A MODELLING SYSTEM TO ASSESS THE AEROSOL SENSITIVITY TO GAS EMISSIONS

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Abstract: In this paper the aerosol sensitivity to NO_x, VOC, SO_2 and NH_3 gas emissions has been investigated over a complex domain in Northern Italy, including Milan metropolitan area. The analysis has been performed comparing eight simulations performed by the Gas Aerosol Modelling Evaluation System (GAMES): the smog episode occurred in 1-5 June 1998, selected as reference case, and seven simulations obtained feeding the modelling system with increasing or decreasing precursor emissions. The study evidences the role played by the photochemical modelling systems in recognizing the photochemical regimes and in providing a support for local Air Quality Authorities in selecting effective emission reduction strategies.

Keywords: Air pollution, Modelling system, Nonlinear systems, Sensitivity analysis, Mathematical models.

1. INTRODUCTION

The evaluation of emission control strategies, aiming at reducing air pollution, requires the analysis of benefits on the air quality of possible and sustainable alternative emissive scenarios, mainly for urban environments. In particular the evaluation should take into account both the primary pollutants, directly emitted by the sources, and the secondary species generated in the troposphere by physical processes and by chemical reactions catalyzed by solar radiation. If the relation between primary compound concentrations and emissions are nearly linear, the secondary pollutant concentrations and precursor emissions are ruled by strongly non linear laws. So that the secondary pollutants (both in gas and aerosol phase) can be affected in different ways by emission changes and the resulting effects can be counterproductive depending on the dominating chemical regime (Meng *et al.*, 1997), (Sillman, 1999). For this reason pollution control strategies should be carefully studied and designed by modelling simulations in order to estimate the global impact on air quality. Several air quality models have been applied at the urban and regional scales (Russel and Robin, 2000). They are generally classified in long term and episodic models. The former ones are devoted to analyze prolonged pollution episodes and are characterized by simplifications in the model structure and in the treatment of the pollution phenomenology to reduce the computational requirements. On the other hand the episodic models, being devoted to shorter time periods, allow a more detailed description of phenomena. Both model classes require big effort to provide input data (meteorological and emission fields, boundary and initial conditions), often computed and estimated in turn by models. To harmonize the pre-processing modules with the transport and chemical model means to design Modelling Systems, supporting (1) the analysis of gas and aerosol phase phenomena, (2) the selection of reliable and effective emission abatement strategies. In this paper the GAMES (Gas Aerosol Modelling Evaluation System), transport, photochemical and aerosol modelling system (Volta and Finzi, in press), has been performed to assess the aerosol sensitivity to NO_x,

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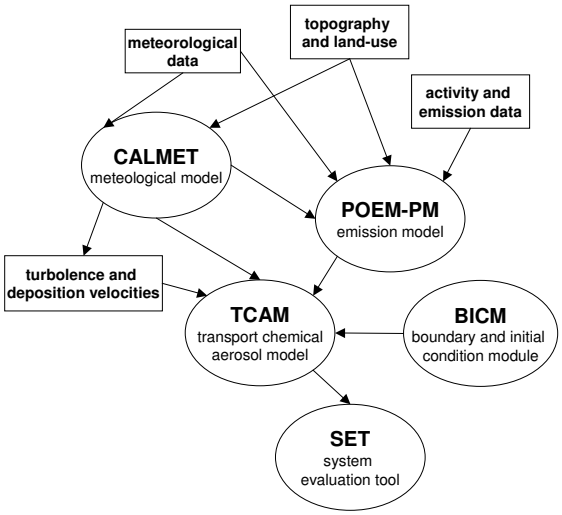

Fig. 1. The GAMES modelling system

VOC, SO_2 and NH_3 gas emissions over a complex domain in Northern Italy.

2. THE MODELLING SYSTEM

GAMES has been designed following the classic scheme of the photochemical multi phase modelling systems. It includes and harmonizes meteorological, emission, transport, chemical models and post-processing tools (Figure 1).

2.1 The meteorological pre-processor

The 3D meteorological fields are provided by means of a three-step procedure. (1) The available local measurements (SYNOP reports and upper air sounding data, wind and temperature profiles) and the ECMWF fields are collected and analysed. (2) The background wind field is reconstructed, adjusting ECMWF model output to local topography. (3) The meteorological CALMET model (Scire *et al.*, 1990) provides 3D wind fields merging background field with measurements and introducing local features revealed by ground-level measurements. Moreover CALMET estimates temperature fields as well as turbulence parameters.

2.2 The emission model

The emission processor POEM-PM (Carnevale *et al.*, in press) has been specifically designed to produce present and alternative emission field estimates by means of an integrated top-down (disaggregating a large scale yearly inventory) and bottom-up (inventorying the polluting activities and applying emission factors) approach . POEM-PM can be applied to the CORINAIR data base 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. Model outputs are the results of four algorithms: the spatial disaggregation, the time modulation, the VOC and the PM splitting. The spatial allocation procedure makes use of surrogate variables, GIS and land-use information. The model can provide hourly emission fields. In accordance with EUROTRAC-2/GENEMIS Project (Friedrich, 1997), 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. The total NMVOC amount is split in SAROAD classes of individual compounds and then lumped in agreement to the chemical mechanism implemented in the transport model. The POEM-PM also estimates size resolved and chemically split particulate matter emission fields. The considered chemical species are the organic carbon (OC), the elementary carbon (EC), SO_4=, NO_3-, H_2O and others (including the heavy metals and the undefined compounds), while the size bins range from 0 to 11.4 μm (Carnevale and Volta, 2003).

2.3 Boundary and Initial Condition Module

Initial and boundary conditions are assigned by BICM for all the species solved by the photochemical model, by means of a measurement extrapolation procedure. The module classifies three different cell types (urban, rural and mountain) on the basis of the domain land use. For each cell type, the typical hourly concentration profiles at ground level are estimated from experimental measurements. The estimated vertical profile is an exponential decreasing function that gradually reduces the concentrations down to a 0.1 factor of the ground values. The ozone vertical profile is estimated establishing the top value and defining a sigmoid profile obtained on the basis of experimental measurements.

2.4 The photochemical model TCAM

The Transport Chemical Aerosol Model is an eulerian photochemical three-dimensional multi phase model (Decanini, 2003), (Decanini and Volta, 2003*b*). It implements the advection-diffusion scheme derived by CALGRID model code (Yamartino *et al.*, 1992). The dry deposition is treated using a resistance-based algorithm which takes into account pollutant (gas and particulate matter) properties, local meteorology and terrain features (Yamartino *et al.*, 1992). The wet deposition module for gas and particles describes dissolution in droplets and precipitation scavenging (Seinfeld and Pandis, 1998). The model implements different chemical mechanisms based both on lumped molecule (SAPRC90, SAPRC97, COCOH97) and on lumped structure (CB-IV) approaches (Kumar *et al.*, 1995). The TCAM model also includes and harmonizes an

aerosol module describing aerosols by means of a fixed-moving approach. The particle is represented as an internal core containing the non-volatile material (elemental carbon, crustal and dust). The dimension of the core is held constant throughout the simulation. The volatile material is supposed to reside in an outer shell of the particle whose dimension is evaluated by the module at each time step on the basis of the total mass and of the total number of suspended particles. Shell and core fractions are supposed to be mixed.

The aerosol module is coupled to COCOH97 chemical mechanism and it describes the dynamics of 21 chemical compounds. The inorganic species are twelve (H_2O, SO_4=, NH_4+, Cl-, NO_3-, Na+, H+, SO_2(aq), H_2O_2(aq), O_3(aq), elemental carbon and other). The organic species are 9, namely a generic primary and 8 classes of secondary organic species, each of them corresponding to one of the Condensable Organic Compounds included in the gas phase chemical mechanism. Such chemical compounds are split in 10 size bins, so that the prognostic variables solved by the module are 210. TCAM describes the most relevant aerosol processes: the condensation, the evaporation (Seinfeld and Pandis, 1998), the nucleation (Jaecker-Voirol and Mirabel, 1989) of H_2SO_4 and the aqueous oxidation of SO_2 (Seinfeld and Pandis, 1998). The TCAM model solves the mass balance equations by means of a splitting operator (Marchuk, 1975). The gas phase chemistry is solved implementing the IEH algorithm (Sun *et al.*, 1994) which treats separately the slowly reacting species and the fast reacting species.

2.5 System Evaluation Tool

GAMES includes a module performing the assessment of the system simulations by statistical and graphical methods. The indexes and the graphics can be obtained for each measurement station or for a limited number of *representative* stations of the domain. The procedure to select the *representative* stations is based on the Cluster Analysis, detecting the similarity of the patterns recorded in different stations in terms of concentration levels and daily shape. SET, processing measurements and simulated concentrations, provides the computation of statistical indexes suggested by EPA (EPA, 1991) and by the EU "Directive 2002/3/EC".

3. BASE CASE SIMULATION

The selected domain, located in Northern Italy, is characterized by complex terrain and high emissions due to urban and industrial areas, a dense road network, agriculture activities. It includes the Milan metropolitan area, the most industrialized and inhabited area of Northern Italy. This domain has been deeply investigated by means of experimental campaigns (Vecchi and Valli, 1999), (Neftel *et al.*, 2002) and modelling assessments (Silibello *et al.*, 1998), (Martilli *et al.*, 2002), (Gabusi and Volta, in press).

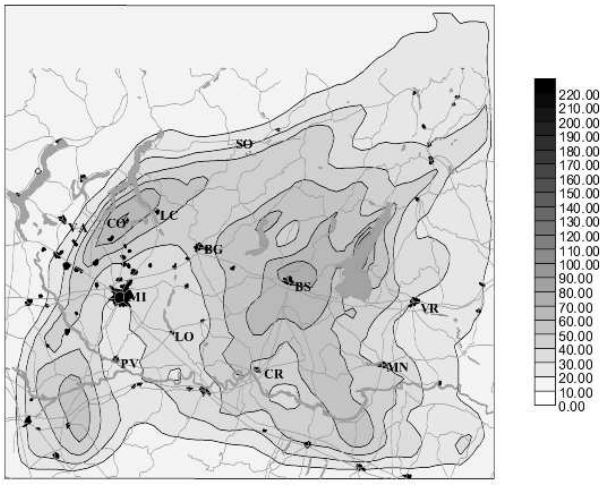

Fig. 2. PM_{10} concentration field (in $\mu g/m^3$) evaluated for June 4 at 16:00.

The GAMES modelling system has been performed over a 240x232 km^2 domain, subdivided according to a grid system having 60 per 58 horizontal cells, with 4 km step size. It has simulated the gas and aerosol production and transport during the June 1-5 1998 episode, monitored by the PIPAPO campaign (Neftel *et al.*, 2002) in the frame of the European EUROTRAC2 project. During the selected period, the strong photo-oxidant activity has been stressed by the stagnating meteorological conditions associated with high solar radiation. The validation analysis of the base case simulation is discussed in (Decanini and Volta, 2003*a*), (Decanini and Volta, 2003*b*), (Volta and Finzi, in press). The modelling simulations evidence that considerable PM levels are spread all over the central and the southern regions of the domain (see Figure 2 showing simulated PM10 at 4 p.m. of June 4th). The highest concentrations are estimated in areas surrounding the main urban ones.

Figure 3 suggests that photochemical pollutants play a relevant role in particulate production and accumulation processes during the analyzed episode. The daily mean concentrations estimated for Bresso (sub-urban monitoring station, North of Milan) show that nitrates increase day by day due to the gas phase production of nitric acid condensing on the particles and taking part to the neutralization equilibria with ammonia.

4. SECONDARY AEROSOL SENSITIVITY TO GAS EMISSIONS

Since in the episode a strong dependence of PM levels on the gaseous species has been found, sensitivity tests of the model have been performed to analyze the impact of control strategies based on primary gas emission reduction. Seven scenarios have been supposed: three scenarios with 50% reduction of the

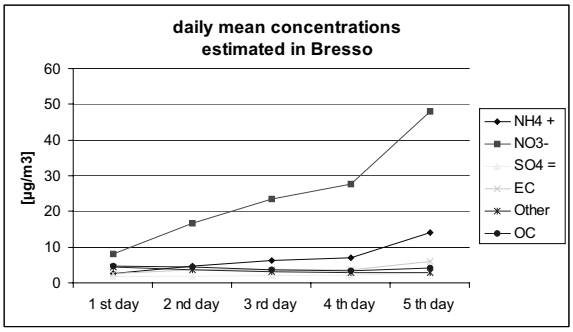

Fig. 3. PM_{10} daily mean concentrations (in $\mu g/m^3$), split in chemical compounds, estimated for Bresso.

emissions of (1) NH_3, (2) NO_x and (3) SO_2, two scenarios with the emissions of NH_3, NO_x and SO_2 respectively (4) decreased and (5) increased of 50%, two scenarios with the VOC emissions respectively (6) decreased and (7) increased of 50%,. In Figures from 4 to 10 the difference fields for PM_{10} between each scenario and the base case are shown, while some numerical evaluations performed for all cases are summarized in Table 1.

Table 1. Maximum and mean calculated PM_{10} concentrations ($\mu g/m^3$) in the domain for June 4, 16:00 and for the whole episode.

	June 4		**episode**	
	Max	*Mean*	*Max*	*Mean*
Base Case	93.05	29.92	199.54	35.30
- NH_3	61.69	22.91	185.26	28.50
- NO_x	89.00	28.84	187.97	33.13
- SO_2	92.58	29.77	197.45	35.25
- VOC	92.62	29.88	198.10	35.14
+ VOC	92.46	29.92	205.27	35.37
- NH_3, NO_x, SO_2	58.72	22.14	168.52	27.20
+ NH_3, NO_x, SO_2	124.14	37.11	239.32	41.22

Considering the first three scenarios (Figure 4 for NH_3, Figure 5 for NO_x and Figure 6 for SO_2), the most efficient in reducing PM_{10} concentrations is the one based on ammonia control. In particular in Brescia (in Figures: BS) metropolitan area, where a peak in the PM_{10} field has been calculated for the base case (Figure 2), the model estimates a reduction of nearly 30% of the concentrations. The advantages of developing strategies based on ammonia control have been underlined also in other literature studies (Nguyen and Dabdub, 2002), (Lurmann *et al.*, 1997).

The most significant effect of NO_x emission control has been calculated for the Brescia area: in this case a decrease of nearly 15% has been estimated. Comparing Figures 4 and 5 it is also interesting to observe that in the northern surroundings of Milan (in Figures: MI) the strategy based on ammonia control is more effective than the NO_x one because the secondary PM production is limited by the availability of the ammonium ion.

As far as SO_2 is concerned, the model has estimated a low sensitivity ($\pm$ 4%) of environment concentrations of particulate matter to the emissions of this species (Figure 6). This result can be explained assuming a relatively low importance of primary emissions of sulphur dioxide in the domain. Also Table 1 allows to conclude that the most effective strategy for the particulate matter reduction in the domain is based on ammonia control followed by nitrogen oxide and sulphur dioxide control.

If NH_3, NO_x and SO_2 strategies are combined, PM_{10} reductions are more evident while an increase of the emissions leads to an increase of PM_{10} as expected (Figures 7 and 8).

Analyzing the sensitivity to VOC it has been found that a modification in the organic species emissions leads to low global effects ($\pm$ 5%). Nevertheless, observing Figures 9 and 10 where the total VOC emitted amount has been respectively reduced and increased, it is interesting to note that the most significative impact has been found, for both cases, in the northern surroundings of Milan, that is the strongest VOC emission source of the domain. The reduction of VOC emissions decreases the secondary organic fraction of the particulate matter. Moreover this area is characterized by a VOC-limited regime as far as the ozone is concerned ((Gabusi and Volta, in press)) suggesting that the responses of both pollutants, O_3 and PM_{10} are related. In fact since in this area a reduction of VOC causes a reduction of ozone, it can be supposed that this strategy also limits the availability of the other photochemical oxidants (mainly $OH\cdot$). The resulting concentrations of the oxidized nitrogen compounds are then reduced and therefore the rate of nitrate PM_{10} formation decreases.

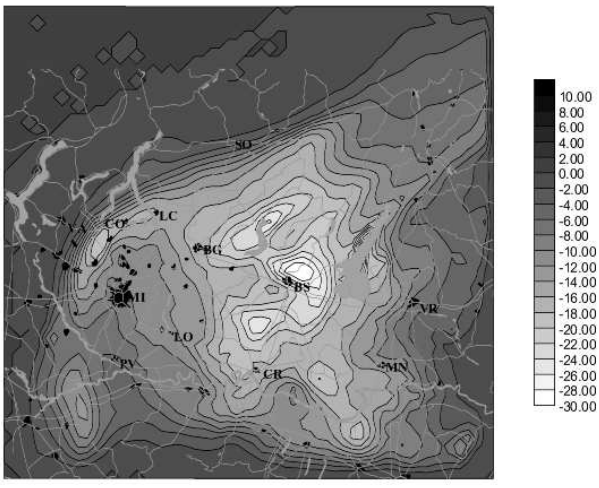

Fig. 4. Difference PM_{10} field (%) evaluated for June 4 1998, 16:00 for the 50% NH_3 reduction scenario.

5. CONCLUSIONS

The GAMES modelling system has been performed to simulate the smog episode monitored in Lombardia

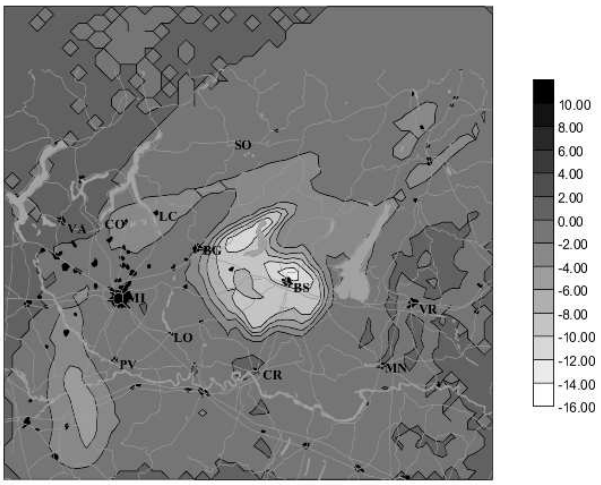

Fig. 5. Difference PM_{10} field (%) evaluated for June 4 1998, 16:00 for the 50% NO_x reduction scenario.

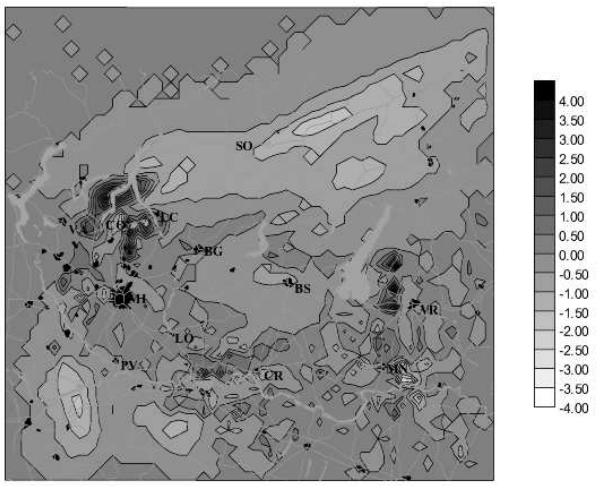

Fig. 6. Difference PM_{10} field (%) evaluated for June 4 1998, 16:00 for the 50% SO_2 reduction scenario.

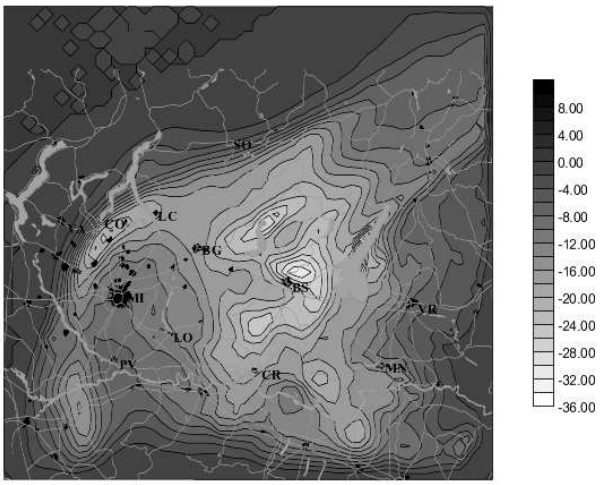

Fig. 7. Difference PM_{10} field (%) evaluated for June 4 1998, 16:00 for the 50% NH_3, NO_x, SO_2 reduction scenario.

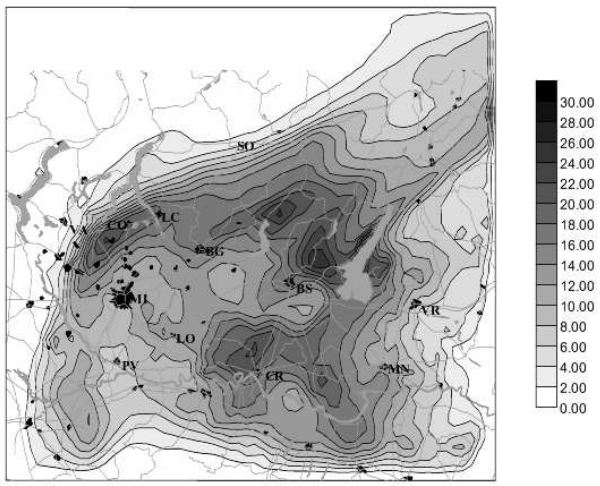

Fig. 8. Difference PM_{10} field (%) evaluated for June 4 1998, 16:00 for the 50% NH_3, NO_x, SO_2 increase scenario.

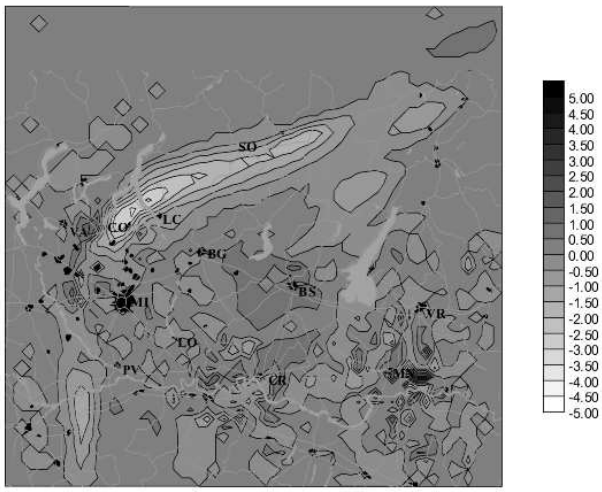

Fig. 9. Difference PM_{10} field (%) evaluated for June 4 1998, 16:00 for the 50% VOC reduction scenario.

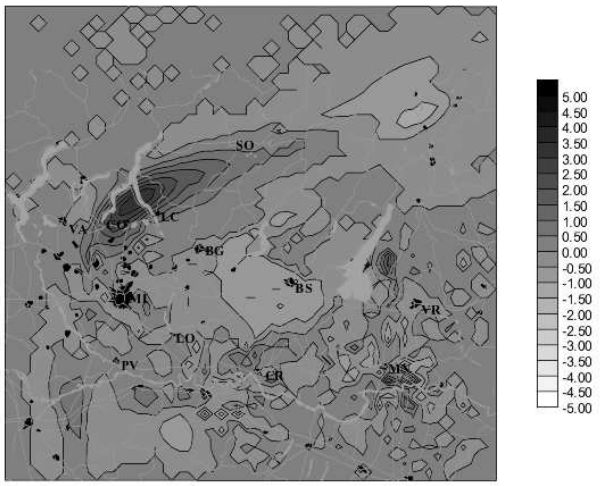

Fig. 10. Difference PM_{10} field (%) evaluated for June 4 1998, 16:00 for the 50% VOC increase scenario.

Region in 1998, 1-5 June during the PIPAPO experimental campaign. The simulations show that (1) a considerable fraction of aerosol concentrations during the simulated episode is due to the production of secondary species, (2) the particulate matter control strategies reducing gas primary emissions have different effectiveness over the domain, (3) the most effective PM reduction strategy is to limit NH_3 and NO_x emissions in a area where the most effective ozone reduction policy is to reduce VOC emissions.

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