ASSESSING THE CONTRIBUTION OF THE MAIN EMISSION SOURCES TO PARTICULATE MATTER CONCENTRATIONS IN THE MILAN AREA

Marco Bedogni[1], *Simone Casadei*[1] *and Guido Pirovano*[2]

[1]Mobility and Environment Agency of Milan, Italy
[2]CESI Ricerca Spa, Milan, Italy

Abstract: On the basis of CityDelta III project input dataset, the CAMx chemical and transport Eulerian model has been applied over a 300x300 km^2 domain focused on the Milan area (Northern part of Italy) for the whole 2004. Model results have been analysed by mean of the Source Apportionment technique at the Milan receptor, in order to discriminate the contribution of several key emission sectors, also distinguishing the most emitting area of Milan from the surroundings. Obtained results highlight that road transport is the most relevant sector, followed by agriculture and domestic heating. The source apportionment analysis also revealed that more than 70% of the PM10 is due to emissions outside of Milan.

Key words: *Source apportionment technique, PM10 modelling, Chemical and Transport Models, Po Valley air quality*

1. INTRODUCTION

Particulate matter is widely recognized as a dangerous pollutant that may lead to serious health effects. The analysis of the PM10 observations (EEA, 2007) confirmed that EU limit values were still exceeded in 2004 and, despite of a clear downward trend in both primary and precursors emissions, no noticeable decreasing trends were put in evidence by the observed time series. In particular, the Milan area and the whole Northern Italy basin are subject to very critical conditions, due to their peculiar meteorological conditions, with frequent exceedances of the air quality standards mainly during winter time.

However, the definition of effective emission reduction policies for PM is a challenging task due to the relevant non–linearity that influences the interactions among the different sources. To this aim, numerical models can represent a very powerful tool, mainly in order to detect the role played by the different emission sources. In particular, CAMx model implements the PSAT algorithm (Particulate Source Apportionment Technology) that allows to discriminate the contribution of different emission sectors and areas to the modelled PM concentrations. PSAT is embedded in CAMx code thus allowing reducing the effect of non-linearity, arising in more traditional approach such as the *zero-out* modelling (i.e. removing a subset of sources).

This work describes the results of the CAMx modelling system simulations, performed for the whole 2004 over a 300x300 km^2 domain focused on the Milan area, and the main conclusions of the analysis carried out by mean of the source apportionment technique.

2. CAMx MODEL AND THE CITYDELTA MODELLING INTERCOMPARISON

Due to the increasing interest in long term simulations with Chemical Transport Models (CTMs) as a support of air quality assessment and policies analysis, the CityDelta model intercomparison exercise (http://aqm.jrc.it/citydelta) has been organized by the Joint Research Centre (JRC-IES) of Ispra, in collaboration with EMEP, IIASA and EUROTRAC, as a contribution to the modelling activities in the CAFE (Clean Air For Europe) Programme (6th Framework Programme). The aim of the CityDelta exercise was to compare the results of different photochemical dispersion models in order to estimate air quality response, namely tropospheric ozone and particulate matter, to local and regional emission variations (Cuvelier et al., 2007; Thunis et al., 2007).

The Comprehensive Air quality Model with eXtensions (CAMx) chemical and transport model (Environ, 2006) has been applied over the Milan domain during all CityDelta phases to simulate both ozone and PM10 (Bedogni et al., 2005; Minguzzi et al., 2005; Angelino et al., 2007). In order to simulate particulate matter concentrations CBIV chemical mechanism (Gery et al., 1989) 1999 version has been used, together with RADM (Chang et al., 1987) mechanism for aqueous phase chemistry. ISORROPIA equilibrium model (Nenes et al., 1998) has been implemented for inorganic sulphate-nitrate-ammonium chemistry, whereas SOAP (Strader et al., 1999) semi-volatile scheme has been used to reproduce secondary organic aerosols.

The acquired knowledge on the CAMx capabilities concerning the Northern Italy context allowed the modelling group to perform specific applications of the PSAT algorithm (Particulate Source Apportionment Technology) in relation to the city of Milan. PSAT algorithm (Yarwood, G. et al., 2004) allows discriminating the contribution of different emission sectors and areas to the modelled PM concentrations. PSAT is embedded in CAMx code, thus allowing reducing the effect of non-linearity, arising in more traditional approach such as the *zero-out* modelling (i.e. removing a subset of sources).

3. DOMAIN AND INPUT DATA

The computational domain (300 x 300 km^2), centred on the city of Milan, includes a large flat area (all the central and most of the western Po Valley) surrounded by mountains along three sides (Fig. 1). It is a complex test bench in which low wind speeds and intense solar radiation cause high ozone episodes during summer and critical PM concentration levels during winter. The domain has been horizontally divided into 5x5 km^2 cells and vertically in 11 varying levels ranging from 30 to 5800 m. More details concerning the computational domain and the modelling system are reported on Angelino et al. (2007).

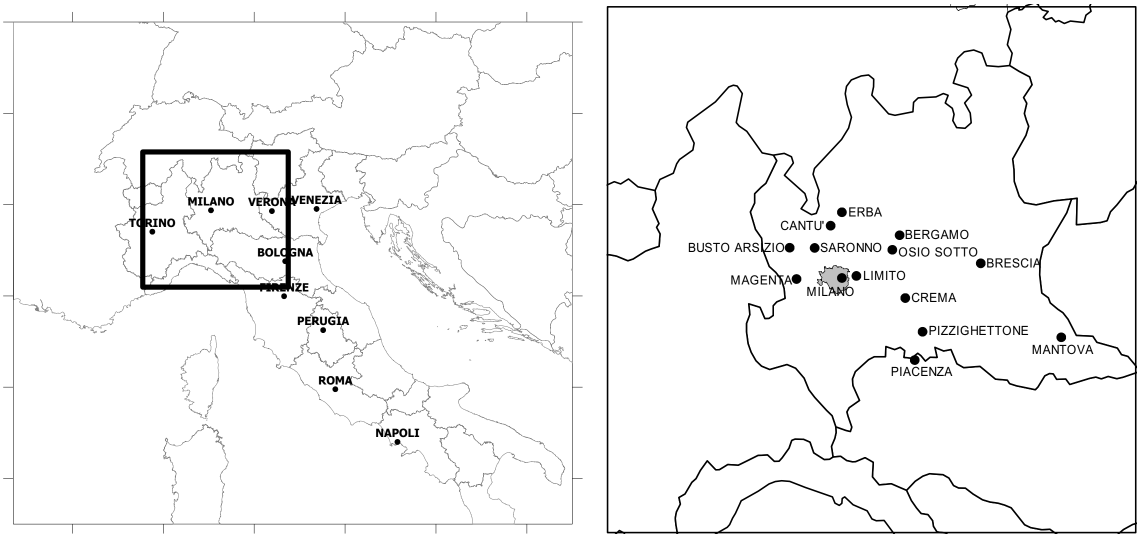

Figure 1. Computational domain and location of the selected air-quality monitoring stations.

Raw input data (emissions, boundary conditions and meteorological fields) have been provided by JRC to all participants of CityDelta III exercise. In particular, emission database over the computational domain was a combination of the EMEP inventory (EMEP, 2006) outside Lombardy Region, with a resolution of 50×50 km^2, and a local inventory covering Lombardy Region at a 5×5 km^2 resolution. But, to perform the simulation here analysed, local emission inventory has been replaced with the 2003 Lombardy Region inventory (ARPA Lombardy and Lombardy Region, 2006). The final inventory includes yearly emission data of NO_X, VOC, CO, NH_3, SO_X, PM10 and PM2.5 for the 11 SNAP sectors and for both area and point sources. Total primary fine particulate emissions are mainly due to domestic heating and road transport that represent respectively 32% and 24% of the whole PM2.5 emissions. As for the SIA gas precursors, NO_X emissions are primarily related to road transport (about 50%), power plants sector is the main responsible of SO_X emissions (45% of the total load), and NH_3 emissions are largely due to agriculture (more than 95% of the total load).

4. VALIDATION

As already mentioned above, within the framework of the different CityDelta phases CAMx model results have been compared to a considerable dataset of observed concentrations over Milan domain. Moreover, the comparison with the results produced by the transport and chemical models participating into the CityDelta exercise demonstrated that the performances of the CAMx model are similar, or sometimes better, to the other ones, putting in evidence that CAMx implements the current state-of-art knowledge on PM10 chemistry and dynamics (Vautard et al., 2007).

Nevertheless the present simulation, being carried out using a different local emission inventory, has been submitted to a new validation process, mainly in relation to the particulate concentrations. The evaluation is focused only on PM10, because in 2004 PM2.5 observations were not available. The validation phase has been performed by comparing daily simulated PM10 concentrations with observations at the available stations. In 2004, 69 PM10 stations were active over the computational domain. A selection of these stations has been carried out: stations outside Lombardy Region have been excluded, because the resolution of the emission inventory is too low (50 km) to make possible a real comparison between measured and simulated values. Likewise, stations classified as "road traffic" or "industrial" as well as stations placed in the mountain part of Northern Lombardy have been excluded, because the resolution of the local emission inventory (5 km) was not fine enough to allow a correct simulation of local scale phenomena by the CAMx model. The 14 remaining monitoring stations (Fig. 1) are all classified either as urban or rural background.

Table 1 summarizes some statistics depicting the model performances, such as mean values, Normalised Mean Bias (NMB), Mean Fractional Bias (MFB), Mean Fractional Error (MFE) and correlation. Statistics outline a particular spatial trend of the model performances, with the best representation of the observed values in the Southern part of Lombardy Region and the worst in the Northern part, where the underestimation of the PM10 annual mean concentrations overcomes 40% and the correlation between observed and modelled PM10 daily mean concentrations is less than 0.6. Over the central part of Lombardy (Milano, Limito, Magenta etc.) CAMx model underestimates annual mean values by around 30% while the correlation ranges from 0.6 to 0.7. The southern monitoring stations (Crema, Pizzighettone, Piacenza) are apparently well represented. Incidentally, the emission inventory resolution seems to have great importance in the simulation of particulate matter: in relation to the monitoring stations located outside Lombardy, CAMx performances are the worst in terms of both mean concentration values and correlation.

As for ozone values (Tab. 1), CAMx performances are quite good, with a correlation value ranging from 0.80 to 0.92 and a general overestimation of the yearly mean value of the daily maximum concentrations (average NMB value over the validation stations: +23%) that becomes smaller if O_X annual mean (i.e. $O_3 + NO_2$) is considered instead of O_3 daily maximum (average NMB value: +10%).

Finally, model evaluation has been extended to the main PM2.5 compounds. Because routinely observation of PM2.5 compounds were not available for the simulation period, CAMx results have been qualitatively compared to the seasonal distribution of some measurement campaigns undertaken in the Milan urban area during 2002 and 2003 (Lonati et al., 2005; Lonati et al., 2007): CAMx results showed a rather good agreement with the measured data in terms of SIA (Secondary Inorganic Aerosols). Nitrate annual mean agrees quite well (10.4 μgm^{-3} modelled, 9.1 μgm^{-3} measured) with a better estimation in winter (13.2 μgm^{-3} modelled, 17.1 μgm^{-3} measured) than in summer when CAMx slightly overestimates (7.6 μgm^{-3} modelled, 4 μgm^{-3} measured). CAMx overestimates the sulphate annual mean (6.4 μgm^{-3} modelled, 3.9 μgm^{-3} observed). Such an overestimation is due to the winter values (10 μgm^{-3} modelled vs 4.9 μgm^{-3} measured), while in summer the model is quite skilful (2.9 μgm^{-3} modelled vs 3.5 μgm^{-3} measured). Ammonium is also well simulated (5.4 μgm^{-3} modelled annual mean vs 2.8 μgm^{-3} measured) with a general overestimation both in summer and winter. Also Elemental Carbon agrees quite well with the annual mean observed value (1.7 μgm^{-3} modelled, 1.4 μgm^{-3} measured) without significant differences during winter and summer; conversely the model underestimates Organic Carbon concentrations and maybe a fraction of primary PM (in this simulation, particulate resuspension phenomena are not included). Consequently, the analysis of chemical composition seems to suggest that the PM10 underestimation, put in evidence by CAMx, is mainly related to the organic fraction and secondly to some processes related to the primary fraction. On the opposite, SIA seem well reproduced. It is worth noting that the apparently well modelled PM10 concentrations over the Southern part of Lombardy could be in fact related to an overestimation of the secondary inorganic fraction.

Table 1. PM10, O_3 and O_x daily concentration values: statistics of CAMx performances.

Station	Observed PM10 annual mean (μgm^{-3})	Modelled PM10 annual mean (μgm^{-3})	Corr.	NMB (%)	MFB (%)	MFE (%)	Modelled O_3 annual mean of the daily max. (ppb)	Corr.	NMB (%)	Modelled O_x annual mean (ppb)	Corr.	NMB (%)
Bergamo	48	20	0.55	-57	-79	82	49	0.84	40			
Brescia	50	22	0.59	-55	-79	81	48	0.83	34	50	0.51	18
Crema	39	38	0.75	-3	-18	44	46	0.90	4	45	0.79	9
Limito	46	36	0.68	-23	-32	47	46	0.91	25	49	0.69	13
Saronno	47	25	0.68	-47	-67	69	50	0.84	8	50	0.77	9
Pizzighettone	43	38	0.63	-13	-30	53	46					
Milano Juvara	51	35	0.71	-31	-42	53	45	0.88	44	50	0.69	0
Magenta	49	34	0.68	-30	-45	57	46	0.80	18	47	0.34	-1
Mantova	47	32	0.68	-32	-54	63	47	0.92	3	43	0.65	3
Busto Arsizio	43	24	0.62	-43	-55	63	49	0.86	12	49	0.69	12
Erba	45	15	0.32	-67	-103	104	54	0.81	38	51	0.68	25
Cantù	42	19	0.50	-53	-76	81	53					
Osio Sotto	46	30	0.63	-32	-41	54	45					
Piacenza	32	32	0.61	6	5	58	47	0.87	23	43	0.73	13
Mean Values	**45**	**29**	**0.62**	**-34**	**-51**	**65**	**48**	**0.86**	**23**	**48**	**0.65**	**10**

For these reasons the source apportionment analysis has been focused on the Milan receptor, in relation to primary PM and SIA components.

5. SOURCE APPORTIONMENT ANALYSIS

CAMx model results have been analysed by means of the Particulate Source Apportionment Technique, in order to discriminate the contribution at the Milan receptor of several key emission sectors such as transport, heating, agriculture and power plants, also distinguishing the most emitting area of Milan from the surroundings. PSAT algorithm has been applied to primary PM, both organic and inorganic, and secondary inorganic aerosol fraction. SOA have not been included because of the clear lack in their reconstruction, above highlighted.

Obtained results (Tab.s 2-3) underline that road transport is the most relevant sector contributing more than 25% to the PM10 yearly mean in Milan, followed by agriculture and domestic heating. The analysis has also highlighted that several areas contribute to the yearly mean concentration in Milan: more than 70% of the PM10 is due to emissions outside of Milan, thus testifying that particulate matter is a basin scale problem. It is worth noting that a relevant contribution comes from the Eastern boundary because of the presence in the Po valley of prevailing winds blowing from east to west, driven by its orographical features. The importance of the Eastern boundary puts in evidence that particulate matter concentrations are influenced also by large scale transport phenomena. During the summer season (not shown), the fraction due the boundary conditions is higher then winter; it could be related to a slight increase of the windy conditions during the summer months, but also to a reduction of the emissions of primary PM (e.g. domestic heating) as well of the efficiency of the processes that give rise to secondary PM (e.g. nitrate and ammonium) inside the domain.

The same analysis carried out for the exceedance days only, i.e. for daily PM10 concentration exceeding 50 μgm^{-3}, shows that the percentage contribution of the Eastern boundary decreases. Indeed, critical episodes usually take place during weak circulation conditions that favour accumulation processes of pollutants produced locally and, at the same time, damp the influence of the long range transport. It is also worth noting that, during the exceedance days, the most relevant contribution does not come from the Milan city, but it is generated by the emissions placed north of the urban area.

The contribution of the PM emitted over the Milan area is 34% of the total annual mean primary particulate concentration in Milan (not shown), and this contribution rises to 57% if all the urbanised area around Milan is taken into account. As for the secondary fraction, on the contrary, the city area accounts only for 11% of the total secondary PM concentration in Milan; in this case, the highest contributions to the secondary fraction (56%) are yielded by the areas belonging to the Lombardy region, where the considerable emissions of particulate matter precursors can be transformed and transported very efficiently towards the Milan receptor.

Table 2. Contribution of each emission area and group to the PM2.5 yearly mean concentration (μgm^{-3}).

	P. Plants	Dom. Heat.	Industry	Road Tr.	Agric.	Other	BC_East	BC_North	BC_South	BC_West	Area
Milan	0.00	1.37	0.49	2.64	0.34	0.69					5.53
Crit. Zone	0.03	1.36	0.75	2.33	0.41	0.53					5.41
North Lomb.	0.66	1.36	1.58	2.58	1.57	0.66					8.41
South Lomb.	0.74	0.20	0.30	0.63	1.40	0.31					3.58
West P.V.	0.13	0.31	0.29	0.59	0.56	0.43					2.31
East P.V.	0.09	0.06	0.06	0.18	0.12	0.11					0.62
Alps	0.00	0.04	0.02	0.11	0.07	0.06					0.30
B.C.							2.63	0.68	0.94	0.87	5.13
Sector	1.64	4.70	3.50	9.07	4.46	2.78	2.63	0.68	0.94	0.87	**31.29**

Table 3. Contribution of each emission area and group to the PM2.5 concentration (μgm^{-3}) computed during the exceedance days.

	P. Plants	Dom. Heat.	Industry	Road Tr.	Agric.	Other	BC_East	BC_North	BC_South	BC_West	Area
Milan	0.01	3.66	1.21	4.96	0.75	1.13					11.70
Crit. Zone	0.07	3.68	2.02	5.23	1.06	1.04					13.09
North Lomb.	2.07	3.47	4.10	5.66	3.84	1.35					20.49
South Lomb.	2.13	0.48	0.69	1.23	3.09	0.53					8.15
West P.V.	0.35	1.03	0.85	1.77	1.78	1.27					7.05
East P.V.	0.22	0.13	0.13	0.34	0.27	0.20					1.29
Alps	0.00	0.10	0.05	0.26	0.18	0.13					0.72
B.C.							4.81	0.82	1.42	1.77	8.81
Sector	4.84	12.54	9.06	19.44	10.97	5.65	4.81	0.82	1.42	1.77	**71.30**

6. CONCLUSIONS

The CAMx chemical and transport model has been applied over a 300x300 km^2 domain centred on the city of Milan with the purpose of studying PM concentration levels by mean of the PSAT tool. The obtained results show a general underestimation of the modelled PM, mainly due to the lack of SOA simulated contribution, but a rather good agreement with the SIA fraction. Source apportionment analysis demonstrate that road transport is the most relevant sector contributing to the PM10 yearly mean in Milan, followed by agriculture and domestic heating. 70% of the PM in the city is due to emissions outside of Milan, and the fraction due to the emissions of the city is mainly primary particulate. Secondary inorganic fraction is related mainly to the emissions of the Lombardy region as a whole, and it reveals that particulate matter in the city is a basin scale problem.

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REFERENCES

Angelino, E., M. Bedogni, C. Carnevale, G. Finzi, E. Minguzzi, E. Peroni, C. Pertot, G. Pirovano and M. Volta, 2007: PM10 Chemical Model Simulations Over Northern Italy in the Framework of the CityDelta Exercise. *Environmental Modeling and Assessment*, DOI 10.1007/s10666-007-9104-8.

ARPA Lombardy and Lombardy Region, 2006: INEMAR, Inventario Emissioni in Atmosfera: emissioni in regione Lombardia nell'anno 2003. Public data, http://www.ambiente.regione.lombardia.it/inemar/inemarhome.htm.

Bedogni, M., V. Gabusi, G. Finzi, E. Minguzzi and G. Pirovano, 2005: Sensitivity analysis of ozone long term simulations to grid resolution. *International Journal Environment and Pollution*, **24**, No. 1/2/3/4, 51-63.

Bedogni, M., S. Casadei, G. Pirovano, G. Sghirlanzoni and A. Zanoni, 2008: Application of Advanced Particulate Matter Source Apportionment Techniques in the Northern Italy Basin. Air Pollution Modeling and its Application XIX, Springer, *in press*.

Chang, J.S., R.A. Brost, I.S.A. Isaksen, S. Madronich, P. Middleton, W.R. Stockwell and C.J Walcek, 1987: A Three-dimensional Eulerian Acid Deposition Model: Physical Concepts and Formulation. *J. Geophys. Res.*, **92**, 14681-14700.

Cuvelier, C., P. Thunis, R. Vautard, M. Amman et al., 2007: CityDelta: A model intercomparison study to explore the impact of emission reductions in European cities in 2010. *Atmospheric Environment*, **41**, 189-207.

EEA, 2007: Air pollution in Europe 1990-2004. *EEA report*, **2**-2007.

ENVIRON Corporation, 2006: User's guide to the Comprehensive Air quality Model with extension (CAMx) version 4.42. *Technical report*.

Gery, M., G. Whitten and J. Killus, 1989: A photochemical mechanism for urban and regional scale computer modelling. *Journal of Geophysical Research*, **94**, 12925–12956.

Lonati, G., M. Giugliano, P. Butelli, L. Romele, and R. Tardivo, 2005: Major chemical components of PM2.5 in Milan (Italy). *Atmospheric Environment*, **39**, 1925–1934.

Lonati, G., S. Ozgen and M. Giugliano, 2007: Primary and secondary carbonaceous species in PM2.5 samples in Milan (Italy). *Atmospheric Environment*, **41**, 4599–4610.

Minguzzi, E., M. Bedogni, C. Carnevale and G. Pirovano, 2005: Sensitivity of long-term CTM simulations to meteorological input. *International Journal Environment and Pollution*, **24**, No. 1/2/3/4, 36-50.

Nenes, A., C. Pilinis and S.N. Pandis, 1998: ISORROPIA: A New Thermodynamic Model for Multiphase Multicomponent Inorganic Aerosols. *Aquatic Geochemistry*, **4**, 123-152.

Strader, R., F. Lurmann and S.N. Pandis, 1999: Evaluation of secondary organic aerosol formation in winter. *Atmospheric Environment*, **33**, 4849-4863.

Thunis, P., L. Rouil, C. Cuvelier, R. Stern, B. Bessagnet et al., 2007: Analysis of model responses to emission-reduction scenarios within the CityDelta project. *Atmospheric Environment*, **41**, 208-220.

Vautard, R., P.H.J. Builtjes, P. Thunis, C. Cuvelier et al., 2007: Evaluation and intercomparison of Ozone and PM10 simulations by several chemistry transport models over four European cities within the CityDelta project. *Atmospheric Environment*, **41**, 173-188.

EMEP WebDab, 2006: EMEP Activity data and emission database. Public data, http://webdab.emep.int.

Yarwood, G., R. Morris, and G. Wilson, 2004: Particulate Matter Source Apportionment Technology (PSAT) in the CAMx Photochemical Model. NATO International Technical Meeting, Banff, Canada (October 2004).