# PM10 TRANSPORT AND DIFFUSION IN NORTHERN ITALY

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**Abstract**: A secondary pollution modelling system for simulating airborne dispersion and chemical reactions is tested over a regional scale domain located in the North-West of Italy, covering areas characterized by different emission levels and where urban and industrial areas are present. This region is often affected by severe pollution episodes, which are driven by anthropogenic emissions and meteorological conditions. The domain is centred over a main plain area, surrounded by elevated Alpine mountains and the sea. This valley is characterized by unfavourable meteorological conditions, such as weak circulation, frequent low wind and stagnant conditions. Moreover the complexity of the orographic site affects both reconstruction of a proper emission picture and meteorology, in particular at the domain borders where the Alpine mountains are located. The model resolution was 5 km and simulations have been run for the whole year 1999. Emissions are derived from different inventories based on different territorial units. The meteorological input was provided by the meteorological model RAMS, whereas the dispersion and chemical reactions were computed by CAMx photochemical model. The results of the simulations are compared together with measured sulphur dioxide, nitrogen dioxide and particulate matter data. Two winter periods were considered because of the PM accumulation processes that take place during the cold season. The simulated PM concentrations are also compared with the results of the same modelling system, applied over a larger scale and at lower (25 km) resolution. CAMx provided quite satisfactorily performances in reproducing the seasonal evolution of PM10, particularly at 5 km resolution. Afterwards, model performances have been compared against $SO_2$ and $NO_2$, two of the most important particulate matter precursors. As expected, the comparison shows a worsening in the model performance moving from gas species to PM10, confirming that some processes related to particulate matter are still missing in chemical transport models. Moreover, PM10 performances proved to be related more to $NO_2$ than $SO_2$. In fact, while the former gives rise mainly to local scale production, sulfate formation is generally related to large-scale processes, hence less linked to the precursor concentration at the receptor point.

***Key words:*** *atmospheric dispersion, secondary pollution, numerical modelling.*

## 1. INTRODUCTION

PM is one of the most significant pollutants with respect to the potential impacts to human health and natural ecosystem, both in terms of critical episodes and as long-term exposures. Consequently, in order to assess the effects of secondary pollution, pollutant exposures on "seasonal" scale and peak concentrations need to be quantified. Modeling systems can represent suitable tools for this purpose (Carmichael et al., 1986; Sokhi et al., 2007).
In this study an integrated modelling system has been developed in order to perform a simulation of the secondary pollution around some main modules: an emission pre-processor developed by our group (Balanzino et al., 2007 a and b), the prognostic non-hydrostatic meteorological model RAMS (**R**egional **A**tmospheric **M**odelling **S**ystem, Pielke et al., 1992), the interface module RAMS-CAMx (Environ. http://www.camx.com/down/support.php) and the Eulerian model CAMx (**C**omprehensive **A**ir Quality **M**odel with e**x**tensions, Environ, 2005). The modelling system was applied over a domain placed in Northwest Italy and covering areas characterized by different emission levels: an industrial high emission level area nearby the cities of Milan, Turin and Genoa and a low emission level rural area, centred on the eastern Piedmont valley which is often affected by very high particulate matter concentrations during wintertime (Vecchia et al., 2004; Giugliano et al., 2005, Finardi et al., 2008; Calori et al., 2006; De Maria et al., 2007). This pollution phenomenon is driven by anthropogenic emissions and meteorological conditions. In fact the Po river valley is very industrialized and populated and it is characterized by strong urban, industrial and traffic emissions; moreover, the presence of the Alps often gives rise to weak circulation and stagnant conditions (Minguzzi et al., 2005; Bonafè et al., 2006). The simulated data are compared against the $NO_2$, $SO_2$, and PM10 measurements. Furthermore the results of a second simulation, carried out on a mesoscale domain including the whole Italian peninsula, were considered for sake of comparison (Costa et al., 2006). The effects due to the different scales and resolutions are analysed and discussed. In fact, grid resolution is widely recognized (Vautard et al., 2007) as a critical parameter, strongly influencing model performances on the one side and computational time on the other one.

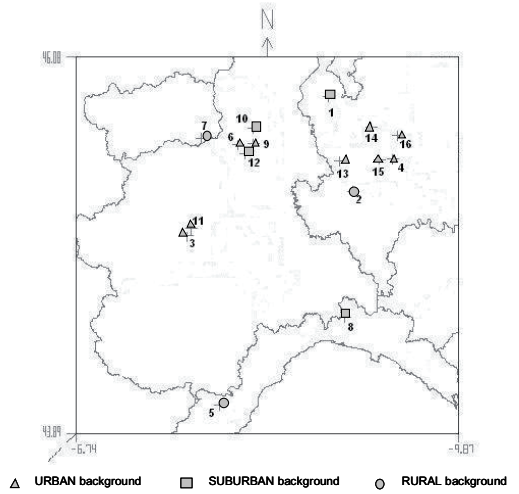


Figure1. Simulation domain and monitoring sites

## 2. METHODOLOGY

The proposed modeling system includes the meteorological model, several interface modules (e.g. RAMS-CAMx), an emission processor, developed by our group (Balanzino et al., 2007 a and b), and the photochemical model. RAMS resolves the full set of the primitive dynamic equations. The model is driven by means of the nudging technique using the ECMWF (European Centre for Medium-Range Weather Forecasts) analysis as initial and boundary conditions. The Mellor and Yamada 2.5 scheme (Mellor and Yamada, 1982) is used in RAMS to compute the turbulence contribution. The RAMS simulation was carried out using one grid only, over a 250x250 $km^2$ domain (Fig. 1) with a horizontal resolution of 5 km and a stretched vertical grid with a minimum grid space of 70 m near the domain bottom. The domain includes different Northwest Italian regions, a portion of Mediterranean Sea, and some European areas and it is characterized by a main plain area (the Po valley), surrounded by elevated Alpine mountains. RAMS provides mean three-dimensional hourly fields of velocity and temperature, as well as the turbulent kinetic energy through interface modules, that are useful tools in order to translate RAMS output meteorological fields to CAMx inputs. The interface modules also provide height/pressure ratio, water vapor, cloud and rainwater content, topography and land use, and the photolysis rates.

Table 1. $PM_{10}$ indeces computed on the mean daily concentration (winter time) at 5 km resolution

| station | period | MEAN_meas ($\mu g/m^3$) | MEAN_camx ($\mu g/m^3$) | R | NMSE | FB |
|---|---|---|---|---|---|---|
| URBAN | first | 59.4 | 36.8 | 0.7 | 0.5 | 0.5 |
| | second | 71.3 | 44.6 | 0.3 | 0.7 | 0.5 |
| SUBURBAN | first | 26.0 | 24.4 | 0.3 | 0.5 | 0.1 |
| | second | - | - | - | - | - |

An emission processing system was designed to produce emission fields. As the data provided by emission inventories are unsuitable for air quality modeling purposes (*Seinfeld, 1988*), the emission processor performs the spatial disaggregation, the temporal modulation, the hydrocarbon speciation and the particulate speciation.

According to the speciation profile (Passant, 2002), total VOC emissions of a specific source are split into single organic species and then lumped into the aggregated classes handled by the model (in our case the required species are 13). Finally the particulate speciation (CARB, 2000) is performed according to the aerosol mechanism options chosen for treating the aerosol size distribution. The scheme actually implemented divides the size distribution into two bins (coarse and fine). Primary species are modeled as fine and/or coarse particles, while all secondary species are modeled as fine particles. According to the model, the PM required species are 15.

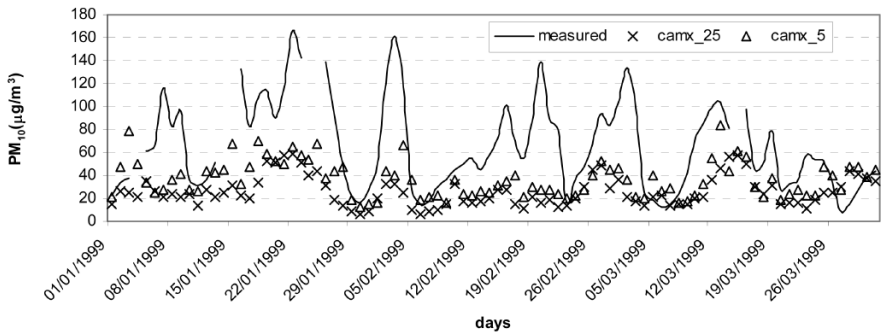


Figure 2. Mean daily $PM_{10}$ concentration at urban site 4 for the first period.

In this study, within the Italian borders, emission data are provided by the regional emission inventories (more detailed than the national inventory) while for European areas and for the portion of Mediterranean Sea emission data have been derived by the EMEP inventory.

The Eulerian photochemical dispersion model CAMx simulates the emission, dispersion, chemical reaction and removal of pollutants in the troposphere by solving the pollutant continuity equation for each chemical species on a system of nested three-dimensional grids. CAMx implements an extended version of the CB-IV mechanism (Whitten et al., 1980, Gery et al., 1989) including aerosol and mercury chemistry and several additional inorganic reactions appropriate for regional modeling conditions. The CAMx simulation area coincides with the RAMS domain (Figure 1) and has been horizontally subdivided into 50x50 cells, with a space resolution of 5 km whereas the vertical domain extends up to 5490 m agl, subdivided into 14 layers of growing thickness. The initial and boundary conditions were provided by a larger scale simulation carried out with a similar modeling system (Costa et al., 2006). The vertical diffusion coefficients have been prescribed on the basis of the turbulent kinetic energy fields provided by RAMS according to the Mellor and Yamada 2.5 scheme.

Both meteorological and photochemical models have been run for the whole year 1999, supplying hourly concentrations fields. Two winter periods (January-March and October-December) were considered for this analysis because of the PM accumulation processes that take place during the cold season. In fact, high PM levels are due to strong emissions and unfavorable meteorological conditions, such as high pressure, weak circulation and episodes of thermal inversion.

## 3. RESULTS AND DISCUSSION

A comparison between observed and predicted sulphur dioxide, nitrogen dioxide and particulate matter levels at different kind of monitoring sites (suburban and urban stations) were performed. However, the limited number of measurements available during 1999 at monitoring sites made difficult the comparison. The stations were grouped by category (urban and suburban) and the standard statistical indices: mean, correlation coefficient (R), normalized mean square error (NMSE) and fractional bias (FB) were calculated for the mean daily PM10 concentration for the simulation at 5 km resolution (Tab. 1). Concerning the first period, the model underestimates the measurements at both the urban and suburban sites. In particular, the correlation coefficient improves when moving from suburban to urban stations, even if, in the suburban case, the model well reproduces the mean values and the FB is smaller. The analysis of the second period statistical indices demonstrates that the model performances are slightly worse at urban sites respect to the first period: the correlation between observed and simulated values decreases, but the FB improves, even if the model still underestimates the measurements. The dashes indicate the cases where the observed PM10 data were not available.

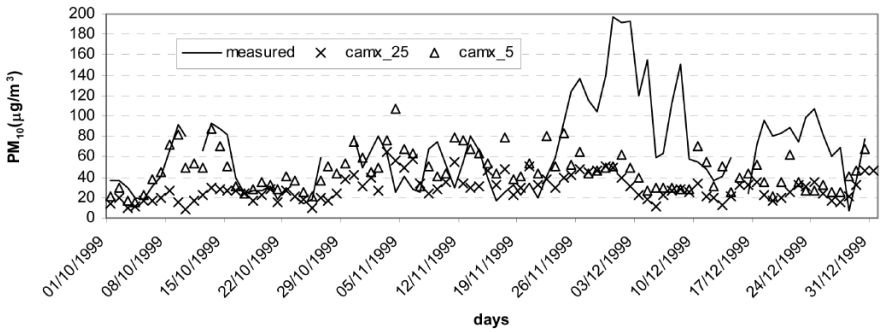


Figure 3. Mean daily $PM_{10}$ concentration at urban site 4 for the second period.

Concerning $NO_2$, $SO_2$ and PM10, the correlation coefficients (R) and the FB are shown in Table 2, for the simulation at 5 km resolution and for both periods at suburban and urban stations. The dashes indicate the cases where the observed $NO_2$, $SO_2$ and PM10 data were not available.

In Table 2, it is possible to observe a similar behaviour for PM10 and $NO_2$: in both periods, when the PM10 correlation increases, the $NO_2$ correlation increases and vice versa. Generally, for both urban and suburban sites, the correlation is better in the first period than in second one. Unlike the previous $NO_2$ case, an opposite behaviour between precursor species $SO_2$ and product species PM10 for urban station (the suburban observed $SO_2$ data were not available) seems to appear (Tab. 2). Generally the model $SO_2$ performances are better in the second period than in first one, while the opposite behaviour occurs for PM10. In the first period, the PM10 correlation between measured and computed value is rather good. Instead, in the second period, the $SO_2$ correlation slightly improves, but PM10 correlation gets worse. Concerning $NO_2$, the FB has an opposite behaviour with respect to the correlation coefficient for urban and suburban stations: in both periods, the FB improves when the correlation gets worse. In general, at both sites, the model underestimates the observed PM10 and $NO_2$ values in both first and second period. It is worth noticing that, in the case of the urban stations, the model overestimates $SO_2$, but underestimates PM10.

Table 2. Correlation coefficients and fractional bias (winter time)

| station | period | R | | | FB | | |
|---|---|---|---|---|---|---|---|
| | | $NO_2$ | $SO_2$ | $PM_{10}$ | $NO_2$ | $SO_2$ | $PM_{10}$ |
| URBAN | first | 0.7 | 0.3 | 0.7 | 0.4 | -0.9 | 0.5 |
| | second | 0.4 | 0.4 | 0.3 | 0.1 | -1.0 | 0.5 |
| SUBURBAN | first | 0.6 | - | 0.3 | 0.3 | - | 0.1 |
| | second | 0.5 | - | - | 0.01 | - | - |

The simulation results were also compared with those of another simulation carried out on a mesoscale domain (1400x1600 $km^2$) having a resolution of 25 km (Costa et al, 2006). Thirteen layers, up to 9000 m a.s.l, compose the vertical structure. The Italian domain simulations have been performed by means of the same modeling system, but

with different input data: Italian emissions have been derived from the Italian official inventory, while chemical initial and boundary conditions have been obtained by the CHIMERE model (Vautard et al., 2005).

Table 3. $PM_{10}$ indices computed on the mean daily concentration (winter time)

| station | period | R_5km | FB_5km | R_25km | FB_25km |
|---|---|---|---|---|---|
| URBAN | first | 0.7 | 0.5 | 0.6 | 0.9 |
| | second | 0.3 | 0.5 | 0.1 | 0.9 |
| SUBURBAN | first | 0.3 | 0.1 | 0.3 | 0.4 |
| | second | - | - | - | - |

Concerning PM10, the comparison between observed and predicted mean daily concentrations at one monitoring urban station (n.4) is presented in the Figures 2 (first period) and 3 (second period) The comparison of the time series highlights that both models generally underestimate the observations and show a good temporal variability, but the high-resolution model better reproduces the measured values, particularly in some critical episodes (e.g. in October and November 1999).

The correlation coefficients (R) and the FB are shown in Table 3, for PM10 and for both grid resolutions at suburban and urban stations. The dashes indicate the cases where the observed PM10 data were not available.

Regarding the urban stations, the high resolution simulation provided higher correlation coefficients than the large scale one, while in the suburban case, the correlation between observed and simulated values decreases for both camx_5 and camx_25. In general, the model performances at the urban sites get worse when moving from the first to the second period. The FB improves using a grid spacing of 5 km that better describes the concentration spatial gradients. Generally, the analysis of the second period statistical indices at urban sites demonstrates that the model (camx_5 and camx_25) performances are slightly worse respect to the first period, except for FB, which improves in the second period, even if the model still underestimates the measurements. The dashes indicate the cases in which the observed PM10 data were not available.

## 4. CONCLUSIONS

A secondary pollution modeling system has been applied to simulate airborne dispersion and chemical reactions on a local scale domain (250x250 $km^2$) placed in the North-West of Italy with a grid resolution of 5 km. The simulation has been run for the whole year 1999. The results of the simulations are compared together with measured sulphur dioxide, nitrogen dioxide and particulate matter data, as well as the results of a simulation performed at a lower resolution. Due to the PM accumulation processes that take place during the cold season, the two winter periods were considered for the analysis. As for PM10, the model shows a general underestimation of the observed concentrations. The mean suburban values are better than the urban one. As a matter of fact the reproduction of winter PM10 concentrations in complex meteorological conditions such that within the Po valley is still a challenge for air quality modeling. As for the particulate matter precursors ($NO_2$ and $SO_2$), a different behaviour is shown at urban and suburban sites: when $NO_2$ gets worse, $SO_2$ slightly improves. In particular the model PM10 performances seem to be more affected by $NO_2$ than $SO_2$: in the first period, the reproduction of winter PM10 concentrations improves when the $NO_2$ correlation increases. The different PM10 dependence from the precursors highlights that, at local level, the relation $NO_2$-nitrate-PM10 is stronger than the corresponding one $SO_2$-sulfate-PM10. This is due to the higher weight of nitrate than sulfate in urban and suburban areas and also to the different formation pathways. In fact, while the former gives rise mainly to local scale production, sulphate formation is related to the oxidation of sulphur dioxide, mostly emitted by large point source, hence its concentration is less linked to the precursor availability at the receptor point.

The performed analysis suggests that varying horizontal grid resolution from 5 to 25 km can influence the PM10 concentrations: both models generally underestimate the observations. However the high-resolution model better reproduces the measured values, particularly in some critical episodes. Moreover the FB improves using a grid spacing of 5 km, which allows better description of the spatial concentration gradients, but this does not involve a better description of the temporal variability, measured by correlation coefficient, which assume comparable values. In fact the temporal variability is related to the model skillfulness in reproducing correctly the daily evolution of the atmospheric circulation over the studied domain. The underestimation for this pollutant may be attributed to a number of factors: the uncertainty in the emission inventories, the spatial resolution considered (also the grid resolution of 5 km may be too low for small sized urban sites) and the adopted aerosol modeling approach which does not include or does not well reproduce some processes such as dust transport; dust resuspension; emission and formation of organic aerosol.

***Acknowledgements:*** *The authors wish to acknowledge ARPA Piemonte Meteorological Service for providing meteorological data and emission inventory, Regione Piemonte ("Bandi regionali sulla ricerca scientifica applicata 2003 e 2004") and Provincia di Alessandria for supporting the project in which this work is carried out. CESI RICERCA contribution has been financed by the Research Fund for the Italian Electrical System under the Contract Agreement between CESI RICERCA and the Ministry of*

*Economic Development - D.G. for Energy and Mining Resources stipulated on June 21, 2007 in compliance with the Decree n.73 of June 18, 2007.*

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