

A simple linear model for estimating ozone AOT40 at forest sites from raw passive sampling data†

Marco Ferretti,^{*a} Fabiana Cristofolini,^b Antonella Cristofori,^b Giacomo Gerosa^c and Elena Gottardini^b

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A rapid, empirical method is described for estimating weekly AOT40 from ozone concentrations measured with passive samplers at forest sites. The method is based on linear regression and was developed after three years of measurements in Trentino (northern Italy). It was tested against an independent set of data from passive sampler sites across Italy. It provides good weekly estimates compared with those measured by conventional monitors ($0.85 \leq R^2 \leq 0.970$; $97 \leq \text{RMSE} \leq 302$). Estimates obtained using passive sampling at forest sites are comparable to those obtained by another estimation method based on modelling hourly concentrations ($R^2 = 0.94$; $131 \leq \text{RMSE} \leq 351$). Regression coefficients of passive sampling are similar to those obtained with conventional monitors at forest sites. Testing against an independent dataset generated by passive sampling provided similar results ($0.86 \leq R^2 \leq 0.99$; $65 \leq \text{RMSE} \leq 478$). Errors tend to accumulate when weekly AOT40 estimates are summed to obtain the total AOT40 over the May–July period, and the median deviation between the two estimation methods based on passive sampling is 11%. The method proposed does not require any assumptions, complex calculation or modelling technique, and can be useful when other estimation methods are not feasible, either in principle or in practice. However, the method is not useful when estimates of hourly concentrations are of interest.

1. Introduction

Despite much criticism,¹ the AOT40 (Accumulated ozone concentrations Over a Threshold of 40 ppb)² is still the regulatory air quality standard used in Europe for evaluating the risk

that ozone poses to vegetation.^{3,4} AOT40 is relatively easy to calculate because it requires only hourly ozone concentration values which can be obtained from conventional monitors. However, ozone measurements are rarely carried out with conventional monitors in remote forest areas, and in most cases passive (diffusive) sampling is used instead (e.g. ref. 5 and 6). Diffusive samplers do not provide hourly ozone values, only time-integrated ozone concentrations for the entire measurement period (typically 1–2 weeks). For this reason, several methods have been devised to estimate AOT40 from ozone values obtained from passive sampling.^{7–11} The effectiveness of these methods has been found to vary, and the methods also rely on additional data and/or assumptions. For example, when AOT40 is calculated from estimations of hourly ozone concentrations made from time-integrated data, a Gaussian distribution⁷ and/or

^aTerraData environmetrics, Via L. Bardelloni 19, I-58025 Monterotondo M.mo (GR), Italy. E-mail: ferretti@terradata.it; Fax: +39 0566 916681; Tel: +39 0566 916681

^bIASMA Research and Innovation Centre, Fondazione Edmund Mach, Via E. Mach 1, 38010 San Michele all'Adige (TN), Italy

^cUniversità Cattolica del Sacro Cuore, Dipartimento di Matematica e Fisica, via Musei 41, Brescia, Italy

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Environmental impact

AOT40 is a regulatory air quality standard used in Europe to estimate the potential risk that ozone poses to vegetation, and it is calculated from hourly concentration values. However, ozone measurements in forested areas are often carried out by passive sampling that provides time integrated mean ozone concentrations that are not suited for AOT40 calculation. Several models have been developed in the past to solve this problem. Here, a simpler linear model was developed and tested to estimate AOT40 from the mean concentration values. Unlike previous modeling approaches, it has very little data requirements and assumptions, and needs very little computational effort. Thus, it permits calculation of AOT40 estimates when other modeling approaches are unfeasible, either in principle or in practice.

Table 1 Description of datasets: site location, type of monitor, years covered, no. of weeks with data completeness >95%, respective mean ozone concentrations and weekly AOT40. Land use is according to the Corine Land Cover classification (<http://www.eea.europa.eu/themes/landuse/clc-download>); ContUrb: continuous urban fabric; DiscUrb: discontinuous urban fabric; Agr/NatV: land principally occupied by agriculture, with significant areas of natural vegetation; Br-LFor: broad-leaved forest; ConifFor: coniferous forest; MixedFor: mixed forest; and NatGrass: natural grasslands

Site	x UTM WGS84	y UTM WGS84	Elevation m a.s.l.	Land use	Monitor	Year	Week (n) > 95%	[O ₃] ppb	AOT40 ppb h
Borgo Valsugana	689 854	5 102 722	380	ContUrb	Conventional	2007–2009	67	30.8	594
Gardolo	663 108	5 107 970	196	DiscUrb	Conventional	2007	26	28.0	467
Monte Gaza	651 389	5 105 081	1'601	NatGrass	Conventional	2007–2009	56	50.5	1'027
Piana Rotaliana	663 134	5 118 479	211	Vineyards	Conventional	2009	26	34.3	738
Riva del Garda	643 094	5 083 639	73	DiscUrb	Conventional	2007–2009	73	33.2	766
Rovereto L.go Posta	658 357	5 084 026	200	ContUrb	Conventional	2007–2009	67	39.6	764
San Michele a/A	664 104	5 118 111	228	DiscUrb	Conventional	2007	18	30.5	498
Trento Parco Santa Chiara	664 449	5 103 237	203	ContUrb	Conventional	2007–2009	72	33.3	756
Andalo	651 000	5 118 000	1'563	ConifFor	Passive	2009	13	75.0	3'192
Bedollo	681 000	5 118 000	1'400	MixedFor	Passive	2007–2009	39	46.8	952
Brentonico	651 000	5 070 000	1'464	Pastures	Passive	2009	13	90.3	4'491
Canal San Bovo	714 000	5 118 000	1'503	Pastures	Passive	2009	12	73.4	3'103
Canazei	714 000	5 148 000	1'588	ConifFor	Passive	2007–2009	40	51.6	1'632
Castello Tesino	699 000	5 118 000	1'549	ConifFor	Passive	2007–2009	37	45.5	1'072
Cles	651 000	5 133 000	1'698	ConifFor	Passive	2007–2009	39	57.2	1'571
Concei	633 000	5 085 000	1'309	MixedFor	Passive	2007–2009	39	65.6	2'479
Condino	618 000	5 085 000	1'856	NatGrass	Passive	2007–2009	36	77.0	3'202
Coredo	666 000	5 133 000	1'389	ConifFor	Passive	2007–2009	40	50.2	1'186
Faedo	666 000	5 118 000	306	Vineyards	Passive	2007–2009	38	44.9	1'395
Folgaria	666 000	5 085 000	1'210	MixedFor	Passive	2007–2009	39	52.9	1'217
Fondo	666 000	5 148 000	1'375	ConifFor	Passive	2007–2009	39	55.3	1'601
Levico Terme	681 000	5 100 000	1'105	Agr/NatV	Passive	2007–2009	38	44.7	845
Peio	633 000	5 133 000	1'675	ConifFor	Passive	2007–2009	38	45.6	813
Pinzolo	633 000	5 118 000	971	MixedFor	Passive	2009	8	44.4	1'028
Predazzo	699 000	5 133 000	1'464	ConifFor	Passive	2007–2009	38	52.8	1'444
Tione	633 000	5 100 000	719	MixedFor	Passive	2009	8	54.1	1'815
Tonadico	714 000	5 133 000	1'854	NatGrass	Passive	2007–2009	40	68.3	2'903
Trento	666 000	5 100 000	359	Br-LFor	Passive	2007–2009	40	43.3	1'128

a standard, constant daily ozone concentration profile⁹ is assumed. Otherwise, additional information – ranging from meteorological^{8,11} and geographical data⁹ to the standard deviation of hourly concentration values – is required. Assumptions and data requirements may not always be met and this may render AOT40 estimation difficult, complex or unfeasible. In order to overcome the complexity of the previous methods, we propose a simple method for estimating AOT40 values from raw passive sampling data. We will demonstrate that the method proposed is able to provide results which are comparable to those obtained by another modelling technique used for forest sites in the sub-alpine Trentino region and across Italy (*e.g.* ref. 6,9 and 12), but with fewer data requirements and assumptions and less computational effort. Our method is grounded in empirical evidence that there is a close relationship between the mean concentration and AOT40 values^{13,14} and is intended as a rapid tool for estimating AOT40 when other approaches^{7–9,11,12} are unfeasible, either in principle or in practice.

2. Methods

2.1 Concept of the study

The present study is based on the ozone concentration data measured by conventional monitors and by passive samplers over the years 2007–2009 in the Trentino region (Autonomous Province of Trento, northern Italy). Around 79% of the forests in this region are located above 600 m a.s.l., while 59% are above

900 m a.s.l. The main urban areas are in the valley bottom. A total of eight conventional monitoring stations managed by the local Environmental Protection Agency (APPA Trento) were considered: seven are located in urban areas in the valley bottom, between 73 and 380 m a.s.l. (Table 1); one (Monte Gaza) is at a higher elevation rural/forest site (1601 m a.s.l.). Raw hourly concentration ($\mu\text{g m}^{-3}$) data for the period from 1st April to 30th September of each year were converted to ppb (standard condition) in order to calculate AOT40 values (ozone

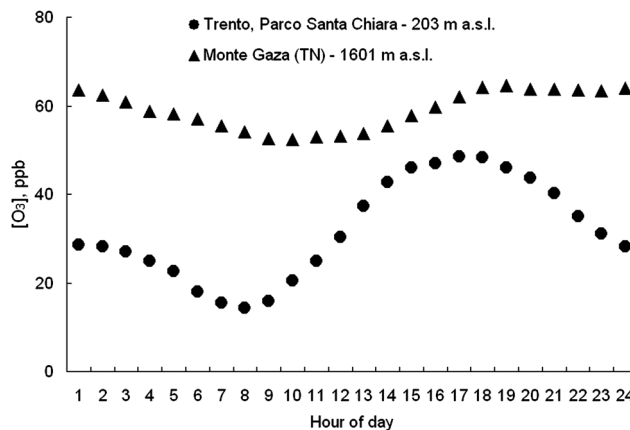


Fig. 1 Typical daily ozone profiles in the Trentino region: mean data from June 2011 for the urban site Parco Santa Chiara in Trento city (dots) and for the high altitude forest site Monte Gaza (triangles).

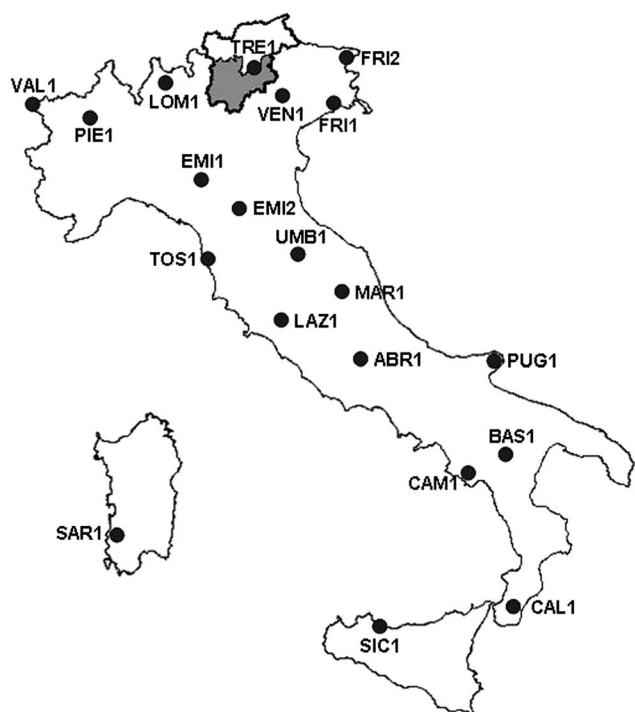


Fig. 2 The Trentino region (in grey) and spatial distribution of the 20 Italian permanent monitoring plots of the CONECOFOR programme where ozone concentrations were measured by passive samplers in the year 2000.

accumulated over a threshold of 40 ppb). Overall, the weekly mean ozone concentrations, as recorded by conventional monitors, ranged from 28.0 to 50.5 ppb; weekly measured AOT40 ranged from 467 to 1027 ppb h for the various monitoring stations.

Weekly mean ozone concentration data were measured by passive samplers located in 15–20 forest sites distributed over the entire 6207 km² of the Trentino region, following a systematic 15 × 15 km grid and belonging to the Level I International Co-operative Programme on Assessment and Monitoring Air Pollution Effects on Forest (ICP Forests) network (for details see Gottardini *et al.*⁶). Measurements covered the period from May

to July of each year (12–14 weeks) (Table 1). Overall, the weekly mean ozone concentrations recorded by passive samplers ranged from 43.3 to 90.3 ppb; weekly AOT40, estimated according to Gerosa *et al.*,¹² ranged from 813 to 4491 ppb h for the various monitoring stations.

The study was organised as follows:

Firstly, a proof of concept was carried out by investigating the relationship between the mean ozone concentration and AOT40 using actual data from conventional monitors, which are expected to be free from major measurement errors. Our intention here is to avoid the problems that one might incur while working with passive sampling data alone. The aim of this first step was to obtain evidence that regression approach is meaningful for calculating AOT40. Due to expected differences in the diurnal pattern of the ozone concentration (*e.g.* ref. 15 and 16) between urban areas (located in the valley bottom in Trentino) and the rural/forest site (located at a higher elevation – see above), data from urban and rural/forest sites were analysed separately. The daily ozone profile of urban areas is characterized by an early-to-mid afternoon peak in concentration and considerable night-time depletion; in rural/forest sites, on the other hand, the night-time concentration remains high¹⁶ (Fig. 1). The resulting regression functions were used to predict weekly AOT40 for these sites on the basis of the mean weekly ozone concentrations.

Secondly, a similar procedure was used to estimate AOT40 from passive monitoring data from Trentino, following Gerosa *et al.*¹² This step was designed to show how well the results of the method presented here compare with those obtained with a recognised modelling technique.^{9,12} It should be pointed out that the latter technique requires information on relative elevation (the difference between the elevation of the monitoring site and the minimum elevation in a 2.5 or 5 km radius), adoption of a standard elevation-based ozone profile model,¹⁵ and the assumption that the daily time course of ozone concentrations is constant over a one-week period.^{9,12}

Thirdly, the method was tested against an independent dataset of ozone concentrations measured with passive samplers at the 20 Italian permanent monitoring plots of the CONECOFOR programme (Fig. 2) during the period of May–July 2000 (13 weeks) and for which AOT40 estimates were available.⁹

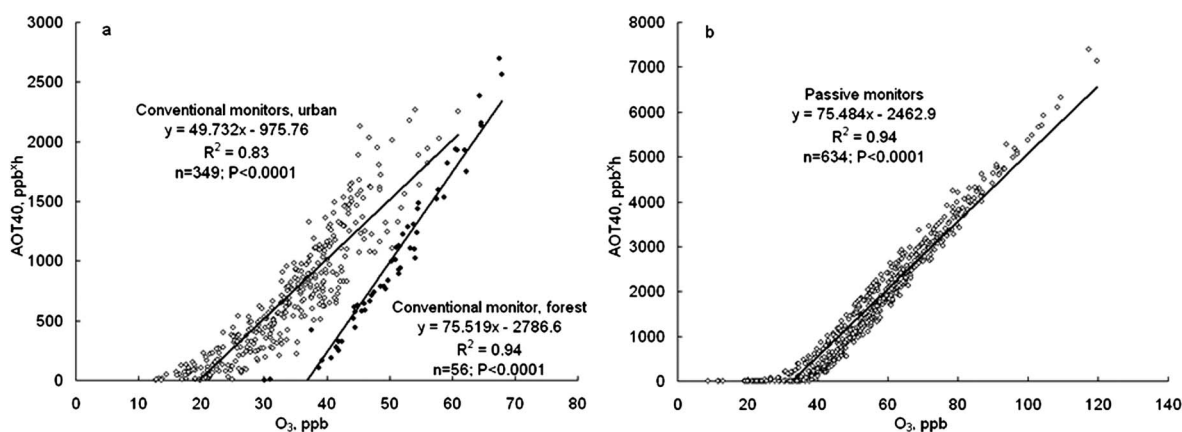


Fig. 3 Regression functions between ozone concentrations and AOT40 values. (a) Regression functions obtained from conventional monitors at urban (empty dots, regression line on the left) and rural/forest sites (black dots, regression line on the right). (b) Regression function from passive monitors.

Table 2 Performance of the regression models and estimates of: (a) measured vs. *F*-estimated AOT40 for conventional monitors and (b) G-estimated vs. *F*-estimated AOT40 for passive samplers

Site category	Sites (<i>n</i>)	Data (<i>n</i>)	<i>R</i> ²	<i>P</i>	RMSE	MAE
(a) Conventional monitor, urban	7	349	0.85	<0.0001	97–302	75–271
(a) Conventional monitor, rural/forests	1	56	0.97	<0.0001	119	93
(b) Passive sampler, rural/forests	20	634	0.94	<0.0001	131–351	99–314

2.2 Data processing

To ensure consistency with passive sampling data, hourly concentration values from conventional monitors were

aggregated and averaged on a weekly basis. Only weekly datasets with completeness of hourly data $\geq 95\%$ were retained. In accordance with EU Directive 2008/50/EU,⁴ hourly exceedances of 40 ppb from 8 am to 8 pm were computed to obtain weekly AOT40 values (referred to as “measured AOT40”). Weekly concentration data from passive samplers were processed to obtain AOT40 values, according to Gerosa *et al.*¹² (referred to as “G-estimated AOT40”).

For both data series (conventional monitors and passive sampling), the weekly AOT40 values were regressed against the mean weekly ozone concentrations using a simple linear regression. The resulting regression functions (*F*) were then used to obtain new AOT40 values based on the mean weekly concentrations (referred to as “*F*-estimated AOT40”). The statistical significance of the linear regressions was evaluated with a

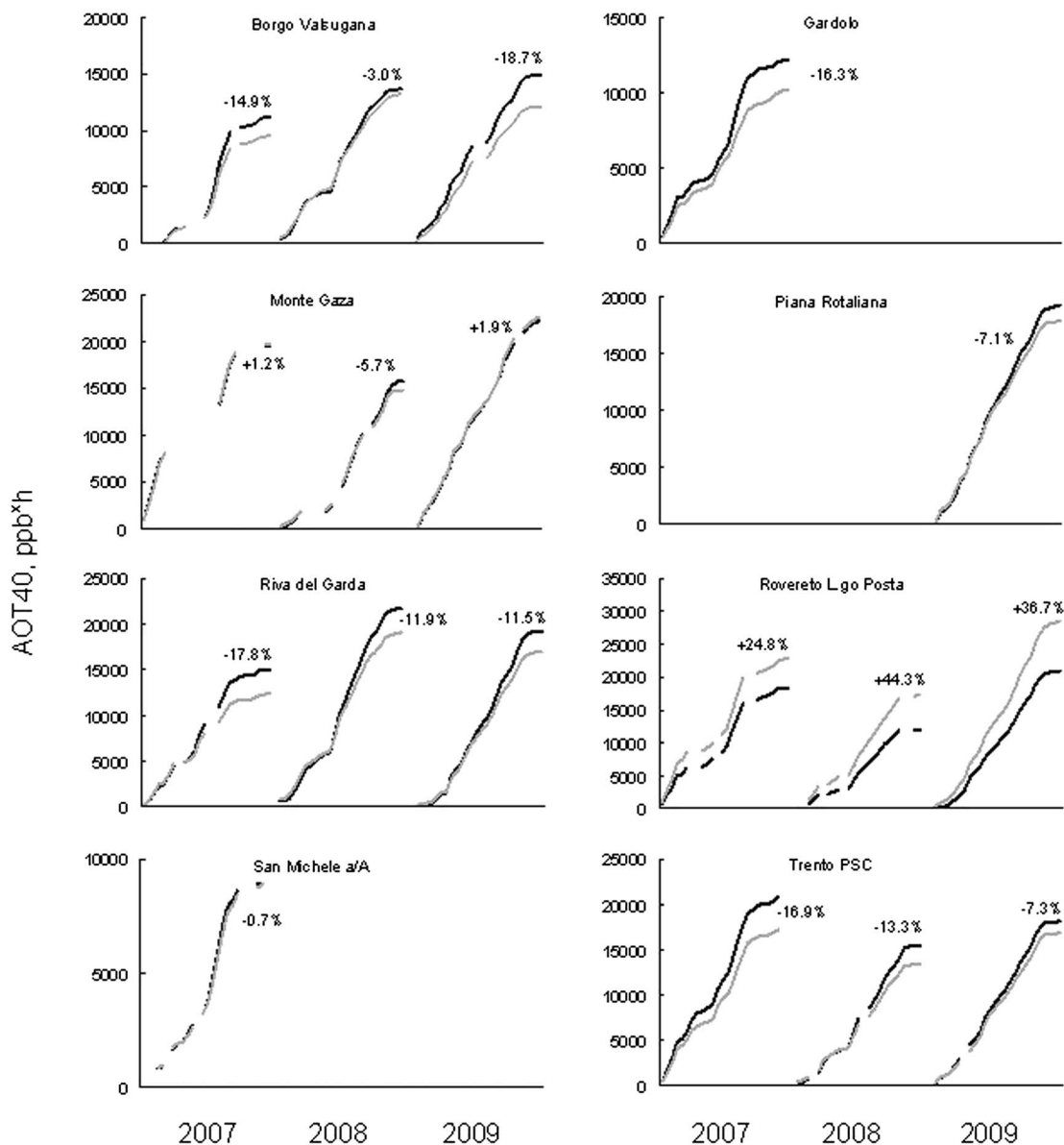


Fig. 4 AOT40 values calculated from data obtained with eight conventional monitors in Trentino (measured AOT40, black lines) compared with values obtained by applying the regression functions shown in Fig. 3a (*F*-estimated AOT40, grey lines) in three different years (when available). Numbers in the graph refer to the percentage differences between seasonally accumulated measured AOT40 and *F*-estimated AOT40.

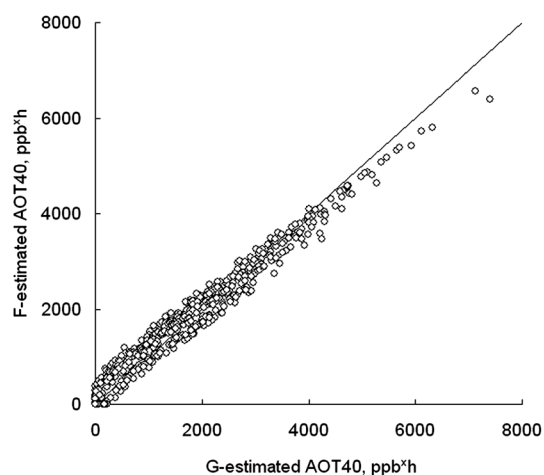


Fig. 5 Weekly *F*-estimated vs. *G*-estimated AOT40, passive monitors in Trentino, northern Italy, years 2007–2009. The 1 : 1 line is reported.

Fisher's test at a 95% probability level. The measured and estimated AOT40 values were compared by calculating the Root Mean Square Error (RMSE), the Mean Absolute Error (MEA) and the percentage difference. Statistical analyses were performed using STATISTICA 9.1 (StatSoft, Inc., Tulsa, USA).

3. Results and discussion

3.1 Proof of concept – conventional monitors

The relationship between ozone concentration and AOT40 has already been reported for different datasets by Mills *et al.*¹³ and Paoletti *et al.*¹⁴ This is confirmed in Fig. 3a which illustrates the relationship between weekly mean ozone concentrations and AOT40 values for urban and rural/forest sites in the province of Trento. Ozone concentrations explain a large part (83–94%) of AOT40 variations, although the slope of the regression is quite

different between urban and forest sites due to the reported differences in daily ozone profile. The higher variability showed at urban sites is due to the inherent differences existing among the seven conventional monitors located at different sites (see Table 1). AOT40 values estimated by the relevant generic functions (*F*) shown in Fig. 3a (*F*-estimated) for urban and forest sites were then compared with measured AOT40 (Table 2, upper part; Fig. 4). As expected from the above results, there is a close relationship between the two datasets ($P < 0.0001$). The root mean square error (RMSE) and mean absolute error (MAE) are reported in Table 2. RMSE varies between 97 and 302 in absolute terms, corresponding in relative terms to 12–26% of measured AOT40; MAE varies between 75 and 271 (absolute values), corresponding to 9–19% of measured AOT40. The lowest RMSE and MAE values, corresponding to the best estimate, were 97 and 75, respectively, for the San Michele all'Adige site; the highest RMSE and MAE values were 303 and 271 and were reported for the Rovereto L.go Posta site. In general, the small difference between RMSE and MAE shows that large errors are unlikely. By way of example, Fig. 4 shows the April–September cumulative course of the measured and *F*-estimated AOT40 for each conventional monitor and year. *F*-Estimated AOT40 values were generally lower than the measured ones, with a maximum underestimation of 9% (Borgo Valsugana, 2009). In only one site (Rovereto L.go Posta) was the *F*-estimated AOT40 higher than the measured AOT40. At this site the three-year mean overestimation was 35%, with the highest RMSE and MAE. The median absolute percentage difference between seasonal values was 13%. It is worth noting that this value is smaller than that reported by Gerosa *et al.*¹² (p. 632) for a similar comparison (19%).

3.2 Proof of concept – passive samplers

The regression functions developed between weekly ozone concentrations obtained from passive samplers at forest sites in

Table 3 Comparison between *F*-estimated and *G*-estimated AOT40 for the period of May–July 2000 at CONECOFOR sites (data for CONECOFOR sites after Gerosa *et al.*⁹)

CONECOFOR site	Long. E	Lat. N	Elevation, m a.s.l.	R^2	P	RMSE	MAE	<i>G</i> -estimated AOT40, ppb h	<i>F</i> -Estimated AOT40, ppb h	<i>F</i> – <i>G</i> -estimated AOT40, ppb h	<i>F</i> – <i>G</i> -estimated AOT40, %
ABR1	10.402900	46.34078	1'500	0.88	<0.0001	401	386	4'099	9'122	5'023	123
BAS1	10.594132	44.95844	1'125	0.99	<0.0001	217	156	17'439	16'884	–555	–3
CAL1	10.622915	42.53802	1'100	0.97	<0.0001	342	333	7'580	11'908	4'328	57
CAM1	10.556363	44.76038	1'175	0.99	<0.0001	219	198	12'471	14'032	1'561	13
EMI1	9.595308	49.52518	200	0.99	<0.0001	306	302	16'048	12'121	–3'927	–24
EMI2	9.669496	48.86040	975	0.98	<0.0001	81	76	7'014	7'866	852	12
FRI1	10.377660	50.76904	6	0.99	<0.0001	478	478	14'909	8'696	–6'213	–42
FRI2	10.412213	51.49680	820	0.99	<0.0001	91	88	10'444	9'304	–1'140	–11
LAZ1	9.737261	47.45346	690	0.99	<0.0001	71	68	7'804	8'541	737	9
LOM1	9.546127	51.21289	1'190	0.99	<0.0001	87	81	5'959	7'015	1'056	18
MAR1	10.363359	47.95266	775	0.99	<0.0001	78	64	15'727	15'638	–89	–1
PIE1	9.427625	50.59399	1'150	0.99	<0.0001	154	148	8'302	10'229	1'927	23
PUG1	10.602369	46.30613	800	0.99	<0.0001	125	112	12'692	13'700	1'008	8
SAR1	9.464162	43.55675	700	0.98	<0.0001	245	234	10'672	12'972	2'300	22
SIC1	10.379967	41.96893	940	0.97	<0.0001	323	269	22'542	21'074	–1'468	–7
TOS1	9.615614	48.18581	150	0.99	<0.0001	282	281	15'031	11'382	–3'649	–24
TRE1	9.691812	51.37093	1'775	0.86	<0.0001	288	261	6'509	9'898	3'389	52
UMB1	10.314899	48.15784	725	0.99	<0.0001	65	58	11'699	12'357	658	6
VAL1	9.341911	50.65317	1'740	0.97	<0.0001	255	247	8'235	11'447	3'212	39
VEN1	10.317576	51.03850	1'100	0.88	<0.0001	261	232	3'651	6'662	3'011	82

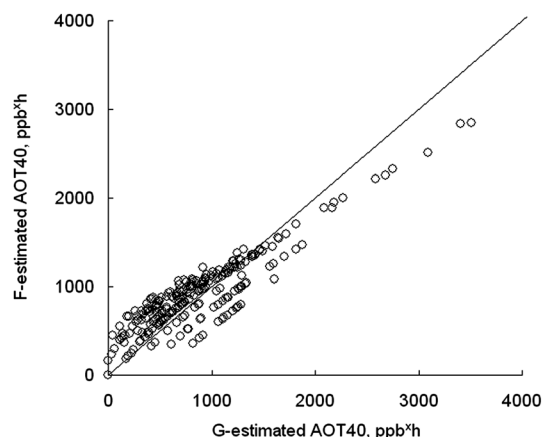


Fig. 6 Weekly *F*-estimated vs. *G*-estimated AOT40, passive monitors at the CONECOFOR plots, Italy, year 2000. The 1 : 1 line is reported.

Trentino and AOT40 estimates obtained according to Gerosa *et al.*^{9,12} are reported in Fig. 3b. Again, the coefficients of determination proved that the mean ozone concentration explains 94% of the variability of AOT40 values ($P < 0.0001$). It is worth noting that the regression coefficients obtained from the conventional monitor at the rural/forest site are quite similar to those obtained from passive samplers at forest sites (Fig. 3a and b). AOT40 values estimated from the generic functions (*F*) shown in Fig. 3b (*F*-estimated) were compared with those estimated according to Gerosa *et al.*¹² (*G*-estimated) (Table 2, lower part). There is a close relationship between the two datasets ($P < 0.0001$); RMSE ranges between 131 and 351, MAE between 99 and 314. In relative terms, they were 7–34% and 6–29% of the *G*-estimates, indicating good precision of estimates. Fig. 5 reports the relationship between the two AOT40 datasets – weekly *G*-estimated and *F*-estimated – and shows data to be very close to the 1 : 1 line.

3.3 Test of concept – independent passive sampling dataset

The generic function developed from the three-year datasets obtained from passive samplers in Trentino were tested against an independent set of data from 20 forest monitoring sites throughout Italy for which *G*-estimated AOT40 was available.⁹ Results are reported in Table 3 for individual sites. R^2 values for weekly AOT40 ranged between 0.86 and 0.99 and all regressions were highly significant ($P < 0.0001$) for individual sites. RMSE ranged between 35 and 325, MAE between 29 and 287; these values are equivalent to 7–127% and 10–123%, respectively, of the weekly AOT40 *G*-estimates. Fig. 6 reports *F*-estimated vs. *G*-estimated AOT40: overall, there is a consistent pattern up to 2000 ppb h per week; higher *G*-estimated values, however, are clearly underestimated. Weekly differences between AOT40 over the May–July period tend to accumulate (Table 3 and Fig. 7): in 10 out of 20 sites the absolute percentage difference between *F*-estimated and *G*-estimated AOT40 was <20% while the median difference across all sites was 11%. This value is again smaller than that reported by Gerosa *et al.*¹² (p. 632) for a similar comparison (19%). However, in two cases the differences between the AOT40 estimates were extremely large (82% at VEN1 and 123% at ABR1).

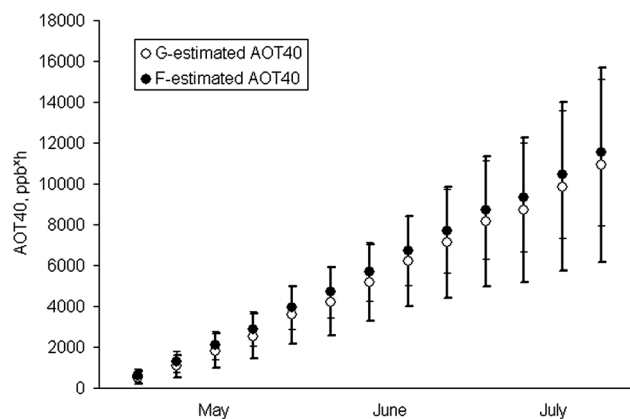


Fig. 7 Mean *G*-estimated AOT40 (filled symbols) for 20 passive sampling sites of the CONECOFOR programme in Italy in individual weeks compared with mean *F*-estimated AOT40 (empty symbols) for the same sites and weeks obtained by the regression functions shown in Fig. 3b. Error bars indicate the standard deviation. Data refer to the period of May–July (x axis) of the year 2000.

4. Conclusions

Weekly AOT40 was demonstrated to be a linear function of the mean weekly ozone concentrations at urban and forest sites, although there were differences between the regression coefficients. A regression equation was therefore calculated and tested to predict weekly AOT40 values at forest sites based on the weekly passive sampling concentration data. The weekly estimates can be used to compute the seasonal (May–July or April–September) AOT40 values. Regression coefficients were very similar for both conventional monitors and passive sampling, thus suggesting that the value of the approach does not depend on the measurement technique. The method provided results comparable to the actual measurements and estimates obtained from more computationally intensive methods. The results were confirmed when the method was applied outside the region of the present study on an independent dataset, and – given the amplitude of geographical and ozone gradients explored – this suggests a possible, much wider application of the generic function for forest sites. For example, passive sampling is carried out at the Level II monitoring sites of the ICP Forests across Europe.¹⁷ In the future, this will offer the chance to test and evaluate the method on a larger dataset.

It is worth noting that the EU Directive 2008/50 considers the use of “objective estimates” and modelling approaches suitable to supplement measurement from conventional monitors. In this line, weekly estimates obtained from passive sampling can be aggregated on a different time resolution, thus allowing relative risk assessment in relation to EU (three months, without distinction between vegetation types) and UNECE (three or six months and/or vegetative period, with difference among crops, semi-natural vegetation and forests) AOT40 standards. While these estimates are to no extent intended to replace information provided by conventional monitors, they can be very useful to obtain information about ozone exposure and for potential risk assessment where monitoring by conventional devices is unfeasible or unsustainable and/or when more complex modelling techniques are unfeasible, either in principle or in practice.

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