Implementing the RAMS-CALMET-CALGRID modelling system to assess seasonal ozone pollution: a five-month application over Tuscany region

G. GUALTIERI[a], F. CALASTRINI[a], C. BUSILLO[a], G. PIROVANO[b]
[a] Laboratory for Meteorological and Environmental Modelling (LaMMA)
CNR-IBIMET
Via Madonna del Piano, 10, 50019 Sesto Fiorentino
ITALY
[b] CESIRICERCA SpA
Dip. Ambiente & Sviluppo Sostenibile
Via Rubattino, 54, 20134 Milano
ITALY
http://www. lamma.rete.toscana.it

Abstract: - The LaMMA laboratory implemented the RAMS-CALMET-CALGRID integrated modelling system to assess ozone pollution at a regional scale. The system is based on the RAMS model meteorological forecasting and the CALGRID photochemical grid dispersion model.

In the present study a comprehensive application of RAMS-CALMET-CALGRID has been carried out on a seasonal basis over Tuscany region, Italy. The study area is 184x224 Km^2 wide, made of a 45x55x12 3-D computational grid, with a horizontal resolution of 4 Km.

The modelling system has been applied in a long term mode on a five-month long (May to September 2002) time period, which can be certainly reckoned as the most original part of the work. As a matter of fact, a number of strong modifications in the original CALGRID code proved to be necessary to let the model run continuously all over the five months of simulation.

The SAPRC-90 “lumped” chemical mechanism developed by Carter has been used to lump all VOCs involved in the O_3 formation and removal processes in the atmosphere. Concerning emission data, all point, line and area sources within the whole region were provided by the regional emission inventory. As far as initial and boundary conditions are concerned, they have been provided as outputs by the CAMx dispersion model applied by CESIRICERCA in the framework of a project aimed at evaluating atmospheric pollution over Italy.

Eventually ozone 1-hour concentrations estimated by CALGRID have been compared with observations provided by the major stations of ozone regional monitoring network, showing a good accuracy degree being performed.

Key-Words: - Ozone, Dispersion models, Modelling system, CALGRID, RAMS, Seasonal concentrations, Tuscany

1 Introduction

Basically, there are two different mechanisms mainly affecting the ozone (O_3) formation in the Planetary Boundary Layer (PBL): the stratospheric ozone intrusion on the one hand, and the ozone production on the other hand, mostly as a result of the transformation of emissions due to combustion processes from within the PBL itself. The first mechanism is a natural one, where O_3 is generated in the stratosphere by photochemical reactions driven by the high-energy solar radiation. A small fraction of this stratospheric O_3 penetrates the PBL, following large convective thunderstorms, and is responsible for the ambient background O_3 concentrations [10]. However, such a stratospheric ozone intrusion accounts for not more than 10% of the whole PBL O_3 concentrations, as the major contribution is due to the second anthropogenic mechanism. As a matter of fact, a large number of chemical reactions is involved in the latter, in which O_3 formation is mainly due to the photolysis of NO_2 deriving from emissions of primary NO. Thus, any reaction converting NO to NO_2 undirectly contributes to the O_3 formation. This role is played by the carbon-containing species, and particularly by some very reactive species, the peroxy alkly radicals, which are generated from the oxidation of the Volatile Organic Compounds (VOCs). Summarizing, the troposheric ozone formation finally result from all chemical reactions involving the anthropogenically emitted O_3 precursor species, such as NO_x and VOCs. However, the complexity of the problem is increased in that there is no linear correlation between precursor emissions and O_3 concentrations, neither in space nor in time, i.e., decreasing one of precursors somewhere may cause an increase in O_3 concentrations elsewhere, and viceversa. What's more, the spatial scales involved in the transportation processes account for hundreds of kilometers, where air parcels may encounter new precursor concentrations and thus cause an O_3 increase in rural areas downwind from industrial or urban areas [2].

With this in mind, the LaMMA laboratory developed an integrated modelling system based on the CALMET [7] and CALGRID [8] models, starting from the RAMS [9] meteorological model forecasting. The implemented RAMS-CALMET-CALGRID system is intended to be as a modelling tool to assess ozone concentrations at a regional scale, and thus to manage such a photochemical pollution problem.

In the present work a comprehensive application of RAMS-CALMET-CALGRID has been made on a seasonal basis over Tuscany region, Italy. This is the sequel of a work previously carried out, dealing with a short term application focused on a four-day O_3 peak episode occurred in Tuscany in July 1997 [4]. This time the modelling system has been applied in a long term mode on a five-month (May to Sept. 2002) period, which can be certainly reckoned as the most original part of the work. As a matter of fact, a number of strong modifications in the original CALGRID code proved to be necessary to let the model run continuously all over the five months of simulation.

2 Model description

2.1 Modelling scheme

The working scheme of the integrated RAMS-CALMET-CALGRID modelling system developed in the present study is shown in Fig.1. This scheme is made of a meteorological section, including the RAMS and CALMET models, a chemical one, based on the PREPEMIT preprocessor, an emission block, downstream linked to the chemical section, and eventually the CALGRID-based dispersion modelling, designed to estimate O_3 concentrations. Besides, geophysical data, i.e., DEM and land use, point and areal emission sources, as well as initial and boundary conditions have been collected and processed to properly feed the modelling system.

CALGRID is an Eulerian transport and diffusion grid model including modules for horizontal and vertical advection/diffusion and dry deposition. It is specifically conceived to manage photochemical pollution, featuring a number of modules to fully reproduce all chemical and photochemical reactions involving the ozone precursor species.

The meteorological input to CALGRID is based on the RAMS archive [3] and is performed thanks to a suitably developed interface connecting RAMS to CALGRID via the CALMET meteorological postprocessor.

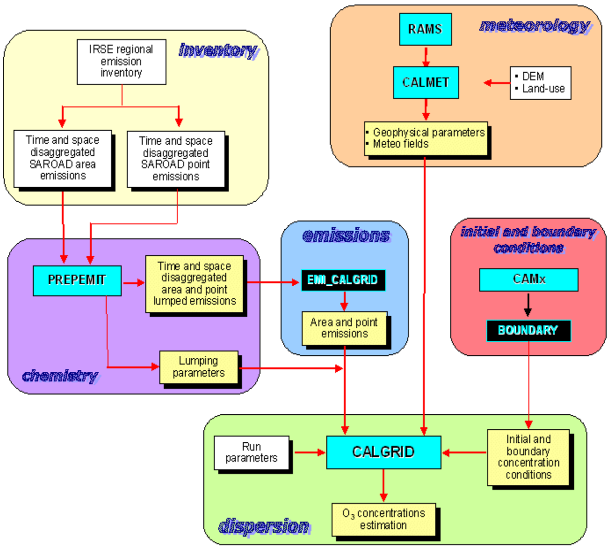


Fig.1: Architecture of the RAMS-CALMET-CALGRID modelling system developed.

An important section of this work included the application of the PREPEMIT chemical preprocessor, which is based on the SAPRC-90 "lumped" chemical mechanism developed by Carter [1]. Starting from time and space disaggregated emission data, PREPEMIT has been used to provide both emissions of all lumped species, and the species-to-group parameters based on different chemical kinematics affecting each involved species. Final emissions to CALGRID have been estimated through the EMI_CALGRID specifically developed utility downstream to PREPEMIT.

Initial and boundary conditions have been derived from national scale fields, provided as outputs by the CAMx dispersion model applied by CESIRICERCA in the framework of the research fund for the Italian electrical sector [5].

2.2 Input setup

Emission data are provided by the "IRSE" regional emission inventory of the Tuscany Region Authority [6]. They include both point and areal sources, as shown in Fig.2. A total number of 380 point sources were taken into account, including all the major industrial chimneys all over the region. Areal emissions are imported by means of a 4-Km spaced grid, which globally is made of 1750 cells.

All main emitting categories were considered, particularly those related to urban and extra-urban traffic, productive and biogenic processes. Areal emissions include ship routes as well.

Starting from emission data, all SAROAD species are arranged by PREPEMIT, whose ultimate result is their lumping into groups according to the different chemical reaction kinematics. In the same time, such species-to-group parameters are produced by PREPEMIT. Summarizing, a total number of 16 species has been used, including NO, NO_2, CO, SO_2 plus 12 VOCs, whose percentual speciation is drawn in Fig.3.

Large scale fields by the CAMx model at a 25-Km resolution all over Italy have been used to define the initial and boundary conditions to CALGRID. A total number of 40 species advected to Tuscany from boundaries has been taken into account. Since in the CAMx application a different lumped chemical mechanism was used, an additional effort was required to perform a chemical conversion when lumping VOCs.

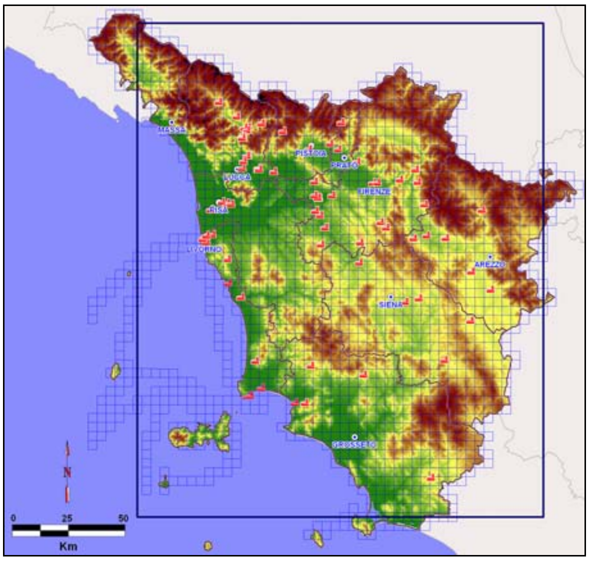


Fig.2: Map of spatial domain overlapped to point and areal emission sources.

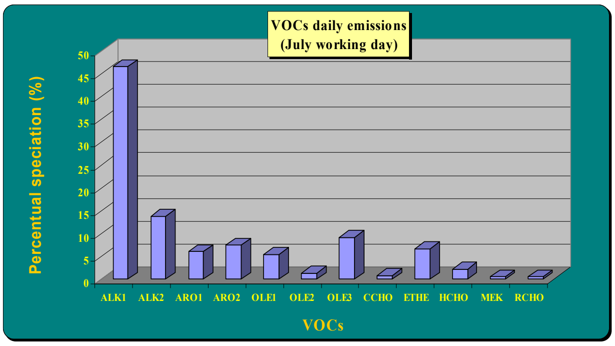


Fig.3: Emission data: VOCs speciation into 12 lumped groups.

3 CALGRID application and analysis of results

3.1 Case-study features

As shown in Fig.2, the study area is 184x224 Km^2 wide, made of a 4-Km spaced 45x55 computational grid with 12 terrain-following vertical levels ranging from 10 to 2860 m a.g.l.

To achieve the ozone seasonal pattern over Tuscany, the long term CALGRID application was carried out from May 1st to September 30rd, 2002, a five-month period with a 1-hour time step which accounted for 3672 hours in all.

Since CAMx initial and boundary conditions refer to the same season, but of a different year (1999), a suitable seasonal adjustment proved to be necessary. A number of runs was performed in order to set the best CALGRID configuration as far as boundary conditions are concerned. Ultimately, seasonally-adjusted CAMx outputs have been used for all species except for O_3, whose boundary conditions were defined as the averages of concentrations measured through the same May to September 2002 time period by all the ozone monitoring stations.

3.2 Spatial pattern of estimated ozone concentrations

A preliminary “overview” analysis of CALGRID results was made in terms of spatial distribution of O_3 concentrations per month over Tuscany. As a sample graphical output, in Fig.4 O_3 hourly averaged concentrations estimated by CALGRID in the month of July 2002 are plotted according to a 3–hourly time step representation.

A general result that can be drawn is the CALGRID capability to reproduce the diurnal cycle of ozone formation and removal, depending on solar activity increasing, in terms of both intensity and timing. A peak in solar activity results in O_3 concentrations, both in localized peak values and spatial extent, as shown in Fig.4.

Besides photochemical features, CALGRID proved to be strongly affected by meteorological conditions, particularly as concerns wind pattern. As a result, O_3 transportation and diffusion is affected by both the synoptic wind situation and local circulations, such as sea-land and slope-valley breezes.

From a numerical point of view, the highest O_3 concentrations are estimated on average in the month of July and secondly in June. In particular, this occurs at h.18. On the other hand, the lowest O_3 levels are estimated in September. It is remarkable that O_3 peaks estimated in May are on average higher than those in August.

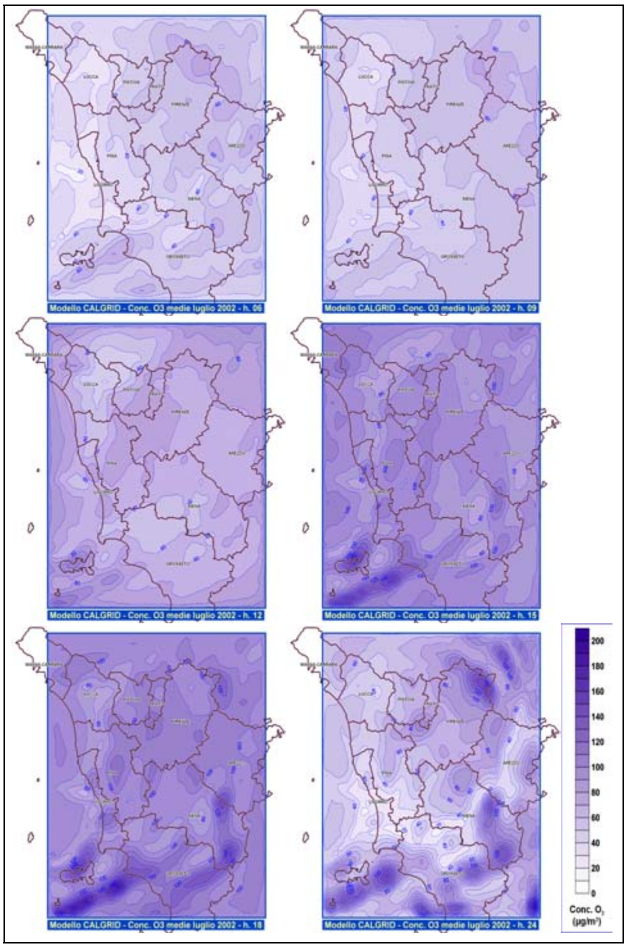


Fig.4: 3-hourly time sequence of O_3 hourly averaged concentrations estimated by CALGRID over Tuscany (July 2002).

3.3 Estimations vs. observations comparison

In order to evaluate CALGRID local performances a comparison of model estimations has been made versus the observations provided by some regional monitoring stations. The latter have been chosen to be as much representative as possible regarding the different features affecting the O_3 accumulation in the lower atmosphere. In particular, six stations were considered for comparison: two urban ones, within the cities of Firenze and Arezzo, two extra-urban (background) stations (Settignano and Maurogordato), in the outskirts close to the cities of Firenze and Livorno, respectively, and two stations (Milani and Serao), located within two industrial districts. Table 1 and Fig.5 summarize all the features of the ozone stations selected for comparison with CALGRID estimations. In addition, Table 1 includes indexes of model computation cells where stations fall and the relative station to grid point distance.

Table 1: Ozone stations selected for local comparison with CALGRID and station to grid point relative distance.

Stat. No.	Station name	City	Typology	CALGRID grid indexes		Station to CALGRID grid point distance (m)
				I	J	
1	Settignano	Firenze	background	28	38	340
2	Boboli	Firenze	urban	26	37	1780
3	Milani	Montelupo F. (FI)	industrial district	22	36	1730
4	Serao	S. Croce (PI)	industrial district	17	36	1700
5	Maurogordato	Livorno	background	8	29	1090
6	Acropoli	Arezzo	urban	39	29	1180

Chemical stations have been chosen taking into account different meteorological regimes as well: the Livorno coastal station is dominated by sea-land breeze circulations, despite the Boboli and Settignano ones, located within or near Firenze and thus representative of inland conditions, as well as the Arezzo station. On the other hand, the Montelupo and S. Croce stations are sited in the Arno valley south-west to Firenze, between inland and sealand, and thus can be affected by both circulation regimes.

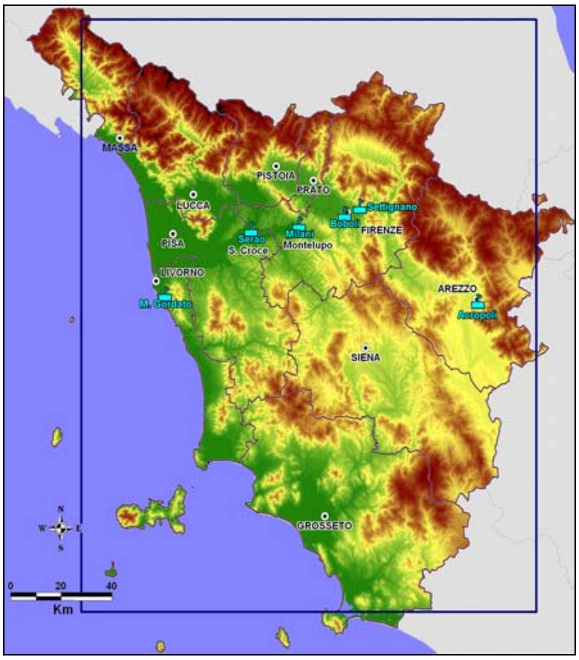
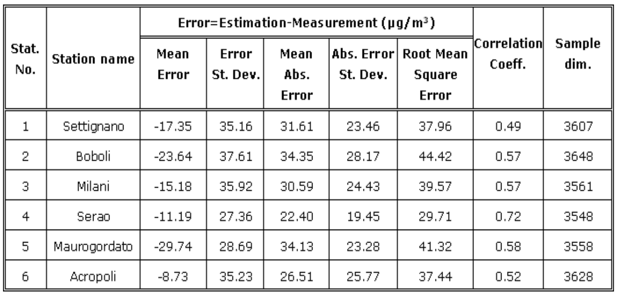


Fig.5: Geographic location of ozone stations selected for local comparison of model results.

Concerning such a comparison, a number of remarks appears to be necessary. First of all, O_3 concentrations are measured at 3 m a.g.l., whereas CALGRID estimations are values vertically averaged from 0 to 20 m a.g.l. Secondly, since CALGRID estimations are expressed in terms of spatial averages over each 4x4 Km^2 wide cell and thus referred to its central point, a substantial spatial distance generally exists between any cell centre and station location (see Table 1).

Table 2: Statistical values of hourly time series of 0 to 20 m O_3 concentrations estimated by CALGRID vs. 3-m observations by all selected stations (May to Sept. 2002).



Stat. No.	Station name	Error=Estimation-Measurement (μg/m³)					Correlation Coeff.	Sample dim.
		Mean Error	Error St. Dev.	Mean Abs. Error	Abs. Error St. Dev.	Root Mean Square Error		
1	Settignano	-17.35	35.16	31.61	23.46	37.96	0.49	3607
2	Boboli	-23.64	37.61	34.35	28.17	44.42	0.57	3648
3	Milani	-15.18	35.92	30.59	24.43	39.57	0.57	3561
4	Serao	-11.19	27.36	22.40	19.45	29.71	0.72	3548
5	Maurogordato	-29.74	28.69	34.13	23.28	41.32	0.58	3558
6	Acropoli	-8.73	35.23	26.51	25.77	37.44	0.52	3628

Although from a strictly spatial point of view it is not correct, a local comparison between model estimations and station measurements has been made as well, which resulted in the statistical values summarized in Table 2. Moreover, Figures 6, 7 and 8 give a sample idea of hourly pattern of estimations vs. observations comparison by the stations of Arezzo, S. Croce and Montelupo, respectively.

Correlation coefficients reported in Table 2 show that the CALGRID highest accuracy is performed by the Serao (S. Croce) station, resulting in a 0.72 value. Good accuracy levels (0.57÷0.58) occur by all other stations as well except for Acropoli (Arezzo) and Settignano (Firenze) ones, where the lower values of 0.52 and 0.49 are achieved, respectively. As a matter of fact, CALGRID tends to underestimate nighttime ozone concentrations, particularly when background stations are considered, i.e. Maurogordato and Settignano. This is due to an overestimation of nighttime NO concentrations over those areas, resulting in a marked ozone scavenging by NO which leads to extremely low nighttime ozone concentrations. Such a behaviour is probably related to whether an overestimation of NO_x emissions or an underestimation of the nighttime boundary layer, causing an overburden of NO near the surface.

On the other hand, another general result is the CALGRID tendency of underestimating the daily peak concentrations. This can be traced back to an underestimation of VOCs emissions or, most likely, and underestimation of their reactivity, which is particularly effective in the daytime.

Apart from the above considerations, CALGRID appears to provide some good performances when compared to observations, particularly with regard to the capability to reproduce the diurnal cycle of ozone formation and removal and the prediction of diurnal peak concentrations. On the other hand, CALGRID good results are remarkable particularly if one considers the low accuracy of some input data used in the application, i.e. the 1999 seasonally-adjusted boundary conditions data (opposite to the 2002 application year) and the not updated emission data.

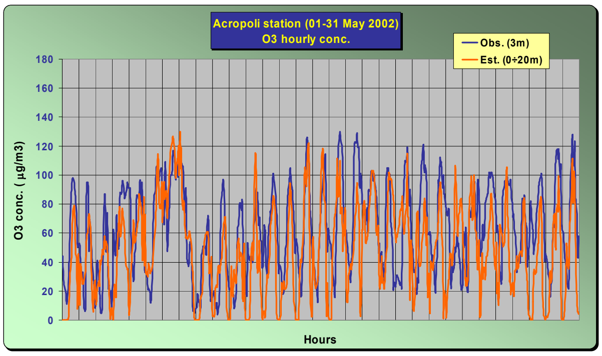


Fig.6: Hourly time series of 0 to 20 m O_3 concentrations estimated by CALGRID vs. 3-m observations by the Acropoli (Arezzo) station (May 2002).

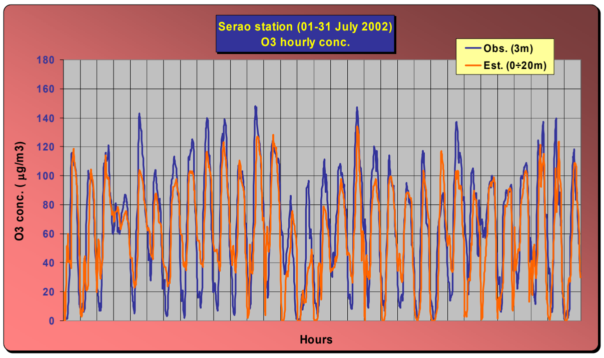


Fig.7: Hourly time series of 0 to 20 m O_3 concentrations estimated by CALGRID vs. 3-m observations by the Serao (S. Croce) station (July 2002).

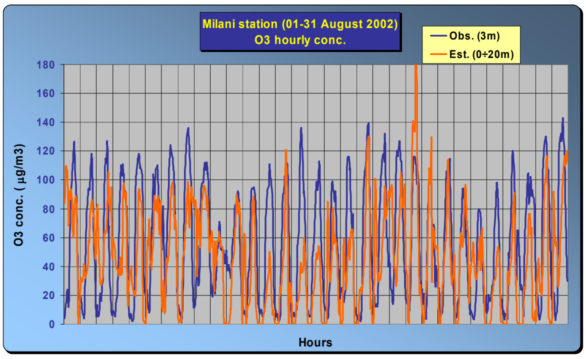


Fig.8: Hourly time series of 0 to 20 m O_3 concentrations estimated by CALGRID vs. 3-m observations by the Milani (Montelupo) station (August 2002).

4 Conclusions

In the present work the implementation of the RAMS-CALMET-CALGRID integrated modelling system has been made to assess ozone pollution at a regional scale. A comprehensive system application has been carried out on a seasonal basis over Tuscany region, Italy. In particular, the modelling system has been applied in a long term mode on a five-month (May to September 2002) period, which can be certainly reckoned as the most original part of the work. As a matter of fact, a number of strong modifications in the original CALGRID code proved to be necessary to let the model run continuously all over the five months of simulation.

Eventually ozone 1-hour concentrations estimated by CALGRID have been compared with observations provided by the major stations of ozone regional monitoring network, although from a strictly spatial point of view such a comparison is not correct. Anyway, CALGRID performances show a high accuracy degree when compared with observations, particularly if one considers the capability of reproducing the diurnal cycle of ozone formation and removal and the prediction of diurnal peak concentrations. On the other hand, CALGRID good results are remarkable, particularly if one considers the low accuracy of some input data used, i.e. the 1999 seasonally-adjusted boundary conditions data (opposite to the 2002 application year) and the not updated emission data. Therefore, a substantial accuracy increase in model performances can be expected whether in the future more accurate input data will be available.

References:

[1] Carter W., *Documentation for the SAPRAC Atmospheric Photochemical Mechanism Preparation and Emissions Processing Programs for Implementation in Airshed Molels,* Appendix C of Final Report, Statewide Air Pollution Research Center, Sacramento, CA, USA, 1988.

[2] Horowitz J.L., *Air quality for urban transportation planning*, MIT Press, Cambridge, MA, USA, 1982.

[3] Gualtieri G., Calastrini F., *A 3-D meteorological archive based on the RAMS model daily forecasting*, Procs. of 6th European Conference on Application of Meteorology, organized by the Meteorology General Office of the Italian Military Airforce, held in Rome, Italy, 15-19 Sept. 2003.

[4] Gualtieri G., Calastrini F., *An integrated meteo-diffusional modelling system to manage atmospheric pollution: an application of combined RAMS-CALMET-CALGRID models to evaluate ozone pollution in the Tuscany region*, Procs. of the 13rd Conference on the Applications of Air Pollution Meteorology With the Air and Waste Management Association, organized by the American Meteorological Society, held in Vancouver, Canada, 23-27 August 2004.

[5] Pirovano G., Pertot C., Costa M.P., *Valutazione dell'inquinamento atmosferico su scala nazionale. Caso base ed analisi di scenario* - Report CESI A5/028375, Milano, Italy, 2005.

[6] Regione Toscana, *Inventario Regionale delle Sorgenti di Emissione in aria ambiente* – Dept. “Politiche Territoriali e Ambientali”, Area “Qualità dell’aria, inquinamento acustico, industrie a rischio, inquinamento elettromagnetico”, Firenze, Italy, 2001.

[7] Scire J.S., Robe F.R., Fermau M.E., & Yamartino R.J., *A User’s Guide for the CALMET Meteorological Model (version 5.0)*, Earth Tech Inc., Concord, MA, USA, 1999.

[8] Scire J.S., Yamartino R.J., Carmichael G., & Chang Y., *A User’s Guide for the CALGRID Meteorological Model (version 1.6b),* Earth Tech Inc., Concord, MA, USA, 1996.

[9] Walko R. L., & Tremback C. J., *RAMS – Regional Atmospheric Modeling System - version 4.3/4.4,* User’s guide, Fort Collins, CO, USA, 2001.

[10] Zannetti P., *Air pollution modeling*. Computational Mechanism Publications, Southampton, UK, 1990.