

Ozone and nitrogen dioxide measurements in the framework of the National Integrated Programme for the Control of Forest Ecosystems (CONECOFOR)

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ABSTRACT

Ozone (O_3) concentrations were monitored using passive (diffusive) samplers at 20 permanent monitoring plots of the National Integrated Programme for Forest Ecosystem Monitoring (CONECOFOR) during late spring and summer from 1996 to 2000. Nitrogen dioxide (NO_2) concentrations were measured at the same sites with passive samplers during the same months in 1999 and 2000. Measurement devices were placed at a short distance from the forest plots in areas with freely circulating air. The exposure period for O_3 passive samplers was 1 week; NO_2 samplers were exposed for 7 days in 1999 and 14 days in 2000. Mean O_3 concentrations recorded by the passive samplers for the monitoring periods in individual years vary from 75.2 to 125.8 $\mu g m^{-3}$, showing considerable differences between individual years and the geographical locations of the monitoring sites. Generally speaking O_3 concentration levels are higher at the plots located in Central and Southern Italy. The highest mean weekly concentrations were recorded in Sicily with a maximum of 171 $\mu g m^{-3}$. No correlation can be found between O_3 levels and elevation if all the plots are considered, while a significant increase of O_3 levels with elevation can be observed in the six plots located in the Alpine region. The correlation analysis of 5-year mean concentration data and plot latitude show a moderately close relationship. Mean weekly concentrations often exceed 65 $\mu g m^{-3}$ as 24 h mean, one of the short term thresholds proposed for the protection of vegetation. Exceedances are recorded in all monitoring periods. At the permanent monitoring observation plot in Sicily this threshold was exceeded in all the years of the study. Mean weekly or fortnightly NO_2 concentration levels are generally very low, with the exception of some plots influenced by nearby emissions or transport phenomena. NO_2 probably exceeds the critical level for the protection of forest ecosystems at one plot only. No correlation was observed between NO_2 and O_3 concentration data.

Key words: air pollution, forest, passive sampler, ozone, nitrogen dioxide

1. INTRODUCTION

Ozone is a secondary pollutant produced in the atmosphere from anthropogenic precursors i.e. nitrogen oxides (NO_x) and volatile organic compounds (VOC) in the presence of sufficient sunlight. O_3 is the most important gas in the photochemistry of the atmosphere and is the primary constituent of photooxidative smog. It can thus be considered an indicator of the overall burden of atmospheric oxidants. The emissions of precursors for O_3 formation over most of Europe are always sufficient to feed photochemical episodes and keep mean concentrations in spring and summer at levels potentially toxic for human and plant health. This means that O_3 levels depend mainly on the meteorological situation (Beck *et al.* 1998). Meteorology affects the formation, transport, deposition and seasonal behaviour of ozone. The meteorological conditions which favour the formation of ozone are intense solar radiation, low wind speed, high temperature and a restricted boundary layer depth. Radiation and temperature drive the chemical reactions producing ozone, while boundary layer characteristics and the absence of wind are the factors which respectively lead to the build-up of precursors and limit their dispersion.

Ozone concentrations are characterised by seasonal and diurnal variations. O_3 formation increases rapidly during springtime and decreases sharply in the second

half of the summer. Its diurnal behaviour during spring and summer is in accordance with sunshine and daily temperature.

In most of Europe the monthly means vary between 20 and 100 $\mu g m^{-3}$. At remote mountainous sites in central Europe, the monthly means can be higher than 100 $\mu g m^{-3}$ (De Leeuw *et al.* 1997). In northern Scandinavia, the maximum monthly mean measured so far is 80 $\mu g m^{-3}$ (Kulmala *et al.* 1998). Background O_3 concentration data in southern European countries are often scarce and fragmentary, although the conditions for intense photochemical ozone formation are met each year in spring and summer, every time a high pressure cell is stable for more than a few days. Data for the Southern Alps collected at various altitudinal transects during the vegetative period show a clear stratification of pollutant concentrations with elevation (Kirchner *et al.* 1994). Above 1500 m a.s.l. weekly mean concentrations may occasionally reach 160 $\mu g m^{-3}$. EMEP monitoring stations located in Italy (at Ispra in the Western Prealps and at Montelibretti near Rome) show mean concentrations varying from 45 to 80 $\mu g m^{-3}$ during the summer. The monitoring stations at remote sites, however, are not nearly numerous enough to draw any conclusions regarding the background ozone concentration field (De Leeuw & Bogman 2001).

Ozone has harmful effects on human health (WHO 1996) and vegetation. The European Union has set limit

values for ozone concentrations for the protection of both human health and ecosystems. The effects of O₃ on plants are well known, and the current scientific knowledge has been discussed in several reviews (Reich 1987; Heath & Taylor 1997; Grulke 2003). The AOT40 index, accumulated exposure above a threshold of 40 ppb (76.2 µg m⁻³) referring to the period from April to September, has been suggested as a means of estimating the potential risks to vegetation due to ambient O₃ (Fuhrer & Achermann 1994). In addition to AOT40, short term thresholds, 200 µg m⁻³ (1 h mean) and 65 µg m⁻³ (24 h mean), have been adopted in the framework of EU Directive 72/92. These limit values are exceeded quite frequently all over Europe (Leeuw *et al.* 1999).

Nitrogen dioxide (NO₂) is a gaseous pollutant which may also be harmful to plants. The effects caused by NO₂ and its reaction products are broad based. Damage to vegetation is usually greater when NO₂ reacts with other gases such as ozone and peroxyacetyl nitrate (PAN) than when NO₂ acts alone (Cowling 1985).

The effects of NO₂ are more subtle than those caused by O₃, and visible symptoms are difficult to recognise. Generally speaking, at remote sites NO₂ can be considered the best represented nitrogen oxide, as NO is rapidly oxidised to NO₂. A threshold concentration (critical level) based on current scientific knowledge for nitrogen oxides (NO + NO₂) of 30 µg m⁻³ as yearly mean has recently been adopted at European level (EU Directive 99/30).

Nitrogen dioxide measurements in forest areas are rather limited at European level (Photooxidant Research Group 1997). In fact, concentration levels at remote sites do not usually exceed the critical level.

Field studies on air pollutants-ecosystem interactions are generally performed at stand level, due to the cost of continuous air quality measurements. In the last two decades, however, approaches based on passive samplers have met with increasing interest in forest ecosystem research (Manning *et al.* 1996; Blum *et al.* 1997; Krochmal & Kalina 1997; Cox *et al.* 1999; Cooper & Peterson 2000; Cox *et al.* 2001; Krupa *et al.* 2001; Bytnerowicz *et al.* 2002a; Bytnerowicz *et al.* 2002b). In fact, passive samplers can provide valuable information regarding the concentration level and distribution of different pollutants. Passive samplers are relatively inexpensive, easy to handle and do not require electricity, and so are a very attractive option for field measurements. On the other hand, they cannot identify short term pollution episodes, and data provided by passive samplers are unsuitable for calculating accumulated exposure over given concentration thresholds (e.g. AOT40). However, statistical approaches have been proposed to estimate AOT40 values from passive sampling (Gerosa *et al.* in press). Characteristics, advantages and disadvantages of passive samplers are discussed in several reviews (Harper & Purnell 1987; Krupa & Legge 2000). In the framework of the National

Integrated Programme for Forest Ecosystem Monitoring (CONECOFOR), concentrations of O₃ and NO₂ were measured at 20 forest sites. This paper reports the results of the monitoring activity carried out from 1996 to 2000.

2. SAMPLING AND METHODS

2.1. Monitoring sites

Ambient concentrations of tropospheric O₃ were measured at a short distance from 20 of the permanent monitoring plots of the National Integrated Program for Forest Ecosystem Monitoring Network (CONECOFOR). A detailed description of the plots is given in Allavena *et al.* 2000. A number of research activities have been in progress since 1996 at the forest plots within the framework of the Level II forest monitoring programme (Regulation EC n. 1091/94) with the aim of characterising and evaluating interactions between ecosystems and air pollution.

Passive samplers are used to monitor air pollutants. O₃ and NO₂ are measured at sites usually situated less than 1 km from the plots. These sites are characterised by freely circulating air. Samplers are exposed at some distance from potential disturbance sources like buildings, trees and roads. A description of the requirements for the exposure of passive samplers is reported in Buffoni & Tita (2002).

2.2. Ozone monitoring

Measurements of weekly average concentrations of O₃ were performed from the middle of June to the end of September from 1996 to 2000. Monitoring was carried out in the framework of two EU pilot projects (96.60.IT.003.0 and 98.60.IT.003.0) aimed at providing a preliminary picture of O₃ levels in Italian forest sites. Monitoring was thus limited in time, and the measurements do not completely cover the vegetative period.

Ozone samplers used at the permanent observation plots of the CONECOFOR network are based on the passive diffusion of a gas to an absorbing medium, in this case cellulose papers impregnated with the well-known dyestuff indigo (C₁₆H₁₂N₂O₂) (Werner 1992). Ozone reacts with indigo to isatin, which can be determined spectrophotometrically. Diffusion tubes are 57 cm long and have a diameter of 45 cm. Ambient O₃ concentration can be determined according to Fick's first law:

$$c = \frac{Q \times l}{D \times A \times t} \quad (1)$$

where *c* is the ambient O₃ concentration (µg m⁻³), *l* the diffusion path (cm), *D* the diffusion coefficient (cm² s⁻¹), *Q* the amount absorbed (µg), *A* the cross section (cm²), and *t* the exposure time (s). Cross section, length of tubes and diffusion coefficient are constant for a sampling system and express the sampling rate (ml min⁻¹) of a diffusive sampler. As the diffusion coefficient of O₃ is

Tab. 1. Mean O₃ concentrations (in $\mu\text{g m}^{-3}$) during the monitoring periods (weeks 24 – 39) at the permanent monitoring plots.

	ABR1	BAS1	CAL1	CAM1	EMI1	EMI2	FR11	FR12	LAZ1	LOM1
1996	70.2	64.8	59.8	72.8	64.7	69.8	59.6	59.6	69.4	-
1997	87.4	107.4	84.2	113.7	94.2	111.5	71.5	80.1	98.8	73.1
1998	82.1	69.0	64.4	74.3	76.4	83.7	76.8	67.8	84.4	67.3
1999	87.4	83.5	78.1	95.0	78.8	91.7	88.9	87.2	88.6	73.4
2000	86.4	99.8	87.0	93.7	92.8	81.2	78.5	80.7	84.7	76.4
'96-'00	82.7	84.9	74.7	89.9	81.4	87.6	75.1	75.1	85.2	72.5
	MAR1	PIE1	PUG1	SAR1	SIC1	TOS1	TRE1	UMB1	VAL1	VEN1
1996	71.4	67.1	68.2	71.2	83.7	63.9	66.0	61.8	67.7	64.3
1997	105.0	83.8	108.5	84.6	122.7	79.2	95.7	91.3	88.3	77.0
1998	76.4	67.2	89.4	68.2	92.1	74.0	80.0	66.3	80.9	70.3
1999	103.2	90.2	95.9	95.2	101.7	89.7	92.0	91.7	89.2	84.3
2000	94.0	83.4	91.6	90.1	103.8	82.7	84.6	87.9	85.5	76.1
'96-'00	90.0	78.3	90.7	81.9	100.8	77.9	83.6	79.8	82.3	74.4

not known, an empirical coefficient is determined by parallel exposure of passive samplers with co-located UV monitors.

Passive samplers were calibrated during the monitoring periods against a continuous O₃ analyser placed in a mountainous Alpine location. Correlation between the two devices shows a statistically significant correlation (Buffoni & Tita 2000; Buffoni & Tita, in press). The same kind of passive samplers were used previously in other monitoring studies in forest areas (Werner *et al.* 1999) and were checked for correlation with continuous UV monitors at several sites (Hangartner *et al.* 1996).

Five samplers were exposed in parallel in 1996, three in 1997, while in the last three years measurements were based on the exposure of one passive sampler only. Operators belonging to the National Forest Service or other regional Agencies were asked to fill in a form specifying the start and end of each exposure period, relevant meteorological events during the week, and the presence of occasional disturbance sources near the measurement sites.

2.3. Nitrogen dioxide monitoring

Continuous NO₂ monitoring is usually carried out by chemiluminescence analysers. The passive samplers adopted (Passam ag) collect NO₂ by molecular diffusion along an inert polypropylene tube. The pollutant is retained by an absorbent (triethanolamine) placed at the end of the tube. The devices (9.5 internal diameter and 74 mm length) are placed in a special shelter to protect them from rain and minimise wind influence. The NO₂ collected is determined spectrophotometrically by the Saltzman method.

Nitrogen dioxide samplers were exposed at the same locations already identified for O₃ measurements. Concentration data were collected from the middle of June to the end of September in 1999 and 2000. Samplers were exposed to ambient air for a duration of 7 days in 1999 and 14 days in 2000. One sampler was used for each exposure period.

3. RESULTS AND DISCUSSION

3.1. Ozone concentration levels

A 5-year dataset of weekly O₃ measurements for the period from the 18th to the 39th of the year is available for the permanent investigation plots of the CONECOFOR network. Less than 4% of data for this period are missing. Mean O₃ concentrations for the different monitoring periods are given in table 1. With the exception of 1996, a year with abnormally low O₃ concentrations, mean O₃ concentrations generally exceed 80 $\mu\text{g m}^{-3}$ at almost all the plots. In certain years characterised by long-lasting high pressure conditions (e.g. 1997), the mean concentrations recorded at several plots are above 100 $\mu\text{g m}^{-3}$. Plot SIC 1, located in Northern Sicily, shows the highest mean values among the plots of the CONECOFOR network in all the years considered, with a maximum in 1997. Generally speaking, the permanent observation plots in Central and Southern Italy show higher mean O₃ concentrations than the plots in the Po Plain and in the Alps.

The 5-year mean O₃ concentration may be adopted to give a preliminary characterisation of O₃ concentration levels. The Alpine plots show mean concentrations between 70 and 85 $\mu\text{g m}^{-1}$, while the concentrations in the Central Apennine plots are generally between 80 and 90 $\mu\text{g m}^{-1}$. In the South, apart from plot CAL1, the mean concentrations recorded are above 90 $\mu\text{g m}^{-1}$.

Using the 5-year mean concentrations as a yardstick, 1996 can be considered as a low O₃ year in all plots, 1997 as a high O₃ year. A detailed discussion about differences in individual years is reported in Buffoni & Tita (in press). The differences in individual years fully justify the general recommendation of a 5-year mean for integrated assessment purposes (Federal Environmental Agency 1996).

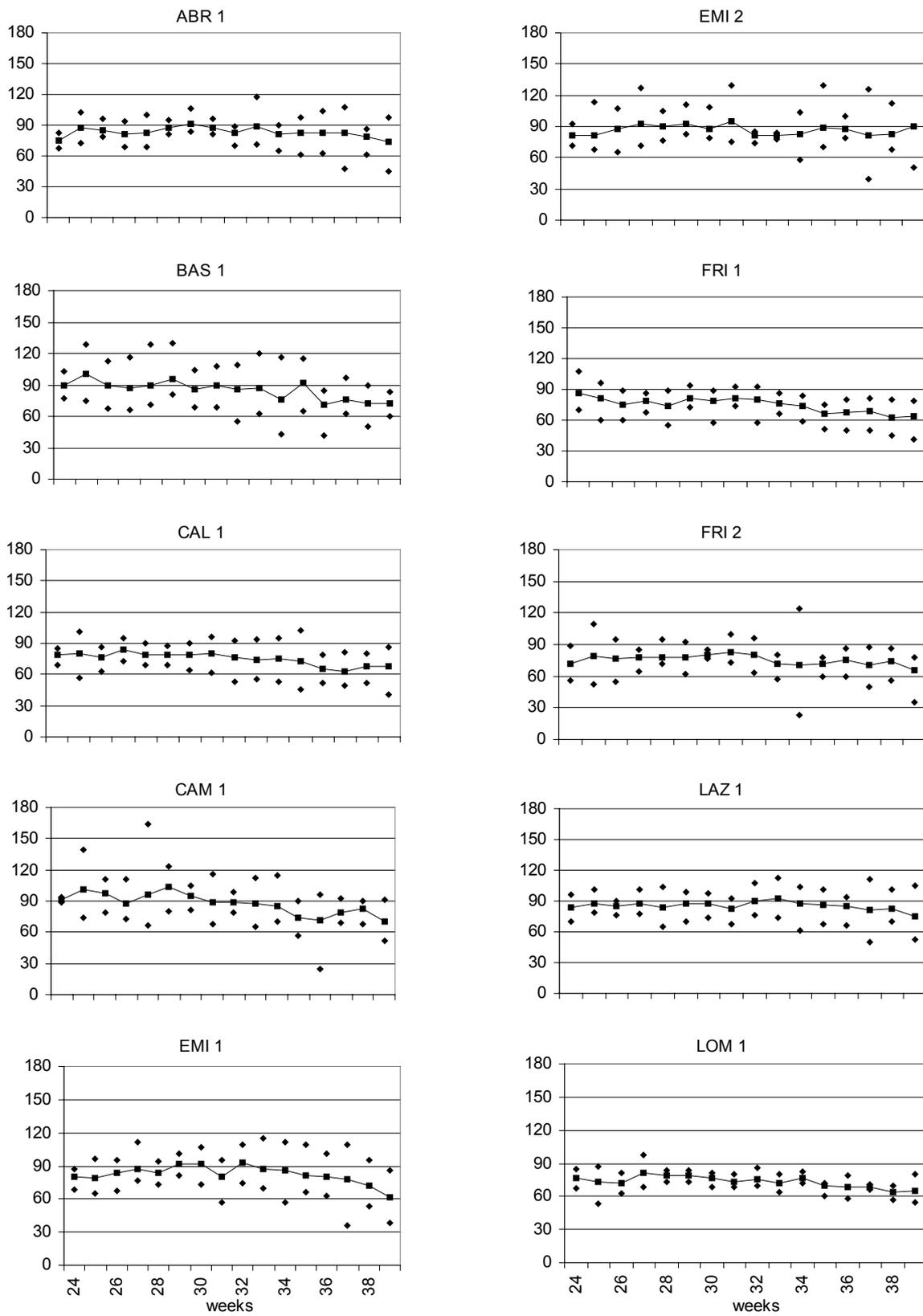


Fig. 1a. Ozone 5-year means, minima and maxima ($\mu\text{g m}^{-3}$) of individual years during the monitoring periods.

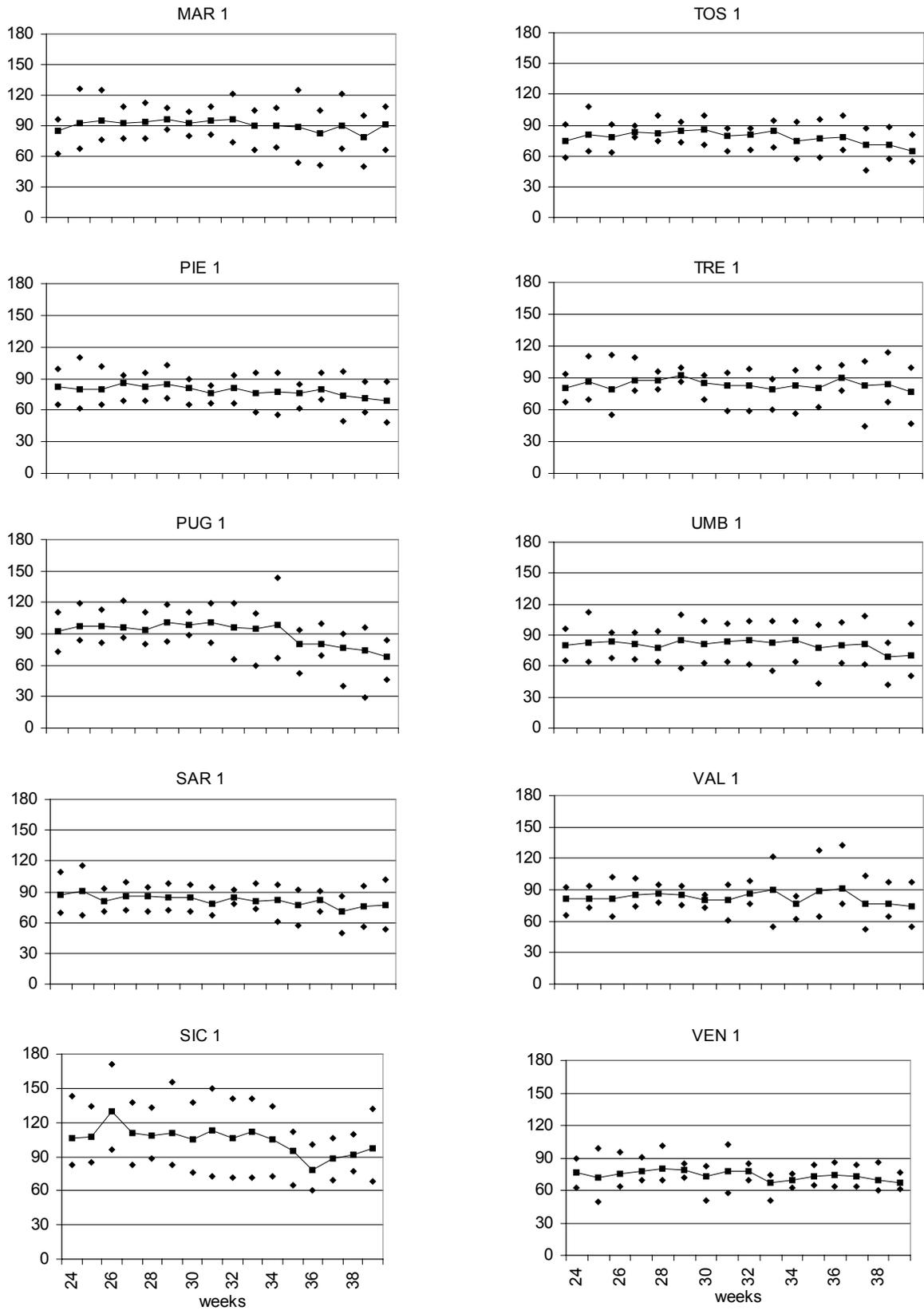


Fig. 1b. Ozone 5-year means, minima and maxima ($\mu\text{g m}^{-3}$) of individual years during the monitoring periods.

Data from four EMEP monitoring stations: Ispra (45°48'N 8°38'E) and Montelibretti (45°48'N 8°38'E), located in Italy, Vorhegg in Austria (46°40'N 12°58'E) and Malta (36°06'N 14°12'E) show substantially consistent average O₃ concentrations for the period June – September in the years of the study. The data from the two Italian stations, 54.6 and 65.0 µg m⁻³ respectively as 5-year means for the same monitored periods, reflect the influence of the nearby urban areas and are generally lower than those of the plots in the same geographical region. The mean O₃ concentration at Vorhegg (78.6 µg m⁻³) is similar to the O₃ levels at the Alpine plots, while the O₃ level recorded in Malta (101.5 µg m⁻³) is comparable with the concentrations observed at the plot located in Sicily (SIC 1).

Figure 1 illustrates the 5-year mean O₃ concentrations at the permanent observation plots and the minima and maxima recorded in the different years for each individual week. At most of the plots the maximum concentrations are recorded in the middle of the summer. Concentrations generally decrease rapidly in September.

In the Alpine region some plots (e.g. LOM1) show very constant concentrations over the period monitored. At plot VAL1, a typical remote mountainous site, concentration variability is very limited, while unsettled weather conditions towards the end of the summer mean that concentrations may vary substantially from year to year. Plot FRI1, located in the Po Plain, is characterised by a relatively low O₃ concentration but high NO₂ levels. The constant O₃ levels observed may reasonably be attributed to the low wind activity typical of this area.

Most of the plots in the inner Apennines show comparable patterns, with variations between individual years limited to ±20% compared to the 5-year mean. Plots located at very open and windy sites like BAS1, CAM 1 and SIC 1, may show a higher variability.

Correlation analysis was performed on O₃ concentration data and geographical parameters of the permanent monitoring plots such as latitude and elevation. A slight correlation can be observed between the 5-year mean concentrations and latitude ($r = 0.50$; $p < 0.01$) (Fig. 2), while the correlation between elevation and the mean of yearly maxima is less evident ($r = 0.51$; $p < 0.05$). No correlation could be found between mean and maximum concentrations with plot elevation. If the 6 plots (VAL1, PI1, LOM1, TRE1, VEN1, FRI2) located in the Alpine region are considered, a statistically highly significant correlation can be observed between plot elevation and the 5-year mean concentrations ($r = 0.84$; $p < 0.001$) (Fig. 3), but not with the O₃ maxima. This result confirms previous observations (Kirchner *et al.* 1994) regarding the stratification of ozone along elevation gradients in Alpine valleys. The same analysis carried out for the plots located in the Central Apennines did not highlight any significant correlation.

Regarding the potential risks for forest ecosystems, a comprehensive analysis of the results of the numerous

research activities carried out in the framework of the CONECOFOR programme is in progress (Ferretti *et al.*, in press) and the exposure of forest ecosystems to O₃ has been evaluated.

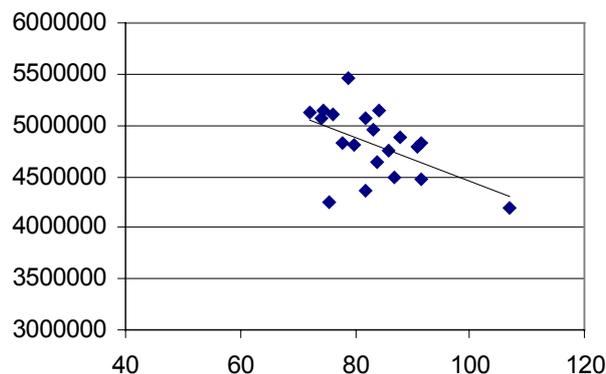


Fig. 2. Relationship between latitude and O₃ 5-year mean concentrations (µg m⁻³).

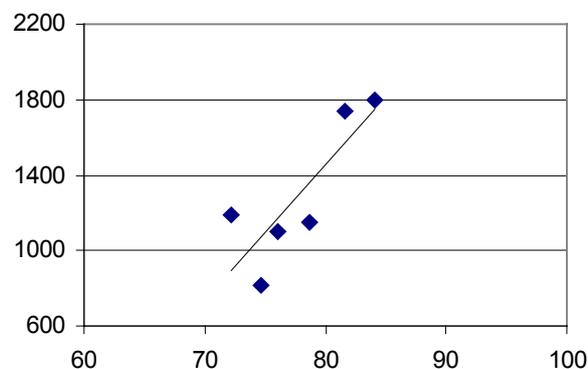


Fig. 3. Relationship between elevation (m) of Alpine plots and O₃ 5-year mean concentrations (µg m⁻³).

If we consider the O₃ concentrations measured at the permanent monitoring plots, some preliminary indications of potential risks related to O₃ levels emerge from the data reported here. In fact, if we look at the threshold for the protection of vegetation compared to the daily mean O₃ concentrations (33.1 µg m⁻³), it is clear that frequent high exceedances occur. A mean weekly concentration above 33.1 µg m⁻³ means that the threshold was not respected on at least one day of the week. Exceedances for the different years considered in this study are reported in table 2. The overall exceedance (1996 – 2000) of the weekly mean concentrations considered ranges from 77.8 to 97.5% of the monitored weeks.

Exceedances were more frequent in 1997 and fewer in 1996. These observations are confirmed by the data collected by continuous analysers at national level (De Leeuw *et al.* 1999). In fact, in 1996 exceedances of the 24 h threshold recorded at urban, suburban and background stations were half those observed in 1997.

Tab. 2. Weekly mean O₃ concentrations (in %) above the threshold of 65 µg m⁻³ in the monitoring periods.

	ABR1	BAS1	CAL1	CAM1	EMI1	EMI2	FRI1	FRI2	LAZ1	LOM1
Missing data	2.6	10.4	1.3	18.2	1.3	7.8	1.3	2.6	0.0	20.8
1996	68.8	62.5	31.3	75.0	31.3	81.3	42.9	25.0	81.3	80.0
1997	100	80.0	100	40.0	100	80.0	73.3	100	100	80.0
1998	100	66.7	53.3	80.0	66.7	93.3	80.0	86.7	100	73.3
1999	100	100	93.3	100	86.7	86.7	100	100	100	93.3
2000	87.5	93.8	100	93.8	100	100	100	100	93.8	93.8

	MAR1	PIE1	PUG1	SAR1	SIC1	TOS1	TRE1	UMB1	VAL1	VEN1
Missing data	3.9	0.0	0.0	0.0	0.0	1.3	0.0	0.0	1.3	2.6
1996	68.8	62.5	68.8	75.0	100	43.8	56.3	31.3	62.5	31.3
1997	100	100	100	100	100	100	100	100	100	100
1998	93.3	86.7	100	80.0	100	93.3	100	80.0	86.7	66.7
1999	100	100	100	100	100	100	100	100	93.3	100
2000	93.8	100	100	100	93.8	100	93.8	100	100.0	81.3

Tab. 3. Mean and maximum NO₂ concentrations (in µg m⁻³) recorded at the permanent monitoring plots in 1999 and 2000.

		ABR1	BAS1	CAL1	CAM1	EMI1	EMI2	FRI1	FRI2	LAZ1	LOM1
1999	mean	2.1	3.1	2.1	1.6	11.3	4.9	31.5	11.1	2.7	2.0
	max	3.2	4.8	6.5	3.6	14.4	23.8	40.9	14.8	4.4	2.9
2000	mean	3.6	n.a.	1.3	1.1	9.2	3.4	30.5	9.8	2.0	1.3
	max	8.9	n.a.	2.7	1.6	13.3	11.6	38.4	12.2	3.5	1.8

		MAR1	PIE1	PUG1	SAR1	SIC1	TOS1	TRE1	UMB1	VAL1	VEN1
1999	mean	3.3	2.7	3.4	2.3	5.7	6.8	2.2	3.8	2.2	3.1
	max	9.4	5.5	5.5	4.2	18.5	12.2	3.5	8.4	4.7	4.5
2000	mean	2.4	2.1	2.6	1.5	4.5	7.2	2.6	5.4	1.4	3.5
	max	2.9	3.9	3.9	2.1	9.3	11.8	9.0	7.1	2.0	5.5

3.2. Nitrogen dioxide concentration levels

Nitrogen dioxide concentrations measured by passive samplers at the permanent observation plots are reported in table 3. Most plots show very low mean concentrations during the spring and summer, with the exception of FRI1, FRI2, EMI1 and EMI2. At these plots the influence of nearby NO_x sources should be considered. Typically, NO₂ concentration are higher during the cold season, and thus the threshold value (critical level) for the protection of ecosystems of 30 µg m⁻³ (NO + NO₂) as an annual mean is probably exceeded at plot FRI1. With reference to the maximum weekly or fortnight concentrations, values above 10 µg m⁻³ up to 25 µg m⁻³ can be recorded occasionally at several plots (EMI1, EMI2, FRI2, TOS1 and SIC1). At plot FRI 1 the concentrations measured are constantly above 25 µg m⁻³. Correlation analysis carried out on O₃ and NO₂ concentration data show no significant (inverse) relationship.

4. CONCLUSIONS

Ozone and nitrogen dioxide measurements were carried out from 1996 to 2000 in the permanent plots of the National Integrated Program for Forest Ecosystem Monitoring Network (CONECOFOR). The data collected give a preliminary picture of the concentration

levels of the most important pollutants in these remote areas. Temporal variations and differences among plots may be used to categorise forest areas and risk levels. Passive samplers here show their potential for air quality assessment in remote areas. However, site selection and standardised procedures for monitoring have to be carefully planned.

Regarding O₃ levels, plots in Central and Southern Italy show generally higher concentrations, probably due to the more intense solar radiation and to a longer duration of daytime. In addition, the more unstable meteorological conditions recorded in the Alpine valleys partly limit the formation of O₃. The collected O₃ data show that the threshold for the protection of ecosystems of 65 µg m⁻³ referred to the 24 h mean was exceeded at all plots. In fact, most of the mean weekly concentrations are above this threshold. Future measurements should include complete coverage of the vegetative period.

Moreover, the O₃ data collected in this study are particularly important, as for some Italian regions this is the only information available regarding O₃ levels and temporal variations in remote areas.

Nitrogen dioxide concentration are generally very low, with mean concentrations below 10 µg m⁻³. The only exception is that of plot FRI 1 located in the Eastern part of the Po Plain. The area is affected by the emission of different nearby NO_x sources (traffic and

industries), and the passive samplers were able to highlight the highly probable exceedance of the critical level for the protection of ecosystems set at 30 $\mu\text{g m}^{-3}$ as yearly mean.

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