CAN A MODELLING SYSTEM BIAS AIR QUALITY POLICY SELECTION?

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Abstract: In the frame of the CityDelta project, three different chemistry-transport models, CALGRID, CAMx and STEM were used to simulate different emission scenarios expected up to the year 2010 over a 300x300 km^2 domain located in Northern Italy. The aim of this work is to show the differences between the results produced by the three models. The results show that the three models give very similar results in terms of mean value, but they could give very different responses if the exceedance day indicator is used.

Keywords: Air Pollution, Decision Support System, Environmental Engineering, Mathematical models, Nonlinear systems

1. INTRODUCTION

In the last decade photochemical smog episodes have become more and more critical over Europe, mainly in the Mediterranean regions, where the sun radiation and the stagnating meteorological conditions occurring in the summer season play a significant role in chemical transformations of urban and industrial pollutant emissions. The cause-effect chain relations between precursors (typically nitrogen oxides (NO_x) and volatile organic compounds (VOC)) and photochemical pollutants (mainly tropospheric ozone, NO_2, PAN and formaldehyde) are thousands and characterized by different reactive times, resulting being complex and non-linear. In order to define and evaluate the emission control strategies for the reduction of photochemical pollution, regulatory Agencies need suitable Decision Support System (DSS). One of the DSS tools is the modelling system, performing the simulation of pollutant dynamics in troposphere. An air quality modelling system is composed by (i) a meteorological preprocessor, simulating the wind and temperature fields and the turbulence parameters over the domain, (ii) an emission processor, estimating emission fields for the emitted species alternative emission scenarios, (iii) a photochemical model that describes the transport and the chemical transformations taking place in the atmosphere. Due to the high complexity of the phenomenon under study the DSS answer could change varying the photochemical model. In this work, the different response to emission change of three different photochemical models are analysed and commented.

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2. THE PHOTOCHEMICAL MODELS

The photochemical models describe the transport phenomena and the main chemical transformations taking place in the atmosphere solving the general continuity equation 1.

$$\frac{\partial C_i}{\partial t} = T_i + R_i + D_i + S_i \tag{1}$$

where:

C_i is the concentration of species i;
T_i is the transport term;
R_i is the reaction term;
D_i is the deposition term;
S_i is the source (emissions) term.

The models need to solve equation 1 for each point of a three-dimensional grid describing the domain under study. This fact complicates the computational problem, enhancing the time costs even more. A simplification could consist in treating the different processes separately as sequential phenomena instead of simultaneous ones. A simple procedure could be based on the definition of the following submodels from equation 1 for the horizontal transport, the vertical transport including emission injection and deposition and, finally, the chemistry:

$$\begin{aligned}
\left[\frac{\partial C_i}{\partial t}\right]_{horiz} &= -v_x\frac{\partial C_i}{\partial x} - v_y\frac{\partial C_i}{\partial y} + K_{xx}\frac{\partial^2 C_i}{\partial^2 x} + \\
&\quad + K_{yy}\frac{\partial^2 C_i}{\partial^2 y} \\
\left[\frac{\partial C_i}{\partial t}\right]_{vert} &= -v_z\frac{\partial C_i}{\partial z} + \frac{\partial}{\partial z}\left(K_{zz}\frac{\partial C_i}{\partial z}\right) + \\
&\quad + S_i - D_i \\
\left[\frac{\partial C_i}{\partial t}\right]_{chem} &= P_i(C) - L_i(C)C_i
\end{aligned} \tag{2}$$

where:

v_x, v_y, v_z are the wind components;
C_i is the concentration of species i;
D_i is the deposition term;
S_i is the source (emissions) term.
K_{xx}, K_{yy}, K_{zz} are the turbulence coefficients;
P_i is the production term due to the chemistry;
L_i is the loss term due to the chemistry.

The models use a similar approach to solve the transport partial differential equations, but they deeply differ in the solution of the chemistry kinetics. Photochemical models could not consider all the chemical reactions taking place in the atmosphere so the chemical modules use the concept of *chemical mechanism* defining a sub-set of reactions and species describing the phenomenon more accurately as possible. Two approaches could be used to define mechanisms:

- in the lumped structure approach, organics are divided into smaller reaction elements, based on the types of carbon bonds in each species;
- in the lumped molecule approach, a particular organic compound or a generalized species is used to represent similar organics.

2.1 CALGRID

The photochemical model CALGRID (Yamartino *et al.*, 1991) is an Eulerian three-dimensional model for gas phase simulations. It implements an advection-diffusion scheme in terrain-following coordinates with vertical variable levels. The CALGRID chemical module implements the SAPRC90 (Carter, 1990) and the CBIV (Gery *et al.*, 1989) mechanisms. The QSSA (Quasi Steady State Approximations) algorithm solves the kinetic equations (Hessvedt *et al.*, 1978). During this intercomparison, the SAPRC90 mechanism was used to describe the chemistry.

The QSSA solver is a fully explicit method based on the assumption that, if the production and loss terms in equation 2 are constant during time step δt, equation 2 can be solved analytically as:

$$C_{i,n} = \frac{P_i}{L_i}(1 - e^{-L_i\delta t}) + C_{i,n-1}e^{-L_i\delta t} \tag{3}$$

The accuracy of this method highly depends on the choice of the time step δt. In case of a high number of equations in the system, the time step could be very small so that the assumption leading to equation 3 is respected. The SAPRC90 chemical mechanism follows a *lumped molecule approach* and it includes 54 species (10 fast reacting species, 30 slow reacting species, 5 constant and 9 steady-state), and 128 reactions(16 photolytic ones). The mechanisms have been evaluated using the results of a variety of environmental chamber experiments. The results show that the scheme is able to simulate maximum ozone concentrations and rates of NO oxidation and ozone formation to within 30% of the experimental measurements for 63% of the simulation tests, although there was a slight bias towards overpreddicting maximum ozone concentrations in experiments designed to represent ambient mixtures (Carter and Lurmann, 1991).

2.2 STEM-FCM

STEM-FCM (Silibello *et al.*, 2001) implements the same advection-diffusion scheme of CALGRID model and takes into account chemical transformations and deposition processes of both gas and aerosol species. The model uses SAPRC90

mechanism with an extended VOC chemistry. As for the kinetic equation integration, STEM-FCM implements the IEH solver (Sun *et al.*, 1994). Finally, aerosol treatment is based on a size-resolved multi-component aerosol module (Wexler *et al.*, 1994). The IEH solver consists in a hybrid implicit-explicit scheme that treats fast and slow reacting species in a different way. The system of fast species is solved by means of the Livermore Solver for Ordinary Differential Equations (Hindmarsh, 1975), which uses an implicit Backward Differentiation Formula method (Wille, 1994). Once the concentration of fast species is obtained for time t, the concentration of slow species is calculated using a second-order Adams-Bashforth scheme (Wille, 1994).

2.3 CAMx

The Comprehensive Air quality Model with extensions (CAMx) (ENVIRON, 2004) is a publicly available computer modelling system for the integrated assessment of photochemical and aerosol air pollution over many scales. CAMx provides the option of using two different chemical mechanisms: SAPRC 1999 (Carter, 2000) version and CBIV 1999 version, modified to model ozone and fine/coarse PM, using RADM (Chang *et al.*, 1987) mechanism for aqueous phase chemistry, ISORROPIA (Nenes *et al.*, 1998)for inorganic sulphate-nitrate-ammonium chemistry, SOAP semi-volatile scheme for secondary organic aerosols. About the gas-phase chemistry solver, the CAMx user can choose either IEH (Implicit-Explicit Hybrid) solver or a fast efficient one developed by ENVIRON and referred to as CMC (ENVIRON, 2004). The two approaches give similar results during the day but differ during the night-time (in this case IEH is more accurate than CMC). In this work, CAMx was applied using the CMC solver with SAPRC99 mechanism. The CMC fast chemistry solver uses an adaptive-hybrid numerical approach to solve the gas-phase chemistry. The fastest reacting species (the radicals) are solved assuming that their concentrations are fully equilibrated to the slower reaction species. The latter are divided into two groups: fast state species with chemical lifetimes of seconds to a few minutes, and slow state species with longer chemical lifetimes. The fast species system is solved using a Runge-Kutta implicit methods, while the concentration of slow species is obtained explicitly. In particular, NO, NO_2 and ozone are always considered fast state species, whereas PAN is considered fast only if its concentration is higher than 1% of NO_2 ones. The SAPRC99 mechanism represent a complete update of SAPRC90. It incorporate more recent reactivity data from a wide variety of VOCs, assigning $\sim$ 400 types of VOCs. It can be used to estimate reactivities for $\sim$ 500 VOC categories. The mechanism was evaluated against the result of approximately 2000 environmental chamber experiments, including test on ozone reactivity predictions.

3. SIMULATION SETUP

In the frame of CityDelta European Project (http://rea.ei.jrc.it/netshare/thunis/citydelta/), the three photochemical models have been applied over a 300 x 300 area centered to Milan (Figure 1). The simulations concerned the 1999 six summer months hourly concentrations of ozone. The emission, the meteorological, initial and boundary condition data set, collected by the Joint Research Centre for the CityDelta project, were processed to provide suitable input fields to the models.

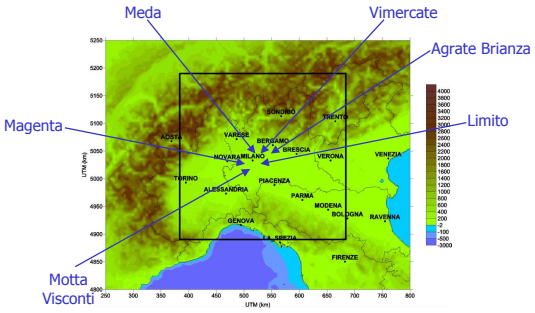

Fig. 1. Simulation domain

4. SIMULATION DISCUSSION

The validation of the three models was performed in relation to the six air quality monitoring stations nearest to the Milan urban area among those selected by JRC (Figure 1). The six months hourly concentrations simulated by the photochemical models were mainly evaluated by means of a statistical tool provided by JRC.

4.1 Validation of the model simulations

The validation analysis for the three models is discussed in (Bedogni *et al.*, 2004*a*), (Bedogni *et al.*, 2004*b*). The results presents a good agreement with data as Figure 2 summarizes.

4.2 Emission Scenario impact assessment

In order to perform a sensitivity analysis with respect to emission change, three different scenarios have been considered. The definition of alternative scenarios was performed by means of two different

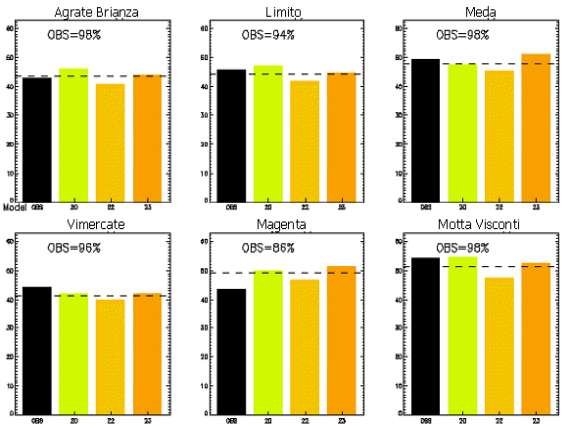

Fig. 2. Mean hourly ozone concentration (ppb) from 8 to 19 hour over the simulation period

kinds of reduction coefficients, calculated by JRC-IIASA (http://www.iiasa.ac.at/rains/index.html). The first scenario (CLE) is related to the emission reduction obtained in the year 2010 by the application of Current LEgislation. The MFR-scenarios take into account the Most Feasible emission Reduction that could be obtained using the best technology for NOx and VOC. Table 1 shows the reduction (in %) of VOC and NOx emissions for the scenarios with respect to the base case. The evaluation of the scenario impact has been performed in relation to 9 points distributed over the domain as presented by figure 3.

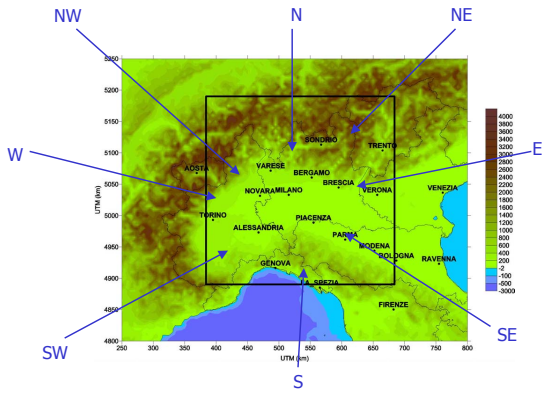

Fig. 3. Position of the nine points used for the emission scenario evaluation

Table 1. Total domain emission reduction for the scenarios with respect to the base case (%)

Scenarios	NOx	VOC
CLE	-36.2	-41.5
MFR(NOx)	-51.1	-41.5
MFR(VOC)	-36.2	-61.1

4.2.1. CLE scenario Figures 4 and 5 show the differences in the mean values and days of exceedance the 50 ppb threshold between the base case and the CLE emission scenario for the three models (STEM(20), CALGRID(22), CAMx(23)).

The complexity and non-linearity of the phenomenon is highlighted by the fact that, even the reduction of the emissions of main ozone precursors are higher than 30%, its concentration decrease is lower than 3 ppb (approximately the 5% with respect to the base case). The differences are higher if the exceedance day indicator is used. The results show that CALGRID and STEM have a very similar behavior even in terms of mean and exceedance. CAMx model mean shows a higher decrease in the North of the domain, where the terrain is more complex, and a smaller reduction near the city of Milan, characterized by higher emissions.

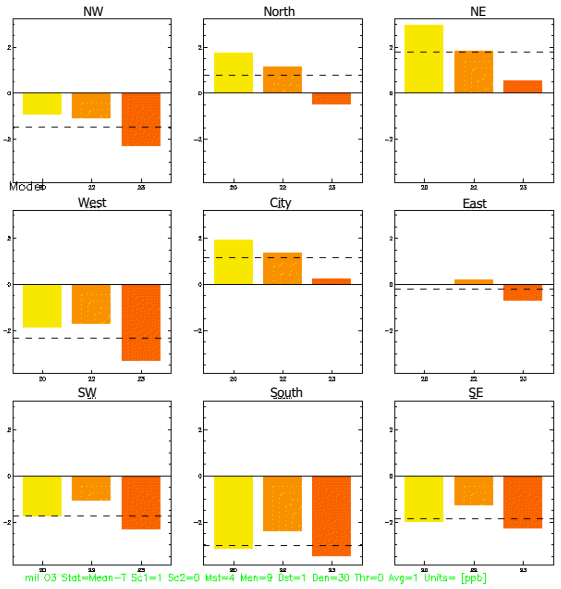

Fig. 4. Mean hourly ozone concentration difference (ppb) over the period between base case and CLE scenario

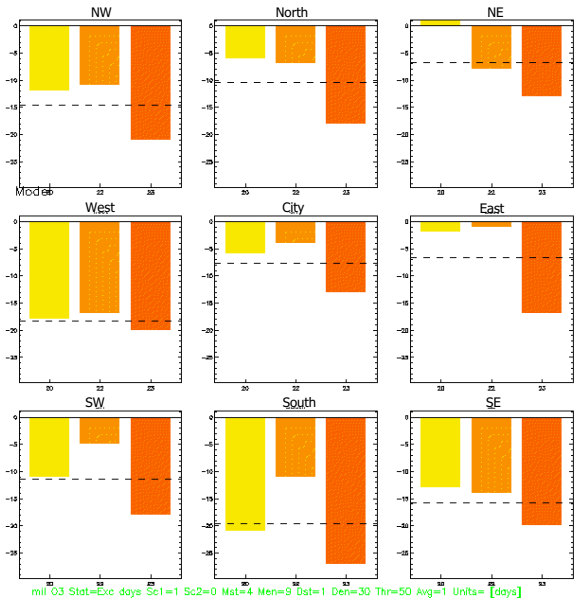

Fig. 5. Number of exceedance day difference (#) between base case and CLE scenario

4.2.2. MFR(NOx) scenario In terms of mean value, the three models show the same behavior when the most feasible reduction scenario is applied to NOx domain emission (Figure 6). The model simulations exhibit a VOC-limited area in the center-north of the domain, where an increase of ozone concentration could be noted, and a NOx-limited area in the south.

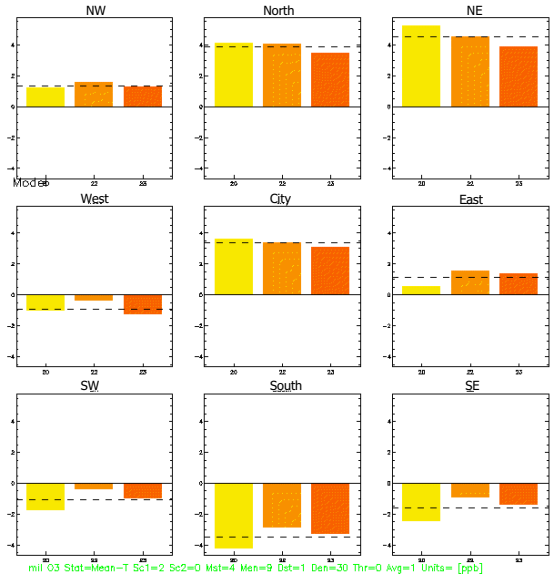

Fig. 6. Mean hourly ozone concentration difference (ppb) over the period between base case and MFR(NOx) scenario

The results of the three models are significantly different in terms of exceedance days (Figure 7), with CALGRID model that reproduces an increase in the exceedance days in the North of the domain.

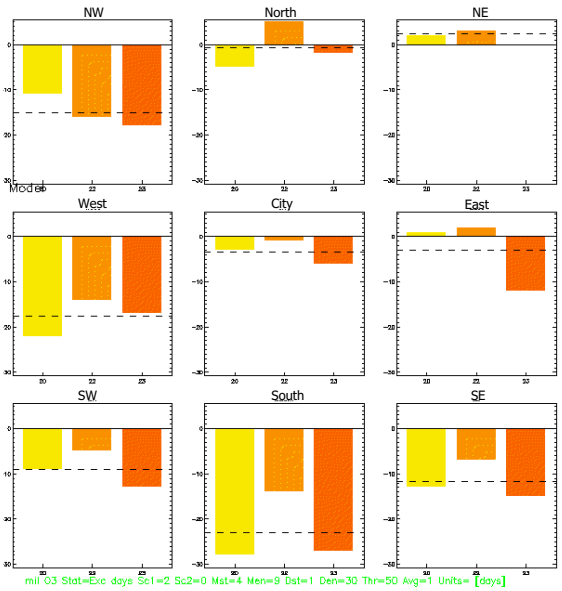

Fig. 7. Number of exceedance day difference (#) between base case and MFR(NOx) scenario

4.2.3. MFR(VOC) scenario The last simulated scenario considers the most feasible reduction applied to VOC emission and it has the highest impact in terms of ozone concentration reduction. CAMx simulation shows that the model is more sensible to the VOC emission change with respect to other models. This behavior is especially showed by the mean over the period (Figure 8) but is confirmed also by the exceedance days (Figure 9).

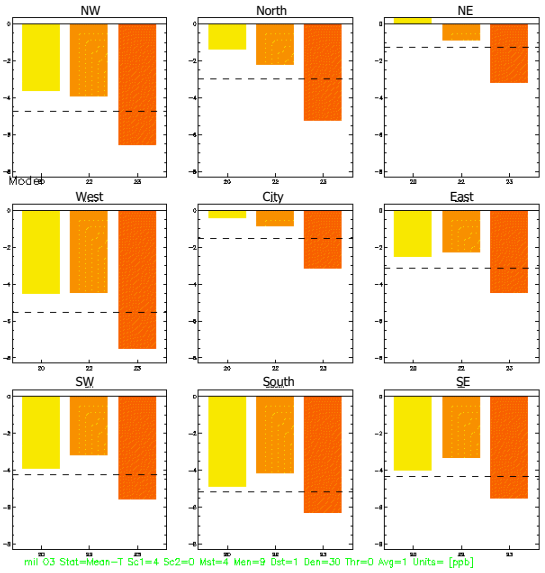

Fig. 8. Mean hourly ozone concentration difference (ppb) over the period between base case and MFR(VOC) scenario

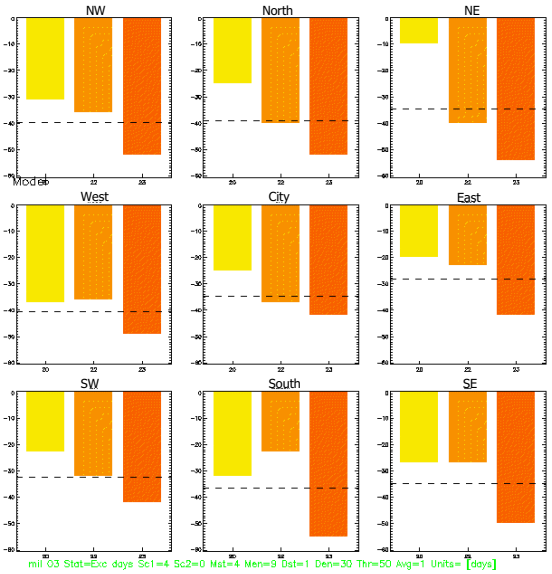

Fig. 9. Number of exceedance day difference (#) between base case and MFR(VOC) scenario

5. CONCLUSION

In this work, a sensitivity analysis on how the modelling system can bias the emission reduction

policy selection is presented. In terms of mean concentration value, the three considered models give very similar responses. The differences are higher if the exceedance day non-linear indicator is used (Table 2).

Table 2. Mean number of exceedance days over the 9 considered points

Scenarios	STEM	CALGRID	CAMx
CLE	-9.78	-8.33	-19.11
MFR(NOx)	-9.56	-4.78	-11.78
MFR(VOC)	–24.89	-31.89	-47.00

CAMx model is more sensible to ozone precursors emission reduction, probably due to the higher reactivity of chemical mechanisms (SAPRC99) and solver (CMC) implemented. STEM and CALGRID results show a lower impact of emission reduction. Moreover, they exhibit different behavior with respect to NOx and VOC reduction. In particular, when the MFR(NOx) scenario is applied, STEM shows a sensitivity very close to that of CAMx, while in the MFR(VOC) case, the exceedance day decreasing calculated by CALGRID is higher than STEM one. These results could be explained by the difference between the chemical treatment of VOC in the two model.

In conclusion, the results show that the application of different photochemical models could bias the emission policy selection. If the control policy concerns the mean value concentration, none of the three models is preferred, while if excedence days is considered as indicator, a conservative choice could drive to the selection of CALGRID model.

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