# A chamber-less approach to derive ozone flux-effect relationships for crops and semi-natural vegetation

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## Introduction

In order to quantify the impacts of ozone ($O_3$) on vegetation and to derive biologically meaningful air quality standards, current discussions focus on a possible transfer of the UNECE AOT40 Level I into a Level II approach. There is a general agreement that the current exposure-concentration approach must be replaced by a flux-oriented one, i.e. *critical levels* have to be replaced by *critical loads* (cumulated fluxes between the atmosphere and the phytosphere).

Because the European *critical levels* as well as the German maximum permissible ozone concentrations (MPOC; Grünhage et al. 2001, VDI 2310 part 6 2002) are based mainly on chamber experiments, they are biased in principle. Generally, exposure-response relationships deduced from chamber experiments show increasing intensities of plant responses with increasing $O_3$ exposure concentration due to the experimental design as illustrated in Fig. 1. This contrasts with observations under ambient conditions where the biological response to ozone exposure does not increase continuously with increasing exposure (e.g. Bugter & Tonneijck 1990, Krupa et al. 1995). Moreover, the chamber environment may affect plant and plant community responses independently of a pollution stress due to the differences in radiation, air temperature and air humidity between chamber and ambient microclimate (e.g. Grünhage et al. 1990). Therefore, similar to the problems with the $O_3$ exposure-response approach, the derivation of flux-response relationships from chamber experiments is also likely to be questionable. Only few free-air fumigation systems for $O_3$ were developed during the last 20 years (e.g. McLeod et al., 1985, 1992, Wulff et al. 1992). However, due to the difficulties to establish defined exposure treatments these systems were not used for deriving exposure-effect relationships.

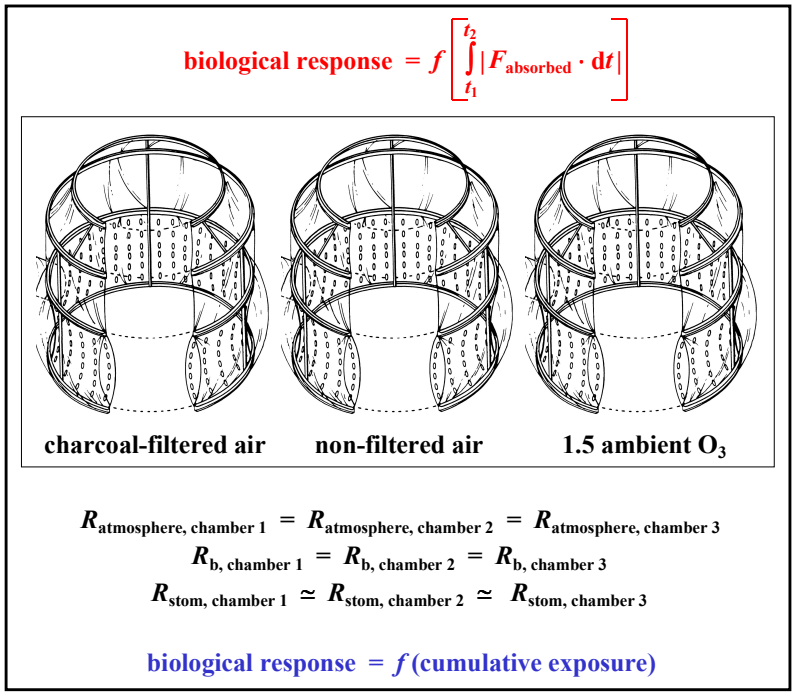


**Fig. 1:** Conventional concept for the derivation of exposure-response relationships

## The SPIDER concept

Taking into account the situation above mentioned, we developed a concept, called SPIDER (**S**imulation of air **P**ollutant **I**njection and its influence on **D**eposition **E**stimation **R**esults), which combines methods of the toxicology of air pollutants and of micrometeorology (Fig. 2).

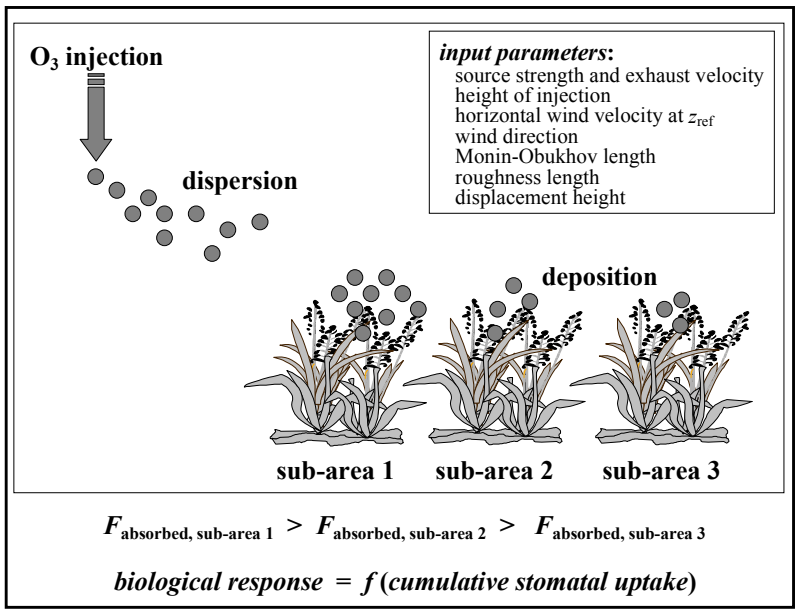


**Fig. 2:** The SPIDER concept for the derivation of flux-response relationships under chamber-less, ambient conditions

As an analogy to free-air fumigation approaches, $O_3$ is released into the atmosphere by an injection system at some height above the canopy under investigation (experimental phase).

Three-dimensional atmospheric dispersion and surface deposition of the $O_3$ released from the point source above the canopy are calculated using the Lagrangian trajectory modelling (cf Grünhage & Jäger 2002; modelling phase).

The point source in the SPIDER model is represented as a nozzle with a given emission velocity. The gas flow is restricted to vertical directions (upward or downward). At present, SPIDER does not consider turbulence effects induced by the nozzle itself. In order to enable "quantifying" of $O_3$ deposited at the canopy, the vegetated area surrounding the pollutant source is divided into rectangular sub-plots as in a chessboard.

Depending on wind direction and velocity several sub-plots can be identified around the point of pollutant release, which show $O_3$ deposition rates above the ambient levels, without any disturbance to the microclimate and micrometeorology. Deposition rates and vegetation responses at these sub-plots can then be easily used to derive $O_3$ flux-effect relationships under ambient conditions taking into account the "conventional" methods to deduce limiting values for risk assessments. It must be noticed, however, that atmospheric chemistry is presently not taken into account in the SPIDER approach. The model results can be biased, for instance, due to the reaction of $O_3$ with NO emitted from soil and with hydrocarbons emitted by the vegetation.

## Model application

The SPIDER approach can be separated into six steps which will be described briefly in the following.

(i) Micrometeorological flux measurements of $O_3$ for quantifying the actual "background deposition"

As the $O_3$ deposition is increased artificially above ambient level, a prerequisite to the derivation of a flux-response relationship is the quantification of the actual $O_3$ "background deposition" by micrometeorological methods (e.g. eddy covariance (EC) technique):

$$F_{\text{total}}(O_3) = \overline{w' \cdot \rho'} + \text{correction terms}$$

with $w'$ the fluctuation of vertical wind velocity and $\rho'$ the fluctuation of $O_3$ concentration.
The EC method requires sufficient horizontal fetch conditions and horizontal homogeneity of vegetation and soil properties (cf e.g. Grünhage et al. 2000).

(ii) Modelling the partitioning of total $O_3$ flux into toxicologically effective stomatal uptake and non-stomatal deposition

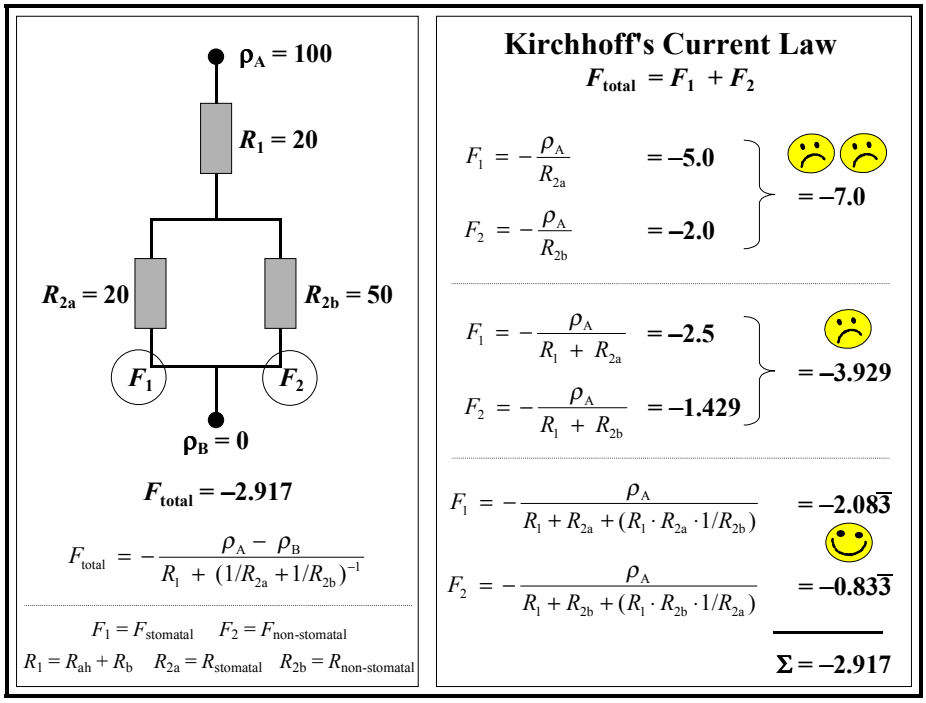


**Fig. 3:** Partitioning of total into partial fluxes according to concepts published recently
(units: $\rho$ in $\mu g \cdot m^{-3}$, $R$ in $s \cdot m^{-1}$, $F$ in $\mu g \cdot m^{-2} \cdot s^{-1}$)

Toxicologically relevant is the amount of $O_3$ absorbed mainly through the stomata, which cannot be measured directly. However, it can be estimated via the calculation of stomatal canopy resistance, $R_{\text{stomatal}}$, using measurements of water vapour fluxes at the experimental site taking into account the ratio of the molecular diffusivities of $O_3$ and $H_2O$. By consideration of Kirchhoff's Current Law (cf Fig. 3), total $O_3$ flux can then be partitioned into stomatal uptake and non-stomatal deposition by:

$$F_{\text{stomatal}} = -\frac{\rho_{O3}}{R_{\text{ah}} + R_{\text{b}} + R_{\text{stomatal}} + \left[ [R_{\text{ah}} + R_{\text{b}}] \cdot R_{\text{stomatal}} \cdot \frac{1}{R_{\text{non-stomatal}}} \right]}$$

and

$$F_{\text{non-stomatal}} = -\frac{\rho_{O3}}{R_{\text{ah}} + R_{\text{b}} + R_{\text{non-stomatal}} + \left[ [R_{\text{ah}} + R_{\text{b}}] \cdot R_{\text{non-stomatal}} \cdot \frac{1}{R_{\text{stomatal}}} \right]}$$

where $R_{\text{ah}}$ is the turbulent atmospheric resistance and $R_{\text{b}}$ the quasi-laminar layer resistance. The non-stomatal canopy resistance, $R_{\text{non-stomatal}}$, can be derived from measurements by:

$$R_{\text{non-stomatal}} = \left[ \frac{1}{-\frac{\rho_{O3}}{F_{\text{total}}} - (R_{\text{ah}} + R_{\text{b}})} - \frac{1}{R_{\text{stomatal}}} \right]^{-1}$$

A description of the calculation procedures can be downloaded from:

http://www.uni-giessen.de/~gf1034/pdf/O3flux.pdf

The sad faces in Fig. 3 denote that these equations used in the present literature are wrong in principle and yield inadequate partial fluxes.

(iii) Artificial $O_3$ injection to increase $O_3$ deposition above ambient level

The size of the area under the influence of $O_3$ and the magnitude of increased $O_3$ deposition depend on the height of injection above the canopy, the source strength and the exhaust velocity but mainly on the horizontal wind velocity near the canopy (cf Grünhage & Jäger 2002): The lower the wind speed, the smaller is the area under the influence of $O_3$ and the higher is the "SPIDER deposition rate". In addition, atmospheric stratification and surface roughness determine the distance of the area with maximum deposition from the point source, as well as the magnitude of deposition in those areas.

(iv) Calculation of additional total $O_3$ flux and additional stomatal uptake

As mentioned above, depending on wind direction and wind velocity several sub-areas with different deposition rates above the ambient level can be found around the source position. An example is given in Fig. 4 for May 1999, the same time period used in Grünhage et al. (2000). Due to practical reasons, an $O_3$ release is proposed between 9:00 and 16:00 CET. The input parameters summarised in Fig. 2 were used for the calculation.

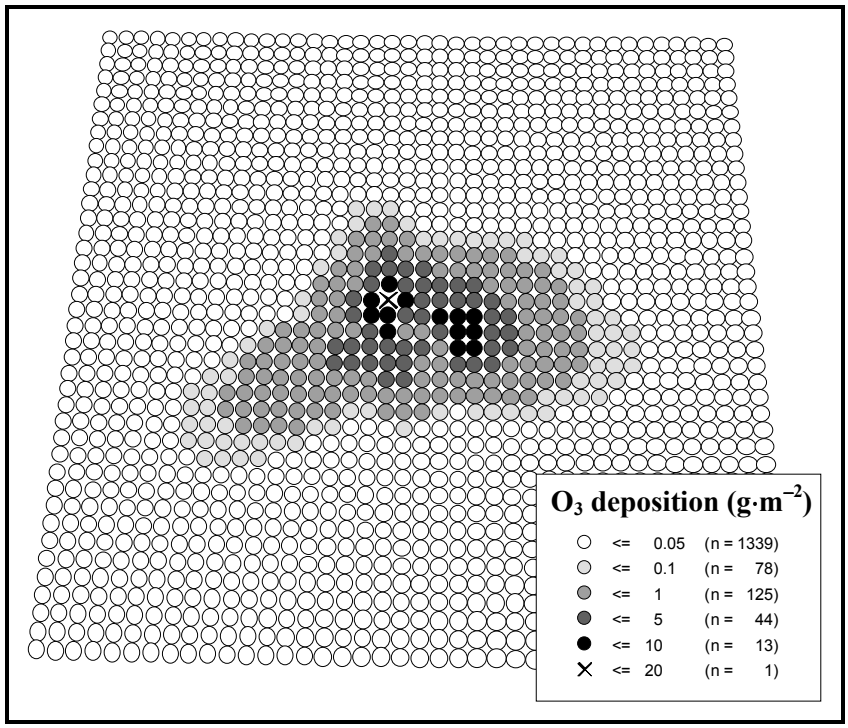


**Fig. 4:** Pattern of additional deposition due to $O_3$ injection for May 1999 at the Linden grassland (cf Grünhage et al. 1996)
(actual background deposition: 0.85 g· $m^{-2}$; sub-plot area: 20 x 20 m, height of injection: 2 m above ground; source strength: 100 µg·$s^{-1}$; exhaust velocity: 2 m· $s^{-1}$; release of $O_3$: 9:00 – 16:00 CET)

A similar partitioning of total $O_3$ flux into toxicologically effective stomatal uptake and non-stomatal deposition like for the actual background $O_3$ deposition is assumed.

A first validation experiment was conducted in August 2002 above a winter wheat field in Braunschweig, Germany. Fig. 5 shows the relative increase in $O_3$ concentration 30 cm above the canopy on August 13. According to the mean wind direction $O_3$ concentrations was increased up to 530 % above ambient level (34 ppb) near the $O_3$ point source by a source strength of 760 $\mu g \cdot s^{-1}$ and an exhaust velocity of 1.1 $m \cdot s^{-1}$.

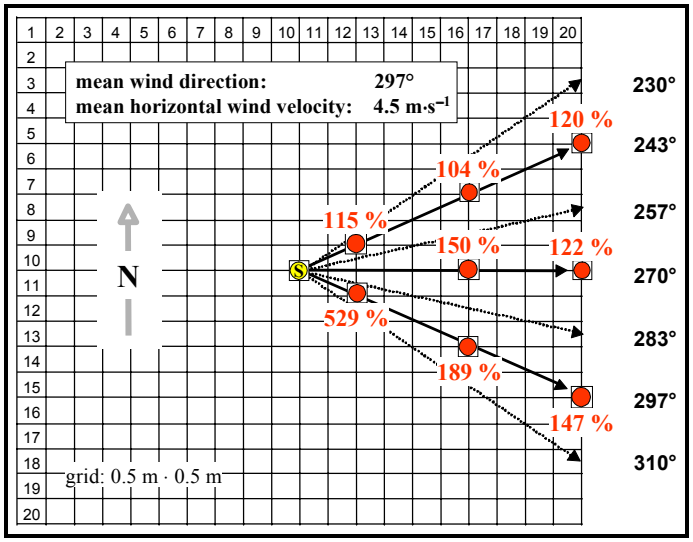


**Fig. 5:** Pattern of relative increase in $O_3$ concentration 30 cm above a winter wheat canopy downwind of an $O_3$ point source

(v) Measurements or observations of the response of the vegetation at the sub-plots with different deposition rates and derivation of a flux/dose-response relationship

(vi) Deduction of critical cumulative $O_3$ fluxes (*critical $O_3$ loads*) to protect the respective vegetation type

After validation of such a dose-response relationship *critical $O_3$ loads* can be applied for site and local scale risk assessments.

It must be noted, however, that any up-scaling to regional, national or European level is associated with an increase in uncertainty. Therefore, models for risk assessments on larger scales must be carefully calibrated by models for site scale risk assessments and not *vice versa*.

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