A FLEXIBLE MODEL SYSTEM FOR AIR POLLUTION SIMULATION AND CONTROL IN NORTHERN ITALY

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INTRODUCTION

Northern Italy, characterised by a complex terrain, high and urban and industrial emissions and a close road net, is often affected by severe photochemical and particulate pollution episodes, mainly during summer season. Photochemical pollution is a mixture of pollutants, as ozone, NO_2, PAN and formaldehyde, produced by a complex chemical reaction system. The main compounds inducing these reactions are NO_x and VOC coming from road traffic and industrial combustion plants. An essential role in the chemical process activation is played by high solar radiation and stagnating meteorological conditions. These prerogatives make photochemical smog to be typically a summer phenomenon, mainly occurring in domains nearby large urban areas. The cause-effect relation between precursors and photochemical pollutant is quite complex and non-linear, due to accumulation processes and to the large number of variables affecting the photo-oxidant production, including their integration and the consequent feedback multiple reactions. As a consequence of such complexity, the development of effective strategies of emission reduction requires suitable tools that enable to take into account all the variables involved in gas and aerosol phenomenology. In this work an integrated modelling system that can include several chemical mechanisms representing the gas-phase photochemistry has been set up to support the analysis of emission control strategies. Preliminary simulations and comparison between the different mechanisms results are presented. The system will be integrated with an aerosol processor and a particle emission module for particulate matter analysis.

APPROACH

The modelling system includes three main processors and post-processing tools:

1. The photochemical model CALGRID, *Yamartino, R.J., et al.* (1992) which is an Eulerian three-dimensional model that implements an accurate advection-diffusion scheme in terrain-following co-ordinates with vertical variable spacing; a resistance-based dry deposition algorithm takes into account pollutant properties, local meteorology and terrain features; the CALGRID chemical module uses SAPRC-90 mechanism, *Carter W.P.L.* (1990), including 54 chemical species with 129 reactions and the QSSA (Quasi Steady State Approximations) solver for the integration of kinetic equations. The CALGRID model has been modified to allow for changes in chemical schemes, *Decanini E. and Volta M.* (2002), and this goal has been obtained implementing the Flexible Chemical Mechanism interface, *Kumar N. et al.* (1995). The chemical mechanisms that have been implemented are two: SAPRC-97, *Carter W.P.L. et al.* (1997), which considers 82 chemical species and 184 reactions; Carbon Bond IV, *Gery M. et al.* (1988), with 37 chemical species and 78 reactions. The original integration solver of the CALGRID, the QSSA, has been substituted with the IEH solver of *Sun P. et al.* (1994) as the FCM makes possible to distinguish among fast and slow reacting species: the IEH integration method uses the implicit scheme LSODE of *Hindmarsh A.C.* (1980) to solve for the fast reacting species and an explicit second order scheme to solve the slow reacting species.
2. The meteorological pre-processor CALMET; *Scire J.S. et al.* (1990), which provides 3D meteorological fields by means of a three-step procedure: the available local measurements (SYNOP reports and upper air sounding data; wind and temperature profiles) and the ECMWF fields are collected and analysed; the background wind field is reconstructed,

adjusting ECMWF output to local topography; finally CALMET model provides 3D wind fields merging background field with measurements, introducing mesoscale features (mountain-valley breezes and local effects) revealed by ground-level measurements and estimating temperature fields as well as turbulence parameters.

3. The emission processor POEM, POllutant Emission Model, *Catenacci G. et al.* (1998) that has been specifically designed to produce present and alternative emission field estimates by means of an integrated *top-down* and *bottom-up* approach. POEM can be applied in particular to the Italian CORINAIR data and considers diffuse and main point sources coming from different activity sectors. Thanks to its technology and fuel-oriented formulation, this emission processor can be used to provide scenarios consistent with new fuel trades and pollutant abatement technologies. In particular, some pollutant control technologies (catalytic converter, zero-emission vehicle) and alternative clean fuels (oxygenated fuels, compressed natural gas), together with their specific slitting profiles, can be taken into account for sustainable air quality improvement strategies. Model outputs are the results of all possible combinations of three steps: the spatial disaggregation, the time modulation and the VOC splitting. As far as the spatial disaggregation the model estimates municipality emissions starting from the province level, then it is possible to distribute emissions on a gridded domain (e.g. adding new sources configuration, deleting point sources, introducing new roads or pedestrian areas and changing road network); the spatial allocation makes use of *surrogate variables*, highly correlated with emissions and defined by means of national and local statistical sources, GIS and land-use information. As far as the time modulation the model can provide emission fields for any assigned time interval on a daily or hourly basis starting from the annual database. In accordance with EUROTRAC-2/GENEMIS Project, fuel use, temperature, degree-days, working time, production cycle, traffic counts and road statistics are the main indicators being used for the temporal modulation of emission activities. Finally the total VOC amount is split into SAROAD classification of individual compounds and then lumped into the emission classes needed by the photochemical mechanism implemented in the transport model. To allow for such flexibility in chemical schemes, the FCM interface has also been implemented in the POEM. The road transport, agriculture and biogenic emissions are estimated by means of the *bottom-up* approach implemented by POEM. The emission fields due to the other sectors are computed by means of the top down approach starting from the CORINAIR90 data set, updated by the last available Italian CORINAIR report (1994).

The system has been designed for air quality policy analysis in the Lombardia basin but also for possible applications to other areas in co-operation with local environmental agencies.

The area under study is located in Northern Italy and includes: the Lombardia Region, with several cities (Milano, Brescia, Bergamo, Varese, Como and others) as well as rural areas in Po Valley (Po is the main Italian river), most of central Alps (up to 4000m on the see level) as well as portions of Southern Swiss. The Southern part of the domain (i.e. the Po Valley) is densely inhabited and includes highly industrialized and intensive agricultural areas. This area is regularly affected by high ozone level, also due to the frequent stagnating conditions, especially during summer months. This fact claims for a careful design of emission abatement strategies, taking into account photochemical regimes, emission and meteorological conditions. The episode chosen to be simulated as base-case for Lombardia is 1-5 June 1998 during the PIPAPO projects measurements campaign performed in the frame of SATURN and LOOP subprojects of EUROTRAC-2. During the selected period, values exceeding the health protection threshold have been recorded in many stations of the regional network. Significant information reports about threshold exceedances were given to the population for many consecutive days. Moreover,

the strong photo-oxidant activity noticed in this period was also stressed by the stagnating meteorological conditions associated with high solar radiation.

Urban air quality impact simulations have been performed with the 2 chemical mechanisms implemented for the base case reference meteorological and emission field in order to point out the differences between the schemes. The simulation domain has been subdivided according to a grid system having: 60 per 58 horizontal cells, with 4km step size, 20 vertical layers of variable thickness (20, 45, 80, 120, 170, 240, 330, 440, 580, 770, 1000, 1300, 1600, 2100, 2700, 3400, 4100, 4900, 5900 and 7000 m). In order to provide boundary and initial conditions the grid cells have been subdivided into three categories (urban, rural and mountain) on the basis of height and land use cover; a ground level value for the concentration of each compound has been derived from available measurements in the surroundings or from literature values. When no data are available a low value of 1.0E-7 ppb has been assumed. The concentrations have been assumed to be constant in time for all compounds, except for ozone, NO and NO_2. Typical hourly profile for the concentrations of such species has also been estimated from available measurements for each cell category. Since the organic chemical species used by the two mechanisms are not the same, a correspondence table to convert the data used in both the two cases has been evaluated: the converting factors are represented in Table 1.

Table 1. Correspondence between SAPRC97 and CB4 organic species

	SAPRC97	**CB4**
Alkanes	ALK1	~3 PAR
	ALK2	~7 PAR
Aromatics	ARO1	TOL
	ARO2	XYL
Ethene	ETHE	ETH
Alkenes	OLE1	OLE+PAR
	OLE2	2ALD2
	OLE3	ISOP
Formaldehyde	HCHO	FORM
Aldehyde	CCHO	ALD2

The vertical profile for the boundary and initial conditions has been derived on the basis of measurements. The ozone vertical distribution has been estimated establishing the top value (60 ppb over 2600 m a.s.l.) and defining a sigmoid profile for each ground level value. For all other species a decreasing profile has been assumed reducing progressively ground-level concentrations. (down to a 0.1 factor at the top of domain of the ground concentrations).

RESULTS AND DISCUSSION

First applications of the modeling system have been realized. The episode chosen as base case for model system validation has been simulated with the 2 chemical schemes implemented in the transport model. The results obtained for O_3 and NO are represented in Figure 1 for an urban site while the same pollutant time series for a rural site are represented in Figure 2.

As far as the urban site is concerned, the simulation performed with the CB4 mechanism shows a good agreement between the ozone concentrations calculated and the experimental data, while the values estimated by SAPRC-97 mechanism are higher. It must be evidenced that the NO time concentration simulated by the SAPRC-97 are lower than the others obtained with the CB4 and this fact can explain the higher concentrations calculated for the ozone.

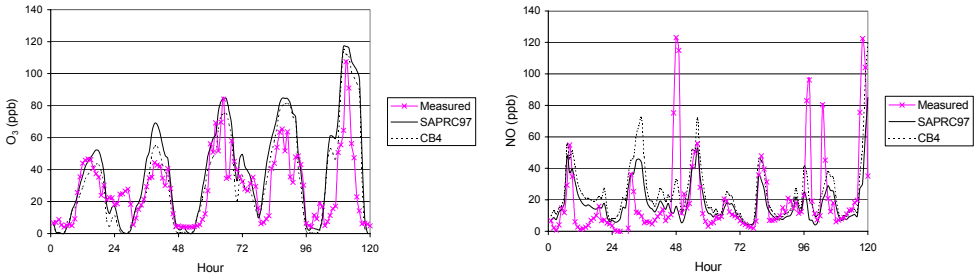

Figure 1. Comparison for O_3 and NO calculated concentrations with the experimental values for an urban site, Milano-Juvara.

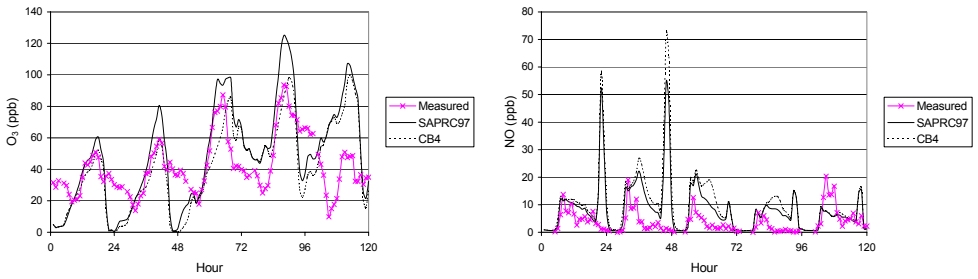

Figure 2. Comparison for O_3 and NO calculated concentrations with the experimental values for a rural site, Erba (CO).

Also in the case of the rural site, the ozone concentrations estimated by the CB4 mechanism show a good agreement with the measured values but only for the first 4 days. In the last day the values obtained for the ozone are too high and this fact is due to a local meteorological effect produced by a wind recirculation at the foot of the Alpes. A comparison among the ozone fields evaluated by the 2 mechanisms at the same hour in the domain is shown in Figure 3.

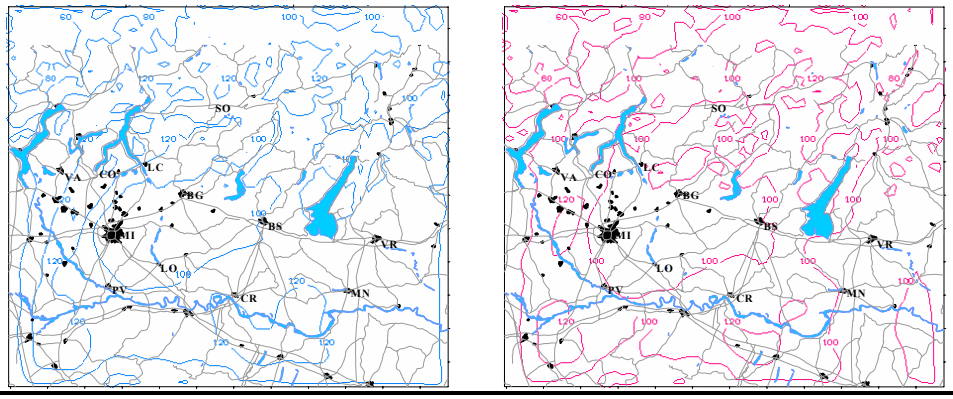

Figure 3. Ozone fields evaluated by the (a) SAPRC97 and (b) CB4 mechanism at 16 PM, June 4, 1998.

In each case the ozone concentration fields show the same distribution. It can be noticed that in the Milano zone the values obtained are lower than in the surrounding area due the higher emission of NO_x. Finally, in the northern and in the southern zone of the domain the ozone concentrations tend to accumulate: this fact is due to a breeze effect produced by the mountains.

CONCLUSIONS

To evaluate the possible strategies of emissions reductions, a flexible and integrated modeling system has been realized. This system is composed of the CALGRID model which has already been applied to the Northern Italy, the POEM emission processor and the CALMET meteorological processor. This system can include several chemical mechanisms. First simulations have been obtained and it has been observed that the various chemical mechanisms implemented show different level of agreement with the experimental data. Next objective is to implement an aerosol processor in the transport model and in the emission processor to evaluate the particulate matter role in the photochemical pollution.

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REFERENCES

Carter, W.P.L., 1990: A detailed mechanism for the gas-phase atmospheric reactions of organic compounds. *Atmos. Environ.*, **24A**, 481-518.

Carter, W.P.L., D. Luo and I.L. Malkina 1997: Environmental chamber studies for development of an updated photochemical mechanism for VOC reactivity assessment. Final report, California Air Resources Board, Sacramento (CA).

Catenacci, G., M. Riva, M. Volta and G. Finzi, 1998: POEM: an emission scenario processor. 2nd SATURN Workshop, Hamburg.

Decanini E. and M. Volta, 2002: Flexible modelling System for Air Pollution Simulation and Control in Northern Italy. ", proc. EUROTRAC SYMP'02.

Gery, M.W., G.Z. Whitten and J.P. Killus, 1988: Development and testing of the CBM-IV for urban and regional modeling. EPA/600/3-88/012, U.S. Environmental Protection Agency, research Triangle Park, NC.

Hindmarsh A.C., 1980: LSODE and LSODEI, two new initial value ordinary differential equation solvers. ACM-SIGNUM Newsletter **15**, N° 4, 10-11.

Kumar, N., F.W. Lurmann and W.P.L. Carter, 1995: Development of the flexible chemical mechanism version of the urban airshed model. Final report STI-94470-1508-FR, California Air Resources Board, Sacramento (CA).

Pandis, S.N., R.A. Harley R.C. Glen and J.H. Seinfeld, 1992: Secondary organic aerosol formation and transport. *Atmos. Environ.* **26A**, N° 13, 2269-2282.

Scire, J.S., E.M. Insley and R.J. Yamartino, 1990: Model formulation and user's guide for the CALMET meteorological model. Report A025-1, California Air Resources Board, Sacramento (CA).

Sun P., D.P. Chock and S.L. Winkler, 1994: An implicit-explicit hybrid solver for a system of stiff kinetic equations. Paper presented at the Air & Waste Management Association 87th Annual Meeting, Cinncinnati, OH, June 19-24.

Yamartino, R.J., J.S. Scire, G.R. Carmichael and Y.S. Chang, 1992: The CALGRID mesoscale photochemical model–I. Model formulation. *Atmos. Environ.* **26A** N° 3 1493-1512.