure data are not available, use standard pressure $p_0$ = 1013,25 hPa. Missing values must be indicated by "-999".

***Spreadsheet "R_canopy"***

In this spreadsheet the water vapor pressure deficit of the atmosphere *VPD* [hPa], the weighting function $\beta$ for canopy development, the Jarvis-Stewart functions for parameterizing the dependence of stomatal aperture on radiation, air temperature, *VPD*, soil moisture and phenological stage the bulk stomatal resistances $R_{\mathrm{c,\,stom,\,H2O}}$ and $R_{\mathrm{c,\,stom,\,O3}}$ and the bulk canopy resistances $R_c(H_2O)$ and $R_c(O_3)$ are calculated. Furthermore, the slope of water vapor pressure saturation pressure curve $s$ [$hPa \cdot K^{-1}$], the specific heat of moist air $c_{\mathrm{p,\,moist\,air}}$ [$m^2 \cdot s^{-2} \cdot K^{-1}$] and the density of moist air $\rho_{\mathrm{moist\,air}}$ [$kg \cdot m^{-3}$] at absolute air temperature $T$ [$T = t_a + 273.15$; K] are calculated.

• *water vapor pressure deficit of the atmosphere*

$$VPD = e_{\mathrm{saturation\,water\,vapor\,pressure}} - e_{\mathrm{water\,vapor\,pressure}} \tag{9}$$

with the saturation water vapor pressure of the atmosphere [hPa; after Magnus]:

actual air temperature $t_a \geq 0$°C: $$e_{\mathrm{saturation\,water\,vapor\,pressure}} = 6.1078 \cdot e^{\frac{17.08085 \cdot t_a}{234.175 + t_a}} \tag{10a}$$

actual air temperature $t_a < 0$°C: $$e_{\mathrm{saturation\,watr\,vapor\,pressure}} = 6.1078 \cdot e^{\frac{22.44294 \cdot t_a}{272.44 + t_a}} \tag{10b}$$

and the water vapor pressure [hPa]:

$$e_{\mathrm{water\,vapor\,pressure}} = e_{\mathrm{saturation\,water\,vapor\,pressure}} \cdot \frac{rH}{100} \tag{11}$$

• *weighting functions for canopy development*

$$\beta = e^{-c_{\mathrm{LAI}} \cdot LAI_{\mathrm{total}}} \tag{12}$$

where $c_{\mathrm{LAI}}$ is the a vegetation type-specific attenuation coefficient [= 0.5; cf discussion in Grünhage et al. (2000)].

• *Jarvis-Stewart functions* (according to Pleijel et al. 2000)

$$f_1(S_t) = 1 - e^{-0.009 \cdot (2 \cdot S_t)} \tag{13a}$$

with $S_t$ actual global radiation [$W \cdot m^{-2}$]

$$f_2(T) = 0.1 \quad \text{if} \quad t_a \leq 14°C$$
$$f_2(T) = (-0.0059 \cdot t_a^{\,2}) + (0.3083 \cdot t_a) - 3.0275 \quad \text{if} \quad 14 < t_a < 39 \tag{13b}$$
$$f_2(T) = 0.1 \quad \text{if} \quad t_a \geq 39°C$$

with $t_a$ actual temperature [°C]

$$f_3(VPD) = 1 \quad \text{if} \quad VPD \leq 0.9\,\text{kPa}$$
$$f_3(VPD) = (-0.4737 \cdot VPD) + 1.4263 \quad \text{if} \quad 0.9\,\text{kPa} < VPD \leq 2.8\,\text{kPa} \tag{13c}$$
$$f_3(VPD) = 0.1 \quad \text{if} \quad VPD > 2.8\,\text{kPa}$$

with $VPD$ actual water vapor pressure deficit [kPa]

$$f_4(\textit{soil moisture}) = 1\,(\text{optimal water supply}) \quad \text{or} \quad 0.7\,(\text{moderate water stress}) \tag{13d}$$

$$f_5(\text{phenological stage}) = 1 - (0.027 \cdot \text{"number of days after anthesis"}) \tag{13e}$$

with $f_5 = 0$ if $[1 - (0.027 \cdot \text{"number of days after anthesis"})] < 0$

$$f_{\text{Jarvis}} = f_1(S_t) \cdot f_2(T) \cdot f_3(VPD) \cdot f_4(\textit{soil moisture}) \cdot f_5(\text{phenological stage}) \tag{13f}$$

with $f_{\text{Jarvis}} = 0.1$ if $[f_1 \cdot f_2 \cdot f_3 \cdot f_4 \cdot f_5] < 0.1$

• *flag leaf stomatal resistance for $H_2O$ and $O_3$*

$$R_{\text{leaf, stom, A}} = \left[ \frac{1}{R_{\text{leaf, stom, min, H2O}}} \cdot f_{\text{Jarvis}} \right]^{-1} \cdot \frac{D_{\text{H2O}}}{D_{\text{A}}} \tag{14}$$

where $D_{H2O}$ is the molecular diffusivity of water vapor [$m^2 \cdot s^{-1}$] and $D_A$ is the molecular diffusivity for a trace gas species A. For water vapor is $D_{H2O}/D_A = 1$, for $O_3$ is $D_{H2O}/D_A = 1.51$. For $S_t = 0$, $R_{\text{leaf, stom}}$ is set to "1E+20 $s \cdot m^{-1}$".

• *bulk canopy resistance for $H_2O$ and $O_3$*

The bulk canopy resistance for $O_3$ is calculated according to eq. (7) with $R_{\text{leaf, mes, O3}} = 0.01\ s \cdot m^{-1}$, $R_{\text{leaf, cut, O3}} = 3 \cdot 10^7\ s \cdot m^{-1}$, $R_{\text{leaf, ext, O3}} = 2000\ s \cdot m^{-1}$ and $R_{\text{soil, O3}} = 375\ s \cdot m^{-1}$ under the assumption that the external plant surfaces are dry and the soil surface is wet. The bulk canopy resistance for $H_2O$ is calculated with $R_{\text{leaf, mes, H20}} = 0$, $R_{\text{leaf, cut, H2O}}$ with $9 \cdot 10^4\ s \cdot m^{-1}$ and $R_{\text{soil, H2O}} = 100\ s \cdot m^{-1}$. As dry external plant surfaces are assumed, $R_{\text{c, ext, H2O}}$ can be neglected.

• *slope of the water vapor pressure saturation curve*

actual air temperature $t_a \geq 0$°C:

$$s \approx e_{\text{saturation vapor vapor pressure}} \cdot \frac{17.08085 \cdot 234.175}{(234.175 + t_a)^2} \quad (15a)$$

actual air temperature $t_a < 0$°C:

$$s \approx e_{\text{saturation water vapor pressure}} \cdot \frac{22.44294 \cdot 272.44}{(272.44 + t_a)^2} \quad (15b)$$

• *specific heat of moist air at constant pressure*

$$c_{\text{p, moist air}} = c_{\text{p, dy air}} \cdot (1 + 0.84 \cdot q) \quad (16)$$

with $c_{\text{p, dry air}} = 1004.67\ \text{m}^2\cdot\text{s}^{-2}\cdot\text{K}^{-1}$

and

$q$ specific air humidity [g·g$^{-1}$]

$$q = \frac{0.622 \cdot e_{\text{water vapor pressure}}}{p - 0.378 \cdot e_{\text{water vapor pressure}}} \quad (17)$$

• *density of moist air at temperature T*

$$\rho_{\text{moist air}} = \rho_{\text{dry air}} \cdot \left(1 - 0.378 \cdot \frac{e_{\text{water vapor pressure}}}{p}\right) \quad (18a)$$

with

$$\rho_{\text{dry air}} = \frac{p}{R_{\text{dry air}} \cdot (273.15 + t_a)} \cdot 100 \quad (18b)$$

and $R_{\text{dry air}}$ the gas constant for dry air (= 287.04 J·kg$^{-1}$·K$^{-1}$)

***Spreadsheet "radiation"***

For estimating the Monin-Obukhov length $L$ (see eqs. 4 - 6) the net radiation balance $R_{\text{net}}$ [W·m$^{-2}$]

$$R_{\text{net}} = \frac{S_t \cdot (1 - \alpha) + \varepsilon \cdot L_d - \varepsilon \cdot \sigma \cdot (273{,}15 + t_a)^4}{1 + (\varepsilon \cdot c_3)} \quad (19)$$

with
- $\varepsilon$ effective long-wave emissivity of the canopy (= 0.97)
- $L_d$ flux density of downward long-wave radiation of the atmosphere [W·m$^{-2}$]
- $\sigma$ Stefan-Boltzmann constant (= 5.669·10$^{-8}$ W·m$^{-2}$·K$^{-4}$)
- $c_3$ heating coefficient of the canopy (= 0.041714; cf Holtslag & van Ulden (1983)

and the soil heat flux density $G$ [W·m$^{-2}$]

$R_{\text{net}} \geq 0$ W·m$^{-2}$:

$$G = 0.4 \cdot e^{-c_{\text{LAI}} \cdot LAI_{\text{total}}} \cdot R_{\text{net}} \quad (20a)$$

$R_{\text{net}} < 0$ W·m$^{-2}$:

$$G = 0.5 \cdot R_{\text{net}} \quad (20b)$$

must be approximated.

In the following a new parameterization of lang-wave radiation of the atmosphere $L_d$, derived for 50.53°N 8.69°E, and a parameterization of maximum possible global radiation at cloudless sky $S_{t,\,ref}$ is described.

• *daytime* $L_d$ (global radiation $S_t > 0$ W·m$^{-2}$):

$$L_{d,\,S_t>0\,W\cdot m^{-2}} = 1.24 \cdot \left( \frac{e_{2m}}{T_{2m}} \right)^{1/7} \cdot \sigma \cdot T_{2m}^{\;4} + A_{dd} + B_{dd} + C_{dd} \qquad (21)$$

with

$$A_{dd} = 20 - 70 \cdot \left( S_t / S_{t,\,ref} \right) \qquad \text{for} \quad S_t \le S_{t,\,ref}$$

$$A_{dd} = -50 \qquad \text{for} \quad S_t > S_{t,\,ref}$$

$$B_{dd} = 2.3 + 1.33 \cdot (rH - 40) \qquad \text{for} \quad rH \ge 40\ \%$$

$$B_{dd} = 2.3 \qquad \text{for} \quad rH < 40\ \%$$

and

$$C_{dd} = -12.3 + 1.1 \cdot (rH - 70) \qquad \text{for} \quad rH \ge 70\ \%$$

$$C_{dd} = -12.3 + 0.6 \cdot (70 - rH) \qquad \text{for} \quad rH < 70\ \%$$

Making use of the relation between measured global radiation and maximum possible solar irradiation, the term $A_{dd}$ accounts for the fact that the sky is not always totally covered, which in fact is assumed by term $B_{dd}$. The dependance of $B_{dd}$ on relative humidity can be derived formally (with the help of some minor simplifications) from the assumption that the sky be completely covered by cumuli, the lower boundary of which can be described by the lifting condensation level (cf Stull 1989). Other than for $A_{dd}$ and $B_{dd}$, no direct physical explanation seems possible for the third term, $C_{dd}$, the contribution of which to the variance, however, is much smaller than that of the other terms. The constants in $A_{dd}$, $B_{dd}$, and $C_{dd}$ were adjusted to minimize the bias resulting from the introduction of the variable parts of these three terms.

• *nighttime* $L_d$ ($S_t = 0$ W·m$^{-2}$):

$$L_{d,\,S_t=0\,W\cdot m^{-2}} = 1.24 \cdot \left( \frac{e_{2m}}{T_{2m}} \right)^{1/7} \cdot \sigma \cdot T_{2m}^{\;4} + A_{dn} + B_{dn} \qquad (22)$$

with

$$A_{dn} = 14 + 10.7 \cdot (rH - 92.5) \qquad \text{for} \quad rH \ge 92.5\ \%$$

$$A_{dn} = 14 \qquad \text{for} \quad rH < 92.5\ \%$$

and

$$B_{dn} = 10 - 20 \cdot \sqrt{T_{2m,\,n-1} - T_{2m,\,n}} \qquad \text{for} \quad (T_{2m,\,n-1} - T_{2m,\,n}) > 0 \ \text{(n: actual data set)}$$

$$B_{dn} = 10 \qquad \text{for} \quad (T_{2m,\,n-1} - T_{2m,\,n}) \le 0$$

where $e_{2m}$ is the actual water vapor pressure (hPa) and $T_{2m}$ is the absolute air temperature (K) at $z$ = 2 m above ground.

• *maximum possible global radiation $S_{t, ref}$*

$S_{t, ref}$ is the astronomic maximum maximum possible global radiation at cloudless sky parameterized according to Kasten & Czeplak (1980)

$$S_{t, ref} = S_{t, cloudless sky} = a_1 \cdot \sin\phi + a_2 \tag{23}$$

where $a_1$ and $a_2$ are empirical coefficients describing the average atmospheric attenuation of short-wave radiation by water vapor and dust at a given site, and $\phi$ is solar elevation. For our field site at 50.53°N 8.69°E $a_1$ and $a_2$ were adjusted to $a_1$ = 1097 W·m$^{-2}$ and $a_2$ = –54 W·m$^{-2}$.

Solar elevation $\phi$ is calculated dependending on latitude, longitude and time according to Lenoble (1993):

$$\sin\phi = \sin\varphi_{geo} \cdot \sin\Delta + \cos\varphi_{geo} \cdot \cos\Delta \cdot \cos\varphi_h \tag{24}$$

mit $\varphi_{geo}$ latitude [radians]
$\Delta$ sun declination [radians]
$\varphi_h$ hour angle [radians]

Sun declination $\Delta$ is given by:

$$\Delta = 0.006918 - 0.399912 \cdot \cos\varphi_d + 0.070257 \cdot \sin\varphi_d - 0.006758 \cdot \cos 2\varphi_d + 0.000907 \cdot \sin 2\varphi_d \tag{25}$$

where the day angle $\varphi_d$ [radians] is:

$$\varphi_d = 2 \cdot \pi \cdot \frac{\text{day of the year} - 1}{\text{number of days in the year}} \tag{26}$$

The hour angle $\varphi_h$ is given by

$$\varphi_h = \pi \cdot \left(1 - \frac{TST}{12}\right) \tag{27}$$

where TST is the True Solar Time [h; decimal system] for the center of the time interval under consideration:

$$TST = GMT + \frac{\lambda_{geo}}{15} + ET - \frac{DT}{2} \tag{28}$$

with GMT Greenwich Mean Time (for Germany: MET – 1)
MET Mean European Time [h]
$\lambda_{geo}$ latitude [degree]
DT duration of time interval [h]

and the equation of time ET:

$$ET = 3.819667 \cdot (0.000075 + 0.001868 \cdot \cos\varphi_d - 0.032077 \cdot \sin\varphi_d - 0.014615 \cdot \cos 2\varphi_d - 0.040849 \cdot \sin 2\varphi_d) \tag{29}$$

***Spreadsheet "MO length"***

The aforementionend atmospheric stability functions for momentum $\Psi_m$ and sensible heat $\Psi_h$ are dependent on the Monin-Obukhov length $L$ (Monin & Obukhov 1954):

$$L = -\rho_{\text{moist air}} \cdot c_{\text{p, moist air}} \cdot \frac{\bar{\bar{\theta}} \cdot u_*^3}{\kappa \cdot g \cdot H} \approx -\rho_{\text{moist air}} \cdot c_{\text{p, moist air}} \cdot \frac{T(z_{\text{ref}}) \cdot u_*^3}{\kappa \cdot g \cdot H} \tag{30}$$

with $\bar{\bar{\theta}}$ average potential temperature of the air layer under consideration (K)

$g$ gravitational acceleration (= 9.81 $m \cdot s^{-2}$)

$H$ turbulent vertical flux density of sensible heat [$W \cdot m^{-2}$]

The average potential temperature of the air layer under consideration can be approximated by an absolute temperature $T(z_{ref})$ measured in the respective air layer. Eq. (30) is iteratively solved in 8 steps. Yielding sufficient accuracy, $H$ is estimated as residual of the energy balance:

$$R_{\text{net}} = H + \lambda E + G \tag{31}$$

where $\lambda E$ is the evapotranspiration rate (latent heat flux density) of the plant/soil system [$W \cdot m^{-2}$]. The latent heat flux density is calculated by the Penman-Monteith approach (Monteith 1965):

$$\lambda E = \frac{s\,(R_{\text{net}} - G) + \rho_{\text{moist air}} \cdot c_{\text{p,moist air}} \cdot \dfrac{VPD}{R_{\text{ah}}(d + z_{0\text{m}}, z_{\text{ref},T}) + R_{\text{b, heat}}}}{s + \gamma \cdot \dfrac{R_{\text{ah}}(d + z_{0\text{m}}, z_{\text{ref},T}) + R_{\text{b, H2O}} + R_{\text{c, H2O}}}{R_{\text{ah}}(d + z_{0\text{m}}, z_{\text{ref},T}) + R_{\text{b, heat}}}} \tag{32}$$

where $\gamma$ is the psychrometric constant (= 0.655 $hPa \cdot K^{-1}$).

$R_{ah}(d+z_{0m}, z_{ref,\,T})$, the turbulent atmospheric resistance between $z_{ref,\,T}$ and $z_1 = d+z_{0m}$, is calculated after eq. (4). In the first iterative step, the M-O length is approximated under neglection of the stability functions $\Psi_m$ und $\Psi_h$ by solving the equation system (4) - (7), (31) and (32). In the following iterative steps the stability functions, which are described in the spreadsheet "stratification", are taken into account. If the friction velocity $u_*$ falls below 0.05 $m \cdot s^{-1}$ during daylight hours or below 0.075 $m \cdot s^{-1}$ during night, a mimimum-$u_*$ is used to maintain a plausible energetic coupling between canopy and atmosphere:

$$S_t \geq 50\ W \cdot m^{-2}: \qquad u_{*,\text{min}} = \max\left(0.05\ ;\ 0.15 \cdot \sqrt{\frac{u(\text{zref})}{z_{\text{ref}}(u) - (d + z_{0\text{m}})}}\right) \tag{33a}$$

$$S_t < 50\ W \cdot m^{-2}: \qquad u_{*,\text{min}} = \max\left(0.075\ ;\ 0.3 \cdot \sqrt{\frac{u(\text{zref})}{z_{\text{ref}}(u) - (d + z_{0\text{m}})}}\right) \tag{33b}$$

For introduction of mimimum friction velocity $u_*$-min see discussion by, for example, Rißmann (1998).

***Spreadsheet "stratification"***

For a given M-O length the following atmospheric stability functions for momentum $\Psi_m$ and sensible heat $\Psi_h$ are calculated using the set of coefficients published by Dyer (1974) with $\kappa = 0.41$.

• *labile atmospheric stratification* ($L$ < 0 m)

$$\Psi_m(\zeta) = 2 \cdot \ln\left[\frac{1}{\phi_m(\zeta)} + 1\right] + \ln\left[\frac{1}{\phi_m^{\ 2}(\zeta)} + 1\right] - 2 \cdot \arctan\left[\frac{1}{\phi_m(\zeta)}\right] \quad (34a)$$

with $\phi_m(\zeta) = (1 - 16 \cdot \zeta)^{-0.25}$

and $\zeta = \dfrac{z - d}{L}$ with $z = z_2' = z_{ref,\,u}$ and $z = z_1 = d + z_{0m}$

and

$$\Psi_h(\zeta) = 2 \cdot \ln\left[\frac{1}{\phi_h(\zeta)} + 1\right] \quad (34b)$$

with $\phi_h(\zeta) = (1 - 16 \cdot \zeta)^{-0.5}$

and $\zeta = \dfrac{z - d}{L}$ with $z = z_2 = z_{ref,\,O3}$ and $z = z_1 = d + z_{0m}$

• *stabile atmospheric stratification* ($L$ > 0 m)

$$\Psi_m(\zeta) = \Psi_h(\zeta) = -5 \cdot \zeta \quad (35)$$

and $\zeta = \dfrac{z - d}{L}$ and $z = z_2 = z_{ref,\,O3}$ and $z = z_2' = z_{ref,\,u}$

and $z = z_1 = d + z_{0m}$

• *neutral atmospheric stratification* ($|L| \to \infty$)

$$\Psi_m = \Psi_h = 0 \quad (36)$$

***Spreadsheet "output"***

Using the aforementioned atmospheric stability functions the friction velocity is calculated according to (5) taking into account $u_{*,\,min}$, as well as the turbulent atmospheric resistance $R_{ah}(d + z_{0m},\ z_{ref,\,O3})$ and the quasi-laminar layer resistance $R_{b,\,O3}$. Ozone exchange between phytosphere and atmosphere near the ground can now calculated by eq. (1). If necessary, ozone concentration can be converted from unit "ppb" to unit " µg·m$^{-3}$" taking into account air temperature and air pressure:

$$1\,\text{ppb} = \frac{\text{molecular weight}}{22.4} \cdot \frac{273.15}{273.15 + t_a} \cdot \frac{p}{1013.25}\ \mu g \cdot m^{-3} \quad (37)$$

with molecular weight$_{O3}$ = 48 g·mol$^{-1}$

$F_{absorbed}(O_3)$, $F_{external\ plant\ surfaces}(O_3)$ and $F_{soil}(O_3)$ [µg·m$^{-2}$·s$^{-1}$] are calculated as follows:

$$F_{\text{absorbed}}(O_3) = - \frac{\rho_{O3}(z_{\text{ref}})}{R_{\text{ah}} + R_{\text{b, O3}} + \frac{R_{\text{leaf, absorbed, O3}}}{LAI_{\text{green}}} + \left( [R_{\text{ah}} + R_{\text{b, O3}}] \cdot \frac{R_{\text{leaf, absorbed, O3}}}{LAI_{\text{green}}} \cdot \left[ \frac{LAI_{\text{total}}}{R_{\text{leaf, ext, O3}}} + \frac{\beta}{R_{\text{soil, O3}}} \right] \right)} \quad (38)$$

with $$\frac{1}{R_{\text{leaf, absorbed, O3}}} = \frac{1}{R_{\text{leaf, stom, O3}} + R_{\text{leaf, mes, O3}}} + \frac{1}{R_{\text{leaf, cut, O3}}} \cong \frac{1}{R_{\text{leaf, stom, O3}} + R_{\text{leaf, mes, O3}}}$$

$$F_{\text{external plant surfaces}}(O_3) = - \frac{\rho_{O3}(z_{\text{ref}})}{R_{\text{ah}} + R_{\text{b, O3}} + \frac{R_{\text{leaf, ext, O3}}}{LAI_{\text{total}}} + \left( [R_{\text{ah}} + R_{\text{b, O3}}] \cdot \frac{R_{\text{leaf, ext, O3}}}{LAI_{\text{total}}} \cdot \left[ \frac{LAI_{\text{green}}}{R_{\text{leaf, absorbed, O3}}} + \frac{\beta}{R_{\text{soil, O3}}} \right] \right)} \quad 39)$$

$$F_{\text{soil}}(O_3) = - \frac{\rho_{O3}(z_{\text{ref}})}{R_{\text{ah}} + R_{\text{b, O3}} + \frac{R_{\text{soil, O3}}}{\beta} + \left( [R_{\text{ah}} + R_{\text{b, O3}}] \cdot \frac{R_{\text{soil, O3}}}{\beta} \cdot \left[ \frac{LAI_{\text{green}}}{R_{\text{leaf, absorbed, O3}}} + \frac{LAI_{\text{total}}}{R_{\text{leaf, ext, O3}}} \right] \right)} \quad (40)$$

The amount of ozone absorbed by the vegetation, $PAD(O_3)_{canopy}$ [$\mu g \cdot m^{-2}$], is calculated by eq. (3) and converted from unit "$\mu g \cdot m^{-2}$" into unit "$nmol \cdot m^{-2}$". Taking into account the leaf area index of non-senescent leaves $PAD(O_3)_{canopy}$ [$nmol \cdot m^{-2}$] is expressed on a unit leaf area basis.

Potential relative yield loss due to $O_3$ stomatal uptake is calculated as follows:

$$\textit{relative yield loss} = 100 - \{100.11 - [(4.314\, m^2 \cdot mmol^{-1}) \cdot (PAD(O_3) - 1\, mmol \cdot m^{-2})]\} \quad (41)$$

with $PAD(O_3)$ on a unit leaf area basis in $mmol \cdot m^{-2}$

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