APPLICATION OF REMSAD AND GAMES MODELLING SYSTEMS ON A PARTICULATE MATTER AND OZONE EPISODE IN MILAN METROPOLITAN AREA

E. Decanini[1], M. Pola[2], F. Polla Mattiot[3] and M. Volta[4]
[1]Scuola Normale Superiore, Pisa, ITALY
[2]D.I.T.S., Università La Sapienza, Roma, ITALY
[3]EniTecnologie S.p.A., Monterotondo Scalo, ITALY
[4]D.E.A., Università degli Studi di Brescia, Brescia, ITALY

INTRODUCTION

Milan metropolitan area is regularly affected by intense photochemical pollution episodes during summer. These events are often coupled to particulate matter (PM) high concentration levels. Attention on PM concentration measurements, related health effects and simulation results has recently raised, and new models for atmospheric dispersion are now available. To study the PM role in atmospheric pollution over Lombardia region, two modelling systems have been applied.

THE MODELLING SYSTEMS

Increased attention has been devoted, in last years, to atmospheric particulate matter (PM) due to adverse health effects that have been observed during prolonged exposure. Particulate matter can be originated by direct emission in the atmosphere (primary) or by reaction of gas phase species, producing low volatility compounds that condense on pre-existing particles or generate new ones by nucleation (secondary). Moreover particulate can also affect ozone concentration and production in the atmosphere both by scattering solar radiation that causes photochemical process, and by modifying ozone precursors that can be absorbed in the condensed phase.

Suitable tools are required to study air pollution. In this work two comprehensive modelling system have been designed and applied to analyse photochemical and aerosols episode.

GAMES MODELLING SYSTEM

The system consists of some main modules:

- The transport, diffusion and gas phase chemical model CALGRID *Yamartino R.J. et al.* (1992) an accurate advection-diffusion scheme in terrain following coordinates with variable vertical spacing and a resistance-based dry deposition algorithm taking into account of pollutant properties, local meteorology and terrain features. The model has been integrated by Flexible Chemical Mechanism *Kumar, N. et al.* (1995): the chemical module has also been updated with COCOH-97 mechanism, *Durlak, S.* (2000) containing 96 chemical species (single and lumped), 8 explicit condensable organic compounds, and 198 reaction. The CALGRID model has also been updated with a module for aerosol transport and a module for aerosol deposition (MAPS).
- The meteorological model CALMET, *Scire, J.S. et al.* (1990), which consists of a diagnostic wind field module and micrometeorological modules for over water and overland boundary layers.
- The aerosol module MAPS, *Durlak, S.* (2000), which allows for size resolved representation of particulate matter based on discrete sections. The module includes gas to particle processes such as condensation and nucleation that change the total size and number concentration of the aerosol population. The model takes into account of various aerosol phase compounds, such as sulphate, nitrate, ammonium, water, chloride, sodium, elemental carbon, primary organic compounds and 8 class of secondary organic compounds. The

module solves explicitly aqueous oxidation of SO_2 by uptake of gases (O_3, H_2O_2 and SO_2) into the aqueous aerosol and aqueous oxidation of the SO_2 to sulphate.

- The emission evaluation model POEMPM provides present and alternative emission field of gas phase compounds, estimated by means of an integrated *top-down* and *bottom-up* approach. The model considers diffuse and main point sources coming from different activity sectors. The POEMPM estimates size resolved and chemically split particulate matter emissions fields from road traffic *Finzi, G. et al.* (2002).

REMSAD MODELLING SYSTEM

The modelling system is constituted by the Regulatory Modelling System for Aerosols and Deposition, (REMSAD 5.0, System Application International, 1998), the 3D meteorological pre-processor (SAIMM), and the flexible emission inventory module, *Finzi, G. et al.* (2002). The REMSAD has been developed to support further understanding of the distributions, sources, and removal processes relevant to the components of atmospheric particulate matter. The REMSAD system is built on the basis of the UAM-V regional air quality model but it is mainly devoted to long term simulations on continental scale. The photochemical mechanism module is a reduced-form version of the Carbon Bond Mechanism (version 4). The intent of the micro-CB mechanism is not to estimates ozone levels, with the precision usually sought in air quality models designed to address the ozone issue *per se*, but rather to provide a physically faithful representation of the linkages between emissions of ozone and PM precursors; the oxidising capacity of the troposphere, represented primarily by the concentrations of radical and hydrogen peroxide; and the rate of oxidation of the nitrogen ozone and sulphur dioxide PM precursors. The process of primary importance for PM applications in REMSAD is sulphate formation. The particle size distribution and the particle mass fractions into eight classes are used in calculating deposition rates for particulate species (both dry and wet deposition). Particulate species are thus grouped: particulate nitrate, primary elemental carbon, primary elemental carbon, aqueous pathway sulphate particles, gaseous pathway sulphate particles, ammonium sulphate, ammonium nitrate, secondary organic aerosols, other as primary fine and coarse particulate matter.

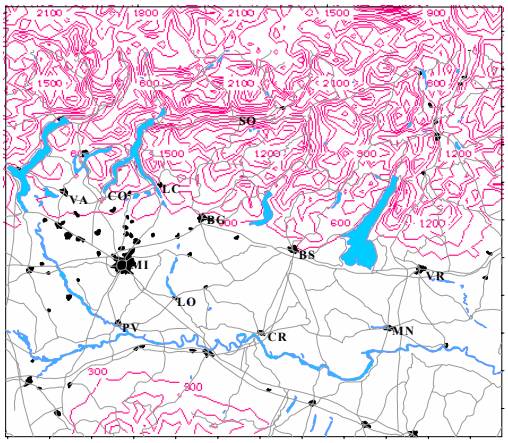

Figure 1. The application domain.

THE SIMULATION DOMAIN AND EPISODE

The systems have been implemented and tested in a domain (Figure1) in Northern Italy including the whole Lombardia Region and some portions of the neighbouring provinces (240x232 km^2). The area has been subdivided according to a grid system having 60 per 58

horizontal cells, with 4 km step size and 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). The episode chosen to be simulated as base-case for Lombardia is 1-6 June 1998 during the PIPAPO projects measurements campaign performed in the frame of SATURN and LOOP subprojects of EUROTRAC-2. Several data concerning gas and aerosol phase (chemical composition and size distribution) have been collected.

RESULTS AND DISCUSSION

To evaluate the performances of the two modelling systems a comparison of the calculated PM and ozone concentrations with the measured values has been done. The results are shown for two monitoring stations for PM10 (Figure 2 and Figure 3) and ozone (Figure 4 and Figure 5). As far as the PM10 is concerned both modelling systems show a time modulation of the estimated concentrations which agrees with the experimental data behaviour; nevertheless the GAMES model estimates the highest concentrations during the night while REMSAD computes the highest values during the afternoon. Estimated PM levels agree with measurements for the first two days of the episode but in the latter days only GAMES follows the increased trend registered by the measuring stations; in the obtained simulations REMSAD seems to underestimate such critical levels. In the case of the ozone the two models reproduce the daily pattern shown by the experimental measurements although GAMES is in agreement also in the peak concentration prediction while ozone levels estimated by REMSAD are lower.

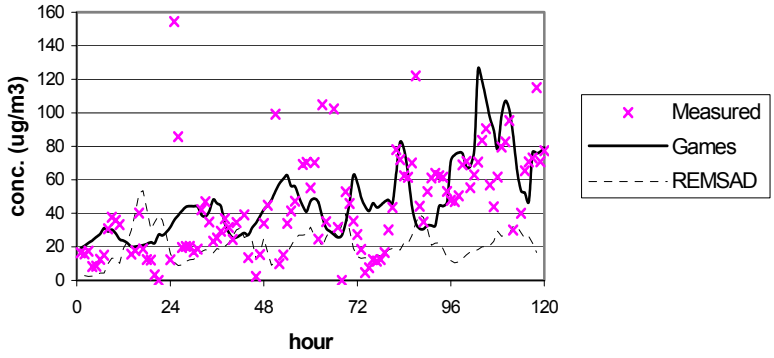

Figure 2. Comparison of PM10 calculated values and experimental data in Milano-Marche.

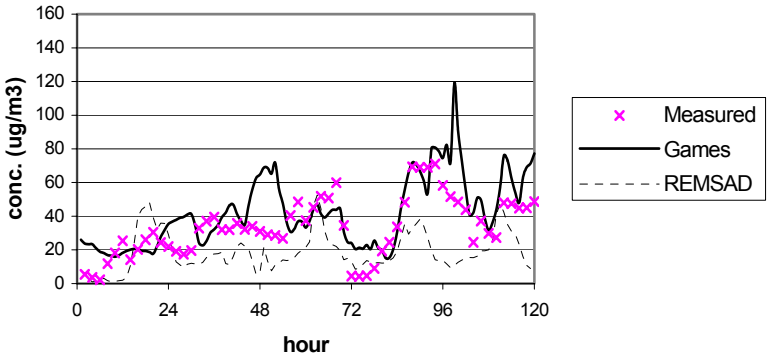

Figure 3. Comparison of PM10 calculated values and experimental data in Erba (CO).

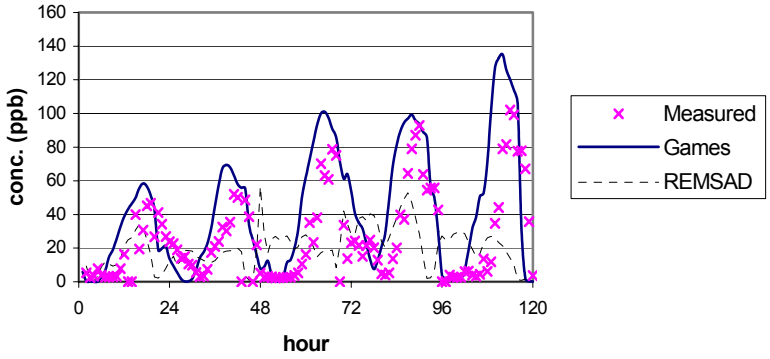

Figure 4. Comparison of ozone calculated values and experimental data in Milano-Marche.

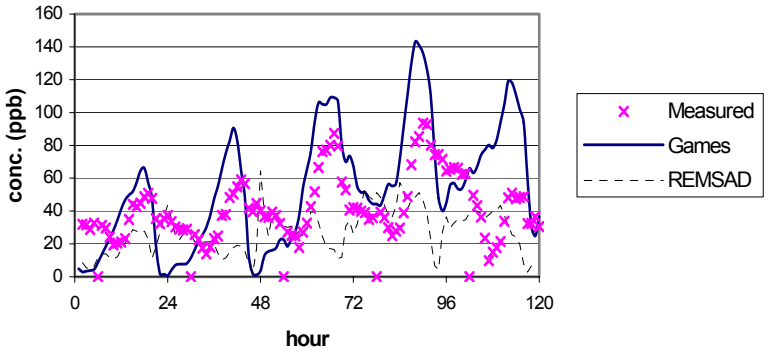

Figure 5. Comparison of ozone calculated values and experimental data in Erba (CO).

The PM10 calculated maps for the same hour of a critical day of the episode are shown in Figure 6. The PM levels are spread all over the central and the southern regions of the domain. The highest concentrations are estimated in areas surrounding the main urban emission sources but this is more evident in the GAMES results than in the REMSAD ones.

The PM spatial distributions suggest that secondary aerosol components play an important role in determining the total amount. The chemical composition of the aerosol estimated by the two model is quite different. REMSAD model tends to convert SO_2 to H_2SO_4 and to generate higher sulphate concentrations that are close to the measured data, available for a suburban station. On the other hand GAMES estimates higher particulate nitrate percentages which are in agreement with experimental data in particular during the last two days. It can be inferred that the equilibrium among water and particulate sulphate and nitrate is different for the two chemical modules.

CONCLUSIONS

A comparison between two different modelling systems for ozone and particulate matter simulation has been performed. Preliminary results show that REMSAD model does not seem to be able to correctly calculate pollutant levels during critical episodes because the gaseous chemical module is too simplified for such applications. On the other hand GAMES, implementing a more detailed scheme for the gas chemistry, shows a better agreement with the experimental PM10 data.

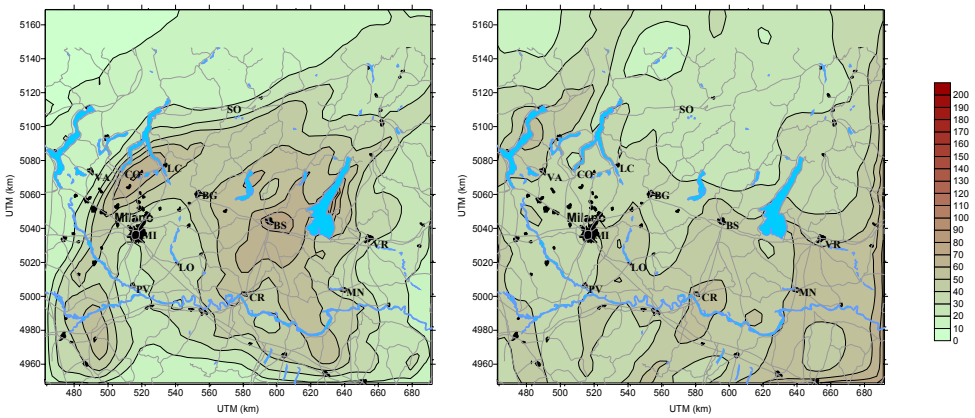

Figure 6. PM10 maps at 16 pm of June 4 1998 calculated by GAMES (a) and REMSAD (b) (units $\mu g/m^3$).

ACKNOWLEDGEMENTS

The authors acknowledge Dr. Fulvio Giavazzi of AgipPetroli-CREA and Prof. Giovanna Finzi of University of Brescia for the support provided to realise this work.

REFERENCES

Durlak, Sue, 2000: http://acd.ucar.edu/~durlak/MAPShome.html

Finzi, G., C. Gariazzo, F. Polla Mattiot and M. Volta, 2002: B.C. nesting approach harmonisation: UAM-V/CALGRID model exercise on Northern Italy. Proc. EUROTRAC'02 SYMP.

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.

S.A.I., 1998: User's guide to the Regulatory Modeling System for Aerosols and Deposition (REMSAD), SYSAPP98-96\42r2, Systems Applications International, Inc., San Rafael, California.

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).

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.