

Estimates of ozone AOT40 from passive sampling in forest sites in South-Western Europe

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Ozone AOT40 estimates from passive sampling predict significantly real time data and reveal frequent exceedance of ozone critical levels for forest vegetation in South-Western Europe.

Abstract

Weekly-fortnightly ozone (O_3) concentrations measured by passive sampling at 81 forest monitoring plots in France, Italy, Spain and Switzerland over the period 2000–2002 were used to estimate the cumulative exposure index AOT40. The estimation method is based on a deterministic model 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) and the time of the day. Estimated AOT40 values ($AOT40_e$) were evaluated against co-located automatic measurement stations and with 14 independent automatic stations located throughout Italy whose weekly mean O_3 values were used to simulate passive samplers. AOT40 can be predicted by modelling passive sampling data (R^2 : 0.90; $P < 0.0001$, SE of estimates: 3271 ppb h), although considerable deviations can occur for individual sites. Estimated AOT40 shows a distinct, significant latitudinal and altitudinal gradient. Taking the 3-year average as a whole, exceedance of critical level of 5000 ppb h occurs at 77–100% of the monitored sites, respectively.

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1. Introduction

Despite several criticisms (e.g. Matyssek and Innes, 1999), ozone (O_3) AOT40 (O_3 accumulated over a threshold 40 ppb, $1 \text{ ppb} = 1 \text{ nl l}^{-1}$) remains the basis for estimating the potential risk of forests due to O_3 and for setting environmental quality objectives within the European Union (EU) and the United Nation Economic Commission for Europe (UN/ECE) (e.g.

Directive 2002/3/EC; UN/ECE, 2004; Karlsson et al., 2003). Under this perspective, the knowledge about AOT40 values at forest monitoring sites is of considerable interest. The AOT40 is defined as the sum of the hourly O_3 concentration exceeding the threshold of 40 ppb over the period 1 April–30 September (a recent revision of the definition consider the vegetative period, which varies according to the species and the geographical location UN/ECE, 2004). For the purposes of this paper, however, we maintained the 1 April–30 September period. Only the daylight hours are considered (global radiation $> 50 \text{ W m}^{-2}$) (Kärenlampi and Skärbi, 1996; Fuhrer et al., 1997). Proper calculation of AOT40 implies the availability of complete hourly O_3 measurements throughout a 6-month period. Unfortunately, hourly O_3 measurement is uncommon in forests, a situation in which passive sampling offers considerable practical advantages (Krupa

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et al., 2001; Cox et al., 2001; Karnosky et al., 2003). Given the nature of the technique, passive sampling typically results in time-integrated data, such as weekly-to-monthly O₃ concentration, which are not consistent with the definition of AOT40 (see Tuovinen, 2002; Ferretti and Gerosa, 2002). For this reason, there were several attempts to estimate hourly values and cumulative or summation indices (as the AOT40) starting from integrated mean O₃ concentration obtained from passive samplers (Gerosa et al., 2001; Krupa et al., 2001, 2003; Krupa and Nosal, 2002; Tuovinen, 2002). In a previous paper, Mazzali et al. (2002) used an iterative procedure based on the functional dependence of O₃ concentrations both on elevation and time of the day (Loibl et al., 1994) to estimate AOT40 in a small (80 × 40 km) domain in Northern Italy on the basis of passive sampling data. In this paper, we test the feasibility of the approach at a larger scale in order to estimate the AOT40 at 81 monitoring plots located in France, Italy, Spain and Switzerland (see Ferretti et al., 2004, submitted for publication, for site location), and on which O₃ was measured by passive sampling in 2000, 2001 and 2002. The method only requires O₃ data obtained by passive sampling, geographical coordinates and elevation of the site. If proven feasible, it will permit AOT40 estimates also in cases where no ancillary data (e.g. meteorological data, Krupa et al., 2003) are available. This will enable an analysis of the potential risk to forests only on the basis of passive sampling as well as the validation of large-scale O₃ models. First results for Italy were presented elsewhere (Gerosa et al., 2003).

2. Materials and methods

2.1. O₃ data and data completeness

Details about O₃ monitoring sites are in Ferretti et al. (2004). Ozone concentrations obtained from passive samplers were collected at all the sites over the period 2000–2002. Data completeness, methods of measurement and reliability of O₃ data are reported by Sanz et al. (submitted for publication). Data completeness over the period 1 April–30 September is crucial for the calculation of AOT40. Data completeness can be expressed in terms of % of days covered by validated data over the reference measurement period. Differences in data completeness between sites and countries occurred mostly because of the different starting dates of passive measurements. For example, in 2002 measurements started at the beginning of April in France and Spain, between 1 and 23 April in Italy and between 29 April and 19 June in Switzerland (Sanz et al., submitted for publication). In addition, cases of invalid data affecting data completeness were reported (Sanz et al., submitted for publication). Here we considered 70% as a threshold for valid data completeness, and we discarded the sites that did not reach this threshold (Table 1).

Table 1
Percent of sites that match the 70% data completeness threshold between 1 April and 30 September for different years and countries

Country	Year					
	N	2000 Valid (%)	N	2001 Valid (%)	N	2002 Valid (%)
France	26	100	26	100	27	100
Italy	20	100	26	96.1	26	100
Spain	12	91.7	12	100	12	100
Switzerland	5	40	5	60	15	93.3

N = number of operational sites for a given year and country.

2.2. Modelling O₃ daily profile

Loibl et al. (1994) and Loibl and Smidt (1996) reported a function describing the hourly O₃ concentration 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:

$$O_3(h_r, t) = a_1 + a_2 e^{-(t-a_3)^2 a_4} \ln \left(\frac{h_r}{100} + \frac{b_1 t^2 + b_2 t + b_3}{b_4 t^2 + b_5 t + 10,000} e^{-b_6 t} \right) \quad (1)$$

where

h_r is the relative altitude in meters (see below),

t is the daytime,

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

The function was obtained as the best fit of a series of O₃ measurements carried out at more than 100 sites distributed over a range of h_r in Austria. Although the function was proven to fit well also for Italian forest sites (Gerosa et al., 2003), it should be considered that it represents a mean, ideal situation which may be subject to interferences, such as O₃ advection from areas with high photochemical production and O₃ depletion by nearby NO_x emissions. For this reason, deviations in hourly concentrations can be expected according to the situation of individual sites, and, at the same site, for individual days.

The pattern of the O₃ daily profile was modeled for each site according to Eq. (1) and using the h_r calculated for each site. Then, the O₃ daily profile was adjusted in order to match the mean O₃ concentration measured by the passive samplers, assumed as the 24-h daily average over the measurement window covered by passive sampling. The resulting O₃ daily profile was replicated for each day of the measurement window (i.e., 7 or 14 days, see Sanz et al., submitted for publication) (Fig. 1). The underlying, simplifying assumption is that the invariance of the daily O₃ profile, which is considered to be constant every day of the measurement period (Mazzali et al., 2002). Thus, deviations between measured and modelled hourly O₃ concentration for individual days are implicit in the assumption of the model, but they are expected to compensate over the April–September period (i.e., the computational period of AOT40) (Fig. 2). In this perspective, passive sampling on a weekly basis is intuitively more suited than on a fortnightly or monthly basis. This is because the strength of the assumption needed is somewhat proportional to the length of the measurement window. As a consequence, the uncertainty of estimates is expected to increase with the length of the exposure period of passive samplers.

The modeled O₃ concentration of the hours with global solar radiation > 50 W m⁻² were considered to estimate AOT40 values (AOT40_c) for each site. When direct radiation measurements were not available, we assumed global solar radiation > 50 W m⁻² occurring between dawn and sunset, and we estimated the number of hours by means of an astronomic model based on latitude, longitude of the site, the calendar date and the time of the day (Strahler, 1984; Scire et al., 1989). In order to avoid underestimation of the AOT40 in the case of missing data, the “raw” AOT40 calculated on the basis of the available measurements was weighted according to (Eq. (2)):

$$AOT40 = \frac{AOT40_{\text{raw}}}{N_{\text{dA}}} N_{\text{dT}} \quad (2)$$

where:

N_{dA} is the number of the valid days of measurements available;

N_{dT} is total number of the days of the measurement period.

2.3. Validation

The validation has been carried out with the following two approaches:

- (i) direct comparison with co-located automatic measurement devices. When there were co-located real time analyzers and passive samplers, measured

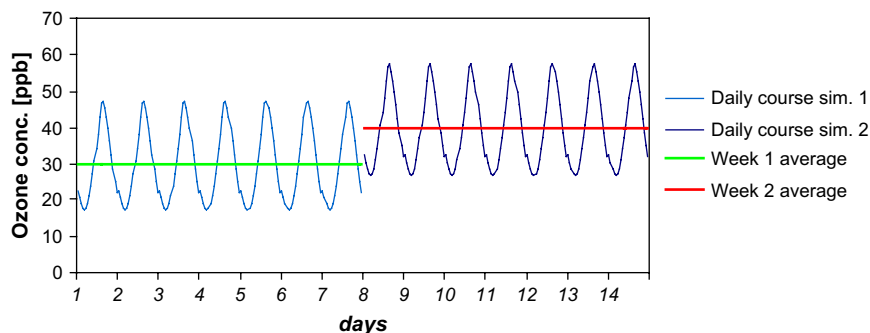


Fig. 1. Example of how does the procedure work: the daily course expected from Eq. (1) is adjusted on a weekly basis in order to obtain a week mean value equal to the weekly ozone concentration obtained by passive samplers.

(AOT40_m) and estimated (AOT40_e) AOT40 values were compared. Direct comparisons were possible for 3 sites in Spain (years 2000–2002, according to the site), 2–3 sites in Italy (years 1996 and 1998 to 2002, according to the site) and 1 site in Switzerland (years 2000–2002) (Table 2).

- (ii) indirect comparison with simulated passive sampling. The O₃ concentration measured on a hourly basis by 14 real time analyzers in Italy was averaged – for each site – over a weekly basis to simulate the passive sampling data (sensu Tuovinen, 2002) (see Gerosa et al., 2003 for site details and data completeness checks). Then, the O₃ daily profile was estimated for each site according to the Eq. (1) and after having calculated the h_t value for each site. AOT40 values were calculated and missing data were managed according to Eq. (2). The aim of the indirect validation is to supplement the dataset in checking the validity of the model and in comparing measured and estimated values.

3. Results

3.1. Performance of the model: validation against automatic measurements

Table 2 reports the data used to verify whether AOT40 estimated by passive sampling data is a significant and precise predictor of actual AOT40 values measured by automatic devices. In Table 2, a “calibration” site means a site where passive samplers and automatic devices were co-located; “simulated” means an automatic device used to simulate a passive sampler as described above. The median absolute percent difference between measured and estimated AOT40 for co-located (“direct”) calibration sites is 13.6% (Table 3). Deviation > 20% (in absolute terms) occurs mostly in specific cases (Table 2):

- (a) when AOT40 is very low (see VAL1, Italy, 1996, and La Peira, Spain, 2001). In these cases, even relatively small differences in terms of ppb h give rise to high % differences. Note that for the Spanish site there are only two series of measurements (14 and 28 days, respectively) and this may also affect the results.
- (b) In case of prolonged exposure periods. For example, median difference between AOT40_m and AOT40_e is 14.4% for sites with 2-week sampling periods and 9.3% for sites with 1-week sampling periods (Table 3). The reason for this is clear when considering Fig. 1. Longer sampling periods will smooth O₃ episodes and can introduce a further approximation in the model.

A second comparison was carried out using simulated passive sampling by weekly O₃ concentration averages obtained from automatic measurements. The median absolute difference is 19.4% (Table 3), i.e., higher than for co-located calibration sites. These data must be interpreted taking into account the nature of the “simulated” passive sampling sites (Gerosa et al., 2003): several sites are located in suburban conditions where nearby anthropogenic NO_x and VOC emissions and/or remixing phenomena may cause O₃ depletion or generate short peak events; on the other hand, O₃ advection may occur downwind of areas with high photochemical production. Both these processes may substantially affect the daily O₃ course, the invariance of which is the basic assumption of the model. Accordingly, Gerosa et al. (2003) report that differences between measured and estimated values were much less for rural and remote sites than in suburban conditions. Fig. 3 reports the complete dataset (co-located + simulated). The model ($R^2 = 0.90$, $P < 0.0001$, standard error of estimate = 3271 ppb h) underestimates AOT40 values below 10,000 ppb h, and tends to overestimate values higher than 40,000 (Fig. 3). Measured and estimated values resulted not

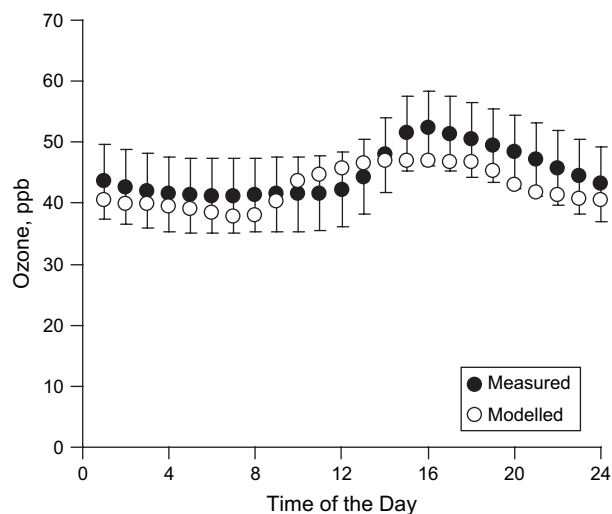


Fig. 2. Modelled hourly O₃ concentration (empty circles) plotted together with measured values (filled circles). Error bars indicate the standard deviation of the mean hourly values over the period 3 April–30 September 2001 ($n = 169$ –172), site VAL1, La Thuile, Northern Italy, 1740 m a.s.l.

Table 2
“Calibration” and “simulated” sites with measured and estimated AOT40 (ppb h)

Site type	Site name	Year	Country	Frequency of passive sampling	AOT measured	AOT40 estimated	Relative difference (%)	Absolute difference (%)
Calibration	VAL1	1996	Italy	1	3621	1676	−53.7	53.7
Calibration	VAL1	1998	Italy	1	7921	7758	−2.1	2.1
Calibration	VAL1	1999	Italy	1	14118	15429	9.3	9.3
Calibration	TOS1	2000	Italy	1	32200	34387	6.8	6.8
Calibration	VAL1	2000	Italy	1	12358	12250	−0.9	0.9
Calibration	Gandia ^a	2000	Spain	2	16717	10072	−39.7	39.7
Calibration	La Peira ^a	2000	Spain	2	12119	12917	6.6	6.6
Calibration	Latecaldo ^b	2000	Switzerland	1	32473	40231	23.9	23.9
Calibration	VAL1	2001	Italy	1	16383	19061	16.3	16.3
Calibration	BOL1	2001	Italy	1	29562	28107	−4.9	4.9
Calibration	Gandia	2001	Spain	2	17625	15083	−14.4	14.4
Calibration	La Peira ^c	2001	Spain	2	1986	459	−76.9	76.9
Calibration	Sant Jordi	2001	Spain	2	25944	19614	−24.4	24.4
Calibration	Latecaldo ^b	2001	Switzerland	1	30944	39317	27.1	27.1
Calibration	VAL1	2002	Italy	1	18101	13770	−23.9	23.9
Calibration	BOL1	2002	Italy	1	28488	29438	3.3	3.3
Calibration	Gandia ^a	2002	Spain	2	12677	11055	−12.8	12.8
Calibration	La Peira ^a	2002	Spain	2	19994	9931	−50.3	50.3
Calibration	Sant Jordi ^a	2002	Spain	2	23533	21826	−7.3	7.3
Calibration	Latecaldo ^b	2002	Switzerland	2	21981	23116	5.2	5.2
Simulated	Fontechiari	1996	Italy	(1)	20946	17602	−16.0	16.0
Simulated	Ispra	1996	Italy	(1)	17579	11614	−33.9	33.9
Simulated	Ispra	1997	Italy	(1)	15766	11143	−29.3	29.3
Simulated	Bormio	1998	Italy	(1)	20897	21421	2.5	2.5
Simulated	Cimetta	1998	Switzerland	(1)	35254	43631	23.8	23.8
Simulated	Erba	1998	Italy	(1)	28489	28891	1.4	1.4
Simulated	Ispra	1998	Italy	(1)	18304	8095	−55.8	55.8
Simulated	Olgiate C.	1998	Italy	(1)	17516	13595	−22.4	22.4
Simulated	Brione	1998	Brione	(1)	18540	18543	0.0	0.0
Simulated	Massenzatico	1999	Italy	(1)	1036	786	−24.1	24.1
Simulated	Fontechiari	1999	Italy	(1)	25547	24199	−5.3	5.3
Simulated	Pieve Teco	1999	Italy	(1)	5594	2240	−60.0	60.0
Simulated	Boccadifalco	1999	Italy	(1)	28312	24167	−14.6	14.6
Simulated	Settignano	1999	Italy	(1)	23352	18819	−19.4	19.4
Simulated	Oss. Napoli	2000	Italy	(1)	5566	3564	−36.0	36.0
Simulated	Leonessa	2000	Italy	(1)	45659	52391	14.7	14.7
Simulated	Chiavenna	2000	Italy	(1)	10122	9455	−6.6	6.6

Data refer to common measurement periods. Frequency of passive sampling is expressed in number of weeks. Frequency reported in brackets indicates a simulated passive sampling. Data from the calibration site VAL1 (1996–2000), TOS1 (2000) and from simulated sites after Gerosa et al. (2003).

^a Occurrence of 1 and 2 months exposure periods for passive sampling.

^b AOT40_m based on a multiplexed sampling device. AOT40_e averaged between estimates from two sets of passive sampling.

^c Only 2 measurement periods available.

significantly different (Wilcoxon matched pair test, $P = 0.054$).

3.2. Estimates of AOT40

AOT40 estimates for the sites of interest are in Fig. 4. Although considerable variation exists, there is a distinct decrease of AOT40 along a latitudinal gradient ($R^2 = 0.51$, $P < 0.001$) and a less marked, still significant increase with elevation ($R^2 = 0.21$, $P < 0.01$). This confirms the pattern reported by Dollard et al. (1995) and the concentration data described by Sanz et al. (submitted for publication). Sites in Northern France have almost always a much lower AOT40 than sites in Italy and Spain. A cluster of sites with very high AOT40 values is also obvious at the border between Switzerland and Italy, a well-known target area for O₃ pollution

Table 3

Median absolute and relative differences between measured and estimated AOT40 for direct and indirect comparisons and for different sampling regimes (when applicable)

Sampling regime	Direct			Indirect		
	N	Median relative difference (%)	Median absolute difference (%)	N	Median relative difference (%)	Median absolute difference (%)
1-Week	11	3.3	9.3	17	−16.0	19.4
2 Weeks and more	9	−14.4	14.4	—	—	—
Total	20	−3.5	13.6	—	—	—

See text for details.

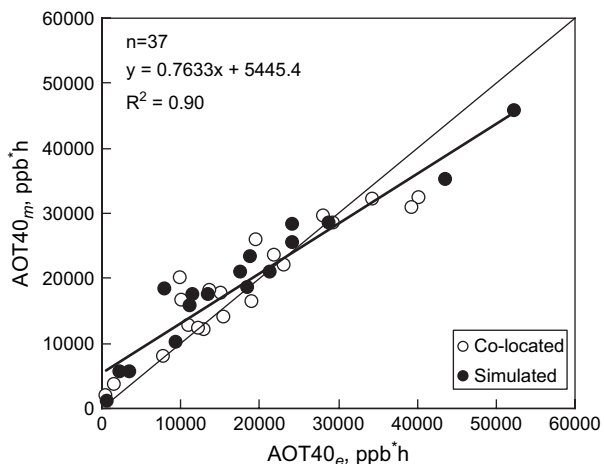


Fig. 3. Measured AOT40 (AOT40_m) regressed against estimated AOT40 (AOT40_e). Sample size, coefficient of determination and regression equation are reported. Bold line: regression function; light line: 1:1 line.

(e.g. Novak et al., 2003) (Fig. 4). Among the sites considered in this paper – which must not be considered a statistically representative sample of the forests within the investigated countries – exceedance of the UN/ECE critical level 5000 ppb h is common (Table 4). Exceedance of the EU target

value set for 2010 (9000 ppb h, calculated over the period May–July) and of the long-term objective (6000 ppb h, calculated as above) is also possible at many sites.

4. Discussion and conclusions

Since AOT40 continues to be the basis for estimating the potential risk of forests due to O₃ and for setting environmental quality objectives within the EU and the UN/ECE, there is a clear need to obtain information about it. A key point is that forest sites in remote areas cannot always be equipped with expensive real time O₃ analysers, which are needed to obtain hourly concentration values. On the other hand, large-scale models (e.g. Jonson et al., 1999; Simpson et al., 2001) are useful for large-scale evaluation and policy negotiations, but model output is not applicable at the site level and therefore cannot be used in risk analysis at local level. This is particularly true when considering the inherent uncertainty associated to large-scale models (see below). In this paper O₃ passive sampling data were used to estimate the AOT40 exposure index for 81 forest sites in France, Italy, Spain and Switzerland. Unlike other approaches (e.g. Krupa et al., 2003), the one presented here only needs concentration values obtained from passive sampling, coordinates of the site, and elevation data

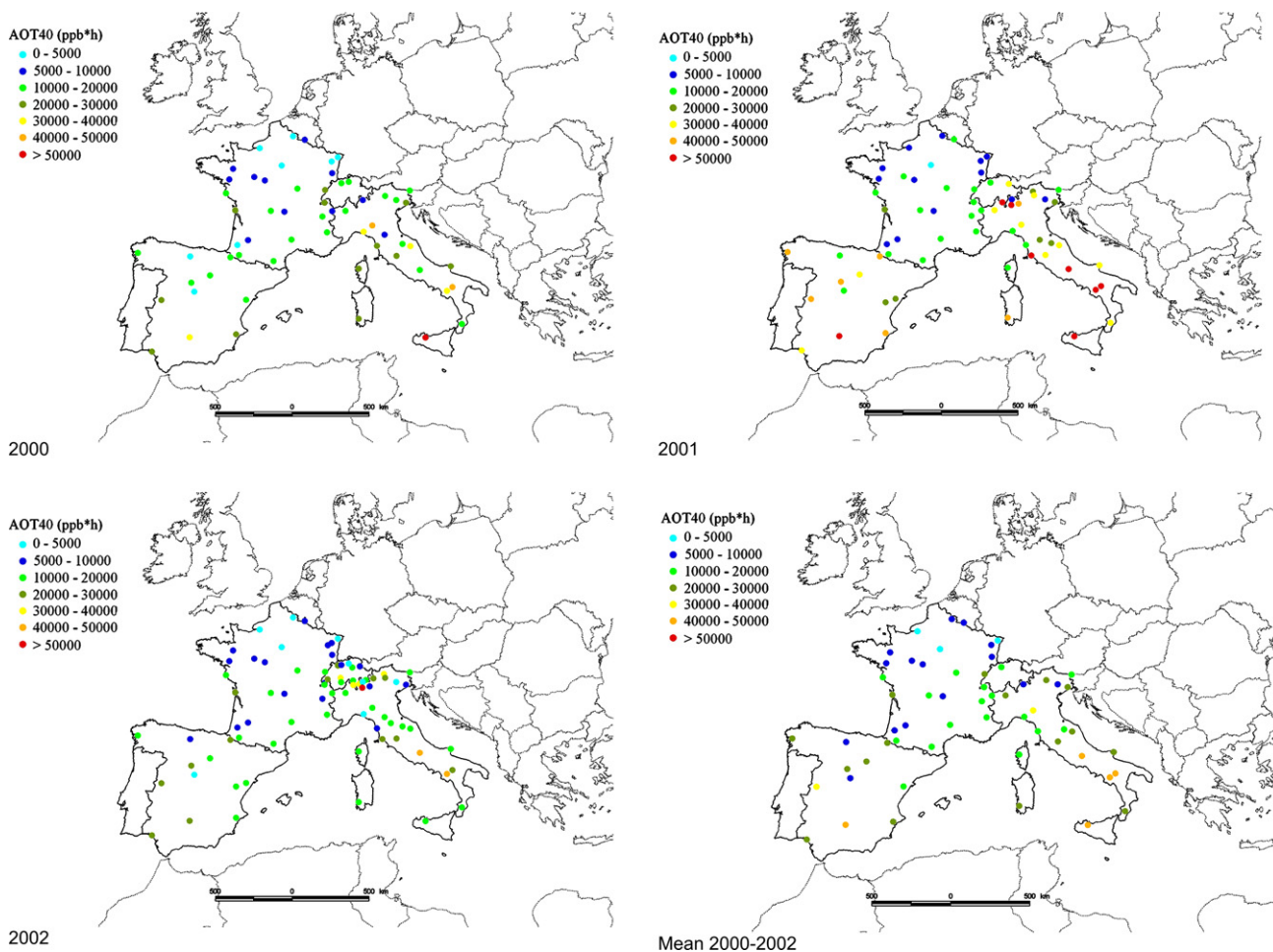


Fig. 4. Estimated AOT40 values for the various sites in the 4 countries involved in the project.

Table 4
Summary information about AOT40 levels at the common sites over the period 2000–2002

	2000	2001	2002	Mean plot values (2000–2002)
Median AOT40 ppm h	10.5–17.1	15.1–21.6	10.6–17.1	12.8–19.4
% Of sites > 10 ppm h	56.1–78.9	61.4–89.5	52.6–78.9	56.1–85.9
% Of sites > 5 ppm h	73.7–100	80.7–100	73.7–100	77.2–100

The range is calculated taking into account the standard error of the estimates (3.27 ppm h). For each plot the standard error was subtracted and added to the actual estimate, thus generating the minimum and the maximum value of the range. Data are reported for individual years and as 3-year average ($N = 57$).

and makes it possible to obtain AOT40 estimates for sites with no infrastructure (e.g. real time analysers, meteorological stations). The main conclusions of this paper are as follows:

- (i) AOT40 modelled according to the Loibl equation is a significant predictor of measured “true” AOT40. Median deviation between measured and estimated AOT40 values was 13.6% at co-located sites (direct comparison) and 16% overall (direct + indirect comparison) with a standard error for estimates of 3271 ppb h. However, considerable differences can occur for individual sites and for individual hours and days at the same site and this suggests that a lot of caution is needed when interpreting the data. The absolute deviation (%) is greater for sites with passive sampling carried out on a 2-week (or longer) basis and for low AOT40 values. This is a clear indication that 1-week should be considered as a target sampling frequency for passive measurements, at least in southern Europe.
- (ii) The estimates reported in this paper represent the first site-related, measurement-based AOT40 dataset available at international level for remote sites in Europe. This can be helpful from different perspective, including the validation of large-scale models and the use of the hourly estimates obtained for O₃ flux calculations (see Schaub et al., in this volume). In this perspective, the precision and accuracy of hourly estimates need to be explored further.
- (iii) Estimated AOT40 suggests that – over the period 2000–2002 – critical levels of 5000 ppb h were exceeded at the majority of the monitored plots and the same is likely for EU target value set for 2010 (9000 ppb h, calculated over the period May–July) and long-term objective (6000 ppb h, calculated as above) is also possible at many sites. However, it is known that proper assessment of risk is best done on a 5-year basis in order to account for remarkable year-to-year variation in the O₃ summer production (UBA, 1996). Since only 3 years of data are considered, our results should be viewed as a preliminary finding. In addition, great care should be used in interpreting these results in terms of potential risk (Hogsett et al., 1997; Karlsson et al., 2003): while it is obvious that high exposure is a factor of risk (and this is the basis for concentration-based critical levels), it is also known that sensitivity to O₃ varies within species according to genetic and phenological characteristics and ecological condition. For example, it is acknowledged that beech provenances from southern Europe are less sensitive to

O₃ (Paludan-Müller et al., 1999); at the same time, ecological conditions in southern European sites (e.g. high VPD) may prevent O₃ uptake, thus limiting the potential for adverse effects (e.g. Emberson et al., 2000). In this case, the combined effect is that the current concentration-based UN/ECE critical level may overestimate O₃ risk for beech forests in southern Europe.

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