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Sammanfattning/Summary

This document is the final report from the workshop on "Establishing Ozone Critical Levels II", held in Hindås, 50 km outside Gothenburg, Sweden, 19-22 November 2002. It contains the overall summary of the conclusions from the workshop, the reports from the three working groups that were active during the workshop as well as all background papers presented at the workshop.

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UNECE CONVENTION ON LONG-RANGE TRANSBOUNDARY AIR POLLUTION

Establishing Ozone Critical Levels II

UNECE Workshop Report

Per Erik Karlsson, Gun Selldén and Håkan Pleijel (editors)

IVL report B 1523

UNECE Workshop in Gothenburg, Sweden, 19-22 November, 2002

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Preface

This document is the final report from the workshop on “Establishing Ozone Critical Levels II”, held in Hindås, 50 km outside Gothenburg, Sweden, 19-22 November 2002. It contains the overall summary of the conclusions from the workshop, the reports from the three working groups that were active during the workshop as well as all background papers presented at the workshop.

We believe that the results of the workshop will be a valuable source of information for many years to come and we hope that the report can be utilized for future work and negotiations under the LRTAP Convention and within the European Union CAFÉ Programme.

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The work performed by the chairpersons and rapporteurs is gratefully acknowledged.

We thank the Hjortviken Conference Centre for excellent service during the workshop.

Last, but not least, we thank all participants for their contributions and the good spirit during the workshop.

Per Erik Karlsson GunSelldén Håkan Pleijel

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Workshop Summary

Bull, K.R., Karlsson, P.E., Selldén, G. & Pleijel, H.

Introduction

The UNECE Convention on Long-range Trans-boundary Air Pollution (UNECE CLRTAP) is one of the central means for protecting the European environment from air pollution. Since its entry into force the Convention has been extended by eight protocols (www.unece.org/env/lrtap). The latest is the 1999 Gothenburg Protocol to Abate Acidification, Eutrophication and Ground-level Ozone. Its review/ revision is envisaged for 2004/2005.

The Workshop on **Establishing Ozone Critical Levels II** was held in Hindås, 50 km east of Gothenburg, Sweden, 19-22 November 2002. The workshop was attended by 95 participants from the following Parties to the Convention: Austria, Belgium, Canada, Czech Republic, Denmark, European Community, Finland, France, Germany, Greece, Italy, The Netherlands, Norway, Poland, Spain, Sweden, Switzerland, United Kingdom and USA.

There were representatives present from the UNECE CLRTAP secretariat (Radovan Chrast and Keith Bull), from WGE (Heinz Gregor, Tor Johannesen and Beat Achermann), from IIASA (Markus Amann), from EMEP (David Simpson), from the International Cooperative Programme (ICP) Vegetation (Gina Mills), from ICP Forest (Georg Krause), from ICP Modelling and Mapping (Maximilian Posch), as well as from EU DG Environment (Marion Wichmann-Fiebig). In addition, there were two participants from private enterprise, Michael Swindoll from CONCAWE (Exxon Mobile Biomedical Sciences), and Ingemar Gottberg from Engelhard Corporation.

The workshop formed another of the series of Convention workshops devoted to develop critical levels for ground-level ozone: Bad Harzburg 1988, Egham 1992, Bern 1993, Kuopio 1996 and Gerzensee 1999.

The aims of the workshop were:
- To review recent progress in the fields of ozone exposure–response relationships for vegetation and ozone exposure modelling and mapping;
- To suggest ozone exposure–response relationships for agricultural crops, natural vegetation and forest trees, based on the most recent available knowledge;
- To suggest specific changes in the text of the Mapping Manual concerning critical levels for ozone.

The meeting consisted of plenary sessions with keynote speakers, poster sessions and working group discussions.

There were three working groups:
1. Agricultural crops
2. Natural vegetation
3. Forest trees

The tasks of the working groups were:

1. To make suggestions for basic principles upon which to base new critical levels;
2. To identify which data sets that will form the basis for new critical levels;
3. To make suggestions for specific texts to be adopted in the Mapping Manual;
4. To make suggestions for principles for regional-scale ozone exposure index modelling.

Results

General.

Substantial progress was made in the development of new ozone critical levels, level II, for agricultural crops, semi-natural vegetation and forests. However, final suggestions for changes in the text of the Mapping Manual were not completed at the workshop. Different working groups were formed to continue this work in the nearest future.

The progress of this work will be presented at the following meetings:

- The ICP Vegetation Task Force meeting, in Slovenia, 27-30 January 2003.

- The TFM Modelling and Mapping meeting in Estonia, 19-23 May 2003.

- The ICP Forest Meeting in Croatia, 24-28 May 2003.

The final suggestions for changes to the text of the Mapping Manual must be submitted for approval to the twenty-second session of the Working Group on Effects in September, 2003. It is important to realise, however, that the suggested text for the Mapping Manual has to be completed well before the WGE meeting in September 2003.

In the over-all procedure of the revision of the Mapping Manual, the coordination of the structure and the care for the consistency of the critical levels chapter is done by an editorial group led by Gina Mills and the overall consistency and the technical realisation of the whole manual is done by "Federal Environmental Agency" in Germany.

Agricultural crops.

<u>Level I</u>

- New "level I" critical levels will be provided for wheat, cotton, tomato, sugar beet and potato.

- Other crops, including watermelon and pulses, may be included if the data are considered sufficiently robust after further statistical analysis.

- The level I critical levels will be the canopy height AOT40 or AOT30 (still to be decided) required for a 5% reduction in yield

- Recommended methods for conversion of ozone concentration from measurement height to canopy height will be included.

- Information on regression statistics, confidence limits, and timing windows for the different climatic regions of Europe will be included

- A table will be provided that assigns approximately 20 species of crops into broad sensitivity categories

Level II

- Level II critical levels and response functions, based on stomatal ozone uptake - yield response relationships, will be recommended for use within the Mapping Manual.

- The models will be based on the multiplicative algorithm of stomatal conductance (g_s) developed by Emberson et al. (2000) from that described by Jarvis (1976), and would be available for wheat and potato.

- It was agreed that the identification and application of a flux-threshold would need to be justified both statistically and biologically, in the former case by re-analysing ozone uptake – yield loss relationships for wheat and potato.

- It was agreed that since the purpose of Level II is to estimate effects it was not absolutely necessary to define a critical uptake/ load. However, since critical levels were developed with the aim of defining acceptable levels of air quality and to retain consistency with the AOT40 Level I approach it was recommended that the stomatal uptake-effect relationship regressions be included in the Mapping Manual with the critical stomatal uptake associated with a 5% yield loss clearly identified.

Short-term critical level

- A simple modified-AOT40 or modified-AOT30 approach, using vapour pressure deficit as the modifying factor, will be used for the revised short-term critical level.

Time schedule

- Outstanding work needed for completion of text for the Mapping Manual on ozone effects on crops has been assigned to responsible persons.

- All new work for level I is to be complete and an update on progress for level II and short-term critical level is to be presented to the ICP Vegetation Task Force meeting, in Slovenia, 27-30 January 2003.

- A first draft of the document is to be completed and ready to be presented in Estonia for consideration by the TF on Modelling and Mapping in May, 2003.

- A final text is to be presented at the twenty-second session of the Working Group on Effects for approval, September, 2003.

Semi-natural vegetation.

Level I

- The distinction between annuals and perennials, which had been proposed at the Gerzensee workshop (Fuhrer & Achermann 1999) in both the period and critical level, was removed.

- The critical level at level I for semi-natural vegetation, should be expressed as exposure index daylight AOT40, and the value should be 3000 ppb.h over a period of up to three months

- A provisional range of uncertainty between 2500 and 4500 ppb.h was suggested, which could be used for sensitivity analysis in mapping and integrated assessment exercises.

- Variable time windows should be used in the mapping procedure to account for different growth periods of annuals and perennials in different regions of Europe.

- The criteria for adverse effects remain effects on growth for perennial species and effects on growth and seed production for annual species.

- The critical level is based on growth reductions of =/>10% and refers to sensitive taxa of grassland and field margin communities.

Level II

- Currently, no flux-based critical levels can be identified for taxa of semi-natural vegetation.

- The development of flux-based critical levels was strongly favoured and it was considered that it would be possible to make significant progress towards a flux-based approach within the next five years.

- The following Level II factors are implicitly considered in the new calculations of exceedance based on the EMEP model, since they influence the modelling of total deposition and concentration at canopy height: phenology, soil moisture deficit, atmospheric conductivity, vapour pressure deficit (VPD), temperature, and management.

- The other most important factors modifying the response of plants to ozone were: community dynamics, the successional stage of a community, the canopy structure, low light (shade) conditions, nutrient status and nitrogen deposition.

The following mapping procedure was suggested:

- The time-window when the vegetation is active is first identified using European phenological models (as in the EMEP model) or local information.

- The AOT40 is calculated over the growth season if this is less than three months.

- Otherwise, the AOT40 over the first three months of the growing season is calculated for those communities in which annuals dominate, while, if perennials dominate the vegetation, the AOT40 is calculated for the three months with the highest ozone concentrations.

Forests.

- There was consensus that the effective ozone dose based on stomatal uptake (flux) of ozone represents the most appropriate approach to setting future ozone Critical Levels for forests trees.

- Uncertainties in the development and application of flux-based approaches to setting level II Critical Levels for forest trees are at present too large to justify their application as a standard risk assessment method at a European scale.

- The AOTx approach should be retained as the recommended method for Integrated Risk Assessment for forest trees at level I in the revised Mapping Manual, as a provisional measure awaiting further refinement of the ozone flux approach.

- The time window over which the AOTx is accumulated should be adapted according to local phenology.

- Both the Critical Level value and the threshold concentration above which ozone exposure (AOTx) is accumulated require re-consideration for some sensitive deciduous tree species under conditions of low temperature and high humidity.

- It is recommended that the ozone flux-based approach and the maximum permissible ozone concentration concept (MPOC) are incorporated within the Mapping Manual, representing alternative methods for Integrated Risk Assessment for forest trees at Level I.

- The inclusion of the MPOC concept into the Mapping Manual requires an update of the MPOC output with more recent literature from 1999 up until now, as well as the validation for sites from countries other than Germany.

- National evaluations should be encouraged to use and to validate the alternative approaches. These evaluations should make use of available data such as those provided by ICP-forests, and will provide experience of application to support their further development.

- The text to be added to the Mapping Manual will be suggested by a sub-group mandated by the workshop.

- An indicative range of 2-6 mmol m^{-2} projected leaf area accumulated over one growing season is proposed as a preliminary flux-based Critical Level I, for protecting sensitive forest trees from the effects of ozone. This value assumes a flux threshold of 1.6 nmol m^{-2} s^{-1}.

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Report from Working Group on Agricultural Crops

Chair: Gina Mills, UK
Rapporteur: Lisa Emberson, UK

The group comprised 25 participants representing countries with a wide geographical spread across Europe, plus Canada and the USA. All were in agreement that much progress had been made since the previous Critical Levels for Ozone Workshop, held at Gerzensee, Switzerland, in 1999. Improvements were recommended for level I and a new level II methodology was agreed upon.

Note on terminology: Many different descriptive words are used for the uptake of ozone by the leaf or canopy. Prior to a decision on standardisation, the following terms will be used in this report:

Stomatal uptake of ozone (sometimes called stomatal flux, flux or stomatal deposition).

Accumulated stomatal uptake of ozone (hourly stomatal uptake integrated over a period of time e.g. three months).

Stomatal uptake threshold (similar concept to 40 ppb in AOT40, an uptake or flux threshold above which the detoxification capacity is exceeded, and measurable responses to ozone can be detected).

Non-stomatal deposition (absorption of ozone by leaf surfaces etc, with no assumed biological effect).

Long-term Critical Level

Level I

There was general agreement that we could suggest modifications to the Level I standard that was most recently agreed at the "Critical levels for ozone – level II" meeting, held in Gerzensee, Switzerland, in 1999. These modifications would be described in the Mapping Manual with the clarification that Level I would offer users the option of applying a most simple standard that could identify the potential risk of ozone damage. However, it would be clearly stated that this standard should not be used for economic evaluation of ozone impacts.

Choice of crop and crop response
In the next few months, it will be possible to define Critical Level I standards for a number of different crop species based on an extensive review of published literature collated by Mills et al. (2002a).

This will be achieved in two ways :

1) For species for which it was felt more robust data exist, a quantitative table will be provided. This will describe critical levels (equivalent to a 5% reduction in yield), and associated regression statistics / confidence limits as a measure of the robustness of the

data. Information describing the number of cultivars studied for each species will be included. The species selected for consideration for quantitative analysis are indicated in Table 1.

2) By providing a table assigning approximately 20 species into broad sensitivity categories according to the Mills et al. (2002a) data.

Table 1: Crops for which response functions may be included in the Mapping manual

Crop	Data considered sufficiently robust	Data require further analysis	Comments
Wheat	✓		
Cotton	✓		
Potato	✓		Additional data may become available within the next 6 months
Sugar beet		✓	Selection of response parameter may require further consideration since sugar content rather than yield may be more important
Soya		✓	
Watermelon		✓	Data from only 1 experimental investigation; new data sources will be investigated. It is recommended that investigations with water melon receive further funding
Turnip		✓	This is an important fodder crop and should be further investigated in the future
Tomato		✓	Additional data may be available via the TOMSTRESS programme.
Pulses		✓	Needs further investigation since more than one species included within this group and genetic variability between cultivars may also significantly affect response to ozone

A note should be made in the Mapping Manual that the functions for these crops represent effects on yield quantity; other crop types may require different response parameters. i.e. visible leaf injury will be more important for crops such as lettuce. However, sufficiently robust quantitative information on yield quality effects is not currently available for recommendation for inclusion in the manual.

Conversion of the ozone concentration from measurement height to canopy height
The importance of applying the critical level and response functions to the ozone concentration at canopy height (i.e. the upper boundary of the quasi-laminar layer) was clearly identified within the Working Group. It was proposed that a simple system for data conversion would be provided for this purpose in the Mapping Manual. This would be an algorithm using wind speed as the driving input variable, that could be used in combination with spatially variable surface resistances derived from the EMEP deposition model.

Selection of threshold for the AOTx parameter
The current Level I, based on AOT40, is intended to "protect for the most sensitive conditions" but use of AOT40 does mean that possible contributions to a biological effect from concentrations below 40 ppb are ignored. In contrast, the stomatal uptake approach, recommended for level II, will incorporate some concentrations below 40 ppb. Although, concerns were raised about changing Level I too dramatically, it was felt in the group that we needed to offer an approach consistent with the stomatal uptake method. The response-functions described by Mills et al. (2002a), will be reanalysed using AOT20 and AOT30; a decision will be made at the ICP Vegetation Task Force Meeting (27-30 January, 2003) as to which threshold to use following consideration of the results by the Task Force.

It may also be possible to support the choice of threshold by plotting data for several species on a graph of number of days exposure versus mean ozone concentration where significant yield reductions have been detected (using the MPOC (Maximum Permissible Ozone Concentration) approach proposed by Grünhage et al., 2001). In such a figure, as the number of days of exposure increases, the mean ozone concentration will decrease; were the function to level at either 30 or 40 ppb this would provide additional support for the use of one or other of the thresholds. By including data from the Mediterranean region in the graph, the range of European crops considered would be increased, thus further increasing our confidence in the choice of threshold. It was agreed that such a function would be available for consideration at the ICP Vegetation Task Force Meeting, 27-30 January, 2003.

Different "time windows" for application of index
It was recommended that the current fixed time interval of May, June and July be replaced by user-defined timing windows appropriate to the climate and crop being considered. An Ad-hoc sub-group was formed containing representatives of the main geographical regions of Europe, to draft this part of the text of the Mapping Manual. The main growth periods for key crops will be suggested for northern Europe, Atlantic central Europe, continental central Europe, western Mediterranean and eastern Mediterranean areas. For each crop and region, a specific "time window" will be suggested with the proviso that this may change according to local agricultural management practices (e.g. irrigation, local sowing and harvesting practices).

Level II

Modified AOTx
The Working Group did not consider it necessary to suggest level II methods based on a modified AOTx approach since sufficient progress had been made with the development of ozone uptake-based response functions. One possible approach, in which ozone concentration is modified by relative uptake was considered. However, as this approach would first necessitate calculation of stomatal uptake of ozone, it would incorporate an unnecessary additional step in the calculation. Where such approaches have been attempted (e.g. by Pleijel et al. 2002), a loss in accuracy of approximately 10% has been shown to occur in the prediction of yield response to ozone.

Ozone uptake-response functions
The Working Group strongly agreed that, given further refinement of the models planned for the next few months, level II critical levels and functions based on stomatal ozone uptake - yield response relationships could be recommended for use within the Mapping Manual. The models

would be based on the multiplicative algorithm of stomatal conductance (g_s) developed by Emberson et al. (2000) from that described by Jarvis (1976).

Choice of crops
Four different response relationships were considered in terms of whether stomatal uptake-based relationships improve on the existing AOT40 functions. These were: wheat, potato, grape and clover.

<u>Wheat</u>
It was agreed to recommended that a stomatal uptake - yield response relationship replace the AOT40 relationship for risk assessment application for wheat. This relationship is based on experiments performed over a period of 9 years with 4 cultivars and in 3 countries (new data from Italy is expected to be added shortly). The wheat stomatal uptake-effect relationship suggested by Pleijel et al. (2002) was derived from a different algorithm from that which is currently used in the EMEP model to estimate stomatal deposition to this cover type (Emberson et al., 2000), referred to as the EMEP/Emberson model in the following text. The following work will be performed to further strengthen the uptake-response relationship and to ensure consistency between the EMEP/Emberson algorithm used to calculate deposition and the Pleijel model used to calculate stomatal uptake-response.

The main differences between these two algorithms are:

1) g_{max}
There was large variability in the values obtained from the literature to derive the EMEP/Emberson model parameterisation. These values require further investigation to assess their suitability for inclusion in deriving g_{max}. This investigation will be based on statistical (no. of g_s measurements including published g_s values) as well as experimental considerations (e.g. instruments used to measure g_s, conditions under which measurements made, provision of information describing the leaf area and gas for which conductance is measured). It was agreed that it may be beneficial to re-visit the literature (especially to search agronomy journals) to find new data.

2) g_{VPD}
There was some variation in the g_{VPD} parameterisation defined in the Pleijel and EMEP/Emberson g_s models. This is predominantly due to the methods of derivation (i.e. boundary line analysis versus step-wise increments in the VPD parameter). Further investigation of the data used to derive the function will be performed to agree on a single g_{VPD} function for use in both models .

3) g_{SWP}
The feasibility of using the EMEP/Emberson model parameterisation for gSWP needs to be assessed since no strong relationships for this parameter exists in the Pleijel model. New data from Ben Gimeno and colleagues may be available early in 2003 from exposure of wheat to ozone in well-watered and drought-stressed conditions, which might be useful for the derivation of g_{SWP}.

4) Time of day and O_3 function
Both these functions exist in the Pleijel model but not in the EMEP/Emberson algorithm. It will be investigated as to whether they are necessary for inclusion in the EMEP/Emberson model by performing additional runs of the Pleijel model with modified parameterisation.

In all cases described above it will be important to define the levels of certainty associated with each of the parameterisations.

Potato

Ozone uptake-effect relationships were considered for potato based on experiments conducted over two years in four countries using one commonly grown cultivar. The r^2 values for the regression of the potato flux-effect relationship were lower than those for the wheat relationship. This is likely to be an artefact of the experimental system in that fewer potato plants can be grown in an open-top chamber compared to wheat (<50 as opposed to c.1,000), meaning that fewer plants are harvested per replicate, exaggerating inter-plant variation in the mean yield per replicate. Nevertheless, the Working Group was confident that a stomatal uptake - yield effect relationship for potato should be included in the Mapping Manual.

Grape

A stomatal uptake-yield response relationship has been derived for grape based on an experiment conducted at one site in Austria over a period of four years (Soja et al. 2002). This experiment provides clear evidence of a carry-over effect of ozone exposure in one season on the yield response in subsequent seasons. The AOT40 parameter performed equally well if not better as compared to cumulative stomatal uptake when related to the different response parameters investigated (grape yield, sugar content), although it should be noted that no different uptake thresholds were considered in the analysis of these data. The Working Group agreed that these were important results, but that a function for grape could not be included in the Mapping Manual because the experiment was only performed at one site in Europe, and the uptake algorithm was not parameterised for local conditions (or evaluated against such parameterisation) with the exception of derivation of g_{max}.

White clover

A stomatal uptake - biomass effect model has been developed for white clover using data from the ICP Vegetation monitoring programme (see Mills et al. 2002b). This has been developed via parameterisation of the multiplicative model using g_s data collected by ICP Vegetation participants from 9 countries. Stomatal uptake is currently calculated for the ozone measurement height of 3m; a transfer function for conversion of ozone concentration to that at the canopy height will be applied before subsequent analysis. Nevertheless, provisional modelling indicated that stomatal uptake appeared to perform better at predicting reductions in biomass (expressed as the biomass reduction in a sensitive compared to a resistant biotype) than the AOT40 index with r^2 values for regression analyses of 0.32 versus 0.57 respectively.

The Working Group agreed that as the white clover biotype system was originally intended to serve as bioindicator of the effect of ambient O_3 concentrations across Europe, it would not be appropriate to include a stomatal uptake-response function based on this system within the Mapping Manual. However, it was also agreed that this unique long-term investigation may provide useful information for supporting, in relative terms, estimates of ozone impacts using stomatal uptake-effect relationships on other crops (in particular those that are irrigated). Indeed, the modelling of the white clover data has already shown that increased stomatal uptake equates to increased damage in ambient air (as opposed to open-top chamber) conditions and thus is supportive of the concept of using a stomatal-uptake based index in response studies.

Non-stomatal deposition

It was noted that cuticular deposition to the leaf was not included in the derivation of the Pleijel stomatal uptake-effect model. Although at the leaf scale this term is not considered as important as published data would suggest it is at the canopy scale, it is advisable to re-calculate the flux-effect model allowing for cuticular deposition to the flag leaf. This can be done using the parameterisation for non-stomatal deposition currently used in the EMEP deposition model. It was also noted that care should be taken over reference to non-stomatal deposition, this should NOT be referred to as cuticular uptake but rather as loss to the cuticle since the former could add confusion concerning the term for policy makers.

Detoxification thresholds

It was agreed that the identification and application of a flux-threshold would need to be justified both statistically and biologically. Biological justification appears to be strong, given the current understanding of the mechanism of internal plant defence mechanisms to instantaneous O_3 absorbed doses. Pleijel et al., 2002, have also performed comprehensive statistical analyses of data for both wheat and potato that strongly supports the use of a flux-threshold. However, some concern was raised as to whether this statistical justification of a threshold was a result of using a particular parameterisation of the g_s algorithm. This concern largely resulted from a previous study published by Pleijel et al. (2000) where using the EMEP/Emberson g_s parameterisation for wheat the relationship between stomatal ozone uptake and yield loss was stronger when no threshold was used. In view of this it was agreed that the parameterisation for both wheat and potato should be reviewed by pooling all available datasets. Once this parameterisation has been agreed the resulting stomatal uptake model will be re-run with both the wheat and potato data and the statistical investigations be re-done using the "new" stomatal uptake data to determine whether a threshold is necessary. It was considered especially important to ensure the use of a threshold is appropriate since the thresholds under consideration (e.g. 4 nmol m^{-2} s^{-1}) would result in approximately a third of the total cumulative ozone uptake over a growing season being discounted in estimates of biological impact.

Additional recommendations for the application of stomatal uptake-based critical levels

- It was agreed that since the purpose of Level II is to estimate effects it was not absolutely necessary to define a critical uptake/ load. However, since critical levels were developed with the aim of defining acceptable levels of air quality and to retain consistency with the AOT40 Level I approach, it was recommended that the stomatal uptake-effect relationship regressions be included in the Mapping Manual with the critical stomatal uptake associated with a 5% yield loss clearly identified. The inclusion of the regression function would allow policy makers to determine varying levels of damage. However, the Working Group wished it to be noted that the 5% value relates to a loss in productivity; and will not necessarily translate into a loss of production which will be dependant upon management practices such as fertilizer input, irrigation etc. As such it was suggested that the term "yield loss" should be replaced with "productivity loss".

- It is recommended that the application of stomatal uptake-response functions, particularly for performing economic assessments, should be performed in consultation with the scientific community. This would ensure that all necessary factors would be included in the modelling and mapping process and could most easily be achieved through communication with the ICP Vegetation.

- It was felt that before the response functions for wheat and potato could be recommended for use across the whole of Europe, confirmation of their applicability should be obtained from scientists working with ozone effects on crops in the Mediterranean (possibly through Ben Gimeno, a member of the ICP Vegetation Steering Committee) as the cultivars used are more typical of northern Europe. It is hoped that new uptake-effect data for wheat from recent experiments conducted in Italy and Spain with local cultivars, will become available for use prior to the drafting of the Mapping Manual.

- Estimates of the degree of certainty and confidence should also be made with respect to application of the flux-response functions by listing and prioritising the uncertainties. Application of the functions should also be supported by sensitivity analysis of the EMEP model. Since these will change over time (as new information becomes available) it was suggested that a web site be established to which users could be directed to obtain information on the current development of the model and associated uncertainties.

- Irrigation should be taken into account when applying the function. This should be possible since the land-cover map used by EMEP to calculate ozone deposition includes the presence/absence of irrigation facilities though assumptions as to the use and effectiveness of such facilities would have to be made. Such assumptions should be noted on consideration of the fact that potato is almost always irrigated whereas wheat irrigation is more variable.

- In areas where the soil is saline (e.g. in some Mediterranean areas), the functions should not be applied since soil salinity will affect g_s in a manner that would not be modelled by the existing algorithm.

- Response parameters can be viewed as either physical (e.g. effects on biomass, yield etc…) or chemical (e.g. effects on sugar, protein, starch content). To date, ozone uptake based indices for wheat and potato have been established for yield effects, however, a number of investigations on these crops have shown both positive and negative effects of ozone on chemical quality. Since such effects may be important for economic assessments of ozone impacts on crops it was agreed that these additional effects caused by ozone should be highlighted, although they could not be quantified at this stage.

Short-term Critical Level

The Working Group agreed that the short-term critical level should be maintained with the specific purpose of identifying the risk of visible ozone injury. This is considered especially important both for providing evidence of effects of ozone on vegetation, and also since it poses a real threat to commercial losses in value of certain crops for which appearance is particularly important (e.g. leafy crops such as lettuce, spinach etc…).

It was recognised that it would be unlikely that emission reductions targets would be set to protect for the short-term critical level. However, the existence of a short-term level would enable evaluation of the effect on incidences of visible injury to be determined from the reductions initiated for protection of the longer term level.

It was agreed that the short-term critical level be modified from that which currently exists in the Mapping Manual. A modified AOT40/AOT30 approach (using a g_{VPD} function) should be used, although a more detailed stomatal uptake algorithm may be available within the next year. Whichever approach is used, the Working Group thought that it should be kept relatively simple so that it is easy to apply on a local scale. It may be necessary to more specifically define the response in terms of the degree of injury suffered (e.g. percentage of leaves injured).

It was proposed that additional data from the Mediterranean region could be presented in the form of an MPOC dose-response relationship to show the large number of commercial crops and cultivars at risk of ozone injury in this area. This could be achieved by plotting the number of days to visible injury against the associated ozone concentration, and against the modified-AOT40 or modified-AOT30.

The potential uses of the short-term critical level should be well defined in the Mapping Manual to avoid misuse.

Priority actions and recommendations for further research

Action List – work to be completed within the next 6 months by members of the Working Group

Timetable

The work described in Table 2 will be completed according to the following schedule:

ICP-Vegetation Task Force Meeting, Slovenia, 27-30 January 2003:
All new work for level I to be completed.
Update on progress for level II and short-term critical level to be presented.

February and March, 2003:
Drafting of text for Mapping Manual

End of April, 2003:
Second draft of document to be completed ready to be presented to the TFM Mapping and Modelling for consideration.

September, 2003:
Final text to be presented at the WGE Meeting for approval.

Table 2: Outstanding work needed for completion of text for the Mapping Manual on ozone effects on crops

Action	**Person(s) responsible**	**Notes**
Level I		
Response functions and stats (4 to 8 crops)	Gina Mills (and Ron Smith)	Further data input from Maria Sanz and Ben Gimeno (tomato, water melon)
Timing intervals (by crop & region)	Ludwig De Temmerman	
O_3 conversion function	Ludger Grünhage	
Crop sensitivity table	Gina Mills	
MPOC function for validation of AOTx threshold	Ludger Grünhage	
Level II		
g_{max}	Lisa Emberson & Håkan Pleijel	Wheat & potato
$g_{VPD} + g_{SWP}$	Lisa Emberson & Håkan Pleijel	Wheat & potato
Other g functions	Lisa Emberson & Håkan Pleijel	Wheat & potato
Non-stomatal deposition / Uncertainties	Juha-Pekka Tuovinen	Wheat & potato
Threshold	Håkan Pleijel, Giacomo Gerosa & Jeremy Barnes	Wheat & potato
New effect data	Ben Gimeno and Giacomo Gerosa	Wheat
Recommendations for future use	Allan Legge & Gina Mills	Wheat & potato
Uncertainty / sensitivity analysis	Ron Smith	Wheat & potato
Short-term critical level		
Modified AOTx function	Håkan Pleijel & Gunilla Pihl Karlsson	Clover
MPOC function for visible injury	Maria Sanz & Ludger Grünhage	Mediterranean species

Research recommendations for the next five years

Further evaluation of flux modelling

- Perform stomatal conductance measurements at micrometeorological flux measurement sites and in free air fumigation systems for use in the further development of up-scaling procedures (from the leaf to the canopy)
- Modelling at different spatial scales – this should allow for fine tuning of regional models using information from local site-specific modelling
- Quantification and process understanding of non-stomatal losses (i.e. non-stomatal deposition)

For all the above, Mediterranean agro-ecosystems should be especially targeted since these are currently least well represented by model evaluation investigations

Ozone uptake-response relationships

- Investigate and develop canopy uptake-response relationships using free air fumigation systems
- Investigate effects on quality (e.g. forage quality of pasture)
- Increase the number of species for which robust flux-response relationships exist (particularly focussing on grape, sunflower, soybean, pulses and tomato) by further experimentation
- Investigate mechanisms and quantification of species-specific detoxification capacity
- Given the expected future decrease in peak ozone concentrations but increase in background ozone levels, further ozone exposure experiments should be performed to investigate the effects of these pollutant profiles on crop species.

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Report from the Working Group on Semi-Natural Vegetation

Chairman: Mike Ashmore
Rapporteur: Jürgen Franzaring

A. General remarks

New scientific findings from ozone fumigation experiments were presented in the working group sessions, which confirmed the rising interest in potential adverse effects of photo-oxidants on the biodiversity of semi-natural vegetations. Since the 1999 Gerzensee workshop (Fuhrer & Achermann 1999), when critical levels for semi-natural vegetation were last considered, most research has continued to be dedicated to central and western European herbaceous plant species. The recent experiments are generally performed under more realistic conditions, because plant communities or mesocosms are being studied instead of single plants and because field fumigation studies are also in progress.

For the time being, most of the studied wild plant species are from grasslands, while annual agriophytes (ruderals) also represent a group of species which have extensively been used in fumigation experiments. This has implications when mapping the exceedance of critical levels for semi-natural vegetation (see section D). The WG noted the paucity of information on the response to ozone of taxa from many types of community and many regions of Europe and considered priorities for future research to address these gaps (see section E) .

The possibility of predicting which plant communities might be sensitive to ozone, in the absence of experimental data, was considered by the WG. Addressing the issue of plant functional types, plant ecological groups and plant traits of wild plant species, it was noted that these may be used to understand why some taxa are more sensitive to ozone than others. There was good agreement in the WG that the legume family (Fabaceae) contains many taxa responding strongly to ozone. The WG thus suggested that plant communities with a high proportion and high cover of legumes are potentially sensitive to ozone, and proposed to identify the relevant grassland communities (see section D). However, overall the conceptual framework for using autecological information to identify sensitive taxa and communities needs to be further developed, and translated into mapping applications.

B. Level I

The WG carefully reviewed the new experimental data and the implications for the critical level, taking into account the criteria for a significant adverse effect. In addition to the new data discussed in the background paper on semi-natural vegetation (Fuhrer et al.), the group identified some additional datasets of value. The group chose to reject some estimates of thresholds for growth effects in the background paper (e.g. the data of Danielsson et al., 1999 for a range of Phleum genotypes), due to high scatter in the critical region of the exposure-response relationship. It was agreed by the WG that the critical level, expressed as exposure index **AOT40**, should be **3000 ppb.h over a period of up to three months**, the value currently used for annuals. The distinction between annuals and perennials, which had been proposed at the Gerzensee workshop (Fuhrer & Achermann 1999), in both the period and critical level, was removed. The criteria for adverse effects remain effects on growth for perennial species and effects on growth and seed production for annual species. The major justification for the revised critical level for perennials was new data from the BIOSTRESS programme, which showed that early-season exposure of perennial species can have significant effects on growth over the whole growing season. This important finding also means that critical AOT40 values for the growing season which are derived from experiments of less than three months duration can be based on the fumigation period only. The critical level is based on growth reductions of =/>10% and refers to sensitive taxa of grassland and field margin communities.

The WG considered the uncertainty in this estimate of the critical level, as, unlike for crops, it was not possible to combine data for the same taxa. Therefore, it was felt useful to provide an indication of the range of estimates of the critical level to protect the most sensitive taxa. The WG suggested a provisional range of uncertainty between 2500 and 4500 ppb.h, which could be used for sensitivity analysis in mapping and integrated assessment exercises. The lower estimate is based on the study of Gimeno et al. as part of the BIOSTRESS programme; the group agreed to re-analyse new data after the workshop before agreeing on the upper limit (see section E). Some concern was expressed within the group that some, but not all, of the studies used to define the critical level and the uncertainty range have not yet been peer-reviewed.

Variable time windows should be used in the mapping procedure (see section D) to account for different growth periods of annuals and perennials in different regions of Europe. It was also noted that the AOT40 calculation for comparison with the critical level should in the future be based on modelled concentrations at the top of the canopy.

The WG was not aware of any evidence for semi-natural vegetation that other exposure indices (AOT30, AOT20) were more suitable than the AOT40 index, as most of the relevant experiments had only considered AOT40. It is not clear if it would be possible to re-analyse the key experimental data to derive values of the critical level using AOT20 and AOT30.

C. Level II

Currently, no flux-based critical levels can be identified for taxa of semi-natural vegetation. However, the WG strongly favoured the development of such approaches and considered that it would be possible to make significant progress towards a flux-based approach within the next five years (see section E). The MPOC (maximum permissible concentration at the canopy top; Krause et al.) approach was not seen as a useful alternative for semi-natural vegetation, because there was concern over pooling data on different species and end-points, as well as the lack of a cumulative component.

The BIOSTRESS project provided results from several groups that confirmed earlier studies that the impacts of ozone are enhanced by competition in mesocosm experiments (Fuhrer et al., Bender et al.,). Information on BIOSTRESS is available at http://www.uni-hohenheim.de/biostress. However, despite efforts being made by several research projects on the effect of ozone on semi-natural plant communities, no general view has emerged with regard to the effect of photo-oxidants on complex communities. It was noted that the presence of leguminous taxa in a plant community enhances its overall sensitivity to ozone. Apart from species composition, the genetic variability of taxa (genera, species, varieties, ecotypes) was also identified as an important factor determining the plant´s response to ozone, e.g. the different sensitivity of provenances and genotypes of, and within, species of the genus *Centaurea* (Bungener et al.).

The WG considered the list of modifying factors of significance for Level II applications in the Mapping Manual and agreed that these needed to be revised. It was noted by the WG that the following Level II factors are implicitly considered in the new calculations of exceedance based on the EMEP model, since they influence the modelling of total deposition and concentration at canopy height:- phenology, soil moisture deficit, atmospheric conductivity, vapour pressure deficit (VPD), temperature, and management. The other most important factors modifying the response of plants to ozone were agreed by the WG to be:- community dynamics, the successional stage of a community, the canopy structure, low light (shade) conditions, nutrient status and N-deposition. Several of these important factors are likely to influence vegetation response through factors other than leaf conductance, and thus a more comprehensive mechanistic approach is needed to identify species and communities as risk.

D. Mapping

The WG discussed approaches and possibilities in mapping critical levels of ozone. Because most of the information available is from grassland species, attempts should be made to locate grasslands in existing European databases on vegetation and land use (e.g. those offered by CORINE and SEI) and merge these maps with the EMEP model. The classification of the European Nature Information System, EUNIS (refer to http://mrw.wallonie.be/dgrne/sibw/Eunis/), may be used for the preliminary identification of those grassland types across Europe for which the critical level is supported by experimental data, and for the identification of grasslands possessing high proportions of legumes and thus at high risk from ozone. EUNIS is already the agreed basis for empirical critical loads of nitrogen and mapping critical levels of ozone may be further developed on the basis of this system in the future.

The scheme in Figure 1 gives an overview on the mapping procedure suggested by the WG. The time-window when the vegetation is active (see Level I, section B) is first identified using European phenological models (as in the EMEP model) or local information. The AOT40 is calculated over the growth season if this is less than three months. Otherwise, the AOT40 over the first three months is calculated for those communities in which annuals dominate, while, if perennials dominate the vegetation, the AOT40 is calculated for the three months with the highest ozone concentrations.

E. Requirements for future developments / recommended future research

The WG discussed requirements for the refinement of critical levels. One prerequisite in the short-term is the publication of scientific findings of current research projects and the re-analysis of existing data-sets in order to re-check the range of uncertainty in the existing critical level. Other work to be performed by members of the WG is the identification of potentially sensitive grasslands using the EUNIS classification and the decision on which land cover maps to use. Other members of the WG agreed to co-operate in improving the parameters of the existing EMEP ozone grassland deposition model (which currently is relevant to productive grasslands of northern Europe) so that it can be used for a wider range of communities.

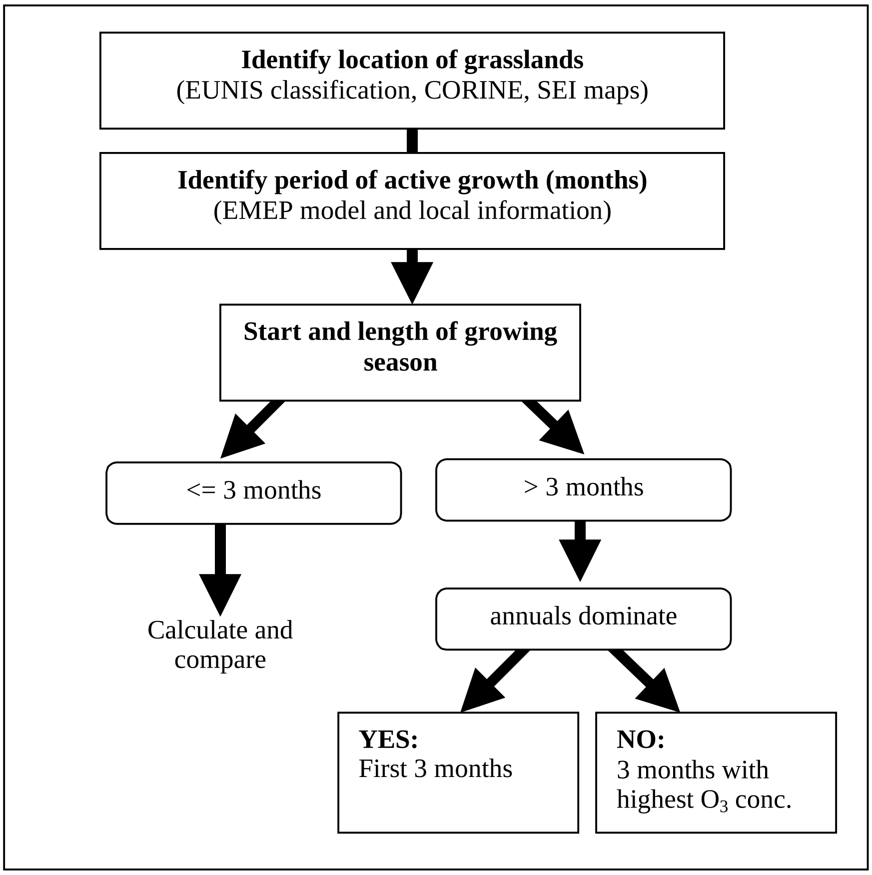

Figure 1. Mapping application for the identification of semi-natural plant communities potentially at risk from ozone (for explanation, see section D).

In the long term (i.e. 4-5 years), the development of a flux approach seems possible in principle for semi-natural plant communities. The first stage will be to develop and test models of total stomatal flux to a wider range of plant communities. A number of appropriate micro-meteorological datasets were identified for refining/testing the EMEP model parameters for these communities. Further micro-meteorological investigations, over a wider range of communities and climatic zones, are needed to extend the range of available field data on ozone flux. Additional work will be needed, including data on stomatal/non-stomatal uptake of ozone in individual species and mixed communities across Europe, if the models are to extended to model stomatal flux to individual species, rather than the whole canopy.

The members of the WG noted the urgent need for much more experimental data to support the development of a critical level approach for ozone. These data should be applicable to a wider range of semi-natural communities across Europe. Large integrated experimental programmes will be needed in the coming years, in which field and mesocosm fumigation experiments, as well as gradient studies, should be performed.

The working group agreed on the urgent need to acquire information on the response of a much greater range of taxa, and discussed specific priorities. It was agreed to be important to obtain more information on the sensitivity to ozone of herbaceous taxa from forest floors and forest edges, for which evidence of visible injury in the field had been obtained (Godzik et al., Ferretti et al.). Experiments relating to eastern European herbaceous plant species and ecosystems are generally lacking, despite studies being performed there on identifying forest species and ecosystems sensitive to ozone in the field. Work on forest floor and forest edge species could be developed with participants of the ICP Forests, who already are recording visible injury to such species. More information from the Mediterranean zone is urgently needed, as only the studies of Gimeno et al., on dehesa, and Elvira et al., Paoletti et al., and Inclan et al. (1999) on shrub species, report responses other than visible injury. Species of arctic and upland regions also need study.

Bioindication methods for ozone and standardised experiments (e.g. transplant studies) should be established and set up at different sites in Europe using monoclonal plants (e.g. *Centaurea*). Such approaches and standard tools using native wild species will help to reduce the biological variability seen in ozone experiments and will assist in refining critical levels for ozone for Europe. ICP Vegetation should co-ordinate these activities and efforts should be made to include participants from eastern and southern European countries in these programmes.

The database already developed by ICP Vegetation for experimental data on semi-natural vegetation allows rapid analysis of information for characteristics related to ozone sensitivity. The WG recommended that this database should be further developed in any future ICP Vegetation programme and used to further assess if plant characteristics, such as specific leaf area ratio and growth rates, as well as ecological strategies, are related to ozone sensitivity. Such experimental databases should be linked to information on species composition in different communities within the EUNIS classification, as methods of identifying and mapping sensitive communities must remain a focus of future work.

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Report from the Working Group on Forests

***Chairman*: Antonio Ballarin-Denti,** *Catholic University of Brescia, Italy*
***Rapporteur*: Elina Oksanen,** *University of Kuopio, Finland*

In establishing ozone critical levels, at Level II, for forests in Europe, one of the main difficulties will be to determine the impact of ozone on mature trees under field conditions. Recent data from dendro-ecological measurements for beech in Southern Germany (see background paper by Dittmar, Elling & Lorenz), for Norway spruce in south Sweden (see background papers by Karlsson et al. and Kivimäenpää et al.), for beech and Norway spruce growing along an ozone pollution gradient in Switzerland (see Braun et al., 1999 and background papers by Braun et al.), and for beech in Italy (see background paper by Ferretti et al.) provide evidence that current ambient ozone concentrations in Europe represent a risk to forest trees.

Data from the Scandinavian region indicate that a lower AOT40 index value (AOT40 ~ 5 ppm.h) is needed to protect the most sensitive tree species, such as birch, in northern Europe, under low VPD conditions and short growing seasons (see background papers by Karlsson et al., Oksanen, Kivimäenpää et al., Manninen, Miettinen & Huttunen). In addition, data from Braun et al. for beech indicate that critical levels should be lower in certain low VPD areas of Switzerland. On the other hand, the current critical level may be too low for some tree species under certain Mediterranean site conditions (see background papers by Paoletti et al., Elvira et al.). Therefore, the current critical level based on AOT40 (the level I approach) needs revising, because mapped exceedances may provide an incorrect assessment of the risk of damage to vegetation by ozone in some areas of Europe.

A flux-based risk assessment method for ozone impact offers the potential for significant improvements over the AOT40 approach, because it quantifies the internal dose of ozone received by plants. Progress has been made towards a first application of flux-based critical levels at a European scale for indicator tree species including birch, beech, oak, and Norway spruce. However, for most tree species and regions it is not yet possible to move from the daylight AOT40 ozone index to a cumulative uptake index, due to a lack of dose-response data sets, as well as uncertainties in the parameterisation and validation of the stomatal flux models for specific species. However, it should be noted that species-specific exposure-response relationships using the AOT40 index are also only available for a limited number of tree species and do not distinguish between different climatic conditions.

The development of an ozone flux approach is still in progress and many research issues remain. In addition, non-stomatal deposition to the cuticle, as well as leaf de-toxification properties have not yet been taken into account in flux based indices. A stepwise procedure is suggested to enable a Level II estimate of ozone critical levels for forests to be made. Developing ozone uptake-biomass response relationships from experimental data with young trees represents the first step towards the establishment of Level II ozone critical levels for ozone impacts on forests. We then need to generate more information on environmental and plant internal factors that modify ozone uptake. Furthermore, there is evidence of a difference in sensitivity between adult and young trees. Therefore, more information is needed on ozone impacts on adult trees (The Kranzberg Forest Experiment, see background paper by Matyssek et al, or epidemiological observations, see Braun et al., 1999 and background paper by Karlsson et al.).

To date, results from re-calculated data and comparisons between AOT40 and cumulative ozone uptake indices do not indicate that a flux-based approach represents an advantage over the AOT40 approach in explaining growth responses (see background papers by Braun, Remund & Flückiger, Karlsson et al.). This may be a result of small ranges in some of the climatic factors that modify stomatal ozone uptake in these experiments, particularly soil moisture deficit. Further refinement and validation of the ozone flux approach needs to be continued on the basis of more comprehensive data sets.

The current uncertainties in the development and application of flux-based approaches to level II critical levels are at present too large to allow their application as a standard risk assessment method at a European scale, within the planned timescale for the next stage of policy assessment. Nevertheless, work on the ozone flux approach must proceed in order to base, on the long term, risk assessment on ecologically meaningful cause-effect relationships.

The three different approaches that are currently available (AOTx, MPOC, and the flux-based concepts) were discussed in greater detail by three *ad-hoc* groups formed at the workshop.

1. AOTx (Accumulated Exposure Over Threshold x)

Limitations of the AOTx approach have been recognized in terms of climatic representativeness and species specificity. In addition, the relatively high threshold of 40 ppb in the AOT40 index may result in uncertainties in estimating exceedances, especially in northern Europe

The group discussed the following questions: What are the arguments for keeping an AOTx? Should we change the recommendations on for example: (a) Time window, (b) Critical level, (c) Threshold concentration

The advantages of AOTx are: 1. Historical data are available in this format, and recalculation using an alternative index could be problematic. 2. Relatively easy to calculate from active monitoring data. 3. Low data requirement, meteorology is not needed.

Recommendations:

- The time window over which the AOTx is accumulated should be adapted according to local phenology.
- For Nordic countries, a lower threshold of 20-30 ppb may be relevant. Alternatively, the current AOT40 critical level value could be lowered for certain sensitive deciduous trees species under low temperature and high humidity conditions.
- A rigorous, scientific re-evaluation of existing data addressing the issues listed above is necessary and will be carried out by a sub-group mandated by this workshop.
- The results of the work by the sub-group will be submitted to the ICP Mapping and modelling Task Force meeting in May 2003.

2. MPOC concept (Maximum Permissible Ozone Concentration)

A concept of risk assessment based on an exposure-response relationship and applied to ambient air concentration measurements has been provided in the background paper of Krause, Köllner & Grünhage (MPOC concept). The data points illustrated in the MPOC dose-response diagram is a qualitative measure of the observed effect. In other words, different plant species with different intensities of response are represented. The MPOC output represents a variety of significant plant responses to ozone exposure, including visible injury, growth and biomass effects. Furthermore, the MPOC output allows classification of the probability for adverse plant response to ozone exposure as one moves from the green (no risk) over the yellow (some probability of negative effects) to red (highest probability of risk) area (See Figure 2 in background paper by Krause et al. and Grünhage et al. (2001).

There is some uncertainty in the exposure-response relationships because of the range of the indicator responses assessed and the short time frame of some of the experiments. Furthermore, information is largely lacking for tree species from the Mediterranean climate and, to date, the concept has only been validated for sites in Germany. Since the MPOC analysis was based on literature up until 1999, it is necessary to update the MPOC output with more recent literature from 1999 up until now.

Recommendations:

- It is necessary to update the MPOC output with the more recent literature from 1999 up until now.
- Countries should be encouraged to validate this framework at a national level following discussion of the concept within ICP-Forests.
- The next step in the improvement of risk assessment using MPOC is to develop a true probability scale (e.g. 10%, 20%, etc. risk of occurrence to a certain ecosystem at a local scale).
- The MPOC approach currently uses a 10% margin of error. Additional work with the same sets of data augmented by more recent observations coupled with a sensitivity analysis can assist in numerical predictions of risk

3. Flux concept

Assessing an effective ozone dose to vegetation based on stomatal ozone uptake now seems to offer a practical and physiologically sound basis for determining ozone effects. The development of a concept based on flux based dose-response relationships should therefore be pursued with the intention of future implementation. Initially this will follow a precautionary principle for sensitive trees in terms of structural and functional responsiveness. In time, this concept will be extended to a range of genotypes and species. There is consensus that this dose-based approach should be the basis of future work.

The approach combines ozone levels and meteorological data from regional and continental models (e.g. EMEP), or from measurements, with a parameterisation of stomatal conductance using accepted algorithms accounting for phenological, climatic and edaphic factors. Important data inputs, which need to be established, include land-use characteristics (leaf area index, stand height, species present, soil moisture retention capacity), and the parameters used in the stomatal conductance model. Further evaluation of the methodologies and data availability for different regions of Europe is required – both the parameterisation of the stomatal flux model (EMEP) and the derivation of the dose-response relationships. In particular, data for Mediterranean species is lacking and may require further data collection.

While AOT40 is based upon empirical observations, flux-based dose-response relationships have a functional basis for explaining ozone effects. This approach allows the combination of avoidance (uptake - stomata) and tolerance (repair and defence mechanisms) with respect to ozone susceptibility.

A preliminary, indicative range of critical ozone uptake of 2-6 mmol m^{-2} projected leaf area, accumulated over one growing season using an hourly flux threshold of 1.6 nmol m^{-2} s^{-1}, is proposed with respect to the induction of ozone effects on tree function and structure, based on the background paper by Karlsson et al.

Conclusions.

There was consensus that the effective ozone dose to forest trees, based on stomatal ozone uptake (flux), should be implemented for future ozone critical levels for forests.

The AOTx approach should be retained within the mapping manual as the recommended method for Integrated Risk Assessment (Level I approach), as a provisional measure as long as the ozone flux approach will need further preparatory refinement prior to practical application. The time window over which the AOTx is accumulated should be adapted according to local phenology. The threshold value for the AOTx, alternatively the value for the AOT40, should be re-considered for certain sensitive deciduous tree species under low temperature and high humidity conditions.

It is recommended that the two new approaches, flux-based concepts and MPOC, are incorporated within the Mapping Manual, representing alternative methods for Integrated Risk Assessment for forest trees at Level I. National evaluations should be encouraged to use and to validate the new approaches to gain experience and to support their further development, making use of data such as those collected by ICP-Forests.

The text to be added to the Mapping Manual will be suggested by a sub-group mandated by the workshop.

Research and monitoring recommendations

It is recommended that research is encouraged in order to further improve the parameterisation of the different stomatal conductance regulating factors (the g-factors) used within the EMEP model deposition module.

ICP Forests is encouraged to continue its monitoring of ozone injury at ICP Forest Level II sites throughout Europe.

An evident need was stated for increasing the number of Level III sites to promote methodologies applicable to level II approaches.

Subgroup

A sub-group was mandated by the workshop to carry out further evaluation of available experimental and field data.

The subgroup should carry out the following tasks:

1. Re-evaluate the threshold used for the AOTx concept, as well as the critical level value, with a special focus on sensitive tree species in geographic regions with a climate characterised by a low VPD.

2. To further develop the MPOC concept regarding an update to include datasets after 1999, as well as a validation of the concept for countries other that Germany.

3. To further analyse data sets available mainly for young trees, to improve the dose-response relationships based on cumulative uptake of ozone (CUO), especially with regard to the adopted g_{max} values and g_{temp} functions and the question of threshold setting for modelling ozone flux. The aim should be to provide definitive values, or a range of values, that can be used to set new ozone critical levels based on CUO for sensitive trees species in terms of structural and functional responsiveness. These values should reflect the current state of knowledge of the CUO levels which affect mature trees.

4. The group should eventually suggest new text for the Mapping Manual.

The outcome of the work of this subgroup, including updated values and text for the Mapping Manual, will be submitted to the ICP Mapping and Modelling Task Force Meeting in May 2003.

The work of the subgroup will be co-ordinated by Per Erik Karlsson, IVL Swedish Environmental Research Institute.

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Introductory remarks

Måns Lönnroth, Managing Director, MISTRA Foundation, Sweden

Dear friends and colleagues,

I appreciate very much the opportunity to give a few introductory remarks at your workshop ”Establishing ozone critical levels II”. This event is an important one, perhaps crucial, in the continuing evolution of what I think is the by far most successful of all international environmental agreements. The UNECE Convention on Long Range Transboundary Air Pollution serves as a model and an inspiration for other similar activities around the world.

I would like to take the opportunity to place this event at Hindås in its perspective. ASTA is funded by MISTRA, the foundation for strategic environmental research. MISTRA was set up in 1994 by the Swedish government with the mandate to fund research that can contribute to solving environmental problems. Our annual budget is some 25 MUSD, taken out of our endowment of, today, some 320 MUSD. MISTRA funds a small number of large scale programmes, each one of which running up to eight years. Our guiding metaphor is the Death Valley. MISTRA-funded research has to cross the Death Valley between research and application. This is a valley full of brilliant ideas that never got anywhere, due to a lack of dialogue between scientists and practitioners. A MISTRA-program, should, on the other hand, make a difference in the world of action.

ASTA is one in a group of programmes aimed at a rather specific type of bridging of the Death Valley: where science serves as input to international environmental agreements. ASTA was recently evaluated after a first four-year period. The international scientific panel was unusually enthusiastic and recommended strongly that MISTRA should fund another four year period. The MISTRA board decided accordingly.

Let us now put the LRTAP convention in perspective. We should start with a simple observation: efforts to control regional air pollution in Europe are now well into the fourth decade. I for one believe that there will be a fifth one as well. Success needs time. And, as we shall see, it is the simultaneous action on and interaction between three arenas that determine success. The three arenas are: science, politics and mitigation. The first decade started in 1969 and ended in 1979. It concerned the right to put the issue on the political agenda. The decade ended with the signing of the convention in 1979. The politics had its roots in east-west relations, Willy Brandt’s ostpolitik, the Helsinki agreement and so on. Politics was the most important arena, together with some science.The second decade started in terms of politics with the Soviet invasion of Afghanistan in 1979 and ended in 1989 with the collapse of the Soviet Union hold of eastern and central Europe. Two crucial events on the political arena of fundamental importance for transforming the environmental problem into a manageable one. In terms of science, politics and mitigation:

- *The science arena*: The crucial concept of critical loads was launched - successfully.
- *The political aren*a: the first flat rate reduction protocols (in particular the first Sulphur protocol from 1985) and the first EU directives, e.g. the directive on large combustion plants were agreed to, after first Germany and later the UK changed their minds.
- *The mitigation arena*: the European electric utility industry and the European automobile industry accepted grudgingly the need for de-sulphurisation and catalytic converters.

The key significance of the second decade is that learning started in earnest. A virtuous circle of action and learning started to move. Somewhat hesitantly but nevertheless.

The third decade saw this virtuous circle gather speed. The decade started with the democratisation of central and eastern Europe which in turn led to serious participation in the convention. The decade ended with the signing of the Göteborg protocol. In terms of our three arenas science, politics and mitigation:

- *Science* has evolved. Look in your book-shelves for the documents we used to write around 1990 and compare them with the sophistication of the multi-source, multi-effect Göteborg protocol.
- *Politics* has also evolved. The expansion of the EU from twelve to fifteen members as well as the modernisation of central and eastern Europe is a fundamental change. Geneva and Brussels are now working together with the active analytical support of Vienna.
- *Mitigation* has evolved. The automobile industry is no longer dragging its feet, and the resistance of the European oil and refinery industry has been broken. The automobile industry has been a valuable ally in forcing the oil industry into accepting high quality fuels.

The third decade ended with the virtuous circle in full speed. The environmental quality is improving, signs of acidification recovery are observed, nitrogen oxide depositions are declining. The problems that weighed heavily during the first and second decade are now improving. At the same time other problems emerge.

So let us speculate about the fourth decade, again in terms of science, politics and mitigation.

- *Science.* The scientific base is changing. The human health aspects are rising on the agenda, in particular the effects from particles but also from ozone and nitrogen oxides. The key concept of the second decade - critical loads - may be replaced by something else that measures the dynamics of recovery. And I would hope that Europe’s cultural heritage would move up the agenda. I for one regard it as a tragedy that buildings and artefacts that have survived for millenia, like the rock carvings in Scandinavia, Acropolis and Trajan’s Colonne as well as the glass windows of the medieval cathedrals still are being sacrificed for economic short-sightedness. A revised concept for ozone effects is also part of the new agenda and I am looking forward to ways to handle ozone effects that are smarter than those used for the Gothenburg Protocol.
- *Politics*, The politics should be reasonably stable, with the EU expanding into some 24 members. The revision planned for 2005 could well be followed by yet another revision around 2010. Countries with large scale agriculture damages and bad city air quality should become crucial. Revising the present ground level ozone criteria AOT40 and taking into account climatic differences across Europe will probably give added political weight to Europe north of the Alps.
- *Mitigation.* Combustion engine technology and fuel quality will move to the top. The automobile industry will dominate, but other combustion engine sources will be drawn into the area.

Will there be a fifth decade? I should think so. One key question is whether the limits of the existing Otto and Diesel engines are being reached or not. The revision planned for 2005 will in all probability have to be revisited in perhaps 2010. Perhaps hybrid systems will be necessary, something like the present Toyota system.

Ground level ozone is already a defining element in the revisions during the fourth decade. A possible fifth decade will probably have to take back ground levels emanating from semi-hemispherical transport of ozone into account. It is quite possible that a fifth decade will widen, both the scientific, the political and the mitigation arenas. A possible fifth decade will also inevitably interact with the measures necessary for the second commitment period of the Kyoto protocol.

So, dear friends, you should not be mistaken. The weight on your shoulders is no small weight. This workshop is an important building block towards the revision of both the Göteborg protocol and the EU clean air directives. Ground level ozone will be one major driver in the efforts to stoke the political pressure for further efforts to reduce air pollution.

Let me make a final remark on the role of science in the formation of environment policy. ”Science gives nature a voice”. This is a nice way of formulating the role of science. Science gives nature a voice in the rooms of decision making, where the voices of business and the existing order of interests otherwise reign. Science and monitoring of effects on human health and the environment are the type of arguments that can be used by governments to force otherwise reluctant economic interests to change their way of investing and in the longer run their policies of technological development. Believe me - I have listened to the discussions. But this transformation of science into policy is not automatic. Scientists frequently despair of the lack of action despite what seems to be incontrovertible evidence. Let me point at two issues which scientists themselves can control.

The first issue concerns what one could call explanatory concepts. Scientific results have to be translated into language that governments can use in legislation. Such concepts inevitably lose the richness and the nuances of scientific knowledge. They often seem crude, at best, or plain wrong, at worst. But they are nevertheless necessary. The concept of critical load is an obvious case. It can and has been critiqued as vague, inexact or worse. Nevertheless it has been highly effective in mobilising the will to act. It is easy to explain and easy to comprehend. A non-expert politician can explain it to a lay man public or a journalist without much problems.

The second issue concerns the need for scientific consensus - a contradiction in terms. Science progresses on disagreement. Dispute is at the heart of scientific discourse. Nevertheless, science in policy making has to be essentially consensual. Dissent breeds inaction. The more open the dissent, the stronger the tendency of the powerful to postpone action. The climate issue is a perfect example, at least in the US. Scientific dissent is particularly problematic in international negotiations. Heads of delegations tend to listen to scientists from their own country and disregard arguments from heads of delegations from other countries - if the respective scientists disagree. Science is much more national than one would like. Problems discovered by scientists from other countries tend to exist only when confirmed by scientists from one’s own country All the more important, therefore, are meetings like these. Fleshing out areas of agreement and areas of disagreements including explaining why are of direct importance for eventual policy.

Dear friends and colleagues, it has been a privilege to give this opening address. I wish you all luck for this workshop.

Thank you for your attention

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PAST, PRESENT AND FUTURE OZONE-RELATED ACTIVITIES UNDER THE CONVENTION ON LONG-RANGE TRANSBOUNDARY AIR POLLUTION

Radovan Chrast (UNECE secretariat), Keith Bull (UNECE secretariat) and
Heinz-Detlef Gregor (Chairman of the Working Group on Effects)

Introduction – the past

The 1979 Convention on Long-range Transboundary Air Pollution (CLRTAP) has long recognized the effects of ground-level ozone as a pollutant and the long-range transport of its precursor gases. Already in the 1988 Protocol on Nitrogen Oxides (NO_x) there is reference to "secondary products" of NO_x (in the preamble), while the 1991 Protocol on Volatile Organic Compounds (VOCs) makes direct reference to ozone and photochemical oxidants throughout its text. However, these early protocols made no attempt to link directly emission reductions by Parties to the effects of ozone, or indeed concentrations of NO_x or VOCs.

Following the successful adoption of the effects-based, 1994 Oslo Protocol on Further Reduction of Sulphur Emissions, the Convention through its subsidiary bodies worked towards a similar effects-based, but multi-pollutant, multi-effect approach that more effectively dealt with emission controls, in particular those linked to the various polluting mechanisms involving NO_x. These activities resulted in the Protocol to Abate Acidification, Eutrophication and Ground-level Ozone that was adopted in Gothenburg (Sweden) on 30 November 1999. It was signed by 31 Parties to the Convention and, as of 15 October 2002, has been ratified by 4 of them. It will enter into force when there are16 ratifications.

By addressing simultaneously acidification, eutrophication and ground-level ozone, the 1999 Gothenburg Protocol is the most comprehensive and sophisticated of the protocols under the Convention. It is also the first of them that specifically addresses the control of ozone pollution.

The basis of the effects-based approach used for the Protocol was the bringing together of quantitative information on effects (critical loads and levels), emissions (from reported information), pollutant loads and levels (calculated from emissions using atmospheric transport models) and costs of abatement technology (described in cost curves for emission abatement for each country). Integrated assessment models used all of these data to develop cost-optimal scenarios that identified maximum benefits to the environment and human health. For such work there was a need for close collaboration between subsidiary bodies under the Convention, in particular, the EMEP Steering Body and its centres, The Working Group on Effects and its International Cooperative Programmes, the Task Force on the Health Aspects of Air Pollution, and the Working Group on Strategies and its Task Force on Integrated Assessment Modelling.

Effects and critical levels – past and present

In developing the 1999 Gothenburg Protocol, the Executive Body recognized the harmful effects of ground-level ozone both to human health, to materials and to trees and other plants. It noted that ozone affects lung function, particularly in children and asthmatics, causes leaf injury in plants, including crops and trees, thus significantly reducing plant growth and crop yield, and causes damage to some, in particular to organic, materials such as paints, rubber and plastics.

However, only limited knowledge and data were available on effects (dose/response) of ozone at the time when the Protocol was negotiated. Hence, relatively simple critical levels of ozone (so-called level I critical levels) were used for integrated assessment modelling, even if it was recognized that they provided only an approximate solution. The simple approach also enabled the integrated assessment modellers to devise a means for using these critical levels in their models – an essential requirement for such an effects-based approach.

The level I critical levels for ozone for vegetation were initially defined on the basis of experimental data from open-top chambers without considering the influence of environmental factors, such as soil moisture or nutrient supply. They were expressed as an AOT40 exposure index (accumulated exposure over the threshold concentration of 40 ppb (parts per billion)) and related to the ozone concentrations measured at the top of the canopy. Figures were used to evaluate the potential damage of ozone to crops, trees and semi-natural vegetation. This was done by comparing actual levels of ozone with the critical levels, i.e. estimating the exceedance of critical levels. The level I approach was attractive because of its simplicity, but it did not consider any biotic or abiotic factors that might influence a plant's sensitivity to ozone and might thus determine the real damage to vegetation. To use a more sophisticated approach in 1999 was not feasible mainly because: (i) scientific knowledge was not sufficiently advanced to provide reliable values; (ii) databases required for the calculations were not available; and (iii) the means for incorporating a level II approach into an integrated assessment model did not exist.

Effects and critical levels – the future

The Executive Body was well aware of the simplistic nature of the level I critical levels used for the 1999 Gothenburg Protocol. This was highlighted by the Working Group on Effects which further indicated that future work, beyond 1999, would be focussing on improving the methodology and working towards a level II approach. This would more accurately assess the impacts caused by the exceedance of critical levels for ozone and provide essential inputs for better economic evaluation of damage due to ozone.

Such a level II approach for vegetation should consider the influence of environmental factors on ozone uptake and effects, for instance by considering the transport of ozone from the atmosphere to the site of action inside leaves and needles. It should also consider differences in the response to ozone between species, and the response of plant communities, which cannot be represented by the individual responses of species present in a mixture.

In 1999, several options for a level II approach were identified at a workshop in Gerzensee, Switzerland. These included:

a) The modification of the level I critical levels for ozone;
b) The modification of the AOT40 values calculated to express the exceedance of the Level I critical levels for ozone using modifying factors;
c) The development of a flux-based approach which addresses the ozone uptake and toxicity of the absorbed ozone dose.

There was general agreement [in all Gerzensee working groups] that the development of a flux-based approach should be the long-term goal of a level II approach, but that many difficulties in its realizing at a larger scale for several receptors types might make it necessary to apply interim solutions, such as the modification of the AOT40 values calculated to express the exceedance of the Level I critical levels for ozone. The direct modification of the level I critical levels was not considered a useful approach since it would imply full re-analysis of the exposure-response part of the Level I approach.

To develop a method based on ozone flux-response relationships, there have been proposals to use a flux model composed of atmospheric, boundary layer, stomatal and non-stomatal resistance components. Since stomatal uptake of ozone by a plant cannot be measured directly, a receptor-specific micrometeorological parametrization is needed. Some details are provided in

EB.AIR/WG.1/1999/12; Proceedings of the workshop: Critical Levels for Ozone – Level II, Gerzensee, Switzerland, April 1999 and the report of the Harrogate expert meeting, June 2002.

The timetable for the future

The Executive Body for the Convention at its nineteenth session (2001) decided “to schedule the review of the Gothenburg Protocol, in view of its expected entry into force in 2003, to commence in 2004” (ECE/EB.AIR/75, para. 69 (a)). The Working Group on Effects at its twenty-first session (2002) approved the updated medium-term work-plan for the further development of the effect-oriented activities, containing, inter alia, the following tasks (EB.AIR/WG.1/2002/4, para 8):

(i) Further develop the ozone flux-effect model for clover, including validation with data from the 2001 and 2002 seasons, and finalize flux-effect models for wheat and potato;

(ii) Establish new (level II) or revised (level I) critical levels of ozone for crops, semi-natural vegetation and trees (at the workshop on establishing ozone critical levels II, November 2002).

The ad hoc expert panel meeting on the methodology for modelling ozone flux and deposition held in June 2002 (Harrogate, United Kingdom) noted that a flux approach, once further developed, would substantially improve the quality/reliability of ozone critical levels. It was suggested that the forthcoming workshop on level II critical levels for ozone (November, Gothenburg, Sweden) should assess possible approaches (e.g. modified level I, level II - fluxes), evaluate advantages and risks/shortcomings of their respective application and propose practical solutions, that are also applicable to integrated assessment modelling.

While the Working Group on Effects at its twenty-first session expressed its satisfaction with the progress already achieved, it also stressed the need for further elaboration of a practical and credible approach for deriving critical levels of ozone (EB.AIR/WG.1/2002/2, paras. 29-32). To this end Mr. M. Amann, representing the Centre for Integrated Assessment Modelling, stressed that, while a flux approach would be a great improvement for the understanding of ozone effects on plants, a number of important questions still remained to be answered. Necessary data have to be gathered and models developed.

Conclusions

Taking into account the timescales for development of integrated assessment models and economic assessments in readiness for the review of the Gothenburg Protocol in 1994, it is essential that decisions are taken now with regard to a practical way ahead for developing and applying critical levels for vegetation. Not only must these reflect current understanding of ozone effects on plants but also they must be applicable to a range of vegetation types across the different climates of Europe. In addition, they must be consistent with transport model outputs and must lend themselves to integrated assessment modelling and economic assessment.

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How Well Can We Model Ozone Fluxes? A Report from the Harrogate Ad-hoc Expert Panel Meeting on Modelling and Mapping Ozone Flux and Deposition to Vegetation

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Abstract

This paper provides an overview of the conclusions and discussion points from the Expert Panel meeting held in Harrogate in June 2002. The meeting concluded that flux-based risk assessment methods for ozone offer for the potential for significant improvements over the AOT40 approach, and that we are now at the stage when the first application of flux-based critical levels on a European scale is possible. The meeting also concluded that it would be possible at the Gothenburg workshop to agree on the first set of flux-response relationships for a small number of major crop and tree species. However, the discussion also identified a number of important limitations and uncertainties in the approach which need to be addressed in a longer-term programme to develop a stronger basis for European scale application of flux-based risk assessment.

1. Introduction

In planning this workshop in Gothenburg, the organisers were aware that flux-based approaches offered one important mechanism of moving towards Level II critical levels for ozone. Since flux is not measured directly in most situations, but is derived from appropriate models, it was clearly important that the quality of the available flux models was assessed prior to the workshop. Furthermore, for flux-based models to be applied in pan-European risk assessment, it was also important to evaluate the availability of appropriate regional scale modelling schemes, and the associated spatial databases, to simulate stomatal flux as part of the total ozone deposition to vegetated surfaces. The Expert Panel meeting, held in Harrogate in June 2002, aimed to address these issues prior to the Gothenburg workshop.

In this paper, I will present the most important conclusions of the Harrogate Expert meeting and some of the key issues raised in the discussions. I will then provide a personal view of the implications of the discussions at Harrogate for this workshop in Gothenburg, and for the implementation of flux-based risk assessment of ozone in a policy context, both in the short- and longer-term.

2. Summary of Key Conclusions

The meeting produced a formal two-page summary of its conclusions, which is included as an annexe to this paper. The full summary report of the meeting can be found at http://www.york.ac.uk/inst/sei/APS. There is also an extensive set of background papers available from the meeting.

Three key conclusions emerged from the workshop:-

(a) Although the AOT40 index has provided a useful indicator of the potential for ozone damage to vegetation, it is likely that the current definition for the critical level for ozone, based on AOT40 for a fixed time interval (the level I approach), provides an incorrect assessment of the regional distribution of the risk of damage to vegetation by ozone across Europe;

(b) Flux-based risk assessment methods offer the potential for improved quantitative evaluation of the impacts of ozone on vegetation across Europe and should be recommended for future application within the Convention;

(c) The deposition and stomatal flux algorithm now implemented within the EMEP photochemical model provides an adequate basis for first estimation and application of flux-based critical levels.

Therefore, a key conclusion of the Harrogate meeting was that we were now in a position to move forward with European flux-based risk assessments. However, the more detailed discussion and recommendations from within the three working groups at the meeting provides a clearer picture of the current status of ozone flux and deposition models, and, in particular, their limitations and the priorities for future research and development.

3. Working Group Discussions and Recommendations

Three working groups were formed to address different aspects of flux and deposition modelling. I will consider in turn the issues identified and the conclusions reached in each of these groups.

3.1 European scale deposition (modelling tools and data requirements)

Initial model runs using the EMEP model (Simpson *et al.*, 2002) clearly demonstrate that the spatial variation of modelled flux to grasslands and crops across Europe is quite different from that of AOT40. This conclusion is consistent with that reached by Emberson *et al.* (2000), using a simpler model framework. Hence the issue of whether to define critical levels in terms of AOT40 or flux is not a trivial academic one – it has major implications for assessment of ozone impacts across Europe. Therefore, this group emphasised that flux models can be used to assess damage to vegetation across Europe in a more realistic manner, and could be used to strengthen socio-economic evaluations. The major weaknesses identified were the difficulty of validating both the stomatal and non-stomatal components of the model, and the low spatial resolution. Key priorities for development of the model in the short-term were identified as improved data on leaf area index, evaluation of land cover datasets, quantification of model uncertainties, further validation of model predictions against field data, and testing of methods for sub-grid modelling.

This working group also made a number of recommendations that are significant for longer-term application of the flux approach. These included:- systematic identification of existing flux datasets for model evaluation; the development of a network of stations within the wider EMEP monitoring network for monitoring dry deposition of ozone and other pollutant gases; encouragement of national flux modelling for inter-comparison with the EMEP model; and efforts to harmonise terminology to communicate the methods more widely to policy makers and the general public.

3.2. Stand-level deposition assessments (quantifying total and stomatal components)

This group considered how well the EMEP model, and other similar models, were able to simulate total deposition and stomatal flux to different types of vegetation, based on micro-meteorological methods. The broad conclusion was that the deposition velocities predicted by the EMEP model were consistent with the measurement data, but that stomatal conductance and flux was more difficult to validate and showed greater variability between model predictions and measurements. The broad conclusions of Working Group 1 - that more comparisons with existing field data and new monitoring networks specifically designed for flux monitoring were needed - were supported, but with some more specific recommendations. A range of existing datasets for validation for different forest species and for natural vegetation were identified, but for crops only data for wheat, soybean and maize were identified. Temperate crops and grasslands were suggested as priorities for future flux monitoring, alongside a greater focus on southern European systems. The importance of better understanding of processes within the canopy, especially in forest stands, was emphasised, as was improved understanding of the fate of ozone on plant surfaces, as recent data show external plant surfaces to be a more important ozone sink that was implied in earlier studies.

3.3 Modelling stomatal conductance and flux effects

This group concluded that there is a good degree of confidence in the flux modelling approach and that for selected species, flux-based critical levels could now be identified. A major focus of the discussion was on identifying those species for which flux-response relationships could now be derived. For crops, the conclusion was to focus on wheat, potato and clover for analysis prior to the Gothenburg workshop. However, the potential for extending this to other crops, such as water melon, bean and tomato was identified, and it was suggested that closer collaboration with the US groups represented at the meeting could extend the range of data available. For forests, six species were identified for which parameterisation of the Emberson *et al.* (2000) flux algorithm might be possible – oak, Norway spruce, birch, beech, Scots pine and Aleppo pine. However, the effects data available all related to sapling studies, and the issue of extending flux-response relationships to mature trees required much further work. For semi-natural vegetation, it was concluded that it is not possible at this stage to derive flux-response relationships. The group concluded that there is a sound physiological basis for using a threshold in calculating accumulated flux over time, and emphasised that flux thresholds derived from theoretical considerations and models should be considered alongside statistical derivations from experimental flux-response data.

There was considerable discussion within this group as to whether the multiplicative flux algorithm developed by Emberson *et al*. (2000) was the most appropriate for flux-effect modelling. It was concluded that the algorithm worked quite well, given its limitations, but that additional terms might be needed, for example for time of day effects because the algorithm tends to overestimate conductance in the afternoon, and for cumulative ozone effects on conductance. Parameterisation is another very important issue. The Emberson *et al.* (2000) algorithm uses parameters derived from literature review, which are employed within the EMEP model to simulate regional-scale deposition and stomatal flux. However, other parameter values may provide better fits to particular experimental datasets. Indeed, there is evidence that the flux-response relationships derived from experimental data may be dependent on the parameters selected for the flux model (Pleijel *et al.*, 2002). Likewise, for species which are widely distributed across Europe, there may be systematic spatial variations in key parameters which need to be considered. In general, there are important issues to be resolved to ensure that there is consistency, or the potential for inter-calibration, if the flux-effect relationships derived from particular experimental datasets are to be linked rigorously to the fluxes predicted across Europe within the EMEP model.

This group also considered alternative approaches to modelling stomatal conductance and flux which should be considered for application in the longer term. One approach would be the use of artificial neural networks (ANNs), but these require large databases for derivation. A more attractive alternative is the Ball *et al.* (1987) conductance algorithm, which has been used in some US modelling programmes. This algorithm is more mechanistic, being based on modelled photosynthetic rates, and is likely to give better predictions for a given dataset; however, it may be more demanding in terms of data requirements for regional-scale modelling.

4. Implications for this Workshop

The Harrogate expert panel meeting provides a very clear signal from deposition and flux experts that we are now in a position where flux-based risk assessment for ozone impacts on vegetation can begin to be applied. It is therefore important that the Gothenburg workshop can provide at least first estimates of flux-response relationships and flux-based critical levels for a limited number of species. It is possible that the current level of uncertainty in flux models and the proposed critical levels will be judged such that these flux-based Level II approaches will not be applied in the formal negotiations within UN/ECE expected from 2004 (see below). However, flux-based critical levels or flux-response relationships recommended from this workshop are absolutely essential to complete the information needed to begin working with this approach, and hence to develop and improve the flux approach for wider application in the policy domain. It will also be very important that the flux models and parameterisations used in deriving these relationships or critical levels are fully documented in the conclusions of the workshop.

5. Linking Flux Estimates at Different Scales

A key issue in applying the flux-response relationships which will be discussed at this workshop is that of scaling from data on individual experimental leaves or plants through site- or local-scale risk assessments to European-scale risk assessment (Grunehage, 2002). The issues in terms of local or European scale assessment relate primarily to the level of detail in the model, the data input requirements and the availability of appropriate parameterisations. A key need is to identify which factors lead to greatest uncertainties in modelling site-specific deposition. For example, Tuovinen *et al.* (2001) showed that the default biological parameterisations used in the EMEP model caused systematic errors in predicting stomatal flux to three specific forest canopies in Scandinavia. However, the use of the regional-scale meteorological input data used by the EMEP model, rather than local data, did not cause significant errors. Tuovinen *et al.* (2001) also emphasise that regional-scale models, such as the EMEP model, can only ever aim to simulate biological processes reasonably well in most cases, with little bias but possibly with large scatter. Of more significance for discussions at the Gothenburg workshop are the issues of scaling from experimental data to models of deposition or stomatal flux to vegetation canopies. These scaling issues can be addressed using standard resistance analogue models to relate, for example, deposition to a vegetation canopy modelled as a single layer using the big leaf approach (as in the EMEP model) to stomatal flux to a specific leaf within the canopy (Grunehage *et al.*, 2000). For example, Grunehage (2002) used the wheat yield-flux relationship of Pleijel *et al*. (2000), which is based on stomatal flux to the flag leaf only, with a model to up-scale from leaf to canopy flux, to estimate the relative yield loss of wheat at an agricultural site in Germany. Hence, in principle such scaling is possible, although robust testing of the methods proposed to relate flux estimates at different scales is required.

6. Implications for Integrated Assessment Modelling

A number of concerns have been raised subsequent to the Harrogate meeting about the application of the flux approach in integrated assessment of the costs and benefits of emission control. These have been most clearly articulated in a short discussion paper by Amann & Schopp (2002). This paper identifies some key missing information that will be provided at the Gothenburg workshop, such as the existence of a threshold flux, but also identifies some more fundamental concerns. These concerns need to be clearly addressed in order for progress to be made in developing an effective approach to flux-based risk assessment. Here, I briefly consider some of the key issues raised by Amann & Schopp (2002), distinguishing two different questions:-

(i). Are there fundamental differences between the flux/deposition approach and the AOT40 approach which make the former more difficult to apply in integrated assessment modelling or less acceptable to policy makers?

(ii). Are the current uncertainties such that we need a further period of development and testing of flux-based approaches before they are applied?

In terms of (i), a number of issues have been raised. Firstly it has been suggested that fluxes should not be used as policy targets, because they are not directly measurable quantities and are less transparent to policy-makers. However, modelled estimates of deposition of nitrogen and sulphur have been routinely used in integrated assessment over the last two decades, despite the same problems existing for the dry deposition component. Indeed, the revised version of the EMEP model being used to model deposition of ozone is based on an integrated deposition module including sulphur dioxide, nitrogen dioxide and ammonia. The key issue therefore may rather be the need to improve the extent of monitoring of dry deposition of ozone, to provide a stronger basis to test the validity of the model.

A second series of issues relate to the underlying spatial data-sets, and the need for sub-grid modelling. These do not seem to be fundamental problems, as ecosystem-specific flux estimates will be provided, as irrigation data is currently available for use with the model, and as simulations could in theory incorporate other components of sub-grid variation. A third argument has been that derivation of vegetation-specific fluxes creates difficulties because different source-receptor relationships have to be derived for each vegetation type. However, methods have been developed to handle ecosystem-specific variation in critical loads of nitrogen and sulphur and their exceedance, using cumulative frequency distributions within grid squares (Hettelingh *et al.,* 2001) and it would seem possible to adapt these to deal with ozone deposition. However, there is one crucial factor which is different for ozone – the need to calculate flux over different periods for different vegetation types and locations. These periods may be quite different within a grid square (e.g. irrigated and non-irrigated crops in Mediterranean areas); in contrast, annual deposition is used for sulphur and nitrogen. It is important to note, however, that other Level II approaches, based on continued use of AOT40, which are under discussion at this workshop, would also lead to the need to calculate ecosystem-specific exceedances over different time periods, and hence this issue is not unique to the flux approach.

However, in terms of (ii), it is clear from the preceding discussion that a number of important issues need to be resolved and investigated further Hence it may well be that the current uncertainties in the development and application of flux-based approaches to Level II critical levels are too great to allow their application within planned timescales for the next stage of policy assessments. It is nevertheless crucial that the Gothenburg workshop provides information and recommendations which can be applied over the coming years in further developing and testing flux-based risk assessment, so that, over an appropriate timescale, this approach can replace AOT40. In practice, this may mean parallel development of the two approaches over the next few years.

Acknowledgements
The successful organisation of the Harrogate meeting resulted from the hard work of three people: Jenny Duckmanton, Lisa Emberson and Monica Machen. We are also grateful to the UK Department of Environment, Food and Rural Affairs for their support of the meeting under contract EPG 1/3/173.

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ANNEXE

CONVENTION ON LONG-RANGE TRANSBOUNDARY AIR POLLUTION

Summary Report

Joint ICP Vegetation/EMEP Ad-hoc Expert Panel Meeting on Modelling and Mapping of Ozone Flux and Deposition to Vegetation, Harrogate, UK; 16-19 June 2002

1. The meeting was organised to assess the current scientific status of methods to model and map the deposition and flux of ozone to vegetation, in the context of their possible application in revised critical level assessments for ozone under the review of the Gothenburg Protocol, provisionally planned for 2004/5. Specific objectives of the meeting were: (i) to evaluate the models of deposition and flux proposed for application in the EMEP photochemical model; (ii) to assess the ability of these models to predict stomatal and non-stomatal deposition of ozone; (iii) to review progress in modelling flux to vegetation in experimental studies in order to derive flux-based critical levels; (iv) to identify key uncertainties in flux and deposition models; and (v) to assess whether flux-based approaches should be recommended for the development and application of a level II methodology for critical levels of ozone within the Convention.

2. Experts in all relevant fields were invited from Europe and North America. The meeting was attended by 37 experts from the following 12 Parties to the Convention: Belgium, Denmark, Finland, Germany, Italy, Netherlands, Norway, Spain, Sweden, Switzerland, United Kingdom, and the United States of America. The Chairpersons of the Working Group on Effects and ICP Vegetation attended, and EMEP and the Coordination Centre for Effects were also represented. A list of participants is attached as Annexe 2 to this report.

3. The first day of the meeting primarily involved presentations in plenary. For the rest of the meeting, three working groups were formed, to discuss:

(1) European scale deposition (modelling tools and data requirements);

(2) stand level deposition assessments (quantifying total and stomatal components); and (3) modelling stomatal conductance and flux-effect relationships.

The reports of these three working groups, which were approved by a final plenary session, form Annexe 1 to this summary report.

4. Three key conclusions were reached by the three working groups, and were approved by the final plenary session:

(a) Although the AOT40 index has provided a useful indicator of the potential for ozone damage to vegetation, it is likely that the current definition for the critical level for ozone, based on AOT40 for a fixed time interval (the level I approach), provides an incorrect assessment of the regional distribution of the risk of damage to vegetation by ozone across Europe;

(b) Flux-based risk assessment methods offer the potential for improved quantitative evaluation of the impacts of ozone on vegetation across European and should be recommended for future application within the Convention;

(c) The deposition and flux algorithm now implemented within the EMEP photochemical model provides an adequate basis for first estimation and application of flux-based critical levels.

5. It was concluded that it would be possible to use flux-response relationships to derive flux-based critical levels for consideration at the workshop on level II critical levels to be held in Gothenburg in November 2002. It was agreed to focus effort on wheat, potato and clover as crop species and on spruce, pine, beech and oak as major tree species. Agreement was reached on the form of the mathematical algorithms to be used to calculate flux when deriving flux-response relationships and critical levels.

6. The presentation from EMEP demonstrated that first tests of the new deposition algorithm within the EMEP photochemical model produced realistic estimates of ozone concentrations. The EMEP model also provides estimates of deposition velocity which are consistent with measurement data, although it was recognised that more intensive evaluation was needed. Results from the EMEP model show large differences between the spatial distribution of AOT40 and modelled stomatal flux across Europe, demonstrating that the choice of flux rather than AOT40 will significantly influence the results of pan-European risk assessments.

7. Each of the working groups recognised that there are many significant uncertainties in the flux modelling approach which need to be resolved, although these need to be assessed alongside the known uncertainties and systematic errors in the current level I AOT40 approach. Each working group considered these uncertainties in the context of work which might be possible in the next six months, and longer-term objectives.

8. In the next six months, in preparation for the Gothenburg Workshop, the key priorities identified were: (i) improvement and evaluation of the land cover database, including the description of leaf area index; (ii) further quantification of model uncertainty and sensitivity; (iii) further testing of deposition module performance, especially for wheat and grasslands; and (iv) improved parameterisation of the stomatal algorithm for flux-effect modelling of high priority species.

9. An additional set of activities was identified to further evaluate and develop the flux-based approach over the next 2-5 years. The major priorities identified were: (i) development of methods for sub-grid and national flux modelling which can be linked to the EMEP model; (ii) further development of the land-cover database; (iii) development of a flux monitoring strategy within the EMEP network to improve data available for model validation; (iv) intensive evaluation of the EMEP model, and especially the stomatal flux term, using micrometeorological datasets; (v) improved parameterisation of non-stomatal components of the deposition module, including more mechanistic understanding; (vi) extension of flux-effect analysis to more crop and forest species, and possibly to semi-natural vegetation; (vii) comparison of the empirical Jarvis multiplicative algorithm used in modelling stomatal conductance in the current EMEP model with alternative more mechanistic algorithms; (viii) improved modelling of soil moisture and vapour pressure deficit effects on stomatal conductance; and (ix) further evaluation of critical fluxes for effects.

In Establishing Ozone Critical Levels II (Karlsson, P.E., Selldén, G., Pleijel, H., eds.). 2003. UNECE Workshop Report. IVL report B 1523. IVL Swedish Environmental Research Institute, Gothenburg, Sweden.

Introducing Ozone Fluxes in Integrated Assessment Modelling

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Much progress has been achieved in modelling the flux and deposition of ozone to vegetation. The summary report of the ICP Vegetation ad-hoc expert panel meeting at Harrogate, UK (June 16-19, 2002) recommends future application of flux-based risk assessment methods within the Convention on Long-range Transboundary Air Pollution.

The present note presents some initial considerations about how flux-based assessment methods could be used in the context of integrated assessment modelling and thus could contribute to future negotiations about emission control strategies under the Convention in a quantitative way.

The scope of integrated assessment modelling

Integrated assessment models such as the RAINS model perform two main functions:

- For a given emission scenario, they can quantify the environmental impacts (the ‘scenario analysis’ mode), and
- For a user-defined environmental target, they can determine cost-effective emission reductions (the ‘optimization’ mode).

The optimization mode has been extensively used in the negotiations for the Gothenburg Protocol to Abate Acidification, Eutrophication and Ground-level Ozone and the Emission Ceilings Directive of the EU, where the model calculated least-cost emission ceilings that achieve the proposed “gap closure” targets for a range of environmental effects. By connecting the effort in reducing emissions with quantitative targets for environmental quality, the optimization mode is at the core of the effect-based approach.

Introducing flux calculations in integrated assessment

From an integrated assessment perspective, progress in flux modeling is highly welcomed because it allows more accurate modelling of ozone concentration levels by taking into account deposition processes in a more realistic way, and because it enables a more accurate assessment of impacts of ozone on vegetation.
In interaction with the full EMEP photochemical model, the scenario-mode calculation of ozone impacts of a given emission scenario seems perfectly feasible with the flux-based methodologies outlined in the meeting report. However, using flux calculations for target setting in the optimization mode raises a number of problems:

- **Methodology to introduce fluxes into integrated assessment needs to be developed.** If it is intended to express environmental targets in terms of fluxes, it is necessary to include the fluxes in the source-receptor relationships that are used by the integrated assessment model, i.e., to quantify how individual emission sources contribute to the ozone flux to a specific vegetation type at a given location. Since fluxes are vegetation-specific, different source-receptor relationships have to be derived for each vegetation type. It cannot be judged at the moment to what extent this will be feasible, but without any doubt significant time and resources will be necessary to study this problem and to derive quantitative results. With the time plan envisaged in the meeting report for producing accepted procedures and results for flux calculations (i.e., two years from now), we estimate that it would take approximately two more years before flux calculations could be fully incorporated in integrated assessment models for target setting.

- **Acceptability of fluxes as policy targets**, as they are not directly measurable quantities: For the practical policy application, experience shows that environmental targets used in integrated assessment need to be intuitive and transparent to decision makers and the public, so that they can be easily understood and thus used as policy targets. The ability to directly measure a target and to judge compliance with the target is an important aspect in this context. If countries are not able to measure how far away they are from the policy target, they are not likely to accept it as a transparent value. Moving from a concentration-based measure to flux-based targets might cause serious problems because of the practical difficulties in measuring fluxes. It appears questionable whether a measure that cannot be directly measured and that is solely based on calculations of a single model could serve as an acceptable policy target.
- **Existence of a threshold value**: We note that the meeting report does not address the question whether 'no-effect' threshold values of fluxes, representing critical levels for ozone (e.g., in analogy to the old AOT40 critical level) or only dose-response curves (as for health impacts from fine particles) are envisaged as the final product. While in principle both types of information could be used for target setting, it would be important for negotiators and integrated assessment modelers to be informed about the envisaged deliverable, so that options for target setting can be explored in time. Again, time is a critical issue, because appropriate methodologies need to be developed.
- **Sub-grid variation**: While regional variation in Europe is improved in comparison to the AOT40 approach, it is noted that the present lack of variations of ozone fluxes within a grid cell puts a serious constraint on the usefulness of fluxes for short-term policy application. Progress has been made in many other fields to address sub-grid variations (e.g., ecosystem-specific depositions, pollution levels in cities, etc.), so that a lack of a comparable approach for fluxes might seriously hamper the credibility of calculations that ignore such variations.
- **Irrigated vegetation**: We assume that the present methodology addresses non-irrigated natural vegetation and crops. For any economic assessment, however, (and therefore for policy applications) irrigated agricultural crops are most important. The meeting report does not specify how flux calculations could be applied for irrigated vegetation. Ignoring the differences caused by irrigation, or ignoring irrigated crops altogether, would create a serious and relevant flaw in an integrated assessment. If flux methods should be used for target setting, developing concepts for handling irrigated crops and compiling appropriate land use databases needs to become a priority.
- **Time is perhaps the most serious problem**. While many of the issues listed above could possibly be resolved at least in principle, there is little time left for developing a practical approach for setting vegetation-related targets for the forthcoming round of policy analysis. The meeting report estimates that level 2 will only be available and fully accepted in two years, i.e., by mid of 2004. If integrated assessment should express environmental targets in terms of fluxes, even more time is needed to explore the relation between fluxes and concentrations and to represent it in the source-receptor relationships. We estimate that this could easily take two years, and would then deliver results in 2006, i.e., too late for CAFE and for CLRTAP.

A possible two-track approach? While we consider these problems are serious in an integrated assessment context, we could imagine a constructive and practical solution by developing, in addition to the proposed flux methodology, a measurable, concentration-based yardstick that captures, for the purposes of target setting, variations in sensitivities caused by differences in ozone fluxes. A potentially useful approach could specify, for a given ecosystem at a given site, a critical concentration threshold that would correspond to a critical flux level. While a ‘modified’ AOTx concept has been proposed, any other statistical measure based on observable ozone concentrations, e.g., in relation to seasonal mean ozone concentrations, is conceivable. Obviously, such a relation needs to be grid-, meteorology- and vegetation-type-specific and would in practice be based on calculations of the EMEP photo-oxidants model.

This measure could be used for target setting in the integrated assessment, which would then be concentration based. Thereby, the interface between atmospheric calculations and impact assessment would be measurable ozone concentrations, in line with what will be required for setting health-related targets to protect the population.

If such a measurable concentration-based concept could be developed, reviewed and implemented within the next year, we could imagine a two-track approach for the next round of policy assessment. This concentration-based measure would be used in integrated assessment models to set environmental targets to drive emission controls and to obtain cost-effective sets of emission reduction measures, while subsequently the full flux calculations could be used in the ‘scenario analysis’ mode to quantify the environmental effects of these emission reductions. To implement this concept, it would be critical to consider the development of the alternative concentration-based measure as a priority activity.

In Establishing Ozone Critical Levels II (Karlsson, P.E., Selldén, G., Pleijel, H., eds.). 2003. UNECE Workshop Report. IVL report B 1523. IVL Swedish Environmental Research Institute, Gothenburg, Sweden.

Ambient flux-based critical values of ozone for protecting vegetation: differing spatial scales and uncertainties in risk assessment

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Abstract

The current European critical levels for ozone (O_3) to protect crops, natural and semi-natural vegetation and forest trees (Level I) are based on exposure-response relationships using the AOT40 exposure index. In the context of the revision of the 1999 UNECE multi-pollutant/multi-effect protocol that is expected in 2004 ("... no later than one year after the present Protocol enters into force"; UNECE, 1999, Article 10), a transition to a flux-based limiting value is currently under discussion. In principle, there are three alternatives for replacing the Level I approach based on European literature and scientific discussions. One alternative is a modified AOT40 index. Because of several uncertainties discussed in the literature during the recent years that approach appears questionable. The second alternative is the German VDI's MPOC (Maximum Permissible O_3 Concentration at the canopy top) concept. In contrast to the current European critical AOT40 levels, MPOC values are based on a significantly higher number of experiments, with more than 30 species for crops and wild plants and 9 species for forest trees. In principle, the MPOC concept can be applied from local, up to the European scale and fulfil the demand for the UNECE abatement strategies. The third alternative under discussion is the flux approach. Here, critical levels will have to be replaced by critical cumulative stomatal uptake (critical absorbed dose). The main problems with that approach can be attributed to uncertainties due to, (1) parameterisation of stomatal conductance (e.g. how representative are they of different geographic regions in Europe in up-scaling from leaf estimates to canopy level, (2) parameterisation of non-stomatal deposition, and (3) the representativeness of species used in flux-effect studies. Nevertheless, establishing realistic flux-effect relationships clearly requires chamber-less experiments, especially for species rich ecosystems, but will have to be based on flux estimates at the canopy level. Compared to Europe, the situation is quite different in North America. Although in general, the flux approach is well accepted by plant effects scientists there, concerted research efforts have not taken place in that direction due to a distinct lack of funding. Furthermore, because of the differences in the approach to setting ambient air quality standards in N. America, it appears very doubtful that policy makers and air quality regulators in the US and Canada will readily accept the overall philosophy.

Introduction

Tropospheric ozone (O_3) poses a critical threat and a challenging problem to present and future world food, fiber and timber production and conservation of natural plant communities, including their species diversity (Krupa *et al.* 2001). Some 50 years of research has taken place in the US on the adverse effects of O_3 on terrestrial vegetation. Based on the numerous studies published and the world literature, a secondary National Ambient Air Quality Standard (NAAQS) is used in the US to protect vegetation against the negative effects of O_3, while a primary standard is designed to protect human health. The current secondary standard in the US is the same as its primary 8-hour standard. That 8-hour standard is considered to be met at an ambient air quality monitoring site, when during a 3-year period, the average of the 4th highest daily maximum 8-hour mean O_3 concentration is $\leq$

0.080 ppm (Federal Register, 1997). The Canada-Wide Standard (CWS) for O_3 is similar to the US, with the exception that the 8-hour average is 0.065 ppm (CCME, 2000).

In comparison to the long history of O_3-vegetation effects research in the US, in Europe, particularly within the UN-ECE (United Nations Economic Commission for Europe) and the European Union, as a consequence of the so-called forest die-back, since the mid 1980s, tropospheric O_3 and its impacts on human health and vegetation has received increasing attention. Although initially critical levels for O_3 were defined as 7-hour means over the growing period (25 ppb, 0900-1600 hrs) to protect vegetation (UN-ECE, 1988), through subsequent discussions, it was changed to an **A**ccumulated exposure index **O**ver a certain **T**hreshold, AOTx, (Fuhrer and Achermann, 1999a; EU, 2002).

However, currently there is a widespread agreement among plant effects scientists that simple air or exposure concentrations measured at some height above the surface are not very useful in relating to the corresponding plant responses, since the true underlying mechanism is the dose taken up or absorbed by the plant canopy (Krupa and Kickert, 1997). The exchange of gases between the atmosphere and the phytosphere (flux) is governed by the ambient O_3 concentration, the turbulent conductivity of the lower atmosphere, and the sink properties of the plants (Grünhage *et al.,* 1997).

Based on that principle, there is a very strong and concerted movement among the scientists within the UN-EU to arrive at a flux-based ambient O_3 air quality critical level(s) to protect vegetation. Although that represents the most desirable strategy, this paper describes some of the scientific concerns and uncertainties in applying such an approach for risk assessments at different spatial scales.

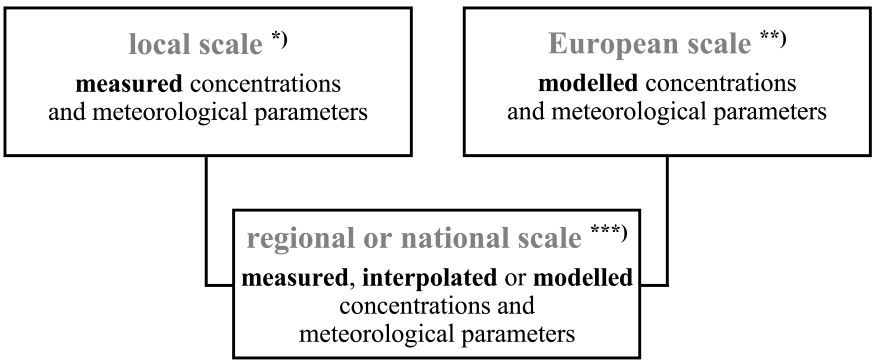
Risk assessments in the US

It is well accepted that ambient O_3 is the most important phytotoxic air pollutant in the US (Krupa *et al.,* 2001). However, at the present time, vegetation related risk assessment in the US is solely based on various exposure statistics derived from the measurements of air concentrations (US-EPA, 1996). A number of statistical yield loss functions were developed for many crops in a variety of locations using open-top chambers (OTCs). The results clearly indicated production losses due to O_3 (US-EPA, 1996). However, considerable variability was observed between and within species, between years, irrigation regimes, and environments. Combining data from 38 species in the U.S. and applying a statistical function to 7 hour mean O_3 concentration data, suggested that 11% of species would exhibit 10% yield loss when exposed to an average of 0.035 ppm and 50% of the cases at 0.050 ppm (US-EPA, 1996). Those levels of O_3 are observed during the growth season at many locations in the US (http://www.epa.gov/castnet/data.html). Extrapolation of those limited data from domesticated species grown in OTCs to the plant kingdom in general, and to the ambient environment, is subject to considerable uncertainty. However, it was estimated that O_3-induced economic damage on 8 major US crops was $1.89-3.3 billion annually (see Mauzerall and Wang, 2001). Similarly, studies have been conducted on the responses of tree seedlings and saplings to O_3 in OTCs showing adverse effects when exposed to a 7 hour average of >0.080 ppm. An analysis of ambient O_3 data for the contiguous US for the 1980-1998 period showed that the average number of summer days per year in which O_3 concentrations exceeded 0.08 ppm (level of the current secondary NAAQS) is in the range of 8-24 in the northeast and Texas and 12-73 in southern California (Lin et al. 2001). However, as with crops, a great deal of uncertainty remains regarding the extrapolation of results from seedling/sapling studies to mature trees, their populations, ambient environments, ecosystems and landscapes (US-EPA, 1996). Because of those considerations economic loss estimates are not currently available for forests and native vegetation. Nevertheless, in its final analysis, EPA's Clean Air Science Advisory Committee concluded that plants are more sensitive and respond more rapidly than humans to O_3 stress and therefore, the secondary NAAQS must be more stringent than the primary. However, "agreement on the level and form of such a standard is still elusive" because of "… important limitations to our understanding of the extent of the responses of vegetation to O_3 under field conditions" (Federal Register, 1997).

Risk assessments in Europe

Spatial Scales

Within the UN-ECE, it is well accepted that risk assessments should be performed at different geographic scales (Fig. 1). The development of protocols on the control of O_3 precursor emissions under the 1979 UN-ECE Convention on Long-Range Transboundary Air Pollution (LRTAP; UN-ECE, 1979) assumes that models will accurately predict the impacts of precursor pollutant control strategies on ambient O_3 concentrations and deposition across Europe. Thus, for use within the EMEP (Cooperative Programme for Monitoring and Evaluation of the Long-range Transmission of Air Pollutants in Europe) photochemical model, a new big-leaf module was developed to estimate O_3 deposition or fluxes on to major vegetation types (Emberson et al., 2000a, b). Using that approach, estimates of the O_3 fluxes were based on large-scale modelled meteorology and concentration fields. Together with cumulative flux-effect relationships, outputs of the EMEP models allowed economic assessments of, for example, O_3-induced crop losses in Europe.



*) Council Directive 96/62/EC on ambient air quality assessment and management
**) UNECE Convention on Long-Range Transboundary Air Pollution
***) Council Directive 96/62/EC on ambient air quality assessment and management
UNECE Convention on Long-Range Transboundary Air Pollution

Fig. 1. Levels of risk assessments, data used and legal basis

Within the UN-ECE Mapping Programme, National Focal Centers are responsible for risk assessments at the national scale using small-scale models and data from monitoring networks (UBA, 1996). EMEP outputs can be used as boundary conditions for such national models. On the other hand, the Council Directives 96/62/EC (EU, 1996) and 2002/3/EC (EU, 2002) require risk assessment on the basis of "fixed continuous measurements" (Article 9 of EU, 2002). Such measurements for pollutant concentrations and meteorological parameters will have to be performed at local as well as on regional or national scales. That implies a network of air quality monitoring stations representative of the different geographic extents (urban, suburban, rural, rural background, and perhaps pristine air, Annex IV of EU, 2002).

Uncertainties in Critical Levels

Critical levels based on the AOT40 Level I approach are subject to several uncertainties:

- They are based on a relatively small number of experiments (17 for crops and 3 for trees; Fuhrer *et al.,* 1997; Kärenlampi and Skärby, 1996);

- Only a very limited number of plant species are considered (wheat as representative of crops and semi-natural vegetation, young (0-3 yr) beech as representative of forest trees), although O_3 response data are available for other species (Grünhage *et al.,* 2001);
- Sometimes the sensitivity of the AOT40 index to variations in the input data appear to be unacceptably high (Tuovinen, 2000; Sofiev and Tuovinen, 2001).

Nonetheless, the UN-ECE Level I approach is simple and therefore attractive, however, it does not consider that the critical levels are related to the O_3 concentrations at the top of the canopy and not at some measurement height above the surface. It also does not consider biotic or abiotic factors that influence vegetation response to O_3. Therefore, to address these shortcomings, at the 1999 workshop on "Critical Levels for Ozone – Level II" (Gerzensee, Switzerland), three different options were identified (Fuhrer and Achermann, 1999b):

- A revision of the Level I values using factors that modify cause-effect relationships;
- A revision of the AOT40 values calculated to identify exceedances of the Level I values, using the modifying factors; and
- The development of a flux-based approach that addresses the O_3 uptake and the toxicity of the absorbed dose.

The approach of using modifying factors (cf Posch and Fuhrer, 1999) may be considered as being empirical. Similarly, based on the aforementioned uncertainties, the flux-based AOT40effective previously proposed by Grünhage *et al.* (1999) and Tuovinen (2000) may be considered in principle as biased.

Derivation of Flux-based Limiting Values

Any flux-based limiting value would require the application of micrometeorological Soil-Vegetation-Atmosphere-Transfer (SVAT) models, the simplest of which follows the big-leaf approach (cf Grünhage *et al.,* 2000). This SVAT model parameterises O_3 exchange $F_{total}(O_3)$ between the atmosphere near the ground and the phytosphere applying a relatively simple resistance scheme taking into account that there are sinks in the plant canopy reducing the O_3 concentration ρ_{O3} to zero (cf Laisk *et al.,* 1989; Wang *et al.,* 1995):

$$F_{\text{total}}(\text{O}_3) = -\frac{\rho_{\text{O3}}(z_{\text{ref}})}{R_{\text{ah}} + R_{\text{b}}(\text{O}_3) + R_{\text{c}}(\text{O}_3)} \tag{1}$$

The parameterisation of the turbulent atmospheric resistance R_{ah} describing the transport properties for O_3 between a reference height ($z_{ref,\,O3}$) above the canopy and the conceptual height ($z = d + z_{0m}$), the sink for momentum (d = displacement height, z_{0m} = roughness length for momentum) and the quasi-laminar layer resistance $R_b(O_3)$ between momentum sink height ($z = d + z_{0m}$) and the O_3 sink height ($z = d + z_{0,\,O3}$) are well accepted (Grünhage *et al.,* 2000). However, higher uncertainties exist for the parameterisation of the bulk canopy resistance $R_c(O_3)$ describing the influences of the plant-soil system on the vertical exchange of O_3.

Total O_3 flux can be partitioned into the flux absorbed by the plant through the stomata and the cuticle and the flux on the external plant surface and the soil (combined non-stomatal deposition). Investigations of cuticular permeability of O_3 and other trace gases show that penetration through the cuticle can be neglected in comparison to stomatal uptake (Kerstiens and Lendzian, 1989a, b; Lendzian and Kerstiens, 1991; Kerstiens *et al.,* 1992), thus:

$$F_{\text{total}}(\text{O}_3) \cong F_{\text{stomatal}}(\text{O}_3) + F_{\text{non-stomatal}}(\text{O}_3) \tag{2}$$

Because stomatal uptake $F_{stomatal}(O_3)$ is the toxicologically effective part of $F_{total}(O_3)$, flux-effect relationships should be based on that component, expressed by:

$$F_{\text{stomatal}}(\mathrm{O_3}) = -\frac{\rho_{\mathrm{O3}}(z_{\text{ref}})}{R_{\text{ah}} + R_{\text{b}}(\mathrm{O_3}) + R_{\text{c, stomatal}}(\mathrm{O_3}) + \left[[R_{\text{ah}} + R_{\text{b}}(\mathrm{O_3})] \cdot R_{\text{c, stomatal}}(\mathrm{O_3}) \cdot \frac{1}{R_{\text{c, non-stomatal}}(\mathrm{O_3})} \right]} \quad (3)$$

In comparison, stomatal uptake calculations proposed for example, by Bermejo *et al.* (2002), Grulke *et al.* (2002) and Mills *et al.* (2002) deviate from known rules of micrometeorology or physics. Calculation of stomatal uptake by multiplying stomatal conductance with O_3 concentrations measured at some height above the canopy, neglects the influence of Rah, Rb(O_3) and Rnon-stomatal(O_3) on O_3 flux, resulting in an overestimation of O_3 uptake (cf Fig. 3 in Grünhage *et al.,* 2002). It should only be interpreted as a first approximation of stomatal loading (potential stomatal loading, PSL). Any partitioning of total O_3 deposition into stomatal uptake and non-stomatal deposition has to take into account Kirchhoff's Current Law which states that the current into a node equals the current out of a node (Kirchhoff, 1845), in other words $F_{\text{total}} = F_{\text{stomatal}} + F_{\text{non-stomatal}}$.

Bulk stomatal resistance $R_{\text{c, stomatal}}(O_3)$ is parameterised via resistance to water vapour taking into account the differences between the molecular diffusivity for water vapour and ozone. In many SVAT models as in the EMEP big-leaf module, the dependence of bulk stomatal resistance for H_2O, on radiation, temperature and the water budgets of the atmosphere and the soil is described by the Jarvis-Stewart approach (Jarvis, 1976; Stewart, 1988). There, $R_{\text{c, stomatal}}(H_2O)_{\text{min}}$ represents the minimum value of the bulk stomatal resistance for water vapour and functions $f_1(S_t)$, $f_2(T)$, $f_3(VPD)$, $f_4(S_M)$ and $f_n(...)$ account for the effects of solar radiation *S*t, temperature *T*, atmospheric water vapour deficit *VPD*, soil moisture S_M and other factors such as the influence of plant development stage or the time of day on stomatal aperture ($0 \le f_i \le 1$; cf Körner *et al.*, 1995; Legge *et al.*, 1995).
$R_{\text{c, stomatal}}(H_2O)$min can be determined directly via micrometeorological water vapour flux measurements at the canopy level or can be estimated by an up-scaling procedure from leaf-level estimates using the leaf area index *LAI* (cf Baldochi *et al.*, 1987; Kelliher *et al.*, 1995; Grünhage *et al.*, 2000). Such up-scaling procedures from leaf or twig-branch to whole canopy as applied in the EMEP big-leaf module, assume that all leaves or classes of leaves at a given time have the same potential for gas exchange and do not consider possible systematic variations in the driving forces at the leaf surface, from leaf to leaf (Jarvis, 1995). While these bottom-up approaches might be suitable for monocultures (e.g. crops), they are not appropriate for multi-species systems such as semi-natural grasslands. In addition to the uncertainties in flux estimates due to the up-scaling procedure, in principle the flux estimates can be biased if the value for the minimum stomatal resistance chosen is not appropriate or the leaf stomatal conductance model is not well calibrated. The coefficients of determination for the three sets of data in Figure 2 illustrate the difficulty with model adequacy. In that case, all measurements were performed using the same clover clone (NC-S; Heagle *et al.*, 1995) that was established following a standard protocol developed by the ICP Vegetation Coordination Centre (UN-ECE, 2001). The results show the basic necessity of validating any parameterisation of stomatal resistance via measurements of canopy level water vapour exchange.

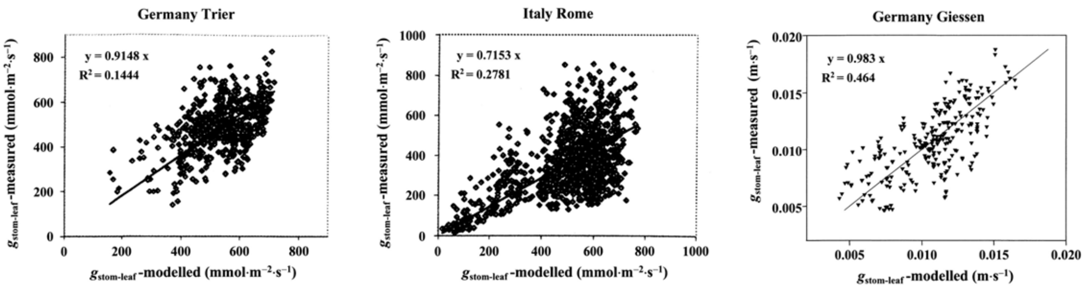


Fig. 2. Comparison between modelled and measured leaf-level stomatal conductance for a white clover clone established at three different sites in Europe (Trier, Germany [n = 775] and Rome, Italy [n = 1063]: Mills et al., 2002; Linden near Giessen, Germany [n = 261]: Gavriilidou *et al.*, 2002)

Even greater uncertainty can be noticed with respect to the parameterisation of $R_{c,\ non\text{-}stomatal}(O_3)$ controlling non-stomatal deposition. $R_{c,\ non\text{-}stomatal}(O_3)$ can be estimated as the residual term in the canopy resistance if bulk stomatal resistance is known (Fowler *et al.*, 2001).
As in the EMEP big-leaf module, respectively $R_{c,\ non\text{-}stomatal}(O_3)$ components are often parameterised by constant values neglecting for example, the influence of surface wetness on the sink properties for O_3. Rondón *et al.* (1993), Coe *et al.* (1995), Fowler *et al.* (2001) and Gerosa *et al.* (2002) observed enhanced O_3 deposition to external surfaces of wheat, moorland and conifer species. Measurements by Fowler *et al.* over moorland at Auchencorth Moss in Southern Scotland showed that during a 2-year period, at a seasonal time scale, non-stomatal deposition dominated (65 %) the overall flux. The reduction of $R_{c,\ non\text{-}stomatal}(O_3)$ with increasing radiation (Fig. 3) and temperature is regarded as evidence of thermal decomposition of O_3 at the leaf surface.

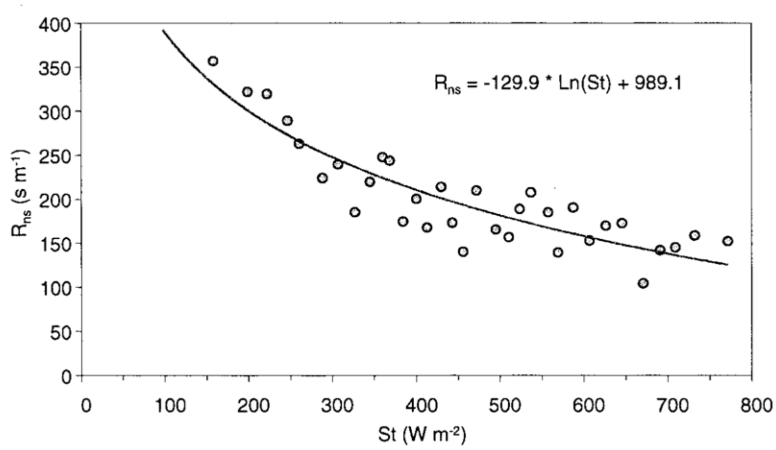


Fig. 3. The relationship between canopy resistance for non-stomatal O_3 deposition (R_{ns}) and global radiation (S_t), Auchencorth Moss daytime data. Stomatal canopy resistances ($R_{c,\ stomatal}$) for dry surfaces were estimated from water flux measurements (Fowler *et al.*, 2001)

Figure 4 illustrates the problem arising from relatively similar dial patterns of $R_{c,\ stomatal}(O_3)$ and $R_{c,\ non\text{-}stomatal}(O_3)$. Deviations can be expected only if there is a strong influence of VPD on stomatal aperture. Comparisons of the three flux patterns in Figure 4 show that the increase in O_3 deposition due to reduced $R_{c,\ non\text{-}stomatal}(O_3)$ during daylight hours (y = 1.144 x) can be compensated by a higher $R_{c,\ stomatal}(H_2O)_{min}$ (y = 1.003 x). This example demonstrates the need for appropriate validation procedures. Nevertheless, after validation of the parameterisation of stomatal behaviour via water vapour flux measurements, it is logical to validate parameterisation of non-stomatal deposition by micrometeorological measurements of O_3 exchange at the canopy level.

Further, modelled O_3 fluxes can be biased if air chemistry is not considered. In the first few meters above the canopy O_3 flux densities can be influenced by reaction with NO emitted from the soil and hydrocarbons emitted by the vegetation (cf Grünhage *et al.*, 2000). While reaction of O_3 with NO can be important in fertilised agriculture, reactions with hydrocarbons may be significant for forest ecosystems.

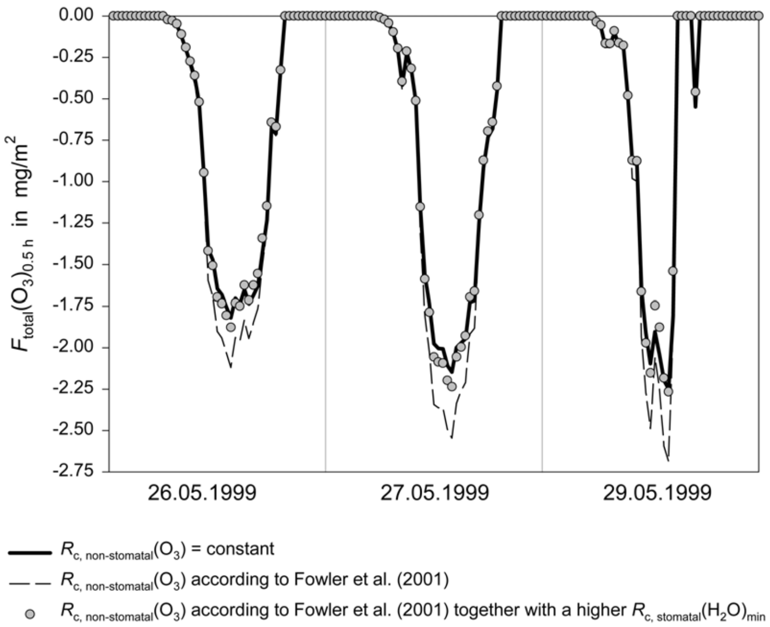


Fig. 4. Dial variation of total O_3 deposition during daylight hours (S_t > 100 $W^{\cdot}m^{-2}$) applying different parameterisations for $R_{c,\ non\text{-}stomatal}(O_3)$ and $R_{c,\ non\text{-}stomatal}(H_2O)_{min}$

Uncertainties in Flux-based Limiting Values

The application of the AOT40 *critical level* (according to its present definition), as well as the German Maximum Permissible O_3 Concentrations (MPOC; Grünhage et al., 2001; VDI 2310 part 6, 2002) requires the determination of O_3 concentrations at the top of the canopy, i.e. at the upper boundary of the quasi-laminar layer ($z = d + z_{0m}$), if the big-leaf approach is applied. Taking into account the aforementioned resistance parameterisations, these concentrations [$\rho_{O3}(d+z_{0m})$] can be recalculated by:

$$\rho_{O3}(d + z_{0m}) = \rho_{O3}(z_{ref}) + \left[F_{total}(O_3) \cdot R_{ah}\right] \tag{4}$$

It should be noted that during conditions that limit stomatal O_3 uptake such as low radiation or high water vapour pressure deficit in the atmosphere, high O_3 concentrations can occur at the top of the canopy without a significant toxicological risk. In other words, O_3 concentrations at the upper boundary of the quasi-laminar layer are not readily coupled to stomatal uptake. Thus, a correction is needed to provide toxicologically effective O_3 concentrations, by taking into account the stomatal opening. Thus, O_3 concentrations at the upper boundary of the quasi-laminar layer should be weighted with the Jarvis-Stewart factors for radiation, temperature, atmospheric water vapour pressure deficit and soil moisture.

The simplest flux-based approach is the German VDI's Maximum Permissible O_3 Concentrations (MPOC) at the canopy top (the conceptual height $z = d+z_{0m}$), rather than at some measurement height above the surface (Grünhage *et al.,* 2001; VDI 2310 part 6, 2002). The MPOC values for crops, semi-natural and natural vegetation and forest trees were derived from a reanalysis, by applying a micrometeorological flux model to the 1989-1999 published data on the effects of O_3 on European plant species (see http://www.uni-giessen.de/~gf1034/ENGLISH/WINDEP.htm).

For characterising the risk to vegetation at a specific site, different types of average O_3 concentrations were calculated based on a ranking in a descending order of all half-hourly or hourly O_3 concentrations at $z = d+z_{0m}$ during the growing season of the year under consideration. Because the MPOC approach does not consider the toxicological effectiveness of the O_3 concentrations at $z = d+z_{0m}$, applying that concept can be interpreted as a 'worst-case' assessment. MPOC-risk evaluation for forests is presented in Krause *et al.* (2002) and for semi-natural vegetation discussed in Bender *et al.* (2002).

At present, the database for the derivation of critical cumulative O_3 fluxes (critical loads) is extremely inadequate. For spring wheat, a flux (stomatal uptake by the flag leaf) - response (relative yield) relationship was calculated from 5 open-top chamber experiments, with two "old" wheat varieties (Pleijel *et al.,* 2000). Additional data on such relationships are available from open-top chamber studies with potato and the grass *Phleum pratense* (Pleijel *et al.,* 2002). Because the relationships are based on leaf flux estimates, it can be argued whether they are representative of the whole canopy. For example, at the beginning of the grain filling period, the leaf area of the flag leaf represents only a fraction (20 - 25 %) of the non-senescent leaf area of the whole canopy (Pleijel *et al.,* 2000). Furthermore, from an economic viewpoint, it is essential that the selected agricultural species be representative of the geographic area. In Germany for example, spring wheat represents approximately 1.6 % of the total area under wheat cultivation. Therefore, spring wheat does not appear to be the appropriate species for evaluating the risk of O_3-induced crop yield loss in that region of Europe.

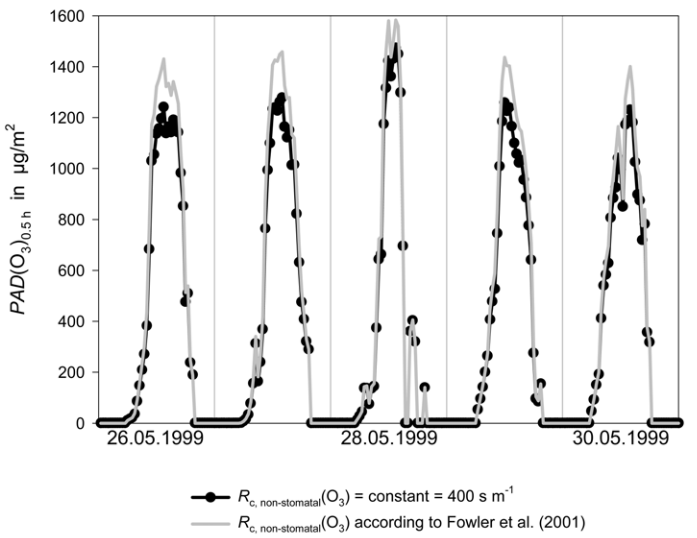
Constant flux thresholds for O_3 uptake as supposed by Pleijel *et al.* (2002), appears questionable from a toxicology view point. At first, the threshold depends on the biological response parameter considered. Secondly, the threshold depends on the capacity of detoxification at the respective growth stage. The relationship between stomatal uptake and effect does not obey the law of reciprocity, according to which equal doses generate equal effects. The same cumulated stomatal O_3 uptake (pollutant absorbed dose *PAD*; Fowler and Cape, 1982)

$$PAD(\mathrm{O}_3) = \int_{t_1}^{t_2} \left| F_{\mathrm{stomatal}}(\mathrm{O}_3) \right| \cdot \mathrm{d}t \tag{5}$$

can cause more injury, shorter the time in which the dose is absorbed. In particular, high $F_{stomatal}(O_3)$ can be a greater hazard if it is in sink with high O_3 concentrations and more so if that temporal relationship is repetitive. Overall, both situations lead to a premature depletion of the detoxification capacity. Consequentially, O_3 fluxes must be weighted and the frequency of the occurrences of sequentially high fluxes must be taken into account (Grünhage and Jäger, 1996, 2001).

Because until now predominant descriptions of exposure/flux-response relationships are based on chamber experiments, in principle they may be considered as biased (Jetten, 1992). Generally, O_3 exposure-response relationships derived from chamber experiments show linear or non-linear relationships between exposure and plant response. This is in contrast to the observations under ambient conditions where biological responses to O_3 exposures do not always increase with increasing exposures (Bugter and Tonneijck, 1990; Krupa *et al.,* 1995). Furthermore, without any changes in the pollution climate, modifications of species composition can take place in species rich ecosystems such as the grasslands, due to the differences in radiation, air temperature and humidity between the chamber and ambient microclimates (cf Grünhage *et al.,* 1990). Equally importantly, responses of a given species in isolation (monocultures) would appear to be quite different than in the presence of a competitor for resources, as in mixed plant communities and ecosystems (Andersen *et al.,* 2001). Therefore it should be emphasized that realistic flux-effect relationships require chamber-less experiments (Grünhage *et al.,* 2002), especially for species rich ecosystems and will have to be based on flux estimates at the canopy level.

The UN-ECE ICP Vegetation clover programme provided an opportunity to derive *critical O_3 loads* under chamber-less, ambient conditions. The provisional *critical absorbed O_3 dose* for the onset of visible injury on the NC-S clover clone derived by Gavriilidou *et al.* (2002) shows that the demand for validation of stomatal parameterisation via water vapour fluxes can also be achieved in pot experiments, if the so-called "oasis effect" (cf Brutsaert, 1984; van Eimern and Häckel, 1984) is taken into account. The oasis effect occurs when hot dry air blows across the edge of an oasis resulting in rapid evapo-transpiration that is governed by the sensible heat from the air and the radiant energy. Accordingly, the evapo-transpiration of a small wet area (well-watered pot of clover) embedded in a dryer environment (semi-natural grassland without irrigation) will be higher than that of an extended wet area. As illustrated in Figure 5, the pollutant-absorbed dose depends strongly on the parameterisation of non-stomatal O_3 deposition. Therefore, all of the aforementioned limiting values for O_3 to protect vegetation may not be realistic, since the values were not verified by independent experiments and by micrometeorological flux measurements.



$$\frac{\text{constant } R_{c,\,\text{non-stomatal}}(O_3)\ \cdot\ 100}{R_{c,\,\text{non-stomatal}}(O_3) \text{ according to Fowler et al.}} =$$

88.7 %	88.9 %	93.6 %	89.1 %	91.2 %

Fig. 5. Variations in absorbed O_3 doses applying different parameterisations for Rnon-stomatal(O_3)

Conceptual degrees of uncertainty for differing limiting values of O_3 to protect vegetation are illustrated in Figure 6.

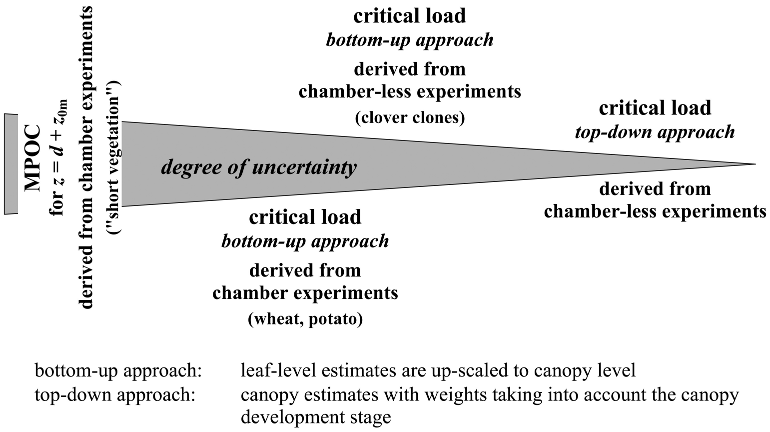


Fig. 6. Conceptual degree of uncertainty in flux-based limiting values for crops and semi-natural vegetation

Uncertainties in flux-based risk assessments

While the European O_3 risk assessments under the convention of LRTAP have a spatial grid scale of 50 km x 50 km, they have to be based on relatively generalised concepts. Site-specific meteorological conditions and pollutant climate are replaced by modelled meteorological and concentration fields, and site-specific ecosystems properties are replaced by those of a vegetation type. It is clear that such a generalised approach must be carefully calibrated by well validated models for site level risk assessments and not *vice versa*. Beside the aforementioned issues, the degree of uncertainty of the EMEP model outputs depends largely on the quality of the land use data and on the prediction of the water balance (soil moisture, atmospheric water vapour pressure deficit) of the respective vegetation types in a specific model grid.

Because the results of risk evaluations on smaller geographic scales require a higher degree of precision, they can be verified directly by for example, the farmers. In Figure 7 flux-based approaches for local scale risk evaluations for crops and natural and semi-natural vegetation are ranked by the degree of their uncertainty.

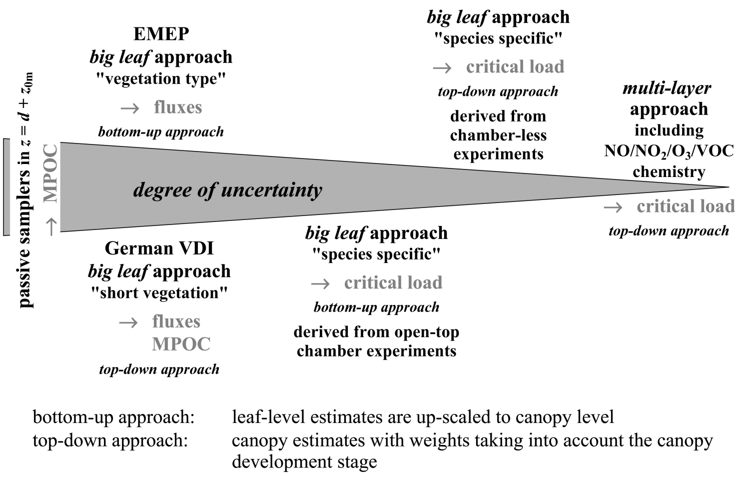


Fig. 7. Conceptual degree of uncertainty in flux-based O_3 risk assessments for crops, natural and semi-natural vegetation using measured data

There is a rapid increase in the use of passive O_3 samplers both in N. America and in Europe providing weekly or biweekly mean ambient concentrations (Krupa and Legge, 2000). Although that technology is evolving to allow the measurements of dial or diurnal concentrations, such an effort while desirable can be labour intensive and in many cases cost limiting when applied to a regional scale. While there are large uncertainties with the application of MPOC, passive samplers allow an inexpensive approach to the first order MPOC risk assessments on a local to regional scale at time scales of ≥ week (cf Krause *et al.*, 2002). Where a greater time resolution is deemed warranted, as in specific flux calculations, the preliminary studies of Krupa *et al.* (2001, 2002) that use either a Weibull probability generator or a multi-variate meteorological model represent the beginning of our efforts to simulate passive sampler data to mimic the continuously monitored frequency distributions of hourly O3 concentrations. Overall, results from such efforts can be incorporated into the EMEP or the German VDI big-leaf model to achieve a greater utility in understanding cause-effect relationships under ambient conditions.

Conclusions

In Europe, all flux-based risk evaluations have to be based on validated models for the different climate zones, considering specific plant species, their cultivars, varieties or genotypes. Taking into account the up-scaling problems, species-specific big-leaf models based on a bottom-up approach are less precise than models based on a top-down approach. Especially for forests or highly fertilised agricultural systems, local risk evaluations based on multi-layer, Soil-Vegetation-Atmosphere-Transfer (SVAT) models, including air chemistry are more appropriate. Such extended models applied at representative sites distributed over Europe can serve as an additional fine tuning instrument for big-leaf models with a one-layered resolution of the vegetation.

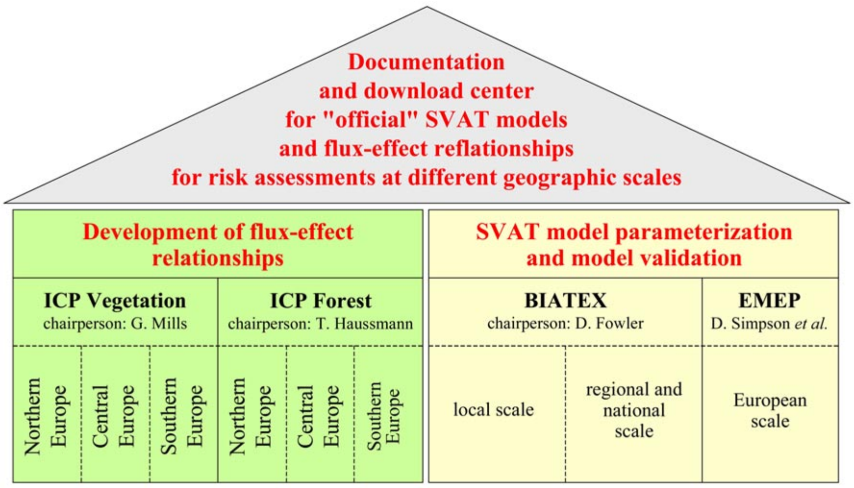


Fig. 8. Organisational chart for the establishment of *critical O_3 loads* and O_3 risk evaluation

Therefore, a network of O_3 flux monitoring sites will have to be established throughout Europe. Because on a relative scale, the existing flux-effect relationships are yet to be validated, future work will have to be directed to establishing cause-effect relationships based on chamber-less experiments at micrometeorological flux measurements sites. Progress in developing realistic risk evaluation procedures can be guaranteed only, if the existing scientific communities can come together. For Europe, integration as illustrated in Figure 8 might be reasonable. While the development of flux-effect relationships can be performed under the leadership of ICP Vegetation and ICP Forest, SVAT model parameterisation and model validation may be achieved by the BIATEX (**BI**osphere/**AT**mosphere **EX**change of Pollutants) community for local to national scales and by EMEP for the European scale. The work of those groups may be consolidated in close co-operation with the ICP Modelling and Mapping Program. It might be desirable to establish a virtual documentation group for "official" SVAT models and flux-effect relationships for risk evaluation at different geographic scales.

However, such an approach if applied to the US and Canada will have to be substantially modified based among others on the differences in the research effort and the technical organisational structure, human health considerations, on the geographic scale-climatic extent and diversity of cultivated species (crops), native vegetation and forest ecosystems. The approach to setting an air quality standard (NAAQS) in the US is very different from the EU (e.g., Federal Register, 1997). Nevertheless, some US scientists from 14 major institutions participating in an ambient O_3 – vegetation effects research project (USDA Multi-State Project # 1013) suggest as an example, that if an ambient flux-based secondary NAAQS were to be identified, although the EU might consider a time-integrated threshold or "critical level", such an approach would not capture the stochasticity of plant compensation and repair of stress that is governed by uptake beyond a point of the plant's resources and ability to cope. An alternative could be to capture the dynamics of maximal deposition and uptake. That can be accomplished by examining the time-integrated frequency distributions of the atmospheric flux and plant uptake using their percentile statistics such as the 90^{th} or the 95^{th} values. That is feasible to administer from a regulatory perspective, since one can develop a "Vegetation Injury Index" based on the conditions for increased O_3 synthesis/transport and period of the day and generalised climatic conditions that likely will produce maximal flux and uptake. However, for validation, such a definition must be coupled to measured plant responses. Nevertheless, it appears very doubtful that policy makers and air quality regulators in the US and Canada will readily accept such an overall philosophy.

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Stomatal Ozone Uptake over Europe: Preliminary Results

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1 Introduction

This chapter reports briefly on some preliminary results on ozone uptake modelling performed within the EMEP photo-oxidant model. This new version (rv1.2β} contains a deposition module which enables explicit calculation of the stomatal component of surface fluxes. The module has previously been examined and tested mainly in an oflline mode, but here we present for the first time calculations made with the module implemented in the full 3-D oxidant model.

This modelling is capable of differentiating between stomatal and non-stomatal ozone flux, and as such enables estimates of the absorbed ozone dose to be made for different vegetation types as a component of total ozone deposition. Stomatal ozone flux (or ozone uptake} is calculated with consideration of vegetation-specific phenology and physiological responses to environmental conditions and as such should improve spatial and temporal assessments of the possible risks of ozone damage to vegetation across Europe.

It should be emphasised that these results are presented for illustration only. Work is still required to improve many of the input databases to the new model, and to deal with soil moisture effects, before proper estimates of ozone uptake can be made. However, the basic features of the ozone uptake modelling are now in place and the results presented here are believed to represent a reasonable first estimate of the spatial and temporal patterns of ozone uptake that exist across Europe.

1.1 Background

An Ad-hoc Expert Panel Meeting on "Modelling and Mapping of Ozone Flux and Deposition to Vegetation" was held in Harrogate, UK, between 16-19th June 2002. This meeting was organised to assess the current scientific status of methods to model and map the deposition and flux of ozone to vegetation, in the context of their possible application in revised critical level assessments for ozone under the review of the Gothenburg Protocol. A main conclusion of the meeting was that there was general agreement that the flux approach represents an improvement on the AOT40 index for the following reasons:

- The existing Level I AOT40 approach is not appropriate for estimates of actual damage and as such should not be used to evaluate economic losses attributable to ozone.
- In contrast, the flux approach, if used in conjunction with appropriate flux-response relationships, could be used to estimate damage and hence economic estimates of ozone impacts.

- There seems to be strong agreement amongst the scientific community regarding the merits of the flux approach for Level II mapping.

2 Brief description of module

The deposition module has been described and evaluated in a series of publications (Emberson et al, 2000a, 2000b, 2001, Simpson et al., 2001, Tuovinen et al., 2001). The deposition flux of ozone at a particular height *z* is calculated as the product of the ozone concentration at height *z* and the deposition velocity, $V_{g,z}$, at that height. $V_{g,z}$ is calculated using a standard resistance approach, where the resistances used are (1) R_a - aerodynamic resistance between *z* and the top of the vegetation canopy; (2) R_b - the quasi-laminar layer resistance to ozone transfer; (3) R_c - the surface (canopy) resistance to ozone.

For the regional ozone modelling, we calculate fluxes to a number of land-cover classes within each grid square, using a sub-grid (mosaic) approach, similar to that used previously for the MADE model (Jakobsen et al., 1997, Simpson et al., 2002). Stability and turbulence are first calculated over each landuse, based upon the vegetation characteristics (roughness length, height, LAI). Deposition velocities are then calculated, which can be multiplied by the oxidant model's O_3 concentrations (both estimated at the same reference height) to estimate both total O_3 flux and the stomatal component.

The surface resistance is controlled by various parameters associated with the ground and external leaf surfaces, and by the stomatal conductance of the leaves. It is the stomatal conductance, G_{sto}, which governs the flux of ozone into the leaf itself, and thus controls ozone uptake to the plant. G_{sto} is calculated using a multiplicative model which has been parameterised for a number of different vegetation types:

$$G_{sto} = G_{max} \cdot f_{age} \cdot max\,\{f_{min}, f_{light} \cdot f_T \cdot f_{VPD} \cdot f_{SWP}\} \qquad (1)$$

where G_{max} is the maximum stomatal conductance (m S^{-1}), f_{min} is the minimum day-time stomatal conductance factor (0-1) and f_x are factors (from 0-1) describing the stomatal conductance relationship with phenology, photon flux density (light), leaf-temperature (T), leaf-to-air vapour-pressure deficit (VPD), and soil-water potential (SWP). For details of the functions used see Emberson et al. (2000b).

It should be noted that this module is used for all depositing gases in the rvl.2β unified model, with stomatal conductances for each gas simply a function of the stomatal conductance for ozone as specified above and the relative molecular diffusivity of the gas relative to ozone. Other conductances are introduced to allow for within-canopy deposition to the ground surface and to the external plant surfaces. These extra terms are often dependant on the solubility of the gas and, in these cases, the conductances are specified for SO_2 and O_3, and interpolated to other gases using reactivity-solubility scaling similar to that suggested by Wesely (1989).

The model structure is sufficiently flexible to include the deposition of gases, such as ammonia, where there is possible emission from the plant canopy alongside deposition, and work to extend the model parameterisation to cover a wider range of gases across Europe is continuing. As part of this process, the deposition model behaviour is being compared to measured flux data from sites in UK, the Netherlands and Germany over a range of vegetation types. In a comparison of ozone fluxes to semi-natural vegetation at Auchencorth Moss (UK), the EMEP model predictions using the 50x50 km^2 meteorology were compared against the deposition calculated from local meteorological observations and against the measured fluxes. There is considerable variability comparing individual observations, as expected, but the overall patterns are encouraging. The hourly median canopy resistances for the month of July 1997 give a diurnal pattern as shown in Figure 1. The box-model version of the deposition module used meteorological data from the Norwegian weather prediction model and compares well with both the measured canopy resistances and those calculated using local meteorology. The high overnight measured canopy resistances are not reflected in output from either model and the canopy resistances from the local meteorology are too high during the period 0900 to 1500. The effect of these discrepancies is being investigated further, but the comparison shows that the EMEP deposition module is achieving a good representation of land use specific canopy resistances.

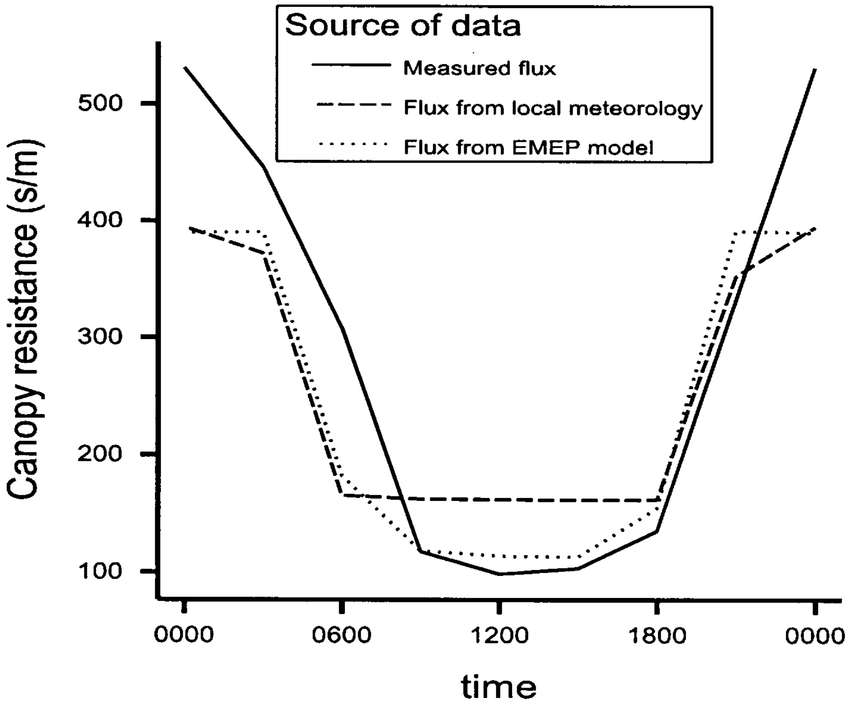

Figure 1: Calculated and Observed R_a for Auchecorth Moss (UK), July 1997

3 Input Data

In order to perform preliminary calculations of ozone flux as presented below, simplified input data have been used. In fact, at the present stage it is probably an advantage to use simple input data in order to better understand the outputs of the ozone uptake modelling. For more realistic estimates of ozone uptake to different vegetation types, these simplified inputs will be replaced by more detailed information.

3.1 Land-use, LAI

The land-use data base used in these calculations is based upon that provided by RIVM and used previously in the MADE model. The RIVM database consists of 8 different vegetation classes. Although the intention is to use a much more detailed land-use database from the Stockholm Environment Institute (SEI) in future, time precluded using this for the present study.

However, the following assumptions have been made in an attempt to mimic the SEI land-use classes and their characteristics as given in Emberson et al. (2000b). The RIVM classes do not distinguish between temperate and Mediterranean vegetation, and have a broad "other" vegetation category. For this preliminary study we have assumed that vegetation (forest, crops) north of 45°N is temperate, and south of 45°N is Mediterranean. North of 60°N we assume that RIVM's "other" category is "Tundra", south of 45°N we assume Mediterranean scrub, otherwise "moorland".
Leaf area index (LAI) is a key parameter in deposition modelling. Values of LAI are currently specified for each vegetation type as detailed in Emberson et al. (2000b). The one exception is that for forests we have reduced LAI (and forest height) by 50% north of the 62°, in order to generate more realistic values for Nordic conditions (e.g. Tuovinen et al, 2001).

3.2 Growing Seasons

Growing seasons for the different vegetation types are currently based upon the default parameterisations given in Emberson et al. (2000b). For crops a latitude-based growing season is used. In the Mediterranean the growing season starts around mid-March, whereas at 60°N it doesn't start until mid-May. This difference in growing season has important implications for the modelling of ozone fluxes.

4 The calculations

Here we present preliminary calculations from the EMEP model rv1.2$\beta$for the year 1999. It should be noted that soil moisture deficits have not been included in the current modelling scheme, this may generate significant overestimations of stomatal flux for some vegetation types, especially in southern Europe. However, the use of this algorithm for irrigated crops in such regions would be expected to provide realistic predictions of stomatal flux.

It should be noted that the calculations are given as nmol O_3 m^{-2} s^{-1} on a ground area basis. The data given in each figure do not represent the actual coverage of a particular vegetation type, but just the presence of such vegetation within the 50 x 50 km^2 EMEP grid. (Where a vegetation category is not present in the RIVM data base for a particular area, no results are shown).

5 Results

Figure 2 illustrates the monthly stomatal uptake of ozone for grasslands over Europe, from March to August. Grasslands were chosen for this first example, since they are present all year round and cover large areas of Europe. The seasonal variation in ozone uptake is apparent, with low ozone uptake values predicted for March followed by progressive monthly increases in ozone uptake to July from when values start to decrease again in August. An important feature of the months when ozone uptake is relatively high is that the fields of calculated uptake are rather uniform. For example, in June ozone uptake values range between 4-6 nmol O_3 m^{-2} s^{-1} over nearly the whole of Europe. (It should be noted though that the incorporation of the soil water deficit component of the modelling algorithm might reduce this uniformity somewhat, as uptake values in the Mediterranean region would be lower.)

Figure 3 illustrates the monthly stomatal uptake of ozone for crops, again from March to August. The strongest feature now is the marked seasonal variation. As mentioned in section 3.2 the growing seasons of crops are determined using a latitudinal model. Thus, the growing season starts in March in the Mediterranean, and not until May for Scandinavian countries. Further, for crops this season is assumed to last 3 months, so that in effect a band of growth moves northwards as the year progresses. This movement is clearly reflected in the uptake patterns in Figure 3.

6 AOT40

Figure 4 shows the AOT40 values predicted by the model for both crops and forests. Values are clearly highest in the Mediterranean areas, and very strong gradients exist between this area and the rest of Europe. These spatial patterns are clearly different from the O_3 uptake patterns for grasslands and crops presented above.

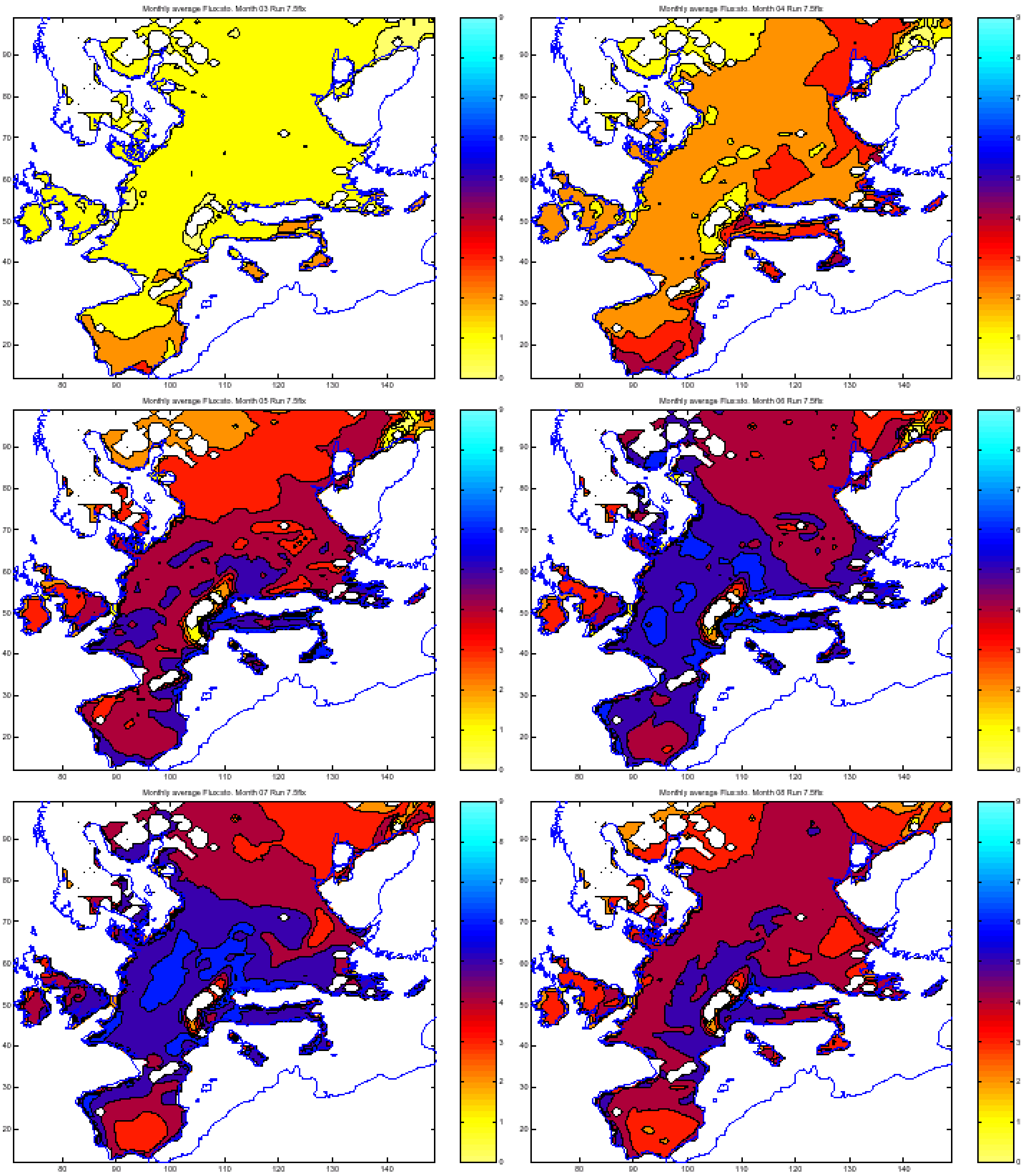

Figure 2: Estimated stomatal fluxes to grasslands, March (top left) to August (bottom right). Units: nmol m^{-2} s^{-1}. Estimates are only available where the landuse data-base shows grasslands, otherwise area is blank.

7 Conclusions

First, the caveats of this study need to be stated. The results are very preliminary, not least because the revised EMEP model is still very new and the deposition module just implemented. Many inputs are provisional, notably the leaf area index values, and growing season calculation. More work is needed to estimate soil moisture deficit. Clearly further analysis of the model outputs are required, and further evaluation of the model should also be performed, especially for areas in southern Europe where the reduction in stomatal conductance due to low humidities and soil moisture is a regular occurrence.

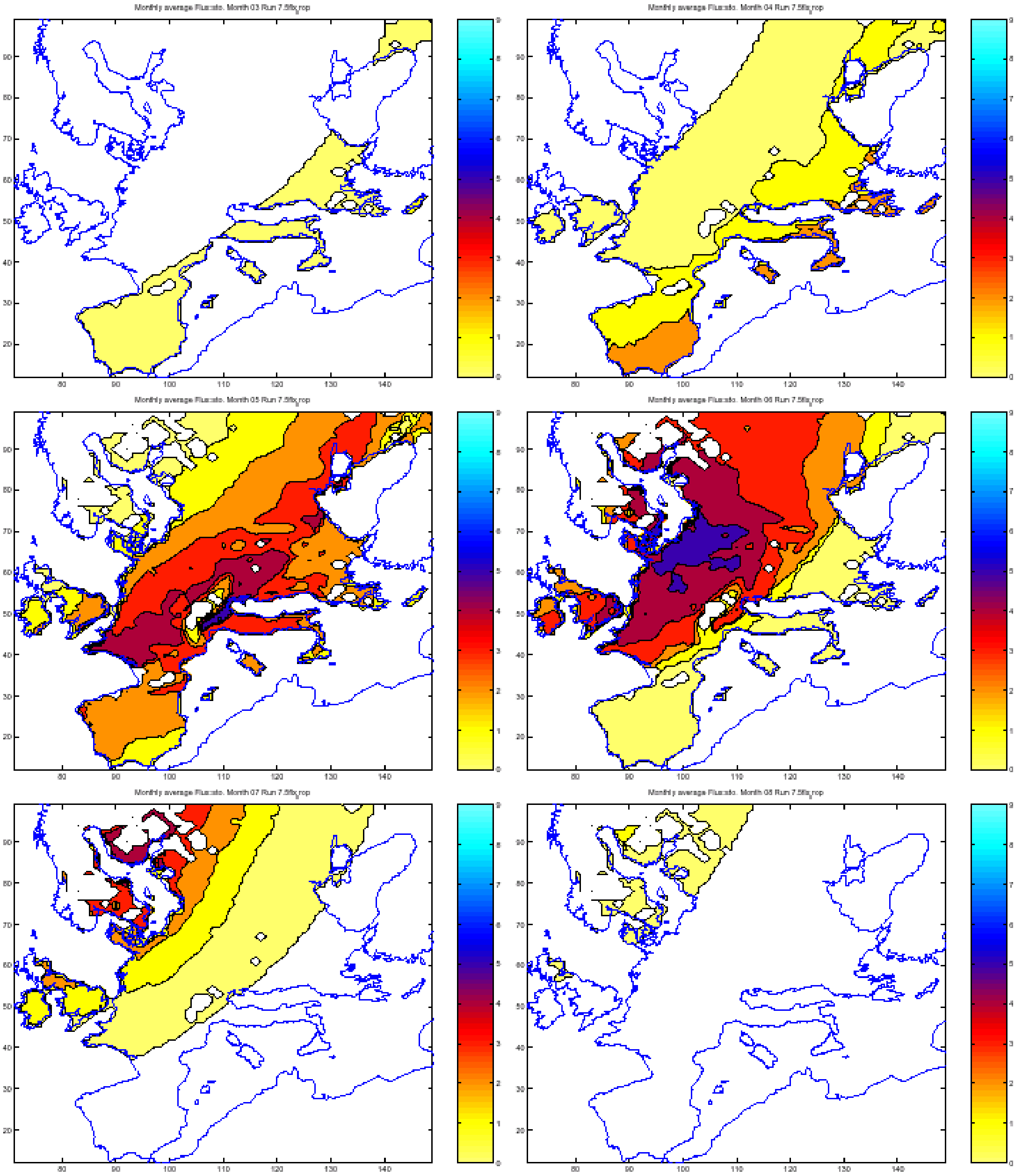

Figure 3: Estimated Stomatal fluxes to crops, March (top left) to August (bottom right). Units: nmol m^{-2} s^{-1}. Estimates are only available where the landuse data-base shows crops, otherwise area is blank.

Despite these caveats, the results presented above do allow a number of important conclusions to be made. It seems clear that patterns of ozone uptake and patterns of AOT40 can be very different. Further, ozone uptake rates seem to have a much more uniform distribution across Europe than AOT40. Similar results were found in an earlier study, based upon the Lagrangian model, of Emberson et al. (2000a) .The difference between AOT40 and uptake is largely due to the fact that in central and southern Europe, the meteorological conditions favour ozone formation (high temperatures, and hence high vapour-pressure deficits) tend to inhibit stomatal conductance and hence uptake.

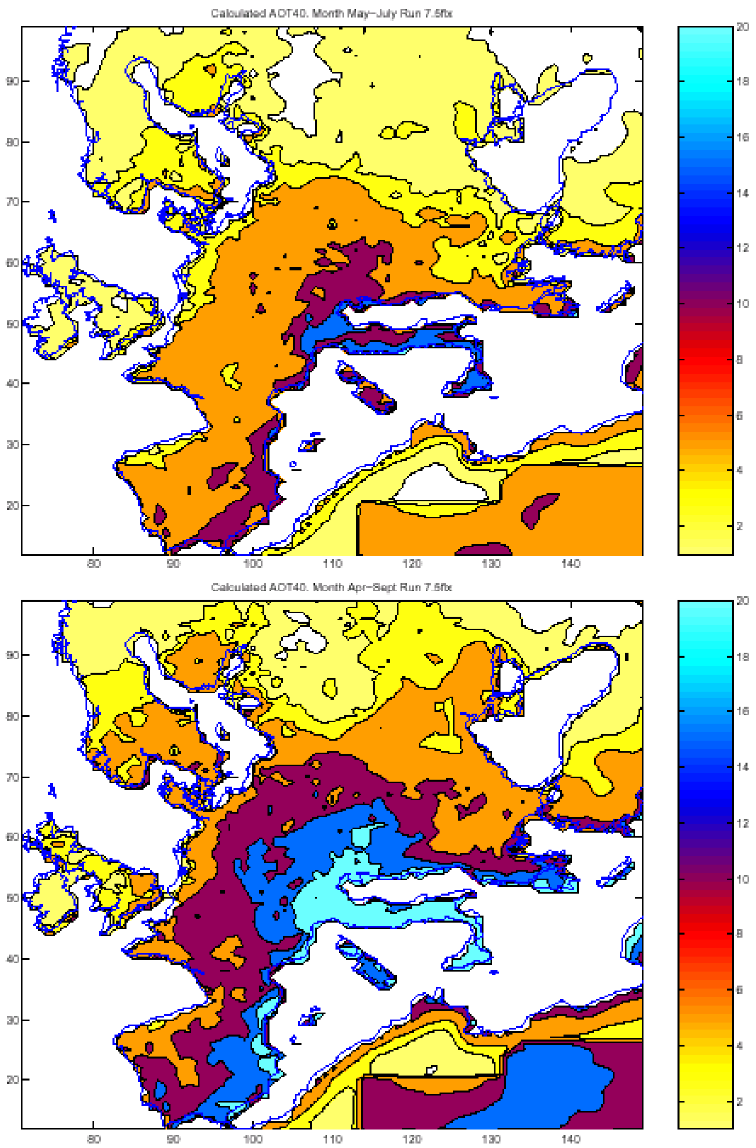

Figure 4: Calculated AOT40 for Crops (top) and Forests (bottom), from EMEP model (rv1.2.β) calculations for 1999. Units: ppm.h

Thus elevated ozone can contribute to the AOT40 index but not to uptake. In northern Europe, on the other hand, the cooler, more humid conditions, tend to promote stomatal conductance under typical conditions causing even moderate ozone concentrations to result in significant levels of ozone uptake to the plant. As such, concentrations of ozone below 40 ppb (common in northern areas), which are discounted entirely in the AOT40 index, may contribute to ozone uptake rates.

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