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# ANNALI

ISTITUTO SPERIMENTALE PER LA SELVICOLTURA

Special Issue on

## OZONE AND FOREST ECOSYSTEMS IN ITALY

Ozone measurements, critical levels and forest response indicators in selected forest ecosystems in Italy over the period 1996-2000

Report 2 of the Task Force on Integrated and Combined (I&C) Evaluation of the CONECOFOR programme

*F.D.C. 425: 180: (450)*



CONECOFOR (*CONtrollo ECOSistemi FOREstali*) is the intensive monitoring programme of forest ecosystems in Italy. The programme is framed within the Pan-European Level II Monitoring of Forest Ecosystems. It is co-sponsored by the European Union (EU) and co-operate with the UN/ECE ICP-Forests and the UN/ECE ICP-Integrated Monitoring. CONECOFOR is managed by the Ministero delle Politiche Agricole e Forestali (MiPAF), D.G. Risorse Forestali, Montane ed Idriche, Divisione V, CONECOFOR Service, acting also as National Focal Center (NFC) of Italy within the EU and UN/ECE programme.

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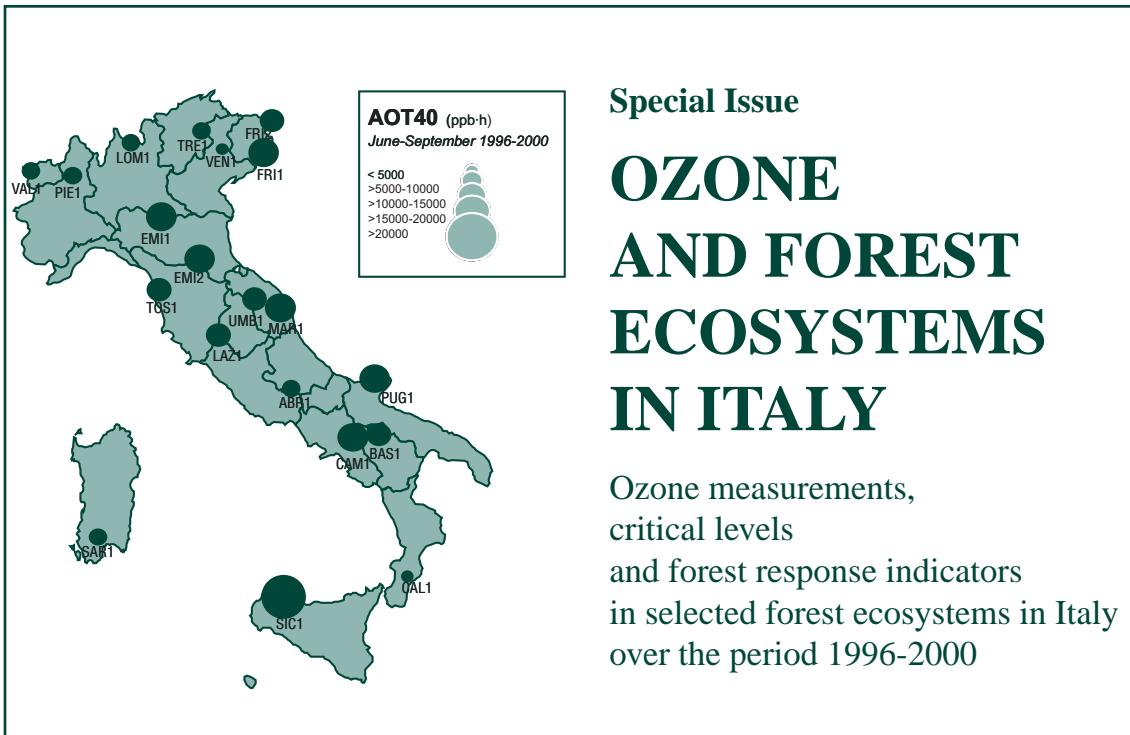
## ERRATA CORRIGE

Page	Location	Errata	Corrigé
Many	Throughout the papers in relation to AOT40 reporting unit	ppb*h <sup>-1</sup> , ppm*h <sup>-1</sup>	ppb*h, ppm*h



# ANNALI

## ISTITUTO SPERIMENTALE PER LA SELVICOLTURA



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# Ozone risk in the permanent plots of the Italian intensive monitoring of forest ecosystems - An introduction<sup>§</sup>

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**Summary** - Ozone ( $O_3$ ) is probably the most dangerous air pollutant for forest vegetation in Italy and elsewhere. Since 1996  $O_3$  has been (and is being) measured by passive dosimeters at the permanent monitoring plots of the CONECOFOR programme in Italy. In the year 2000, a 2001-2005 strategy for an integrated evaluation of forest intensive monitoring data was adopted by the CONECOFOR Task Force. Within this strategy, top priority was given to risk analysis in relation to  $O_3$  exposure. This paper provides basic information on the CONECOFOR programme, on  $O_3$  formation and regimes in Italy, on reported effects of  $O_3$  on forests, on the overall evaluation strategy adopted for the data generated by the programme, and on the steps undertaken to carry out Risk Analysis in relation to  $O_3$ .

**Key words:** *forest ecosystem, integrated evaluation, intensive monitoring, ozone, Risk Analysis.*

**Riassunto** - Il rischio ozono per le aree permanenti del monitoraggio intensivo degli ecosistemi forestali in Italia - Un'introduzione. L'ozono ( $O_3$ ) è probabilmente il contaminante atmosferico più pericoloso per le foreste in Italia ed altrove. Dal 1996,  $O_3$  viene misurato mediante dosimetri passivi presso le aree permanenti del programma CONECOFOR. Nel 2000 la Task Force del programma ha formalmente trovato accordo sulla strategia di valutazione dei dati per il periodo 2001-2005. Entro tale strategia, la valutazione del rischio connesso all'esposizione all' $O_3$  rappresenta la massima priorità. Questo articolo fornisce informazioni di base sul programma CONECOFOR, sulla formazione ed i regimi tipici per l' $O_3$  in Italia, sugli effetti conosciuti dell' $O_3$  sulle foreste, sulla strategia di valutazione integrata del programma CONECOFOR e sui passi compiuti per la valutazione di rischio connessa all' $O_3$ .

**Parole chiave:** *ecosistemi forestali, valutazione integrata, monitoraggio intensivo, ozono, analisi di rischio.*

The impact of tropospheric ozone ( $O_3$ ) on crops, natural vegetation and forest ecosystem is of considerable concern in Europe and elsewhere (FOWLER *et al.* 1999; FUMAGALLI *et al.* 2001; MATYSSEK and INNES 1999; MILLS *et al.* 2000; KARENLAMPI and SKARBY 1996). Current  $O_3$  levels (DE LEEUW and BOGMAN 2001), model predictions (FOWLER *et al.* 1999; ASHMORE *et al.* 2002; NEGTAP 2001) and a wide range of field and experimental evidence (see the section on effects of  $O_3$  on forest ecosystems in this paper) seem to justify the above concern. Mapped exceedance of the former critical level AOT40 10 ppm·h<sup>-1</sup> ( $O_3$  accumulated over threshold 40 ppb = 0.04 ppm, FUHRER *et al.* 1997) suggests that a considerable proportion of the European forest area is to be considered at risk (HETTENLING *et al.* 1996). However, these data are not easy to interpret in terms of effects: for example, while the occurrence of visible  $O_3$  injury seems to be widespread (INNES *et al.* 2001), effects on growth are much less certain (*e.g.* SPIECKER *et al.* 1996; BRAUN *et al.* 1999; FERRETTI *et al.*

2002). Yet, current  $O_3$  levels in Europe underscore the importance of the  $O_3$  issue in a monitoring programme aimed at assessing and monitoring air pollution effects on forests. Actually, it has been acknowledged that "especially the lack of  $O_3$  data is a serious limitation for the EU Intensive Monitoring (Level II) database" (DE VRIES 2000 p. 27). In addition,  $O_3$  data are highly relevant to other issues that are the object of important political agreements, like tropospheric chemistry changes and regional  $O_3$  formation (see *e.g.* the CLRTAP multi-pollutant, multi-effect directive; the UN Biodiversity Convention; the EU Habitat Directive; the EU acidification strategy, the UN/ECE CLRTAP, the EU Air Quality directive) (DE VRIES 2000 p. 19). This is the reason for which a section of the 2003 report about the intensive monitoring of forest ecosystems in Europe is devoted to  $O_3$  (EC - UN/ECE 2003).

Concern over  $O_3$  effects is especially great in the Alpine region (WOTAWA and KROMP-KOLB 2000) and in southern Europe (LORENZINI *et al.* 1994; SANZ and MILLAN 2000; PONT and FONTAIN 2000; NOLLE *et al.*

<sup>§</sup> Paper subject to review by members of the *ad-hoc* referees committee

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2002), especially in relation to the marked latitudinal (DOLLARD *et al.* 1995; MILLS *et al.* 2000) and altitudinal (BRONNIMANN *et al.* 2000) gradient of  $O_3$  and to some peculiar local dynamics (SANZ and MILLAN 1997; MILLAN *et al.* 2000; BAUMANN *et al.* 2001). Visual symptoms caused by  $O_3$  have been first observed on crops (*e.g.* LORENZINI *et al.* 1984) and subsequently observed and confirmed on trees, shrubs and herbs in parts of Eastern Europe, Greece, Italy, Spain and Ticino (Switzerland) (BYTNEROWICZ *et al.* 2001; MANNING *et al.* 2002; VELISSARIOU *et al.* 1992; SKELLY *et al.* 1999; SANZ and MILLAN 2000; COZZI *et al.* 2000; COZZI *et al.* 2002). In Italy there is a situation conducive to both acute and chronic  $O_3$  effects, with  $O_3$  peak values reaching about 200 ppb (*e.g.* FERRETTI *et al.* 1998) and the former AOT40 critical level of  $10\text{ppm}\cdot\text{h}^{-1}$  being frequently exceeded (ANGELINO *et al.* 1997; BUSSOTTI and FERRETTI 1998). This situation seems to be confirmed by the first data collected at the Permanent Monitoring Plots (PMPs) of the Italian intensive forest monitoring programme (termed CONECOFOR, see below) which reveal  $O_3$  weekly 24 hour averages often higher than 40 ppb, with peaks of 87 ppb (BUFFONI and TITA 2000) (Fig. 1). However, at the same time high  $O_3$  values can be counteracted by a suite of factors likely to reduce the sensitivity of trees and natural vegetation to  $O_3$ ; among others, Vapour Pressure Deficit (VPD) and soil moisture can be very important modifying factors under Mediterranean conditions (EMBESON *et al.* 2000). For this reason, while high concentrations do not mean *per se* effects on vegetation, they certainly create the basis for a concern and  $O_3$  was acknowledged as the top priority for the Integrated and Combined (I&C) evaluation of the data generated by the intensive monitoring data in Italy (FERRETTI 2000).

The aim of this paper is to introduce the second report of the I&C Task Force. In the following chapters we will briefly (i) provide basic information on the CONECOFOR programme, (ii) review some features characterizing the  $O_3$  situation in Italy, (iii) review the effects of  $O_3$  on forest ecosystems, and (iv) illustrate some basic features of the evaluation process that will be developed through the various sections of the present volume.

## The Conecofor programme

### Set up

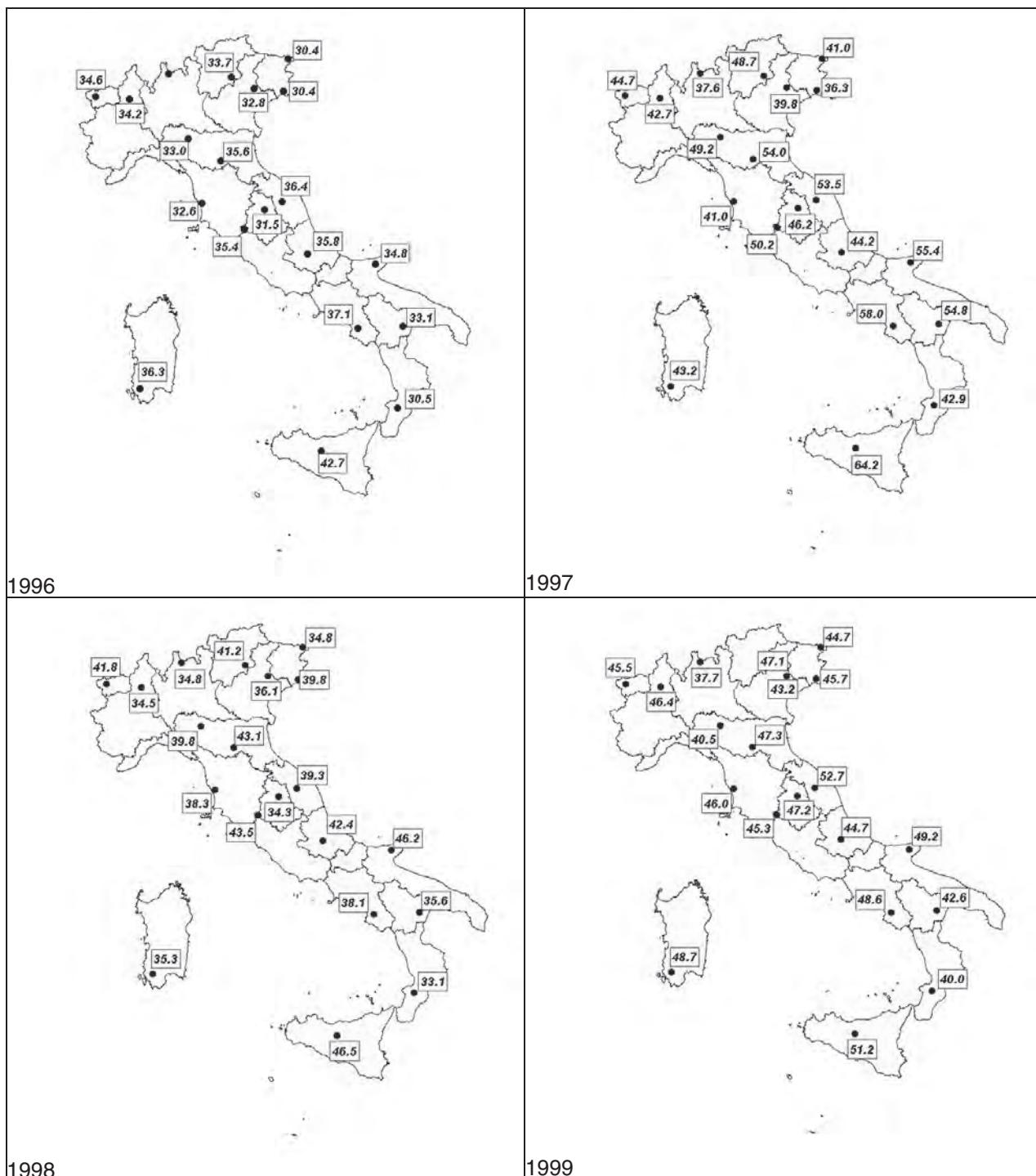
The Italian programme for the monitoring of

forests ecosystems (Italian acronym: CONECOFOR) started in 1995 within the framework of the intensive forest monitoring programme launched by the European Commission (Regulation EC n. 1091/94) and carried out under the auspices of the United Nation Economic Commission for Europe (UN/ECE) (ALLAVENA *et al.* 1999). CONECOFOR can be considered both an extension and an integration of several projects already existing at sub-national level in Italy. In 1995, 20 permanent monitoring plots (PMPs) were either incorporated from existing projects or selected and established *ex novo*. More recently, new PMPs have been incorporated and now CONECOFOR includes 28 PMPs (Table 1, Fig. 2). In all cases the locations of the PMPs were selected on a preferential basis. Therefore, while the PMPs of the programme are installed within important Italian forest ecosystem types, they should be considered as a series of case studies and not a statistically representative sample of the Italian forests. This implies that the results obtained through the programme do not allow for formal statistical inferences, *e.g.* findings cannot be extrapolated to sites other than those being monitored. However, it is reasonable to suggest that consistency of results from different sites spread throughout Italy can provide circumstantial evidence of trends that may occur also to other sites.

### Investigations

While the results are difficult to extrapolate to other sites, the major benefit of the intensive monitoring programme is to concentrate at the same site a number of different investigations that provide the basis for the integrated assessment. The investigations carried out at the PMPs include crown condition assessment, chemical content of soil and leaves/needles, deposition chemistry, gaseous air pollutants (mostly  $O_3$ ), tree growth, meteorological measurements and ground vegetation and remote sensing (Tables 2-3). Soil solution, streamflow chemistry, Leaf Area Index and litterfall are also investigated in several PMPs. Details relating to set up of the different investigations, data collection and first results are in ALLAVENA *et al.* (2001); an overview of the first results obtained is reported by FERRETTI (2000) and by MOSELLO *et al.* (2002).

The data collected from all the CONECOFOR PMPs are submitted to the European Commission and to the UN/ECE International Cooperative Programme



**Figure 1-** Mean seasonal ozone concentrations (in ppb) recorded by passive samplers in the period June-September 1996 - 1999 at the permanent monitoring plots of the CONECOFOR programme (after Buffoni and Tita 2000).  
*Concentrazioni medie stagionali di ozono (in ppb) rilevate con campionatori passivi nel periodo Giugno-Settembre 1996 - 1999 presso le aree di indagine permanenti (da Buffoni e Tita 2000).*

on Assessment and Monitoring Air Pollution Effects on Forests (ICP-Forests). Since 1997, 11 of the PMPs are also included among the sites of the International Co-

operative Programme on Integrated Monitoring of Air Pollution Effects on Ecosystems (ICP-IM) (Table 1).

**Table 1 -** Permanent Monitoring Plots (PMPs) of the CONECOFOR programme over the period 1995-2002. Asterisks indicate those PMP incorporated in the ICP-IM. In brackets: cases of PMPs installed outside the CONECOFOR programme and subsequently incorporated in the programme. The report will concentrate on the first 20 PMPs, e.g. those operational since 1995.  
*Aree permanenti del programma CONECOFOR nel periodo 1995-2002. Gli asterischi indicano le aree permanenti incorporate in ICP-IM. Tra parentesi: i casi di aree installate indipendentemente da CONECOFOR e successivamente incorporate nel programma. Il rapporto si concentrerà sulle prime 20 aree, cioè quelle attive dal 1995.*

PMP no.	Code	Lat	Long	Alt (m)	Main tree species	1995	1996	1997	1998	1999	2000	2001	2002
1	ABR1*	415051	133523	1500	Fagus sylvatica	+	+	+	+	+	+	+	+
2	BAS1	403638	155225	1125	Quercus cerris	+	+	+	+	+	+	+	+
3	CAL1*	382538	161047	1100	Fagus sylvatica	+	+	+	+	+	+	+	+
4	CAM1	402558	152610	1175	Fagus sylvatica	+	+	+	+	+	+	+	+
5	EMI1*	444306	101213	200	Quercus petraea	+	+	+	+	+	+	+	+
6	EMI2*	440631	110700	975	Fagus sylvatica	+	+	+	+	+	+	+	+
7	FRI1	454734	130715	6	Q.robur/Carpinus betulus	+	+	+	+	+	+	+	+
8	FRI2	462928	133536	820	Picea abies	+	+	+	+	+	+	+	+
9	LAZ1*	424950	130010	690	Quercus cerris	+	+	+	+	+	+	+	+
10	LOM1*	461416	93316	1190	Picea abies	+	+	+	+	+	+	+	+
11	MAR1*	431738	130424	775	Quercus cerris	+	+	+	+	+	+	+	+
12	PIE1	454055	80402	1150	Fagus sylvatica	+	+	+	+	+	+	+	+
13	PUG1	414910	155900	800	Fagus sylvatica	+	+	+	+	+	+	+	+
14	SAR1	392056	83408	700	Quercus ilex	+	+	+	+	+	+	+	+
15	SIC1	375432	132415	940	Quercus cerris	+	+	+	+	+	+	+	+
16	TOS1*	433034	102619	150	Quercus ilex	+	+	+	+	+	+	+	+
17	TRE1*	462137	112942	1775	Picea abies	+	+	+	+	+	+	+	+
18	UMB1	432757	122757	725	Quercus cerris	+	+	+	+	+	+	+	+
19	VAL1*	454326	65555	1740	Picea abies	+	+	+	+	+	+	+	+
20	VEN1	460326	120156	1100	Fagus sylvatica	+	+	+	+	+	+	+	+
21	ABR2	415409	142100	980	Q. cerris/C. betulus/Abies alba								+
22	LAZ2	415051	133523	190	Quercus ilex								+
23	LOM2	455726	100753	1150	Picea abies	(+)	(+)	(+)	(+)	+	+	+	+
24	LOM3	455441	93017	1250	Fagus sylvatica					+	+	+	+
25	TOS2	425212	104634	30	Quercus ilex	(+)	(+)	(+)	(+)	+	+	+	+
26	TOS3	434418	113422	1170	Fagus sylvatica	(+)	(+)	(+)	(+)	+	+	+	+
27	BOL1*	463516	112604	1740	Picea abies	(+)	(+)	(+)	(+)	(+)	+	+	+
28	LIG1	442410	92730	1290	Fagus sylvatica						(+)		+



### Tropospheric ozone formation and chemical regimes in Italy

$O_3$  formation occurs, in the troposphere, as a result of the photolysis of  $NO_2$  at wavelengths  $< 424$  nm (SEINFELD and PANDIS 1998),



where J is the constant of the  $NO_2$  photolysis and M represents a third molecule (e.g.  $N_2$ ,  $O_2$  or another) that absorbs the excess vibrational energy and stabilizes the  $O_3$  molecule formed. The  $O_3$  formed rapidly reacts with the nitrogen monoxide to reform

**Figure 2 -** The location of the Permanent Monitoring Plots of the CONECOFOR programme. Circles: PMPs operational since 1995 and considered within the report; squares: PMPs that have joined the programme at a later stage and not covered by the report. Map prepared by D. Rocchini, Dept. Environmental Sciences, the University of Siena.  
*Localizzazione delle aree permanenti di monitoraggio del programma CONECOFOR. I cerchi indicano le aree permanenti operative all'interno del programma sin dal 1995 considerate nel presente rapporto; i quadrati indicano le aree permanenti che sono state incorporate successivamente, ma che non sono considerate nel presente rapporto. Mappa preparata da D. Rocchini, Dip.to Scienze Ambientali, Università di Siena.*

**Table 2 -** Investigation categories carried out at the PMPs of the CONECOFOR programme. In brackets: cases of investigations formerly undertaken outside the CONECOFOR programme. Note: individual investigations may have covered only part of the 1995-2000 period. Details about the nature of the various investigations are provided by Allavena et al. (2001) and Ferretti (2000).  
*Categorie di indagini portate avanti presso le aree permanenti del programma CONECOFOR. Tra parentesi: indagini iniziate prima dell'incorporazione nella rete CONECOFOR. Nota: alcune indagini possono non avere coperto l'intero periodo 1995-2000. I dettagli sulla natura delle varie indagini sono riportati da Allavena et al. (2001) e Ferretti (2000).*

PMP no.	Code	Tree condition	Soil chemistry	Foliage chemistry	Tree growth	LAI	Litterfall	Ground vegetation	Deposition chemistry	Ozone meas.	Meteo meas.	Remote sensing
1	ABR1	+	+ (1)	+	+	+	+	+	+	+	+	+
2	BAS1	+	+	+	+	+	+	+		+	+	+
3	CAL1	+	+	+	+	+	+	+	+	+	+	+
4	CAM1	+	+	+	+	+	+	+	+	+	+	+
5	EMI1	+	+	+	+	+	+	+	+	+ (3)	+	+
6	EMI2	+	+	+	+	+	+	+	+ (2)	+	+	+
7	FRI1	+	+	+	+	+	+	+	+	+ (3)		+
8	FRI2	+	+	+	+	+	+	+	+ (2)	+	+	+
9	LAZ1	+	+ (1)	+	+	+	+	+	+ (2)	+	+	+
10	LOM1	+	+	+	+	+	+	+	+	+	+	+
11	MAR1	+	+	+	+	+	+	+	+	+		+
12	PIE1	+	+	+	+	+	+	+	+ (2)	+	+	+
13	PUG1	+	+	+	+	+	+	+		+ (3)		+
14	SAR1	+	+	+	+	+	+	+		+ (3)		+
15	SIC1	+	+	+	+	+	+	+	+	+		+
16	TOS1	+	+	+	+	+	+	+	+	+ (3)	+	+
17	TRE1	+	+	+	+	+	+	+	+ (2)	+	+	+
18	UMB1	+	+	+	+	+	+	+		+		+
19	VAL1	+	+	+	+	+	+	+		+ (3)	+	+
20	VEN1	+	+	+	+	+	+	+	+	+	+	+
21	ABR2											
22	LAZ2											
23	LOM2	+	(+)	+	+		+	+	+	+		
24	LOM3	+		+			+	+	+	+	+	
25	TOS2	+	(+)	+	+	+	+	+	+	+	+	
26	TOS3	+	(+)	+	+	+	+	+	+	+		
27	BOL1	+	(+)	+	(+)	(+)	(+)	+	+ (2)	+	+	+
28	LIG1	+	(+)	(+)				+	+	+	+	

(1) plus soil solution chemistry

(2) plus streamflow chemistry

(3) plus  $\text{SO}_2$

$\text{NO}_2$  according to



where K is the constant of reaction speed.

During sunny periods the above reactions achieve the so-called *photostationary equilibrium* and the *steady-state*  $\text{O}_3$  concentration can be estimated by

$$[\text{O}_3] = \frac{J[\text{NO}_2]}{K[\text{NO}]} \quad (\text{iv})$$

The  $J/K$  ratio varies during the day, depending on temperature (influence on  $K$ ) and the intensity of the ultraviolet component (290-420 nm) of the solar radiation (influence on  $J$ ).

During daytime the  $\text{O}_3$  can also photolise to produce both ground-state ( $\text{O}$ ) and excited singlet

**Table 3 -** Year of start, number of PMPs considered, sampling regime and available repetitions for the various investigations. Only the plots starting in 1995 and the period 1995-2000 are considered.

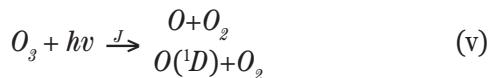
*Anno di avvio, numero di aree interessate, regime di campionamento, ripetizioni e unità di riporto dei dati per le varie indagini. Sono considerati solo il periodo 1995-2000 e le aree attive dal 1995.*

Investigation category	Start (1)	No. of PMPs	Sampling regime	Repetition available
Soil chemistry	1995	20	10y	95
Tree growth	1996-1997	20	5y	97, 00
Foliage chemistry	1995	20	2y	95, 97, 99
Ground vegetation	1996	20	1y	96, 98, 99, 00
Tree condition	1996	20	1y	96, 97, 98, 99, 00
LAI	1997	20	1y	97, 98
Litterfall	1997	20	1m	97, 98
Ozone measurements	1996	20	1w	96, 97, 98, 99, 00
Deposition chemistry	1998	13	1w	98, 99, 00
Soil solution chemistry	1999	2	1w	99, 00
Streamflow chemistry	1998	2	1w	98, 99, 00
Meteo measurements	1998	9	continuous	98, 99, 00

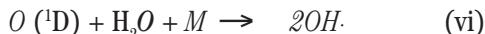
(1): plots installed before the set up of the CONECOFOR programme may have longer time series

y: year; m: month; w: week

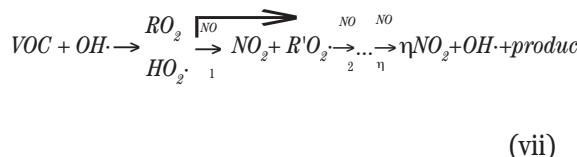
(O(<sup>1</sup>D)):



Once formed O(<sup>1</sup>D) collides with a third molecule (M), removing the excess energy and quenching O(<sup>1</sup>D) to its ground-state O, forming the hydroxyl radical (OH<sup>·</sup>)



The role of this radical is very important because of its high reactivity with most atmospheric trace species, allowing their removal from the lower atmosphere. An example is the reaction of the OH radical with VOC (Volatile Organic Compounds) that leads to the formation of RO<sub>2</sub> and HO<sub>2</sub> radicals, that may react with NO to produce NO<sub>2</sub> and other intermediate radicals. Once formed, these ones reacts with NO to produce NO<sub>2</sub> and other intermediate radicals. The result of such process is a chain reaction, with OH<sup>·</sup> as the chain carrier, leading to a number  $\eta$  of NO<sub>2</sub> molecule produced, for each molecule of VOC consumed, without consuming OH. Hydroxyl radical is then available to react with other VOC molecules (DIMITRIADES 1996), and so restarting the chain:



According to (iv), the steady-state O<sub>3</sub> concentration is proportional to the NO<sub>2</sub>/NO ratio and so the effects of such chain is to produce  $\eta$  O<sub>3</sub> molecules for each NO molecule available. At high NO<sub>x</sub> (=NO+NO<sub>2</sub>) levels this process is interrupted by the reaction of nitrogen dioxide with hydroxyl radical to form nitric acid:



From the discussion above, it is clear that the relationship between O<sub>3</sub> and its precursors (NO<sub>x</sub> and VOC) is far to be linear. The comprehension of the O<sub>3</sub>

behaviour calls in fact to an improved understanding of the relationship and the sensitivity of ambient O<sub>3</sub> concentration to volatile organic compounds and nitrogen oxides emissions at regional scale.

At this purpose, SILMAN (1995) found that VOC and NO<sub>x</sub> predictions are correlated with concentration values of certain species or 'indicator ratios', involving peroxides and total reactive nitrogen NO<sub>y</sub> (= NO<sub>x</sub> + NO<sub>2</sub>; where NO<sub>2</sub> represent the sum of oxidation products of NO<sub>x</sub>, i.e. NO<sub>2</sub> = HNO<sub>3</sub> + organic nitrates + inorganic nitrates). Following the so-called *indicator approach*, three chemical regimes can be generally distinguished (SILMANN 1995, 1999):

- *VOC sensitive*, typical of urban areas, characterised by an increase of O<sub>3</sub> concentrations with increasing VOC emissions (and an unchanged or decreasing O<sub>3</sub> with increasing NO<sub>x</sub> emissions);

- *NO<sub>x</sub> sensitive*, typical of rural areas, characterised by an increase of O<sub>3</sub> concentrations with increasing NO<sub>x</sub> emissions (and an unchanged or decreasing O<sub>3</sub> with increasing VOC emissions);

- and *transitional*, that can be thought as a broad region that divides the previous two regimes.

An example is given in Table 4, reporting literature threshold values for NO<sub>x</sub> sensitivity of some indicators.

Atmospheric simulation models are the typical tools that can be employed to quantitatively estimate the relationship between ambient O<sub>3</sub> and precursors emissions. The use of 3D Eulerian models to set-up emission control strategies is although affected by some uncertainties, mostly related to the availability and the quality of input data: uncertainties in emission inventories, in the horizontal and vertical dispersion and in boundary conditions. Recent studies suggest to encourage the direct use of measured values of indicators, to identify photochemical regimes and to provide an indirect method to evaluate the accuracy of model predictions (e.g. see the project web sites of EUROTRAC-2 LOOP: <http://loop.web.psi.ch/> and the VOTALP I and II <http://www.boku.ac.at/imp/votalp/>). Specifically, the analysis of differences between weekday and weekends O<sub>3</sub> concentrations may help in identifying which pollutant reduction strategy (NO<sub>x</sub>

**Table 4 –** Threshold values for  $\text{NO}_x$ -sensitivity of afternoon concentrations or ratios.  
*Valori soglia per la sensibilità ad  $\text{NO}_x$  delle concentrazioni pomeridiane o dei rapporti di concentrazione.*

Indicator	Threshold	Reference
$\text{NO}_x$	< 10 to 25 ppb	Milford et al., 1994; Sillman, 1995
$\text{O}_3/\text{NO}_2$	> 6 to 11	Sillman, 1995
$\text{HCHO}/\text{NO}_2$	> 0.2 to 0.4	Sillman, 1995
$\text{H}_2\text{O}_2/\text{HNO}_3$	> 0.3 to 0.5	Sillman, 1995

or VOC control) can be more effective in reducing ambient  $\text{O}_3$  concentration. In northern California, as an example, the presence of higher weekend  $\text{O}_3$  concentrations suggests that the need for VOC control is greater than the one for  $\text{NO}_x$  control (ALTSCHULER *et al.* 1995).

The local abundance of  $\text{O}_3$  precursors is not the only factor that can explain the accumulation of  $\text{O}_3$  in the lower troposphere. Two other processes need to be considered: horizontal advection and the vertical mixing of pollutants. The first process is particularly relevant for the Italian peninsula, characterised by the occurrence of breezes, induced by topographic features (mountain-valley and land-sea). Such local wind regimes have the effect of recirculating  $\text{O}_3$  and other pollutants (mainly  $\text{NO}_2$ ) present in "aged" plumes coming from urban areas and other relevant sources. This is especially true for smaller domains, where concentrations at the inflow play an important role, since they largely determine the levels of species with lifetimes similar (or even smaller) to the time required to cross the entire domain. In such areas is fundamental to take into account the pollutants emitted locally as well as the recirculation of "aged" air coming from surrounding regions. Vertical mixing is an important process for large areas in Italy, characterised by the frequent occurrence of high pressure situations, especially in the Northern Po Valley, that inhibit the vertical dispersion and the dilution of pollutants in the atmosphere.

#### **Integrated assessment approach**

The above considerations evidence the complexity and the synergic effects of the factors that affects the formation of ground-level  $\text{O}_3$  and the consequent exposure of ecosystems. This complexity has been recognised by the Council Directive 96/62/EC on ambient air quality assessment and management, that introduced the concept of "integrated assessment", with two main objectives in mind:

- 1) to assess air quality, with respect to limit values and alert thresholds, throughout the territory of the Member States;
- 2) to take measures to ensure that a plan or programme is prepared or implemented to attain the limit value within a specific time (in areas where levels are higher than the limit value), or maintain the levels of pollutants below the limit values and to preserve the best ambient air quality, compatible with sustainable development (in areas where levels are lower than the limit value).

According to the Directive, air quality assessment and the evaluation of emission control strategies for  $\text{O}_3$  should be done by means of a combination of measurements, emission inventories and modelling techniques.

The successive Directive 2001/81/EC, derived from Gothenburg Protocol of 1 December 1999, under the United Nations Economic Commission for Europe (UN-ECE) Convention on Long-Range Transboundary Air Pollution (LRTAP) to abate acidification, eutrophication and ground-level  $\text{O}_3$ , fixes national emission ceilings for the Member States to be reached by the year 2010 at the latest. These emission ceilings have the purpose to meet broadly the following interim environmental objectives: to respect the critical loads for acidification; to respect the health- and vegetation-related critical levels for ground-level  $\text{O}_3$  exposure. Forested areas, generally characterised by  $\text{NO}_x$  sensitive regimes, are affected by the atmospheric transport of manmade  $\text{O}_3$  precursors but also by direct emissions of hydrocarbon from vegetation, often comparable to or even larger than anthropogenic ones. Biogenic hydrocarbons (BIOVOC) are reactive in presence of atmospheric oxidants, as is the case of isoprene, that is highly reactive with OH,  $\text{NO}_3$  and  $\text{O}_3$ . Primary and secondary BIOVOCs can also condense, depending on their volatility, to pre-existing aerosol participating to complex heterogeneous phase reactions that lead to the formation of so-called SOA (Secondary Organic Aerosols).

#### **Modelling techniques**

For the UN-ECE LRTAP convention source-receptor relationships need to be calculated, in order to quantify the effects of VOC and  $\text{NO}_x$  reductions in individual countries on the seasonally averaged  $\text{O}_3$  concentrations (as well as AOT or alternatives, *e.g.* flux) over target areas. For this purpose, the EMEP

model (SIMPSON 1992) was developed and it is currently the only model framework routinely doing this work on a continental scale, at a 50x50 km<sup>2</sup> horizontal resolution.

The increasing availability of experimental data (meteorological and chemical), high resolution emission inventories and modelling tools (detailed emission models, GIS - geographic information systems, photochemical models, and so on) makes it now possible to properly evaluate the effects of urban plumes and local phenomena that can not be fully captured by the coarse grid adopted by EMEP model. A review of regional photochemical model applications in Italy (resolution of few km), made in 1999 (DESERTI *et al.* 2001; available on line <http://www.sinanet.anpa.it/documentazione/default.asp>), evidenced a larger number of studies in Lombardia (SILIBELLO *et al.* 1998) and Emilia-Romagna regions (DESERTI *et al.* 2001), and still preliminary applications in Liguria, Lazio, Piemonte, Toscana, Puglia and Campania regions (DESERTI *et al.* 2001; LIGUORI *et al.* 2001). At authors' knowledge, only two Alpine sites have been investigated by using photochemical models: Valtellina - a valley situated in Lombardia region (PIROVANO *et al.* 2001; 2002) - and Valle d'Aosta (ARIANET 2001). In the first case a modeling system based on CALGRID model (YAMARTINO *et al.* 1992) has been applied, while the second study has been performed by using a modelling system integrating meteorological models, emission inventory processors and an updated version of STEM-II chemical transport model (CARMICHAEL *et al.* 1991). As an example from the latter study, Figure 3 shows a comparison between observed and predicted O<sub>3</sub> levels at La Thuile (Valle d'Aosta, the site where the CONECOFOR PMP VAL1 is located), where a three days O<sub>3</sub> episode was considered on a domain of 40x60 km<sup>2</sup>, with an horizontal resolution of 1 km. The example confirms the capability of an integrated modelling system, fed with adequate data, to correctly reproduce observed O<sub>3</sub> concentrations and, in perspective, to estimate AOTs.

#### Evidences from monitoring data

The idea of analysing weekday and weekend O<sub>3</sub> concentrations to detect photochemical regimes, previously mentioned, has been also investigated for Italian sites. Hereafter (Figure 4) we give an example considering weekday and weekend hourly values for July 1999, at some monitoring stations distributed

over Italy. The analysis of this figure evidence maximum O<sub>3</sub> levels at Carate Brianza (close to Milano urban area) and at Isola Serafini (rural site), both located in Po valley, confirming the abundance of O<sub>3</sub> precursors in this area. The VOC *sensitive* regime that seems to characterize the first station, evidencing higher O<sub>3</sub> values during weekends, has been also revealed for the Milano metropolitan area applying the indicator approach to O<sub>3</sub> fields computed under different NOx and VOC emission patterns (SILIBELLO *et al.* 1998, 2000). This preliminary analysis of ambient O<sub>3</sub> data suggests the adoption for VOC control for this area, but certainly claims for a deeper analysis of meteorological, emission and air quality data, helped by the use of realistic modelling tools, in order to set up effective O<sub>3</sub> abatement strategies.

At Isola Serafini, the higher weekday values seem to be related to the influence of NO<sub>2</sub> rich plumes coming from major urban areas in Po Valley that, once photolised, lead to the increase in the afternoon O<sub>3</sub>.

A characteristic "background" behaviour is otherwise observed at La Thuile station (CONECOFOR PMP VAL1), with the absence of a marked diurnal O<sub>3</sub> patterns and no differences between weekend and weekday time series. Intermediate patterns between urban and background sites are shown at the other sites, where differences between weekend and weekdays are probably due to a combination of both meteorological processes (breezes, extension of the mixing layer, *etc.*) and local emissions (*e.g.* increase of traffic emissions during weekend at vacation areas, *etc.*). This supports once more the importance of

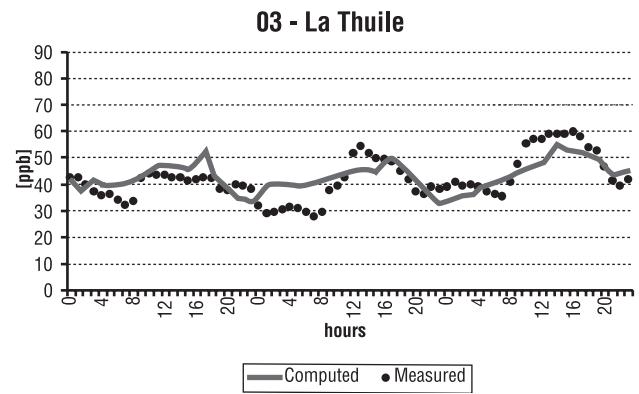
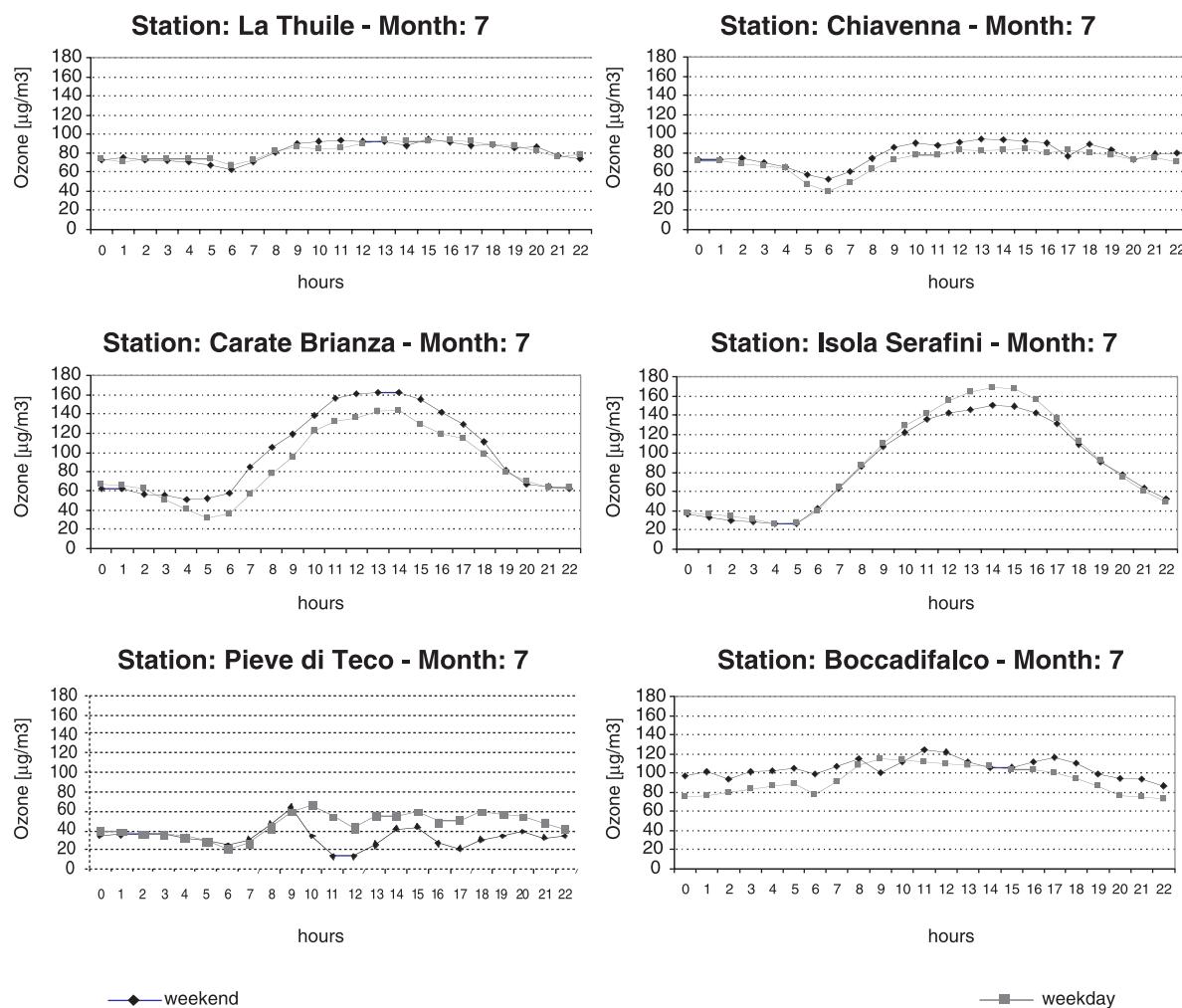


Figure 3 - Example of a comparison between observed and predicted ozone levels at La Thuile (PMP VAL1) for the period 10-12/08/2000.

*Esempio di comparazione tra livelli osservati e previsti presso il sito di La Thuile (PMP VAL1) nel periodo 10-12/08/2000.*



**Figure 4** - Mean hourly ozone at different monitoring stations from North (La Thuile - Valle d'Aosta) to South Italy (Bocca di Falco - Sicilia) during July 1999.  
*Medie orarie delle concentrazioni presso differenti stazioni di monitoraggio da Nord (La Thuile - Valle d'Aosta) a Sud Italia (Bocca di Falco - Sicilia) nel Luglio 1999.*

an integrated approach to the problem, allowing to identify relationships between precursors emissions and ambient concentrations on the basis of both monitoring data and modelling systems results.

#### Final remarks

The innovative approach implied in integrated assessment procedures, introduced by Directive 96/62/EC, represents an useful framework by which assess and manage air quality. This framework combine measurements, emission inventories and modelling techniques, enabling to: 1) consider the effects of different processes that affect  $O_3$  levels (meteorology, emissions, transport from surrounding regions, etc.)

and 2) evaluate *a priori* the effects of different emission control strategies on predicted  $O_3$ . Model- and observation-based methods used to identify the photochemical regimes provide useful information about the effectiveness of emission strategies to reduce vegetation exposure to ground-level  $O_3$ . As shown in the example given for Milano urban area, the analysis of weekday and weekend  $O_3$  concentrations gives indications for emission control strategies design that are coherent with the ones obtained by means of  $NO_x$ -VOC- $O_3$  sensitivity modelling study. Moreover, preliminary application of photochemical models to forest sites in Italy seems to provide calculated  $O_3$  levels rather close to the observed ones. This suggests

the possibility of a more intensive application of such tools to different forested areas and long time periods (April - September), in order to estimate the spatial distribution of the vegetation exposure indexes over large areas. A wider application of photochemical models under a broad range of different emission and meteorological conditions should also increase our knowledge about such tools and their capability of representing real-world conditions.

## Ozone and forest ecosystems: an overview

The first report of field damage on forest trees by  $O_3$  was made in 1961 (BERRY and RIPPERTON 1963) but  $O_3$  had been identified as "a menace to agriculture" since 1959 (RICH 1964). As scientific research devoted increasing attention to the topic, many excellent reviews discussing the effects of  $O_3$  at cellular, biochemical, physiological, morphological and ecosystem levels became available. The aim of this section is to give an overview of  $O_3$  effects on different aspects of forest tree life and on ecosystem functions and processes, including the results of the most recent scientific research.

### ***Molecular mechanisms of $O_3$ toxicity***

The essential elements of the biological mechanisms of  $O_3$  toxicity have been reported by GUDERIAN *et al.* (1985). Damage produced by photochemical oxidants is the result of biochemical and physiological reactions occurring in the mesophyll. Decisive factors in causing these injuries are: the quantity of  $O_3$  that spreads from the atmosphere to the reactive sites within the leaf and the speed at which it spreads. The flux of  $O_3$  is a function of the chemical and physical characteristics it encounters during the transition from gaseous to liquid phase. Physical structures can hinder its flux; chemical reactions can capture  $O_3$  and other photochemical molecules between the gaseous and the liquid phase.

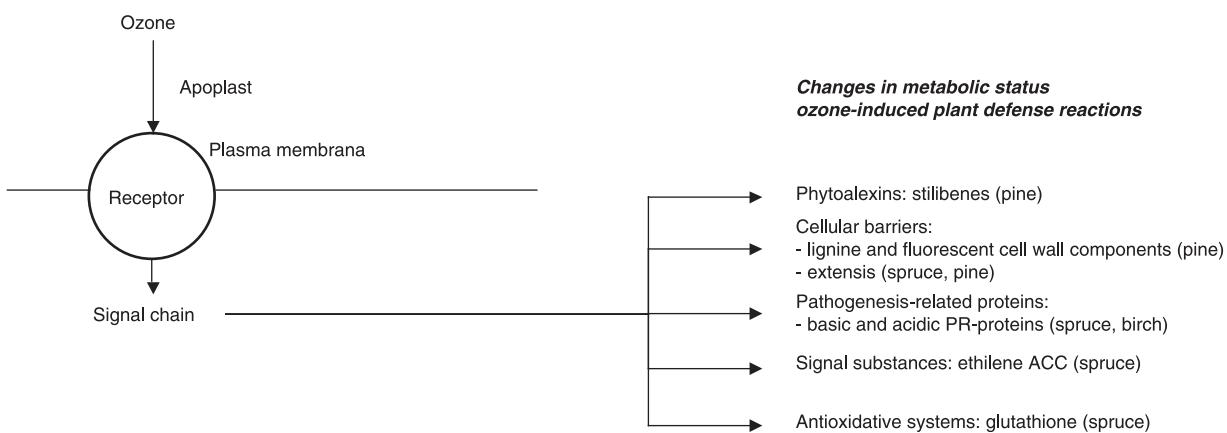
The flux of  $O_3$  in the mesophyll is considered a factor capable of predicting foliar injury, although the onset of damage does not always correlate with the quantity of  $O_3$  absorbed.  $O_3$  flows to the cellular sites where the injury occurs in the liquid phase. This transition through phenomena of diffusion and/or flux crosses the hydrated cellular walls, the membranes and the cytosol. Along the way  $O_3$ -consuming scavenger reactions can be triggered. In cellular solutions, a

variety of organic molecules, such as unsaturated fatty acids, sulphhydryls and ring compounds, are  $O_3$ -sensitive. The reaction of these compounds with  $O_3$  produces hydrogen peroxide ( $H_2O_2$ ) and free radicals, against which the cell is protected by scavenging and buffering mechanisms, such as superoxide dismutase, peroxidase, glutathione and phenolic compounds. These processes are to a large extent extracellular and occur within the wall (apoplast), while the general  $O_3$  induced defensive reactions in plants are summarized in Fig. 5. Consequently, the development of the apoplastic fraction - and therefore the density of the mesophyll - provides a substrate in which the toxic action of  $O_3$  can be neutralized (LYONS *et al.* 2000). The plant's capacity to activate similar detoxifying mechanisms, combined with the intensity of the  $O_3$  flux in the leaves, explain the different degrees of sensitivity.

### ***Ultrastructure damage***

Organelle membranes, and especially chloroplast membranes, are considered the most sensitive targets to  $O_3$  injury. The most common ultrastructure alterations are thylakoid swelling and plastoglobule production, while the stroma becomes granulated and markedly electron-dense (SUTINEN *et al.* 1990; SELLDÉN *et al.* 1996; HOLOPAINEN *et al.* 1996). This type of damage has been observed both in conifer needles and in broadleaf leaves. It can be detected when the symptoms are visible, but is also considered a "previsual" manifestation, *i.e.* it allows one to ascertain the presence of  $O_3$  impact even without the presence of visible symptoms. Damaged chloroplasts are followed by collapse of mesophyll cells and - sometimes - also of epidermal cells. In broadleaved trees the injury mainly affects the palisade mesophyll: this is considered a specific symptom attributable to  $O_3$ , since other pollutants tend to damage the lacunar tissue cells nearest the substomatal chambers. The collapsed cells are isolated from the healthy ones through compartmentalization processes, including the formation of a layer of callose (GRAVANO *et al.* 2003). EVANS *et al.* (1996) found a proportionality between the percentage of foliar surface affected by visible symptoms and the percentage of collapsed mesophyll cells, although cellular damage is normally greater than visible injury.

Other alterations of the apoplastic fractions have also been described: they can be considered



**Figure 5-** General scheme of the ozone-induced plant defense reactions (from Sandemann 2000, redrawn). In brackets the forest trees in which the reaction has been localized.  
*Schema generale delle reazioni di difesa indotte dall'ozono (da Sandemann, 2000, ridisegnato). Tra parentesi le specie su cui è stata identificata la risposta.*

acclimatization mechanisms. They include thickening of the cell walls and production of pectic substances, thickening of cuticles which become imbibed in phenolic substances such as tannins or phenylpropanoids (GÜNTHARDT-GOERG *et al.* 2000; BUSSOTTI *et al.* in press). Another category of alterations affects the photosynthetate transport system, with the collapse of phloem cells and the accumulation of primary starch in the chloroplasts (GÜNTHARDT-GOERG *et al.* 1997; SODA *et al.* 2000). Lastly, a great deal of attention has been devoted to the condition of outer surfaces, in relation to the action of both O<sub>3</sub> and other pollutants (TURUNEN and HUTTUNEN 1990). It is mainly the epicuticular and epistomatal waxes of conifer needles that lose their normal fibrillar network structures, with possible consequences on the efficiency of stomatal regulation. These alterations have been considered as markers useful in recognizing the effects of atmospheric pollution in the field.

#### Effects on physiology

According to GUDERIAN *et al.* (1985) and DARRAL (1989), O<sub>3</sub> attacks the chloroplast membrane, thus hindering the photosynthesis processes connected to it. It further inhibits the carboxylic activity of Rubisco reducing the fixation of CO<sub>2</sub>. Chlorophyll reduction caused by O<sub>3</sub> has been reported in several species. Chlorophyll *b* is more sensitive than chlorophyll *a*. The reduction can also occur in chronic exposure conditions or - in a previsual phase - even without the presence of symptoms. Reduced photosynthesis can be the effect of permanent injury to the mesophyll or of a

temporary stomatal restriction (stomatal conductance is inhibited by high concentrations of O<sub>3</sub>; MINNOCCI *et al.* 1999). In this last case, full physiological function is restored as soon as exposure ceases. An important question concern the O<sub>3</sub>-induced decoupling between assimilation and stomatal conductance reported for broadleaves (MATYSSEK *et al.* 1991; FLAGER *et al.* 1994; TJOELKER *et al.* 1995; CLARK *et al.* 1996; PAOLETTI *et al.* 2002) that may be suggested also for conifers (WIESER *et al.* 2002).

Photosynthesis reduction also brings about a reduced production of soluble sugars and starch, which in turn reduces growth (GUDERIAN *et al.* 1985); important resources are subtracted from the growth process to be used in the detoxification process. Furthermore, sugars and primary starch are retained in the leaves (probably due to the collapse of the leaves' phloem system, WELLBURN and WELLBURN 1994) and the transport of photosynthetates towards the roots is inhibited. Thus root growth is also reduced and the normal ratio between above- and below-ground structures is altered, in favour of the former. Reduced sugar and starch reserves in the roots can explain the alteration of the symbiotic relationship with the soil's fungine component (mycorrhizae) causing a reduction in the efficiency of mineral nutrition.

A loss of photosynthetic efficiency in individual leaves does not necessarily imply a similar loss of efficiency for the whole tree, in that certain compensation mechanisms may be set in motion (cf. KOLB *et al.* 1998). In fact, not all the leaves of an adult tree or of a forest stand are affected the same way.

Usually it is the most exposed leaves that suffer the greatest damage. Unaffected leaves develop a greater photosynthesis efficiency so that neither the tree's overall productivity nor its growth are jeopardized (TEMPLE and MILLER 1994). Damaged leaves undergo premature ageing processes and may be shed and replaced by new, more efficient leaves.

#### **Visible symptoms**

In forest species, visible foliar symptoms attributable to chronic exposure to  $O_3$  were described for the first time in the mountain pines of the south-west of the United States, a region with a Mediterranean-type climate (FOX and MICKLER 1995; MILLER and McBRIDE 1999). The symptoms included chlorotic mottle, or local yellowing, degenerating into necrotic patches; in the more severe cases needle tip necrosis was also observed. The chlorotic mottle was usually associated with degeneration of the mesophyll cells (EVANS and LEONARD 1991; EVANS and FITZGERALD 1993).

Among the woody species of European flora and the Mediterranean basin, basal plane pines appear to be the ones most sensitive to the action of photo-oxidants. The best known symptoms have been described in *Pinus halepensis* in Spain and Greece (BARNES *et al.* 2000), and in Italy (SODA *et al.* 2000). The needles of *Pinus halepensis* develop a typical symptomatology (chlorotic mottle) similar to that described by the American researchers mentioned above. In broadleaved trees the symptoms vary: ranging from stipules (small, punctiform necroses, ivory-white to brown-red or black in colour, which gradually merge into larger necrotic patches), to flecks (reddish spots), and reddening (leaves changing colour earlier than in normal autumn processes). These symptoms tend to affect the upper side of the leaves, in the interveinal areas. The symptomatology has been illustrated in a number of atlases and photoguides (cf. SKELLY *et al.* 1987; FLAGLER 1998; INNES *et al.* 2001) both for North America and Europe. In Europe the most widespread and evident symptoms validated by Open Top Chamber experiments (VANDERHEYDEN *et al.* 2000) were reported in southern Switzerland (SKELLY *et al.* 1998, 1999) and in areas near the Italian border (COZZI *et al.* 2000; GRAVANO *et al.* 2000). Other communications are for sites in east Europe (BYTNEROWICZ *et al.* 2001; MANNING *et al.* 2002). In the Mediterranean region the majority of cases reported are in Spain (SKELLY *et al.* 1999) where

several evergreen shrubs have also been found to be sensitive. In Italy widespread symptoms have been observed in Tuscany (BUSSOTTI *et al.* 2001), Umbria (COZZI *et al.* 2002), Lombardy (COZZI *et al.* 2000) and Piedmont (PAOLETTI *et al.* 2003). Table 5 shows the woody species on which symptoms have been observed in Italy.

#### **Effects on growth and reproduction**

The effects on growth were reviewed recently by BORTIER *et al.* (2000) and DIZENGRIMEL (2001). The growth of seedlings of the most important European broadleaves (birch, beech, poplar) is generally negatively affected by  $O_3$  within one exposure season. The range of growth reduction differs by provenances (LARSEN *et al.* 1990; WULFF *et al.* 1996), families (TAYLOR 1994) and also by clones (PÄÄKKONEN *et al.* 1993).

In coniferous species, although negative effects of  $O_3$  on growth have been reported, the responses were variable and inconsistent (PYE 1988). The results of several seasons of  $O_3$ -exposure indicate that trees show carry-over effects on growth and suggest that it is most likely that observed effects in trees reflect cumulative processes developed over several growing seasons. In the case of species with multiple flushing growth (like Douglas-fir), this characteristic contributed to the variability in the tree responses.

As previously stated,  $O_3$  reduces root growth more than shoot growth in most species, due to an altered carbon allocation within the plant, thus leading to a reduced photosynthetate supply to roots

**Table 5 -** Tree and shrubs showing ozone-like symptoms in Italy.  
*Specie di alberi ed arbusti che hanno mostrato sintomi attribuibili ad ozono in Italia.*

Species	Region in which symptoms were recorded, and bibliographic reference (see below)
<i>Acer pseudoplatanus</i>	Lombardy (1); Piedmont (8)
<i>Allianthus altissima</i>	Tuscany (1, 5); Lombardy (1, 2); Umbria (3)
<i>Carpinus betulus</i>	Lombardy (1)
<i>Corylus avellana</i>	Lombardy (2)
<i>Fagus sylvatica</i>	Lombardy (1, 2)
<i>Fraxinus excelsior</i>	Lombardy (1, 2, 6); Piedmont (8)
<i>Fraxinus ornus</i>	Tuscany (1); Umbria (3)
<i>Pinus halepensis</i>	Tuscany (7)
<i>Populus</i> spp.	Po Valley (4)
<i>Prunus avium</i>	Lombardy (1, 2, 6); Piedmont (8)
<i>Robinia pseudoacacia</i>	Piedmont (8)
<i>Rosa</i> spp.	Lombardy (1, 2)
<i>Salix</i> spp.	Lombardy (1)
<i>Sambucus</i> spp.	Lombardy (1)
<i>Tilia</i> spp.	Lombardy (1)
<i>Ulmus glabra</i>	Lombardy (2)

(1) Bussotti *et al.*, 2001; (2) Cozzi *et al.*, 2000; (3) Cozzi *et al.*, 2002; (4) Fumagalli *et al.*, 1989; (5) Gravano *et al.*, 1999; (6) Gravano *et al.*, 2000; (7) Soda *et al.*, 2000; (8) Paoletti *et al.* (in press).

(DARRALL 1989). In a few plants however shoots may be more affected than roots (*e.g.* KRESS and SKELLY 1982 in loblolly pine), while in Scots pine there is no difference at all (ANTTONEN and KÄRENLAMPI 1995). Reduced root growth and carbon allocation can alter the functioning of rhizosphere organisms.  $O_3$  alone or in a combination with other pollutants ( $O_3+SO_2$ ;  $O_3+acid$  deposition) not only induces quantitative alterations like reduction of the number of infected short roots and the growth rate of mycorrhizae (KYOVIITA *et al.* 1999), but also produces qualitative changes in mycorrhizae (DIAZ *et al.* 1996).

Information on the effects of  $O_3$  on the sexual reproduction of forest trees is very scarce (BLACK *et al.* 2000). Direct effects of  $O_3$  are known only on pollen germination and pollen tube growth in conifers from *in vitro* (BENOIT *et al.* 1983) and in field (HOUSTON and DOCHINGER 1977) studies. The germination percentage and average pollen tube growth was greater in pollen gathered from the low pollution incidence area. Reduced cone production (MILLER 1973) and decreased seed number and weight (HOUSTON and DOCHINGER 1977) were observed in ponderosa pine in San Bernardino Mountains, although the effects could not be specifically attributed to  $O_3$ .

### **Effects on crown condition**

The relationship between predictive variables (site characteristics, seasonal and environmental factors) and crown condition (defoliation and discoloration) was investigated with multivariate statistics (see the review by SEIDLING 2000). In particular, the possible influence of  $O_3$  was examined both in European forests as a whole (KLAP *et al.* 2000) and in individual countries (Switzerland: DOBBERTIN *et al.* 1997; ZIERL 2002; Netherlands: HENDRIKS *et al.* 1997; United Kingdom: INNES and BOSWELL 1988; INNES and WHITTAKER 1993 and MATHER *et al.* 1995). A study of the influence of  $O_3$  on beech crown architecture, based on Roloff's classification, was performed by STRIBLEY and ASHMORE (2002).

In European forests as a whole, the main correlations between  $O_3$  levels (expressed as AOT60) and crown condition (expressed as defoliation) in the forest species included in the CONECOFOR programme were found in *Quercus ilex* and *Fagus sylvatica* (KLAP *et al.* 2000). No correlations were found in the case of deciduous oaks or *Picea abies*. In *Quercus ilex* the correlation between crown status

and  $O_3$  concentration is only slightly significant, but this finding may be biased by the small number of observations carried out. The significance is much greater in the case of *Fagus sylvatica*.

As far as the national studies are concerned, the following conclusions can be drawn:

- in Switzerland the findings showed that defoliation of all species between 1985 and 1994 was greater in areas with higher levels of  $O_3$  and  $SO_2$ , with lower winter temperatures and in sites with a lower humus content (DOBBERTIN *et al.* 1997). ZIERL (2002) applied a hydrological model to Swiss forests that simulates stomatal aperture, and thus  $O_3$  absorption. The findings support the hypothesis that  $O_3$  contributes to defoliation, although the Author underscored the wide range of factors that may modify the response by the different species (first and foremost the effectiveness of their detoxifying processes); he also questioned the value of defoliation itself as a reliable response parameter;
- in the United Kingdom, defoliation values of Norway spruce, Scots pine, English oak, pedunculate oak and beech varied in relation to the annual levels of  $O_3$  (INNES and BOSWELL 1988). These findings were further confirmed by MATHER *et al.* (1995) in beech and Scots pine.
- in the Netherlands  $O_3$  was found to be one of the factors explaining both defoliation levels and foliar Mg and Al content in English oak and Scots pine (HENDRICKS *et al.* 1997).

### **Interactions between $O_3$ and other stress factors**

#### **Drought**

Stomatal activity is considered the key element determining the sensitivity of a particular species to  $O_3$  (GUDERIAN *et al.* 1985; GRULKE 1999; EMBERSON *et al.* 2000). Usually the prevailing weather conditions in most parts of Italy induce a marked reduction in stomatal conductance during the height of summer. The highest levels of  $O_3$  experienced in the field usually coincide with the time that non-managed plants suffer the greatest degree of water deficit, and their stomata are closed. However, the behaviour of individual species varies considerably in their capacity to tolerate drought before resorting to stomatal closure. As a consequence, those species that exhibit the greatest ability to maintain, or reactivate, gas

exchange under conditions of water stress, might be expected to be the most affected by  $O_3$ .

GRULKE (1999) reports that genotypes of Jeffrey pine (*Pinus jeffreyi* Grev. et Balf.) differ in their sensitivity to  $O_3$  as a result of differing stomatal responses under conditions of soil water deficit. Thus, soil moisture availability is considered an important factor modifying the impact of  $O_3$  (GUDERIAN *et al.* 1985), while, *vice versa*, the partial stomatal closure induced by  $O_3$  can lead to the avoidance of drought injury (MAURER *et al.* 1997). The way in which the combination of  $O_3$  and soil water deficit affect stomatal conductance are, however, much more complex than might at first be perceived. Recent evidence suggests that exposure to the pollutant can impair stomatal performance under conditions of soil water deficit (MAIER-MAERCKER 1998). The outcomes in terms of cost/benefit analysis probably depend, at least to some extent, on genotype and on the timing and intensity of the stress to which the plant is exposed (MAIER-MAERCKER 1998).

#### Nutrients

An adequate level of fertilization is not a necessary prerequisite to improve  $O_3$  tolerance (MAURER and MATYSSEK 1997; MAURER *et al.* 1997), yet in optimal edaphic conditions trees can accelerate their foliar turnover, replacing inefficient, injured leaves with new, more photosynthetically active leaves; but the overall condition of the tree suffers the effects of premature aging, since this behaviour requires a huge investment of mineral nutrients. On the other hand, a limited mineral nutrition can improve the content of secondary metabolites (GUTSCHICK 1999) and, ultimately, the plant's antioxidant ability to defend itself against  $O_3$ , thus delaying the premature loss of leaves. It's not certain that this is truly an advantage, since injured leaves tend to have an increased respiratory activity and a reduced photosynthetic one.

#### Light

Several investigations (TJOLKER *et al.* 1995; FREDERICKSEN *et al.* 1996; GÜNTHARD-GOERG *et al.* 1997) suggest that the effect of  $O_3$  is more pronounced in low light or shaded environments. The mechanisms of resistance to  $O_3$  injury in sun versus shade leaves involves both structural and physiological differences. Sun leaves are normally thicker, have denser mesophyll and higher light saturated photosynthetic rates than

shade-acclimated leaves. The amount of cell surface available to interact with  $O_3$  is lower in shade leaves, thus limiting the effectiveness of the detoxifying processes. Furthermore, the processes whereby many scavengers are formed are stimulated by light (cf. MATYSSEK and INNES 1999). On the other hand, located as they are on the external part of the plant, sun leaves offer a measure of protection to shade leaves, intercepting most of the deposited  $O_3$ .

#### Frost

In evergreen species, the reduced sugar production caused by the inhibition of photosynthesis can influence the efficiency of osmotic regulation. Moreover, direct damage to the lipidic composition on cell membrane may occur. This in turn can alter the hardening processes that provide a defence mechanism against frost. This aspect has been studied in *Picea abies* (WOLFENDEN and WELLBURN 1991) and *Pinus halepensis* (WELLBURN and WELLBURN 1994).

#### Other pollutants

The mixture of  $O_3$ + $SO_2$  has a synergistic effect in producing visible injury on leaves of *Pinus strobus* (DOCHINGER and DELKSAR 1970) and on total biomass of *Pinus halepensis* (DIAZ *et al.* 1996). DOTZLER and SCHÜTT (1990) found that fumigation with low concentrations of  $O_3$ + $SO_2$  affects young Norway spruce although visible symptoms may not occur. Mixed  $O_3$ + $NO_2$  fumigations of forest tree seedlings have a growth-reducing effects (KREES and SKELLY 1982; MOOI 1983);  $NO_2$  applied in a sixfold ambient concentration neither significantly compensated for a low soil N supply nor greatly modified the effect of  $O_3$  (GÜNTHARDT-GOERG *et al.* 1996).

Some evidence of a compensation effect of elevated  $CO_2$  on  $O_3$  toxicity was provided by studies on oak (MANES *et al.* 1998; BROADMEADOW and JACKSON 2000), beech (GRAMS and MATYSSEK 1999), *Prunus serotina*, *Fraxinus pennsylvanica* and *Liriodendron tulipifera* (LOATS and REBECK 1999), poplar (SCHWANZ and POLLE 2001) and birch (VANHATALO *et al.* 2001). The processes involved in the compensation of the  $O_3$  effects are thought to be: reduction of stomatal conductance, metabolic compensation in the mesophyll (GRAMS *et al.* 1999) and the provision of increased amounts of substrate for  $O_3$  detoxification (ALLEN 1990) by means of increased leaf density. Conversely, no buffering effect was seen in coniferous species as Norway spruce

(LIPPERT *et al.* 1997), Scots pine (UTRIAINEN *et al.* 2000) and *Pinus halepensis* (KYTOVIITA *et al.* 2001).

#### Pathogens and phytophagous insects

There is evidence that  $O_3$  alters parasitism via the effects it exerts on host plants (SANDERMANN 2000). The interactions between  $O_3$  and tree pathogens are summarized in Fig. 6.  $O_3$  stress changes may either increase or decrease the susceptibility to the pest agent. The time in which the parasite infestation occurs may be not the same of the  $O_3$  exposure, but may be delayed (memory effect, see Fig. 7).

Generally infections by obligate pathogens, which require a healthy host for successful invasion, are decreased in intensity by  $O_3$  exposure. On the contrary plants weakened by  $O_3$  damage are especially susceptible to facultative parasitic fungi that can benefit from damaged host cells and from disordered transport mechanisms (BEARE *et al.* 1999). However, several factors like tissue age, timing of exposure to  $O_3$  and period in the disease cycle during which  $O_3$  exposure occurs, may influence and alter this general pattern.

Direct effects of  $O_3$  on feeding, growth and survival of insects were observed only in fumigation experiments and the results are contradictory: for example the effect of  $O_3$  on aphid dynamics stimulated the growth of *Phylloxaphis fagi* on beech seedlings (BRAUN and FLÜCKIGER 1988), but had no influence on the survival, reproduction and development of the cottonwood aphid *Chaitophorus populincola* (COLEMAN and JONES 1988a).

The indirect effects are connected to the changes in the physical or biochemical properties of

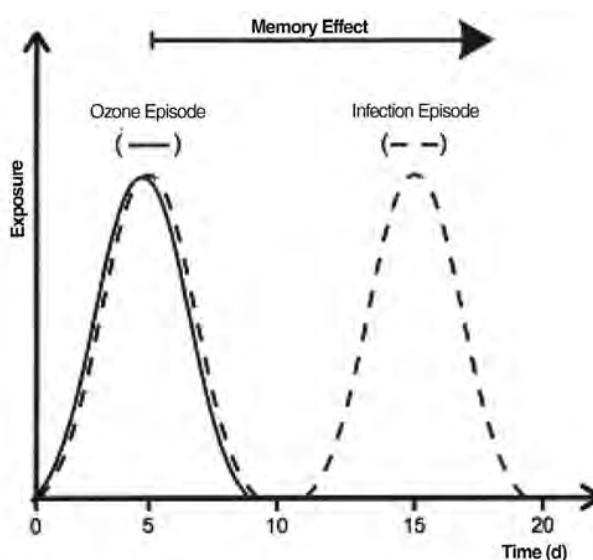


Figure 7 - Epidemiological scenario for ozone-biotic disease interactions (from Sandermann, 2000).

Scenario epidemiologico per l'interazione tra ozono e malattie di origine biotica (da Sandermann, 2000).

leaves (COLEMAN and JONES 1988b) and trunk (COBB *et al.* 1986) which affect the palatability of the foliage or change the host condition. The research conducted in the ponderosa pine forests of the San Bernardino Mountains - subjected to high levels of atmospheric oxidants from Los Angeles urban complex - revealed some mechanisms of the relationships between *Pinus ponderosa* and bark beetle populations (*Dendroctonus ponderosae* and *D. brevicornis*). It has been suggested that the reason bark beetles showed a preference for injured trees was linked to changes in resin and sapwood characteristics (reduction of oleoresin exudation pressure; quantity, rate of flux and increased propensity of oleoresin to crystallize and a reduction in phloem and sapwood moisture content) that facilitated bark beetle activity particularly in the concentration and establishment phases (COBB *et al.* 1968).

#### Effects on ecosystem processes, patterns and dynamics

There are only a few reports in the literature concerning  $O_3$  effects on forest ecosystem function, and they deal exclusively with litter decomposition. No apparent influences of  $O_3$ -exposed litter on the decomposition process was found by FENN (1991) and BOERNER and REBECK (1995); on the other hand, FINDLAY *et al.* (1996) reported that early-abscised leaves from  $O_3$ -exposed *Populus deltoides* saplings had higher N

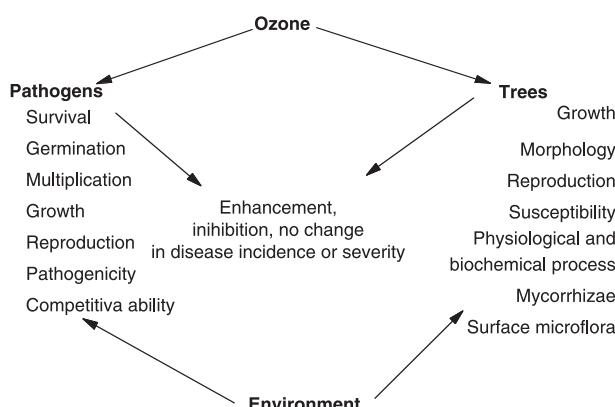


Figure 6 - Interactions between ozone and pathogens of trees.  
Interazioni tra ozono e patogeni.

contents and decomposed at a slower rate than leaves from control plants. In the same studies, leaves from  $O_3$ -exposed plants that abscised at the normal time had a lower N content and lower specific leaf mass than control leaves, but decomposed at the same rate as leaves from control plants. The differences in the results may be attributed to several causes, like the different incubating environments in the litterbags, the decaying environment, the effect of  $O_3$  on the fungal populations of the decomposer community both on the leaf phyllosphere and litter layer that reduces and/or alters the composition of phyllosphere fungal populations (FENN *et al.* 1989; MAGAN *et al.* 1995).

#### **Final remarks**

The literature on  $O_3$  effects on forest trees is wide-ranging, although only a very small part of the findings are useful in understanding the natural ecological processes occurring in forests. The most important limitation lies in the fact that experiments are usually performed on young and small trees whose physiology is very different from that of adult trees (KOLB *et al.* 1998). Seedlings are usually considered more sensitive than adult trees due to their greater stomatal activity, but in some cases (*Quercus rubra*) the findings suggest the opposite (KELLY *et al.* 1995). Further problems are raised by the complex relationships between the tree and external factors (ecological conditions and interactions with other organisms) which we still know little about. It is easy to understand why, when we try to investigate one factor at a time, the findings are often contradictory. It is important to bear these limitations in mind when we attempt to interpret ecological hazards in the field.

### **The I&C evaluation system and the $O_3$ Risk Analysis**

#### ***The I&C evaluation system***

In 1998 the National Focal Center (NFC) of Italy (based at the Ministry of Agricultural and Forestry Policies, Division V) decided to develop a formal evaluation system for the data generated by the Italian intensive forest ecosystem monitoring programme (CONECOFOR) (GRUPPO DI ESPERTI CONECOFOR-I&C 1998). The first step was to establish a Task Force (TF) set up by the team leaders of the various investigations carried out at the Permanent Monitoring Plots (PMPs) of the programme (see Table

2). The TF agreed upon a general concept for the evaluation system. The evaluation system was termed Integrated and Combined (I&C) because it is an attempt (i) to integrate data on different indicators, collected according to different sampling regimes and to different metrics; and (ii) to combine different evaluation perspectives in a cohesive and consistent evaluation system. Detailed information on the I&C evaluation system has been published elsewhere (e.g. FERRETTI *et al.* 2000; FERRETTI 2000, 2002).

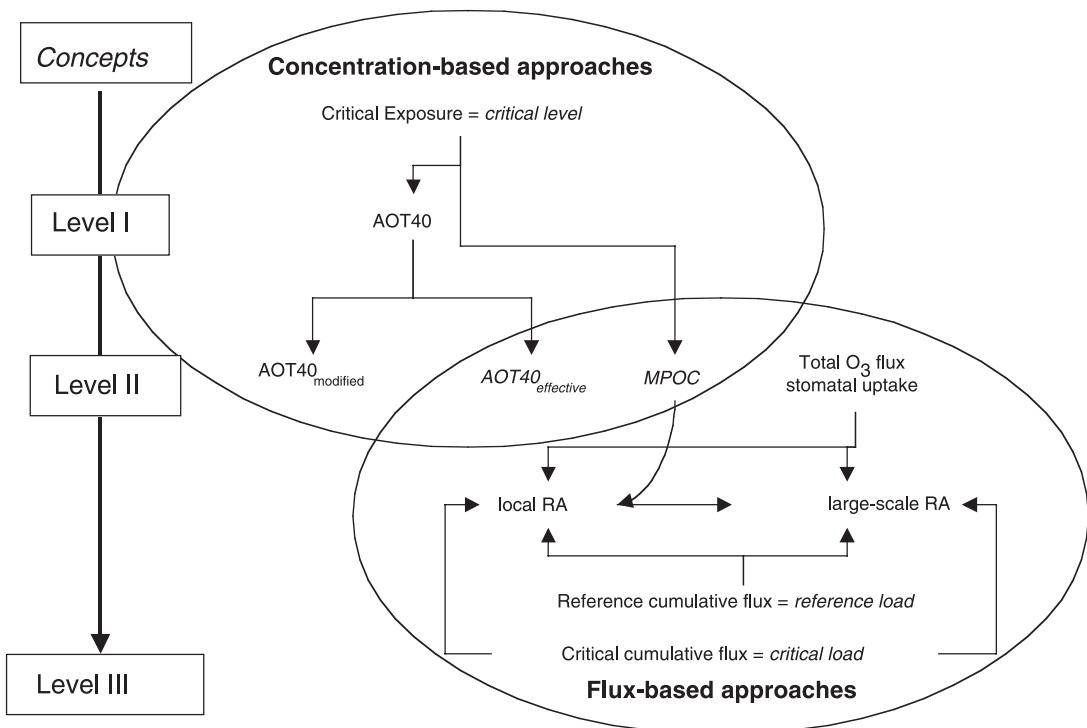
The I&C TF recognized three major issues of concern for the evaluation system: (i) the need to evaluate and identify the actual and potential risk status of the various PMPs in relation to air pollution, atmospheric deposition and meteorological stress; (ii) the need to evaluate, identify and quantify changes in biological, chemical, and physical ecosystem status and (iii) the need to evaluate and identify the determinants of changes. All together, the above issues constitute the framework within which the I&C evaluation system was established. Operatively, three categories of analysis were identified, each dealing with one of the issues mentioned above: Risk Analysis (RA), Status and Changes analysis (S&C) and Nature of Change analysis (NoC) (see FERRETTI 2000 for details). Within the 2001-2005 strategy plan, risk analysis in relation to  $O_3$  exposure was set as the first priority of the I&C system (TASK FORCE I&C 2001).

#### ***Risk Analysis in relation to $O_3$***

Within the I&C system, RA can be defined as a process aimed at evaluating "the effects of an environmental change on a valued natural resource" and at interpreting "the significance of those effects in the light of uncertainties identified in each component of the assessment process" (HUNSAKER *et al.* 1990). This general definition needs to be placed within the context of current approaches in assessing the risk of  $O_3$  exposure (HOGSETT *et al.* 1997). Risk assessment in relation to  $O_3$  is a topic in rapid evolution (see e.g., GRÜNHAGE and JÄGER 2002). There are, however, two main approaches: the concentration-based approach (called Level I) and the flux-based approach (Level II). The way for a "Level III" approach has been already suggested (GRÜNHAGE and JÄGER 2002) (Fig. 8).

#### ***Concentration-based approach***

The current approach in evaluating the potential



**Figure 8** - The progression of concepts from exposure, concentration based-approach (Level I) in relation to a critical level toward a flux-based approach (Level III) in relation to a critical load (after Grünhage and Jäger, 2002, with modifications). AOT40: ozone accumulated over threshold 40 ppb; AOT40modified= AOT40 modified according to Posch and Fuhrer, 1999, taking into account vapour pressure deficit, soil moisture and wind speed; AOT40effective=AOT40 modified according to Grünhage (1999) and Tuovinen (2000). MPOC=maximum permissible ozone concentration (Grünhage et al., 2001). Both AOT40effective and MPOC are based on concentrations at the canopy level. RA: Risk assessment.

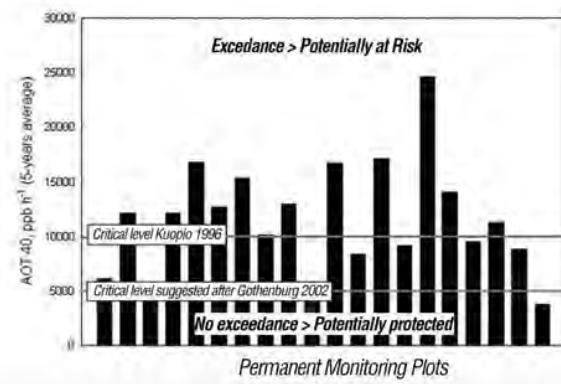
*Progressione dei concetti per la valutazione di rischio: dall'approccio basato sull'esposizione (Livello I) in relazione ad un livello critico verso un approccio basato sul flusso (Livello III) in relazione ad un carico critico (da Grünhage e Jäger, 2002, con modifiche). AOT40= ozono accumulato sopra la soglia di 40 ppb; AOT40 modified= AOT40 modificato secondo Posch and Fuhrer, 1999, tenendo conto del deficit di pressione di vapore, del deficit di umidità del suolo e della velocità del vento. AOT40 effective=AOT40 modificato secondo Grünhage (1999) e Tuovinen (2000). MPOC=massima concentrazione di ozono ammissibile (Grünhage et al., 2001). Sia AOT40 effective che MPOC sono basati sulle concentrazioni di ozono a livello della chioma.*

$$AOT40 = \sum_{\substack{[O_3]_i > 40 \text{ ppb} \\ \text{globrad} > 50 \text{ W/m}^2}} ([O_3]_i - 40) \quad (\text{ix})$$

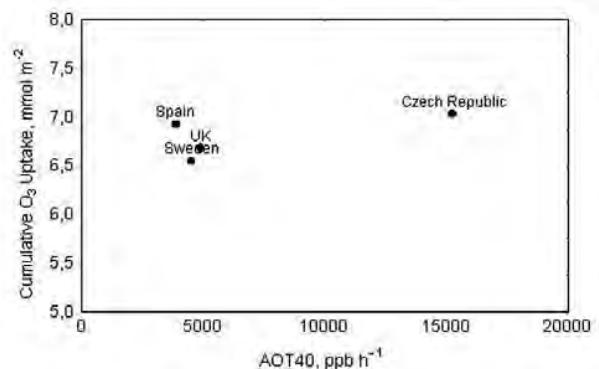
risk for vegetation due to O<sub>3</sub> exposure is based on the critical level concept. A critical level is defined as "the concentration of a pollutant in the atmosphere above which direct adverse effects on receptors, such as plants, ecosystems or materials, may occur according to current knowledge" (UN/ECE 1988). Although different approaches exist (e.g. GRÜNHAGE *et al.* 2001; VDI 2002), the adopted critical level for O<sub>3</sub> is based upon a cumulative exposure index, the AOT40 (UBA 1996). The AOT40 for forests is defined as the O<sub>3</sub> Accumulated Over Threshold 40 ppb (1 ppb=1 nl l<sup>-1</sup>), calculated for the daylight hours over the growth period (April-September).

This index was developed after a series of workshops held in Bad Harzburg (1988), Egham (1992), Bern (1993) and Kuopio (1996) (UN/ECE 1988; ASHMORE and WILSON 1992; FUHRER and ACKERMANN 1994; KARENLAMPI and SKARBY 1996; PORG 1997). At the Kuopio workshop, the critical level AOT40 for forests was set at 10000 ppb·h<sup>-1</sup>. This threshold was established as provisional on the basis of open top chamber experiments with beech (*Fagus sylvatica* L.) and Norway spruce (*Picea abies* (L.) Karsts.) seedlings

and equals the exposure for which a reduction of 10% of biomass at 95% p-level can be inferred from the fitting function (SKARBY and KARLSSON 1996). To assess the potential risk for a given site/region, the comparison with such a threshold should be done on the basis of the 5-year average AOT40. In practice, this means that - for a given site - a potential risk occurs when the 5 year average AOT40 exceeds 10000 ppb·h<sup>-1</sup> (Fig. 9). Following a new workshop recently held near Gothenburg (Sweden), new data sets for birch, beech, spruce, oak and Aleppo pine have been examined in detail (KARLSSON *et al.* 2003). Following these results, new critical levels were suggested for species with different levels of sensitivity. In particular, a critical level of AOT40 5000 ppb·h<sup>-1</sup> was suggested to protect "the most sensitive species under the most sensitive conditions", although there is a clear warning against the use of such a threshold for estimating O<sub>3</sub> risks under field conditions. Actually, there are several limitations when one transfers the potential risk estimated according to AOT40 to a more concrete risk assessment (see *e.g.* PORG 1997; NEGTAP 2001; FUHRER and ACHERMANN 1999; EMBERSON *et al.* 2000). The most important of these is the strong body of evidence suggesting that O<sub>3</sub> effects on plants do not depend on exposure, but rather on the amount of O<sub>3</sub> that enters the plant (the "physiologically effective dose", SAMUELSON 2001). As high O<sub>3</sub> concentrations tend to occur under meteorological conditions that reduce O<sub>3</sub> uptake (*e.g.* high vapour pressure deficit in the Mediterranean area), sites with high O<sub>3</sub> exposure and



**Figure 9 -** Example of the application of the exposure-based approach to identify plots potentially at risk where the critical levels AOT40 are exceeded.  
*Esempio di applicazione dell'approccio basato sull'esposizione per identificare i plot potenzialmente a rischio ozono dove i livelli critici AOT40 vengono superati.*



**Figure 10 -** Example of comparison between total cumulative O<sub>3</sub> uptake for beech and AOT40 at four EMEP grid squares (drawn on the basis of the data by Emberson *et al.* 2000).

*Confronto tra l'assorbimento cumulato di O<sub>3</sub> per il faggio ed i valori di AOT40 in quattro quadrati EMEP (disegnato su dati di Emberson *et al.* 2000).*

sites with high O<sub>3</sub> uptake do not necessarily coincide (GRÜNHAGE *et al.* 1997; EMBERSON *et al.* 2000) and this is an important drawback for a RA based on the critical level approach.

#### Flux-based approach

Although the value of the AOT40 index in providing a first indication of the potential risk for a given site cannot be denied, it is widely acknowledged that effects are more likely to be related to actual O<sub>3</sub> uptake by plants (REICH 1987; SAMUELSON 2001) rather than to exposure. In other words, O<sub>3</sub> effects are related to the portion of O<sub>3</sub> that enters the plants and this portion is not always proportional to the exposure (Fig. 10). This "physiologically effective dose" primarily depends on stomatal conductance, which is a function of number, size and aperture of the stomata (SAMUELSON 2001). Each of these components of stomatal conductance is highly variable within- and between species, according to a number of factors like the position within the canopy layer, phenology, irradiance, temperature, vapour pressure deficit, soil moisture deficit (SAMUELSON 2001; EMBERSON *et al.* 2000). As direct measurements of O<sub>3</sub> uptake are uncommon (for example they are not carried out at the PMPs of the CONECOFOR programme), modelling according to the resistance analogue principle is often adopted to calculate the flux of O<sub>3</sub> from the atmosphere to the substomatal cavity (EMBERSON *et al.* 2000). In practice, the O<sub>3</sub> flux density is directly related to the concentration gradient and the resistance to transfer

between two points (*e.g.* the height in the atmosphere at which  $O_3$  concentration is described and the substomatal cavity, within which  $O_3$  concentration is assumed to be zero):

$$F_{O_3} = v_{dO_3} \cdot [O_3] \quad (\text{x})$$

where

$$v_{dO_3} = \frac{1}{r_a + r_b + r_c} = G_{O_3} \quad (\text{xi})$$

with  $G_{O_3}$  being the stomatal conductance to  $O_3$ , and where

$r_a$  = aerodynamic resistance, the resistance to  $O_3$  transfer between the height above the plant canopy at which the  $O_3$  concentration is measured or modelled and the top of the canopy with mass transfer occurring mainly via mechanical and thermal turbulence

$r_b$  = quasi-laminar sublayer resistance, the resistance to  $O_3$  transfer across the still layer of air referred to as the quasi-laminar boundary layer found adjacent to the leaf surface where transfer occurs via molecular diffusion.

$r_c$  = stomatal resistance (equal to  $1/g_s$ , with  $g_s$  being the stomatal conductance to water vapour).

Each of the above resistance terms needs to be modelled according to a specific module, which requires data on physical characteristics of the atmosphere and on structural and biological attributes of the canopy (see EMBERSON 2002 in ANONYMOUS 2002). Modelling the flux of  $O_3$  from the atmosphere to the substomatal cavity is therefore complex and data intensive (see EMBERSON 2002 in ANONYMOUS 2002; FERRETTI and GEROSA 2002 in ANONYMOUS 2002) and this is probably the most important disadvantage of this approach.

#### Work plan and structure of the report

This report aims to provide an evaluation of the potential risk of the CONECOFOR PMPs in relation to  $O_3$  and to explore whether any  $O_3$ -related effect is detectable on the vegetation at the CONECOFOR plots. Ideally, this implies covering the following steps:

- estimating  $O_3$  exposure (measurement of  $O_3$  concentration at a given site and calculation of

exposure indices);

- estimating  $O_3$  uptake (measurement/estimation of the portion of atmospheric  $O_3$  that enters the plant);
- estimating the actual  $O_3$  effects (the quantification of the detrimental effects of  $O_3$  on a given indicator of plant performance and/or health condition after having removed the effects due to other factors).

Unfortunately, each of these steps contains several sources of uncertainty and is further complicated by the fact that responses to  $O_3$  vary in relation to the species being considered and to a number of environmental variables (HOGSETT *et al.* 1997). In addition, estimating  $O_3$  uptake is data intensive and the CONECOFOR plots are not always equipped for this aim. Actually, the RA reported in the following sections of this report is based on observational studies carried out on a network of sites that was not designed for investigating  $O_3$  effects. This means that (i) sites were not selected and installed according to an *ad hoc* experimental design and (ii) the data collected by the programme may not cover all the data requirements called for by *e.g.* the Level II RA reported above. With these limitations, and under the perspective reported above, this report will proceed as follows:

- (i) report  $O_3$  concentration levels measured by passive sampling over the period 1996-2000 at the PMPs of the CONECOFOR programme. This is the work of BUFFONI and TITA (2003 this volume)
- (ii) investigate the relationships between  $O_3$  concentration and climatic and geographical factors by means of multivariate analysis. This will be done by AMORIELLO *et al.* (2003 this volume)
- (iii) estimating the vegetation exposure to  $O_3$  over the period 1996-2000. The estimation of AOT40 starting from data obtained by passive sampling is reported by GEROSA *et al.* (2003 this volume).
- (iv) report and evaluate available data in order to estimate the potential sensitivity of vegetation (*e.g.*, species composition, canopy structure, site conditions,...) to  $O_3$  at the various PMPs. This will be done by ALIANIELLO *et al.* (2003 this volume).
- (v) analyse the potential and the limitations of available data in view of implementing the  $O_3$  flux approach (GEROSA and ANFODILLO this volume).

- (vi) estimate the effects of current O<sub>3</sub> levels on tree growth, crown condition and foliar symptoms on trees, shrubs and herbs (for details see BUSSOTTI *et al.* and FERRETTI *et al.* 2003b, both this volume).
- (vii) provide a summary of the actual and potential effects of O<sub>3</sub> on the vegetation at the CONECOFOR PMPs, and also address the achievements, problems and perspectives identified by the O<sub>3</sub> Risk Analysis (FERRETTI *et al.* 2003a this volume).

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# Ozone measurements by passive sampling at the permanent plots of the CONECOFOR programme<sup>§</sup>

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**Summary** - In the framework of the National Integrated Programme for Forest Ecosystem Monitoring. (CON.ECO.FOR.) measurements of ambient ozone ( $O_3$ ) concentrations were carried out by passive samplers at 20 Italian forest plots over the summer seasons 1996 – 2000. Passive samplers exposed at a co-located continuous monitoring station were checked for correlation with data from an automatic  $O_3$  analyser. Mean weekly concentrations highlight relevant concentrations at all the plots. Permanent investigation plots located in Central and Southern Italy show the highest values. The five year mean  $O_3$  concentrations referred to the summer period varies from 37 to 50 ppb. Differences between individual years are also significant. 1997 and 2000 can be considered high ozone years, while 1996 was characterised by low  $O_3$  concentrations at all plots. Results from the analysis of variance performed for plots located in the different geographical areas show significant differences. Passive samplers show to be a suitable method to determine  $O_3$  levels at remote sites and to describe temporal differences among the plots. These devices do not need electricity, allow a free choice of exposure sites, are easy to handle and technical personnel is not needed at the sampling site. The low temporal resolution of passive samplers represents the main disadvantage for their use in forest health studies.

**Key words:** ozone, passive samplers, monitoring, analysis of variance.

**Riassunto** – Misurazioni di ozono mediante analizzatori passivi presso le aree permanenti del programma CONECOFOR. Nell'ambito del Programma Nazionale Integrato per il Controllo degli Ecosistemi (CON.ECO.FOR.) sono state condotte misure di ozono mediante campionatori passivi presso 20 aree forestali. Sono attualmente disponibili le concentrazioni misurate dalla tarda primavera alla fine dell'estate relative a 5 anni (1996 – 2000). I campionatori sono stati inoltre esposti presso una stazione di monitoraggio in continuo per verificare la correlazione con i dati rilevati da un analizzatore in continuo. I valori di concentrazione media settimanale denotano livelli rilevanti presso tutte le aree. Le aree di indagine permanenti situate in Italia Centrale e Meridionale mostrano i valori più alti. Le concentrazioni medie calcolata sui 5 periodi di misura estivi variano tra 37 e 50 ppb. Le differenze tra singoli anni sono pure rilevanti. Il 1997 e il 2000 possono essere considerati anni ad elevato ozono specie presso le aree al Nord e al Sud, mentre il 1996 si caratterizza per le più ridotte concentrazioni presso tutte le aree. I risultati dell'analisi della varianza condotta sulle aree appartenenti a diverse regioni geografiche evidenziano differenze significative. I campionatori passivi risultano essere un metodo adeguato per la determinazione dei livelli di  $O_3$  troposferico in siti remoti e per evidenziare differenze nel tempo tra le diverse aree. Questi dispositivi non richiedono allacciamento elettrico, consentono di scegliere liberamente il sito di misura, sono semplici da utilizzare e non richiedono personale tecnico per il loro impiego. La scarsa risoluzione temporale rappresenta il principale svantaggio nel loro uso nelle indagini sulle condizioni delle foreste.

**Parole chiave:** ozono, campionatori passivi, monitoraggio, analisi della varianza.

Ambient concentrations of tropospheric ozone ( $O_3$ ) represents one of the main environmental threats for the European forest ecosystems. Over large areas of the Mediterranean countries the emission of  $O_3$  precursors and the high solar radiation during spring and summer months result in high concentrations of this pollutant and critical levels for the protection of forest ecosystems and agriculture crops are often exceeded (BUTKOVIC *et al.* 1990; HETTENLINGH *et al.* 1997; POSCH *et al.* 1999). Nonetheless information about levels, spatial and temporal distribution of  $O_3$  are often scarce and fragmentary. In particular, the lack of data outside the urban areas can be observed in several countries of the Mediterranean area which, however, due to the favourable conditions for  $O_3$  formation (high emission of  $O_3$  precursors and solar radiation) should be carefully monitored. For example, in Italy background monitoring stations are relatively scarce in number and are prevalently loca-

ted in the Northern part of the country. According to a first inventory of the National Agency for Environmental Protection (ANPA, 2000) there are 297  $O_3$  monitoring stations in Italy. Two regions (Puglia and Molise) have no  $O_3$  continuous monitor at all, one region has only one  $O_3$  monitor. The rural background stations are 33 and are located in 13 out of 20 regions.

In recent years, the availability of air quality data has become a recognised need in order to evaluate plant exposure to air pollutants at remote sites. However, due to the high costs of the equipment and maintenance, real time automatic measurements are difficult to be implemented under the field condition of forest monitoring programmes. In recent years, the use of passive samplers has received increasing attention (Cox *et al.* 1999; KRUPA and LEGGE 2000) and passive samplers have been largely adopted for the implementation of several studies and national networks regarding forest health and to evaluate air pollution impact on forest ecosy-

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stems (BLUM *et al.* 1997; COX *et al.* 2001; BYTNEROWICZ *et al.* 2002a). For example, passive samplers can be used in forest health monitoring to categorise forest plots and for cause – effect analysis referred to forest health responses (COX *et al.* 1999). In particular, the diffusive sampling technique (see below) has become a tool of increasing importance for the implementation of the EC air quality measurements and for the reduction of air pollution (DE SAEGER *et al.* 2001) and the use of passive samplers is a recommended method within the ICP Forests Programme for areas where active, automatic monitoring of air pollutants is not feasible (UN/ECE-ICP Forests 2002). Passive sampling is also recognised by national laws of different European countries, including Italy.

The relatively low costs and the flexibility of placement of passive sampling devices represent the major advantages of this approach. Passive samplers are devices that contain no moving parts and rely on the principle of gas diffusion (HARPER and PURNELL 1987). Although passive samplers were designed initially for personal exposure monitoring and for exposition of few hours, they are now being tested and used over duration up to a few weeks for use in ecological monitoring studies.  $O_3$  passive (diffusive) samplers are today available with different reactants as nitrate, indigo compounds and 1,2-di-(4-pyridyl)ethylene (DPE) used in badge samplers or diffusion tubes. A comprehensive review of the different approaches based on passive sampling of gaseous air pollutants is reported in KRUPA and LEGGE (2000). Passive samplers allow the quantification of average pollutant concentrations over a sampling period. However, there is a significant temporal variability in the occurrence of  $O_3$  and mean (weekly or fortnight) concentrations cannot explain the underlying dynamics of this pollutant during individual days. Critical levels, on the other hand, rely on recording hourly concentrations and modeling approaches are needed to make use of the

information provided by passive samplers. Actually, the main disadvantage of passive samplers highlighted in several studies is a poor temporal resolution of measurements as only a cumulative value over the sampling period (1 – 2 weeks) is given. Attempts are being made to use mean concentrations from passive samplers to reconstruct real-time  $O_3$  concentrations and to evaluate plant exposure to ambient ozone (BYTNEROWICZ *et al.* 2002b; KRUPA and LEGGE 2000; COX *et al.* 2001).

In the framework of the CON.ECO.FOR Programme, the monitoring of air pollutants at the permanent monitoring plots (PMPs) has three main objectives: (i) to collect information on the presence of phytotoxic air pollutants in forest ecosystems, (ii) to improve the knowledge of the spatial and temporal distribution of these pollutants, and (iii) to estimate the risk of direct and indirect effects of air pollution on forest ecosystems. In the present paper the  $O_3$  monitoring activities carried out in the framework of the CON.ECO.FOR. programme are presented. In view of the importance of  $O_3$  and the relative limited information about  $O_3$  levels in remote areas, the importance of the  $O_3$  measurements carried out in the framework of the CONECOFOR Programme goes beyond the objectives of the present study as it can contribute to a better knowledge of  $O_3$  levels, spatial and temporal distribution in Italy.

## Methods and materials

### Data collection

$O_3$  measurements were carried out at 20 permanent monitoring plots (PMPs) of the CON.ECO.FOR. programme. Plots are located at sites which differ substantially in latitude, altitude and synoptic meteorology (see Table 1 in FERRETTI *et al.* 2003). Measurement sites were identified near (generally 200-300 m apart) the PMPs of the CON.ECO.FOR. project during spring 1996 and were not subjected to changes during the period

**Table 1** - Site characteristics for passive samplers exposition.  
*Caratteristiche dei siti per l'esposizione dei campionatori passivi.*

Free air circulation	Minimum distance from trees	2 x average height of the near forest stand
Distance from emission sources	Minimum distance from buildings	20 m from isolated trees 2 x height of the nearest building
	Minimum distance from roads	200 m from motorways 100 m from other roads 10 – 20 m from forest tracks or country roads No parking areas
	Minimum distance from agricultural activities	50 m from farms 50 m from orchards or vineyards No direct exposition at cultivated fields Fence or attended area No direct exposition near footpaths Information to the public
Protection of exposition devices		

1996 – 2000. The requirements for the measurements sites are reported in Table 1. The sites identified are characterised by free air circulation and absence of obstacles in the proximity of the measurement devices. In particular, a minimum distance from trees and buildings and from roads was required. Important aspects to be considered are also the presence of potential disturbance sources as traffic and agricultural activities, and the risk of vandalism. In general, measurements were performed at the same elevation as the permanent plot in the forest. The majority of the measurements sites selected meets the requirements reported in Table 1. In one case, due to the presence of a continuous forest cover, the minimum distance from trees is less than required (CAL1). In two cases the prescribed position in elevation could not be assured: TOS1 measurement site is lower than the permanent forest plot, while the measurement device near the plot SIC1 is placed on a fire look-out tower above the canopies of the plot. The passive samplers were placed, generally, at 1.80m from the ground; the tubes were mounted on a 2.20 m tall pole and held in position by tie-rods. A detailed description of permanent plot locations and characteristics is given in FERRETTI (2000).

The  $O_3$  measurements were carried out using passive samplers developed at the University of Munich, Department of Forest Bioclimatology and Immission Research (WERNER 1992; HANGARTNER *et al.* 1996). These diffusive tube type samplers rely on the reaction of indigo with ozone to isatin which can be determined spectrophotometrically. The tubes (PVC) are 570 mm long and have a diameter of 70 mm. Indigo impregnated paper filters are placed in the tubes and kept in position (450 mm from mouth) by a rod. A detailed description of the exposition device is given in WERNER (1992) and BUFFONI and TITA (2000). The selected samplers were tested in the framework of two projects (KIRCHNER *et al.* 1994) and are currently used in forest health studies (WERNER 1999).

Samplers were exposed weekly (168 hours) to ambient air. Substitution of samplers was carried out by local operators of the National Forest Service or personnel of other institutions taking part to the CONECOFOR programme. Operators were asked to fill a form regarding start and end of each exposure period, relevant meteorological events during the week and the presence of eventual disturbance sources near the measurement sites.

Samplers placed in air tight plastic bags were sent periodically to the operators and were returned

in the same bags to the laboratory after exposition. Each sampler was coded according to the plot number, the replication number and the week of exposition.

In 1996 five replications were exposed in parallel, in 1997 three, while in the following three years one passive sampler only was exposed at each site.

At one site correlation of passive samplers with continuous measurements was checked by simple regression procedure to fit a linear model.

### **Data analysis**

Mean weekly concentrations recorded by passive samplers during the monitoring periods at the permanent plots in the years 1996 – 2000 have been processed in order to categorise the plots for  $O_3$  levels and temporal variations. In particular the presence of significant differences among years and plots has been carried out performing a multifactor analysis of variance for the concentration data. The method used to discriminate among the means was Fisher's least significant difference (LSD) procedure (STATGRAPHICS PLUS 1998)

## **Results and discussion**

### **Performance of passive sampling and potential noise Data completeness**

As passive samplers needs only a careful handling and conservation, the amount of missing data is very low (Table 2). Missing data are generally due to mistakes in handling and reporting, failure in delivering timely the samplers to the local operators or unavailability of local personnel. The overall amount of missing data over the five monitoring periods is limited to 3.9% of the total planned measurements. Relevant number of missing data can be observed at the plot CAM1 in 1997 as local operators were not available, while LOM1 did not take part to the monitoring activities in 1996.

### **Calibration against real time analysers and data reliability**

At short distance from the PMP VAL1 passive samplers were paralleled by a co-located continuous measurement station equipped with an UV absorption analyser and data checked for correlation. Figure 1 presents a comparison of weekly mean concentration data from a continuous analyser and passive samplers collected near the permanent plot VAL1 in 1996, 1999 and 2000. Differences from the continuous measurements are in most cases generally limited to  $\pm 5\%$ . In few cases differences exceeded  $\pm 10\%$  but no significative influence

**Table 2-**  $O_3$  concentration data capture (in %) in the period 1996-2000 at the permanent monitoring plots.  
*Dati validi di concentrazione di  $O_3$  (in %) rilevati nel periodo 1996-2000 presso le aree di indagine permanenti.*

Year Period	1996 15/6-30/9	1997 17/6-1/10	1998 16/6-29/9	1999 4/5-28/9	2000 2/5-3/10
ABR1	100	100	100	90	90
BAS1	93.8	75.0	81.3	95.5	95.5
CAL1	93.8	100	100	100	100
CAM1	93.8	37.5	81.3	100.0	100
EMI1	100	100	100	95.5	95.5
EMI2	100	81.3	93.8	77.3	95.5
FRI1	93.8	100	100	100	100
FRI2	87.5	100	100	100	100
LAZ11	100	100	100	100	90.9
LOM1	-	100	100	100	100
MAR1	87.5	93.8	93.8	100	100
PIE1	93.8	81.3	68.8	100	95.5
PUG1	100	93.8	100	100	100
SAR1	100	100	100	100	100
SIC1	100	100	100	100	100
TOS1	100	100	100	100	100
TRE1	100	100	100	100	100
UMB1	100	100	100	100	95.5
VAL1	100	100	100	90.9	90.9
VEN 1	100	100	93.8	100	100

of meteorological conditions could be found.

Ozone concentrations measured with passive samplers from May to September 2000 show to be significantly correlated with  $O_3$  concentrations recorded by continuous measurements. The correlation is close to what recorded in 1996 and 1999 (BUFFONI and TITA 2000). Additional data from parallel measurements of continuous analysers and indigo passive samplers from this study and from other studies for fortnight exposures at several Italian sites (KIRCHNER *et al.* 1994) are reported in Table 3.

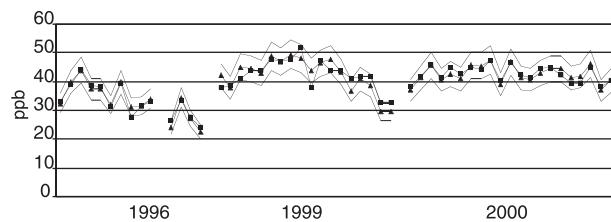
Pooled standard deviations (TAYLOR 1990) are reported in Table 4. Data for 1996 when five replications were exposed are lower than 1.5 at all plots while in 1997 (three replications) pooled standard deviation is lower than 5.5 at all plots with the exception of PUG1. Concentration ranges for measurements carried out in 1996 – 2000 are reported in the same table. These data highlight substantial differences among the plots and monitoring years. Maximum weekly concentrations may reach 80-90 ppb, while minima are below 20 ppb.

#### Potential source of noise

It should be noted that as the ratio of moles ozone forming isatin is not known exactly,  $O_3$  indigo passive samplers have to be calibrated by lots with measurements from a continuous analyser. Factors such as temperature, wind velocity and relative humidity, which can

effect passive samplers efficiency, in the operational phase cannot be controlled. Therefore it is recommended that indigo passive samplers be used at sites with similar characteristics or more continuous analysers should be available for calibration purposes. Sensitivity of these passive samplers to other oxidants (*e.g.* PAN) is not known while possible interactions with  $NO_x$  are excluded and indigo samplers are used also in urban areas (WERNER and FABIAN 1996). In the present study the favourable conditions for indigo passive samplers (low humidity) have probably played a positive role as the correlation found is similar or even closer (1996) than that found in previous calibrations (HANGARTNER *et al.* 1996). Meteorological conditions at the other plots different from VAL1 appear not to have been adverse for the use of indigo passive samplers.

From a theoretical point of view, temperature will influence diffusion through the tube for  $0.2\% * ^\circ C^{-1}$ . Also atmospheric pressure may affect samplers for  $0.1\% * mbar^{-1}$ . Wind velocity may cause turbulences at the open end of the tubes shortening the laminar flow path.



**Figure 1-**  $O_3$  mean weekly concentration (in ppb) recorded by passive samplers (■) and a continuous monitor (▲) located at short distance from the permanent plot VAL1. Continuous lines indicate monitor concentrations  $\pm 10\%$ .  
*Concentrazioni medie settimanali di ozono (in ppb) rilevate mediante campionatori passivi (■) e un analizzatore (▲). Le linee continue indicano le concentrazioni rilevate dall'analizzatore  $\pm 10\%$ .*

**Table 3-** Comparison of parallel measurements of  $O_3$  concentrations taken by passive and an automatic monitor at the permanent observation plot VAL 1 and at other Italian sites.  
*Confronto di misure di concentrazione di  $O_3$  condotte in parallelo mediante campionatori passivi e un analizzatore automatico presso l'area di indagine permanente VAL 1 e altri siti italiani.*

Site	Year	Exposure (days)	r	p	Reference
La Thuile, Aosta	1996	7	0.97	<0.001	Buffoni and Tita 2000
La Thuile, Aosta	1999	7	0.86	<0.001	Buffoni and Tita 2000
La Thuile, Aosta	2000	7	0.90	<0.001	this study
Montelibretti, Rome	1996	7	0.84	<0.002	Unpublished data
Bolzano	1993	14	0.94	<0.005	Kirchner <i>et al.</i> 1994
Trento	1993	14	0.87	<0.005	Kirchner <i>et al.</i> 1994
Alpe Scoccia (VB)	1993	14	0.90	<0.005	Kirchner <i>et al.</i> 1994
Milano	1992	14	0.96	<0.005	Kirchner <i>et al.</i> 1994

Referring to the absorptive fixation of gas molecules air humidity will reduce at an unknown rate the absorptivity thus modifying the sampling rate. In field measurements, however, these effects are generally difficult to characterize. From an operational point of view, when a large number of operators are involved, several other factors, which cannot be controlled, may also influence the samplers characteristics and efficiency (handling, conservation, transport, etc.). In general the approach adopted appears suitable for a first assessment of spatial and temporal variations of  $O_3$  concentrations at remote areas. The definition of standardised procedures for the different operations connected with the use of passive samplers is of extremely importance.

#### $O_3$ concentration levels

Mean weekly  $O_3$  concentrations for the different monitoring seasons are reported in Table 5. As monitoring periods started at the beginning of May in 1999 and 2000 while for the previous periods data were collected beginning from the third week of June, mean values for the common period are reported. In most cases  $O_3$  concentrations measured in May and in the first two weeks of June in 1999 and 2000 are lower than the mean value of the following 16 weeks, with the exception of SIC1 in the first year. Referring to the common period of 16 weeks mean values measured over the monitoring periods vary from 30.4 ppb (FRI1 in 1996) and 30.5 ppb

(CAL1 in 1996) to 62.9 ppb (SIC1 in 2000). The highest concentrations during the five years here considered were always recorded in Sicily at the plot SIC1.

In general plots located in Central and Southern Italy present higher  $O_3$  concentrations than the plots sited in the Alps or in the Po valley. In the Central Alps the plot LOM1, located in small scarcely populated valley, reported the lowest concentrations among the plots in Northern Italy. Direct  $O_3$  measurements and a model application confirm the rather protected character of the site where  $O_3$  is limited to local photochemical activity and not influenced by transportation phenomena (PIROVANO *et al.* 2000).

In Central Italy the lowest values were recorded at the plots TOS1 and UMB1, in the South at the plot CAL1. For TOS1 this result was unexpected as the plot is located a few kilometres away from the urban area of Livorno and from a major industrial area. For TOS1 and CAL1 the effect of forest canopies on  $O_3$  levels cannot be excluded as the two measurement sites are located at short distance from a dense forest stand.

Referring to the weekly mean concentrations of individual years at some plots differences between minima and maxima are very low (less than 20 ppb) for all monitoring periods as, for example, FRI1 and SAR1. At other plots (*e.g.* CAM1) the same differences exceed in most of the monitoring periods 35 ppb.

Figure 2 reports the five year  $O_3$  mean concentra-

**Table 4 -** Average weekly ozone concentrations (in ppb) recorded by passive samplers in the period 1996-2000 at the permanent monitoring plots.  
*Concentrazioni medie settimanali di ozono (in ppb) rilevate mediante campionatori passivi presso le aree di indagine permanenti.*

Year Period	1996 15/6-30/9		1997 17/6-1/10		1998 16/6-29/9	1999 4/5-28/9	2000 2/5-3/10
	Range	Pooled standard deviation	Range	Pooled standard deviation			
ABR1	22.7-48.5	0.54	40.0-49.7	3.97	34.5-54.3	35.0-60.0	32.4-59.2
BAS1	21.5-41.7	0.73	42.8-65.8	3.58	21.8-42.7	37.7-48.2	39.0-73.8
CAL1	20.4-35.8	0.39	35.8-52.1	2.03	27.4-37.2	27.1-48.6	38.0-54.6
CAM1	12.51-49.7	0.60	46.4-83.6	5.51	33.8-44.6	42.5-70.8	34.2-69.3
EMI1	18.5-41.6	0.36	30.8-57.0	2.25	27.3-46.8	29.3-48.7	40.2-64.0
EMI2	19.8-42.9	0.42	34.8-65.7	3.58	36.2-66.1	39.1-57.5	39.9-46.9
FRI1	20.8-40.3	0.48	25.8-42.6	5.23	30.0-47.8	38.0-54.6	31.3-48.7
FRI2	12.0-44.3	0.29	37.2-62.9	2.36	22.1-43.4	38.4-55.6	32.5-51.2
LAZ11	25.2-44.6	0.59	43.4-56.7	3.26	37.1-50.0	39.3-51.9	31.9-62.4
LOM1	-	-	27.1-49.8	2.01	27.7-40.7	32.2-44.5	26.6-44.3
MAR1	25.2-46.8	0.70	42.9-63.6	3.27	32.0-45.3	48.6-64.4	40.2-57.4
PIE1	24.4-44.7	0.89	32.7-49.4	4.51	31.2-37.4	37.3-56.2	30.4-50.6
PUG1	14.9-45.4	0.82	43.0-73.1	11.10	36.1-57.2	42.8-60.5	37.9-59.7
SAR1	25.3-43.9	0.58	36.0-50.4	1.86	27.2-39.5	42.8-58.6	35.2-52.3
SIC1	33.5-52.9	1.21	38.7-87.1	3.61	37.6-66.7	45.3-71.7	27.1-89.4
TOS1	23.5-40.2	0.70	31.4-48.2	3.79	29.2-43.5	37.0-55.0	31.9-47.1
TRE1	22.8-43.8	1.21	41.3-58.2	3.05	34.5-48.2	39.8-56.8	23.6-51.7
UMB1	21.4-44.1	0.52	39.2-55.8	2.53	27.1-38.3	37.8-57.0	37.0-47.6
VAL1	26.5-44.0	0.76	35.6-52.8	3.01	32.2-62.2	38.0-67.6	33.6-46.7
VEN1	25.6-42.3	0.26	31.0-46.0	4.66	30.4-51.7	36.5-50.6	39.8-47.1

tion (1996-2000) for the period from 24<sup>th</sup> week of the year to the 39<sup>th</sup>. 1996 can be considered as a low O<sub>3</sub> year at all plots (Table 5). Data from two EMEP monitoring station, Ispra (Varese) and Montelibretti (Rome) show coherent average O<sub>3</sub> concentrations for the period June – September 1996 of 26.4 and 30.5 ppb, respectively. On the other hand, for most of the plots 1997 and 2000 have to be considered high ozone years. The recommendation for a five year average for integrated assessment purposes (UBA 1996) appears thus fully justified.

Table 6 reports the maximum weekly concentration recorded in the period 1996-2000 at the permanent investigation plots. Data range from 40 to 87 ppb. These data reflects in most cases the differences among plots already highlighted for the mean concentrations over the monitoring periods.

Figure 3 reports temporal variations of O<sub>3</sub> mean weekly concentrations at the plot monitored. Generally, patterns among different years vary substantially due to the influence of climatic conditions. There are, however, exception as at most of the plots located in the Alps or at the plot TOS1 in Central Italy. From this point of view plots can be divided in three groups: areas with rather constant weekly concentrations, areas with occasional high means, and areas with complex patterns and frequent sudden variations in weekly mean concentration. These different pattern are reasonably due to both climate and transport of polluted air masses.

At the plot SIC1, located downwind of major



**Figure 2-** Five year (1996 - 2000) mean O<sub>3</sub> concentration (weeks 24 – 39) at the permanent investigation plots. At the plot LOM1 concentration data refer to the period from 1997 to 2000.  
*Concentrazioni medie quinquennali (1996 – 2000) di O<sub>3</sub> (settimane 24 - 39) presso le aree di indagine permanenti. Presso l'area LOM1 le misure di concentrazione sono riferite al periodo 1997-2000.*

**Table 5-** O<sub>3</sub> mean weekly ozone concentrations (in ppb) recorded by passive samplers in the period 1996-2000 at the permanent forest plots.  
*Concentrazioni massime settimanali di O<sub>3</sub> ozono (in ppb) rilevate mediante campionatori passivi nel periodo 1996-2000 presso le aree di indagine permanenti.*

Year Period	1996 15/6-30/9	1997 17/6-1/10	1998 16/6-29/9	1999		2000	
	4/5-28/9	15/6-28/9	2/5-3/10	13/6-3/10			
ABR1	35.8	44.2	42.3	45.5	44.6	45.8	44.6
BAS1	33.1	54.8	35.6	42.6	42.6	55.6	53.9
CAL1	30.5	42.9	33.1	23.5	39.8	46.2	46.0
CAM1	37.1	58.0	38.1	48.9	48.5	51.0	49.4
EMI1	33.0	49.2	39.8	39.6	40.2	52.2	49.3
EMI2	35.6	54.0	43.1	45.8	46.8	42.7	41.9
FRI1	30.4	36.3	39.8	43.6	45.4	38.9	39.4
FRI2	30.4	41.0	34.8	44.1	44.5	40.2	40.7
LAZ1	35.4	50.2	43.5	45.0	45.2	44.6	44.2
LOM1	-	37.6	34.8	38.0	37.5	37.7	37.4
MAR1	36.4	53.5	39.3	52.9	52.7	49.2	49.5
PIE1	34.2	42.7	34.5	45.0	46.0	43.7	43.0
PUG1	34.8	55.4	46.2	47.9	48.9	49.7	48.5
SAR1	36.3	43.2	35.3	47.6	48.6	46.4	46.4
SIC1	42.7	64.2	46.5	56.8	51.9	62.9	60.4
TOS1	32.6	41.0	38.3	46.4	45.8	41.5	42.4
TRE1	33.7	48.7	41.2	46.8	33.7	44.0	43.7
UMB1	31.5	46.2	34.3	47.7	46.8	43.4	42.8
VAL1	34.6	44.7	41.8	47.0	45.5	40.0	40.4
VEN1	32.8	39.8	36.1	42.1	43.0	43.4	43.3

**Table 6-** Maximum weekly ozone concentrations (in ppb) recorded by passive samplers in the period 1996-2000 at the permanent forest plots.  
*Concentrazioni massime settimanali di ozono (in ppb) rilevate mediante campionatori passivi nel periodo 1996-2000 presso le aree di indagine permanenti.*

Year Period	1996 15/6-30/9	1997 17/6-1/10	1998 16/6-29/9	1999 4/5-28/9	2000 2/5-3/10
ABR1	48.5	49.6	54.2	60.0	59.2
BAS1	41.7	65.7	42.7	48.2	73.8
CAL1	40.8	52.1	37.2	48.6	54.6
CAM1	49.6	83.6	44.6	70.7	69.3
EMI1	41.5	57.0	46.7	48.7	64.0
EMI2	42.8	65.7	66.0	57.5	51.0
FRI1	40.2	42.5	47.8	48.9	48.7
FRI2	44.2	62.9	43.3	55.6	51.2
LAZ11	44.6	56.7	50.0	51.8	62.4
LOM1	-	49.8	40.6	44.5	44.3
MAR1	46.8	63.6	45.3	64.3	57.4
PIE1	44.6	49.4	37.4	56.2	50.6
PUG1	45.3	73.0	57.1	60.4	59.7
SAR1	43.9	50.3	39.5	58.6	68.5
SIC1	52.9	87.0	66.7	71.7	89.4
TOS1	40.2	48.1	43.5	55.0	53.1
TRE1	43.8	58.2	48.1	56.7	51.7
UMB1	44.0	55.8	38.3	56.9	49.3
VAL1	43.9	52.8	62.1	67.5	46.7
VEN1	42.2	46.0	51.7	50.6	47.1

emission sources of precursors the effects of breezes in transport phenomena has been already highlighted (BISTACCHI *et al.* 1996). Apparently these transport mechanisms are not constant phenomena giving rise to relevant differences in concentration from one week to the following. The particular location of the passive samplers at SIC1, placed on the roof of a fire look-out tower and very exposed to wind direction changes, may also contribute to the sudden variations of  $O_3$  concentrations.

The constant regimes are recorded in most cases at plots located far from emissions sources of  $O_3$  precursors (PIE1, LOM1, VEN1) but this cannot be considered a general rule. Other factors such as ventilation seem also to play a relevant role. FRI1 located in the Venetian Plane is characterised by  $O_3$  concentrations lower than plots located in mountainous areas and by small variations during the monitoring periods. Here  $O_3$  scavenging due to the vicinity of  $NO_x$  sources and wind scarcity during the summer season are probably the two factors explaining the relatively low  $O_3$  concentration and the temporal patterns at this site.

In order to highlight differences among years and plots which can drive the analysis of interactions between  $O_3$  and forest ecosystems, permanent plots were grouped according to their geographical locations:

Northern Italy - VAL1, PIE1, LOM1, TRE1, VEN1, FRI1, FRI2, EMI1, EMI2  
 Central Italy - TOS1, LAZ1, UMB1, MAR1, ABR1, SAR1  
 Southern Italy - CAM1, BAS1, PUG1, CAL1, SIC1.

The two-way analysis of variance for the mean weekly concentration recorded at all plots for the monitoring periods 1996 – 2000 indicates significant differences between individual years (Table 7). In 1996 concentrations were significantly lower than the other years for all geographical areas, while 1998 can be described as a year with concentrations near the overall average and significantly different from the others (Table 8). 1997 and 2000 are significantly different from the other years in the northern and southern plots, while in the plots located in Central Italy the same is true for 1997 and 1999.

The same analysis of variance was performed between sites, grouped for their geographical location (Table 9). For plots in Northern Italy results indicate differences between the sites, with significant lower concentrations at the plot LOM1, FRI1, FRI2 and VEN1, all located in the north-eastern region, show concentrations which differ significantly from the other here considered plots. PIE1 and VAL1 in the north-west together with EMI1 form another group of plots with significantly different concentrations.

Referring to Central Italy the analysis identifies three groups of plots: TOS1 and UMB1 (lower concentrations), MAR1 (highest concentrations) and, finally, ABR1, SAR1 and LAZ1.

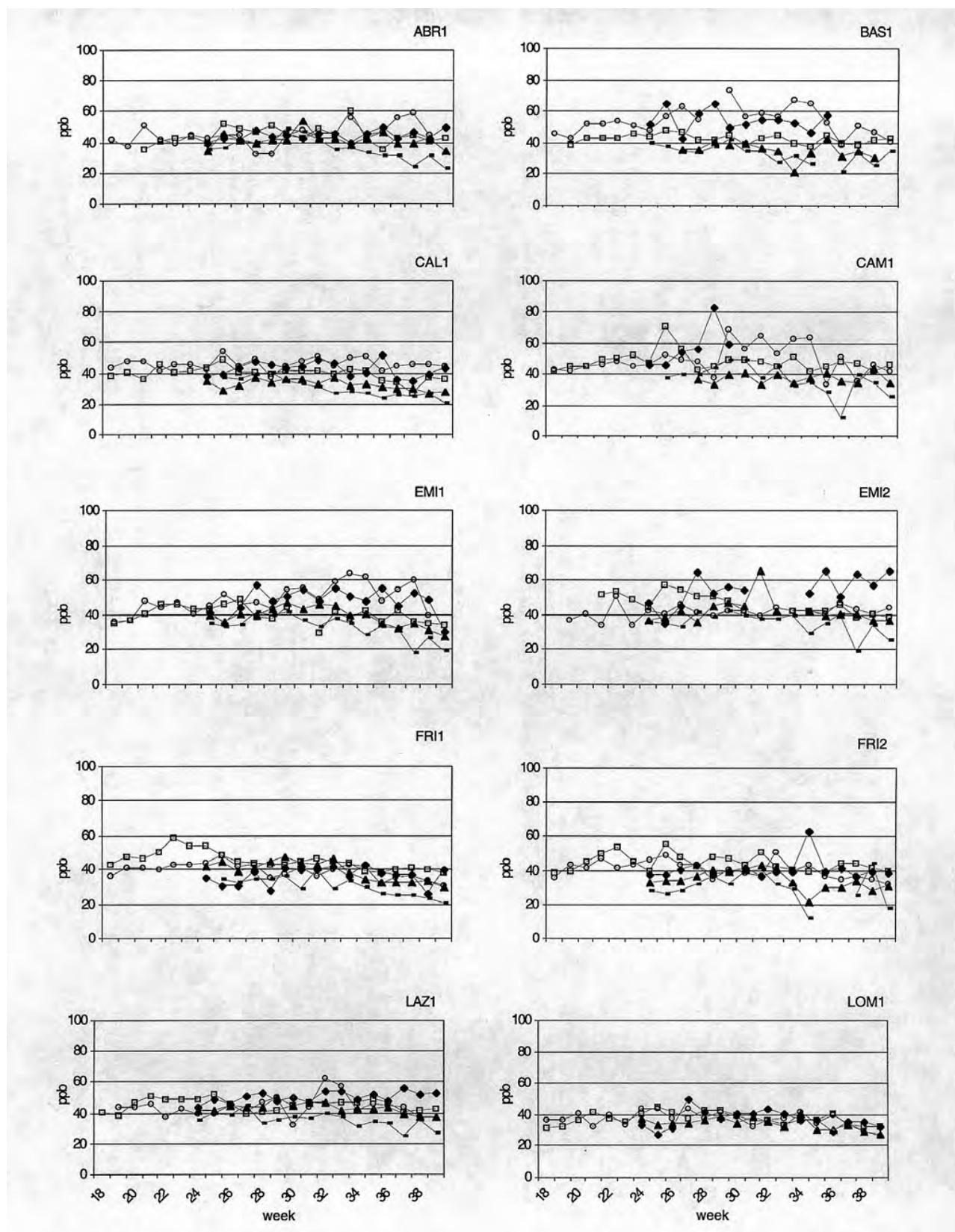
In the South concentrations at CAL1 (lowest values) and SIC1 (highest values) are significantly different from the other plots.

These data underline the regional character of  $O_3$  pollution. Differences among groups of plots and individual years can reasonably be explained as the result of different meteorological conditions, transport phenomena and site characteristics.

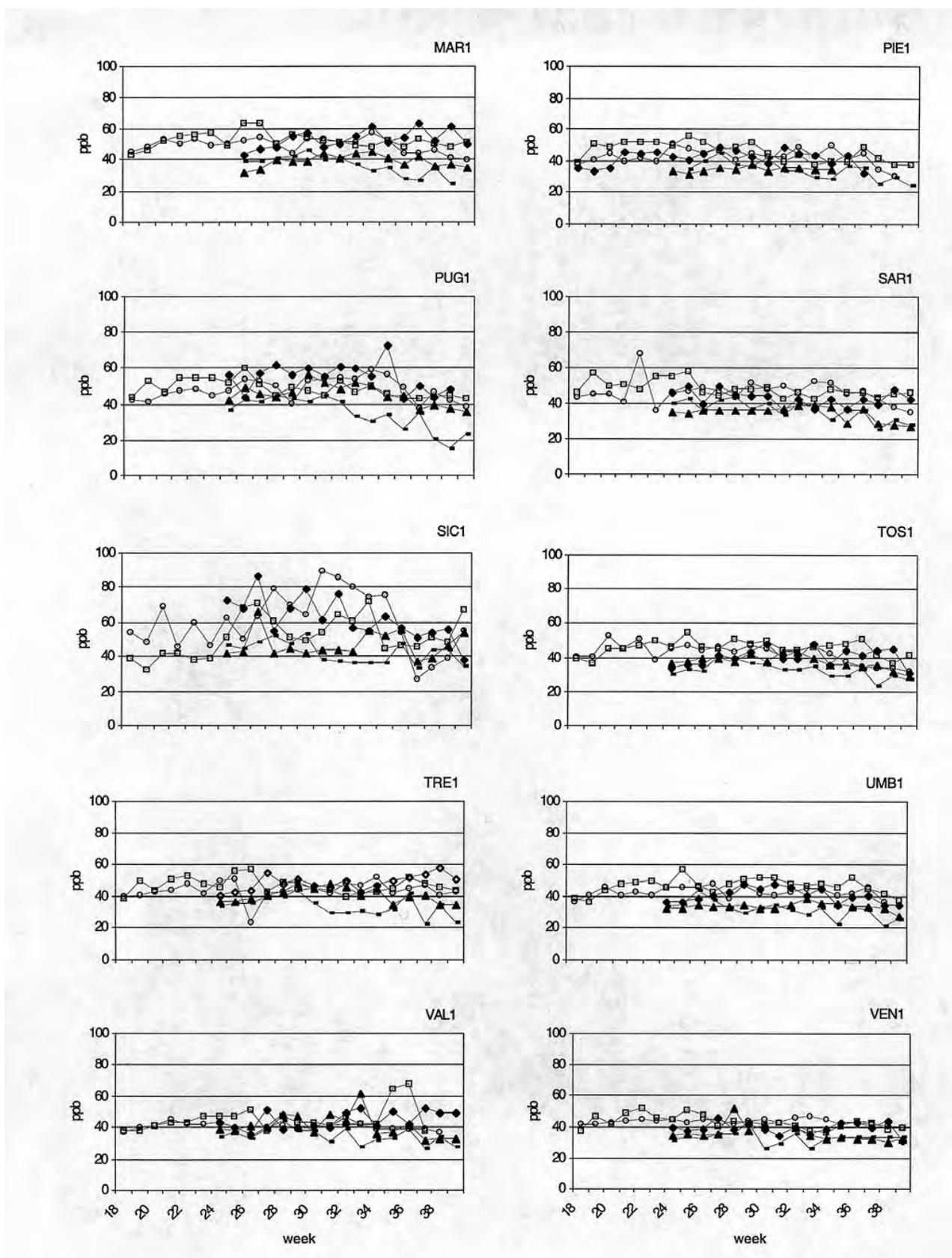
Although risk analysis for forest ecosystem is based on calculation of the accumulated exposure over a threshold of 40 ppb (AOT40) to assess tree exposure to  $O_3$  (Level I) and on  $O_3$  uptake by trees (Level II), ranking of monitoring plots according to  $O_3$  levels may contribute to evaluate forest health assessment data or to pin-point the areas where potentially risks may be the highest.

## Conclusions

The  $O_3$  monitoring programme carried out at the



**Figure 3-**  $O_3$  mean weekly concentrations at the permanent forest plots in 1996 (-), 1997 (♦), 1998 (▲), 1999 (□) and 2000 (○).  
*Concentrazioni medie settimanali di  $O_3$  presso le aree di indagine permanenti negli anni 1996 (-), 1997 (♦), 1998 (▲), 1999 (□) e 2000 (○).*



**Table 7-** Analysis of variance performed on years and permanent investigation plot grouped for geographical location.  
*Analisi della varianza condotta su anni e aree di indagine permanenti suddivisi per aree geografiche.*

Plot Location	Source	Sum of squares	Df	Mean square	F-ratio	p-Value
Northern Italy	Year	15138.3	4	3784.57	93.71	0.0000
	Plot	5870.61	8	733.827	18.17	0.0000
Central Italy	Year	13853.2	4	3463.31	112.41	0.0000
	Plot	2678.67	5	535.935	17.39	0.0000
Southern Italy	Year	20485.6	4	5121.4	76.60	0.0000
	Plot	10191.3	4	2547.83	38.11	0.0000

**Table 8-** Significance of differences in  $O_3$  concentrations between individual years in Northern, Central and Southern Italian permanent forest plots (different letters indicate significant differences ( $p<0.001$ ) as determined with the least significance distance (LSD) test.  
*Significatività delle differenze in concentrazione di  $O_3$  tra singoli anni nelle aree permanenti dell'Italia settentrionale, centrale e meridionale (lettere differenti indicano differenze significative ( $p<0.001$ ) come determinato dal test della minima distanza significativa (LSD).*

	Northern Italy	Central Italy	Southern Italy
1996	a	a	a
1997	b	b	b
1998	c	c	c
1999	d	b	d
2000	b	d	b

**Table 9-** Significance of differences in  $O_3$  concentrations between individual sites in Northern, Central and South Italian permanent forest plots. Different letters indicate significant differences ( $p<0.001$ ) as determined with the least significance distance (LSD) test.  
*Significatività delle differenze in concentrazione di  $O_3$  tra singoli siti dell'Italia settentrionale, centrale e meridionale. Lettere differenti indicano differenze significative ( $p<0.001$ ) come determinato dal test della minima distanza significativa (LSD).*

	Northern Italy	Central Italy	Southern Italy
LOM1	a	TOS1	a
FRI2	b	UMB1	a b
FRI1	b	ABR1	b c
VEN1	b c	SAR1	c
PIE1	c d	LAZ1	c
VAL1	d e	MAR1	d
EMI1	d e		
TRE1	e f		
EMI2	f		

permanent investigation plots of the CON.ECO.FOR. network shows the efficacy of indigo passive samplers to give a first estimate of  $O_3$  concentration levels at remote sites. Temporal variations and differences among plots may be used for a categorization of ozone risk at the considered forest areas.

Correlation coefficients of measurements carried out with passive samplers and continuous monitors reported in literature and data from parallel measurements carried out during this study highlight their potential for air quality assessment in remote areas. Site selection and standardised procedures in monitoring activities have to be carefully planned and carried out. Data of five year monitoring of  $O_3$  concentration recorded by passive samplers at the permanent plots show high  $O_3$  levels at all plots and particularly at the plots located in Central and Southern Italy. These data highlight the need for further investigations and for measurements with instruments with higher temporal resolution for an assessment of short-time concentrations. The identification of significant differences in  $O_3$  concentration levels among years and plots may contribute to direct properly the assessment of  $O_3$  effects on forest ecosystems. Moreover the  $O_3$  data collected in this study have a particular relevance as they represent for some Italian regions new, original information regarding  $O_3$  levels and temporal variations in remote areas.

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# Ozone levels and meteorological variables at the permanent monitoring plots of the CONECOFOR programme in Italy<sup>§</sup>

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**Abstract** – Weekly ozone ( $O_3$ ) values measured at the permanent monitoring plots (PMPs) of the CONECOFOR programme were processed together with meteorological data. The aims were: (i) to identify the most important factors associated to  $O_3$  concentrations and (ii) to identify common patterns (if any). Principal Component Analysis and multiple regression models were used. The results of both techniques were rather similar at the site level and identified three major categories of variables associated to  $O_3$ : an energetic, radiative component (solar radiation and its proxies, i.e. number of hours of sunshine and temperature) which accounts for 40-50% of the variance; a wind-related component (13-20% of variance explained), and a third component that varied from site to site.

**Key words:** ozone, Italy, forests, meteorological variables, site factors, principal component analysis, multiple regression.

**Riassunto** – Livelli di ozono e variabili metereologiche presso le aree permanenti CONECOFOR in Italia. I valori settimanali di ozono misurati nel periodo 1996-2000 presso le aree permanenti CONECOFOR sono stati analizzati in relazione ai dati meteorologici raccolti presso gli stessi siti. Per l'analisi statistica sono state utilizzate l'analisi delle componenti principali e la regressione multipla. I risultati di entrambe le tecniche utilizzate sono stati simili ed identificano tre principali categorie di variabili associate alle concentrazioni di ozono: una di natura energetica (radiazione solare e suoi proxy) che spiega circa il 40-50% della varianza; una collegata al vento, che spiega circa il 13-20% della varianza; ed una di altra natura che cambia di volta in volta e di sito in sito.

**Parole chiave:** ozono, Italia, foreste, variabili meteorologiche, fattori stazionali, analisi componenti principali, regressione multipla.

The origin, dynamics and concentration levels of tropospheric ozone ( $O_3$ ) depends on a variety of chemical and physical factors and on their interactions (e.g. PORG 1997). For example, while the formation of photochemical  $O_3$  is the result of the sunlight-initiated oxidation of VOCs (volatile organic compounds) in the presence of  $NO_x$  (Nitrogen oxides, NO and  $NO_2$ ), the occurrence of high  $O_3$  concentration is frequent in rural/ remote and/or high altitude sites, e.g. sites where the emission of  $O_3$  precursors is negligible (e.g. COYLE *et al.* 2002). This is due – in general – to phenomena of transport of either  $O_3$  or  $O_3$  precursors from emission areas to downwind located rural/forest areas. In this context, the wind regime and the geographic characteristics of the surrounding region are important factors affecting the  $O_3$  levels at a given site

(FOWLER *et al.* 1999; MILLÁN *et al.* 1992, 1997).

Despite the considerable complexity of air masses dynamics, the dependence of  $O_3$  levels and diurnal cycles upon geographical and meteorological characteristics was substantiated by different, well-documented  $O_3$  patterns. For example, a latitudinal gradient ( $O_3$  increases from North to South Europe, DOLLARD *et al.* 1995) and an altitudinal gradient (high-altitude sites tend to have a smoothed 24-hour profile of  $O_3$  concentrations, LOIBL and SMIDT 1996; BRONNIMANN *et al.* 2000) were reported. Acknowledging and quantifying the relationships between  $O_3$  concentrations and other characteristics of the environment is important when considering the current structure of  $O_3$  monitoring networks and the necessity to know or estimate  $O_3$  exposure of forest sites. Automatic monitoring devices are most frequently located in

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urban areas, with very little coverage of rural and forest areas (DE LEEUW and BOGMAN 2001; DESIATO *et al.* 2000; PORG 1997; MILLÁN *et al.* 2000). While the use of passive sampling can help the collection of data over natural and remote areas (KRUPA *et al.* 2001), it may not always be practicable on a large scale with sufficient sampling density. On the other hand, mapping of O<sub>3</sub> levels only on the basis of the measured values at some, sparse sites without considering the local topography and the role of meteorological parameters can result in considerable errors (COYLE *et al.* 2002). This is especially true for Italy because of different O<sub>3</sub> regimes (see FERRETTI *et al.* 2003) and the knowledge of the relationships between O<sub>3</sub> and environmental parameters can be helpful to identify the nature of O<sub>3</sub> pollution at a given site. Therefore, the identification of the most important factors associated to O<sub>3</sub> concentrations and the identification of the relationship between environmental parameters and O<sub>3</sub> concentration at the site scale can be a considerable step forward towards the development of effective, tailored predictive models and mapping.

Several papers have reported the relationships occurring between O<sub>3</sub> and meteorological parameters, like air temperature, relative humidity, wind speed and radiation (*e.g.* FREDERICKSEN *et al.* 1996; OLSZYNA *et al.* 1997). This paper illustrates an explorative statistical analysis carried out on O<sub>3</sub> and meteorological data collected at the Permanent Monitoring Plots (PMPs) of the CONECOFOR programme over the period 1996-2000. The aim is the identification of the most significant

variables (at a given PMP and in general) related to the measured O<sub>3</sub> levels.

## Materials and Methods

### Collection, quality and reliability of data

The description of the PMPs of the CONECOFOR programme is provided by ALLAVENA *et al.* (2000). Within the CONECOFOR programme, meteorological data are collected in the open field (generally at no more than 2 km from the plot) and below the forest canopy in the actual plot. For the purposes of this paper, only open field data are considered. The measured parameters include: Air Temperature (AT) at 10, 2 and 0.1 m, respectively; Relative Humidity (RH) at 10, 2 and 0.1 m, respectively; Soil Temperature (ST) at 0.2 m; Solar Radiation (SR) at 2 m; Precipitation (PR) at 2 m; Snow Depth (SD) at 5 m; Wind Speed (WS) at 10 and 2 m; Wind Direction (WD) at 10 m. Details relating to the technical apparatus and the routine procedures are provided by AMORIELLO *et al.* (2000). Data have been checked for their completeness and plausibility according to FERRETTI and NIBBI (2000). Table 1 provides an overview of the periods covered by the measurements over the years 1996-2000. Several gaps exist, especially during 1997 and 1998. Actually, the summer season (*i.e.* the season important for O<sub>3</sub>) was covered at one PMP in 1996 (TOS1), at two in 1997 (LAZ1 and TOS1), at six in 1998 (ABR1, EMI1, FRI2, LAZ1, TOS1 and TRE1) and at all the 11 PMPs equipped with the meteorological devices in 1999 and 2000.

**Table 1** - Periods (day and month) covered by meteorological measurements at the CONECOFOR PMPs over the years 1996-2000.  
*Periodi (giorno e mese) coperti dalle misurazioni meteo le aree permanenti CONECOFOR negli anni 1996-2000.*

No.	PMP	1996				1997				1998				1999		2000	
		from	to	from	to	from	to	from	to	from	to	from	to	from	to	from	to
1	ABR1					1/12	31/12			1/1	21/10	1/11	31/12			1/1	31/12
2	BAS1															1/1	31/12
3	CAL1															1/5	31/12
4	CAM1															1/1	31/12
5	EMI1							1/11	31/12			1/1	31/12			1/1	31/12
6	EMI2															1/1	31/12
7	FRI1															1/1	31/12
8	FRI2															1/1	31/12
9	LAZ1					1/5	17/11	25/11	31/12			5/6	31/12			1/1	31/12
10	LOM1							1/1	31/12			1/1	31/12			1/1	31/12
11	MAR1															1/1	31/12
12	PIE1															16/11	31/12
13	PUG1															1/1	31/12
14	SAR1															1/1	31/12
15	SIC1															1/1	31/12
16	TOS1	1/1	31/12			1/1	2/10	22/11	16/12			1/1	14/4	20/6	31/12	1/1	31/12
17	TRE1							1/1	31/3	21/4	10/5	26/9	6/12	1/1	31/3	21/4	10/5
18	UMB1															1/1	31/12
19	VAL1	1/1	14/2	16/5	13/6	20/6	27/6	1/1	4/6	10/6	31/12					1/1	31/12
20	VEN1															1/1	31/12

**Table 2 - Completezza dei dati alle varie aree permanenti ed ai vari anni. I dati sono espressi come % di dati effettivamente raccolti rispetto al totale dei dati attesi. Abbreviazioni nel testo.**

N.	PMP	1996					1997					1998					1999					2000								
		PR	AT	RH	WS	WD	SR	ST	PR	AT	RH	WS	WD	SR	ST	PR	AT	RH	WS	WD	SR	ST	PR	AT	RH	WS	WD	SR	ST	
1	ABR1	100	100	100	100	100	100	100	100	100	100	100	100	100	100	73	73	73	73	63	63	73	90	88	90	90	63	79	90	
2	BAS1															86	86	86	86	86	86	86	97	97	97	97	97	97	97	
3	CAL1	100	100	100	100	100	100	100	100	100	100	100	100	100	100	98	98	98	98	98	98	98	100	100	100	100	89	89		
4	CAM1															99	99	99	99	99	99	99	94	93	93	94	94	84	94	
5	EMI1	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100		
6	EMI2															100	100	100	100	100	100	100	98	98	98	98	98	96	96	96
7	FRI1															100	100	100	100	100	100	100	96	96	96	96	96	88	88	85
8	FRI2	100	100	100	100	92	100	100	100	100	100	100	100	100	100	94	96	96	96	96	96	96	71	100	100	99	100	100	99	
9	LAZ1	100	100	100	100	92	100	100	100	100	100	100	100	100	100	85	83	83	83	83	83	83	100	99	99	99	99	99	99	
10	LOM1	68	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100		
11	MAR1																													
12	PIE1																													
13	PUG1																													
14	SAR1																													
15	SIC1																													
16	TOS1	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100		
17	TRE1																													
18	UMB1	83	83	83	83	83	83	83	83	83	83	83	83	83	83	100	100	100	100	100	100	100	100	100	100	100	100	100	100	
19	VAL1																													
20	VEN1																													

The overall meteorological data, and their completeness, for the periods covered by the measurements are reported in Table 2. In general, the completeness is high: exceptions are the year 1999 at ABR1, CAL1, PIE1 and TRE1 as well as some parameters at TOS1 in the year 2000 (Table 2). Table 3 reports the plausibility of the data expressed as % of plausible records in relation to overall data. Within the available data sets, plausibility is always high. Table 4 reports an overview of the main parameters over the period May-September in the years 1996-2000 for the PMPs used for this study.

Data on collection, quality and reliability of O<sub>3</sub> data are provided by BUFFONI and TITA (2000, 2003).

### Hypothesis and objectives

In order to study the multivariate relationships between meteorological measurements and O<sub>3</sub>, principal component analysis and multiple regression were used for purely explorative/descriptive purposes, based on two considerations: firstly, that the measurement plots were not selected according to an experimental design specifically conceived for the elaboration of a predictive model; secondly, the size of the measurement sample is not sufficient for the validation of the model (see below).

The objectives of the investigations are:

- An evaluation of the capacity to describe mean weekly O<sub>3</sub> levels without direct passive measurements, by means of regression models;

**Table 3 -** Data plausibility reported as % of plausible records out of total number of records.

*Plausibilità dei dati riportata come % di dati plausibili sul totale dei dati raccolti.*

N.	PMP	1996	1997	1998	1999	2000
1	ABR1		100	100	100	100
2	BAS1				100	100
3	CAL1					100
4	CAM1					100
5	EMI1		100	100	100	100
6	EMI2				100	100
7	FRI1					100
8	FRI2				100	100
9	LAZ1		100	100	100	100
10	LOM1		100	100	100	100
11	MAR1				100	100
12	PIE1					100
13	PUG1					100
14	SAR1					100
15	SIC1					100
16	TOS1	100	100	100	100	100
17	TRE1			100	100	100
18	UMB1	83	83	83	100	100
19	VAL1	100	100	100	100	100
20	VEN1					100

**Table 4 -** Summary of the various parameters (mean values for AT, SR, RH, WS; sum for PR) over the period May-September in different years and PMPs.  
*Dati riassuntivi (medi per AT, SR, RH, WS; somma per PR) del periodo maggio-settembre per anno e per ogni area.*

PMP	Year	AT (°C)	SR (W/m <sup>2</sup> )	RH (%)	PR (mm)	WS10 (m/s)	WS2 (m/s)
01-ABR1	1998	13.0	218	72	364.4	4.7	3.8
	1999	13.2	230	74	186.4	4.4	3.6
	2000	14.0	252	67	206.4	4.6	3.7
03-CAL1	1999	16.6	206	77	517.3	1.6	0.9
	2000	16.1	216	78	476.2	1.6	0.9
05-EMI1	1998	19.7	195	67	296.8	1.5	1.0
	1999	20.2	205	70	232.2	1.4	1.0
	2000	21.1	233	66	205.0	1.5	1.1
06-EMI2	1999	15.8	175	79	369.7	1.8	0.5
	2000	15.8	241	74	237.6	1.8	0.5
08-FRI2	1998	13.9	172	83	826.2	1.1	0.9
	1999	13.8	179	90	886.3	1.1	0.8
	2000	13.6	202	88	501.2	1.1	0.8
09-LAZ1	1997	18.3	218	67	205.6	1.6	1.2
	1998	18.5	214	68	279.8	1.7	1.2
	1999	17.8	221	72	331.6	1.6	1.1
	2000	18.5	260	68	135.8	1.7	1.1
10-LOM1	1997	13.1	186	83	828.6	-	
	1999	13.6	208	84	-	0.6	
	2000	13.8	224	72	993.1	-	0.7
12-PIE1	2000	13.2	205	79	1319.0	1.8	1.6
16-TOS1	1996	19.0	213	81	280.0	-	0.6
	1997	20.9	218	79	154.0	-	1.4
	1998	20.9	217	83	215.5	-	0.6
	1999	20.0	192	90	182.5	-	0.5
17-TRE1	1997	9.2	166	80	535.6	-	
	1999	11.2	141	82	500.4	-	
	2000	11.7	175	74	656.2	-	
19-VAL1	1996	10.1	199	66	252.0	2.1	
	1997	11.3	201	66	312.2	2.1	
	1998	11.5	204	63	299.6	2.5	
	1999	11.3	218	70	558.8	2.3	
	2000	11.6	239	62	332.7	2.5	

AT = Air Temperature at 2m; SR = Solar Radiation; RH = Relative Humidity at 2m;  
 PR = Precipitation; WS = Wind Speed at 10 m (at 2 m for TOS1).

- The identification of local phenomena and their quantification;
- To provide indications on the measurements that it would be useful to perform, in order to improve the structure of the current regression model.

It is important to bear in mind that for the purposes of the explorative analysis the individual years of observation (and therefore their differences) were considered separately; whereas for the estimate of the regression parameters all observations were considered as originating from the same population. In fact, the variability of mean yearly O<sub>3</sub> concentrations is considerable (VECCHI and VALLI 1999; SANDRONI *et al.*

1994; LORENZINI *et al.* 1994) and the accepted practice (UBA 1996) is to refer to periods of at least five years in elaborating estimates and models. This was the practice which this study attempted to follow, compatibly with the data available.

#### Statistical methods

Two different statistical analyses were performed with different datasets: principal component analysis (PCA) and multiple regression analysis. Hereafter a brief summary of the two methods and relevant input data is given. Common to both analyses is the use of weekly averages of both meteorological data and derived parameters. This is because O<sub>3</sub> concentration data by passive sampling are available as weekly averages: therefore weekly averages of the meteorological parameters are needed to have fully paired data sets.

#### *The Principal Component Analysis approach*

Principal component analysis (PCA, JOLIFFE 1985) involves a mathematical procedure that transforms a number of (possibly) correlated variables into a (smaller) number of uncorrelated variables called principal components. The first principal component accounts for as much of the variability in the data as possible, and each succeeding component accounts for as much of the remaining variability as possible. Traditionally, PCA is performed on standardized data if the variances of individual variates differs greatly, or if the units of measurement of the individual variates differ; otherwise raw data can be used.

The objectives of principal component analysis are:

- To discover or to reduce the dimensionality of the data set.
- To identify new meaningful underlying variables.

To get an indication of the importance of each component and of its interpretation we must know what kind of output we obtain from a PCA. We can simply say that, given a sample of *m* cases and *n* variables in a matrix, say A, finding its principal components involves finding three matrixes U,  $\Sigma$ , and V so that

$$A = U \Sigma V'$$

The matrices have the following properties:

$$U [m \times n] \text{ and } V [n \times n]$$

are orthogonal matrices. The columns  $u_i$  of U = [u<sub>1</sub>,

...,  $u_n$ ] are the *left singular vectors*, and the columns  $v_i$  of  $V [v_1, \dots, v_n]$  are the *right singular vectors*.

$$\Sigma[n \times n] = \text{diag}(\sigma_1, \dots, \sigma_n)$$

is a real, nonnegative, and diagonal matrix. Its diagonal contains the so called *singular values*  $\sigma_i$ , where  $\sigma_1 \geq \dots \geq \sigma_n \geq 0$ .

The singular values contained in  $\Sigma$  give the relative importance of the components and are useful to compute the fraction of the total variance explained by the corresponding component.

$V$  contains the loadings, *i.e.* the coefficients of the linear combination of the original variables that defines the principal components;  $U$  contains the scores, *i.e.* the  $m$  cases projected in this space. The plots of the loadings and of the scores are a very useful aid to the interpretation of the principal components in terms of the original variables.

#### Input data

The variables adopted for the PCA are based on the data collected between the years 1996-1999 (Table 5) at the PMPs listed in Table 6. Given the high frequency of missing data (see Table 1, 2 and 6), PMPs, years and variables were selected according to the following criteria:

- minimize missing data (Table 6);

**Table 5 -** Parameters used in PCA. Codes used in the text, definition and reporting units.  
*Parametri utilizzati nella PCA. Codici utilizzati nel testo, definizioni ed unità di misura.*

Code	Definition	Unit
numorecalma	Weekly hours of calm	n
numoresole	Number of hours of solar radiation (SR) > 0	n
OPPioggiamed	Weekly mean rainfall	mm
OPPioggiasomma	Weekly sum of rainfall	mm
ORadsolMed	Mean solar radiation	$W \cdot m^{-2}$
OFTaria2Med	Mean air temperature at 2 m	$^{\circ}C$
OFTaria2Somorelince	Weekly sum of temperature at 2 m with SR > 0	$^{\circ}C$
OFUmid10Med	Mean relative humidity at 10 m	%
OFUmid2Med	Mean relative humidity at 2 m	%
OFVe10VelMed	Mean wind speed at 10 m	$m \cdot s^{-1}$
OFVe2Med	Mean wind speed at 2 m	$m \cdot s^{-1}$

- maximize the amount of information. A preliminary PCA was carried out to select those variables that, while having similar information potential to others, maintain a higher sample size;
- make use, when possible, the variables with the highest correlation with  $O_3$  (Table 7).

As an example, Figure 1 reports the loadings of the first two components in relation to the entire dataset available (all PMPs, all years). When interpreting the figure, the following criteria should be considered:

- position with respect to 0 is a relative one. PCA determines the direction, not the way of the axis:

**Table 6 -** Number of weekly records available for each PMP and year.  
*Numero di misure settimanali disponibili per sito, per variabile ed anno.*

PMP	Year	numorecalma	numoresole	OPPioggia med	ORadsol Med	OFTaria2 Somorelince	OFVe10Vel Med	OFVe2 Med	OFTaria2 Med	OFUmid10 Med	OPPioggiasomma	OFUmid Med
TOS1	96	19	19	19	19	19	0	19	19	0	19	19
VAL1	96	0	0	16	19	0	19	0	19	0	16	18
LAZ1	97	19	19	19	19	19	19	19	19	19	19	19
LOM1	97	0	19	19	19	19	0	0	19	0	19	19
TOS1	97	15	15	13	16	15	0	16	16	0	13	16
TRE1	97	0	0	18	19	0	0	0	19	0	18	19
VAL1	97	19	19	19	19	19	19	0	19	0	19	19
ABR1	98	19	19	19	19	19	19	19	19	19	19	19
EMI1	98	19	19	19	19	19	19	19	19	19	19	19
FRI2	98	19	18	19	19	18	19	19	19	19	19	19
LAZ1	98	19	19	19	19	19	19	19	19	19	19	19
LOM1	98	0	0	0	0	0	0	0	0	0	0	0
TOS1	98	15	15	18	19	15	0	19	19	0	18	19
TRE1	98	0	0	4	0	0	0	0	5	0	4	5
VAL1	98	19	19	19	19	19	19	0	19	0	19	19
ABR1	99	21	21	21	21	21	21	21	21	21	21	21
CAL1	99	23	23	23	23	23	23	23	23	23	23	23
EMI1	99	22	20	22	20	20	22	22	22	22	22	22
FRI2	99	23	23	23	23	23	23	23	23	23	23	23
LAZ1	99	22	22	22	22	22	22	22	22	22	22	22
LOM1	99	23	22	0	23	22	0	0	23	0	0	23
TOS1	99	23	23	23	23	23	0	23	23	0	23	14
TRE1	99	0	0	17	11	0	0	0	19	0	17	19
VAL1	99	13	23	23	23	23	23	0	23	0	23	23
Total		352	377	414	433	377	286	283	448	206	414	438

- the way is not significant for individual variables;
- the closer the variables, the higher the correlation;
- variables on opposite sides of the origin of the axis are negatively correlated;
- variables located along perpendicular lines are not correlated (independent).

According to the above criteria, the following variables were not considered:

- OFPioggiamed**, **OFUmid10Med** and **OFUmid2Med**. They are considered as “represented” by **OFPioggiasomma** (positive correlation), **numoresole** and **OFRadsolMed** (negative correlation).
- OFTaria2Med**. **OFTaria2Somorelucce** which is more directly correlated to  $O_3$  was considered instead.
- OFVe10VelMed**. **OFVe2Med**, which allows a larger data set, was used instead. This choice implied the exclusion of site VAL1: on the other hand it allowed us to complete 1996-1999 data series for TOS1.

At the end, the variables selected for PCA are: solar radiation (**OFRadsolMed**), sum of rainfall (**OFPioggiasomma**), no. of hours with wind calm (**numorecalma**), no. of sun hours (**numoresole**), wind speed at 2 m (**OFVe2Med**), sum of air T at 2 m during the daylight hours (**OFTaria2Somorelucce**).

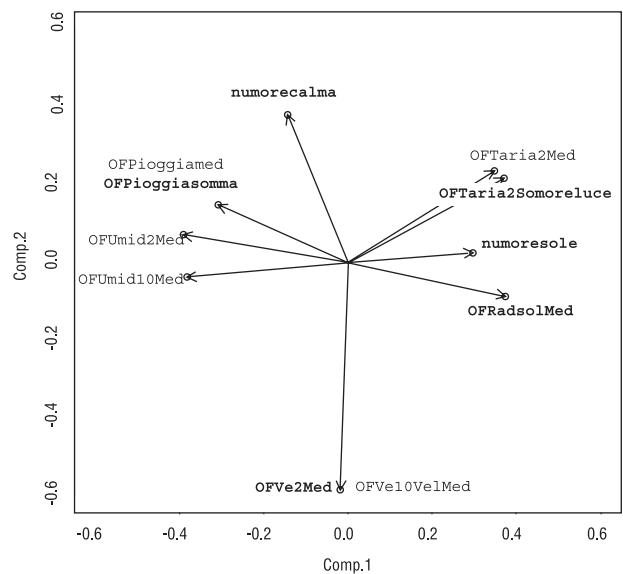
#### The multiple regression approach

The study of the response variable,  $O_3$ , as a function of some independent variables, the meteorological parameters, is carried out through correlation analysis and regression analysis. Correlation is concerned with

**Table 7-** Correlation coefficients between ozone and meteorological parameters used in the PCA.

*Coefficienti di correlazione tra ozono e parametri meteorologici utilizzati nella PCA.*

Variable code	<i>r</i>
numorecalma	-0,24
numoresole	0,45
<b>OFPioggiamed</b>	-0,41
<b>OFPioggiasomma</b>	-0,41
<b>OFRadsolMed</b>	0,57
<b>OFTaria2Med</b>	0,38
<b>OFTaria2Somorelucce</b>	0,44
<b>OFUmid10Med</b>	-0,50
<b>OFUmid2Med</b>	-0,45
<b>OFVe10VelMed</b>	0,09
<b>OFVe2Med</b>	0,10



**Figure 1 -** Loading plot with all the variables reprinted in Table 5. In bold: those variables selected for the PCA. See text for details.  
*Loading plot delle variabili riportate in Tabella 5. In grassetto le variabili scelte per la PCA.*

measuring the relationship or strength of association among sets of variables. Regression is concerned with prediction, that is, the ability to build a statistical model which uses information about a set of independent or predictor variables in order to estimate the expected value of some dependent or response variable.

The assumptions necessary for regression analysis are:

- normality: at each fixed X the probability density function for Y is identical, and it follows a normal distribution with mean  $\mu_y$  and variance  $\sigma^2_y$ ;
- linearity: it specifies the functional relationship between X and Y.
- independence: the observed Y values are independent of one other for each value of X.
- homoscedasticity: the variation or scatter about with respect to the line of regression  $\sigma^2_y$  are constant for all values of X.

The multiple linear regression model, containing  $p$  predictor variables, can be expressed in matrix form as:

$$Y = X\beta + \varepsilon$$

where  $Y = n \times 1$  column vector of observations  $Y_i$  ( $i = 1, \dots, n$ );  $X = n \times (p+1)$  matrix of quantitative independent variables;  $\beta = (p+1) \times 1$  column vector of parameters ( $\beta_0, \beta_1, \dots, \beta_p$ );  $\varepsilon = n \times 1$  column vector of

residual terms  $\varepsilon_i$ . The term  $\varepsilon_i$  represents scatter above and below the regression equation.

Using the method of least squares, we wish to obtain the  $\beta$  estimate, a  $(p+1) \times 1$  column vector of sample regression coefficients, so that the resulting sample regression provides the best linear fit to the observed data.

To measure the strength of the linear relationship between between Y and X, we use the coefficient of multiple determination  $R^2$  (it ranges from 0 to 1), which represents the proportion of the total variation in Y explained by the set of  $p$  independent variables selected.

#### Input data

Standard parameters measured at 2 m height (10 m for wind speed) over the period May-September in the years 1996-2000 at 5 PMPs were considered (Table 8). Unfortunately, it was not possible to apply statistical analysis for all 11 sites with meteorological data, as in some cases there were problems as to completeness of data and/or measured parameters. For example, at TOS1 wind data at 2 m were used, since standard wind data at 10 m were not measured.

A first stage was to explore the relationships between  $O_3$  and the measured variables.

The correlation analysis highlighted several variables that can explain the variability of  $O_3$  within the regression model. In particular, air temperature and humidity are known to be often correlated with  $O_3$  and can be considered as indicators of  $O_3$  formation. Indeed, these two parameters appear to be the effect, but not the cause, of climatic condition (solar radiation, precipitation, wind, seasonal trend, etc.).

The variables to be used in the multiple regression model were selected taking into account *a priori* knowledge about meteorological variables and their interrelationships. The selected variables were SR, PR, and wind data (calm, WS e wind direction). The frequency of wind direction was transformed into two new variables: direction with significant positive corre-

**Table 9-** Transformation of wind direction data. See text for details.  
*Trasformazione dei dati della rosa dei venti. Vedi il testo per i dettagli*

PMP	DIR_POS	DIR_NEG
CAL1	N-NW Sea	E, SE, S, SW Mountains
EMI1	SW (1998-99), E (2000-01) Anthropic	NE (1998-99), W (2000-01) Non anthropic
FRI2	N, NW, W, SW Anthropic	S, SE, E Mountains
LAZ1	NE, N, NW Anthropic	SE, S, SW Non anthropic
TOS1	-	-

lation with  $O_3$  (DIR\_POS) and direction with significant negative correlation with  $O_3$  (DIR\_NEG) ( $P < 0.05$ ). Depending on the PMP, the source of positive direction can be of anthropogenic or marine origin; on the other hand, the source of negative direction is almost always determined by mountains or large natural areas (Table 9). As DIR\_POS and DIR\_NEG are highly (negatively) correlated, they cannot be used together in the same multiple regression.

## Results

### The PCA approach

The likely, strong influence exerted by local site condition on  $O_3$  levels led us to perform PCA for each individual PMP. Table 10 reports the loadings per each site and the percent variance explained by the first three components. On average ca. 71% of the variance is explained by the first two components. In detail:

- the first component is always proportional to  $O_3$ , OFRadsolMed, numoresole, OFTaria2Somorelucce and inversely proportional to OFPioggiasomma. This component can be qualified as the “radiative component” and explains 44-67 % of the total variance;
- the second component is linked to OFVe2Med and numorecalma (see PMPs ABR1, CAL1, FRI2, LAZ1, TOS1, and, to a lesser extent, EMI1). It provides information that is obvious also from Fig.1. This component can be qualified as the “wind component” and explains 13-23 % of the total variance;
- the third component in each site seems to be linked to  $O_3$  of different origin, which is not always predictable by means of the meteorological measurements. It explains 9-14% of the variance and can include either  $O_3$  occurring late in the season, up to early Autumn (and this may explain the non- or low correlation with OFRadsolMed) and  $O_3$  formed elsewhere

**Table 8 -** Years and plots considered in the Multiple Regression Analysis.  
 Anni e PMP considerati nell'analisi di regressione multipla.

PMP	1996	1997	1998	1999	2000
03-CAL1				X	X
05-EMI1			X	X	X
08-FRI2			X	X	X
09-LAZ1		X	X	X	X
16-TOS1	X	X	X	X	

and subsequently transported at sub-national scales.

Besides the analysis of the loadings, the temporal development of the scores was investigated. As an example, Figure 2 reports the seasonal variation of the scores for PMP TOS1 in each year for each individual principal

**Table 10-** Loadings of the various parameters for the first three principal components for each site and percent variance explained. From this table it is possible to explain the various principal components in terms of their actual meaning.

*Pesi delle misure meteorologiche sulle prime tre componenti principali per sito e varianza spiegata dalle stesse componenti nei medesimi siti. A partire da questa tabella e' possibile interpretare le componenti principali in termini delle misure meteorologiche.*

PMP	Variable	Comp.1	Comp.2	Comp.3
ABR1	ozone	-0.31	0.22	0.84
	OFRadsolMed	-0.49	-0.04	-0.19
	OPPioggiasomma	0.40	0.09	-0.07
	numorecalma	0.18	-0.75	0.27
	numoresole	-0.44	-0.02	-0.41
	OFVe2Med	0.30	0.60	0.00
	OFTaria2Somoreluge	-0.44	0.10	0.07
Variance explained		54%	17%	11%
CAL1	ozone	-0.45	0.08	0.38
	OFRadsolMed	-0.46	-0.09	0.00
	OPPioggiasomma	0.38	0.25	-0.54
	numorecalma	0.33	-0.33	0.59
	numoresole	-0.46	0.15	-0.12
	OFVe2Med	-0.10	0.76	0.16
	OFTaria2Somoreluge	-0.34	-0.47	-0.41
Variance explained		54%	20%	10%
EMI1	ozone	-0.39	-0.31	0.01
	OFRadsolMed	-0.44	-0.13	-0.03
	OPPioggiasomma	0.30	-0.36	0.87
	numorecalma	0.25	-0.80	-0.42
	numoresole	-0.40	-0.29	0.09
	OFVe2Med	-0.40	0.13	0.16
	OFTaria2Somoreluge	-0.43	-0.16	0.16
Variance explained		67%	13%	9%
FRI2	ozone	-0.35	-0.20	0.60
	OFRadsolMed	-0.50	-0.06	-0.11
	OPPioggiasomma	0.34	0.29	0.76
	numorecalma	0.00	-0.73	0.15
	numoresole	-0.46	0.06	0.06
	OFVe2Med	-0.30	0.59	0.01
	OFTaria2Somoreluge	-0.46	-0.02	0.16
Variance explained		53%	23%	9%
LAZ1	ozone	-0.32	-0.05	-0.59
	OFRadsolMed	-0.54	0.02	0.11
	OPPioggiasomma	0.27	0.41	-0.20
	numorecalma	0.04	-0.69	0.46
	numoresole	-0.51	0.17	0.17
	OFVe2Med	0.11	0.55	0.58
	OFTaria2Somoreluge	-0.51	0.13	0.15
Variance explained		44%	18%	14%
TOS1	ozone	-0.33	-0.28	-0.68
	OFRadsolMed	-0.48	-0.11	0.17
	OPPioggiasomma	0.38	-0.04	-0.44
	Numorecalma	0.23	-0.66	-0.23
	Numoresole	-0.46	-0.19	0.09
	OFVe2Med	-0.18	0.65	-0.50
	OFTaria2Somoreluge	-0.48	-0.11	-0.06
Variance explained		51%	19%	11%

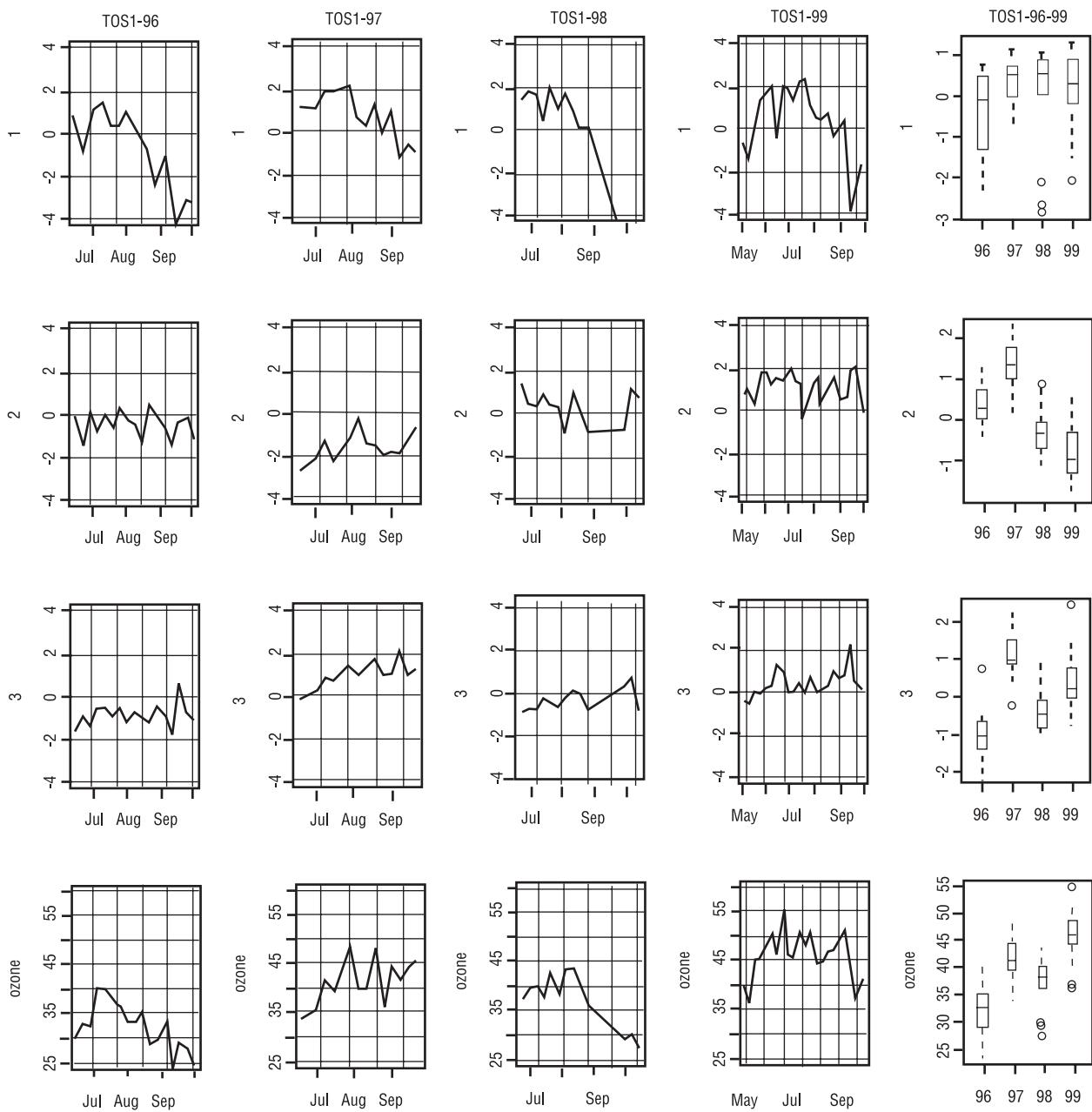
component and for  $O_3$ . Interestingly, the component mostly associated to the inter-annual variation of  $O_3$  levels is not the "radiative component", but the third component (see Figure 2, last column). This may suggest two considerations: first, the weather (in particular solar radiation) drives the seasonal  $O_3$  trend and level. This is in agreement with what we know about  $O_3$  formation in the troposphere (SEINFELD and PANDIS 1997). Second, the occurrence of correlations between  $O_3$  levels and wind calms and/or rainy weeks, provides evidence that other phenomena may take place, especially at the end of Summer/early Autumn, and varying year by year (see Figure 2, individual years and the whole period in the last column).

The fact that the variability of the third component is in line with the variability of  $O_3$  (Fig. 2 last column) suggests that this component may contribute to explain inter-annual variations. However, this interpretation remains uncertain: for example, the correlation between  $O_3$  and solar radiation is stronger in late summer/early autumn, suggesting that the non-radiative component is active especially during the summertime (Table 11).

The potential influence of the third component on the role exerted by the radiative component is suggested by another element. Fig. 3 reports the cross-correlation between  $O_3$  and solar radiation and the autocorrelation between  $O_3$  values at subsequent weeks at PMP TOS1. Usually, the  $O_3$  weekly mean exhibits low autocorrelation values and an irregular autocorrelation pattern, like the data for 1997 in Fig. 3. An exception is the year 1998, when the  $O_3$  autocorrelation values were quite high and with a regular pattern for all the sites. Interestingly, in this year, the correlation values between  $O_3$  and solar radiation were also the highest (see Fig. 3) and this was true for all investigated PMPs. Evidence of this is obvious also from Fig. 2 (see the years 1997 and 1998 with the seasonal course of  $O_3$  and the first and third principal component). This suggests that, when the radiative component exerts a stronger control on  $O_3$  levels, the latter has a more regular temporal pattern (high temporal autocorrelation). Together with the data reported in Table 11 and Fig. 2, the above findin-

**Table 11-** Coefficient of determination ( $R^2$ ) calculated after the regression  $O_3$  vs. solar radiation for each PMP and period.  
*Coefficiente di determinazione ( $R^2$ ) della regressione ozono vs radiazione solare per stagione e sito.*

Period	ABR1	CAL1	EMI1	FRI2	LAZ1	TOS1
May-August	0.03	0.56	0.32	0.12	0.08	0.13
September-October	0.53	0.33	0.49	0.35	0.65	0.54



**Figure 2** - Time development of the scores of the first 3 principal components and ozone (from top to bottom) over the period 1996-1999 at the PMP TOS1. The first 4 columns report the seasonal trend; the last column reports the inter-annual trend.

*Andamento temporale degli scores delle prime tre componenti principali e dell'ozono (dall'alto verso il basso) per il periodo 1996-1999 al PMP TOS1. Le prime 4 colonne riportano l'andamento stagionale per ogni singolo anno; l'ultima colonna riporta le variazioni inter-annuali.*

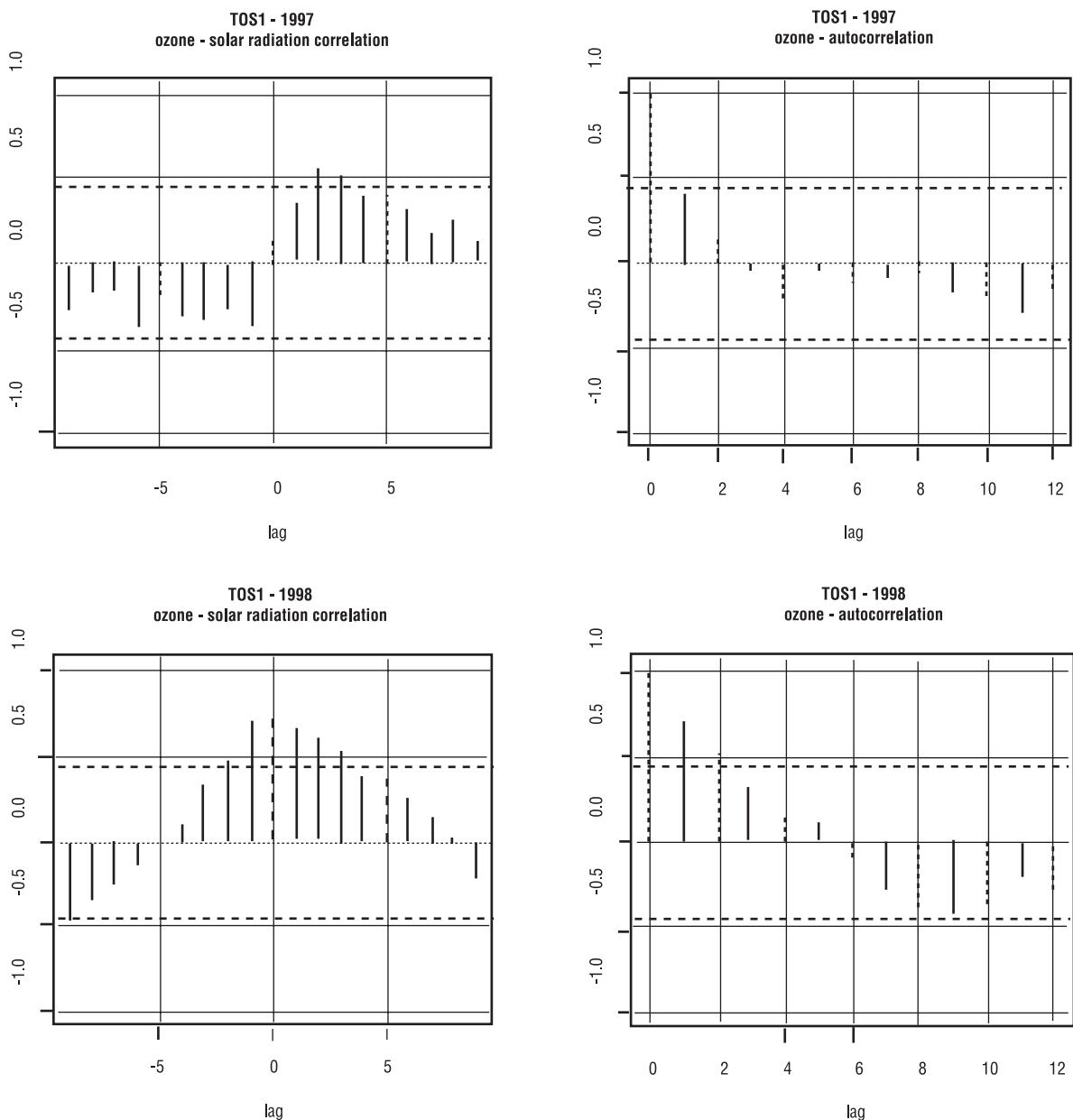
gs suggest that there are other factors that may control the  $O_3$  levels; that these factors are more active during summertime; and that they "disturb" the expected relationship between  $O_3$  and the radiative component.

#### ***The multiple regression approach***

Two different multiple regression models were built. The first one (Table 12) considered the following variables: SR, PR, WS10, CALM, DIR\_POS (10

m); the second one (Table 13) considered the variables SR, PR, WS2, numorecalma (with WS 2<0.5 m/s). Three different procedures were used to test the models: backward, forward and stepwise procedures provided nearly identical results.

Results are not always consistent even for the same PMPs in different years. This is because the individual, annual trends are always different. This is probably due to the high variability of  $O_3$  between



**Figure 3 -** Cross-correlation with solar radiation (left) and ozone autocorrelation (right) for the PMP TOS1. Lag: unit shift of the time series of the data (weeks).  
*Cross-correlazione con la radiazione solare media ed autocorrelazione dell'ozono nel sito TOS1. lag: shift in unita' temporali sulla serie storica (in questo caso settimane) utilizzato per il calcolo della correlazione.*

PMPs and years. The same results were obtained when considering the wind data collected at 2 m height. This is in line with the third principal component of the PCA results. In both models solar radiation and wind data are the most significant predictors, while precipitation has a minor role.

To improve the models, the following interactions were adopted at a second stage: SR-AT, RH-PR, WS10-DIR\_POS, WS10-DIR\_POS-CALM.

The significance of the interactions was tested by a Generalised Linear Model (GLM). There was a general improvement with  $R^2$  increasing by 10 %, in both the global model (all PMPs together) and in the model for each PMP. While in the global model the interaction between SR-AT was the most important one, the individual PMPs have different features. In particular

- interactions were not significant for PMPs CAL1 and LAZ1;

**Table 12-** Variables that appeared significant after multiple regression model 1 (wind measured at 10 m height). Variables are ranked according to significance level.  
*Variabili significative risultanti dall'applicazione del modello di regressione lineare multipla con i dati di vento a 10m (in ordine di significatività).*

PMP	Variabili significative (p-value)	R <sup>2</sup>
CAL1	SR (<0.0001)	0.44
EMI1	DIR_POS (<0.0001), WS10 (0.0005), PR (0.0184), CALM (0.0451)	0.70
FRI2	CALM (0.0016), SR (0.0035), WS10 (0.0309)	0.45
LAZ1	CALM (0.0078), WS10 (0.0235), DIR_POS (0.0439), PR (0.0555)	0.24
Global	SR (<0.0001), WS10(0.0361)	0.33

- interaction SR-AT was significant for PMP EMI1 (P= 0.01);
- interactions SR-AT and PR-RH were significant for PMP FRI2 (P= 0.06 and P= 0.01, respectively);

This study provides evidence that some variables not considered in the first stage may contribute to the understanding of the phenomenon being investigated, if considered interactively with other variables. A possible further step would be to understand better the interactions in order to identify a mathematical formulation.

## Conclusions

The O<sub>3</sub> concentration data were analysed in order to identify possible relationships with meteorological data. The approach suffers limitations due to the limited number of sites available and the nature of the network (*e. g.* the ability of the site to represent wind fields on a regional scale). Yet, through regression analysis and PCA, it was possible to sum up the dependence of O<sub>3</sub> from meteorological parameters by means of three factors:

- an energy factor, of a radiative nature, which probably also depends on the thermal level of energy utilization and/or on daily solar radiation;
- a transportation factor (direction, intensity and frequency of the wind), whose interaction is not linear;
- a removal factor (rain).

The data collected confirm the role of air temperature and solar radiation as factors controlling O<sub>3</sub> levels, on a seasonal scale; they also highlight the presence in all sites of at least one further source of O<sub>3</sub>, statistically not correlated to the O<sub>3</sub> of local photochemical origin. This source would appear to be related to the wider fluctuations of O<sub>3</sub> values on a weekly basis, primarily during the hottest months of the year, and to the mean yearly values. The presence *in situ* of other phenomena,

**Table 13-** Variables that appeared significant after multiple regression model 1 (wind measured at 2 m height). Variables are ranked according to significance level.  
*Variabili significative risultanti dall'applicazione del modello di regressione lineare multipla con i dati di vento a 2 m (in ordine di significatività).*

PMP	Variabili significative (p-value)	R <sup>2</sup>
CAL1	SR (<0.0001), numorecalma (0.0012), WS2 (0.0229)	0.34
EMI1	WS2 (0.0070), numorecalma (0.0468)	0.48
FRI2	SR (<0.0001)	0.34
LAZ1	numorecalma (0.0028), SR (0.0369), WS2 (0.0402)	0.22
TOS1	SR (0.0045), WS2 (0.0351), numorecalma (0.0643)	0.21
Global	SR (<0.0001), numorecalma (0.0012), WS2 (0.0229)	0.34

over and above local photochemical production, suggests that a global model for multiple linear regression (*i.e.* one valid for all sites) for ozone will only explain 30-35% of the variance.

In view of the results obtained from the regression models, and in order to predict more accurately the weekly concentrations of O<sub>3</sub>, it appears necessary to characterize the sites in a more detailed manner. In particular, atmospheric circulation in relation to surrounding conditions (*e.g.*, presence of pollutant sources such as towns, industries or roads, topography..) needs to be described in details.

Alongside fairly marked common characteristics there are, however, some individual situations (*e.g.* sites displaying almost exclusively photochemical phenomena, such as EMI1). In view of this, it may be important to identify groups of similar sites, to model as groups, so as to increase the statistical accuracy of the estimates.

One of the main limitations in the analyses performed was the scarcity of datasets available, covering (and not always completely) only a few years and a limited number of sites. This did not allow for the construction of valid model, *i.e.* one with a predictive value. Despite these limitations, however, the results obtained through regression analysis are promising and suggest that it will be possible to obtain more conclusive findings when more datasets are available.

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# Vegetation exposure to ozone at the permanent monitoring plots of the CONECOFOR Programme in Italy: estimating AOT40 by means of passive samplers<sup>§</sup>

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**Abstract** – Ozone ( $O_3$ ) concentrations are being measured at all the CONECOFOR Permanent Monitoring Plots by means of passive samplers. Measurements with passive samplers typically result in data about weekly-to-monthly  $O_3$  concentrations which are not consistent with the definitions of cumulative exposure indices, like the AOT40 adopted in Europe ( $O_3$  Accumulated Over Threshold 40 ppb). This paper describes an empirical method adopted to estimate AOT40 values at 20 forest sites in Italy starting from mean concentrations obtained from passive samplers. The method is mainly based on the  $O_3$  data from passive samplers collected at all the sites since 1996 and on the Loibl function, which describes the  $O_3$  daily profile as a function of relative altitude (the difference between the altitude of the site and the lowest altitude within a 5.0 km radius). When the theoretical daily profile for the site is known, then it is adjusted to match the weekly  $O_3$  concentration measured by the passive sampler, assumed as 24 hours daily average. The underlying, simplifying assumption is that daily  $O_3$  profile are the same every day of the week. Then, daily exceedance of 40 ppb are computed and summed for each week. Weekly mean  $O_3$  concentrations from passive samplers were validated against co-located automatic measurement stations (when present). Estimated AOT40 values ( $AOT40_e$ ) were evaluated either against co-located automatic measurement stations (when present) and with a series of independent measurement stations located throughout Italy whose weekly mean  $O_3$  values were used to simulate passive samplers. Results show a good correlation ( $R^2: 0.93$ ) between measured and estimated AOT40 values. The median absolute difference between measured and observer values is 15.4% (6.03% at rural and remote sites). The model tends to underestimate for AOT40 values up to 30000 ppb $^{*}h^{-1}$ , while over estimation seems to occur for higher AOT40 values.

**Key words:** ozone, passive samplers, AOT40 estimates, forests, Italy.

**Riassunto** –Esposizione della vegetazione all'ozono presso le aree permanenti CONECOFOR in Italia: stima dell'AOT40 a partire dai campionatori passivi. L'ozono ( $O_3$ ) viene misurato mediante dosimetri passivi presso tutte le aree permanenti del programma CONECOFOR. Le misurazioni tramite dosimetri passivi forniscono valori medi settimanali-mensili che non sono coerenti con le definizioni degli indici cumulativi di esposizione della vegetazione, come l'AOT40 utilizzato in Europa ( $O_3$  Accumulated Over Threshold 40 ppb). Questo articolo descrive un metodo empirico utilizzato per stimare i valori di AOT40 a partire dalle concentrazioni misurate dai dosimetri passivi. Il metodo è basato essenzialmente sulle concentrazioni settimanali misurate e sulla funzione di Loibl che descrive il profilo giornaliero delle concentrazioni di ozono in funzione della quota relativa (la differenza tra la quota del sito di misura e quelle più bassa in un intorno di 5 km di raggio). Una volta conosciuto, il profilo giornaliero dell'ozono viene centrato sulla media settimanale delle concentrazioni misurate dai passivi, assunte come media delle 24 ore. L'assunzione che semplifica è quella di considerare il profilo giornaliero dell'ozono come costante per tutti i 7 giorni della settimana. Le stime dei valori di AOT40 sono state validate sia in relazioni a quanto misurato da analizzatori co-locati, sia in relazione ad una serie di misurazioni indipendenti sparse in tutta Italia. I risultati mostrano una buona relazione tra valori misurati e stimati ( $R^2: 0.93$ ). La differenza media assoluta tra valori stimati e misurati è risultata di 15.4% (6.03% per i soli siti rurali e remoti). Il modello tende a sottostimare per valori misurati di AOT40 inferiori a 30000 ppb $^{*}h^{-1}$  ed a soprastimare per valori superiori.

**Parole chiave:** campionatori passivi, ozono, AOT40, foreste, Italia.

Risk analysis in relation to the effects of ozone ( $O_3$ ) on forests is a complex matter (e.g. HOGSETT *et al.* 1997; EMBERSON *et al.* 2000; SAMUELSON 2001; SAMUELSON and KELLY 2001), but it always needs to know the actual exposure of vegetation to  $O_3$ . Even this basic steps (measurement of ozone concentration to calculate  $O_3$  exposure) is problematic in the forest environment due to a number of practical and therefore financial constraints. In this context, passive

samplers offer considerable advantages for measuring  $O_3$  concentrations ( $[O_3]$ ) in remote areas like forests (KRUPA *et al.* 2000) and – within the intensive forest monitoring programme in Europe – they are suggested “as the main method within the ICP-Forest programme for sites that do not currently monitor ozone using active samplers” (BFH 2000). However, given the nature of the technique, measurements with passive samplers typically result in data about weekly-to-monthly  $O_3$

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concentrations which are not consistent with the definitions of cumulative indices currently being used to estimate the exposure of vegetation to  $O_3$ . In Europe, for example, the Level I risk analysis for natural, semi-natural vegetation, forests and crops is based on the AOT40 index (MILLS *et al.* 2000; KARENLAMPI and SKARBY 1996, FUHRER *et al.* 1998). The  $O_3$  Accumulated Over Threshold 40 ppb (AOT40) is defined as the sum of the hourly  $[O_3]$  exceeding the threshold of 40 ppb over the period April, 1<sup>st</sup>-September, 30<sup>th</sup>. Only the daylight hours are considered (global radiation  $> 50 \text{ W m}^{-2}$ ) (KARENLAMPI and SKARBY 1996):

$$AOT40 = \sum ([O_3]_i - 40) \quad (1)$$

$[O_3]_i > 40 \text{ ppb}$   
 $globrad > 50 \text{ W m}^{-2}$

Although the value of the AOT concept as well as the value of the 40 ppb threshold were questioned (*e.g.*, MATYSSEK and INNES 1999), AOT40 is still the basis for estimating the potential risk of forests due to  $O_3$ , and to set environmental quality objectives within the European Union (EU) and the United Nation Economic Commission for Europe (UN ECE). According to the above definition, proper calculation of AOT40 values implies the availability and completeness of hourly  $[O_3]$  through a six month period. When only weekly mean  $[O_3]$  are available - as in the case of passive samplers - there is an obvious discrepancy between data requirements and data availability (see TUOVINEN 2002; FERRETTI and GEROSA 2002). For this reason, attempts were and are being made to develop statistical and empirical methods to estimate exposure indices like the AOT40 adopted in Europe starting from mean concentrations obtained from passive samplers (KRUPA *et al.* 2001; TUOVINEN 2002). This paper describes an empirical method adopted in Italy to estimate 1996-2000 AOT40 values at 20 Permanent Monitoring Plots (PMPs) of the Italian network of intensive monitoring of forest ecosystems (Italian acronym: CONECOFOR). Unlike the procedure described by TUOVINEN (2002), we report data from actual passive samplers measurements carried out at 20 forest sites from 1996 to 2000. The validation of the AOT40 estimates were made both against 2 co-located automatic measurement devices and 15 automatic devices whose weekly averages were used to simulate passive samplers. Data about the monitoring technique, values and reliability of weekly mean  $[O_3]$  are reported

by BUFFONI and TITA (this volume).

## Materials and methods

### Outlook

The method is based on the following data and steps (Fig. 1):

- (i) the weekly mean  $[O_3]$  data obtained from passive samplers collected at all the sites over the period 1996-2000 (see BUFFONI and TITA 2000; BUFFONI and TITA this volume) (steps 1-2, Fig. 1). Data, methods of measurement and reliability of  $[O_3]$  are reported by BUFFONI and TITA (this volume);
- (ii) the modelling of daily  $[O_3]$  profile (step 3, Fig. 1). This has been carried out on the basis of a function (hereafter referred to as the Loibl function, LOIBL and SMIDT 1996) which describes the  $O_3$  daily profile as a function of relative altitude (the difference between the altitude of the site and the lowest altitude within a 5 km radius) (see below);
- (iii) calculation of estimated AOT40 ( $AOT40_e$ ) and validation against the measured AOT40 ( $AOT40_m$ ) (steps 4-5, Fig. 1). When the theoretical daily profile for the site is known, then the hourly exceedance of 40 ppb can be computed (step 6, Fig. 1) (see below).

### Modelling $O_3$ daily profile

The modelling of  $[O_3]$  daily profile is based upon the evidence that it is strongly related to the altitude. LOIBL *et al.* (1994) and LOIBL and SMIDT (1996) reported a function describing the hourly  $[O_3]$  as a function of the relative altitude ( $h_r$ ) of the site, *i.e.* the difference between the altitude of the concerned site and the lowest altitude within a 5 km radius:

$$O_3(h_r, t) = a_1 + a_2 e^{-(t-a3)^2 a_4} \cdot \ln \left( \frac{h_r}{100} + \frac{b_1 t^2 + b_2 t + b_3}{b_4 t^2 + b_5 t + 10000} e^{-b_6 t} \right) \quad (2)$$

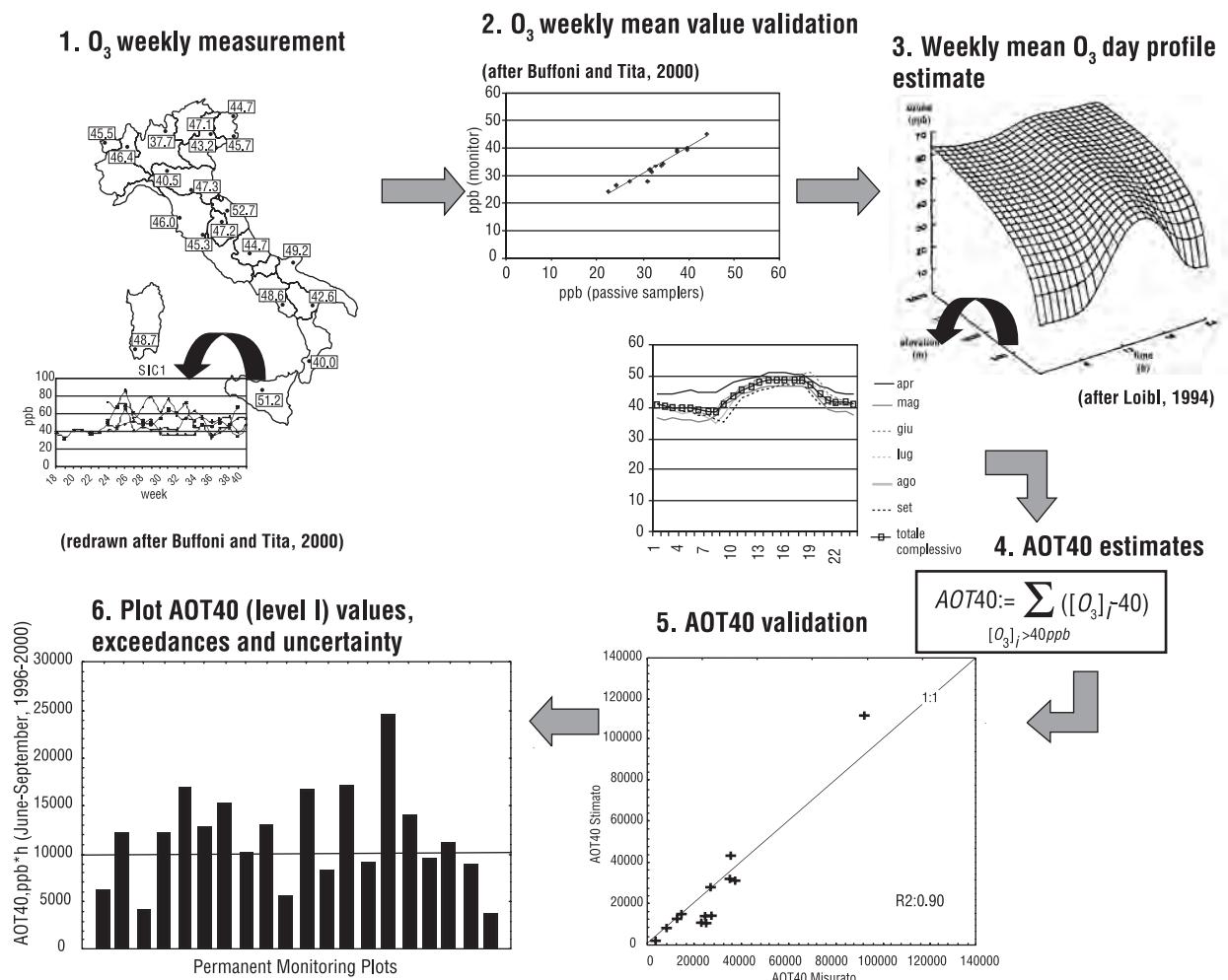
where

$h_r$  is the relative altitude in meters,

$t$  is the daytime,

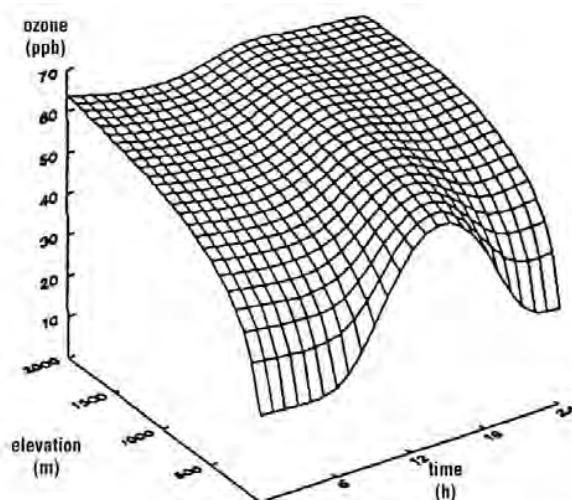
$a_1, a_2, a_3, a_4$  and  $b_1, b_2, b_3, b_4, b_5$  and  $b_6$  are coefficients obtained from the fitting.

The shape of the function is in Fig. 2. It was obtained as the best fit of a series of  $O_3$  measurements carried out at more than 100 rural sites in Austria distributed over a range of  $h_r$ . The shape of the function fits quite



**Figure 1** – The steps undertaken to estimate AOT40 values and exceedance of the  $10000 \text{ ppb-h}^{-1}$  threshold. See text for details.  
*Stadi intrapresi per la stima dei valori di AOT40 e dell'eccedenza della soglia di  $10000 \text{ ppb-h}^{-1}$ . Ulteriori dettagli nel testo.*

well also for the Italian sites (see Fig. 3). However, it should be considered that the (2) represents a mean, ideal situation which may be subjected to interferences due to e.g.  $O_3$  advection from areas with high photochemical production,  $O_3$  depletion by nearby  $NO_x$  emissions and so on. For this reason, some deviation can be expected according to the situation of individual sites. The  $[O_3]$  daily profile has been modelled for each PMP on the CONECOFOR programme according to equation (2) and using the  $h_r$  calculated for each site. Then, the  $[O_3]$  daily profile was adjusted in order to match the weekly  $[O_3]$  measured by the passive samplers, assumed as the 24 hours daily average (see example in Fig. 4). The resulting  $[O_3]$  daily profile was replicated for each day of the week. The underlying, simplifying assumption is that daily  $[O_3]$  profile is considered to be the same every day of the week.



**Figure 2** –  $O_3$  daily profile as a function of the relative altitude (after Loibl and Smidt 1994).  
*Profilo giornaliero di  $O_3$  in funzione della quota relativa (da Loibl e Smidt 1994).*

### Estimation of the AOT40 values (AOT40<sub>e</sub>)

The modelled [O<sub>3</sub>] daily profile was the basis to estimate weekly-to-seasonal AOT40 values (AOT40<sub>e</sub>) for each PMP. Only the [O<sub>3</sub>] of the hours with global solar radiation > 50 W·m<sup>-2</sup> were considered. When direct radiation measurement were not available, we assumed global solar radiation > 50 W·m<sup>-2</sup> occurring between the dawn and the sunset, and we estimated the hours to be considered by means of an astronomic model based on latitude, longitude of the site, the calendar date and the time of the day. Hourly [O<sub>3</sub>] were used in equation (1) to calculate daily-to-weekly AOT40 values. To calculate the AOT40 values over the concerned period, weekly AOT40 (AOT40<sub>w</sub>) values were computed and then summed according to equation (3)

$$AOT40 = \sum_w AOT40_w \quad (3)$$

In order to avoid underestimation of the AOT40 in case of missing data, the “raw” AOT40 calculated on the basis of the available measurements has been weighed by a coefficient given by the reciprocal of the ratio between the number of the weekly measurements available N<sub>wA</sub> and the total number of the weeks of the period N<sub>wT</sub> (eq. 4)

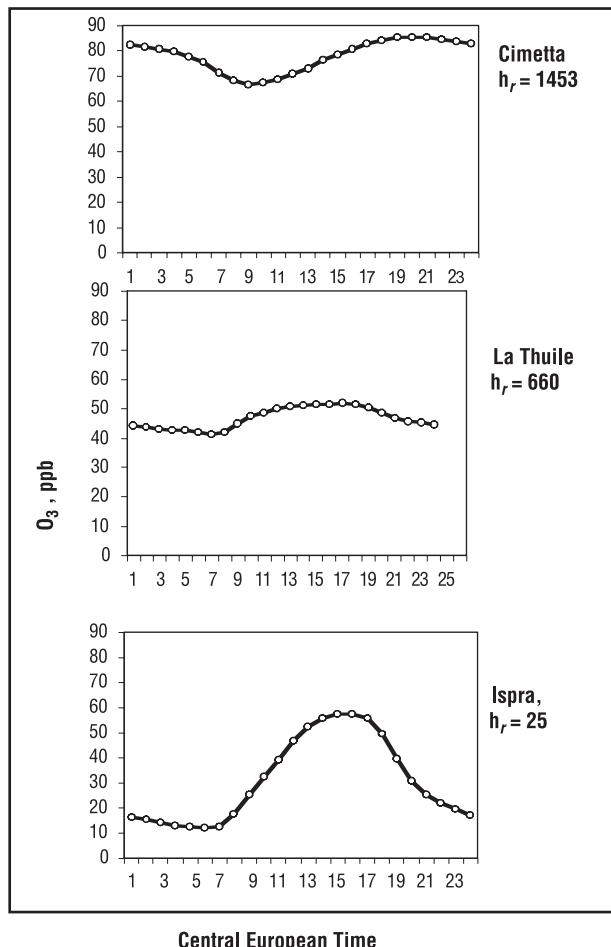
$$AOT40 = \frac{AOT40_{\text{raw}}}{N_{wA}} N_{wT} \quad (4)$$

For the purposes of this report, the AOT40 calculation period was considered between June and September. This is because the measurements with passive samplers were carried out for the period June-September, with only the year 2000 being covered from May to September. As the usual period for AOT40 calculation is April-September, this will lead to an underestimation of AOT40. To give an idea of the extent of underestimation, AOT40 values for both periods were calculated for the validation sites equipped with automatic devices.

### Validation

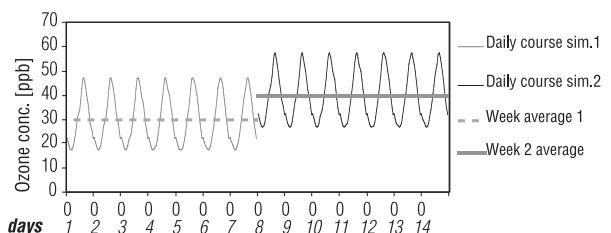
The procedure described above adopted several assumptions which were unavoidable given the nature of the [O<sub>3</sub>] data collected at the CONECOFOR PMPs. This makes necessary a validation of the estimates obtained by the model. The validation has been carried out following two approaches.

*Direct comparison with co-located automatic measurement devices*



**Figure 3 -** Mean summer daily O<sub>3</sub> daily profile at three of the validation sites located at various h<sub>r</sub>. Data refer to 1997 (Ispra and Cimetta) and 1998 (La Thuile). Dots represent the actual measurements.

*Profilo giornaliero medio estivo dell'ozono a tre dei siti di validazione situati a diverse h<sub>r</sub>. I dati si riferiscono al 1997 (Ispra e Cimetta) ed al 1998 (La Thuile). I punti rappresentano le misurazioni.*



**Figure 4 -** An example of how does the procedure work: the daily course is adjusted on a weekly basis in order to obtain a week mean value equal to the weekly ozone concentration obtained by passive samplers.

*Esempio di applicazione della procedura: il profilo giornaliero delle concentrazioni viene aggiustato su base settimanale in modo da ottenere una concentrazione media uguale a quella misurata dai campionatori passivi.*

**Table 1 –** Sites equipped with automatic devices considered for the validation of AOT40 estimates. Location (state/region, longitude/latitude, altitude, and relative altitude  $h_r$ ), data series (year), data capturing (%), AOT40 values over two different periods (April-September and June-September) and ratio between the two.

*Siti equipaggiati con analizzatori automatici considerati per la validazione delle stime di AOT40. Localizzazione (stato/regione, latitudine e longitudine, quota relativa  $h_r$ ), serie di dati (anno), completezza dei dati (%), valori di AOT40 per due periodi (Aprile-Settembre e Giugno-Settembre) e rapporto tra i valori di AOT40 sui due periodi.*

Site name	State/Region	Site type	Longitude	Latitude	Altitude (m asl)	$h_r$ (m asl)	Year	Data capturing (%)	AOT40 Measured Apr.-Sept.	AOT40 Measured Jun.-Sept.	AOT40 ratio Apr-Sept./ Jun-Sept.
Osservatorio Napoli	Campania	Suburban	14.15.16	40.51.46	145	145	2000	98.3	6506	5566	1.17
Massenzatico	Emilia-Romagna	Rural/Suburban	10.41.49	44.43.45	41	9	1999	93.3	1165	1036	1.12
Fontechiari	Lazio	Rural	13.40.48	41.40.48	375	68	1996	92.7	26137	20946	1.25
Fontechiari	Lazio	Rural	13.40.49	41.40.49	375	68	1999	78.3	33548	25547	1.31
Leonessa	Lazio	Suburban	12.57.36	42.34.12	900	150	2000	92.5	60670	45659	1.33
Pieve di Teco	Liguria	Suburban	07.54.50	44.02.56	256	89	1999	88.8	6965	5594	1.25
Bormio	Lombardia	Rural	10.22.00	46.28.00	1200	111	1998	96.0	34907	20897	1.67
Chiavenna	Lombardia	Rural/Suburban	09.23.45	46.19.12	333	113	2000	94.9	14125	10122	1.40
Cimetta	Lombardia	Rural	08.47.00	46.11.85	1650	1453	1997	84.2	84354	46068	1.83
Erba	Lombardia	Rural/Suburban	15.17.44	50.73.13	325	77	1998	95.0	37195	28489	1.31
Ispra	Lombardia	Suburban	08.37.02	47.49.00	225	25	1996	100.0	24480	17579	1.39
Ispra	Lombardia	Suburban	08.37.01	46.49.00	225	25	1997	100.0	22887	15766	1.45
Ispra	Lombardia	Suburban	08.37.00	45.49.00	225	25	1998	100.0	27123	18304	1.48
Olgiate Comasco	Lombardia	Suburban	08.58.00	45.46.00	415	154	1998	87.0	24364	17516	1.39
Boccadifalco	Sicilia	Rural/Suburban	13.18.10	38.07.10	141	131	1999	95.1	45063	28312	1.59
Brione	Ticino (CH)	Rural	14.83.05	51.13.20	480	283	1998	74.0	26559	18540	1.43
Gabbro*	Toscana	Rural	11.24.46	43.29.57	253	253	2000	77.9	42996	32200 (1)	1.34
Settignano	Toscana	Rural/Suburban	11.19.28	43.47.15	195	147	1999	92.0	28782	23352	1.23
La Thuile**	Valle d'Aosta	Rural/Remote	06.55.55	45.43.26	1660	660	1996	89.1	5227	3621	1.44
La Thuile**	Valle d'Aosta	Rural/Remote	06.55.56	46.43.26	1660	660	1997	97.2	4377	1175	3.73
La Thuile**	Valle d'Aosta	Rural/Remote	06.55.57	47.43.26	1660	660	1998	94.6	24282	10583	2.29
La Thuile**	Valle d'Aosta	Rural/Remote	06.55.58	48.43.26	1660	660	1999	99.5	20509	14118 (2)	1.45
La Thuile**	Valle d'Aosta	Rural/Remote	06.55.59	49.43.26	1660	660	2000	99.3	15886	12358 (1)	1.29

\*close to PMP TOS1

\*\*close to PMP VAL1

(1) from May, 2nd, to September, 30th

(2) from May, 4th, to September, 30th

When a co-located measurement device was present (Fig. 5; Table 1), then the measured ( $AOT40_m$ ) and the estimated ( $AOT40_e$ ) AOT40 values were compared. This was possible in a strict sense only in the case of PMP VAL1. With some caution, also the comparison between the passive sampling at PMP TOS1 and the automatic  $O_3$  analyser at Gabbro (available year: 2000) can be accepted as the two sites are at similar altitude, rural/remote condition and ca. 2 km apart.

#### Indirect comparison by simulated passive sampling

The other validation procedure is similar to the one adopted by TUOVINEN (2002) and is based on the existing  $O_3$  automatic measurement devices which were used to simulate the passive sampler data.  $O_3$  monitoring in Italy is carried out by means of automatic measurement devices managed by the Regional and Provincial Environmental Protection Agencies (ARPA/APPA, Agenzie Regionali/Provinciali Protezione Ambiente) under the umbrella of the National Thematic Centre on Atmosphere, Climate and Emissions (CTN-ACE) of

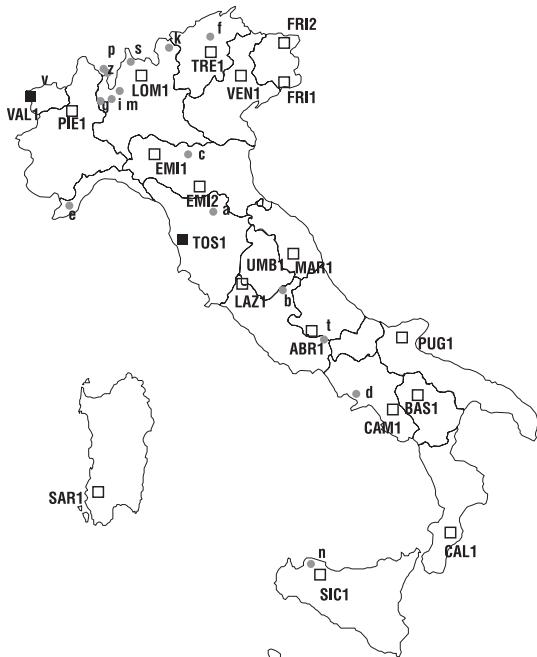
the National Environmental Protection Agency (ANPA, now APAT). DESIATO *et al.* (2000) provide an overview of the existing air pollution monitoring networks in Italy. Upon specific request, ANPA provided the available  $[O_3]$  data collected at a number of  $O_3$  monitoring devices located throughout Italy. In formulating the request, the preference was given to devices located under rural-remote condition. However, the limited availability of devices under such condition forces us to consider also suburban sites.  $[O_3]$  data were checked for completeness, and only data series with data capturing  $> 70\%$  were retained. Fig. 5 and Table 1 reports details about location, measurement year, data capturing and AOT40 values at the monitoring sites that survived the data completeness check. The  $[O_3]$  averaged over a weekly basis (24 hours average) were used to simulate the passive sampler weekly data. Then, the  $[O_3]$  daily profile was estimated according to the equation (2) and after having calculated the  $h_r$  value for each site. AOT40 values were calculated according to (1) and (3). Missing data were managed according to (4).

## Results

### Performance of the model: validation against automatic measurements

#### Direct comparison with co-located automatic devices

Direct comparison between estimates by passive samplers and measurements by automatic devices was possible only for the PMP VAL1 (years 1996, 1997, 1998, 1999, 2000) and at a lesser extent for TOS1 (year 2000) (Table 1). The 1997 data at VAL1 were rather anomalous, in particular over the period June-September with a measured AOT40 1175 ppb $\cdot$ h $^{-1}$  (see Table 1). The anomalous character of the 1997 dataset is shown by the high ratio between the AOT values measured over the period April-September and June-September. Actually, the reported O<sub>3</sub> concentrations were lower in summer than in winter and this is *per se* an indication of poor



**Figure 5 –** Location of the validation sites. Empty squares: CONECOFOR PMPs; full dots: location of the ANPA-ARPA-APPA O<sub>3</sub> automatic measurement devices; full squares: O<sub>3</sub> devices and PMPs are co-located. The letters indicate the O<sub>3</sub> sites: a Settignano, b Leonessa, c Massenzatico, d Napoli Osservatorio, e Pieve di Teco, f Renon, g Ispra, z Cimetta, i Olgiate Comasco, k Bormio, m Erba, n Boccadifalco, p Brione, q Gabbro, v La Thuile, s Chiavenna, t Fontechiari. See also table 1.  
*Localizzazione dei siti di validazione. I siti CONECOFOR sono rappresentati con quadratini vuoti, le stazioni ANPA con cerchietti pieni. Quando coincidono appare un quadrato pieno. Legenda stazioni: a Settignano, b Leonessa, c Massenzatico, d Napoli Osservatorio, e Pieve di Teco, f Renon, g Ispra, z Cimetta, i Olgiate Comasco, k Bormio, m Erba, n Boccadifalco, p Brione, q Gabbro, v La Thuile, s Chiavenna, t Fontechiari. Vedi anche Tab. 1.*

**Table 2 -** Direct comparison between AOT40 values measured by automatic devices and values estimated by processing data obtained by co-located passive samplers.

*Comparazione diretta tra AOT40 misurati da analizzatori automatici e valori stimati processando i dati ottenuti da campionatori passivi collocati nelle vicinanze.*

PMP	O <sub>3</sub> site name	year	Common measurement period	AOT <sub>m</sub>	AOT <sub>e</sub>	AOT <sub>m</sub> -AOT <sub>e</sub> ppb $\cdot$ h $^{-1}$	AOT <sub>m</sub> -AOT <sub>e</sub> (%)
TOS1	Gabbro	2000	2 May - 30 Sept.	32200	34387	-2188	6,79
VAL1	La Thuile	1996	15 June-30 Sept.	3621	1676	1945	-53,71
VAL1	La Thuile	1998	16 June-30 Sept.	7921	7758	163	-2,06
VAL1	La Thuile	1999	4 May - 30 Sept.	14118	15429	-1311	9,29
VAL1	La Thuile	2000	2 May - 30 Sept.	12358	12250	108	-0,87

data quality. Problems with the 1997 dataset were confirmed by a statistical analysis based on the probability distribution function of the AOT40 values: assuming a log-normal distribution function, the estimated probability to obtain the 1997 AOT40 value is 3.83458·10<sup>-6</sup> (if distribution is estimated considering the 1997 dataset) or 1.7049·10<sup>-11</sup> (if distribution is estimated without the 1997 dataset). For these reasons, the year 1997 was not considered in the study. Table 2 reports the data for both VAL1 (1996, 1998, 1999, 2000) and TOS1 (2000). At VAL1 high deviation occur for low AOT40 values (3621 ppb h $^{-1}$  in 1996); for the remaining years, deviations ranges from -2.06 to +9.29%. At TOS1 there is only one year available, and a deviation of 6.79% was recorded.

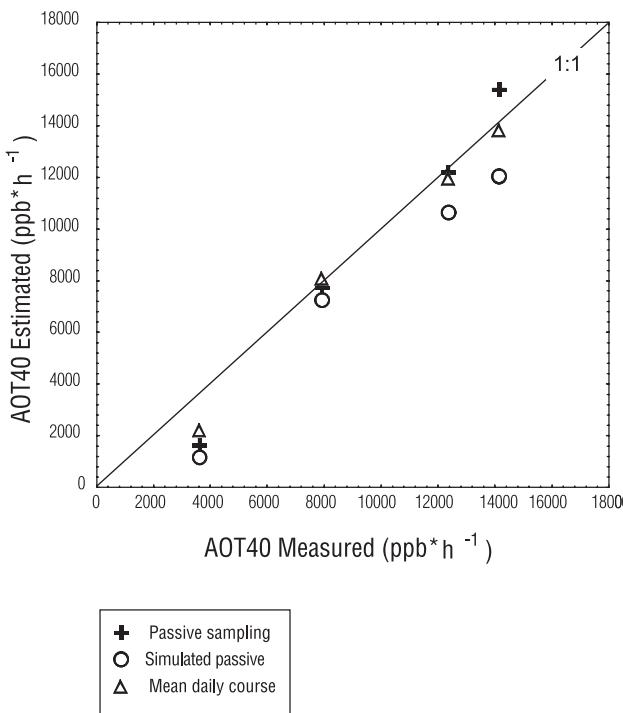
The PMP VAL1 was also used to test whether AOT40 estimates obtained from weekly [O<sub>3</sub>] averages calculated from automatic measurements can simulate the passive sampling. Measured AOT40 values and AOT40 estimated according to three different methods (passive sampling, simulated passive sampling, and 4 year average daily course) are in Fig. 6. Although simulated passive sampling seems to underestimate the measured AOT40 values as well as the AOT40 estimated by actual passive sampling, differences were not significant (Mann-Whitney U test, P>0.05).

#### Indirect comparison by simulated passive sampling

Weekly [O<sub>3</sub>] averages obtained from automatic measurements were used to simulate the passive sampling. Table 3 reports the comparison between the measured and estimated AOT40 values. The median disagreement is -15.97%, thus showing an overall underestimation of the measured AOT40. This is in line with the findings reported above and is particularly true for the suburban sites, while the performance for rural/remote sites is much better (Table 4).

#### Performance of the model: a synthesis

The statistics about the differences between mea-



**Fig. 6 –** Direct comparison between the AOT40 measured by real time analysers and the AOT40 estimated from passive sampling, simulated passive sampling and mean daily course obtained by real time analysers at VAL1. Each data point represent a year.

Comparazione diretta tra i valori di AOT40 misurati dagli analizzatori automatici e quelli stimati dai campionatori passivi, dai campionatori passivi simulati e dal giorno medio ottenuto dai dati degli analizzatori automatici per il l'area VAL1. Ogni punto rappresenta un anno.

**Table 3 -** Indirect comparison between AOT40 values measured by automatic devices and values estimated by processing data obtained by the same devices in order to simulate passive samplers.

Comparazione indiretta tra AOT40 misurati da analizzatori automatici e valori stimati processando i dati ottenuti dagli stessi analizzatori in modo da simulare i campionatori passivi.

Site name	Year	$AOT_m$ ppb·h⁻¹	$AOT_e$ ppb·h⁻¹	$AOT_m - AOT_e$ ppb·h⁻¹	$AOT_m - AOT_e$ (%)
Osservatorio Napoli	2000	5566	3564	2001	-35,96
Massenzatico	1999	1036	786	250	-24,13
Fontechiari	1996	20946	17602	3345	-15,97
Fontechiari	1999	25547	24199	1348	-5,28
Leonessa	2000	45659	52391	-6731	14,74
Pieve di Teco	1999	5594	2240	3354	-59,96
Bormio	1998	20897	21421	-524	2,51
Chiavenna	2000	10122	9455	667	-6,59
Cimetta	1998	35254	43631	-8378	23,76
Erba	1998	28489	28891	-402	1,41
Ispra	1996	17579	11614	5965	-33,93
Ispra	1997	15766	11143	4623	-29,33
Ispra	1998	18304	8095	10209	-55,77
Olgiate Comasco	1998	17516	13595	3922	-22,39
Boccadifalco	1999	28312	24167	4146	-14,64
Brione	1998	18540	18543	-3	0,02
Settignano	1999	23352	18819	4533	-19,41

**Table 4 -** Differences between measured and estimated AOT40 values according to the site type. Relative differences reports the actual sign of the differences. Absolute differences report the absolute value of the differences.

Differenze tra valori di AOT40 misurati e stimati in relazione al tipo di sito. Le differenze relative si riferiscono alle differenze considerate con il loro segno originale. Le differenze relative fanno riferimento ai valori assoluti delle differenze.

Site type	N	Indirect comparison		Direct+Indirect comparison	
		Median relative differences (%)	Median absolute differences (%)	Median relative differences (%)	Median absolute differences (%)
Rural and remote	10	0.02	5.28	-0.43	6.03
Rural/suburban	5	-14.64	14.64	-14.64	14.64
Suburban	7	-33.93	33.93	-33.93	33.93

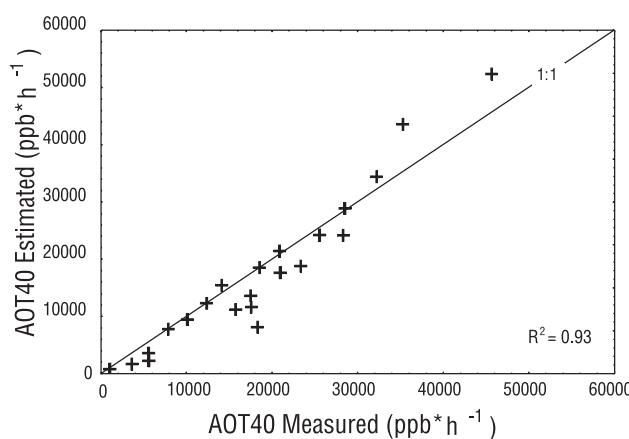
sured and estimated AOT40 values (direct + indirect comparisons) are reported in Table 4 and 5. Although the maximum disagreement between measured and estimated AOT40 may reach 60%, the median difference is 10.62%. The model seems to underestimate AOT40 values below 30000 ppb·h⁻¹, while overestimation seems to occur for higher values (Fig. 7). The model seems to perform much better under rural/remote condition: this may be due to the fact that, under suburban condition, there could be phenomena of  $O_3$  avvection from areas with high photochemical production,  $O_3$  depletion by nearby  $NO_x$  emissions and/or remixing that may generate peak events. These events may substantially affect the daily  $O_3$  course, the regularity of which is the basic assumption of the model.

#### Estimates of the AOT40 values (AOT40<sub>e</sub>) for the CONECOFOR monitoring plots

Table 6 and Fig. 8 report the June-September AOT40 values estimated (AOT40<sub>e</sub>) for the 20 CONECOFOR plots over the period 1996-2000. As recommended by the Task Force for Mapping dell'UN/ECE (UBA 1996), the 5-years average AOT40<sub>e</sub> is reported for each site in Fig. 8. Within the examined PMPs, the AOT40 values increase between 250 and 1250 m asl (Fig. 9). However, the Alpine sites (mostly located above 1200 m asl) show the lower values. High values (15000-20000 ppb·h⁻¹) usually occur at the sites in the Apennine Mountains and on the East coast. The sites on the East coasts have intermediate values (10000-15000 ppb·h⁻¹). The highest AOT40 values are in Sicily (> 25000 ppb·h⁻¹) (Figs. 8 and 10).

**Table 5 -** Descriptive statistics of the AOT40 measured and estimated and of the differences between the two. Differences are reported as relative (in both ppb $\cdot$ h $^{-1}$  and %) and absolute (in %). *Statistica descrittiva dei valori di AOT40 misurati e stimati e delle differenze tra i due. Le differenze sono riportate in termini relativi (sia in ppb $\cdot$ h $^{-1}$  che in %) e assoluti (in %).*

Parameter	N	Mean	Median	Min	Max	25°	75°
AOT40 Measured	22	18577	17942	1036	45659	10122	25547
AOT40 Estimated	22	17348	14512	786	52391	8095	24167
Difference, ppb $\cdot$ h $^{-1}$	22	1229	1007	-8378	10209	-402	3922
Difference, % (rel)	22	-14.61	-10.62	-59.96	23.76	-29.33	1.41
Difference, % (abs.)	22	19.93	15.36	0.02	59.96	5.28	29.33



**Figure 7 -** AOT40: comparison between measured (x axis) and estimated (y axis) values.  
*Confronto tra AOT40 misurato (asse x) e stimato (asse y).*

### Exceedance of the AOT40 critical level

Table 6 reports the exceedances of the mean (1996-2000) AOT40 $_e$  values as well as the accumulated exceedance over the 5 years (sum of the annual exceedances). Even if the data refers to a 4 months period (e.g. 67% of the usual computational period) in most cases the critical levels 10000 ppb (KUOPIO 1996) and 5000 ppb (GOTHENBURG 2002) are exceeded at least in one year. In some other cases, the AOT40 $_e$  is very close to the critical levels. Only in a limited number of cases (PMPs: LOM1, VEN1 and CAL1), the former critical level of 10000 ppb $\cdot$ h $^{-1}$  was never exceeded. However, considering the conservative nature of the estimate (only 4 months) and the ratio between the AOT40 calculated over the 6 and 4 months period (Table 1), it is very likely that exceedance may occur at a larger number of sites.

Exceedance may reach up to 4.5 times the former critical level of 10000 ppb $\cdot$ h $^{-1}$ , as in Sicily (PMP SIC1). When the data relevant to May are available (e.g., year 2000), it is shown that exceedances may occur as early as July. As an example, Fig. 11 reports the accumulation curves of the year 2000. The critical level is exceeded by the first week of July at several sites (namely BAS1, EMI1, FRI1, MAR1, SIC1 TOS1).

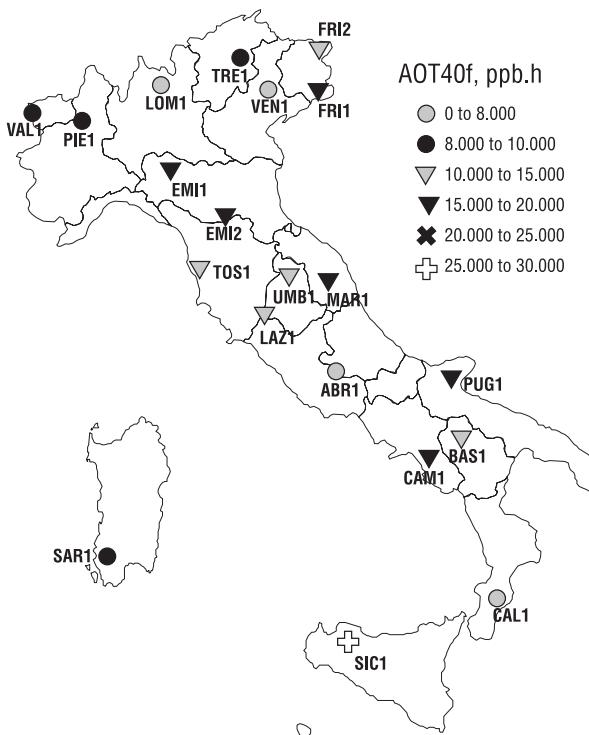
### Conclusions

Weekly [O<sub>3</sub>] data collected by passive samplers were used as input data to estimate the AOT40 exposure index. As AOT40 is based on the sum of the hourly values

**Table 6 -** AOT40 estimated for the PMPs of the CONECOFOR programme over the period 1996-2000. The mean 1996-2000 value, the exceedance of the mean value and the accumulated exceedance (the sum of the annual exceedances) over the 1996-2000 period are reported.  
*Valori di AOT40 stimati per i PMP del programma CONECOFOR per il periodo 1996-2000. Sono riportati anche il valore medio 1996-2000, l'eccedenza del valore medio e l'eccedenza cumulata (somma delle eccedenze annuali) per il periodo 1996-2000.*

PMP	AOT40 1996	AOT40 1997	AOT40 1998	AOT40 1999	AOT40 2000	Mean AOT40 1996-2000	Exceedance of the mean AOT40 10000 ppb $^{-1}$	Accumulated exceedance 1996-2000
ABR1	2022	6960	5341	10017	8392	6546	0	17
BAS1	1961	30438	3374	9911	26433	14423	4423	36871
CAL1	565	7683	208	4137	9354	4389	0	0
CAM1	5920	36927	4095	19960	19553	17291	7291	46439
EMI1	7214	26799	13935	15025	26462	17887	7887	42221
EMI2	4764	29708	12876	18694	10878	15384	5384	32157
FRI1	6996	13121	17572	25127	17831	16129	6129	33651
FRI2	3941	12280	6236	17951	12440	10570	570	12671
LAZ1	4339	22786	12638	15792	13592	13829	3829	24807
LOM1	NM	6462	3203	6205	7086	5739	0	0
MAR1	5237	27973	7058	29239	19232	17748	7748	46444
PIE1	3153	10442	2085	16744	11003	8686	0	8190
PUG1	4933	31430	16154	20458	18632	18322	8322	46675
SAR1	3101	9633	1918	17061	14293	9201	0	11354
SIC1	9492	45578	14333	27858	32196	25891	15891	79965
TOS1	6048	14964	11781	23458	16908	14632	4632	27111
TRE1	1441	16745	6305	15536	9654	9936	0	12281
UMB1	2177	16938	2765	20383	15873	11627	1627	23194
VAL1	1780	11480	8793	15108	10073	9447	0	6661
VEN1	431	4013	2648	8675	4253	4004	0	0

NM: not measured

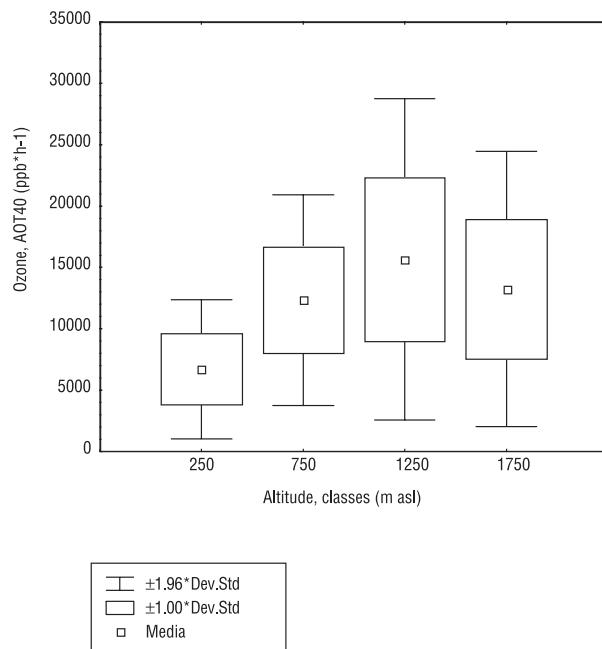


**Figure 8 –** Estimated 1996-2000 AOT40 values for the CONECOFOR plots considered in the study.

*Valori medi stimati di AOT40 per il periodo 1996 alle aree CONECOFOR considerate nello studio.*

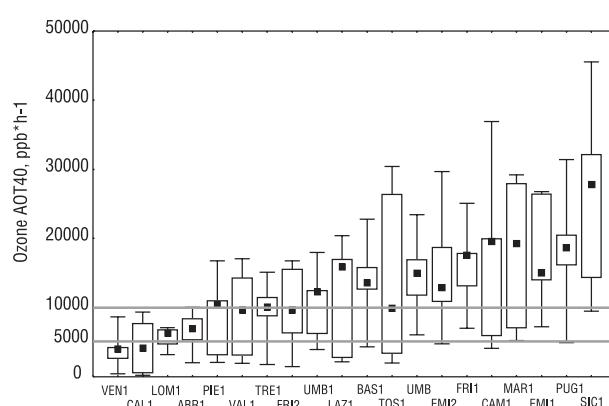
that exceeds 40 ppb during the daylight hours,  $[O_3]$  daily profile needs to be modelled. Modelling was carried out according to the Loibl equation which describe the  $[O_3]$  daily profile as a function of the geographical position, the calendar date and the time of the day. The weekly mean of  $[O_3]$  estimated by passive sampling was used to adjust the daily profile in order to match the  $[O_3]$  recorded at any given site. Median deviations between measured and estimated AOT40 values was 15.36 %: however, considerable differences can occur and this suggest that a lot of caution is needed when interpreting the data. The model seem to underestimate low to medium (up to 30000 ppb·h<sup>-1</sup>) AOT40 values and this, coupled with a reduced computational period resulted into a conservative estimate of the exposure of the concerned PMPs to  $O_3$ . Yet, compared to the low costs of the passive sampler technique, the results are interesting and demonstrated the possibility to collect basic data about the forest exposure to pollutants at relatively low costs.

Bearing in mind the limitations described above, the estimated AOT40 values suggest that most of the PMPs of the CONECOFOR program are exposed to  $O_3$  that exceeds the critical limits of 10000 and 5000 ppb·h<sup>-1</sup>



**Figure 9 –** Mean AOT40 values according to 4 altitude classes of the CONECOFOR plots: 0-500 m (labelled 250); >500-1000 (labelled 750); >1000-1500 m (labelled 1250) and >1500-2000 (labelled 1750).

*Valori medi di AOT40 in funzione di 4 classi di quota delle aree CONECOFOR: 0-500 m (indicata 250); >500-1000 (indicata 750); >1000-1500 m (indicata 1250) and >1500-2000 (indicata 1750).*



**Figure 10 –** Distribution of yearly values (min, max, median, 25th and 75th percentile) of AOT40 values over the period 1996-2000 for the CONECOFOR.

*Distribuzione dei valori annuali di AOT40 per il periodo 1996-2000 alle aree CONECOFOR riportati come valori annuali minimo, mediano, massimo ed intermedi.*

set to protect forest vegetation. This is particularly true when considering the conservative nature of the AOT40 estimates. Although there is a considerable variability between the sites, in some cases the critical level is exceeded already at the beginning of July. While the values of AOT40 5000 or 10000 ppb in a Risk Analysis can be questioned from several point of view, this paper confirm that high exposure levels may occur in South European forests, thus suggesting that  $O_3$ -related adverse effect may occur.

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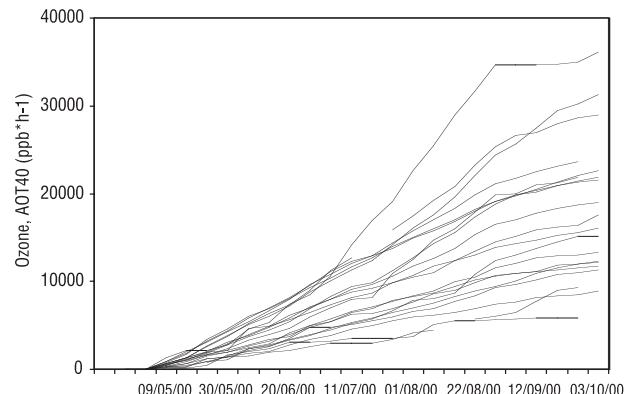
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**Figure 11 – Accumulation of AOT40 values for the 20 PMPs over the period May, 2nd-October, 3rd, 2000.**  
*Accumulo dei valori di AOT40 nel periodo 2 maggio-3 ottobre 2000.*

# Factors influencing vulnerability and response to ozone of the vegetation at the permanent monitoring plots of the CONECOFOR programme in Italy<sup>§</sup>

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**Abstract** – A number of factors control the potential vulnerability of forest vegetation to ozone ( $O_3$ ). They include inherent vulnerability due to species composition, a suite of structural attributes (tree density, canopy height, leaf area index (LAI), foliage displacement within the canopy), and physical factors like soil moisture and vapour pressure deficit. Additional factors (nutritional status, deposition of other pollutants) may counteract or exacerbate the effects of  $O_3$ . This paper reports some of the factors that can be important in determining  $O_3$  vulnerability of the permanent monitoring plots (PMPs) of the CONECOFOR programme. Some PMPs have a rather high frequency of  $O_3$ -sensitive species (>30%). Forest structure varies very much between PMPs. Canopy layers with characteristics that favour gas exchange (tall trees, high LAI, low tree density and crowns located in the very upper portion of the trees) occur mostly in beech high forests located in various parts of Italy. Relative evapotranspiration ( $RE_e$ ) and VPD data reveal conditions more favourable to  $O_3$  uptake in PMPs located in northern Italy. No particular nutritional problem was detected by soil and foliar analysis, although N deposition was reported to be high. When considered together, compositional, structural, meteorological and  $O_3$  exposure data suggest that PMPs located in high forests in Northern Italy may be at potential higher risk for adverse effects than sites in central and southern Italy.

**Key words:** forest structure, Italy, N deposition, nutrients, ozone, relative transpiration, species sensitivity, VPD.

**Riassunto** – Fattori che influenzano la vulnerabilità e la risposta all'ozono della vegetazione nelle aree permanenti del programma CONECOFOR. Molti fattori controllano la vulnerabilità di un sito forestale all'ozono ( $O_3$ ). Essi includono la vulnerabilità innata alla sensibilità delle specie che compongono la cenosi, un insieme di attributi strutturali (densità, altezza delle chiome, localizzazione del fogliame nello strato delle chiome, leaf area index - LAI) e fattori fisici come umidità del suolo e deficit di pressione di vapore (VPD). Altri fattori (stato nutrizionale, deposizione di altri inquinanti) possono contrastare o accenetrare gli effetti dell' $O_3$ . In questo articolo vengono esposti i dati relativi ad alcuni fattori importanti al momento di valutare la vulnerabilità di un determinato sito all' $O_3$ . In alcuni siti la frequenza di specie sensibili sul totale delle specie presenti è risultata superiore al 30%. La struttura dei boschi è risultata variare molto tra le aree permanenti. Coperture con caratteristiche in grado di favorire gli scambi gassosi (alberi alti, alti valori di LAI, basse densità del bosco, chiome localizzate molto in alto sul tronco) sono più frequenti nelle aree in fustae di faggio, dal nord al sud Italia. I dati di evapotraspirazione relativa ( $RE_e$ ) e VPD indicano condizioni più favorevoli all'assorbimento di  $O_3$  nel Nord Italia. Nessun particolare problema nutrizionale è stato rilevato dalle analisi di suolo e foglie, con l'eccezione di elevate deposizioni di N. Valutati nel loro complesso i dati indicano che, sebbene l'esposizione ad  $O_3$  sia a volte più bassa che altrove, le aree permanenti localizzate nelle fustae del nord Italia sembrano soggette ad un maggiore rischio potenziale di effetti avversi dovuti all' $O_3$  rispetto alle aree del centro e sud Italia.

**Parole chiave:** deposizione di N, sensibilità specifica, Italia, nutrienti, ozono, struttura forestale, traspirazione relativa, VPD.

Exposure to ozone ( $O_3$ ) in the CONECOFOR Permanent Monitoring Plots (PMPs) was reported by BUFFONI and TITA (2003) and GEROSA *et al.* (2003) as mean concentration and estimated AOT40 (Figure 1). The AOT40 values ( $O_3$  Accumulated Over Threshold 40 ppb) is defined as the sum of hourly [ $O_3$ ] exceeding the threshold of 40 ppb over the period 1 April–30 September; only daylight hours are considered (KARENLAMPI and SKARBY 1996). Data reported by GEROSA *et al.* (2003) identified a number of sites where the former

and the newly suggested critical levels (10 and 5 ppm  $h^{-1}$ , respectively, KARLSSON *et al.* 2003b) are exceeded on an annual and 5-years basis, thus suggesting potential risk to vegetation. However, there are several factors that can influence the vulnerability and response of forests to  $O_3$  (REICH 1987). Species composition, species diversity, genetic variability, age, canopy structure, nutrient supply, deposition of other pollutants, meteorological characteristics (*e.g.*, wind velocity, vapour pressour deficit-VPD), soil moisture and drought stress all have a role

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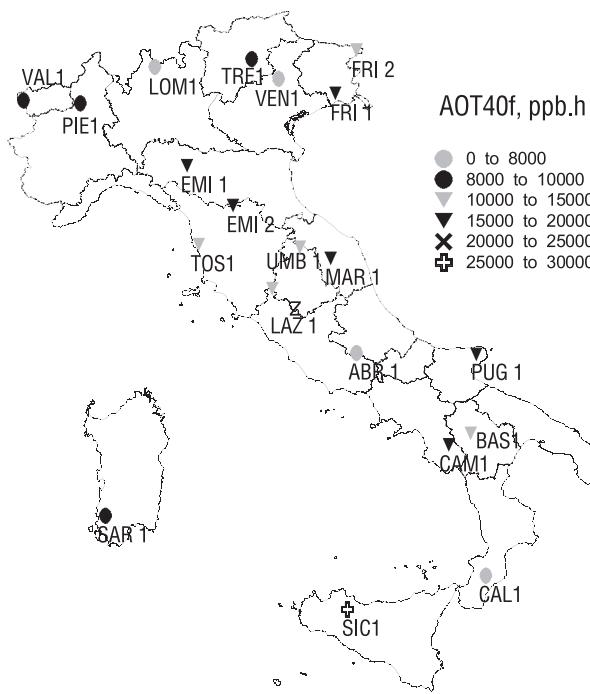
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**Figure 1** – Estimated 1996-2000 AOT40 values for the CONECOFOR plots considered in the study (after Gerosa *et al.*, 2003). Data are calculated as mean of 5 June-September AOT40 values (1 for each year 1996-2000).

*Valori medi stimati di AOT40 per il periodo 1996 alle aree CONECOFOR considerate nello studio (da Gerosa *et al.*, 2003). I dati sono stati calcolati come media dei 5 valori di AOT40 riferiti al periodo Giugno-Settembre di ciascun anno.*

in determining deposition and uptake of  $O_3$ , sensitivity of a given site to the effects of  $O_3$  and the response of vegetation (CHAPPELKA and FREER-SMITH 1995; CHAPPELKA and SAMUELSON 1998; KOLB *et al.* 1997; PALUDAN-MÜLLER *et al.* 1999; WIESER and HAVRANEK 1995; WIESER *et al.* 2002). However, not all potential factors were investigated at the PMPs of the CONECOFOR programme; for example, species genetic diversity, soil moisture deficit and stomatal conductance. Yet, a look at available data for possible influencing factors is useful in order to identify sites where high  $O_3$  exposure is accompanied by conditions conducive to  $O_3$  effects on vegetation. The aim of this paper is therefore to provide an overview of factors that may have a role in determining the sensitivity of vegetation to  $O_3$ . This also makes it possible to present data that were used as explanatory variables for the subsequent studies on the effects of  $O_3$  on crown transparency and growth (FERRETTI *et al.* 2003 this volume). The following sections of this paper provide data and information on:

(i) plant species composition at the various PMPs and their expected sensitivity to  $O_3$ . There is evidence that community structure may be impacted by oxidants (*e.g.*, DAVIDSON and BARNES 1998) and shifts in community structure due to  $O_3$  have been reported (*e.g.* BARBO *et al.* 1998 and references therein). Potential effects of  $O_3$  on vegetation at a given site should consider potential loss in diversity due to species- genotypes selection induced by different  $O_3$  sensitivity (*e.g.* ARMENTANO and BENNET 1992) and the reduction of biomass production (*i.e.*, the response indicator used in dose-response studies aimed at defining critical  $O_3$  levels, KARLSSON *et al.* 2003a). For example, a site with a high number of  $O_3$  sensitive species, each with low cover values, is likely to be impacted in terms of biodiversity. On the other hand, a site with just one species sensitive to  $O_3$ , but which constitutes all of the dominant storey, is likely to be impacted more in terms of growth reduction. Both effects are important and worthy of consideration.

(ii) The structural characteristics of the canopy layer in PMPs. Height, density, LAI and displacement of foliage in the canopy layer are important determinants of the canopy roughness (SHAW and PEREIRA 1982), which partly drives gas exchange and therefore  $O_3$  uptake. In addition, the response of trees to  $O_3$  depends very much on avoidance, defense, compensation and repair characteristics, which are linked to water relationships and therefore to age and size of trees.

(iii) Nutrient availability of soils and trees. Nutrition can have a role in determining plant response to  $O_3$ . DAVIDSON and BARNES (1998) reported mutual influences (mineral nutrients alter the response to  $O_3$ , and  $O_3$  alters the mineral content of the leaves). They also reported that the effects of  $O_3$  was greater in nutrient-deficient plants. There are controversial evidence about the role of nutrition on the occurrence of the effects of  $O_3$  on trees. For example, GUNTHARDT-GOERG *et al.* (1996) reported that leaf injury and leaf loss, as well as effects on specific leaf weight and bark radius of *Populus x euramericana* (Dode) Guinier cuttings were not modified by the soil N regime. BIELENBERG *et al.* (2001) reported increasing rates of  $O_3$ -induced senescence with decreasing N availability in *Populus trichocarpa* A. Henry x *maximowizii* Torr. and Grey. MANNINEN *et al.* (2002) did not find any effects of  $O_3$  on terpenoid concentrations in the wood of Scots pine (*Pinus sylvestris* L.) and Norway spruce (*Picea abies* L. Karst.) at low, optimum and high N supply. UTRIANEN and HOLOPAINEN (2002) reported enhanced effects on

photosynthetic activity of Scots pine needles when acute exposure to O<sub>3</sub> was combined with low P concentration in needles (<1 mg g<sup>-1</sup>).

(iv) Deposition of acidifying compounds and nitrogen (N). N deposition has clear connections with the nutrient balance of the ecosystems and has been reported as a possible cause of increased growth of forests in Europe (SPECKER *et al.* 1996) and its possible role as a factor moderating the potential adverse effect of O<sub>3</sub> was reported (*e.g.* TAKEMOTO *et al.* 2001).

(v) Drought stress and high VPD at the various PMPs: since soil moisture deficit (SMD) had not been measured at CONECOFOR PMPs, drought stress indices were considered in terms of proxy indicators, to estimate where constraints in water supply may occur and where  $O_3$  uptake may be constrained (e.g. LEFOHN *et al.* 1997). On the other hand, VPD is acknowledged to be an important factors regulating  $O_3$  uptake (e.g. EMBERSON *et al.* 2000), especially under Mediterranean climate conditions.

## Materials and methods

### *Data collection*

Table 1 reports the investigations relevant to the various factors discussed in this paper. Hereafter brief information is given on data collection methods, while details are available in ALLAVENA *et al.* (1999), FERRETTI (2000), MOSELLA *et al.* (2002).

### *Ground vegetation*

The reference data used for the investigation were collected according to the Manual compiled by the Expert Group on Ground Vegetation Assessment (DUPOUËY 1998). Namely, specific cover of tree, shrub and herb layers assessed on 12 10x10 m sampling units (SU) inside the fenced PMPs, and on 12 SU in the surrounding area, were used (see CANULLO *et al.* 1999a, b; CAMPETELLA and CANULLO 2000). The vegetation data (which excludes the PMP coded SIC1) collected in summer 1999 in all the PMPs of the CONECOFOR network were used for most of the processing; the 2000 and 2001 data were collected in a subset of the PMPs and consequently only used to verify the stability of the basic information.

Forest and canopy structure

The basic growth variables were surveyed in winter 1996-1997: diameter at breast height (DBH) of all trees from a DBH threshold of 3 and 5 cm in coppices

**Table 1 –** Availability of data for each investigation and plot combination.

0= no; 1=yes

*Disponibilità di dati per ciascuna combinazione indagine/plot.*

0=no; 1=sig

and high forests, respectively, and height of 50 to 60 randomly distributed trees over the full DBH range. Tree species were determined and individual social position was estimated according to Kraft (AMORINI and FABBIO 1997, 2000). Stand age assessment was done on the basis of tree cores taken in the buffer zone of PMPs. The same trees were used for canopy depth assessment. Basal area, mean and dominant DBH were calculated; mean and top heights were estimated by plot-specific allometric functions (FABBIO and AMORINI 2000). Leaf Area Index (LAI) and Diffuse-Non-Interceptance (DIFN, an estimate of canopy gaps) were also measured in most plots (CUTINI 2000).

#### *Soil and foliar nutrients*

Soil samples were collected in 1995. Sampling was carried out in 5 replicates in the organic horizon and in 4 layers of the mineral horizon (0-10 cm; 10-20 cm; 20-40 cm; 40-80 cm). The 5 replicates were mixed together to obtain a combined sample. Sampling, classification and determinations were carried out according to the ICP-Forest Manual, 1994 edition. Details on analytical techniques are in ALIANIELLO *et al.* (2000, 2002).

Leaves and needles were collected from upper third part of the crown of five trees per PMP in the years 1995, 1997, 1999. Samples were collected, shipped and analysed separately for each tree. For details on analytical techniques, see MATTEUCCI *et al.* (2000, 2002).

#### *Nitrogen deposition*

The chemistry of atmospheric deposition was measured in 15 of the 20 PMPs in the network. Sampling was performed weekly from January 1998 to December 2000 in open field (OF) and in the plot (throughfall and stemflow); for details, see MOSELLO *et al.* (2002). Chemical analyses included all major anions and cations, pH, conductivity and total nitrogen.

#### *Meteorological variables*

Only data from OF stations near PMPs (generally at no more than 2 km) were considered. These stations were in line with the World Meteorological Organisation standards (W.M.O. 1969). Soil evaporation and plant transpiration, necessary to calculate water balance, were not measured at any plot, but estimated by simulation models. Due to problems with missing data, drought stress indices could not be calculated for all 15 PMPs considered, and only climatic data of 10 PMPs were considered (see AMORIELLO *et al.* 2003 this volume).

#### *Data completeness, quality and reliability*

Methods to evaluate completeness, quality and reliability of data are described by FERRETTI and NIBBI (2000). Table 2 reports data quality parameters for deposition, foliar nutrients, ground vegetation and growth. Meteorological data are not reported as they are presented by AMORIELLO *et al.* (2003 this volume). Data completeness was generally high. One plot (SIC1) was not covered by the ground vegetation survey. The percentage of deposition samples within ionic balance classes 1 and 2 and calculated conductivity vs. measured conductivity in classes 1-2 was over 90% on the average (MOSELLO *et al.* 2002). The quality of foliar analysis was tested taking into account the various ring-tests carried out at European level over the period 1995-1999. Problems may occur for all the elements examined, and are particularly frequent with P (Table 2).

Minimum, maximum and mean relative errors on mean value estimates (expressed as % of the mean values at 95% confidence level) are reported in Table 3 for throughfall volume (used as an indicator of reliability of deposition estimates), foliar analysis, vegetation (number of species and cover) and growth (DBH). Available data on soil and stand structure were reported by FERRETTI and NIBBI (2000). The mean relative error was always lower than 20%. In particular, estimates of throughfall volume had a relative error of 8.7%; mean concentrations of N, P, Ca, Mg and K have mean relative errors ranging from 6 (N) to 16 % (K) and may reach values up to 47%; estimates of number of species and species cover had a mean relative error between 10

**Table 2 –** Data quality parameters for deposition (% of samples in ionic balance and conductivity classes 1-2), foliar nutrients (data completeness and % of samples showing problems with some elements), vegetation (data completeness) and growth (data completeness).

*Completezza dei dati per deposizioni (% di campioni nelle classi di bilancio ionico e conducibilità 1-2), nutrienti fogliari (completezza dei dati e % di campioni con problemi per i vari elementi), vegetazione (completezza dei dati) ed accrescimenti (completezza dei dati).*

Investigation	Indicator	Period	% of samples		
			Min	Mean	Max
Deposition	Ionic balance 1-2	1998-2000	72	92	100
	K20 meas. Vs calc. 1-2	1998-2000	89	99	100
Foliar analysis	Data completeness	1995-1999	80	90	95
	N problems	1995-1999	0	29	84
	P problems	1995-1999	0	58	84
	Ca problems	1995-1999	0	32	95
	Mg problems	1995-1999	0	30	90
	K problems	1995-1999	0	29	81
Vegetation	Data completeness	1999	0	95	100
	Data completeness	1997	100	100	100

**Table 3 –** Relative error (in % of the mean value) for each indicator of deposition, foliar analysis, vegetation and growth.  
*Errore relativo (in % della media) per i vari indicatori di deposizione, nutrienti fogliari, vegetazione, accrescimenti.*

Investigation	Indicator	Period	Relative error %		
			Min	Mean	Max
Deposition	Volume	1998-2000	2.30	8.69	33.80
	N	1995-1999	2.89	6.12	11.31
	P	1995-1999	2.89	12.34	26.00
	Ca	1995-1999	3.14	12.92	39.40
	Mg	1995-1999	6.41	16.07	47.54
	K	1995-1999	5.36	11.49	25.78
Vegetation	N of species	1999	6.19	11.52	18.65
	Cover	1999	1.64	10.09	19.88
Growth	DBH	1997	2.60	5.50	10.70

and 12 %; DBH measurements have a relative error of 6.3%.

### Data processing

#### Ground vegetation

Presence and frequency of  $O_3$  sensitive species (OSS) (see annex A; ICP-Forests, 2001) were evaluated. Species were considered in three layers: trees, shrubs and herbs. The limits of each layer were not fixed, but depended on the physiognomy of the stand. The herb layer included herbs and ligneous species under the upper limit of the taller herbaceous species. For each PMP and layer, mean OSS frequency in the SU, relative OSS participation in species composition, and OSS cover were calculated. In theory, these descriptors can be used to define the spatial extent of possible impact of  $O_3$  in the PMP (diffuse or localized), and the potential effect of  $O_3$  on species richness and on plant cover, respectively. The above descriptors were combined into a tentative index for each PMP. The index can be regarded as an estimate of the potential vulnerability of the 2500  $m^2$  PMP to  $O_3$  injury compared to a potential maximum, *i.e.*, the vulnerability of a phytocoenosis with all three vegetation layers consisting only by OSS and with full cover. Vulnerability is defined as a combination of adaptability and exposure (KLEIN 2000). Here, adaptability to  $O_3$  can be regarded as depending on non-sensitive plant species. An index of  $O_3$  vulnerability (*OV*) was calculated. This index is based on the “weight” of OSS in the tree (*t*), shrub (*s*) and herb (*h*) layers, by means of a simple combination:

$$OV = (sp.o_t \cdot cov.o_t \cdot fr.o_t) + (sp.o_s \cdot cov.o_s \cdot fr.o_s) + (sp.o_h \cdot cov.o_h \cdot fr.o_h)$$

where:

*sp.o* is the relative importance of OSS in the entire sample (number of OSS/total number of species),  
*cov.o* is OSS relative cover ( $\sum$  OSS cover/ $\sum$  species co-

ver) and *fr.o* is OSS relative frequency in the sample quadrats (OSS occurrences/total species occurrences).

The tree and shrub layers exclude climbing species as *Hedera helix*, *Smilax aspera*, *Clematis vitalba* and *Tamus communis*. Species nomenclature follows PIGNATTI (1982).

#### Forest and canopy structure

The following dimensional and structural attributes were considered: age, tree density, number of tree species in the PMP, number of tree species in the dominant storey, canopy cover, canopy depth, DIFN, vertical structure, total basal area, basal area of the dominant storey, tree height, top height, LAI. Differences between mean to top height (which provides an indication about the canopy layer at the interface with the atmosphere), as well as the Leaf Area Density (LAD) (*i.e.* the ratio of LAI to canopy depth) were calculated.

#### Soil and foliar nutrients

Values for the composite soil sample are presented for each soil attribute and PMP. For leaf/needle, mean 1995-1999 values are reported for each element\*plot combination in order to find out the overall nutritional status of the plot.

#### N Deposition

Deposition data per each PMP, chemical species ( $NO_3$ ,  $NH_4$ , organic N and total N), and flux (openfield and in the plot) were calculated. Data were reported for two different time windows (entire year and April-September) as three year average and for each individual year.

#### Meteorological data

Data were processed to calculate indices of water stress and VPD which may substantially influence the uptake of  $O_3$ . Water enters the forest by precipitation; some enters the root zone of the soil and some evaporates into the air. Precipitation (Pr) as rainfall measured at 2 m height was considered. The potential loss of water from the root zone to the atmosphere occur by evaporation from the soil surface ( $E_s$ ) and by transpiration of the plant ( $E_t$ ). The total evapotranspiration E is the sum of these two terms:

$$E = E_s + E_t$$

The amount of moisture in the root zone may vary from field capacity (the maximum amount of moisture

that can be held in soil against the force of gravity) to wilting point (the value below which moisture cannot be extracted from the soil by the roots of plants).

Soil moisture W can be written (MINTZ *et al.* 1993):

$$W_f = W_i + Pr - E$$

where  $W_f$  (mm) is root-zone moisture at the end of the week and  $W_i$  (mm) is root-zone moisture at the end of the preceding week. E (mm) is actual weekly evapotranspiration. Unfortunately, no actual measurement of transpiration were made, and E can only be estimated from the empirical relationship:

$$E = \beta_{TS} \cdot E^*$$

where:

- $E^*$  is weekly potential evapotranspiration, calculated with Thornthwaite formula (THORNTHWAITE 1948):

$$E^* = 3.733 \cdot (10 \cdot T_w / I)^a F$$

where I is annual heat index:

$$I = \sum_w (T_w / 5)^{1.514}$$

with  $T_w$  is mean weekly temperature ( $^{\circ}$ C);

$$a = 675 \cdot 10^{-9} \cdot I^3 - 771 \cdot 10^{-7} \cdot I^2 + 1792 \cdot 10^{-5} \cdot I + 0.49239$$

F is the Thornthwaite correction factor depending on month and latitude.

•  $\beta_{TS}$  ( $0 < \beta_{TS} < 1$ ) is the coefficient of transpiration plus soil evaporation and is a function of the soil wetness ratio (root moisture W to the root-zone storage capacity,  $W^*$ ):

$$\beta_{TS} = 1 - \exp(-6.8 \cdot W / W^*)$$

When  $W < W^*$ , root-zone soil moisture is below field capacity and  $P-E$  is the change in root-zone moisture. When  $W_i + Pr - E > W^*$ , final root zone moisture  $W_f$  is equal to  $W^*$  and  $W_i + Pr - E - W^* > 0$  is the surplus. If moisture storage capacity is small, on the cessation of precipitation, evapotranspiration quickly drops to a small fraction of its potential rate. If  $W^*$  is large, evapotranspiration remains close to its potential rate for longer after precipitation has stopped. Larger root-zone

storage capacity means larger annual evapotranspiration and smaller annual surplus and drainage. Three indices were calculated:

*First index:* If root-zone storage capacity  $W^*$  is available, relative transpiration ( $E/E^*$ ), calculated as ratio of actual to potential transpiration, may be a suitable index of drought stress. This index consists gives good representation of soil water availability.

*Second index:* When it is not possible to estimate soil water balance, the difference between precipitation and potential evapotranspiration can be used as a simple indicator of drought stress. As problems of drought stress are most likely to appear in summer, the  $P-E^*$  index can be applied on a monthly base. If precipitation is less than potential evapotranspiration, reduced transpiration can be assumed, as not all the amount of water needed is available to the plants.

*Third index:* the relative evapotranspiration index,  $RE_T$  (AMORIELLO *et al.* 2000):

$$RE_T = 100 \cdot \sum_w E_w / \sum_w E_w^*$$

where  $E_w$  and  $E_w^*$  are actual and potential weekly evapotranspiration.  $E_w$  has been defined by the following approximation:

$$E_w = \begin{cases} E_w^* & \text{if } E_w^* < P_w \\ P_w & \text{if } E_w^* > P_w \end{cases}$$

where  $P_w$  is weekly precipitation. This index is a simple indicator of drought stress, but does not consider soil water reserve is. It represents maximum possible water deficit.

Vapour pressure deficit (VPD) is calculated as the difference between saturated vapour pressure ( $e_a$ ) and actual vapour pressure ( $e_d$ ) at the prevailing air temperature. When relative humidity RH is known, VPD can be obtained from the following equation (BURMAN and POCHOP 1994):

$$VPD = e_a - e_d = e_a \cdot (1 - RH/100)$$

Saturated vapour pressure (in kPa) is derived from the equation (FAO 1976):

$$e_a = 0.61121 \cdot (1.00072 + 3.2 \cdot 10^{-5} P + 5.9 \cdot 10^{-9} PT) \cdot \exp \left[ \frac{18.729 - \frac{T}{227.3}}{T + 257.87} \right]$$

where  $P$  is pressure in kPa and  $T$  is air temperature in °C.

Pressure values of all PMPs were obtained by linear regression ( $R^2 = 0.99$ ) based on existing data available for each 100 m of altitude  $h$ :

$$P = 5 \cdot 10^{-6} \cdot h^2 - 0.01197 \cdot h + 101.35$$

An error of 10 % in pressure values leads to an error < 0.03 % in saturated vapour pressure.

## Results and discussion

### Species sensitivity to $O_3$

The number of species found in the PMPs of the network varied (Table 4), with a minimum (on the average) occurring in beech forests and a maximum (on the average) in oak and mixed deciduous stands. In general, the number of OSS increased with the total number of species in the PMP ( $r=0.68$ ,  $P<0.05$ ), a pattern that partly reflects the differences between stand types. Using the stand typology reported by CAMPETELLA and CANULLO (2000), the mean percentage of OSS was higher in PMPs with mixed deciduous or holm-oak (Table 4). The number of OSS found in the PMPs in 1999 and its allocation in trees, shrubs and perennial herbs is shown in Figure 2. *Fagus sylvatica* was the most frequent OSS in all layers (see Appendix B). In the shrub layer, there was the same contingent plus a few heliophilous species; the herb layer was dominated by young woody OSS: perennial herbs are scarcely repre-

sented, with the exception *Mycelis muralis*. Dominant tree species obviously had the higher cover values. A comparison with the summer data collected in 2000 and 2001 confirmed that the number of OSS and their structural distribution remain stable.

A preliminary estimate of the potential maximum vulnerability of the phytocoenosis to  $O_3$  injury, as the  $O_3$  vulnerability index ( $ovi$ ), is reported in Table 5, where PMPs are ranked according to the  $ovi$ . It should be noted that the values are only valid for the CONECOFOR sites. From the distribution of  $ovi$  values (Table 5), relative theoretical  $O_3$  risk classes can be obtained by percentile analysis:

$ovi < 0.40$  (<25<sup>th</sup> percentile)= low risk

$0.4 < ovi < 1.864$  (25<sup>th</sup>-75<sup>th</sup> percentile)= medium risk

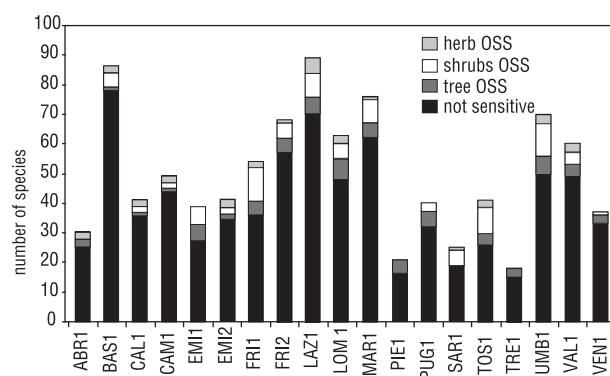
$ovi > 1.864$  (>75<sup>th</sup> percentile)= high risk.

The major determinant of potential  $O_3$  vulnerability is obviously the MTS (Appendix B). A first group of PMPs above the 75<sup>th</sup> percentile included mostly pure beech stands: EMI2, ABR1, VEN1, PIE1. Their theoretical vulnerability is due to dominance of sensitive tree species. In EMI2, *Fagus sylvatica* has a considerable weight also in the shrub layer. VAL1 also belongs to this group, its tree layer consisting exclusively of sensitive conifers *Larix decidua* and *Picea abies*. The tree layer is also important in PMPs FRI1, TRE1, CAL1 and FRI2: however, the high cover of a relatively low number of sensitive species in the shrub layer determined a lower  $ovi$ . Lower values of  $ovi$  define a group

**Table 4 -** Total number of species collected at the PMPs and percentage of ozone sensitive species (OSS).

*Numero totale di specie raccolte alle aree permanenti e percentuale di specie ritenute ozono sensibili (OSS).*

PMP	MTS	Total N. of species	% OSS
ABR1	Beech	30	16,66
BAS1	Oak	86	9,30
CAL1	Beech	41	12,19
CAM1	Beech	49	10,02
EMI1	Oak	39	30,76
EMI2	Beech	41	17,07
FRI1	Mix.Decid.	54	33,33
FRI2	Spruce (fir)	68	16,17
LAZ1	Oak	89	21,34
LOM1	Spruce (fir)	63	23,80
MAR1	Oak	77	19,48
PIE1	Beech	21	23,80
PUG1	Beech	40	20,00
SAR1	Holm-Oak	25	24,00
TOS1	Holm-Oak	41	36,58
TRE1	Spruce	18	16,66
UMB1	Oak	70	28,57
VAL1	Spruce	60	18,33
VEN1	Beech	37	10,81



**Figure 2 -** Total number of species, number of  $O_3$  sensitive species (OSS) separated into trees, shrubs and herbs per each PMP (1999 data).

*Numero totale di specie, divise per  $O_3$  tolleranti ed  $O_3$  sensibili, quest'ultime ancora suddivise tra alberi, arbusti ed erbe.*

**Table 5 -** Results of OVI index, based on the “weight” that the ozone sensitive species respectively have in the tree(1), shrub(2) and herb (3) layers, by means of simple combination. See the text for the explanations.

*Risultati dell'indice OVI, basato sul “peso” che le specie ozono sensibili hanno rispettivamente nello strato arboreo, arbustivo ed erbaceo. Vedi il testo per ulteriori spiegazioni.*

PMP	Tree Index	Shrub Index	Herb Index	OVI - Vulnerability Index	Theoretic vulnerability
EMI2	1,0000	1,0000	0,0001	2,0001	
PIE1	1,0000	1,0000	0,0001	2,0001	
ABR1	1,0000	1,0000	0,0001	2,0001	
VAL1	1,0000	1,0000	0,0000	2,0000	
VEN1	1,0000	1,0000	0,0000	2,0000	
FRI1	1,0000	0,7271	0,0004	1,7275	
TRE1	1,0000	0,6235	0,0000	1,6235	
CAL1	1,0000	0,2799	0,0000	1,2799	
CAM1	0,6148	0,3853	0,0000	1,0002	
FRI2	0,7351	0,2490	0,0000	0,9841	
LOM1	0,5132	0,3180	0,0000	0,8312	
PUG1	0,6147	0,0622	0,0000	0,6769	
EMI1	0,0505	0,5366	0,0001	0,5871	
UMB1	0,0351	0,3922	0,0001	0,4275	
TOS1	0,1216	0,2507	0,0001	0,3724	
BAS1	0,0000	0,3208	0,0000	0,3208	
MAR1	0,0508	0,2610	0,0001	0,3119	
LAZ1	0,0022	0,0489	0,0001	0,0512	
SAR1	0,0058	0,0228	0,0000	0,0286	

of stands which share some common features: all have a highly structured community (SAR1, TOS1) and/or are still influenced by recent release from intense disturbance. Species diversity and a large amount of non-sensitive or moderately sensitive species (*Quercus* and sclerophyllous species) make for a negligible role of the tree layer and a relatively higher vulnerability in the understorey.

The role of the tree canopy in determining the potential sensitivity of the site should also be considered in the light of its higher exposure to  $O_3$ , its role as  $O_3$  scavenger and driver of  $O_3$  deposition and uptake. The latter role will be examined in the next chapter. With respect to the two former roles, the sensitivity of the shrub and herb layers is interesting in the framework of different scenarios of development of the canopy layer. MILLER (1973) reported changes in the canopy structure of *Pinus ponderosa* due to  $O_3$ . He suggested that they may create different conditions in the stand environment which may have a series of consequences that can affect successional dynamics. Gaps in the tree layer always change light conditions of the understorey, initiating a new successional dynamics. However, these dynamics may be subject to different external pressure: for example, at CAL1 gaps may allow potential  $O_3$  injury to one species in the shrub layer (*Fagus sylvatica*),

which shows high frequency and a relative high mean cover. In FRI1 the same scenario can lead to potential  $O_3$  injury in 14 species (with relatively low frequencies and cover) in the shrub layer and 16 species in the herb layer.

### Forest and canopy structure

The dimensional and structural characteristics of tree stand communities are summarized in Table 6. The PMPs are in forest stands that have aged differently according to the species and the management system: PMPs within high forests are mostly adult stands of Norway spruce and beech, whereas PMPs in coppices are mostly oak stands that may be regarded as young forest crops. The structure of the canopy layer is important for an understanding of its roughness, which is a key factor in gas exchange (Monin Obukhov theory of similarity): in general, the greater the roughness, the higher the gas exchange and the potential for  $O_3$  uptake. PMPs in homogeneous forest areas resulted almost always into a full canopy cover without permanent gaps; lower canopy cover values (70%) are only reported for SIC1 and BAS1, where concurrent disturbances (grazing) reduced the original stool density. In closed forests, canopy roughness is mostly determined (although not linearly, MONTEITH and UNSWORTH 1990) by tree density, tree height, LAI and displacement of foliage within the canopy layer (SHAW and PEREIRA 1982). Tree height is also important in terms of water use efficiency.

### Tree height, vertical structure and diversity

Tree height is important for water use efficiency and as a determinant of canopy roughness. Under closed canopy conditions, the higher the canopy layer, the greater the roughness; in addition, for a certain species, taller trees tend to be less efficient in water use. Given the close relationship between mean tree height and age ( $R^2=0.70$ ), Norway spruce and beech PMPs (i.e. the tree species in older stands) had the greatest tree heights. Vertical stand structure of the various PMPs is typically one-storied, while a patchy, subordinate layer is often present into high forests; a two-storeyed structure is recognized in a few coppice forests due to the release of standards. A clearly differentiated stand structure (even-aged to stratified and irregular) was only observed in alpine Norway spruce stands. In most cases, one or two species comprise the dominant storey. The ratio of number of tree species in each PMP and their presence into the dominant storey is mostly determined by the irregular thinning characterizing the past management

of high forests and suspension of harvestings in the coppice PMPs (stored coppices and transitory crops). Tree density is generally high in all PMPs. The proportion of the main (dominant) storey over the full standing crop in terms of basal area ranges from 53% to 93% in spruce-dominated forests, according to the diversity of vertical stand structure, i.e. from one-storeyed to uneven-aged and irregular stands. The parameter shows a much more limited variation in one-storeyed beech PMPs (60%) and Turkey oak-dominated stands (62%). More variable values occur in mixed deciduous oaks and holm oak PMPs.

#### Tree density, canopy cover, canopy gaps

Although canopy cover was always >70%, tree density varies considerably in relation to management and stand age, which in turn were interrelated, as younger stands always host coppice or transitory crops. Thus, stored coppices and transitory crops aged 35-75 years had the highest tree densities (739-4540 trees per ha), whereas 80-200 yrs old high forests had 228 - 1043 trees per ha. With few exceptions, canopy gaps were <10%.

#### Leaf Area Index, top height and canopy depth

LAI values varied with species and, within species, according to stand age and basal area. Older beech PMPs had the highest values, while there was little difference in LAI between oak and Norway spruce stands. The difference between top and mean tree height describes the upper canopy layer i.e. the contact surface between the forest and the atmosphere. With few exceptions (LOM1 and MAR1), this difference was about 5 m or less and depended mainly on stand age, tree species composition and management system. The ratio of crown depth to tree height provides an indication of foliage displacement in the canopy layer. Norway spruce generally had the highest values (foliage displaced over a larger portion of the trunk), whereas beech and oak were similar. In general, the ratio decreased with increasing mean height, although a clear trend is obvious only for beech ( $R^2=0.66$ ).

#### Summary of results

The above findings are summarized in Table 7. We assumed that canopy roughness increased with increasing tree height and LAI, and decreased with increasing tree density and tree height/canopy depth ratio. We also gave the same weight to each factor. Thus data in Table 7 have been expressed in relation to the maximum of each data series and re-scaled (when necessary) to obtain

**Table 6 -** Mensurational parameters of the PMPs arranged according to the main tree species. Age: average age in the dominant storey; DIFN: Diffuse Non Interception. BA: Total Basal Area; BA dominant Basal Area in the dominant storey; Mean and Top height: mean and dominant height values. Minimum dbh surveyed = 3 cm in coppice; = 5 cm in high forests and transitory crops. *Caratteristiche dendrometriche delle aree permanenti elencate secondo la specie principale.*

PMP code	Main Tree Species	Vertical stand structure	Management system	Age yrs	Tree density n*ha <sup>-1</sup>	Tree species in PMP no.	Tree species in the dominant storey no.	MTS dominant storey %	Canopy cover %	DIFN	Mean height m	Top height m	Canopy depth m	LAI m <sup>2</sup> m <sup>-2</sup>	LAD m <sup>2</sup> m <sup>-3</sup>	BA total m <sup>2</sup>	BA dominant m <sup>2</sup>
FRI2	<i>Picea abies</i>	one-storeyed	high forest	110	532	2	1	100	80	0.05	29.1	32.6	18.2	3.98	0.22	52.90	28.13
LOM1	<i>Picea abies</i>	stratified	high forest	80	1043	9	8	65	80	0.03	18.3	26.1	17.0	4.29	0.25	40.23	35.68
TRE1	<i>Picea abies</i>	one-storeyed	high forest	200	393	2	1	100	80	0.11	28.2	30.5	22.5	2.94	0.13	53.89	35.59
VAL1	<i>Picea abies</i>	irregular	high forest	150	745	2	2	70	70	0.09	20.0	24.7	14.3	3.26	0.23	50.18	46.75
ABR1	<i>Fagus sylvatica</i>	one-storeyed	high forest	110	899	1	1	100	90	0.02	19.5	24.6	9.3	4.67	0.50	40.09	32.64
CAL1	<i>Fagus sylvatica</i>	one-storeyed	high forest	110	333	2	1	100	80	0.03	24.1	28.6	14.1	4.36	0.31	39.90	22.39
CAM1	<i>Fagus sylvatica</i>	one-storeyed	high forest	100	228	2	1	100	90	0.01	26.8	28.0	14.1	4.1	0.38	47.57	32.68
PUG1	<i>Fagus sylvatica</i>	one-storeyed	high forest	85	940	7	2	97	90	0.01	22.6	27.5	14.2	5.43	0.38	43.77	22.87
VEN1	<i>Fagus sylvatica</i>	one-storeyed	high forest	110	345	1	1	100	95-100	0.01	23.9	25.2	7.4	5.25	0.71	34.64	15.34
EMI2	<i>Fagus sylvatica</i>	two-storeyed	stored coppice	40	4540	2	2	99	95-100	0.03	9.9	15.2	9.3	4.61	0.50	35.77	20.36
PIE1	<i>Fagus sylvatica</i>	one-storeyed	transitory crop	55	1213	4	2	93	90	0.03	16.1	20.0	11.1	4.24	0.38	28.93	16.18
BAS1	<i>Quercus cerris</i>	two-storeyed	transitory crop	65	917	3	2	95	85	0.03	16.9	20.2	7.0	4.44	0.38	40.85	23.96
SIC1	<i>Quercus cerris</i>	one-storeyed	transitory crop	50	855	3	2	98	70	0.13	14.6	16.9	6.8	2.44	0.36	25.01	14.58
UMB1	<i>Quercus cerris</i>	one-storeyed	transitory crop	75	739	8	2	99	95-100	0.04	25.5	28.5	10.9	4.11	0.38	33.92	21.38
LAZ1	<i>Quercus cerris</i>	one-storeyed	stored coppice	35	1629	2	1	100	90	0.07	13.3	16.8	5.1	3.38	0.66	23.94	14.99
MAR1	<i>Quercus cerris</i>	two-storeyed	stored coppice	35	4233	11	3	94	90	0.03	12.1	20.4	5.7	4.43	0.78	35.84	23.40
EMI1	<i>Quercus spp.</i>	two-storeyed	stored coppice	45	2057	6	2	57	95-100	0.07	14.2	19.1	12.6	3.35	0.27	25.68	20.21
FRI1	Mixed broadleaves	one-storeyed	stored coppice	45	1126	6	5	29	85	0.02	17.1	22.4	9.7	4.44	0.46	24.05	15.11
SAR1	<i>Quercus ilex</i>	two-storeyed	stored coppice	50	1710	5	1	100	90-95	0.02	13.5	18.7	8.6	4.04	0.46	40.54	24.95
TOS1	<i>Quercus ilex</i>	two-storeyed	stored coppice	50	2380	13	7	71	85	0.04	11.1	15.4	6.2	5.22	0.84	26.21	8.07

**Table 7 –** Relative scores for tree density, mean height, canopy depth and LAI. Tree density and canopy depth were also rescaled using the reciprocal of their relativized values in order to have consistent meaning (direction) with the other indexes.

*Valori relativizzati della densità degli alberi, altezza media, profondità della chioma e LAI. La densità degli alberi e la profondità della chioma sono state anche riscalate attraverso il reciproco del loro valore relativizzato in modo da avere un significato (direzione) coerente a quello degli altri indici.*

PMP	MTS	Tree density	Mean height	Canopy depth	LAI	Synthesis
ABR1	<i>Fagus sylvatica</i>	0,80	0,67	0,49	0,86	2,82
BAS1	<i>Quercus cerris</i>	0,80	0,58	0,56		
CAL1	<i>Fagus sylvatica</i>	0,93	0,83	0,38	0,80	2,94
CAM1	<i>Fagus sylvatica</i>	0,95	0,92	0,44		
EMI1	<i>Quercus spp.</i>	0,55	0,49	0,05	0,62	1,71
EMI2	<i>Fagus sylvatica</i>	0,00	0,34	0,00	0,85	1,19
FRI1	<i>Quercus spp.</i>	0,75	0,59	0,40	0,82	2,55
FRI2	<i>Picea abies</i>	0,88	1,00	0,33	0,73	2,95
LAZ1	<i>Quercus cerris</i>	0,64	0,46	0,59	0,62	2,31
LOM1	<i>Picea abies</i>	0,77	0,63	0,01	0,79	2,20
MAR1	<i>Quercus cerris</i>	0,07	0,42	0,50	0,82	1,80
PIE1	<i>Fagus sylvatica</i>	0,73	0,55	0,27	0,78	2,33
PUG1	<i>Fagus sylvatica</i>	0,79	0,78	0,33	1,00	2,90
SAR1	<i>Quercus ilex</i>	0,62	0,46	0,32		
SIC1	<i>Quercus cerris</i>	0,81	0,50	0,50	0,45	2,27
TOS1	<i>Quercus ilex</i>	0,48	0,38	0,40	0,96	2,22
TRE1	<i>Picea abies</i>	0,91	0,97	0,15	0,54	2,58
UMB1	<i>Quercus cerris</i>	0,84	0,88	0,55	0,76	3,02
VAL1	<i>Picea abies</i>	0,84	0,69	0,24	0,60	2,36
VEN1	<i>Fagus sylvatica</i>	0,92	0,82	0,67	0,97	3,38

**Table 8 –** Nutrients concentration on the organic layer (0) and on the surface mineral layer (1).

*Concentrazioni di nutrienti nello strato organico (0) e nello strato superficiale del suolo minerale (1) delle aree permanenti.*

PMP code	Layer	Ctot g kg <sup>-1</sup>	Ntot g kg <sup>-1</sup>	C/N	P tot mg kg <sup>-1</sup>	C/P	K tot mg kg <sup>-1</sup>	Ca tot mg kg <sup>-1</sup>	Mg tot mg kg <sup>-1</sup>	pH (CaCl <sub>2</sub> )	Ac-Exc cmol kg <sup>-1</sup>	ECA cmol kg <sup>-1</sup>	ECB cmol kg <sup>-1</sup>	CEC cmol kg <sup>-1</sup>	Base saturation%
ABR1	0	354	15.03	23.01	1098	323	3278	14727	3508	5.02					
	1	103	7.04	13.09						4.09	1	0.09	11.07	12.05	93
BAS1	0	117	7.05	15.06	809	145	4346	7540	3699	5.06					
	1	37	3.01	11.09						4.06	1	0.07	5.09	6.06	89
CAL1	0	309	17.02	18.00	1353	228	3022	11920	4172	5.00					
	1	113	8.04	13.04						4.03	4	3.07	5.01	8.09	58
CAM1	0	289	16.01	18.00	1733	167	8080	20860	5626	5.06					
	1	164	12.02	13.04						5.05					
EMI1	0	352	15.07	22.04	707	498	1355	12544	2136	4.03					
	1	40	2.05	16.01						3.08	4	4.03	2.02	6.04	34
EMI2	0	145	8.01	17.08	598	242	3352	5926	9872	4.09					
	1	36	3.00	11.08						4.01	2	2.05	3.08	6.03	60
FRI1	0	286	17.05	16.03	898	318	3485	12541	4877	4.08					
	1	39	3.04	11.03						4.03	2	1.08	8.02	10.00	82
FRI2	0	434	19.00	22.09	906	480	1724	4592	1109	3.04					
	1	105	6.07	15.06						3.02	11	11.05	2.01	13.06	15
LAZ1	0	256	9.03	27.5	880	291	3347	8519	3181	4.09					
	1	23	1.08	12.09						4.00	4	4.00	1.09	6.00	33
LOM1	0	131	6.06	19.08	874	150	2547	3299	5559	3.05					
	1	95	5.03	18.00						3.09	5	4.09	2.05	7.04	34
MAR1	0	151	7.06	19.09	420	359	3642	26589	2749	6.07					
	1	24	1.06	14.08						6.06	0	0.02	7.07	7.09	97
PIE1	0	59	3.03	18.00						3.07	7	7.02	0.05	7.07	6
	1	25	1.08	14.04						4.02					
PUG1	0	472	17.05	26.9	1148	411	1651	19313	1943	5.00					
	1	61	5.01	11.09						4.08	1	0.07	7.05	8.02	92
SAR1	0	126	7.09	16.00	952	132	4834	8963	2685	5.08					
	1	47	3.05	13.03						5.08	0	0.02	9.03	9.05	98
SIC1	0	87	5.04	16.03	797	110	2589	6116	2179	6.00					
	1	44	3.02	13.08						4.09	0	0.06	8.05	9.01	94
TOS1	0	398	12.09	30.8	587	678	1740	16572	31512	5.06					
	1	87	5.01	17.01						6.08	0	0.02	19.07	19.09	99
TRE1	0	512	16.08	30.5	656	780	1349	6490	569	3.03					
	1	270	9.04	28.7						3.00	3	3.08	14.00	17.08	79
UMB1	0	129	7.04	17.04	767	168	6179	27950	9815	6.06					
	1	49	3.08	12.06						6.07					
VAL1	0	392	13.00	30.2	1003	390	2097	10416	1860	4.00					
	1	16	0.08	19.09						3.09					
VEN1	0	281	14.05	19.03	1117	251	2767	7906	2743	4.08					
	1	41	3.05	11.08						4.03	3	2.09	4.03	7.02	60

consistent meaning in relation to canopy roughness. Then, data were summed to obtain a score (Table 7). In general, beech high forests had the highest scores. In particular, the beech PMP VEN1 was the site with the highest values of canopy roughness, and was therefore potentially more favourable to gas exchange, a condition that favours O<sub>3</sub> uptake. It was mostly due to low tree density, low canopy depth and high LAI.

#### Nutrient supply: soil and foliage

Soil data are summarized in Table 8. Low pH values seldom occur in Italian PMPs. Basic cations (K, Ca, Mg) generally have optimum concentrations. Nutrient holding capacity of soil examined is generally good. Low values of Cation Exchange Capacity (CEC) were found at BAS1, EMI1, EMI2 and LAZ1. Very low Exchangeable Base Cations (EBC), which indicates a low availability of basic cations (K, Ca and Mg), was found only at PIE1. N and P have been reported to play a role in determining plant response to O<sub>3</sub>. N concentration in the organic layer was always below 20 g kg<sup>-1</sup>, and in six cases below

8 g kg<sup>-1</sup>. Low C/N ratios in organic layer indicate rapid mineralisation of organic matter and hence high N availability. The C/N ratio decreases from organic to the mineral layers. The average C/N values observed should ensure sufficient availability of N. Phosphorus availability is determined by the decomposition rate of soil organic matter (SOM), which is indicated by the C/P ratio. A low C/P ratio is an index of high P availability. Total P concentrations were below 700 mg kg<sup>-1</sup> in only two PMPs (EMI2 and MAR1) and above 1000 mg kg<sup>-1</sup> in six PMPs. The C/P ratio was elevated at PMPs EMI1, FRI2, TOS1 and TRE1, suggesting limited availability of P.

**Table 9 –** Foliar nutrients: 1995-1999 mean and standard deviation (SD). In some cases, not all the 3 survey years were available (see asterisks). Data are in mg g<sup>-1</sup> d.w.

*Nutrienti fogliari: media e deviazione standard (SD) 1995-1999. In alcuni casi non erano disponibili tutti i 3 anni di indagine (vedi asterischi). I dati sono in mg g<sup>-1</sup> s.s.*

PMP no.	Code	Statistic	N	P	Ca	Mg	K
1	ABR1	Mean	25,10	1,47	15,06	2,49	7,27
		SD	2,28	0,39	3,38	0,37	3,74
2	BAS1(*)	Mean	22,62	1,43	11,73	2,90	14,57
		SD	0,09	0,40	2,24	1,00	5,38
3	CAL1(*)	Mean	22,45	1,33	9,63	1,91	6,55
		SD	1,21	0,22	1,21	0,68	0,27
4	CAM1	Mean	25,22	1,26	16,65	2,73	13,82
		SD	0,77	0,32	1,45	1,05	4,60
5	EMI1	Mean	22,29	0,57	8,75	2,09	5,26
		SD	6,27	0,14	1,24	0,12	1,39
6	EMI2	Mean	24,94	1,12	12,20	2,47	9,70
		SD	6,08	0,28	4,32	0,21	1,68
7	FRI1 Hornbeam	Mean	22,64	0,87	12,02	3,43	7,89
		SD	2,50	0,15	1,15	0,39	1,80
7	FRI1 (Pedunculate oak**)	Mean	24,50	1,00	11,93	3,58	7,24
		SD	5,44	0,24	0,93	0,96	1,73
8	FRI2	Mean	13,27	0,78	8,56	1,56	5,33
		SD	1,84	0,19	2,06	0,39	1,48
9	LAZ1	Mean	25,67	1,00	8,15	2,04	9,81
		SD	4,74	0,33	2,10	0,74	2,40
10	LOM1	Mean	13,28	1,60	6,48	1,28	4,86
		SD	2,42	0,17	1,71	0,08	1,03
11	MAR1(*)	Mean	22,60	0,62	15,65	1,57	9,45
		SD	2,22	0,03	5,88	0,40	0,08
12	PIE1 (**)	Mean	30,48	0,98	8,40	1,85	9,90
		SD	6,77	0,55	0,06	0,56	0,29
13	PUG1(*)	Mean	25,28	1,67	9,98	3,15	11,05
		SD	1,69	0,07	1,14	0,31	5,85
14	SAR1 (*)	Mean	14,98	0,87	6,13	1,46	6,33
		SD	4,04	0,24	1,25	0,03	2,84
15	SIC1	Mean	22,31	1,11	10,34	2,02	10,17
		SD	3,09	0,43	2,19	0,78	1,50
16	TOS1	Mean	12,79	0,86	4,64	3,86	5,59
		SD	0,47	0,02	0,75	0,80	0,52
17	TRE1	Mean	14,17	1,21	5,52	1,14	8,03
		SD	2,35	0,41	0,24	0,33	2,99
18	UMB1	Mean	22,86	1,38	11,44	1,29	9,46
		SD	1,01	0,33	2,65	0,57	1,82
19	VAL1 (***)	Mean	10,17	1,11	9,28	1,31	5,86
		SD	1,20	0,43	0,27	0,23	0,43
20	VEN1	Mean	23,32	1,57	13,08	2,84	10,65
		SD	0,43	0,36	5,10	1,84	5,38

(\*) 1995 and 1997 only  
 (\*\*) 1995 and 1999 only  
 (\*\*\*) 1997 and 1999 only

Foliar nutrient concentrations are shown in Table 9. As reported by MATTEUCCI *et al.* (2000), nutrient supply of trees at the various PMP is sufficient in most cases, with some instances of high N values in beech (*e.g.*, PIE1) and deciduous oak (*e.g.* LAZ1). The ratio of elements also generally suggests good nutritional balance.

#### Nitrogen deposition

Nitrogen is often a limiting nutrient for vegetation in pristine areas and N deposition, coupled with warmer temperature, may lead to increased tree growth (thus counteracting O<sub>3</sub> effects, TAKEMOTO *et al.* 2001) and vegetation changes (FENN *et al.* 1998; BOBBINK *et al.* 1998). Atmospheric overload of nitrogen in Northern Italy has already been highlighted in studies on surface waters (MOSELLO *et al.* 2001). Long-term trend of deposition chemistry show that sulphate and acidity are decreasing, while nitrogen is fairly constant over the last twenty years (ROGORA *et al.* 2001).

Among CONECOFOR PMPs, the highest ammonium and nitrate concentrations were measured in sites located in northern Italy (LOM1, PIE1, EMI1, EMI2). Deposition fluxes depends also on the amount of precipitation, which shows a general decreasing pattern from North to South (Table 10). Comparison of precipitation in the three years of the study showed highest values in 2000 in northern Italy, with peaks of 2517 and 2978 mm y<sup>-1</sup> (PIE1 and LOM1, respectively). In the central and southern stations, variability between years was not so pronounced, with the wettest year being 1999. Open field deposition of the different species of nitrogen (Table 11) showed mean values of the same order for N-NO<sub>3</sub> and N- NH<sub>4</sub> in three year period, with organic nitrogen (ON) showing the highest variability between plots. Mean percentage contributions of N-NO<sub>3</sub>, N- NH<sub>4</sub>, and ON in all the considered plots were 39, 39, 22%, respectively. The highest open field total nitrogen (TN) deposition was measured in PIE1 and LOM1 (151 and 121 mmol m<sup>-2</sup> y<sup>-1</sup>, respectively) (Table 11). The lowest flux was measured in the northeastern Italy in plots FRI2 and TRE1 (60 and 61 mmol m<sup>-2</sup> y<sup>-1</sup>) and in Sicily (45 mmol m<sup>-2</sup> y<sup>-1</sup>). Compared with OF deposition, N deposition in the plot showed the same or slightly lower values for N- NH<sub>4</sub> and higher values for N-NO<sub>3</sub> and ON, with higher TN deposition. Mean percentage contributions of TN were 32, 41 and 29% for N-NO<sub>3</sub>, N- NH<sub>4</sub>, ON. Peak values were measured in PIE1 and EMI1 (172 and 210 mmol m<sup>-2</sup> y<sup>-1</sup>). Inter annual variability of annual TN deposition (Table 12) was high in the case

of TRE1, PIE1, EMI1, EMI2 and SIC1 (relative standard deviation, RSD > 20%); in the remaining stations RSD was less than 10%.

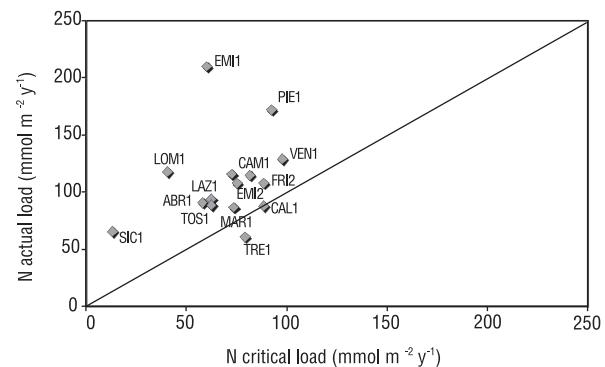
As nitrogen is a major nutrient, it is important to quantify N flux during the period of plant growth. Of course the period is not the same throughout Italy, being shorter in the Alps (June-September, changing with altitude) and longer in the south and islands. We chose the period of April-September for all stations. Precipitation data and the deposition of different forms of nitrogen are shown in Tables 10 and 11. Mean precipitation was about 50% of the annual total, with higher values in northern plots (60-65 %) and lower in the South (minimum 28% in Sicily). The corresponding deposition of TN, as percentage of annual deposition, was 55 and 57 % for open field and in plot, respectively. The highest percentages (70-80%) were obtained for LOM1, VEN1 and PIE1, and the lowest for SIC1 and ABR1.

The sensitivity of the PMPs to acid and nutrient nitrogen deposition, expressed in terms of critical loads, are reported by FERRETTI *et al.* (2000). While acidification does not seem to be a problem due to favourable geology and low acid input (MOSELLO and MARCHETTO 2000), the critical loads of N are largely exceeded in most of PMPs. Actual values are more than twice the critical loads in SIC1, PIE1, LOM1, FRI1, LAZ1, EMI1 and ABR1 (Figure 3). In several sites, the critical load of nitrogen is even exceeded in the period April-September alone. A separate indication of excessive N load comes from nitrate concentrations in streams (STODDARD 1994; TRAAEN and STODDARD 1995). Stream water samples collected close

**Table 10** - Amount of precipitation (mm) during the three study years and in the period April-September.

*Volumi di precipitazione (mm) nei tre anni esaminati nel periodo Aprile-Settembre.*

	Year			April-September				
	1998	1999	2000	mean	1998	1999	2000	mean
ABR1	1236	1127	1340	1234	500	397	400	432
CAL1	1257	1848	1602	1569	393	765	726	628
CAM1	850	1428	1124	1134	518	569	422	503
EMI1	619	995	813	809	363	445	448	419
EMI2	-	1433	1382	1408	417	408	413	413
FRI1	-	961	-	961	-	472	-	472
FRI2	1423	1646	-	1535	956	955	371	761
LAZ1	933	1085	945	988	398	443	345	395
LOM1	1593	2157	2978	2243	1210	1262	1739	1404
MAR1	1038	1166	-	1102	423	579	-	501
PIE1	1252	2036	2517	1935	919	1296	1605	1273
SIC1	582	544	458	528	184	120	139	148
TOS1	717	1006	1330	1018	375	353	423	384
TRE1	1065	1093	1244	1134	786	737	718	747
VEN1	1203	1530	-	1367	902	991	-	946



**Figure 3** - Comparison between actual and critical loads of nitrogen.  
*Confronto tra carico critico stimato per l'N e deposizione effettiva di N.*

to PIE1, FRI2, EMI2 and LAZ1 showed intermediate level of N saturation for stream FRI2, LAZ1 and EMI2, and much higher levels for PIE1. These values are in line with N deposition data: 108, 94, 108, 172 mmol m<sup>-2</sup> y<sup>-1</sup>, respectively.

#### Drought stress and vapour pressure deficit

Comparison of all the proposed indices was possible only for LAZ1. Table 13 shows drought stress indices calculated over four summer seasons. Total values of (P-E\*) were always negative, with a maximum (-216.1 mm) in 2000 and a minimum (-174.7 mm) in 1998. Indeed, the Italian pluviometric trend is characterised by a deficit in precipitation during the growing season. These results confirm our previous considerations about the inadequacy of this index. Analysis on a weekly basis may improve its sensitivity, but we could not express it with a single value.

**Table 11** - Mean deposition of the three species of nitrogen during the three years period (mmol m<sup>-2</sup> a<sup>-1</sup>) and during the period April-September (mmol m<sup>-2</sup> (6 months)<sup>-1</sup>).

*Deposizione media delle tre specie di N durante i tre anni esaminati (mmol m<sup>-2</sup> a<sup>-1</sup>) e durante il periodo Aprile-Settembre (mmol m<sup>-2</sup> (6 months)<sup>-1</sup>).*

	Year			TF+SF			Apr-Sep		
	OF			TF+SF			OF		
	NO <sub>3</sub>	NH <sub>4</sub>	ON	NO <sub>3</sub>	NH <sub>4</sub>	ON	NO <sub>3</sub>	NH <sub>4</sub>	ON
ABR1	21	15	9	23	31	27	10	8	4
CAL1	28	18	28	30	16	42	10	10	12
CAM1	29	21	21	57	23	35	13	12	8
EMI1	35	52	13	57	103	24	19	22	14
EMI2	46	32	20	55	34	29	17	14	11
FRI1	38	38	10	40	40	36	20	23	5
FRI2	28	27	14	43	25	38	16	16	9
LAZ1	26	19	14	45	18	31	13	10	8
LOM1	45	47	43	45	29	43	34	38	14
MAR1	33	50	14	41	17	29	15	35	4
PIE1	65	63	22	76	64	31	47	51	16
SIC1	18	10	17	14	22	30	7	4	1
TOS1	31	35	7	44	43	8	15	20	7
TRE1	23	31	24	20	17	25	13	18	9
VEN1	39	47	17	49	51	30	30	39	11

**Table 12** - Inter annual variability of total N deposition in the open field and in the plot (mmol m<sup>-2</sup> a<sup>-1</sup>).  
 Variabilità interannuale delle deposizioni di azoto totale all'aperto e sottochioma.

	OF (year)				OF (Apr-Sep)				In the plot (year)				In the plot (Apr-Sep)			
	1998	1999	2000	mean	1998	1999	2000	mean	1998	1999	2000	mean	1998	1999	2000	mean
ABR1	47	51	40	46	18	28	11	19	-	91	-	91	-	54	-	54
CAL1	55	96	73	75	18	56	25	33	86	86	93	88	40	49	53	47
CAM1	48	89	73	70	31	40	29	34	92	116	136	115	50	62	52	54
EMI1	92	108	103	101	45	49	71	55	168	264	198	210	93	100	134	109
EMI2	-	100	98	99	-	40	43	42	-	116	99	108	-	56	37	47
FRI1	-	86	-	86	0	48	-	24	-	116	-	116	-	63	-	63
FRI2	69	68	44	60	51	42	29	41	84	128	110	108	67	89	-	78
LAZ1	59	74	42	58	30	34	28	31	83	97	102	94	43	39	34	39
LOM1	72	151	140	121	75	71	112	86	117	114	123	118	99	97	95	97
MAR1	106	87	-	96	65	43	-	54	85	90	-	87	48	43	-	46
PIE1	112	176	165	151	93	131	120	115	124	207	184	172	104	156	138	133
SIC1	56	54	25	45	14	14	7	12	71	96	30	66	40	6	24	23
TOS1	67	58	78	68	44	33	35	37	102	85	80	89	74	38	55	56
TRE1	56	97	80	78	34	51	37	41	47	55	82	61	29	28	47	35
VEN1	94	111	-	103	79	81	-	80	136	123	-	129	119	85	-	102

The index E/E\* was almost 50 %, with a maximum (55 %) in 1999 and a minimum (42 %) in 1998. It was not always correlated with pluviometric deficit (P-E\*) when non-useful precipitation occurs. The weekly trends are shown in Figure 4. Wide annual variability in the critical period (end of May to mid September) was due to variability of precipitation. Drought risk was high in certain weeks: E/E\* was below 20 % for 5 weeks in 1997, 6 weeks in 1998, 4 weeks in 1999 and 5 weeks in 2000.

The relative evapotranspiration index  $RE_T$  ranged from 31 % to 38 %. These values are lower than those of E/E\*, confirming that  $RE_T$  represents the maximum limit of possible water deficit. The range of variability of  $RE_T$  is lower than that of E/E\*, but the annual trend was the same.

Table 14 shows drought stress indices, calculated for summer seasons in the various PMPs. The total values of the (P-E\*) index were always positive for Alpine PMPs (FRI2, PIE1, TRE1, and 1999 of VAL1) and  $RE_T$  values were high indicating a low risk of drought stress. For all other PMPs, especially TOS1, (P-E\*) was negative and  $RE_T$  very low, suggesting that trees suffered water shortage.

The quality of the calculated drought stress indices depends on the quality of input parameters and on the accuracy of potential evapotranspiration estimate. Thornthwaite's formula generally underestimates this parameter, but is the only internationally acknowledged formula based on simple meteorological parameters measured on a routine basis (e.g. air temperature). We could not use other formula (such as Penman-Montheith, which is probably more accurate), because they require more complex meteorological parameters that were not measured in all the PMPs.

**Table 13** - Drought stress indices, calculated on 4 summer seasons, for the PMP LAZ1.

Indici di stress idrico, calcolati per 4 periodi estivi, per l'area LAZ1.

Starting data	Ending data	Drought stress indices						
		P	E*	First		Second		
				E	E/E*	P-E*	RE <sub>T</sub>	
mm	mm	mm	mm	%	mm	mm	%	
17/06/97	28/10/97	231,8	427,1	218,2	51	-195,3	139,3	33
16/06/98	26/10/98	280,0	454,7	192,4	42	-174,7	139,9	31
04/05/99	12/10/99	265,4	457,2	253,7	55	-191,8	202,3	38
02/05/00	03/10/00	255,2	471,3	250,4	53	-216,1	164,0	35

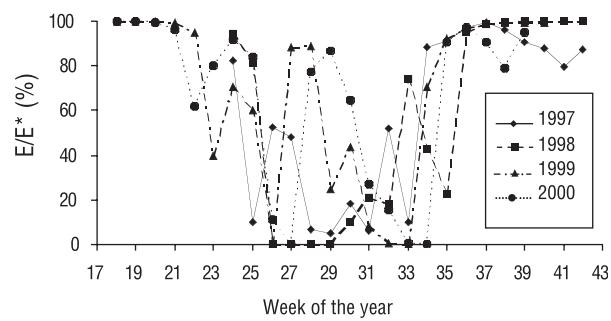
P= precipitation

E\*= potential weekly evapotranspiration

E= actual weekly evapotranspiration

P-E\*= difference between P and E\*

RE<sub>T</sub>= relative evapotranspiration



**Figure 4** - Weekly trends of drought stress index E/E\* (4 summer seasons) for PMP LAZ1 area.

Andamenti settimanali dell'indice di stress idrico E/E\* (4 periodi estivi) per l'area permanente LAZ1.

The calculated indices were considerably different in LAZ1. The E/E\* index is the most powerful, but it requires soil parameters, such as field capacity and wilting point, which were not always measured.  $RE_T$  is underestimated with respect to E/E\*, so it indicates higher stress. It represents the maximum limit of possible water deficit, but does not provide accurate values of drought stress. It is nevertheless a useful indication, in the absence of other information about soil water availability. The (P-E\*) index is the simplest: it does not consider soil water availability or non-useful precipitation, lost because of surplus.

VPD values are reported in Table 15 as seasonal mean and maximum for each PMP and year. Apart from considerable fluctuation from year to year, VPD values tend to decrease with altitude. Specifically, VPD is relatively low at high elevation sites in northern Italy (LOM1, PIE1, VAL1) and high at low elevation sites in central Italy (TOS1, LAZ1, EMI1). Between these two extremes, they were modulated according to latitude and elevation, two parameters that also control temperature, an important driver of VPD. This pattern was similar to that of  $RE_t$ .

### Summing up

In the previous sections we looked at several ecological characteristics of the PMPs. The effect of some of them (e.g., soil and foliage nutrition) on the vulnerability of the vegetation to  $O_3$  is controversial; on the other hand, the effect of others (N deposition) on plant response may counteract harmful effects of  $O_3$ . In both cases data related to these factors need to be accounted for when attempting to establish relationships between  $O_3$  and response indicators like growth and crown condition (see FERRETTI *et al.* 2003). Since N deposition may be important to moderate the effects of  $O_3$ , the position of each PMP according to N deposition and AOT40 over the period 1998-2000 (GEROSA *et al.* 2003) is shown in Figure 5. There is considerable scatter, although the highest AOT40 often go with low N deposition values in almost every stand type. The situation for deciduous oaks is interesting: the PMP EMI1 (main tree species: *Quercus petraea*) had high AOT40 (18470 ppb h<sup>-1</sup> on a four months basis) and the highest N deposition (210 mmol m<sup>-2</sup> a<sup>-1</sup>). On the other hand, SIC1 (main tree species: *Quercus cerris*) had AOT40 (24795 ppb h<sup>-1</sup> on a four months basis) and low N deposition (66 mmol m<sup>-2</sup> a<sup>-1</sup>). Because of the different species and soil characteristics, the different N deposition rates were not associated with different N content

**Table 14** - Drought stress indices, calculated on summer seasons, for all areas.  
*Indici di stress idrico, calcolati nei periodi estivi, per tutte le aree.*

Area	Starting data	Ending data	P mm	E* mm	Drought stress indices		
					Second	Third	
					P-E* mm	E mm	RE <sub>T</sub> %
ABR1	16/06/98	26/10/98	346,4	370,4	-23,6	125,7	34
	04/05/99	12/10/99	254,6	423,9	-169,3	181,2	43
	02/05/00	03/10/00	248,6	414,2	-165,6	99,0	24
CAL1	04/05/99	12/10/99	371,9	494,5	-122,7	197,8	40
	02/05/00	03/10/00	470,0	466,7	3,3	111,5	24
EMI1	16/06/98	26/10/98	297,0	483,3	-186,3	139,6	29
	04/05/99	12/10/99	345,2	604,0	-258,8	201,4	33
	02/05/00	03/10/00	308,0	600,7	-292,7	219,8	37
	04/05/99	12/10/99	369,7	503,6	-133,9	223,5	44
EMI2	02/05/00	03/10/00	262,4	477,6	-215,2	171,6	36
	16/06/98	26/10/98	839,6	367,0	459,2	290,3	79
	04/05/99	12/10/99	824,6	467,3	357,3	340,9	73
FRI2	02/05/00	03/10/00	571,6	468,0	103,6	264,6	57
	17/06/97	28/10/97	231,8	427,1	-195,3	139,3	33
	16/06/98	26/10/98	280,0	454,7	-174,7	139,9	31
LAZ1	04/05/99	12/10/99	265,4	457,2	-191,8	202,3	38
	02/05/00	03/10/00	255,2	471,3	-216,1	164,0	35
	02/05/00	03/10/00	1319,0	445,6	873,4	305,5	69
PIE1	15/06/96	21/10/96	331,0	439,6	-108,6	151,8	35
	17/06/97	30/09/97	154,0	374,7	-220,7	96,0	26
	16/06/98	26/10/98	321,0	448,2	-127,2	118,8	27
TOS1	04/05/99	12/10/99	40,3	587,7	-547,4	40,3	7
	17/06/97	28/10/97	535,6	317,2	218,4	262,5	83
	04/05/99	12/10/99	704,8	457,7	247,1	335,2	73
TRE1	02/05/00	03/10/00	656,2	472,0	184,2	340,4	72
	15/06/96	21/10/96	252,0	263,2	-11,2	144,4	55
	17/06/97	28/10/97	312,2	347,0	-34,8	163,8	47
VAL1	16/06/98	26/10/98	299,6	357,2	-57,6	170,5	48
	04/05/99	12/10/99	558,8	440,1	118,7	300,7	68
	02/05/00	03/10/00	332,7	431,2	-98,5	193,8	45

P= precipitation

E\* = potential weekly evapotranspiration

E= actual weekly evapotranspiration

P-E\* = difference between P and E\*

RE<sub>T</sub> = relative evapotranspiration

of foliage (see Table 9), however in the long-term and at the  $O_3$  exposure levels in question, it seems plausible that N deposition may influence the way in which the vegetation at the two PMPs reacts to  $O_3$ .

Species composition, forest structure and meteorological factors have a clearer connection with the potential  $O_3$  vulnerability of a site. The frequency of  $O_3$  sensitive species is a clear indication of potential effects of  $O_3$  on species diversity, while high OVI values indicate a more general vulnerability, driven by the MTS of the dominant storey and thus more relevant to ecosystem productivity. Forest structure is important for  $O_3$  deposition and gas exchange: stands with low density, tall trees, high LAI and foliage on the upper part of the tree trunks are more subject to intense gas exchange due to increased friction velocity determined by atmospheric turbulence. On the other hand, atmospheric conditions leading to  $O_3$  effects not only include high  $O_3$ , but also high  $RE_t$  (low potential for soil drought stress) and low

**Table 15 –** Max and mean VPD values at the various PMPs.  
*Valori medi e massimi di VPD ai vari PMP.*

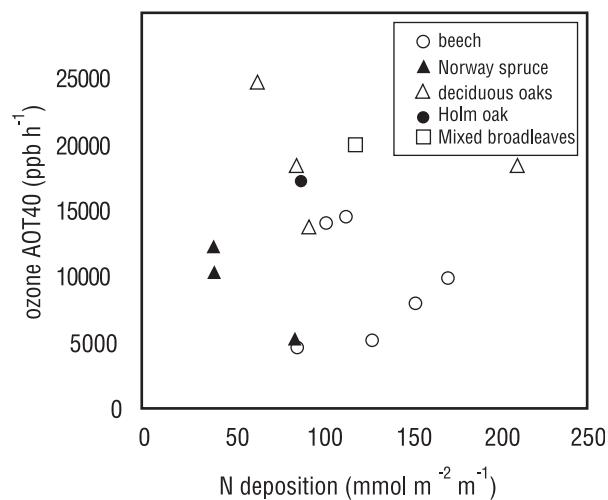
Area	Altitude (m)	Year	VPD max (kPa)	VPD med (kPa)
ABR1 OF	1560	1998	3,013	0,515
		1999	3,199	0,424
		2000	3,4	0,527
ABR1 IP	1490	1997	2,132	0,348
		1998	2,749	0,441
		1999	2,571	0,358
		2000	3,231	0,592
CAL1 OF	990	1999	4,186	0,432
		2000	3,653	0,441
CAL1 IP	1000	1999	2,971	0,501
		2000	3,371	0,459
		1998	4,165	0,894
EMI1 OF	200	1999	4,214	0,794
		2000	4,289	0,809
		1998	3,828	0,786
EMI1 IP	200	1999	3,659	0,545
		2000	3,872	0,551
		1999	2,769	0,412
EMI2 OF	860	2000	3,779	0,507
		1999	2,291	0,363
		2000	3,247	0,461
FRI2 OF	850	1998	3,572	0,3
		1999	3,207	0,176
		2000	3,008	0,21
FRI2 IP	820	1998	2,846	0,244
		1999	2,789	0,17
		2000	2,643	0,243
LAZ1 OF	675	1997	3,809	0,695
		1998	5,852	0,822
		1999	4,007	0,627
		2000	4,931	0,784
LAZ1 IP	690	1997	3,033	0,587
		1998	4,396	0,702
		1999	3,698	0,553
		2000	4,539	0,726
LOM1 OF	1188	1997	2,416	0,274
		1999	2,416	0,271
		2000	3,094	0,477
PIE1 OF	1320	2000	2,381	0,374
		2000	2,06	0,307
		1996	-	0,417
PIE1 IP	1155	1997	-	0,51
		1998	-	0,454
		1999	-	0,246
TRE1 OF	1796	1999	3,157	0,213
		2000	4,46	0,409
TRE1 IP	1780	1999	1,739	0,131
		2000	1,957	0,198
VAL1 OF	1660	1996	2,819	0,452
		1997	2,738	0,505
		1998	3,349	0,577
		1999	2,624	0,439
		2000	2,772	0,564
VEN1 IP	1158	1999	1,226	0,124
		2000	2,205	0,152

VPD (few constraints to  $O_3$  uptake). These aspects are summarized in Table 16. Each variable is reported in relative terms in relation to its own maximum (the original data are reported in the various tables of this paper). VPD max expressed as its reciprocal in order to have consistent meaning with  $RE_t$  and AOT40. Variables are grouped in three categories: variables defining inherent vegetation sensitivity to  $O_3$  (frequency of sensitive species and OVI), variables influencing  $O_3$  uptake (forest

structure,  $RE_t$  and VPD max), and variables defining  $O_3$  exposure (AOT40). Two synthetic scores were then calculated to identify the situations with a potential risk of adverse effects due to concomitance of high  $O_3$  exposure and conditions favourable to  $O_3$  uptake. We gave the same weight to each of the three terms. Unfortunately, a complete dataset was not available for all sites, so it was not possible to calculate the synthetic score for all PMPs. The first score is related to a potential risk in terms of  $O_3$  effects on plant diversity. It was calculated by summing the relative values of frequency of  $O_3$  sensitive species,  $RE_t$ , VPD max and AOT40. The second score reflects a more general risk in terms of forest condition. It was calculated by summing the relativized values of OVI, structure,  $RE_t$ , VPD max and AOT40. In general, the potential for adverse effects seems to be higher in PMPs located in northern and central Italy, though their AOT40 values were not always high.

## Conclusions

Composition, structure, nutrition, and climate vary widely among the PMPs of the CONECOFOR programme. Besides  $O_3$  exposure, species composition, canopy structure and climatic conditions are known to be important determinants of potential sensitivity of a given site to  $O_3$ . On the other hand, N deposition has been acknowledged as a factor that may moder-



**Figure 5 –** Annual mean of N deposition and June-September AOT40 over the period 1998-2000 at the PMPs of the CONECOFOR programme.

*Media annuale della deposizione di azoto e valori medi di AOT40 (periodo Giugno-Settembre) nel periodo 1998-2000 alle varie aree permanenti del programma CONECOFOR.*

**Table 16** – A tentative synthesis of the compositional, structural, meteorological factors and ozone exposure. Data have been relativized so they are non-dimensional. Increasing values indicate more favourable condition for  $O_3$  effects. See text for details.  
*Una sintesi preliminare dei dati di composizione, struttura e meteorologia ed esposizione ad  $O_3$ . I dati sono stati relativizzati rispetto al loro massimo e sono adimensionali. Valori crescenti indicano condizioni più favorevoli agli effetti dell' $O_3$ . I dettagli sono riportati nel testo.*

PMP no.	Code	Inherent vegetation vulnerability		Factors affecting gas exchanges			Ozone exposure	Potential risk for adverse effects on	
		Sensitive species	OVI	Canopy Structure	Ret	VPDmax		Plant diversity	Overall forest condition
1	ABR1	0,46	1,00	2,82	0,45	0,31	0,25	1,47	4,84
2	BAS1	0,25	0,16				0,56		
3	CAL1	0,33	0,64	2,94	0,42	0,26	0,17	1,18	4,42
4	CAM1	0,27	0,50				0,67		
5	EMI1	0,84	0,29	1,71	0,43	0,24	0,69	2,20	
6	EMI2	0,47	1,00	1,19	0,53	0,31	0,59	1,89	3,61
7	FRI1	0,91	0,86	2,55			0,62		
8	FRI2	0,44	0,49	2,95	0,92	0,31	0,41	2,08	5,08
9	LAZ1	0,58	0,03	2,31	0,45	0,22	0,53	1,78	3,53
10	LOM1	0,65	0,42	2,20		0,38	0,22		
11	MAR1	0,53	0,16	1,80			0,69		
12	PIE1	0,65	1,00	2,33	0,91	0,42	0,34	2,31	4,99
13	PUG1	0,55	0,34	2,90			0,71		
14	SAR1	0,66	0,01				0,36		
15	SIC1			2,27			1,00		
16	TOS1	1,00	0,19	2,22	0,30		0,57		
17	TRE1	0,46	0,81	2,58	1,00	0,26	0,38	2,10	5,04
18	UMB1	0,78	0,21	3,02			0,45		
19	VAL1	0,50	1,00	2,36	0,70	0,35	0,36	1,91	4,77
20	VEN1	0,30	1,00	3,38			0,15		

ate the potential detrimental effects of  $O_3$ . Presence, frequency and distribution of species known to be  $O_3$  sensitive were investigated. The mixed deciduous and holm oak forests seemed potentially more sensitive from the point of view of species diversity, as they host the highest share of  $O_3$  sensitive species. On the other hand, in view of known sensitivity of beech, PMPs with beech as main tree species in terms of canopy cover and basal area, are the communities with the highest potential risk in terms of productivity. Although a dense forest canopy may protect shrubs and herbs from exposure to  $O_3$ , changes in the species and structural may affect the future impact of  $O_3$  on a given site. The available data enables us to identify canopy conditions that may favour gas exchanges. Determinants of canopy roughness such as LAI, tree density, tree height and canopy depth vary and PMPs located in beech high forests were mostly the ones with conditions more conducive to increased gas exchange. No particular problem was found with soil or foliage nutrition, whereas N deposition was reported to exceed critical loads at several sites. On the other hand, high VPDmax values and low  $RE_t$  (an indicator of the potential maximum soil water shortage) were found to vary according to latitude and altitude. For this reason, PMPs located in northern Italy appear the most sensitive ones, although the dataset available make it not possible to include sites as SIC1, the southernmost and most  $O_3$  exposed PMP of the CONECOFOR network.

Within the limits of the present dataset, a tentative evaluation of the species composition and expected vulnerability, forest structure, VPD,  $RE_t$  and  $O_3$  exposure indicates that, despite relatively lower AOT40 values, highforests in northern Italy may have a higher potential for adverse effects on vegetation due to  $O_3$ . This is consistent with recent findings (e.g KARLSSON *et al.* 2003a) and provide further evidence of the necessity of a comprehensive approach to understand  $O_3$  effects on forests. In particular, extension (complete the coverage of the PMPs) and integration (included different measurement height in all the measurement sites) of the meteorological measurements are essential.

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**Appendix (A)** [http://www.gva.es/ceam/ICP-forests/icp\\_forests.htm](http://www.gva.es/ceam/ICP-forests/icp_forests.htm)

**LIST OF EUROPEAN OZONE SENSITIVE SPECIES**

(Trees, shrubs and perennial herbs included in Flora Europaea) - **Native species** in *italics*; **foreign species** in normal print.

<i>Abies cephalonica</i>	<i>Lonicera etrusca</i>	<i>Sambucus nigra</i>
<i>Acer campestre</i>	<i>Lonicera implexa</i>	<i>Sambucus racemosa</i>
<i>Acer granatense</i>	<i>Lonicera xylosteum</i>	<i>Sequoia</i> <i>giganteum</i>
<i>Acer platanoides</i>	<i>Morus alba</i>	<i>Solanum</i> <i>sodomeum</i>
<i>Acer pseudoplatanus</i>	<i>Morus nigra</i>	<i>Solidago</i> <i>canadensis</i>
<i>Acer saccharinum</i>	<i>Mycelis muralis</i>	<i>Sorbus</i> <i>aria</i>
<i>Agrimonia eupatoria</i>	<i>Myrtus communis</i>	<i>Sorbus aucuparia</i>
<i>Ailanthus altissima</i>	<i>Oenothera rosea</i>	<i>Sorbus domestica</i>
<i>Alchemilla xanthochlora</i>	<i>Onobrychis viciifolia</i>	<i>Sorbus mugeotii</i>
<i>Alnus glutinosa</i>	<i>Ostrya carpinifolia</i>	<i>Stachys officinalis</i>
<i>Alnus incana</i>	<i>Parthenocissus quinquefolia</i>	<i>Succisa pratensis</i>
<i>Alnus viridis</i>	<i>Picea abies</i>	<i>Symporicarpos albus</i>
<i>Anthyllis cytisoides</i>	<i>Picea glauca</i>	<i>Syringa vulgaris</i>
<i>Arbutus unedo</i>	<i>Pinus banksiana</i>	<i>Thalictrum minus</i>
<i>Artemisia vulgaris</i>	<i>Pinus cembra</i>	<i>Tilia cordata</i>
<i>Aruncus dioecus</i>	<i>Pinus contorta</i> v. <i>latifolia</i>	<i>Tilia platyphyllos</i>
<i>Asclepias syriaca</i>	<i>Pinus halepensis</i>	<i>Trifolium pratense</i>
<i>Atropa bella-donna</i>	<i>Pinus nigra</i>	<i>Tsuga canadensis</i>
<i>Berberis vulgaris</i>	<i>Pinus pinaster</i>	<i>Tsuga heterophylla</i>
<i>Betula pendula</i>	<i>Pinus pinea</i>	<i>Ulmus glabra</i>
<i>Buxus sempervirens</i>	<i>Pinus ponderosa</i>	<i>Ulmus minor</i>
<i>Calamintha grandiflora</i>	<i>Pinus strobus</i>	<i>Valeriana montana</i>
<i>Calystegia sepium</i>	<i>Pistacia lentiscus</i>	<i>Verbascum sinuatum</i>
<i>Carpinus betulus</i>	<i>Pistacia terebinthus</i>	<i>Veronica urticifolia</i>
<i>Centaurea nigrescens</i>	<i>Plantago lanceolata</i>	<i>Viburnum lantana</i>
<i>Centaurea paniculata</i>	<i>Plantago major</i>	<i>Viburnum opulus</i>
<i>Cistus salviifolius</i>	<i>Polygonum bistorta</i>	<i>Viburnum tinus</i>
<i>Clematis flammula</i>	<i>Populus alba</i>	<i>Viburnum x bodnantense</i>
<i>Clematis spp.</i>	<i>Populus nigra</i>	<i>Vinca difformis</i>
<i>Clematis vitalba</i>	<i>Populus tremula</i>	<i>Vitis vinifera</i>
<i>Colutea arborescens</i>	<i>Prunus armeniaca</i>	
<i>Convolvulus arvensis</i>	<i>Prunus avium</i>	
<i>Cornus alba</i>	<i>Prunus dulcis</i>	
<i>Cornus mas</i>	<i>Prunus persica</i>	
<i>Cornus sanguinea</i>	<i>Prunus serotina</i>	
<i>Corylus avellana</i>	<i>Prunus spinosa</i>	<i>Agrostis</i>
<i>Crataegus laevigata</i>	<i>Prunus virginiana</i>	<i>Aquilegia</i>
<i>Crataegus monogyna</i>	<i>Pseudotsuga menziesii</i>	<i>Betula</i>
<i>Cystisus heterochrous</i>	<i>Pyrus malus</i> subsp. <i>malus</i>	<i>Calystegia</i>
<i>Dittrichia viscosa</i>	<i>Quercus robur</i>	<i>Campanula</i>
<i>Epilobium angustifolium</i>	<i>Reseda odorata</i>	<i>Carya</i>
<i>Epilobium collum</i>	<i>Reynoutria japonica</i>	<i>Forsythia</i>
<i>Epilobium hirsutum</i>	<i>Rhamnus alaternus</i>	<i>Hieracium</i>
<i>Euonymus europaeus</i>	<i>Rhamnus catharticus</i>	<i>Lamium</i>
<i>Euphorbia dulcis</i>	<i>Ribes alpinum</i>	<i>Myosotis</i>
<i>Fagus sylvatica</i>	<i>Ricinus communis</i>	<i>Populus</i> (clones)
<i>Frangula alnus</i>	<i>Robinia pseudoacacia</i>	<i>Ribes</i>
<i>Fraxinus angustifolia</i>	<i>Rosa canina</i>	<i>Rosa</i>
<i>Fraxinus excelsior</i>	<i>Rubia peregrina</i>	<i>Rubus</i>
<i>Fraxinus ornus</i>	<i>Rubus fruticosus</i>	<i>Sambucus</i>
<i>Fraxinus pennsylvanica</i>	<i>Rubus idaeus</i>	<i>Spiraea</i>
<i>Fraxinus spp.</i>	<i>Rubus spectabilis</i>	<i>Trifolium</i>
<i>Geranium sylvaticum</i>	<i>Rubus ulmifolius</i>	
<i>Hippophae rhamnoides</i>	<i>Rudbeckia laciniata</i>	
<i>Juglans nigra</i>	<i>Rumex obtusifolius</i>	
<i>Juglans regia</i>	<i>Rumex pulcher</i>	
<i>Laburnum alpinum</i>	<i>Salix alba</i>	
<i>Lamiastrum galeobdolon</i>	<i>Salix caprea</i>	
<i>Lapsana communis</i>	<i>Salix daphnoides</i>	
<i>Larix decidua</i>	<i>Salix glabra</i>	
<i>Ligustrum ovalifolium</i>	<i>Salix pentandra</i>	
<i>Ligustrum vulgare</i>	<i>Salix purpurea</i>	
<i>Liriodendron tulipifera</i>	<i>Salix viminalis</i>	
<i>Lonicera caprifolium</i>	<i>Sambucus ebulus</i>	

Besides other genera containing sensitive species include:

<i>Agrostis</i>
<i>Aquilegia</i>
<i>Betula</i>
<i>Calystegia</i>
<i>Campanula</i>
<i>Carya</i>
<i>Forsythia</i>
<i>Hieracium</i>
<i>Lamium</i>
<i>Myosotis</i>
<i>Populus</i> (clones)
<i>Ribes</i>
<i>Rosa</i>
<i>Rubus</i>
<i>Sambucus</i>
<i>Spiraea</i>
<i>Trifolium</i>

## Appendix (B) -

### FREQUENCY OF THE OZONE SENSITIVE SPECIES IN ALL PERMANENT PLOTS (1999 DATA), FOR THE TREE AND SHRUB LAYERS

	Abr1	Bas1	Cal1	Cam1	Emi1	Emi2	Fri1	Fri2	Laz1	Lom1	Mar1	Pie1	Pug1	Sar1	Tos1	Tre1	Umb1	Vall1	Ven1
<b>Tree Layer</b>																			
<i>Acer campestre</i>								18	2		3		3						1
<i>Acer pseudoplatanus</i>									2				2						
<i>Arbutus unedo</i>															12	10			
<i>Betula pendula</i>									3		16								
<i>Carpinus betulus</i>							24		3		5		15						3
<i>Cornus mas</i>											1								3
<i>Corylus avellana</i>								2			4								
<i>Crataegus monogyna</i>	1		1												1		1		
<i>Eagus sylvatica</i>	24		24	24		24		13		7		24	24						24
<i>Fraxinus ornus</i>					24						24				24		4		
<i>Fraxinus oxyacarpa</i>								9											
<i>Larix decidua</i>									1		2								21
<i>Ostrya carpinifolia</i>															10		4		
<i>Picea abies</i>									24		19						24		21
<i>Pinus cembra</i>																	5		
<i>Pinus pinaster</i>																	1		
<i>Prunus avium</i>																	1		
<i>Prunus spinosa</i>								1											
<i>Quercus robur</i>								14											
<i>Robinia pseudoacacia</i>			1																
<i>Sorbus aria</i>										1	1								
<i>Sorbus aucuparia</i>									1				2						
<i>Sorbus domestica</i>		2									6						1		
<i>Ulmus minor</i>								10								1			
<i>Vitis vinifera</i>																2			
<b>Shrub Layer</b>																			
<i>Acer campestre</i>							12		2		10		1					4	
<i>Acer pseudoplatanus</i>	2							2		6									
<i>Arbutus unedo</i>															17	6			
<i>Carpinus betulus</i>							21		11		12		8					2	
<i>Clematis vitalba</i>					1	1									1		6		
<i>Cornus mas</i>	1								8		12							22	
<i>Cornus sanguinea</i>							6				1							3	
<i>Corylus avellana</i>							15	1			19							1	
<i>Crataegus monogyna</i>	20		1				17				15				1	3		11	
<i>Crataegus oxyacantha</i>									6		2							12	
<i>Euonymus europaeus</i>	2						14		3									4	
<i>Eagus sylvatica</i>	11		12	16		24		9		11		17	23						1
<i>Fraxinus excelsior</i>										1									
<i>Fraxinus ornus</i>							12				22				20		15		
<i>Fraxinus oxyacarpa</i>								1											
<i>Laburnum alpinum</i>										1									
<i>Larix decidua</i>																		2	
<i>Igustrum vulgare</i>	5						15												
<i>Lonicera caprifolium</i>									1									8	
<i>Lonicera etrusca</i>															3		13		
<i>Ostrya carpinifolia</i>																			
<i>Picea abies</i>								2		20							15		14
<i>Prunus avium</i>							1	2											1
<i>Prunus spinosa</i>								5	1		1				1				1
<i>Rosa canina</i>	8																		
<i>Rubia peregrina</i>																		1	
<i>Rubus ulmifolius</i>							1		2		7				15		7		
<i>Sorbus aria</i>										5	5								1
<i>Sorbus aucuparia</i>								1		7						1			1
<i>Sorbus domestica</i>					1				5		9						1		1
<i>Tilia platyphyllos</i>													1						
<i>Ulmus minor</i>							9												
<i>Viburnum lantana</i>									1										
<i>Viburnum opulus</i>								2											
<i>Viburnum tinus</i>										1									
<i>Vitis vinifera</i>									1										

Appendix (B) - *continued*

FREQUENCY OF THE OZONE SENSITIVE SPECIES IN ALL PERMANENT PLOTS (1999 DATA), FOR THE HERB LAYERS

	Abr1	Bas1	Cal1	Cam1	Emi1	Emi2	Fri1	Fri2	Laz1	Lom1	Mar1	Pie1	Pug1	Sar1	Tos1	Tre1	Umb1	Val1	Ven1
<b>Herb Layer</b>																			
<i>Acer campestre</i>				1		22					9		8					2	
<i>Acer platanoides</i>	1																		
<i>Acer pseudoplatanus</i>		4				1		10		13		2	2					1	1
<i>Agrimonia eupatoria</i>		2							2										
<i>Arbutus unedo</i>																2			
<i>Atropa belladonna</i>	1																		
<i>Berberis vulgaris</i>							2											1	
<i>Betula pendula</i>									1										
<i>Carpinus betulus</i>						20			18		9		7						
<i>Clematis vitalba</i>					1				4		3			10	1		14		
<i>Cornus mas</i>		2							6		11						20		
<i>Cornus sanguinea</i>						3			1										
<i>Corylus avellana</i>					4		10	18			13						1		
<i>Crataegus monogyna</i>	16		1			11	1	13			11						6		
<i>Epilobium angustifolium</i>																	5		
<i>Euonymus europaeus</i>	6	1				23			6								8		
<i>Euphorbia dulcis</i>						1	9		1	4							2		
<i>Fagus sylvatica</i>	20		14	23			24		8		12	3	13	24				18	
<i>Fraxinus excelsior</i>										9								5	
<i>Fraxinus ornus</i>				24					1		16				18		15		
<i>Fraxinus oxycarpa</i>						3													
<i>Geranium sylvaticum</i>																	10		
<i>Juglans regia</i>			2							4									
<i>Laburnum alpinum</i>																			
<i>Lamiastrum galeobdolon</i>		23				1	23				1								
<i>Lapsana communis</i>	3																		
<i>Larix decidua</i>																2			
<i>Ligustrum vulgare</i>	11						19												
<i>Lonicera caprifolium</i>				2					21			5							
<i>Lonicera etrusca</i>																21			
<i>Lonicera implexa</i>													3						
<i>Lonicera xylosteum</i>						12				14									
<i>Mycelis muralis</i>	19		16	23			11		1	1	1						6		
<i>Ostrya carpinifolia</i>																2			
<i>Picea abies</i>				1			20			12		1			20		20		
<i>Pinus cembra</i>															13		1		
<i>Pinus pinaster</i>									1										
<i>Pinus strobus</i>									8										
<i>Prunus avium</i>				4												6			
<i>Prunus spinosa</i>					4		4			11				1		3			
<i>Robinia pseudoacacia</i>			3																
<i>Rosa canina</i>		23																	
<i>Rubia peregrina</i>														1	23				
<i>Rubus idaeus</i>			11							10							2	5	
<i>Rubus ulmifolius</i>						1		4		12		24	11	21		15			
<i>Salix caprea</i>									1										
<i>Sambucus nigra</i>			1									1				1	3		
<i>Sambucus racemosa</i>										1									
<i>Sorbus aria</i>					2				4								1		
<i>Sorbus aucuparia</i>	1						20			18		23				11	9		
<i>Sorbus domestica</i>			2						12		14						1		
<i>Stachys officinalis</i>									16		1				1		3		
<i>Tilia platyphyllos</i>												3							
<i>Trifolium pratense</i>	11		2					19									1		
<i>Ulmus minor</i>										12				3					
<i>Veronica urticifolia</i>																			
<i>Viburnum opulus</i>						3													
<i>Viburnum tinus</i>															1				
<i>Vitis vinifera</i>															1				

## Appendix (C)

ACRONYM	EXPLANATION
ABR1....VEN1	Codes of the Permanent Monitoring Plots
AOT40	(Ozone) Accumulated Over a Threshold 40 ppb
CEC	Cations Exchange Capacity
CONECOFOR	CONtrollo ECOsistemi FORestali (forest ecosystems monitoring)
DBH	Diameter at Breast Height
DIFN	Diffuse Non-Interception
EA	Exchangeable Acidity
EAC	Exchangeable Acid Cation
EBC	Exchangeable Base Cations
LAI	Leaf Area Index
MTS	Main Tree Species
OF	Open field
ON	Organic Nitrogen
OSS	Ozone sensitive species
OVI	Ozone Vulnerability Index
PMP	Permanent Monitoring Plots
REt	Relative Evapotranspiration
RSD	Relative Standard Deviation
SMD	Soil Moisture Deficit
SOM	Soil Organic Matter
SU	Sampling Units (ground vegetation survey)
TN	Total Nitrogen
VPD	Vapour Pressure Deficit

Note : symbols of chemical elements and symbols for formulas are not considered here.

# Modelling stomatal uptake of ozone: data requirements and applicability to the CONECOFOR PMPs in Italy<sup>§</sup>

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**Abstract** – It is accepted that ozone ( $O_3$ ) effects on vegetation depend mostly on the amount of  $O_3$  uptake rather than on exposure. As direct measurement of uptake is unfeasible under most forest conditions, modelling is needed. There are different modelling approaches (e.g. diagnostic and prognostic models) which follow the electric resistance analogue principle. Unfortunately, these models are data intensive and implementation can be difficult in sites which are not designed for canopy-atmosphere flux studies. The paper provides an overview on modelling approaches, identifies data requirements and evaluates the actual chance for implementing a flux approach in the CONECOFOR PMPs.

**Key words:** *flux, forest monitoring, Italy, ozone, stomatal conductance.*

**Riassunto** – Modellizzazione dell'assorbimento di ozono: requisiti dei dati e applicabilità alle aree permanenti CONECOFOR in Italia. E' ormai accettato che gli effetti dell'ozono dipendono dalla dose assorbita più che dall'esposizione. Dato che la misura diretta dell'assorbimento di  $O_3$  non è praticabile nella maggior parte delle situazioni forestali, è necessario un approccio modellistico. Esistono diversi possibili approcci (es. modelli prognostici e diagnostici) che seguono il principio dell'analogia con la resistenza elettrica. Sfortunatamente, questi modelli sono molto esigenti in termini di dati e la loro implementazione può essere difficile in siti che non sono stati progettati per studi di flussi pianta-atmosfera. Questo articolo fornisce un panorama sugli approcci modellistici, identifica i requisiti in termini di dati necessari e valuta le possibilità applicative dei modelli di flusso alle aree permanenti CONECOFOR.

**Parole chiave:** *conduttanza stomatica, flusso, monitoraggio foreste, ozono.*

Estimated ozone ( $O_3$ ) exposure of the permanent monitoring plots (PMPs) of the CONECOFOR programme often exceeds the critical levels set to identify areas where vegetation can be considered at risk (KARLSSON *et al.* 2003b; GEROSA *et al.* 2003 this volume) (Figure 1). However, there is evidence that – especially under xeric environmental condition - concentration-based critical levels are not well related to the effect on vegetation, which is more closely related to the fraction of  $O_3$  that enters the plant (the  $O_3$  flux) (EMBESON *et al.* 2000a, b; GRULKE *et al.* 2003). Unfortunately, in the majority of cases the stomatal fluxes of  $O_3$  (and, thus, the dose of pollutant that reaches the plant) cannot be measured directly. To estimate the fluxes one normally uses models, generically called SVAT models (Soil-Vegetation-Atmosphere-Transfer models) (GRÜNHAGE *et al.* 2003). These models are usually data intensive and their data requirement needs to be examined in detail, especially when considering that routine monitoring programmes like the CONECOFOR were not *ad hoc* designed to address such a topic (see FERRETTI *et al.* 2003). Recently, two workshops were held under the

auspices of the UN/ECE Convention on Long-Range Transboundary Air Pollution (CLRTAP) to address the topics of  $O_3$  flux and the Level II approach to critical levels (ANONYMOUS 2002; KARLSSON *et al.* 2003a). Both workshops acknowledged the value of the flux approach and recommended implementation. Yet, it was also obvious that there are still uncertainties surrounding some aspects of the modelling (GRÜNHAGE *et al.* 2003) and questions about data requirements that may limit applicability to field situations. For example, BRAUN *et al.* (2003) argue that, in their experiment, "the calculation of  $O_3$  flux did not represent an advantage over AOT40 in explaining growth response" and this was partly due to the inherent difficulties in flux calculation. This paper will address the question of data requirements and the applicability of the flux approach to the CONECOFOR PMPs.

## Modelling $O_3$ uptake: an overview

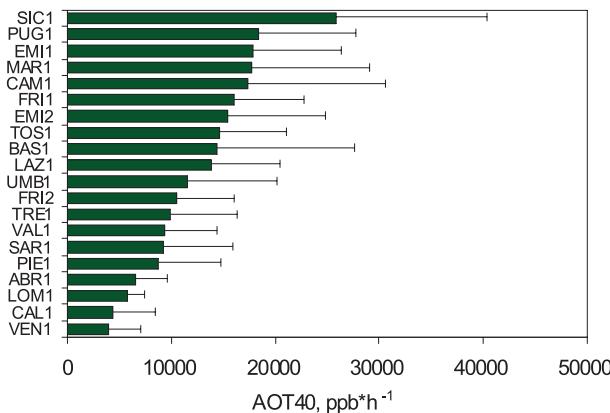
### Modelling $O_3$ uptake

It is necessary to distinguish between *diagnostic* and *prognostic* models (FINZI *et al.* 2001). Diagnostic

<sup>§</sup> Paper subject to review by members of the *ad-hoc* referees committee

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**Figure 1** – Estimated 1996-2000 mean AOT40 values for the CONECOFOR plots (after GEROSA *et al.* 2003, this volume). Error bars represent the standard deviation. See FERRETTI *et al.* (2003, this volume) for the location and characteristics of the plots.  
*Valori medi stimati di AOT40 per il periodo 1996-2000 alle aree CONECOFOR (da GEROSA *et al.* 2003, questo volume). Le barre di errore rappresentano la deviazione standard. Localizzazione e caratteristiche dei plot sono riportati da FERRETTI *et al.* (2003, in questo volume).*

models are characterized by a top-down approach: stomatal flux is derived directly from flux measurements (total fluxes of  $O_3$  and water) without resorting any assumptions on stomatal behaviour. They are not forecasting models but they provide information to parameterize and calibrate forecasting models. Prognostic models are, on the contrary, forecasting models, based on a bottom-up approach: either the stomatal and total fluxes are predicted by modelling the plant's physiological behaviour (especially the stomatal behaviour) through a suitable parameterisation and by using a relatively little set of meteorological input parameters. Some prognostic models describe the stomatal behaviour in leaves and then scale it up to represent the entire canopy (upscaling); others use data relating to a stand and thus describe the stomatal function of the canopy as a whole, the so-called "bulk models".

Despite differences in the details of parameterization, all these models follow the same potential-resistance electrical analogy in the description of fluxes (MONTEITH and UNSWORTH 1990; GARRATT 1994). This analogy is formulated based on the integration of the "turbulent diffusion" equation (Eq. 1) :

$$\text{Eq. 1} \quad \Phi = -K_c \cdot \frac{\partial \bar{C}}{\partial x_i}$$

where  $C$  is the mean  $O_3$  concentration and  $K_c$  the coefficient of turbulent "diffusion". This coefficient,

unlike what may be suggested by the formal analogy with molecular diffusion, is not constant, rather it varies along the  $x_i$  axis in response to several different factors. In the case of forest ecosystems, where  $x_i$  coincides with the vertical height from the ground,  $K_c$  increases with  $x_i$ , the speed of wind, surface roughness and temperature. Furthermore, within the equilibrium sublayer above the canopy, the constant relationship of fluxes to height implies that an increase of  $K_c$  is matched by a reduction of the  $O_3$  gradient. The constancy of fluxes in the surface atmospheric layer, or rather, in the equilibrium sublayer above the canopy, is an assumption that can be experimentally verified in the majority of cases. By definition, in this sublayer the measurement of the vertical flux  $F$  of a scalar entity at a height  $z_m$  above the canopy reflects the exchange at the surface ( $z = 0$ ) or at the top of the canopy.

Integrating Eq. 1 along the height, from the surface to a reference height  $z_{ref}$  we obtain:

$$\text{Eq. 2} \quad \Phi \cdot \int_0^{z_{ref}} \frac{1}{K_c} dz = C(z_{ref}) - C(0)$$

(flux  $\Phi$  appears outside the integration sign because of the hypothesis of the constancy of fluxes in the surface layer). The quantity under the integration sign, which has the physical dimensions of the inverse of a velocity (s/m), is defined total resistance  $R_{tot}(z_{ref})$ , and its reciprocal, a conductance, is the deposition velocity (m/s) introduced by CHAMBERLAIN (1953) :

$$R_{tot}(z_{ref}) = \int_0^{z_{ref}} 1/K_c dz \quad v_d(z_{ref}) = 1/R_{tot}(z_{ref})$$

Eq. 2 thus becomes

$$\Phi = \frac{C(z_{ref}) - C(0)}{R_{tot}(z_{ref})}$$

or

$$\Phi = v_d(z_{ref}) \cdot [C(z_{ref}) - C(0)]$$

where the analogy with Ohm's law ( $I = \Delta V/R$ ) is evident: the flux is the analogue of an electric current and the concentration of a potential. It is a very versatile analogy since the total resistance can be expressed as

the outcome of a combination of other resistances mounted in series or in parallel, each representing the different processes influencing the deposition flux. Yet, unlike electrical networks, here the resistances vary during the daytime and the values they express are a reflection of both the turbulent characteristics of the atmosphere and the activity of the vegetation.

### The big-leaf model

Although models with highly complex arrangement of the various resistances can be formulated, the most used model is the so-called *big leaf* model (Hicks *et al.* 1987) with a fairly simple subdivision of the  $R_{tot}$ . This model assumes that, as far as vertical exchanges of matter and energy are concerned, the entire canopy is considered as a single large leaf at a conceptual height  $z=d+z_{0x}$  above ground. In this model,  $d$  is the height of the canopy's zero plane ( $\approx 2/3$  of the height of the individual trees) and  $z_{0x}$  is the roughness length of the canopy for the transfer of a given scalar entity  $x$ , like momentum, heat and matter (Figure 2a). The conceptual height changes if one considers the fluxes of momentum, of sensitive (H) and latent heat (LE), and matter:  $d+z_0$  represents the height from the ground at which wind within the canopy (which acts as a sink for momentum) is 0, and in general differs from height  $d+z_{0H}$ , a lower height, from which evaporation and sensible heat fluxes appear to originate (source for H and LE, latent heat) or at which  $(d+z_{0O_3})$  the  $O_3$  concentration seems to disappear since the pollutant is absorbed by the vegetation (which acts as an  $O_3$  sink). Since the turbulent transport processes of heat and matter are so similar, the height  $d+z_{0O_3}$  is usually assumed to be identical to height  $d+z_{0H}$ .

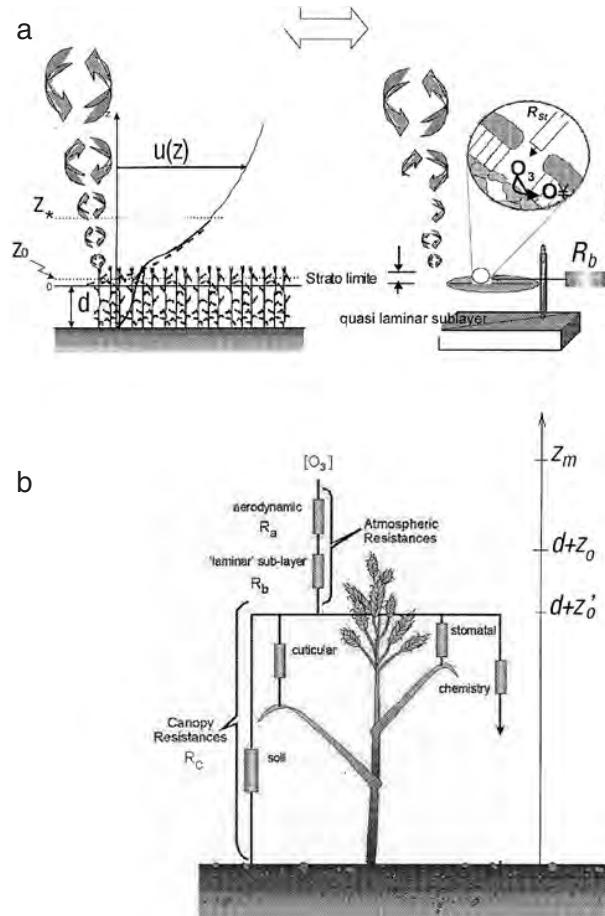
The differences observed in the turbulent transport of momentum and matter above permeable canopies suggest the existence of a thin layer, positioned between height  $d+z_{0H}$  and  $d+z_0$ , where transport cannot be turbulent since there is an absence of wind, but must necessarily be diffusive. For this reason it is called *quasi-laminar* or *viscous sublayer*. Total resistance to vertical  $O_3$  transfer consists of three main resistances, arranged serially:

$$R_{tot}(z) = R_a(d+z_0, z) + R_b + R_c$$

where  $R_a$  and  $R_b$  are two atmospheric resistances and  $R_c$  is the resistance of the whole exchange surface

(plant-soil system in the big leaf model).

$R_a(d+z_0, z)$  represents *aerodynamic resistance* encountered by  $O_3$  (but equally any scalar entity such as temperature, vapour concentration,  $CO_2$  or anything



**Figure 2 –** The resistance analogue principle – a) from the canopy to the Big Leaf model; - b) a resistance network, characterized by increasing complexity. The two profiles, *i.e.* the full and dotted lines, in Figure a) respectively represent the real wind profile above the stand and the theoretical wind profile obtained by logarithmic approximation using the Monin-Obukhov similarity theory. In all figures:  $d$  is the height of plane zero, *i.e.* the top of the canopy;  $z_0$  is the length of roughness for momentum;  $z_0'$  is the length of roughness for heat and gaseous exchanges (it is the homologue of  $z_{0H}$  and  $z_{0O_3}$ ); and  $z_m$  is the height at which measurement is performed. Lastly,  $R$  indicates the various different resistances to deposition (after PORG 1997).

*L'analogia resistiva – a) dalla copertura al Big Leaf model; - b) una rete resistiva a complessità crescente. I due profili a linea piena e tratteggiata in figura a) rappresentano rispettivamente il profilo di vento reale sopra il popolamento vegetale e quello teorico ottenuto come approssimazione logaritmica dalla teoria della similarità di Monin-Obukhov. In tutte le figure  $d$  rappresenta l'altezza del piano zero costituito dal top della canopia;  $z_0$  la lunghezza di rugosità per il momento;  $z_0'$  la lunghezza di rugosità per il calore e gli scambi gassosi (omologo di  $z_{0H}$  e  $z_{0O_3}$ ) e  $z_m$  l'altezza di misura. Le diverse resistenze alla deposizione sono indicate con  $R$  (da PORG 1997).*

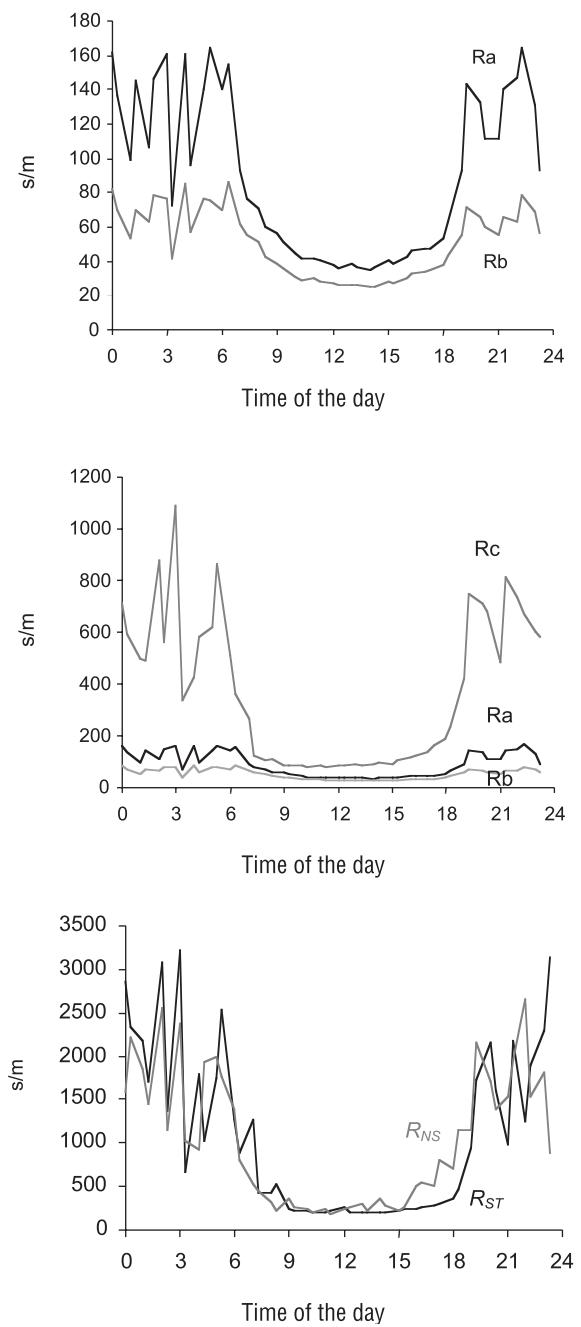
else) during the turbulent transport from height  $z$  to height  $d+z_0$  (the sink for momentum), and reflects the thermodynamic and mechanical features of the atmosphere, mainly atmospheric stability and wind.

$R_b$  is the overall resistance encountered by  $O_3$  in its diffusion through the quasi-laminar sublayer and thus depends on the molecular diffusivity of  $O_3$  in the air, and also to some extent on the intensity of the turbulence above. It is important to stress that, while the value of  $R_a$  is the same in each scalar entity, the value of  $R_b$  changes according to the scalar entity being considered: for example, if instead of  $O_3$  we examine the transport of another gas such as water vapour.

$R_c$  is the surface resistance or canopy resistance to  $O_3$  deposition; it includes all those processes connected to the influence of the plant-soil system on the vertical exchanges of  $O_3$ , and in particular  $O_3$  absorption by the vegetation or its destruction on the surfaces. It is strongly influenced by the physiological (e.g. stomatal functioning, water availability) and phenological (architecture, LAI) characteristics of the vegetation and the soil.  $R_c$  is 2-3 times greater than the two atmospheric resistances, signalling the important role played by vegetation and surfaces in the deposition process. The lesser relative importance of the two atmospheric resistances (Figure 3) explains why, despite different parameterizations of  $R_a$  and  $R_b$ , all authors end up by determining substantially similar flux values. In theory,  $R_c$  can be broken down into any number of parallel sub-resistances, each one representing a different deposition path. In the case of vegetation, a first, natural subdivision is that between stomatal ( $R_{st}$ ) and non-stomatal ( $R_{ns}$ ) components of the deposition, both relating to the canopy:

$$R_c^{-1} = R_{st}^{-1} + R_{ns}^{-1}$$

The stomatal resistance to  $O_3$  " $R_{st}$ " can also be seen as the sum of a real resistance against the diffusion of the gas operated by the stomata and another resistance to penetration into the mesophyll cells. Experimental evidence, however, suggests that the resistance by the mesophyll cells is very weak and is therefore often neglected (TINGY and TAYLOR 1982; LEUNING *et al.* 1979a, b; PLÖCHL *et al.* 1993). Another path of  $O_3$  penetration is through leaf cuticles, occurring in parallel to the



**Figura 3 –** An example of mean daily variation of canopy resistance values in a crop (Hordeum sp.):  $R_a$ ,  $R_b$ ,  $R_c$  e  $R_{st}$  e  $R_{ns}$ . See text for details.

*Esempio di variazione media giornaliera dei valori delle diverse resistenze (riferite alla canopy) in una coltura agraria (orzo):  $R_a$ ,  $R_b$ ,  $R_c$  e  $R_{st}$  e  $R_{ns}$ . Dettagli nel testo.*

stomatal path, which has equally been proved to be of negligible importance (LAISK 1989; KERSTIENS *et al.* 1992).

The non-stomatal resistance to  $O_3$  " $R_{ns}$ " can also be broken down into several parallel components,

so that different aspects can be considered, such as  $O_3$  destruction by outer plant surfaces ( $R_{EXT}$ ), soil deposition, or quantity consumed in chemical reactions:

$$R_c^{-1} = R_{ST}^{-1} + R_{EXT}^{-1} + R_{SOIL}^{-1} + R_{CHEM}^{-1}$$

$R_{EXT}$  to  $O_3$  is generally quite high and depends on the development of the plants (actual LAI): its value is usually around 750-1000 s/m (BALDOCCHI *et al.* 1987). It has been estimated that, in the case of agricultural crops, this deposition path influences less than 5% of the total  $O_3$  flux (MASSMAN 1993).

$R_{SOIL}$  is an integration of 3 other resistance mounted in series:  $R_{InCanopy}$  that is influenced by the aerodynamic effects within the canopy, a resistance to the crossing of the soil boundary layer ( $R_{bSoil}$ ) and a resistance that describes in an aggregate manner deposition on the soil and on any vegetation present.

Since  $O_3$  is not readily soluble in water, a moist soil generally presents a higher resistance to deposition than a dry soil, with an order of difference that has been defined as follows: 1000 s/m in a moist soil and 100 s/m in a dry one, according to WESELY and HICKS (2000), or 500 and 100 s/m according to ERISMAN *et al.* (1994), or even 600 and 400 according to BROOK *et al.* (1999).

#### Parameterization

Parameterization and the calculation of the individual resistances to  $O_3$  deposition derive directly from fluid dynamics equations and from the K-theory (or similarity theory, MONIN and OBHUKOV 1954). Different formulations can be provided for each resistance (for example, integral form and differential form), but they are always formally equivalent (cfr. the sources for the formulations, *e.g.* MONTEITH and UNSWORTH 1990; GRÜNHAGE *et al.* 2000; HICKS *et al.* 1987). Table 1 shows an overview of the most common parameterizations for resistances to  $O_3$  depositions. In brief,  $R_a$  is calculated integrating

**Table 1 –** An overview of the most common parameterization of resistance to ozone deposition. See text for details and symbols. (Continued)  
*Rassegna delle più comuni parametrizzazioni per le resistenze alla deposizione di ozono. Per il significato dei vari simboli si rimanda alla simbologia. (Continua)*

Resistance	Parameterization	Reference
$R_a$	$R_a = \int_{d+z_0}^{z_m} \frac{\Phi_H(\zeta)}{ku^* z} dz$ $\Phi_H(\zeta) = [1 + 5\zeta] \quad \text{se } \zeta = (z - d)/L > 0;$ $\Phi_H(\zeta) = [1 - 16\zeta]^{-1/2} \quad \text{se } \zeta \leq 0$	DYER 1974 CIESLIK 1998
	$R_a = \frac{1}{k \cdot u_*} \left[ \ln\left(\frac{z-d}{z_0}\right) - \Psi_H\left(\frac{z-d}{L}\right) + \Psi_H\left(\frac{z_0}{L}\right) \right]$ $\Psi_H(\zeta) = -5\zeta \quad \text{se } \zeta \geq 0; \quad \Psi_H(\zeta) = 2 \ln\left[\frac{1}{\Phi_H(\zeta)} - 1\right] \quad \text{se } \zeta < 0$	GRÜNHAGE and HAENEL 1997
$R_b$	$R_b = \frac{2}{k u^*} (Sc / Pr)^{2/3}$	All canopies
	ozone $Sc \approx 1.07; Pr \approx 0.72$	THOM 1972;
	$H_2O$ and heat $Sc/Pr \approx 0.9$	HICKS <i>et al.</i> 1987
	$R_b = \frac{1.45}{k u^*} Re^{0.24} Sc^{0.8}$	Forage meadows(Pastures) GARLAND <i>et al.</i> 1983
	$R_b = \frac{1.9}{k^2 u^*} (u^*)^{-2/3}$	Thin pasture MASSMAN 1993
	$R_b = \frac{7.3}{u^*} Re_*^{0.25} Sc^{0.5} - 5$	Rigid soils and surfaces CHAMBERLAIN 1984
$R_{ST, ozono}$	$R_{ST} = 1.65 \cdot R_{ST_{H2O}}$	<i>continue</i>

... cont'd

Resistance	Parameterization	Reference
$R_{STH2O}$ , evaporation	<b>Diagnostic models:</b> Penman-Monteith solved for $R_{STH2O}$	
	$R_{STH2O} = \frac{R_a + R_{bH2O}}{\gamma} \left[ \frac{\Delta \cdot (Rn - G) + \frac{\rho c_p VPD}{R_a + R_{bH2O}}}{\lambda E} - \Delta - \gamma \right]$	MONTEITH 1981
	with $VPD = [e_s(T) - e]$ $e_s(T) = 611 * \text{EXP}(17.269 * (T-273) / (T - 36))$ $\Delta = \frac{\partial e_s(T)}{\partial T} = e_s(T) * (17.269 * 237) / ((T - 36) ^ 2)$ possible correction $R_{STH2O} = R_{STH2O,PM} - \frac{R_b}{\gamma} \left( 1 - \frac{\Delta \cdot H}{\lambda E} \right)$	MURRAY 1967 THOM 1975
	Chamberlain's electric analogue: $R_{STH2O} = \frac{\rho c_p}{\gamma} \cdot \frac{e_s(T_0) - e_{zm}}{\lambda E} - R_a - R_{bH2O}$	MONTEITH, 1981; BALDOCCHI <i>et al.</i> 1987
	with $T_0 = \frac{H}{\rho c_p} (R_a + R_b) + T_{zm}$	
	<b>Prognostic models:</b> $R_{STH2O}^{-1} = R_{STH2O,MIN}^{-1} \cdot f_1(St) \cdot f_2(T) \cdot f_3(VPD) \cdot f_4(SM)$	JARVIS 1976; STEWART 1988
$R_{InCanopy}$ , ozone	$R_{InCanopy} = \frac{b LAI h}{u^*} + c$ b=14 e c=0 tall crops	VAN PUL and JACOBS 1994

the reciprocal of  $K_c$ , the turbulent diffusion coefficient for  $O_3$ , between height  $d+z_0$  and reference height  $z_m$ , and  $K_c$  is obtained by means of the K-theory:

$$K_c = \frac{k u^* z}{\Phi_c(\zeta)}$$

where  $k$  is the von Kármán coefficient,  $u^*$  is the friction velocity,  $z$  the height and  $\Phi_c(\zeta)$  is the non-dimensional Monin-Obukhov's similarity function accounting for the  $K_c$  dependance from the atmospheric stability conditions.

There are several different empirical calculations that can be used to establish  $R_b$  according to the different type of vegetation examined; the most commonly used is that suggested by THOM (1975) and by HICKS *et al.* (1987) (Table 1).

In diagnostic models, stomatal resistance to  $O_3$ ,  $R_{ST}$  is deduced from water fluxes, *i.e.* from stomatal resistance to evaporation  $R_{STH2O}$  weighted in relation

to different coefficients of molecular diffusion of  $O_3$  and vapour in the air.

In prognostic models,  $R_{STH2O}$  is calculated as a response to solar radiation, to temperature and to the balance of water in the atmosphere and in the soil, applying the Jarvis-Stewart physiological approach (JARVIS 1976; STEWART 1988). Stomatal conductance (the reciprocal of the corresponding stomatal resistance:  $R_{STH2O}^{-1}$ ) is modelled based on a theoretical maximum value, specific for the vegetation being considered, re-scaled according to the product of functions (with values between 0 and 1) that describe the limiting action being exerted on stomatal aperture by the range of environmental factors (Table 1).

In diagnostic models, the canopy's resistance to evaporation is obtained by the relative term contained in the Penman-Monteith equation (MONTEITH 1981), based on the energy balance on the surface/crop and in particular on the measurements of atmospheric moisture, temperature and latent heat flux. The formulation and derivation of the Penman-Monteith

equation can be found in good text books on agrometeorology (*e.g.* CECCON and BORIN 1995), while its solution for  $R_{STH2O}$  is shown in Table 1. If, as in the case of the original formulation of the Penman-Monteith equation,  $R_b$  is neglected and  $R_a$  is obtained through measurements of wind velocity ( $R_a$  for momentum) and not of heat fluxes, one may overestimate  $R_{ST}$  by 25-30% (CALLANDER and WOODHEAD 1981). For cases such as these, THOM (1975) has suggested a correction mechanism (Table 1).

An alternative diagnostic approach consists in deducing the stomatal resistance to evaporation using an electrical analogy, based on the different levels of moisture between the atmosphere and the crop and on the observed evaporation flux (Table 1). In the stomata the air is considered water saturated, but at a temperature that is different from the external temperature, and which must be estimated through the heat fluxes.

Constant values are usually attributed to resistances  $R_{EXT}$ ,  $R_{SOIL}$  and  $R_{CHEM}$ , although it is not always easy to choose between the wide range of values available in the literature.

For other resistances (*e.g.*  $R_{InCanopy}$ ,  $R_{bSoil}$ ) other formulations exist, with an higher degree of uncertainty and usually species-specific.

Resistances operated by the vegetation can be related to the entire canopy or to individual leaves: in the latter case, they must be scaled in relation to the LAI of the plant. If the results are to be even more realistic, they should be scaled to the portion of the LAI exposed to light (there are sophisticated energy transfer models that can be used to determine transfer within a canopy according to its architecture: see for example BALDOCCHI *et al.* 1987 e GRÜNHAGE *et al.* 2000). The simplest scaling procedure, which is also the least accurate, yields the value of canopy resistance dividing individual leaf resistance by total LAI or portion of LAI exposed to sunlight.

If one prefers, one can also describe all the formulae illustrated here in terms of conductances, defined as the reciprocals of their respective resistances, *i.e.*  $G = R^{-1}$ .

Once the network of resistances has been constructed (Figure 2), by applying Kirchhoff's law it is possible to determine the  $O_3$  concentration at each level and its flux. The flux is constant where resistances are mounted in series, whereas it is divided into different branches when the resistances are in parallel. In the

latter case, the  $O_3$  flux in each branch is the result of the ratio between  $O_3$  concentration at the beginning of the branch and the sum of the resistances downstream from that point. This yields the partition of the total flux and the determination of the quota of  $O_3$  that enters the stomata, *i.e.* the dose of  $O_3$  that the plant receives.

#### **Dual-source and multi-layer variants**

A first variant to big-leaf models is to distinguish between vegetation and soil sources/sinks of energy and matter, and thus examine them separately (SCHUTTLEWORTH and WALLACE 1985; MASSMAN 1992). This first variant can be applied in situations of open canopy, or scattered canopy, where the contributions of soil and vegetation to  $O_3$  deposition cannot be all lumped together in a single big leaf. Unlike the big-leaf approach, in the dual-source model the resistances of the laminar sublayer of the canopy ( $R_{bCanopy}$ ) do not implicitly include overall air resistance within the canopy ( $R_{InCanopyAir}$ ). The derivation of the latter has been discussed, for example, in MCNAUGHTON and VAN DEN HURK (1995).

A second variant consists in subdividing the vegetation into planes and adopting a big-leaf model for each plane, plus a plane for the soil. These models are called multi-layer models (*e.g.* MEYERS *et al.* 1998) and require an accurate description of the canopy's architecture as well as sophisticated sub-models to calculate wind profiles and radiation transfer within the canopy.

All models described above are one-dimensional models. Some three-dimensional models, intended primarily to be applied to closed and open forest stands (*e.g.* WANG and JARVIS 1990), have been developed, but they call for a very large number of input parameters. Descriptions of these models can be found in the literature (WANG and JARVIS 1990). In any case, GRÜNHAGE *et al.* (2000) conclude that, for the purpose of evaluating the  $O_3$  risk of a forest stand, since the time window involved is so wide (the entire growth season), simpler one-dimensional models such as Big-Leaf and Dual-Source appear to be more suitable.

#### **Modelling $O_3$ uptake: applicability to the CONECOFOR Permanent Monitoring Plots**

##### **Diagnostic or prognostic models?**

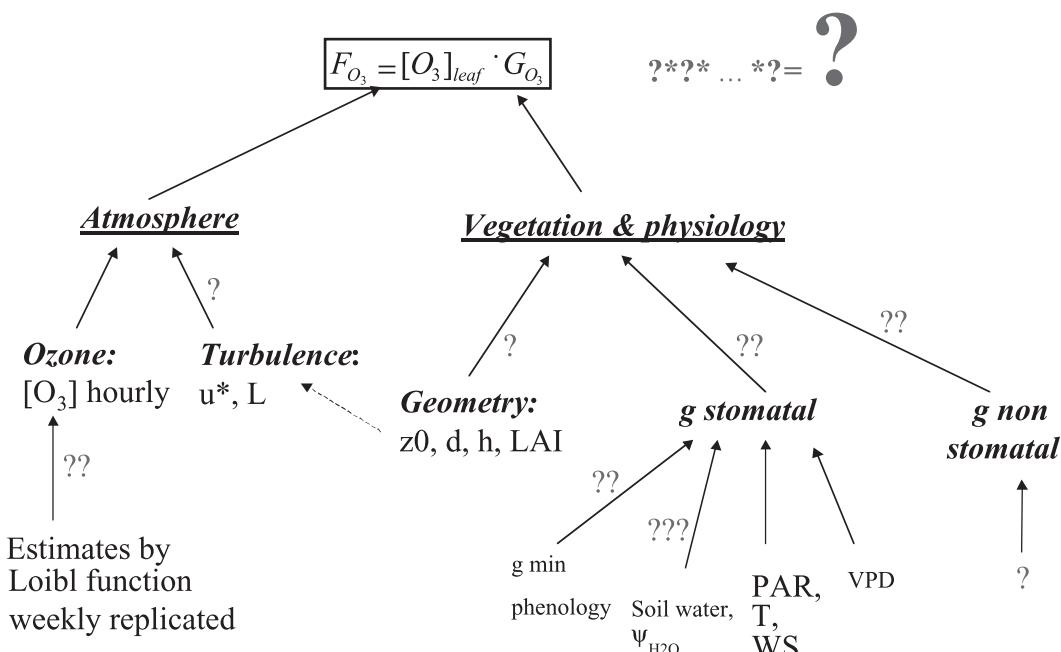
Diagnostic and prognostic models meet different needs and are therefore suitable for different

applications. Diagnostic models are used basically to analyze energy and matter (gaseous species) flux measurements performed with micrometeorological techniques above the different types of vegetation. The purpose of this type of model is essentially to meet research needs, since it enables researchers to investigate the dynamics of the various parameters involved and to determine the correct values for prognostic models' parameterisation. In particular these models are necessary for the determination of those functions that can limit stomatal conductance (the  $f$  functions of Jarvis-Stewart) that are used in prognostic models (GEROSA *et al.* 2003; EMBERSON *et al.* 2000b). The prognostic models, on the other hand, are more useful in practical application, and can provide evaluations and estimates on broad territorial scales. The objectives of a network of intensive monitoring like CONECOFOR by definition have a greater affinity to research purposes than to generic territorial assessment, and thus the application of a diagnostic model would appear to be more appropriate. It is indeed natural to expect that an intensive monitoring project should yield information that makes subsequent application possible (*e.g.*, provide correct parameter values to develop prognostic models suitable for Southern Europe

conditions). Unfortunately, over the period 1996-2000 (*i.e.*, the time window covered by the present I&C report), there were no direct measurements of fluxes, either of  $O_3$  or of water and energy in the PMPs and this makes it impossible to apply a diagnostic model. For this reason, the application of a prognostic model should be considered.

## *A possible modelling approach: description and data requirements*

A possible solution is to implement an extremely simple model, referring to the entire canopy, that takes into account the aggregate of the two paths of  $O_3$  deposition on the vegetation: stomatal and non-stomatal. Total flux of  $O_3$  between atmosphere and forest canopy is obtained as the product of the  $O_3$  concentration present at the top of the canopy ( $(O_3)_{d+z=0}$ ) and the conductance of the canopy itself; or rather the reciprocal of the canopy's  $O_3$  resistance (Figure 4). The former expresses the atmospheric features, including primarily photochemical production of  $O_3$  and turbulence, which causes  $O_3$  to be transported towards the surface, but also  $O_3$  deposition on the ecosystem as a whole, which influences the vertical gradient of  $O_3$  and ultimately the very concentration of



**Figure 4 –** Organization of the model, the parameters needed for implementation and the uncertainties. Number and size of question marks represent the degree of uncertainty. See text for the meaning of the symbols.  
*Organizzazione strutturale del modello, parametri necessari e livelli di incertezza. Numero e dimensioni dei punti interrogativi rappresentano i livelli di incertezza. Vedi il testo per il significato dei vari simboli.*

$O_3$  present in the canopy. The latter, on the other hand, is an expression of the features of the vegetation and its activity: it includes all those factors (climatic, biological, phenological, geometrical and chemical) that influence both stomatal uptake ( $g_{ST}$ ) and non-stomatal deposition ( $g_{NS}$ ). Stomatal conductance determines the fraction of  $O_3$  deposition more strictly linked to biological effects and the modelling of its behaviour; it is thus of crucial importance in any forecast of the  $O_3$  dose absorbed by the vegetation and any risk evaluation performed on such a basis.

Non-stomatal conductance, on the other hand, determines the quantity of  $O_3$  that is intercepted by non-transpiring plant surfaces and by the soil, with which it reacts destroying itself. The importance of this path of elimination of  $O_3$  was emphasized only recently (COE *et al.* 1995; TUOVINEN *et al.* 1999; GEROSA *et al.* 2003a) and any study that does not take it into consideration will inevitably produce a substantial overestimate of  $O_3$  fluxes and of expected risks. The model must therefore consist of three sub-models: an atmospheric one, a physiological one and a non-stomatal one. The atmospheric sub-model will describe  $O_3$  transport to the forest canopy as performed by turbulence and will need the availability at least of the following input parameters:  $u^*$ , hourly  $O_3$  concentrations measured at the top of the canopy.

The "physiological" sub-model, using a Jarvis-type model (1976), should describe the behaviour of foliar stomatal conductance and, after an appropriate upscaling, of canopy resistance  $R_c$  as well (*e.g.* EMBERSON *et al.* 2000a). In order to do this it will need input data relating to the quantity of solar radiation (PFD), the intensity of the wind, temperature, atmospheric moisture (VPD), water availability in the soil (SMD) or the water potential of the plant ( $\Psi_{H2O}$ ), phenology. Information on the geometry of the forest cover (LAI, height, roughness  $z_0$ , displacement height  $d$ ) will all be essential both for upscaling and for the configuration of the atmospheric model.

The "non-stomatal" sub-model, on the other hand, requires data on the concentration of any scavengers of  $O_3$ , such as for example NO produced by the activity of microbes in the soil or volatile organic compounds released by the vegetation itself, and resistances to  $O_3$  deposition in the soil and cuticles.

### Problems and uncertainty

#### Availability of meteorological and micrometeorological parameters

The parameters of atmospheric turbulence and stability, such as friction velocity  $u^*$ , the Monin Obukhov length  $L$ , and the flux of sensible heat  $H$ , indispensable if we are to calculate aerodynamic resistance  $R_a$  and quasi-laminar sublayer resistance  $R_b$  in the "atmospheric" sub-model, are not measured in any PMP (it would be necessary to dispose of *eddy covariance* measurements). Only a few plots (see AMORIELLO *et al.* 2003 this volume) envisage wind velocity and temperature measurements performed at two levels (for example, 2 and 10 m) that may enable us to estimate these parameters by repetition or with empirical procedures such as those suggested by VAN ULDEN and HOLTSLAG (1985). In this case, it is necessary to choose *a priori* the form of the function

$$\Phi_H(\zeta)$$

which describes the dependence of atmospheric turbulence on stability; and this further increases the uncertainty of the result of the assessment.

At the CONECOFOR PMPs, meteorological parameters are usually measured in a clearing and not above the canopy, as would be preferable for a correct assessment of the fluxes. This influences the turbulence parameters considerably, since it is well-known that the roughness of an herbaceous cover is markedly different from the roughness of a forest canopy (think for example of the logarithmic profile of wind). Measurements in the clearing also rules out the condition of horizontal homogeneity necessary to apply the similarity theory and to the establishment of flux constancy on which it is based.

#### Availability of hourly ozone concentration measurements from the top of the canopy

$O_3$  data requirements have been discussed by FERRETTI and GEROSA (2003). Problems arise in relation to the time resolution and location of measurements. At the CONECOFOR PMPs,  $O_3$  concentration is measured by passive sampling (BUFFONI and TITA 2003 this volume). This means that only mean weekly concentration data are available. This temporal scale is too broad to enable us to estimate the fluxes of  $O_3$ , since we know that  $O_3$  concentrations undergo a marked daily variation. In order to estimate  $O_3$  fluxes we would need at least hourly concentration measurements. It may be solved

by adopting the same technique used in estimating AOT40 (GEROSA *et al.* 2003 this volume). However, in this case, the uncertainties in the application of the Loibl function (GEROSA *et al.* 2003 this volume), would be much greater than for the estimation of AOT40 on a seasonal basis. The same measurements with passive samplers also imply another uncertainty because of their positioning which is not always ideal: outside the plot, sometimes quite far away or at different altitudes, and only in one case above the forest canopy (BUFFONI and TITA 2003 this volume). This last aspect is of crucial importance since  $O_3$  concentration displays a sensitive positive gradient above all kinds of vegetation cover. Passive samplers were placed at a standard height of 2m above ground. For the majority of agricultural crops this may be sufficient, but for forests it is necessary to estimate the  $O_3$  concentrations above the canopy which, in the case of CONECOFOR PMPs ranges from 9.9 to 29.1 m (mean height) (ALIANIELLO *et al.* 2003 this volume).

The problem of estimating  $O_3$  concentration at the height of the canopy, based on concentrations measured at a greater height, has already been addressed by several authors (*e.g.* GRÜNHAGE *et al.* 2001; TUOVINEN 2000; PLEIJEL 1998). But the procedure cannot be applied to the reverse situation, *i.e.* when you need to estimate the concentration at a height greater than the height at which the measurements were performed, since it is necessary to know the value of resistances to deposition and in particular of the surface resistance  $R_c$  which, as is well known, depends on the type of vegetation and on its physiological activity. The value of these resistances determines the profile of the  $O_3$  concentrations above a given vegetation cover. As a result, the concentration gradients above a meadow or above a clearing may differ considerably from those above a forest canopy. It is therefore preferable to dispose of actual measurements of  $O_3$  concentration above the canopy.

When, however, these data are not available, an approximate estimate of the  $O_3$  concentration at the top of the canopy ( $z_{top}$ ) can be obtained from the wind and temperature measurements performed a few metres away ( $z_m$ ), above a clearing, based on the calculation of the coefficient of turbulent diffusion of  $K_H$  heat and passing through the finite differences of the flux-profile equation (MONTEITH and UNSWORTH 1990):

$$K_H = \frac{k u_* z_m}{\Phi_H(\zeta)}$$

$$[O_3]_{ztop} = [O_3]_{zm} + (z_{top} - z_m) \cdot H / K_H$$

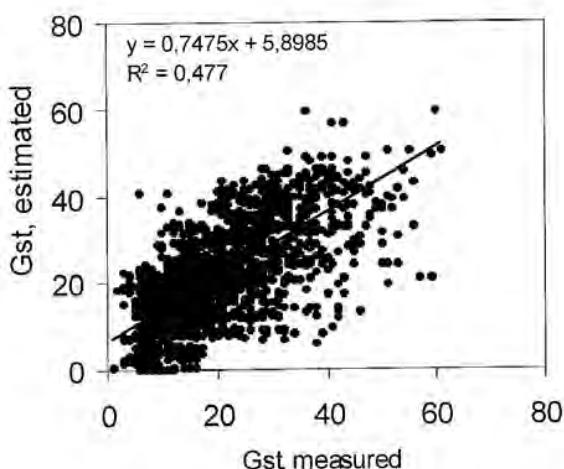
where  $H$  is the flux of sensible heat (which is in itself another crucial parameter that needs to be estimated),  $k$  is the von Kármán constant and  $\Phi_H(\zeta)$  is the Monin-Obukhov similarity function, with  $\zeta = (z/L)$ , the analytical formulation of which can be taken from DYER (1974).

An empirical approach to estimate  $O_3$  concentration at the top of the canopy can be based on circumstantial evidence and could consist in the use of empirical correction coefficients applied to the  $O_3$  concentration at the ground level. For example, the ratio between the concentration measured at canopy height and measurement height was reported to vary between 1.10 and 1.29 (BROADMEADOW, pers. comm.; GEROSA *et al.* 2001; KRAUSE *et al.* 2002), according to the height, the LAI, the canopy roughness and the wind velocity at the site being considered.

#### *Availability of a reliable parameterization of stomatal conductance*

A reliable parameterization of stomatal conductance is perhaps the most problematic aspect. First of all, any parameterization is highly species-specific and is therefore not very effective at describing mixed stands. In any case, even the spectrum of the main species growing in the CONECOFOR plots (Norway spruce, 4 PMPs; beech, 7 PMPs; Turkey oak, 5 PMPs; European oak, 1 PMP; sessile oaks and hornbeam, 1 PMP; holm oak, 2 PMPs) requires the use of several parameterizations. Unfortunately, the parameters of  $g_{max}$  (or  $R_{min}$ ) drawn from the literature are not always representative of the ecotypes and the environmental conditions of the CONECOFOR plots.

The determination of the limiting functions (Jarvis-Stewart's f functions) calls for the availability of continuous measurements of both conductances and of the environmental parameters considered. Among the latter, soil water content, a factor that can exert a markedly limiting function especially in the Mediterranean environment, is the one that would be most necessary, alongside the measurement of the plant's water potential. When attempts are made to make up for this lack of information by using estimates coming from the application of simple "bucket" sub-models to the different soil types and data on rainfall, temperature and wind, the result is merely to introduce further uncertainties into the overall model. Generally, the uncertainties associated with parameterization of stomatal conductance are fairly high. For example,



**Figure 5** - Comparison between measured and modelled stomatal conductance in adult Norway spruce trees located between 1000 and 2000 m a.s.l. in the Alps. Data are in  $\text{mmol m}^{-2} \text{sec}^{-1}$ .  
*Confronto tra dati misurati e modellati di conduttanza stomatica in piante adulte di abete rosso situate tra 1000 e 2000 m s.l.m. nelle Alpi. I dati sono espressi come  $\text{mmol m}^{-2} \text{sec}^{-1}$ .*

an attempt to model  $g_{st}$  was carried out for Norway spruce trees growing between 1000 and 2000 m asl in the Alps, a situation similar to Norway spruce PMPs of the CONECOFOR programme (see FERRETTI *et al.* 2003 this volume). The model (based on the Jarvis multiplicative model) was based on air temperature, time of the day, vapour pressure deficit and soil water content expressed as % of soil water holding capacity. Figure 5 reports the comparison between measured (1849 measurements carried out by different devices: PPSystem and ADC LCi) and modelled  $g_{st}$ . The portion of the variance explained is 47% and the considerable scatter of Figure 5 is a clear demonstration of the high degree of uncertainty surrounding estimates of  $g_{st}$ .

We further need to consider that this refers only to foliar stomatal conductance. Further uncertainties will be encountered when up-scaling to the whole canopy. Here, even the validation is problematic as we shall need to have flux measurements above the canopy, which require the use of sophisticated techniques such as eddy covariance. Upscaling from the individual leaf to the entire canopy thus remains an open problem.

Further problems are linked to phenology (leaf/needle unfolding, laminar distension, seasonal evolution of LAI, optimal function and ageing), which will need to be incorporated into the model, and other aspects related to the diversity of individual leaves,

to adaptation or resistance to other stress factors, primarily to water shortage.

#### Parameterization of non-stomatal deposition and stand geometries

Parameterization of non-stomatal deposition is still unclear in the case of forest stands, while some information is available in the case of agricultural crops (GEROSA *et al.* 2003a, b). It has been suggested that it may play an important role in forests as well, especially in environments that exert a limiting function on stomatal aperture, such as Mediterranean environments. Thus, a priority in research in this field is to obtain measurements that can help us understand the nature of these processes. On the other hand, some information about canopy geometry (*e.g.* LAI) is already available for the CONECOFOR plots.

#### Conclusions

Modelling stomatal uptake of O<sub>3</sub> is a complex matter. There are a large number of uncertainties - and of considerable importance - involving different measurements of input parameters as well as model parameterizations. Further, many approximations would be necessary and their propagation (additive or multiplicative propagation?) is unknown. For all these reasons, for the time being, it has been decided to stop attempt at applying a model for the estimation of O<sub>3</sub> fluxes at the CONECOFOR plots, since the results of such a model would not offer acceptable reliability and would not be verifiable at all.

In the near future, a process aiming at elaborating such a model would need first of all to strengthen intensive monitoring efforts, equipping at least some of the Level II plots with *eddy covariance* flux measurement and continuum analyzers recording hourly O<sub>3</sub> concentrations above the canopy. The recent incorporation in the network of the site BOL1 and the possible cooperation with other sites of the EUROFLUX project (see for example the site Collelongo, located close to the CONECOFOR site ABR1) will provide the chance for a step ahead. Furthermore, it would be important to extend and improve the focus of meteorological measurements. In this perspective, the data collected could contribute not only to a more accurate parameterization of flux models to be applied in CONECOFOR plots, but also be useful to the international scientific community (see GRÜNHAGE *et*

al. 2003) to validate large-scale models. This will help the study of those still unclarified aspects relating to the deposition of pollutants on vegetation in natural environments.

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# Ozone-like visible foliar symptoms at the permanent monitoring plots of the CONECOFOR programme in Italy<sup>§</sup>

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**Abstract** — Visible symptoms attributed to ozone on leaves of several plant species (trees, shrubs and herbs) were recorded in many sites across Europe. In Italy the programme for the assessment of ozone-like symptoms started in 2001 in the Level II beech monitoring plots. In 2002 the programme was implemented further and a total of 10 plots were included. The results were affected by the sampling design proposed by the European manual (which does not ensure site representativity) and by the time window of the survey. The onset and spread of symptoms depends onto site conditions (soil moisture) that enhance the stomatal uptake, light conditions and (most important) species composition, than on ambient levels of ozone.

**Key words:** *Leaf symptoms; Light Exposed Sampling Site; modifying factors; ozone.*

**Riassunto** — Sintomi fogliari visibili attribuibili all'ozono presso le aree permanenti del programma CONECOFOR in Italia. In molti siti europei sono stati osservati sintomi fogliari visibili su specie arboree, arbustive ed erbacee. In Italia la valutazione dei sintomi fogliari da ozono sulle aree di Livello II ha avuto inizio nel 2001. Nel 2002 il programma è stato implementato e sono state prese in considerazione complessivamente 10 aree. I risultati sono stati influenzati dal disegno di campionamento (che non assicura la rappresentatività a livello di sito) e dal periodo in cui l'indagine è stata effettuata. L'insorgenza e lo sviluppo dei sintomi dipendono dalle condizioni ecologiche della stazione che favoriscono l'assorbimento stomatico e della composizione specifica piuttosto che dai livelli di ozono.

**Parole chiave:** *Sintomi fogliari, Light Exposed Sampling Site, fattori modificanti, ozono.*

In 1997, the UN/ECE report on the first 10 years of monitoring activities assessing the effects of atmospheric pollution on forest conditions stated that in the future "ozone is the main pollutant that we need to consider" (MÜLLER-EDZARDS *et al.* 1997). For this reason, specific investigations, cofinanced by the European Union, were implemented within the Level II monitoring strategy. Investigations aimed at measuring ozone ( $O_3$ ) concentrations with passive samplers and at assessing  $O_3$ -induced visible foliar symptoms on spontaneous vegetation. An European common manual was developed and adopted in 2001 (ICP-FORESTS 2001, available also on [www.gva.es/ceam/ICP-forests/](http://www.gva.es/ceam/ICP-forests/)) and specific Training Courses are organized every year. Since 2001, field investigations for the detection of symptoms have been organized in a number of European countries. For example, in 2001 data were collected in Austria, France, Germany, Greece, Italy, Spain, Switzerland and United Kingdom.

Visible  $O_3$ -induced symptoms in plant species (tree, shrub and herb) have been described by a number of authors (SKELLY *et al.* 1987; FLAGLER 1998; INNES *et al.* 2001) and reported in Italy by COZZI *et al.*

(2000, 2001, 2002) and BUSSOTTI *et al.* (2003a). Conifers developed yellowish spots with ill-defined contours (chlorotic mottle), which in the most severe cases can degenerate into necrosis. Broadleaved trees develop a wide range of symptoms specially on the adaxial surface of the leaf, affecting the interveinal areas. According to the classification of KRUPA *et al.* (1998), acute injuries includes bleaching (small unpigmented necrotic spots), flecking (small necrotic areas), stippling (tiny punctuate spots where a few palisade cells are dead or injured, and may be white, black, red or red purple) and bifacial necrosis. Chronic injury consists in pigmentation (leaves turn red-brown to brown as pigments accumulate), chlorosis (due to chlorophyll breakdown) and premature senescence.

These responses are typical for oxidative stresses, but not specific for  $O_3$  (SCHRAUDNER *et al.* 1997; WOHLGEMUTH *et al.* 2002). Cell death is an hypersensitive response well known in relation to fungal and bacterial infections (HEATH 2000); whereas pigmentation (anthocyanins and phenolics) may be interpreted as a protective response against high light radiations (TATTINI *et al.* 2000; STEYN *et al.* 2000) and low site fertility (GUTSCHICK 1999). Because

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of the large range of uncertainty in evaluating symptoms on natural vegetation, they need to be confirmed and validated. The symptoms can be considered confirmed and validated once they have been reproduced in trials in controlled and/or semi-controlled environments (SKELLY *et al.* 1999; VANDERHEYDEN *et al.* 2001; NOVAK *et al.* 2003). A further validation criterion consists in using microscopy techniques to identify specific responses (GÜNTHARDT-GOERG *et al.* 2000; GRAVANO *et al.* 2002, 2003; VOLLENWEIDER *et al.* 2003).

The European visible symptom monitoring programme in Level II permanent plots was established in 2001 and is considered an optional action. It consists in two distinct phases:

- assessment of symptoms on the leaves of the Main Tree Species (MTS). This is performed every other year on a sub-sample of the leaves of upper-canopy branches collected for chemical nutrient analysis;
- assessment of symptoms on the Light Exposed Sampling Sites (LESS), *i.e.* on plants in sampling plots that do not grow under the forest canopy, adjacent to Level II plots, *i.e.* forest edge. This investigation is performed annually. For the purposes of this survey, preference is given to plots already included in meteorological Level II surveys and equipped with passive  $O_3$  samplers.

The common manual and the  $O_3$  symptom photo-documentation can be found at the following website: <http://www.gva.es/ceam/ICP-forests/>. Further pictorial atlases were published by SKELLY *et al.* 1987; FLAGLER 1998; INNES *et al.* 2001; SANZ *et al.*, 2002; SCHAUB *et al.* 2002.

## Methods

Two surveys were conducted in Italy in 2001 and 2002. The first was carried out between the end of July and the end of August 2001 on a selection of plots, for the most part located in beech forests: ABR1, CAL1, CAM1, EMI2, LOM3, PIE1, TOS3 and VEN1 (Fig. 1; for details see FERRETTI *et al.* 2003a this volume), chosen for the following reasons:

- among all the species represented in Level II plots, beech is the only one that is more or less uniformly distributed in a North-South gradient;
- among Level II MTS in Italy, beech is the only species in which  $O_3$  symptoms have been reported and validated (even if beech can be considered less sensitive than other forest tree species, VANDERHEYDEN *et al.* 2001);

- beechwoods in Italy, due to their specific ecological conditions (soil moisture), can be considered potentially more sensitive to  $O_3$  than other ecosystems;

- many of the species (both woody and herbaceous) belonging to beechwoods vegetation are known to be  $O_3$  sensitive.

The survey was conducted both on the MTS and on a LESS, identifying one in each Level II plot; MTS foliar assessment was performed at the same time as samples were collected for chemical nutrient analysis in all plots except EMI2, where leaves were sent to the Department of Plant Biology at the University of Firenze. Site selection criteria followed in determining the LESS are given in Table 2. A second survey was conducted in 2002, only on LESS. The following permanent plots were considered: CAM1, CAL1, LOM3, TOS3 and VEN1 using the same LESS as in 2001, integrated with additional new LESS in the PMPs in LOM3 and VEN1 (Fig 1). Note that EMI1 and TOS1 were not present in the



Figure 1 - The location of the Permanent Monitoring Plots of the CONECOFOR programme (after Ferretti *et al.* 2003a, this volume). Circles: PMPs operational within the programme since 1995; squares: PMPs that have joined the programme at a later stage. The PMPs considered for the present study were: ABR1, CAL1, CAM1, EMI2, LOM3, PIE1, TOS1, TOS3 and VEN1.  
*Localizzazione delle aree permanenti di monitoraggio del programma CONECOFOR. I cerchi indicano le aree permanenti operative all'interno del programma sin dal 1995; i quadrati indicano le aree permanenti che sono state incorporate successivamente. Le aree permanenti considerate nel presente studio sono: ABR1, CAL1, CAM1, EMI2, LOM3, PIE1, TOS1, TOS3 and VEN1.*

Table 1 – Position, topographical features and species composition of the Light Exposed Sampling Sites.

\* Species considered sensitive according to the pan-European list at [www.gva.es/ceam/ICP-Forests/](http://www.gva.es/ceam/ICP-Forests/)

*Localizzazione, posizione topografica e composizione specifica dei siti LESS.*

\* Specie considerate sensibili secondo la lista pan-Europea disponibile presso [www.gva.es/ceam/ICP-Forests/](http://www.gva.es/ceam/ICP-Forests/)

PMP	Altitude m asl	Water availability	Orientation	Species present in the LESS	
				Species considered sensitive*	Other species
ABR1	1450-1500	Moist	South West	<i>Fagus sylvatica</i>	<i>Digitalis micrantha, Verbascum spp.</i>
CAL1	950-1000	Moist	West	<i>Fagus sylvatica, Rubus spp., Crataegus monogyna</i>	<i>Pteridium aquilinum, Euphorbia amygdaloides, Vinca minor, Hedera helix, Cytisus scoparius</i>
CAM1	1150-1200	Moist	North West	<i>Fagus sylvatica, Rubus spp.</i>	<i>Cyclamen hederifolium, Digitalis micrantha, Daphne laureola, Galium spp., Pteridium aquilinum, Viola reichembachiana</i>
EMI1	200	Dry	South	<i>Fraxinus ornus Prunus spinosa, Prunus avium, Rubus spp.</i>	<i>Castanea sativa, Corylus avellana, Quercus cerris, Quercus petraea, Rosa canina, Ulmus minor, Juglans regia, Plantago lanceolata, Vinca minor</i>
EMI2	950-1000	Arid	North East	<i>Prunus spinosa</i>	<i>Crataegus oxyacantha</i>
LOM3	1200-1250	Moist	East	<i>Acer pseudoplatanus, Betula pendula, Fagus sylvatica, Fraxinus excelsior, Laburnum alpinum, Rosa canina, Rubus idaeus, Salix caprea, Stachys spp., Centaurea nigra</i>	<i>Rosa canina, Astrantia major, Cirsium spp., Gentiana spp., Geranium nodosum, Helleborus niger, Mycelis muralis, Petasites spp., Veronica urticifolia</i>
LOM3 LESS 2				<i>Fagus sylvatica, Acer pseudoplatanus, Viburnum lantana</i>	<i>Alnus incana, Salix glabra, Lonicera sp.</i>
PIE1	1150-1200	Moist	West	<i>Picea abies</i>	<i>Vaccinium vitis idaea, Pteridium aquilinum, Gentiana kochiana, Quercus petraea</i>
TOS1 LESS 1	110-115	Dry	South East	<i>Robinia pseudoacacia, Vitis vinifera, Rubus sp.</i>	<i>Clematis vitalba</i>
TOS1 LESS 2	110-115	Moist	North	<i>Ailanthus altissima, Rubus sp.</i>	<i>Arum italicum, Beta vulgaris, Clematis vitalba, Hedera helix, Ligustrum vulgare, Lythrum salicaria, Mentha arvensis, Pteridium aquilinum</i>
TOS3	1100-1150	Moist	North East	<i>Acer pseudoplatanus, Fagus sylvatica, Rubus idaeus, Rubus ulmifolius</i>	<i>Abies alba, Alnus cordata, Daphne mezereum, Ilex aquifolium, Pinus nigra, Quercus cerris, Euphorbia amygdaloides, Geranium robertianum, Potentilla micrantha, Prenanthes purpurea, Pteridium aquilinum, Sanicula europaea, Senecio fuchsii, Silene dioica, Viola reichenbachiana</i>
VEN1 LESS 1	1100-1150	Moist	South	<i>Fagus sylvatica, Picea abies, Rubus spp., Salix caprea, Sambucus racemosa, Lamium spp.</i>	<i>Abies alba, Cirsium spp., Dactylis glomerata, Daphne spp., Fragaria vesca, Lonicera sp., Petasites alba</i>
VEN1 LESS 2	1400-1450	Dry	South	<i>Acer pseudoplatanus, Fagus sylvatica, Fraxinus excelsior, Picea abies, Rubus sp.</i>	<i>Ostrya carpinifolia, Sambucus racemosa, Rosa canina</i>

2001 survey. Characteristic features of the LESS are given in Table 1.

## Results

### Symptoms on MTS and LESS

The main findings are shown in Table 3. Ozone-induced symptoms in the MTS (beech) were observed only in 1 plot (VEN1). These symptoms consisted in brownish interveinal stippling only on the adaxial surface of the leaf. All samples, from all plots, displayed injuries caused by other agents, both abiotic (hail) and biotic (both insects as *Thyspocyba cruenta*, *Mikiola fagi* and fungi as *Apiognomonia* spp.).

In the LESS, symptomatic species were observed in VEN1 and LOM3 in 2001 and 2002, in TOS1 and TOS3 only in 2002 (Table 3). Symptoms were validated based

on descriptions available in the literature (HARTMANN *et al.* 1995; HANISCH and KILZ 1990; BERGMANN *et al.* 1999; INNES *et al.* 2001; SANZ *et al.* 2002; MANNING *et al.* 2002) and on website (<http://www.gva.es/ceam/ICP-forests/>; <http://www.ozone.wsl.ch>). In other cases, reference is made to similar species, belonging to the same genus. For example, in *Centaurea nigra* the symptoms are similar to those described on *Centaurea jacea* (BUNGENER *et al.* 1999) and *Centaurea paniculata* (SKELLY *et al.* 1999). Several symptomatic species were also found near the LESS, but not within the defined perimeter. In LOM3, on 23 July 2001, O<sub>3</sub>-like injuries were observed only on the herb species *Astrantia major*, *Helleborus niger* and *Centaurea nigra*. On the date of the second observation, 24 August, during the “*2<sup>nd</sup> ICP-Forests Training Course on the Assessment of Ozone Injury*” other woody species

Table 2 – Selection criteria followed for the Light Exposed Sampling Sites (LESS).  
 Criteri selettivi per l'individuazione dei siti LESS.

Criteria	Best option	Second option	Third option
Position	Forest edge, near a passive ozone sampler and meteo station	Forest clearing >0,2 ha, near the instrumentation	Forest clearing >0,5ha, far from the instrumentation
Distance from a Level II plot	<3Km, difference in altitude <100m	<3Km, difference in altitude 100m -300m	<3Km, difference in altitude 100m -300m
Exposure	Full light exposure, same exposure as Level II plot	Full light exposure	Full light exposure
Species composition	Presence of MTS as established regeneration and/or rootsuckers	Absence of MTS, but presence of ozone-sensitive species included in the European list	Absence of MTS and of species in the European list
Presence of disturbances	Absence of roads used by motor vehicles; Absence of disturbing elements such as animal grazing, silvicultural and agricultural practices, land movements	Absence of roads used by motor vehicles; Absence of disturbing elements such as animal grazing, silvicultural and agricultural practices, land movements	Absence of roads used by motor vehicles; Absence of disturbing elements such as animal grazing, silvicultural and agricultural practices, land movements

were diagnosed as symptomatic by course participants from other countries: *Fagus sylvatica* (European beech), *Rubus idaeus* (raspberry bush), *Rosa canina* (bramblerose), *Salix glabra* (willow), *Fraxinus excelsior* (European ash), *Laburnum alpinum* (Scotch laburnum), *Acer pseudoplatanus* (sycamore maple). In 2002 the assessment at LOM3 was done more than a month later (10 Sept.) and several species were found as symptomatic (Table 3). Table 4 describes the kind of symptom found on each species; Table 5 lists the species (more numerous!) found as symptomatic at the same PMPs, but outside the LESS.

A limitation affecting the interpretation of the data is that no estimate of AOT40 values was done for the years 2001 and 2002 (see GEROSA *et al.* 2003 this volume). However, the relationships between the mean AOT40 of the period 1996-2000 and the values for each individual year in the same period are acceptable (mean  $R^2=0.54$  when comparing the individual years with the average figure relating to the remaining years). As an example, Fig. 2 shows the relationship between mean values 1996-1999 and the value for the year 2000 ( $R^2 = 0.62$ ) for each PMP. It is thus reasonable to assume that sites with high average AOT40 values over the period 1996-2000 tend to have high values also in the years 2001 and 2002. If this can be considered a reasonable assumption, Table 1 shows that symptoms on MTS occur at the site with the lowest estimated exposure (VEN1). Interestingly, this is the same PMP that also shows unexpected reduced basal area increment (BAI) at low exposure and that provides the datapoints that determine most of the crown transparency trend reported for beech (FERRETTI *et al.* 2003b this volume). On the basis of the data reported in Table 3, no direct

relationship is obvious between estimated mean AOT40 (June-September) and symptom expression. However, the increase in the number of symptomatic species occurred between 2001 and 2002 at LOM1 and VEN1 is consistent with the change of the assessment date (assessment in 2002 were carried out 40-50 days later than in 2001) which ultimately means that plants were subjected to an higher cumulated exposure in 2002.

## Discussion

The problems and uncertainties linked to the surveys of  $O_3$  symptoms were reviewed by FERRETTI and Cozzi (2002). The first experience in Italy on a national

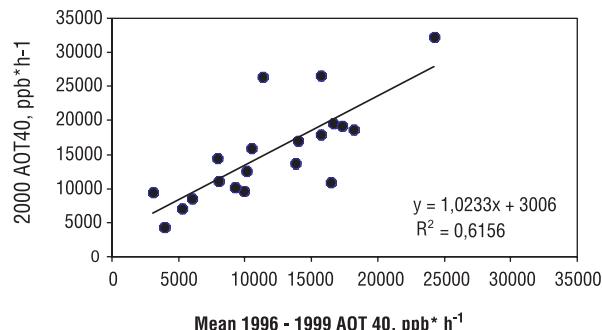


Figure 2 – Relationship between mean 1996-1999 AOT40 for each CONECOFOR Permanent Monitoring Plot and values for the year 2000. The mean  $R^2$  value for the various combination of individual years and the average of the remaining years (e.g., average 1997-2000 vs. 1996; average 1996, 1998, 1999, 2000 vs. 1997 and so on) is 0.54.

Relazione tra i valori medi di AOT40 per il periodo 1996-1999 ed il valore 2000 alle varie aree permanenti. Ogni punto rappresenta un'area permanente. Il valore medio di  $R^2$  calcolato per le varie combinazioni di medie pluriennali e valori annuali (es. media 1997-2000 vs. 1996; media 1996, 1998, 1999, 2000 vs. 1997 e così via) è 0,54.

basis confirms these concerns. Assessment of symptoms on MTS leaves is difficult to be implemented on all Level II plots, since the majority of plots are made up of species whose symptomatology is unknown or not yet validated experimentally. Species in European Level II plots whose symptomatology are known, and are been experimentally validated, are just Aleppo pine, beech and birch. Spruce, although considered sensitive (cfr. SANDERMANN and WELLBURN, 1996; symptoms are also shown in [www.gva.es/ceam/ICP-forests/](http://www.gva.es/ceam/ICP-forests/)), displays a fairly dubious symptomatology. Among all the species represented in Level II plots only beech is unanimously considered sensitive, although the wide range of symptoms documented in the literature (bronzing and stippling, cfr. INNES *et al.* 2001; SANZ *et al.* 2002) may give rise to uncertainties in field observation. Particularly, bronzing may be caused from high UV radiations (HARTMANN *et al.* 1995).

One of the most important question is about the value and relevance of MTS and LESS data. The selection of the LESS on the basis of the European common manual lead to an unreplicated, non-statistically selected sampling site, which, by definition, is representative of itself only. Previous investigations (COZZI *et al.* 2001,

2002) provide clear evidences that  $O_3$  like symptoms have an high variability within the same species even at local (meters) scale. Similar problems arise with symptoms on MTS, as MTS trees are not selected according to a statistical design. Thus, data provided by the current LESS and MTS are not representative for the PMP site and do not allow formal comparisons neither in space (between PMPs) nor in time (between two assessment occasions). Under these condition, even the comparison between symptoms (either on MTS and LESS vegetation) and  $O_3$  exposure risks to provide biased results. Quantitative, comparable informations can be obtained by selecting an adequate number of observation site according to sound statistical procedures (FERRETTI and EHRARDT 2002b; COZZI *et al.* 2001). For this reason, we suggest that the sampling system adopted for surveys on LESS and MTS need to be reconsidered.

The methodology followed envisages an assessment of the species considered sensitive (according to a pan-European list, <http://www.gva.es/ceam/ICP-forests/>) present in the LESS; but not all these species display clear and experimentally validated symptoms. The major problems arise among the herbs: herbs are probably more sensitive to  $O_3$  (cfr., for example, BERGMANN *et al.*

**Table 3 -** Survey 2001 – 2002 on  $O_3$ -like foliar symptoms on MTS (Main Tree Species) and LESS (Light Exposed Sampling Site) in Italy. Symptomatic species. For the species composition of LESS, see Table 1. For the AOT40 concept see Gerosa *et al.* (2003 this volume).  
*Indagini 2001 – 2002 sui sintomi fogliari attribuibili ad  $O_3$  in MTS (specie principale del plot) e LESS (sito di campionamento esposto alla luce). Specie sintomatiche. Per la composizione specifica del LESS, vedi Tab. 2. Per i dettagli su concetto e calcolo di AOT40 vedi Gerosa *et al.* (2003, in questo volume).*

PMP	Survey Date	Year 2001		Year 2002		Estimated AOT40 June-September, 1996-2000
		MTS Symptoms	LESS Symptomatic species	Survey Date	LESS Symptomatic species	
ABR1	August, 21	None	None	Not assessed	Not assessed	6546
CAL1	August, 28	None	None	September, 5	None	4389
CAM1	August, 30	None	None	August, 28	None	17291
EMI1	Not assessed	Not assessed	Not assessed	September, 17	None	17887
EMI2	August, 1	None	None	Not assessed	Not assessed	15384
LOM3	July, 23	None	<i>Astrantia major, Centaurea nigra, Helleborus niger</i>	September, 10	<i>Fagus sylvatica, Fraxinus excelsior, Acer pseudoplatanus, Laburnum alpinum, Rosa canina, Rubus idaeus, Astrantia major, Centaurea nigra, Geranium nodosum, Mycelis muralis, Veronica urticifolia</i>	N.A.
PIE1	July, 27	None	None	Not assessed	Not assessed	8686
TOS1	Not assessed	Not assessed	Not assessed	September, 23	<i>Ailanthus altissima, Beta vulgaris, Rubus sp., Vitis vinifera,</i>	14632
TOS3	July, 21	None	None	July, 22	<i>Rubus ulmifolius</i>	N.A.
VEN1	August, 29	Yes	<i>Lamium spp., Rubus spp.</i>	September, 10	<i>Acer pseudoplatanus, Fagus sylvatica, Picea abies, Sambucus racemosa</i>	4004

**Table 4 –** Species symptomatic in the LESS (2001 and 2002 survey), with description of the symptoms. The column “Listed” indicates if that species is listed (Y) or not (N) in the pan-European list of the sensitive species; (G) indicates when species of the same genus are considered sensitive by the list.

*Specie risultate sintomatiche nel LESS (indagini 2001 e 2002) e descrizione dei sintomi. La colonna “Listed” indica se la specie è elencata (Y) o no (N) nella lista pan-Europea delle specie sensibili; G indica invece se altre specie dello stesso genere sono indicate nella lista.*

Species	Listed	Kind of symptom
<i>Acer pseudoplatanus</i>	Y	Interveinal red stippling and/or browning and reddening
<i>Ailanthus altissima</i>	Y	Small necrosis white ivory and/or red stippling
<i>Astrantia major</i>	N	Interveinal decoloration of the upper leaf surface
<i>Beta vulgaris</i>	N	Interveinal reddening of the upper leaf surface
<i>Centaurea nigra</i>	G	Interveinal reddening of the upper leaf surface
<i>Fagus sylvatica</i>	Y	Brown stippling on the upper leaf surface
<i>Fraxinus excelsior</i>	Y	Dark stippling and browning on the upper leaf surface
<i>Geranium nodosum</i>	G	Interveinal decoloration of the upper leaf surface
<i>Helleborus niger</i>	N	Interveinal decoloration of the upper leaf surface
<i>Laburnum alpinum</i>	Y	Interveinal browning of the upper surface
<i>Lamium spp.</i>	N	Interveinal decoloration of the upper leaf surface
<i>Mycelis muralis</i>	Y	Small whitish necrosis on the upper leaf surface
<i>Picea abies</i>	Y	Chlorotic mottle in needles >1 year old
<i>Rosa canina</i>	Y	Dark stippling and browning on the upper leaf surface
<i>Rubus idaeus</i>	Y	Interveinal reddening of the upper leaf surface
<i>Rubus spp.</i>	Y	Interveinal reddening of the upper leaf surface
<i>Sambucus racemosa</i>	Y	Dark stippling on the upper leaf surface
<i>Veronica urticifolia</i>	Y	Dark stippling and browning on the upper leaf surface
<i>Vitis vinifera</i>	Y	Red-brown stippling on the upper leaf surface

1999; MANNING *et al.* 2002), but they are also greatly more sensitive to environmental stress. Further, herbs have only rarely been considered in cause-effect experiments. The numerous “new findings” listed in Tables 4 and 5 among the herbaceous species suggest that much work is needed to understand the symptom-response of herbs under elevated ambient O<sub>3</sub> concentrations. Time window is another key issue: the different results obtained at LOM3 (and at a lesser extent at VEN1) in 2001 and 2002 may be largely explained with the different assessment date. Lastly, BUSSOTTI *et al.* (2003b), reporting the findings of the 2<sup>nd</sup> ICP-Forests Training Course on the Assessment of Ozone Injury, highlighted the considerable level of uncertainty existing between observers in identifying and quantifying O<sub>3</sub>-like symptoms.

## Conclusions

The surveys carried out in 2001 and 2002 have shown the presence of symptoms attributable to O<sub>3</sub> in several plots of the CONECOFOR network and on a variety of species. Barring a few exceptions, these symptoms are of limited intensity. In particular, symptoms on adult beech trees were found only in VEN1; here, according to the data available, the levels of O<sub>3</sub> appear to be lower, but there are presumably ecological conditions such as soil moisture and canopy structure that may favour

a greater uptake (ALIANIELLO *et al.* 2003; SCHAUB *et al.* 2003). SMITH *et al.* (2003) reporting the data of the US bioindicator network observed that in dry years, even with high O<sub>3</sub> levels, the occurrence of symptoms was lower than in moist years. The surveys enabled researchers to highlight some problems in the methodology related to symptom identification, to the characteristic features of the species and to the currently applied sampling and assessment methods. The Italian system used to detect O<sub>3</sub> symptoms in and around the CONECOFOR PMPs is therefore being re-elaborated to take into consideration the problems mentioned above. The aspects on which attention needs to be focused are:

- selection of PMPs according to a risk assess-

**Table 5 –** Species find as symptomatic out the LESS (2001 and 2002 survey), with description of the symptoms. The column “Listed” indicates if that species is listed (Y) or not (N) in the pan-European list of the sensitive species; (G) indicates when species of the same genus are considered sensitive by the list.

*Specie sintomatiche identificate fuori dai LESS (indagini 2001 e 2002), e descrizione dei sintomi. La colonna “Listed” indica se la specie è elencata (Y) o no (N) nella lista pan-Europea delle specie sensibili; G indica invece se altre specie dello stesso genere sono indicate nella lista.*

Species	Listed	Kind of symptom
<i>Alchemilla vulgaris</i>	G	Dark stippling on the upper leaf surface
<i>Acer platanoides</i>	Y	Interveinal browning of the upper surface
<i>Aruncus dioicus</i>	Y	Interveinal browning and stippling of the upper surface
<i>Atropa belladonna</i>	Y	Whitish necrosis on the upper leaf surface
<i>Cardamine heptaphylla</i>	N	Dark stippling and browning on the upper leaf surface
<i>Carpinus betulus</i>	Y	Dark stippling and browning on the upper leaf surface
<i>Centaurea nigrescens</i>	G	Interveinal reddening of the upper leaf surface
<i>Clematis vitalba</i>	Y	Interveinal reddening of the upper leaf surface
<i>Cornus mas</i>	Y	Interveinal reddening of the upper leaf surface
<i>Corylus avellana</i>	Y	Interveinal browning of the upper surface
<i>Crataegus monogyna</i>	Y	Dark stippling on the upper leaf surface
<i>Euforbia dulcis</i>	Y	Interveinal decoloration of the upper surface
<i>Eupatorium cannabinum</i>	N	Dark stippling on the upper leaf surface
<i>Fraxinus ornus</i>	Y	Dark stippling and browning on the upper leaf surface
<i>Heracleum sphondylium</i>	N	Dark stippling and browning on the upper leaf surface
<i>Lapsana communis</i>	Y	Dark stippling and browning on the upper leaf surface
<i>Lonicera caprifolium</i>	Y	Interveinal reddening of the upper leaf surface
<i>Mercurialis perennis</i>	N	Dark stippling on the upper leaf surface
<i>Ostrya carpinifolia</i>	Y	Dark stippling on the upper leaf surface
<i>Parthenocissus quinquefolia</i>	Y	Interveinal reddening of the upper leaf surface
<i>Pinus halepensis</i>	Y	Chlorotic mottle in needles >1 year old
<i>Prunus avium</i>	Y	Interveinal reddening of the upper leaf surface
<i>Prunus spinosa</i>	Y	Dark stippling and browning on the upper leaf surface
<i>Robinia pseudacacia</i>	Y	Dark stippling on the upper leaf surface
<i>Rumex alpinum</i>	G	Interveinal reddening of the upper leaf surface
<i>Rumex sanguineus</i>	G	Interveinal reddening of the upper leaf surface
<i>Salix alba</i>	S	Dark stippling and browning on the upper leaf surface
<i>Salvia glutinosa</i>	N	Interveinal decoloration of the upper leaf surface
<i>Sambucus ebulus</i>	Y	Dark stippling on the upper leaf surface
<i>Scrophularia nodosa</i>	N	Dark stippling and browning on the upper leaf surface
<i>Sorbus aucuparia</i>	Y	Dark stippling and browning on the upper leaf surface
<i>Thalictrum minus</i>	N	Dark stippling on the upper leaf surface
<i>Tilia cordata</i>	Y	Dark stippling on the upper leaf surface
<i>Ulmus glabra</i>	Y	Dark stippling and browning on the upper leaf surface
<i>Ulmus minor</i>	Y	Dark stippling and browning on the upper leaf surface
<i>Urtica dioica</i>	N	Small whitish necrosis on the upper leaf surface
<i>Valeriana officinalis</i>	N	Dark stippling and browning on the upper leaf surface
<i>Veronica urticifolia</i>	Y	Dark stippling and browning on the upper leaf surface
<i>Viburnum lantana</i>	Y	Interveinal reddening of the upper leaf surface

ment ( $O_3$  concentrations, presence of sensitive species, ecological conditions that may favour foliar  $O_3$  uptake);

- design the monitoring to obtain quantitative data at each Level II plot;
- preliminary survey of the composition of the flora on the forest edge and in open field areas, so as to identify the most sensitive species among the ones present (bioindicators);
- validation of symptoms by means of experiments in controlled environments and microscopy observations.

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# Ozone exposure, crown transparency and basal area increment at the permanent monitoring plots of the CONECOFOR programme in Italy<sup>§</sup>

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**Abstract** — Ozone ( $O_3$ ) is known to affect a number of plant and ecosystem functions and processes. The data collected at the monitoring plots of the CONECOFOR programme may only enable evaluation of effects on tree crown condition and growth. Analysis is however difficult due to the nature of the study and limitations in network design. Median annual crown transparency (CT) over the period 1996-2000 and Basal Area Increment (BAI) between 1997 and 1999 were modelled using a number of predictors selected after PCA. Models were statistically significant for CT (Norway spruce,  $P=0.002$ ; beech,  $P=0.0007$ ) and BAI (all species, standardized values,  $P=0.017$ ). Residual (measured – predicted) analyses were then carried out to investigate whether a relationship exists between  $O_3$  AOT40 exposure index and deviation from expected CT and BAI. CT residuals were found to increase with AOT40 in beech ( $R^2=0.54$ ,  $P<0.05$ , while remaining unchanged for Norway spruce ( $R^2=0.016$ , ns)). BAI residuals were negatively related with AOT40 in the case of Norway spruce ( $R^2=0.47$ , ns) and deciduous oaks ( $R^2=0.99$ ,  $P<0.05$ ), but not for beech. In brief, relationships between  $O_3$  AOT40, CT and BAI were not unequivocal, however the significant reduction of BAI residuals in oaks as well as the indications of a possible impact of  $O_3$  on certain beech sites is noteworthy. Availability of additional data and sites in the next few years, together with developments in flux modelling, will enable a closer examination of the present indications.

**Key words:** *observational study, ozone, AOT40, crown condition, growth.*

**Riassunto** — Esposizione ad ozono, trasparenza della chioma e incrementi di area basimetrica nelle aree permanenti del programma CONECOFOR in Italia. L'ozono è riconosciuto come un agente in grado di influenzare numerose funzioni e processi sia a livello di pianta che ecosistema. I dati raccolti alle aree permanenti del programma CONECOFOR permettono di indagare gli effetti sullo stato delle chiome e sugli accrescimenti. Le indagini sono tuttavia problematiche in relazione a limitazioni dovute alla natura dello studio ed al disegno della rete. La trasparenza della chioma (CT) e l'incremento di area basimetrica (BAI) sono state dapprima modellati secondo un insieme di predittori selezionati dopo una analisi delle componenti principali (PCA). I modelli sono risultati significativi sia per la trasparenza della chioma (abete rosso,  $P=0.002$ ; faggio,  $P=0.0007$ ) che per l'incremento di area basimetrica (tutte le specie, valori standardizzati,  $P=0.017$ ). I valori dei residui (misurato – predetto) sono stati quindi studiati in relazione ai valori di AOT40. I residui di CT aumentano all'aumentare di AOT40 nel caso del faggio ( $R^2=0.54$ ,  $P<0.05$ ), ma rimangono invariati nell'abete rosso ( $R^2=0.016$ ). I valori di BAI sono risultati correlati negativamente con l'AOT40 nel caso dell'abete rosso ( $R^2=0.47$ , ns) e delle querce decidue ( $R^2=0.99$ ,  $P<0.05$ ), ma non per il faggio. In sintesi, la relazione tra esposizione ad  $O_3$  AOT40, CT e BAI appare controversa. Tuttavia, la diminuzione di accrescimenti nelle querce e l'evidenza di un possibile impatto su alcune aree a prevalenza di faggio necessitano di approfondimenti che saranno possibili nei prossimi anni quando – insieme ai progressi nelle modellistica dei flussi - sarà disponibile un dataset più completo.

**Parole chiave:** *studi osservazionali, ozono, AOT40, chiome, accrescimenti.*

Ozone ( $O_3$ ) effects on forests are various in nature and concern several tree and ecosystem compartments (MATYSSEK and INNES 1999; see also the overview by FERRETTI *et al.* 2003b this volume). Ozone concentrations at the Permanent Monitoring Plots (PMPs) of the CONECOFOR programme in Italy were measured by passive sampling over the period 1996-2000 (BUFFONI and TITA 2003). Five-year averages of weekly June-September concentration levels varied between 29.5 and 53.1 ppb, with maximum weekly values reaching 87 ppb (BUFFONI and TITA 2003). On the basis of concentrations measured by passive sampling,

GEROSA *et al.* (2003 this volume) attempted to estimate the exposure in terms of AOT40 ( $O_3$  Accumulated Over a Threshold of 40 ppb, FUHRER *et al.* 1997). They estimated that mean 1996-2000 June-September AOT40 at measurement height at the various PMPs ranged between 4004 and 25891 ppb h<sup>-1</sup>. The critical levels set at the Kuopio (10000 ppb h<sup>-1</sup>, KÄRENLAMPI and SKÄRBI 1996) and at the Gothenburg workshops (5000 ppb h<sup>-1</sup> for sensitive tree species under sensitive environmental condition, KARLSSON *et al.* 2003b) were frequently exceeded, raising the question of potential risk for the forest vegetation in the PMPs. Unfortunately, there are several limita-

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tions when attempting to estimate  $O_3$  effects by means of routine monitoring programmes like CONECOFOR (FERRETTI *et al.* 2003a). In general, these programmes are observational studies carried out in a network of sites. Their aim is generic monitoring rather than specific investigation of the effects of  $O_3$ . This means that sites were not selected and installed according to an *ad hoc* experimental design: this is a clear limitation because the strength of inference of observational studies depends essentially on their design (EBERHARDT and THOMAS 1991). There are several consequences of lack of experimental design: (i) Permanent Monitoring Plots (PMPs) are located in forests with different species assemblages, different ages, and different site conditions; (ii) allocation of PMPs to possible  $O_3$  levels may be not balanced, and certain combinations of species- $O_3$  levels, for example, may be over or under represented. GEROSA *et al.* (2003) report very different exposure ranges for PMPs with various species, deciduous oaks being mostly at high exposure levels ( $>16000-30000$  ppb  $h^{-1}$ ) and Norway spruce at low levels ( $>5000-13000$  ppb  $h^{-1}$ ); (iii) sampling numbers may not be adequate for proper statistical analysis and interpretation (FERRETTI 2000b) and (iv), no true control is available: the results can only be interpreted on a speculative basis.

A second series of consequences is that data collected by the programme may not cover all the data requirements necessary to calculate proper exposure and response indicators. For example, difficulties and uncertainties encountered in calculating  $O_3$  flux (KARLSSON *et al.* 2003b; GEROSA and ANFODILLO 2003 this volume) led us to keep the AOT40 value as exposure indicator, although its limits are well known (*e.g.* FUHRER *et al.* 1997; KARLSSON *et al.* 2003a). Concerning the response of ecosystems, indicators available in the CONECOFOR programme include tree growth (estimated by basal area increment), crown transparency and visible foliar symptoms. Foliar symptoms of trees, herbs and shrubs are presented by BUSSOTTI *et al.* (2003) in another chapter: they are collected mostly on a qualitative basis and are not suitable for statistical analysis. Quantitative data are available for basal area increment and crown transparency (FABBIO and AMORINI, 2000, 2002; BUSSOTTI *et al.* 2000, 2002). Unfortunately, these indicators are unspecific and depend on a variety of factors, including species, genetic features, site conditions, soil properties, climate and weather, current and past forest management and competition for resources. These factors can be regarded as noise when attempting to

evaluate  $O_3$  effects. Thus, although  $O_3$  may contribute to explain changes in the indicators mentioned above (see the various references provided by FERRETTI *et al.* 2003 this volume) the nature of the study, the lack of experimental design and absence of a true control make it necessary to take into account this noise and to remove as much of it as possible.

In the above framework, the aim of this paper is to investigate relationships between  $O_3$  exposure (in terms of AOT40), crown transparency (CT) and basal area increment (BAI) after having removed as many effects of other site and environmental factors as possible.

## Methods

### Assumptions and limitations

In this paper, a number of attributes (crown transparency, foliar nutrients, soil properties,..) averaged at plot level are used as predictor and/or response indicators. This approach requires a number of assumptions. The most important one concerns the ability of the available data to provide reliable, unbiased estimates of population parameters (proportions, means and totals) at plot level. While BAI data are based on a full census of the trees in the plot (and therefore they provide the real value of the attribute of concern at the site), crown transparency and the various other predictors are based on sampling (see FERRETTI and NIBBI 2000). Since the extent to which sampling designs adopted are able to provide reliable, unbiased estimates of the population parameters is questionable (FERRETTI and CHIARUCCI 2003), caution is needed when examining the data reported hereafter.

A second limitation is that PMPs were selected on a preferential basis, unrelated to any explicitly defined target population: they are individual case studies and any conclusions will not be applicable to sites other than those being considered.

A third limitation concerns the reliability of CT data. CT is assessed visually by surveyors and is prone to observer bias (*e.g.* INNES 1988). Italian surveyors underwent intensive training and field checks which enabled major sources of uncertainty to be controlled and documented. Although BUSSOTTI *et al.* (2002) reports a clear improvement of surveyor performance in time, the possibility of subjective assessment remains.

### Approach and statistical methods

Mean plot BAI of the trees of the main tree species

(MTS) in the dominant storey over the period 1997-1999 and plot median annual MTS crown transparency (CT) (years: 1996, 1997, 1998, 1999, 2000) were used as response indicators. BAI is the closest indicator to the one used to establish  $O_3$  critical levels, namely changes in biomass (KARLSSON *et al.* 2003a, b). Crown transparency is the most common indicator of tree condition in Europe (FERRETTI 1998). The hypothesis being verified is whether deviations from expected BAI and CT were related to  $O_3$  exposure. In theory, deviations from expected values should occur at different levels of exposure according to the sensitivity of the MTS being considered. For CT, increasing residuals are expected at increasing  $O_3$  exposure (positive slope of the regression); for BAI, decreasing residuals are expected at increasing  $O_3$  exposure (negative slope of the regression). Three steps were undertaken. First, BAI and CT were modelled according to a number of predictors using multiple regression models (see below). A key problem here is the ratio of the number of variables (the predictors) to the number of cases (the PMPs). This is especially true for BAI (one mean value per plot). To overcome this problem, we used Principal

Component Analysis (PCA) to identify the minimum set of variables to be included in the model (see below). The performance of the models in terms of statistical significance and variance explained was tested by direct comparison with measured data. Second, residuals (measured-predicted) were calculated. Third, CT and BAI residuals (measured – predicted) were analysed in relation to AOT40 (various settings, see below).

#### Predictors and response indicators

The variables considered for the PCA and subsequently included in the various models are in Table 1. Table 2 summarises BAI, CT and AOT40 at each PMP. Original data for the site, basal area increment, soil, foliage, N-deposition, meteorological variables and crown transparency are reported by FERRETTI (2000a), MOSELLO *et al.* (2002) and ALIANIELLO *et al.* (2003). Details about data quality and reliability are provided by FERRETTI and NIBBI (2000), BUFFONI and TITA (2003), AMORIELLO *et al.* (2003), ALIANIELLO *et al.* (2003). Table 3 reports standardized values for BAI 1997-1999, CT 1996-2000, damage to foliage 1998-1999 and foliar nutrients 1995-1999. We first considered site data (elevation, age, stem density,

**Table 1 –** Variables considered in PCA and subsequently used to model BAI and CT.  
Variabili utilizzate nella PCA e successivamente selezionate per i modelli di BAI e CT.

Code	Unit	Definition	No of plots covered	Used in PCA	Used in model
CT std	number	Standardized 1997-1999 median crown transparency	20	BAI	BAI
FD std	number	Standardized 1998-1999 median foliar damage extent	20	BAI	
Alt	m	Elevation a.s.l.	20	BAI	CT (beech)
Age	years	Age of the dominant storey in 1995	20	BAI	
Management system	Categories	Management regime	20		BAI, CT
StemDen	n ha <sup>-1</sup>	number of trees per ha	20	BAI, CT	BAI, CT (beech, Norway spruce)
Topsoil C/N	number	C/N ratio in topsoil in 1995	20	BAI, CT	CT (beech, Norway spruce)
BaseSat	%	Base saturation of mineral soil in 1995	16	BAI, CT	
CEC	CM kg <sup>-1</sup>	Cation Exchange Capacity in 1995	16	BAI, CT	
Prec	mm	Mean summer precipitation over the period 1998-2000	15	BAI, CT	BAI, CT (beech)
REt	%	Relative evapotranspiration, summer 1998-2000	10	BAI, CT	
N	mg/g	Mean foliar concentration of N over the period 1995-1999	20	CT	CT (beech)
P	mg/g	Mean foliar concentration of P over the period 1995-1999	20	CT	CT
Ca	mg/g	Mean foliar concentration of Ca over the period 1995-1999	20	CT	(Norway spruce)
Mg	mg/g	Mean foliar concentration of Mg over the period 1995-1999	20	CT	CT (beech)
K	mg/g	Mean foliar concentration of K over the period 1995-1999	20	CT	CT (beech)
SumCaMgK std	number	Standardized sum of Ca, Mg and K	20	BAI	
N/P std	number	Standardized ratio between mean values of N and P	20	BAI	
N/Ca std	number	Standardized ratio between mean values of N and Ca	20	BAI	BAI
N/Mg std	number	Standardized ratio between mean values of N and Mg	20	BAI	
N/K std	number	Standardized ratio between mean values of N and K	20	BAI	
DepN	mmol* m <sup>-2</sup> per year	Throughfall of N over the period 1998-1999	15	BAI, CT	BAI

**Table 2 –** General plot information and summary data for basal area increment (BAI), crown transparency (CT) and AOT40.  
*Informazioni generali sui PMP e dati di sintesi su incremento di area basimetrica (BAI), transparenza della chioma (CT) ed AOT40.*

PMP	Altitude, m a.s.l	Main Tree Species	Age dominant storey, years	Stem density, n ha <sup>-1</sup>	Mean BAI, dominant storey 1997-2000 m <sup>2</sup> ha <sup>-1</sup>	Mean of median Crown Transparency 1996-2000 %	Mean AOT40 1996-2000, ppb h <sup>-1</sup>
ABR1	1500	<i>Fagus sylvatica</i>	110	879	2,08	20,50	6546
BAS1	1125	<i>Quercus cerris</i>	65	868	0,84	21,67	14423
CAL1	1100	<i>Fagus sylvatica</i>	110	321	0,55	34,00	4389
CAM1	1175	<i>Fagus sylvatica</i>	100	228	1,52	25,00	17291
EMI1	200	<i>Quercus petraea</i>	45	2205	2,17	19,00	17887
EMI2	975	<i>Fagus sylvatica</i>	40	4172	2,13	20,50	15384
FRI1	6	<i>Q.robur/Carpinus betulus</i>	45	1098	1,64	15,63	16129
FRI2	820	<i>Picea abies</i>	110	576	1,80	14,57	10570
LAZ1	690	<i>Quercus cerris</i>	35	1617	1,85	15,00	13829
LOM1	1190	<i>Picea abies</i>	80	1019	3,23	13,10	5739
MAR1	775	<i>Quercus cerris</i>	35	4364	3,21	18,00	17748
PIE1	1150	<i>Fagus sylvatica</i>	55	1187	1,16	24,00	8686
PUG1	800	<i>Fagus sylvatica</i>	85	993	2,13	12,50	18322
SAR1	700	<i>Quercus ilex</i>	50	1540	1,40	15,00	9201
SIC1	940	<i>Quercus cerris</i>	50	839	0,97	14,00	25891
TOS1	150	<i>Quercus ilex</i>	50	2327	0,78	29,50	14632
TRE1	1775	<i>Picea abies</i>	160	389	1,20	12,00	9936
UMB1	725	<i>Quercus cerris</i>	75	731	1,38	23,10	11627
VAL1	1740	<i>Picea abies</i>	150	736	2,32	23,50	9447
VEN1	1100	<i>Fagus sylvatica</i>	110	345	0,98	15,00	4004

management regime), tree condition data (crown transparency), soil data (topsoil C/N ratio, base saturation, cation exchange capacity), meteorological data (precipitation, relative evapotranspiration), foliar nutrient data (concentrations and concentration ratios) and deposition data (throughfall deposition of N). Management regime was subsequently dropped from the list because it is a nominal variable and it is well related to other variables like age and stem density. We then used PCA to identify the minimum set of variables to be included in the model. Due to the limited dataset and the fact that some variables were not measured in all PMPs, we did not use any formal stepwise procedure to select variables to include in the BAI and CT models. For the same reasons, the direct use of principal components (PCs) was unsuccessful (e.g.  $R^2=0.41$ ;  $P=0.74$ , for the BAI model). Rather, PCA was used to identify the variables best related to the individual PCs (*i.e.* variables contributing the most explained variance within each PC). This allowed us to select the variables that maximize the performance of the model in terms of explained variance without unacceptable loss of PMPs for the analysis due to lack of data. After PCA, only variables with eigenvalue  $> 1$  and, within each PC, only variables with correlation coefficients  $> 0.7$  were considered for the regression models. Among them, priority was given to variables measured in a large number of plots and/or with better confidence. For example, stem density and age are well related ( $R^2=0.64$ ;  $P<0.01$ ; Fig. 1) and both are in the same PCs: PC1 and PC3 of the CT study for Norway spruce and beech respectively and PC1 of the

BAI study (see Tables 3 and 4). However, stem density was actually measured whereas age was estimated: thus stem density was selected for the models instead of age. Similarly, although relative evapotranspiration ( $RE_t$ ) is related to summer precipitation ( $R^2=0.59$ ;  $P<0.05$ ), the latter was measured on a larger number of PMPs: we therefore preferred summer precipitation. The relationship between age and stem density is also related to forest management practices, as younger broadleaf stands originated from coppicing (see ALIANIELLO *et al.* 2003 this volume).

Crown transparency was used as response indicator in the relevant study and as predictor in the BAI study. Response indicators include mean plot basal area increment (BAI) of the trees of the main tree species (MTS) in the dominant storey over the period 1997-1999 and plot median annual MTS crown transparency (years: 1996, 1997, 1998, 1999, 2000). Ozone Accumulated Over a Threshold of 40 ppb (AOT40) was obtained starting from weekly  $O_3$  concentration values measured by passive sampling (BUFFONI and TITA 2003 this volume) and modelled by GEROSA *et al.* (2003 this volume). Different AOT40 were used for the BAI and CT studies: AOT40 averaged over the period 1997-1999 (*i.e.*, the time between the two subsequent BAI measurements) and AOT40 up to the assessment date for each PMP and year, respectively. These data were obtained after further processing of the data by GEROSA *et al.* (2003b) and are subjected to the same uncertainties. In the context of this paper, a critical issue is also the ability of the  $O_3$  measurement sites to provide fully valid data for the

PMP where BAI and CT were measured. The two sites do not always coincide (see BUFFONI and TITA 2003), and, under complex terrain conditions, even a small distance between  $O_3$  measurement sites and PMPs may result in considerable difference in  $O_3$  levels.

Another deficiency of the available dataset is the lack of information about genetic variability between the various MTS populations in which PMPs are installed. This is particularly important for beech, whose PMPs range from the Alps (es. PIE1, VEN1) to the southern Apennines (es. CAL1) and for which significantly higher variability was reported for southern provenances (BUCCI *et al.* 1999). Different response of southern provenances to  $O_3$  and to water stress (which is particularly important for  $O_3$  effects) were also reported (PALUDAN-MÜLLER *et al.* 1999; TOGNETTI *et al.* 1995).

#### CT study

The response indicator used for this study was the annual median plot crown transparency of the MTS. Medians were calculated for each plot and year over the period 1996-2000. Details of crown transparency assessment methods at the CONECOFOR PMPs are provided by BUSSOTTI *et al.* (1999, 2002). CT is usually assessed on 30 trees per PMP selected in the dominant storey following a spiral starting in the centre of the PMP. Annual CT values provide a larger dataset (35 cases for beech, 25 for Turkey oak and 20 for Norway spruce) enabling species-specific analysis. Actually, defoliation of Norway spruce, Scots pine, English oak, pedunculate

oak and beech were found to vary in relation to the annual levels of  $O_3$  (INNES and BOSWELL 1988). These findings were further confirmed by MATHER *et al.* (1995) in beech and Scots pine. A limitation here is possible temporal autocorrelation of the data that may violate the underlying assumption of data independency.

Holm oak plots (TOS1 and SAR1) and one mixed broadleaves plots (FRI2) were not considered because of their reduced data set. Selection of possible predictors considered annual data for foliar chemistry: when data were lacking for a certain year at a certain PMP, the 1995-1999 PMP mean value was used.

#### BAI study

BAI data are only relevant to a spatial dimension, as there is only one value per PMP. The study can only consider mean plot values over the period between the two measurement of stem circumference carried out in autumn-winter 1996-1997 and 1999-2000, respectively. Measurements were carried out on all living trees in the PMP: BAI was computed as the difference between the two measurements only for trees of the dominant storey that were alive at both times. Dominant trees are believed to be less influenced by current competition for resources and more exposed to  $O_3$ . The median relative error in circumference measurements was 5.4% in 1996-1997 and 5.3% in 1999-2000. The fact that there was only one mean BAI per plot meant that no more than 20 cases were available for the study. Further stratification according to MTS will sharply reduce the number

**Table 3 –** Species-standardized values for basal area increment (BAIstd), crown transparency (CTstd), damage to foliage (DFstd) and foliar nutrients N, P, Ca, Mg, K (Nstd to Kstd).

*Valori standardizzati sulla specie di incremento di area basimetrica (BAIstd), transparenza della chioma (CTstd), danni alle foglie (DFstd) e nutrienti fogliari N, P, Ca, Mg, K (da Nstd a Kstd).*

PMP	Main Tree Species	BAIstd	CTstd	DFstd	Nstd	Pstd	Castd	Mgstd	Kstd
ABR1	<i>Fagus sylvatica</i>	0,902	-0,24	1,13	-0,061	0,517	0,966	-0,003	-1,061
BAS1	<i>Quercus cerris</i>	-1,095	0,74	-1,47	-0,494	1,241	0,238	0,876	1,796
CAL1	<i>Fagus sylvatica</i>	-1,508	1,83	1,73	-1,100	-0,052	-0,832	-1,220	-1,358
CAM1	<i>Fagus sylvatica</i>	0,020	0,37	-0,65	-0,014	-0,337	1,493	0,501	1,635
EMI1	<i>Quercus petraea</i>	0,555	0,06	0,92	-0,747	-1,335	-0,967	-0,157	-1,452
EMI2	<i>Fagus sylvatica</i>	0,981	-0,37	-0,58	-0,124	-0,906	0,019	-0,045	-0,061
FRI1	<i>Q. robur/Carpinus betulus</i>	-0,103	-0,71	1,64	0,948	-0,047	0,319	1,744	-0,762
FRI2	<i>Picea abies</i>	-0,390	-0,07	0,00	0,312	-1,170	0,627	1,359	-0,493
LAZ1	<i>Quercus cerris</i>	0,158	-0,32	-0,27	1,846	-0,047	-1,210	-0,220	0,135
LOM1	<i>Picea abies</i>	1,270	-0,18	0,00	0,318	1,259	-0,558	-0,243	-0,828
MAR1	<i>Quercus cerris</i>	1,845	-0,52	-0,27	-0,510	-1,185	1,823	-0,820	0,009
PIE1	<i>Fagus sylvatica</i>	-0,547	0,37	-0,58	2,048	-1,475	-1,240	-1,346	0,021
PUG1	<i>Fagus sylvatica</i>	0,981	-1,34	-0,76	0,010	1,330	-0,716	1,382	0,495
SAR1	<i>Quercus ilex</i>	0,710	1,48	-0,71	0,707	0,707	0,707	-0,707	0,707
SIC1	<i>Quercus cerris</i>	-0,934	-1,09	-0,27	-0,732	0,282	-0,324	-0,246	0,261
TOS1	<i>Quercus ilex</i>	-0,710	0,16	0,71	-0,707	-0,707	-0,707	0,707	-0,707
TRE1	<i>Picea abies</i>	-1,090	-0,47	0,00	0,826	0,104	-1,105	-1,044	1,435
UMB1	<i>Quercus cerris</i>	-0,425	1,84	-0,27	-0,310	1,091	0,121	-1,177	0,013
VAL1	<i>Picea abies</i>	0,210	1,24	0,00	-1,456	-0,192	1,037	-0,072	-0,114
VEN1	<i>Fagus sylvatica</i>	-0,830	-0,61	-0,29	-0,759	0,923	0,310	0,732	0,330

of cases. To model BAI, all the PMPs were therefore considered together. To do this, species-specific data (BAI, foliar nutrients, crown transparency, damage to leaves/needles; Table 3) were standardized according to the formula:

$$Xstd_{ij} = \frac{(x_{ij} - \bar{x}_j)}{s}$$

where:

$Xstd_{ij}$  is the new, standardized value of BAI in the plot  $i$  of MTS  $j$ ,  
 $x_{ij}$  is the measured value of MTS  $j$  in plot  $i$ ,  
 $\bar{x}_j$  is the mean value of the  $n$  plots of MTS  $j$ ,  
 $s$  is the standard deviation of the  $n$  plots of MTS  $j$ .

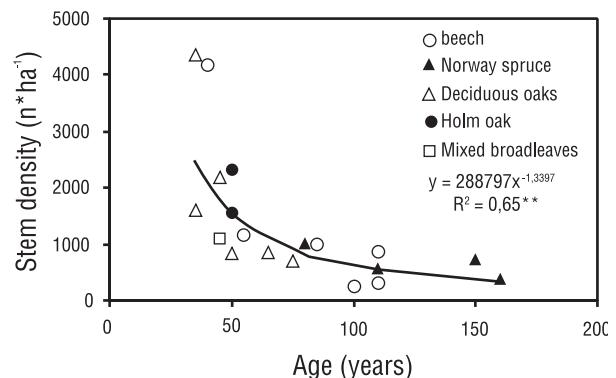
Distinction between PMPs according to MTS will be made when presenting and discussing the results.

## Results and discussion

### Ozone exposure and crown transparency

#### Crown transparency model

The variables listed in Table 1 were subjected to PCA (rotation method: varimax with Kaiser normalization) (Table 4). Different models were investigated for beech, Norway spruce and Turkey oak. Due to changes in the Manual adopted by the surveyor, no consistent data on effects of recognized crown damage are available for all PMPs and years, so they were not used in the PCA. However, available information about damage that may affect crown transparency itself and/or its assessment was used at a later stage to discriminate cases to be



**Figure 1 –** Stem density and estimated age at the various PMPs of the CONECOFOR programme. Different species are identified by different symbols. Regression equation and coefficient of determination of the whole data set is reported. \*\*=P<0.01. *Densità dei fusti ed età stimata alle varie aree permanenti del programma CONECOFOR. Le varie specie sono identificate da diversi simboli. Vengono riportati il coefficiente di determinazione e l'equazione di regressione del set completo dei dati. \*\*=P<0.01.*

included in the analysis (see below).

For beech, the 3 PCs explained 90.8 % of the variance. Elevation, stem density, topsoil C/N ratio, foliar concentration of Ca and summer precipitation were selected as predictors: the model was significant ( $R^2=0.57$ ;  $P=0.0007$ ; Fig. 2). For spruce, the 2 PCs explained 89.8 % of the variance. Stem density, topsoil C/N ratio, foliar concentration of Ca and P were selected as predictors: the model was significant ( $R^2=0.66$ ;  $P=0.002$ ; Fig. 2). It is worth noting that age resulted a significant predictor for both beech and Norway spruce, however since age was significantly related to stem density (see Fig. 1), the latter was used in the model (see Materials

**Table 4 –** PCA to select variables for the CT model. Correlation coefficients and portion of variance explained by each component.  
*PCA per selezionare le variabili per il modello CT. Vengono riportati i coefficienti di correlazione e la porzione di varianza spiegata da ciascuna singola componente.*

Variable code	Beech			Principal Components			Turkey oak		
	1	2	3	1	2	1	2	3	
Alt	0.952	-0.226	0.189	0.642	0.738	-0.977	-0.205	0.037	
Age	0.354	-0.269	0.873	0.990	0.085	NC	NC	NC	
StemDen	-0.223	0.258	-0.914	-0.956	0.274	0.187	0.916	-0.315	
Topsoil C/N	0.979	-0.001	0.162	0.976	0.182	0.982	-0.147	0.087	
BaseSat	0.977	-0.141	-0.100	0.995	0.014	-0.843	0.484	-0.201	
CEC	0.866	-0.457	0.164	0.982	-0.161	-0.992	-0.083	-0.065	
Prec	-0.563	0.383	0.726	-0.846	0.516	0.668	0.701	-0.222	
Ret	-0.227	0.816	0.503	-0.584	0.791	0.571	0.771	-0.251	
N	0.360	0.356	-0.716	0.539	0.355	0.584	0.154	0.699	
P	0.330	0.421	0.706	-0.368	0.884	-0.081	0.098	0.867	
Ca	0.777	0.444	-0.343	-0.357	-0.875	-0.345	0.884	-0.057	
Mg	0.122	0.871	-0.019	-0.182	-0.836	-0.070	-0.903	-0.025	
K	-0.108	0.851	-0.415	0.806	0.360	-0.012	-0.381	0.697	
DepN	0.877	0.386	0.196	-0.955	-0.267	0.952	0.293	-0.069	
Variance explained	39,6	28,0	23,2	60,3	29,5	46,3	33,5	11,6	

and methods). The models were as follows:

$$CT_{\text{beech}} = -0.0014 \text{StemDen} - 0.0109 \text{Prec} - 4.6145 \text{C/N} - 1.4408 \text{Ca} + 0.0421 \text{Alt} + 89.87$$

$$CT_{\text{Norway spruce}} = 0.00433 \text{StemDen} + 0.70752 \text{C/N} + 6.1328 \text{P} + 2.90639 \text{Ca} - 34.97$$

Despite the large percentage of variance explained by the 3 PCs (91.4 %, Table 4), no significant model able to predict CT was identified for Turkey oak. Therefore, no further analysis was undertaken.

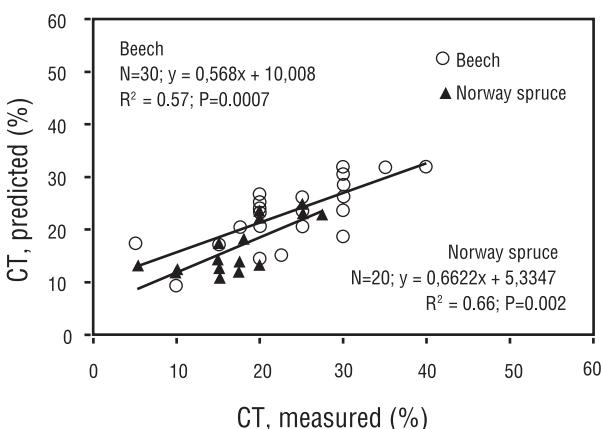
#### Ozone exposure and crown transparency

Residuals (measured – predicted) were calculated for plots in which the selected predictors were available. To avoid misinterpretation, PMPs for which damage to tree crowns due to recognized biotic/abiotic factors (insects, fungi, hail, drought) as well as potential observer bias were reported (BUSSOTTI *et al.* 2002) were not considered in the analysis. The exclusion resulted in a sharply reduced dataset for beech (the species for which most injury was reported), with only 8 cases remaining. For spruce, the effect of recognized biotic/abiotic factors was obvious in a limited number of cases (BUSSOTTI *et al.* 2002). Residuals were regressed against AOT40 until the date of assessment. In the case of beech, data points only covered the lowest exposure (AOT40 < 5000 ppb h<sup>-1</sup>). In this exposure range there was a clear trend, and residuals increased with increasing AOT40

( $R^2=0.54$ ,  $P<0.05$ ) (Fig. 3). It is worth noting that three of the six data points of beech are from the PMP VEN1 (years 1996, 1997, 2000): this was the only CONECOFOR beech site in which foliar symptoms on adult trees were recorded (BUSSOTTI *et al.* 2003). For Norway spruce, there is a considerable scatter, especially for data points below AOT40 5000 ppb h<sup>-1</sup>, and no trend can be detected ( $R^2=0.016$ , slope: -0.0001) (Fig. 3).

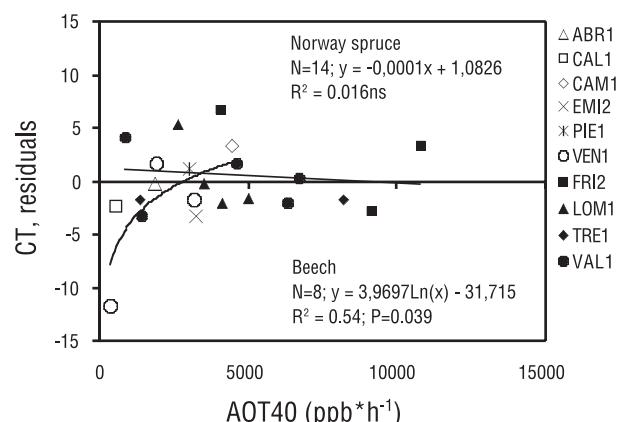
#### Discussion

Previous investigations have looked at relationships between O<sub>3</sub> and CT in larger sample sizes at national and European level (INNES and BOSWELL 1988; INNES and WHITTAKER 1993; MATHER *et al.* 1995; HENDRICKS *et al.* 1997; INNES *et al.* 1997; DOBBERTIN *et al.* 1997; ZIERL 2002; KLAU *et al.* 2000). The findings from Switzerland seem to support the hypothesis that AOT40 can be a significant predictor of defoliation (a somewhat alternative expression of CT) of different species in Level I plots. However, a number of other factors were found to be significant, and the reliability of CT as a response indicator was also questioned (DOBBERTIN *et al.* 1997; ZIERL 2002). Correlation coefficients between AOT40 and CT were influenced by site condition (plots with high modelled stomatal conductance have higher and positive  $r$  values) and changed from year to year according to the species



**Figure 2** – Measured and predicted CT at the various assessment occasion at the PMPs of the CONECOFOR programme. Different species are identified by different symbols. Equation of regression, coefficient of determination and P level for each species is reported.

*Trasparenza della chioma misurata e stimata alle varie valutazioni annuali presso le aree permanenti del programma CONECOFOR. Le varie specie sono identificate da diversi simboli. Vengono riportate l'equazione di regressione, il coefficiente di determinazione ed il livello di probabilità per ciascuna specie.*



**Figure 3** – CT residuals plotted against AOT40 values up to the date of the assessment. Open symbols: beech; Full symbols: Norway spruce. Different PMPs are identified by different symbols and listed on the right side of the graph. Number of datapoints, regression equation and coefficient of determination are reported. ns: not significant.

*Residui di CT in relazione ai valori di AOT40 fino alla data di valutazione. Simboli aperti: faggio; simboli pieni: abete rosso. Le varie aree sono identificate da diversi simboli ed elencate nella parte destra della figura. Vengono riportati il numero di coppie di dati, l'equazione di regressione ed il coefficiente di determinazione. ns: non significativo.*

being considered (ZIERL 2002). No attempt was made to incorporate the effects of known causes of defoliation and/or other relevant stress factors, although ZIERL (2002) acknowledges that "to achieve better results, probably all possible causes... have to be simultaneously included into multivariate statistical analyses". Results from the United Kingdom show that defoliation values of Norway spruce, Scots pine, English oak, pedunculate oak and beech varied in relation to annual levels of  $O_3$  (INNES and BOSWELL 1988). These findings were further confirmed by MATHER *et al.* (1995) in beech and Scots pine. HENDRICKS *et al.* (1997) report  $O_3$  to be one of the factors explaining both defoliation levels and foliar Mg and Al content in English oak and Scots pine in the Netherlands. The above investigations seem to support the hypothesis that  $O_3$  may be a significant factor to explain CT. The evidence for such significance was much less obvious in our study. According to our data, a large portion of the variance (up to 90 %) of CT values can be explained by site factors for beech and Norway spruce: the role of  $O_3$  on CT seems limited in any case. This can depend on different causes: one is certainly related to limitation of the dataset, discussed above. A second likely cause is that the above studies considered data from climatic situations quite different from Italy. In particular, factors counteracting  $O_3$  uptake (high VPD, low soil moisture) can be expected to be much less important in the Netherlands, Switzerland and UK than

**Table 5 –** PCA to select variables for the BAI model. Correlation coefficients and portion of variance explained by each component.  
*PCA per selezionare le variabili per il modello BAI. Vengono riportati i coefficienti di correlazione e la porzione di varianza spiegata da ciascuna singola componente.*

Variable code	Principal Components				
	1	2	3	4	5
CT	0,023	0,198	-0,061	0,827	-0,030
DF	-0,101	-0,120	0,383	0,818	-0,202
Alt	0,804	0,162	-0,180	-0,082	0,066
Age	0,951	0,236	0,047	-0,014	0,034
StemDen	-0,812	0,051	-0,024	-0,210	0,306
Topsoil C/N	0,167	0,658	0,475	-0,242	0,118
BaseSat	-0,060	0,054	0,906	-0,048	-0,266
CEC	0,229	0,576	0,691	-0,148	0,014
Prec	0,388	0,119	-0,742	-0,235	-0,384
Ret	0,583	0,159	-0,591	-0,457	-0,029
SumCaMgK std	0,399	0,651	-0,293	0,289	0,143
N/P std	-0,071	-0,071	-0,054	-0,078	0,917
N/Ca std	-0,087	0,957	-0,053	0,045	-0,108
N/Mg std	0,399	0,651	-0,293	0,289	0,143
N/K std	0,151	-0,406	-0,286	0,712	0,133
DepN	-0,274	-0,699	-0,080	0,105	0,116
<i>Variance explained</i>	20,4	19,6	18,6	15,7	8,5

in Italy, although ZIERL (2000) stated that soil moisture is the major factor controlling the response of stomata during the growing season.

#### **Ozone exposure and growth**

##### *BAI model*

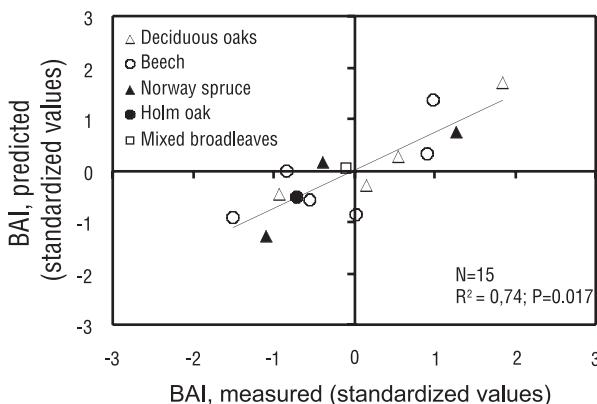
The variables listed in Table 1 were subjected to PCA (rotation method: varimax with Kaiser normalization). The results are reported in Table 5 as correlation coefficients between each variable and each particular PC. Overall, the 5 PCs explained 82.9% of the variance. When only variables with correlation coefficients  $> 0.7$  were considered, stem density, mean summer precipitation, median crown transparency, N/Ca and N/K were selected as predictors for the BAI (Table 3). The model is as follows:

$$\text{BAI}=0.0005\text{StemDen}+0.0009\text{Prec}+0.576\text{N/Ca}_{\text{std}}+0.372\text{N/K}_{\text{std}}-0.359\text{CT}-1.3616$$

The model was significant and still explained a large portion of the variance ( $R^2=0.74$ ;  $P=0.017$ ; Fig. 4); however, performance was much better for the subset of Norway spruce and deciduous oak data ( $R^2=0.80$  and  $R^2=0.88$ , respectively) than for beech ( $R^2=0.56$ ), as is evident from the scattered distribution of the beech data points.

#### *BAI and ozone*

Residuals (measured – predicted) were calculated for plots in which the selected predictors were available and plotted against mean AOT40 values over the period 1997-1999 (Fig. 5). As expected, data points for different species had different distribution along the x axis. Data points for Norway spruce were in the lowest range of exposure (5000-13000 ppb  $h^{-1}$ ); data points for deciduous oaks were always in the highest exposure range (16000-30000 ppb  $h^{-1}$ ). Considering these species, BAI residuals decreased with increasing AOT40, which is consistent with the hypothesis being investigated. Significant relationships were only found for oaks ( $R^2=0.99$ ,  $P<0.05$ ), although the limited number of data points suggests caution in interpreting this result. Within these limits, the fact that the trends occur at different AOT40 levels in the two data sets is consistent with the expected sensitivity of Norway spruce and deciduous oaks and the location of their PMPs in Italy. Under the revision process of AOT40, Norway spruce is considered a sensitive conifer, with a critical level recently suggested at 5000 ppb  $h^{-1}$ . Norway spruce plots are all in the Alps, an area of Italy where drought



**Figure 4** – Measured and predicted BAI (standardized values) at the various PMPs of the CONECOFOR programme. Different species are identified by different symbols. Coefficient of determination and P level for the whole data set is reported.

*Valori di incremento di area basimetrica (BAI) misurati e stimati alle varie aree permanenti del programma CONECOFOR. Le varie specie sono identificate da diversi simboli. Viene riportato il coefficiente di determinazione ed il livello di probabilità per il set completo dei dati.*

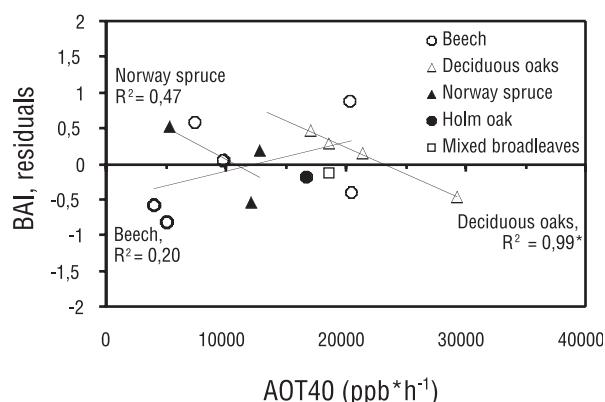
stress (probably the most limiting factor for  $O_3$  uptake) is limited during the vegetative period (see ALIANIELLO *et al.* 2003). No definition of the  $O_3$  sensitivity of *Quercus cerris* (the most frequent deciduous oak species at the CONECOFOR PMPs) is provided, however deciduous oaks like *Quercus petraea* and *Q. robur* are considered moderately sensitive broadleaves in the revision process of the ICP mapping manual (<http://www.oekodata.com/pub/mapping/manual/mapman3.pdf>). Unlike Norway spruce plots, those with deciduous oaks are located mostly in central and southern Italy, where  $O_3$  uptake may be limited by drought stress. This may explain why effects only seem to occur at high-very high exposure levels.

Beech data points encompass a wide range of exposure (4000-20000 ppb  $h^{-1}$ ) and are very scattered, with apparently increasing (although not significantly) BAI residuals at increasing  $O_3$  exposure ( $R^2=0.20$ ) (Fig. 3). This pattern contradicts the expected trend and is mostly related to the unexpected behaviour (in the light of the hypothesis being considered) of three PMPs: CAL1 and VEN1 (negative BAI residuals at low exposure levels) and CAM1 (positive residuals at high exposure).

#### Discussion

Results for Norway spruce and deciduous oaks were in line with the hypothesis being investigated, although the large amount of variance explained by

“traditional” site factors suggests that the effect of  $O_3$  on BAI is limited to maximum 12-20% of the observed variance. The results obtained for beech were in contrast with the hypothesis being tested. This is particularly important as beech is considered to be a sensitive deciduous broadleaf species. BRAUN *et al.* (1999) attempted to study relationships between  $O_3$  and growth in mature beech trees (65-175 years old) growing on 57 permanent plots located in North Switzerland between 260 and 1120 m asl. They used a multilinear regression approach with a number of predictors ( $O_3$  dose, N deposition, base saturation, acid deposition, diameter 1991, social position, position within the stand, crown projection area, crown transparency). They found that “ $O_3$  was negatively correlated with stem increment” and that “the maximum  $O_3$  dose for the 4-year period was a better predictor for growth than the average dose” (BRAUN *et al.* 1999). Although significant correlations were found, the partial correlation plots reported in the paper showed considerable scatter. With our more limited data set, no meaningful relationship between BAI and AOT40 was obvious for beech. There could be different explanations for this. First, limitations to  $O_3$  uptake (*e.g.*, soil moisture deficit, VPD) are likely to be greater in Italy than in Switzerland. The greater the limitation, the greater is the difference between exposure and uptake, which may explain the the different results obtained. In this context, the limited data set and the fact that some of



**Figure 5** – BAI residuals plotted against mean 1997-1999 AOT40 values. Different species are identified by different symbols. Coefficient of determination is reported for beech (N=6), Norway spruce (N=3) and deciduous oaks (N=4). \* = P<0.05.

*Residui dei valori di BAI in relazione alla media 1997-1999 di AOT40. Le varie specie sono identificate da diversi simboli. Vengono riportati i coefficienti di determinazione per faggio (N=6), abete rosso (N=3) e querce decidue (N=4). \* = P<0.05.*

our beech PMPs were located in southern Italy (where limitation to  $O_3$  uptake is likely to occur, see ALIANIELLO *et al.* 2003 this volume) may be a partial explanation.

Second, available information over the time window considered (1997-1999) revealed a number of potential factors that may have influenced the growth of beech in the various PMPs (BUSSOTTI *et al.* 2002) but were not fully considered in the BAI model. In particular, beech plots showed the most frequent and obvious damage from hail, leaf defoliators and leaf miners (BUSSOTTI *et al.* 2002). Part of this damage (leaf defoliators) was routinely recorded by the crown transparency score; similarly, hail was reported to cause crown transparency at PIE1 in 1997. These effects are thus included in the model under the crown transparency predictor. On the other hand, leaf miners and some instances of hail damage were not recorded by the crown transparency score, as leaves were in most cases still on the trees (BUSSOTTI *et al.* 1998). Slight damage to foliage by hail was reported at VEN1 (18% of leaves affected in 1998) while leaf miner *Rynchaenus fagi* affected 40% of foliage in CAL1 in 1998 and 1999. Leaf damage was not included in the BAI model, because – on the whole - its role as predictor was less than crown transparency (see Table 5). However, the fact that leaf damage are mostly concentrated in beech suggests a possible reason for the reduced performance of the BAI model for this species (see above) and – at least in part - for the unexpected behaviour of VEN1 and – especially - CAL1. No peculiar conditions were reported for CAM1, and its deviation from expected BAI remains unexplained. As mentioned above, the role of genetic variability cannot be considered as no data are available.

Third, the inclusion of CT in the BAI model may be also a confounding factor for the detection of  $O_3$  effects. If  $O_3$  affects CT, increased CT may cause a reduction in BAI, but, since CT is included in the BAI model, the expected negative relationship between  $O_3$  and BAI residuals may not occur. On the basis of the data discussed earlier in this paper, there is evidence of an effect of  $O_3$  on CT of beech. Such effect was concentrated on VEN1, one of the PMPs with “unexpected behaviour” of BAI in relation to AOT40.

Fourth, BUFFONI and TITA (2003 this volume) report potential problems with the location of the passive sampler at CAL1, which may have resulted in an underestimation at  $O_3$  levels.

## Conclusions

Establishing relationships between specific environmental stressors and the response of forest ecosystems may encounter substantial problems when based on field observational studies on networks which were not designed *ad-hoc*. The large number of confounding factors and the absence of true controls limit the outcome of any analysis. In our study, further limitation arose from the limited number of case studies available and from the environmental stressor of concern,  $O_3$ , which poses a different set of problems. We use  $O_3$  exposure (AOT40) as stress indicator. It is known that AOT40 may not be a good indicator to predict  $O_3$  effects on plants, but estimates of  $O_3$  flux were not possible within this report (see GEROSA and ANFODILLO 2003 this volume). The value of AOT40 was recently reconsidered by KARLLSON *et al.* (2003b) who used a larger experimental database to detect dose-response relationships between various AOTx (20, 30, 40, 50 ppb·h<sup>-1</sup>) and growth, in terms of biomass reduction. AOT40 provided the best coefficient of determination and it was suggested “to be retained as the recommended method for integrated risk assessment for forest trees”.

The AOT40 estimates used in this paper were based on a June-September time window and are most at measurement height, not at crown height. According to the data provided by GEROSA *et al.* (2003b), the ratio of April-September to June-September AOT40 ranges between 1.12 and 1.83. In addition, the ratio of concentration measured at canopy height to measurement height ranges between 1.10 and 1.29 (BROADMEADOW pers. comm.; GEROSA *et al.* 2001; KRAUSE *et al.* 2002), depending on height, LAI, wind velocity and canopy roughness in the site. All together, this means that actual AOT40 values could be much higher than those used in this paper.

All the above limitations must be taken into account when evaluating the results of this paper. BAI and CT were used as response indicators of effects of  $O_3$  exposure. The portion of variance explained by traditional site and environmental factors was large (82% for BAI; 89-91% for CT) and this means that crown condition and growth are largely determined by site factors, nutritional status, competition and meteorological factors. Residuals (measured-predicted) were calculated and analysed in relation to AOT40 values estimated for the various PMPs. In general, the results were affected by the limited data set, and only qualitative considerations

were possible. When known cases of noise due to the action of biotic and abiotic factors and observer bias were removed, CT residuals remain unchanged for Norway spruce at different AOT40 levels and increased significantly in beech, even at apparently low exposure levels. However, the fact that exposure levels seem low should be considered in relation to the AOT40 values used in the analysis (see above). The behaviour of beech is consistent with the hypothesis being considered and with the nature of beech, considered as a sensitive deciduous species (KARLSSON *et al.* 2003b). In particular, it is worth noting that three of the six datapoints of beech are from the PMP VEN1, a site with the most favourable canopy structure for gas exchange (see ALIANIELLO *et al.* 2003), unexpectedly reduced BAI residuals (see below) and the only CONECOFOR beech site in which foliar symptoms were recorded on adult trees (BUSSOTTI *et al.* 2003). Together, these independent findings suggest potential impact of O<sub>3</sub> at VEN1.

Significant decreases in BAI residuals were only observed for deciduous oaks, i.e. the PMPs located in the highest exposure range. BAI residuals also decreased (although not significantly) for Norway spruce, but not for beech. This contradicts the working hypothesis and may be due to different factors, including site specific conditions, other biotic and abiotic stressors not considered by the model (notably leaf damage by leaf miners), genetic factors, and factors controlling O<sub>3</sub> uptake. The uncertainty surrounding O<sub>3</sub> measurements (including their representativity in relation to PMP), AOT40 estimates (GEROSA *et al.* 2003) and their interaction is superimposed on the CT and BAI studies.

With all the constraints on the analysis described above, there is limited evidence of a general, direct relationship between O<sub>3</sub> exposure and the response indicators considered. This is confirmed by the large portion of variance in CT and BAI explained by traditional site factors. Certain indications (the BAI reduction in deciduous oaks, the increase CT in beech, the occurrence of different, consistent findings at VEN1) are nevertheless of interest and deserve further studies. Future measurement for basal area (planned for the year 2004-2005), inclusion of new sites and – in particular – the acquisition of meteorological parameters and the development of routine, less data-intensive methods for O<sub>3</sub> flux estimates are essential for closer examination of O<sub>3</sub> effects on the Italian intensive monitoring plots.

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# Ozone levels, actual and potential effects on the vegetation at the permanent monitoring plots of the CONECOFOR programme in Italy

## - Achievements, problems and perspectives<sup>§</sup>

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**Abstract** – The 2<sup>nd</sup> Integrated and Combined (I&C) evaluation report of the CONECOFOR programme provides data on ozone exposure and effects on forest ecosystems in Italy. This paper summarizes the various findings of the contributors to the present volume. Emphasis is placed on major results as well as difficulties and problems encountered. Some possible improvements are identified.

**Key words:** *forest ecosystem, integrated evaluation, intensive monitoring, Italy, ozone, Risk Analysis.*

**Riassunto** – Livelli ed effetti reali e potenziali dell'ozono sulla vegetazione delle aree permanenti CONECOFOR– Risultati, problemi, prospettive. Il secondo rapporto di valutazione Integrata e Combinata (I&C) del programma CONECOFOR è il primo tentativo di fornire un quadro documentale dell'esposizione e degli effetti dell'ozono sugli ecosistemi forestali in Italia. Vengono riassunti e sottolineati i risultati, i problemi e le difficoltà riferite dai vari autori del presente volume. Vengono identificati alcuni miglioramenti possibili e le prospettive di lavoro.

**Parole chiave:** *analisi di rischio, ecosistemi forestali, Italia, monitoraggio intensivo, ozono, valutazione integrata.*

There is increasing evidence that tropospheric ozone ( $O_3$ ) has an impact on crops, natural vegetation and forest ecosystems in Europe and elsewhere (BYTNEROWICZ *et al.* 2003; FOWLER *et al.* 1999; KARLSSON *et al.* 2003; MATYSSEK and INNES 1999; MILLS *et al.* 2000). Yet, in general,  $O_3$  concentration is not measured at forest sites and remote areas (DE LEEUW and BOGMAN 2001). Ozone only recently became an issue in the EC and UN/ECE forest monitoring programme, and an *ad-hoc* group on Ambient Air Quality was formed to promote and harmonize methods to measure  $O_3$  concentration in remote areas and to assess visible foliar symptoms (EC – UN/ECE 2003).

The Italian intensive forest monitoring programme CONECOFOR (Italian acronym of CONtrollo ECOsistemi FORestali, Forest Ecosystem Monitoring) was launched in 1995 within the framework of the intensive forest monitoring programme sponsored by the European Commission (Regulation EC n. 1091/94) and

carried out under the auspices of the United Nations Economic Commission for Europe (UN/ECE) (ALLAVENA *et al.* 1999). Ozone measurement by passive sampling started in 1996 at all 20 permanent monitoring plots (PMPs) installed at that time. In 1998 the National Focal Center (NFC) of Italy (based at the Ministry of Agricultural and Forestry Policies, Division V) decided to develop a formal evaluation system (GRUPPO DI ESPERTI CONECOFOR-I&C 1998). The first step was for the team leaders of the various investigations carried out at the PMPs to establish a Task Force (TF). The TF agreed upon a general concept for an Integrated and Combined (I&C) evaluation system and developed a strategy plan for 2001-2005 in which risk analysis in relation to  $O_3$  exposure was the first priority (FERRETTI 2002). The various papers in this volume provide the outcome of the risk analysis in relation to  $O_3$ . This paper attempts to summarize achievements, problems and perspectives.

<sup>§</sup> Paper subject to review by members of the Task Force for the Integrated and Combined (I&C) evaluation of the CONECOFOR data.

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## Achievements

### Ozone levels in remote areas of Italy: 5 years of data

Up to now, knowledge about  $O_3$  levels in remote forest sites in Italy was limited (DESIATO *et al.* 2000). The 1996-2000 data set of  $O_3$  concentration measured by passive sampling at the PMPs of the CONECOFOR programme represents the first consistent, validated, robust dataset for remote forest sites in Italy. Data obtained by passive sampling were in general comparable with those from automatic devices ( $r$  values between 0.84-0.97, see BUFFONI and TITA 2003). BUFFONI and TITA (2003 this volume) report that 5-year weekly mean concentrations over the period June-September ranges between 29.5 ppb (LOM1) and 53.1 ppb (SIC1). In general, concentrations were higher in central and southern Italy, although there were considerable variations between PMPs and years. Ozone concentration data were further processed by AMORIELLO *et al.* (2003) in relation to meteorological variables. Principal Component Analysis (PCA) and multiple regression models confirmed relationships between  $O_3$  and an energy component (solar radiation, number of hours of sunshine, temperature) which accounted for 40-50 % of the variance, and a wind-related component (13-20% of the variance). A third component varied widely between sites and its nature was not fully understood.

### Development of methods: from weekly concentration to AOT40 estimates

Weekly concentrations are useful, but are not consistent with the definition of exposure indices used to identify potential risk for vegetation, like the  $O_3$  Accumulated Over a Threshold of 40 ppb (AOT40, FUHRER *et al.* 1997; KARLSSON *et al.* 2003). The AOT40 index is based on hourly values which can only be obtained by direct measurement; alternatively, AOT40 can be estimated by statistical techniques (*e.g.* TUOVINEN 2002). An "empirical" method to estimate AOT40 values starting from weekly data was developed and tested by GEROSA *et al.* (2003 this volume). When compared to measured data, the method was found to perform reasonably well in comparison with automatic devices ( $R^2=0.93$ ; median difference: 15.4%; median difference for remote sites: 6.08%). This is an important achievement that will enable further work in the direction of estimates of AOT40 for Italy.

### Evidence of potential $O_3$ risk to forest ecosystems: exceedance of critical levels

On a 5-years basis, estimated June-September AOT40 ranges from 4000 to 26000 ppb  $h^{-1}$  (GEROSA *et al.* 2003). These results provide documented evidence that critical levels (both the one set in Kuopio, 1996, and the new one suggested in Gothenburg, 2002, for sensitive vegetation under sensitive conditions) are exceeded at many PMPs throughout Italy. It is worth noting, however, that this critical level may identify a potential risk, not the actual risk which depends on a suite of site and environmental factors (KARLSSON *et al.* 2003), which are in part discussed by ALIANIELLO *et al.* (2003 this volume). Yet data about AOT40 exceedances are important, especially since they are estimated at site level, and not obtained by large-scale modelling. The AOT40 estimates provided by GEROSA *et al.* (2003) are conservative in that they are relevant to 2/3 of the usual computational period. In addition, most AOT40 values are estimated starting from concentration at measurement height (2 m), not at canopy height. Actual April-September AOT 40 values at canopy height may therefore be much higher than those reported by GEROSA *et al.* (2003).

### Evidence of $O_3$ effects on forest ecosystems: visible symptoms, crown condition, growth

The assessment of ozone effects on the vegetation at the PMPs considered by the programme was carried out by two approaches. One approach was the assessment of visible  $O_3$ -like symptoms on trees, herbs and shrubs (BUSSOTTI *et al.* 2003 this volume). In 2001 and 2002,  $O_3$ -like symptoms were identified on 57 species, some of which were not yet in the pan-European list of sensitive species (BUSSOTTI *et al.* 2003). Among symptomatic species, there were important tree species, including *Acer platanoides*, *A. pseudoplatanus*, *Carpinus betulus*, *Fagus sylvatica*, *Picea abies*, various *Fraxinus* species, *Tilia cordata* and *Ulmus minor*.

The second approach was based on modelling crown transparency and basal area increment (BAI) of dominant trees of the main tree species (MTS) in the various PMPs and subsequent statistical analysis of residuals (measured – predicted) vs. AOT40 (FERRETTI *et al.* 2003b). The results are controversial and strongly influenced by the limitation of the available datasets. Increasing crown transparency at increasing level of AOT40 was reported for beech, but not for Norway spruce. Decreasing BAI with increasing AOT40 was reported for Norway spruce and deciduous oaks, but not

for beech. The failure to obtain consistent relationships between  $O_3$  and response indicators was attributed to several reasons: the role of modifying factors (EMBESON *et al.* 2000), the uncertainty surrounding  $O_3$  measurements and AOT40 estimates (BUFFONI and TITA 2003; GEROSA *et al.* 2003), the nature of the response indicators used (especially crown transparency, DOBBERTIN *et al.* 1997). An approach based on ozone flux will certainly be more effective for Risk Assessment. Unfortunately, GEROSA and ANFODILLO (2003) show that, at the current stage of the programme, flux calculation with an acceptable level of confidence was not possible.

#### ***“Side benefits”: results obtained are relevant not only to $O_3$***

A number of additional results were obtained. Cooperation between scientists is always an achievement, and it was strengthened in the various meetings and bilateral contacts held to prepare this report. Insight in factors potentially affecting tree and ecosystem condition was another important achievement. Results reported by FERRETTI *et al.* (2003b this volume) also provide clear indications about the role of site and environmental factors affecting growth and tree condition: in particular an interesting relationship was found between soil/foliar mineral nutrition and crown transparency of beech and Norway spruce. This will be further used in future to investigate cause-effect relationships.

## **Problems**

### ***Calibration sites: more are needed***

Passive sampling for monitoring  $O_3$  in forests offers practical advantages, but an adequate validation system is essential. Among the sites considered in this report, co-located automatic measurement devices able to provide data for validation/calibration existed at only one location (VAL1). At another location (TOS1), the device was within 2 km radius with respect to the passive sampler. These two devices are useful (see BUFFONI and TITA 2003; GEROSA *et al.* 2003), but need to be supplemented for a stronger calibration. New sites recently added to the programme (namely BOL1) are equipped with active monitors, and additional co-located monitors would be of great help, especially in southern Italy.

### ***Design of the monitoring programme: the need for an improvement***

In the first I&C report (FERRETTI 2000) it was stated that "...allocation of monitoring plots over a

wide array of ecological situations (for example: different prevailing tree species) has been proven to be a strong limitation for data analysis... one should think whether – at national level - is useful to spread monitoring sites over a wide range of forests, with very different species composition, stand structure, history, age and site condition...". It "can cause constraints in data analysis unless long or very long time series will be available". Almost all the various chapters of the present report again emphasize the limitation of the dataset, exacerbated by fragmentation of sites due to very different stand types (see FERRETTI *et al.* 2003a). We think this is a serious question that needs to be addressed: although every ecosystem is unique and great care should be taken when grouping PMPs, a minimum of stratification is needed to favour data analysis. At the same time, adequate sample numbers should be ensured for each stratum.

### ***Measurement priorities: learn from the current results***

The most serious problems encountered are relevant to the implementation of a flux-based approach to risk analysis. Such problems came from the lack of suitable data in terms of variables measured, as well as their spatial coverage and temporal resolution. The lack of hourly  $O_3$  concentration is the most obvious problem, although it was partially solved by the approach of GEROSA *et al.* (2003). The problem of  $O_3$  concentrations needs also to be addressed from the perspective of measurement location with respect to horizontal distance from the PMP and height of measurement. In particular, concentration at canopy level are essential. Concern about meteorological data has already been expressed by FERRETTI (2000). Data on the physical properties of soil and the atmosphere (either measured directly or estimated from other data) remains a major problem (GEROSA and ANFODILLO 2003) and the possibility of extending and intensifying meteorological data collection should be seriously considered. The need for soil data (especially data on soil water content) should be addressed soon: the next soil survey (foreseen for the period 2005-2006) provides an opportunity to collect the basic data needed.

## **Perspectives**

### ***Continue to measure $O_3$***

The 5-year data discussed in this volume forms the basis for robust evaluation of forest exposure to  $O_3$  at the beginning of the CONECOFOR programme

(baseline condition). The data confirm that  $O_3$  is a factor of concern for Italian forests. We therefore think that  $O_3$  measurement needs to be continued and improved in terms of time and spatial coverage and consistency (*e.g.* time resolution, horizontal and vertical location) to meet the requirements of flux modelling. This will enable changes in  $O_3$  exposure to be estimated and a better insight into the effects of  $O_3$  on forests.

#### ***Improve meteorological, soil and response indicators***

From the above it is evident that efforts are needed to improve meteorological and soil variables measurements, as well as assessment of response indicators. Crown transparency and basal area increment are non specific indicators of tree condition, whereas visible foliar symptoms are more specific, but with less clear implications with trees' performance. Only an integrated approach which use all the available data, may provide valuable insights into the impact of  $O_3$  on forests. However, the current manner of the visible symptoms survey cannot provide quantitative data for statistical analysis: improvements in sampling design and measurement scales are essential. We recommend revision of the current approach in favour of a more effect-oriented monitoring programme.

#### ***"Ozone at intensive monitoring plots in southwestern Europe": an opportunity for a step ahead***

In July 2002, the EU decided to cofinance a joint project between France, Italy, Spain and Switzerland. The aim of the project is to evaluate  $O_3$  levels, risks, actual and potential effects on the vegetation at the intensive monitoring plots of southwestern Europe. The project will provide a chance for a step ahead in the assessment of  $O_3$  effects on forest vegetation under real field conditions and using routine monitoring data.

## **Conclusions**

Italian research activities in the field of ozone effects on vegetation started 20 years ago with field observation of crops and have subsequently developed into a number of studies on crops, natural and seminatural vegetation, and forests, with connections at international level. In this respect, Italian scientists are involved in the most important international initiatives aimed to assess and monitor air pollution effects on forests and vegetation carried out under the auspices of the EU and UN/ECE. However, no attempt has been made for summarizing existing data. This second report

of the I&C Task Force is the first attempt at national level to provide a consistent, documented overview of  $O_3$  risk in selected forest ecosystems in Italy. On a 5-year basis, and under conservative condition, results presented here show that the PMPs of the CONECOFOR programme are exposed to  $O_3$  AOT40 ranging from 4000 to 26000 ppb  $h^{-1}$ , with maximum yearly values around 45500 ppb  $h^{-1}$ . A number of plant species were found to have  $O_3$ -like symptoms, and likely effects on growth and crown condition of trees, though not always clear, were nevertheless reported. These data confirm that  $O_3$  is a potential factor of risk for Italian forests. Effects of  $O_3$  on forest are also relevant to other major issues on the environmental agenda: among them, conservation of biodiversity (species affected by  $O_3$  may be less fit in competition), forest health and vitality ( $O_3$  effects on crown transparency), and C sequestration ( $O_3$  effects on growth) are all issues of concern for sustainable forest management, which may therefore be impacted by  $O_3$ . Together with the above risk, these connections demonstrate the need to continue and possibly improve monitoring. The way ahead was indicated by recent workshops held in Harrogate (June 2002) and Gothenburg (November 2002), as well as the recent update of the mapping manual of the UN/ECE programme. Although AOT40 is still maintained as an indicator of potential risk, there is a clear demand for a flux-based approach and this should be the direction of our future work. The cooperative project on  $O_3$  levels and effects in southwestern European forests recently cofinanced by the EU may provide the opportunity to go in this direction.

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