

## PM10 LONG-TERM ASSESSMENT OF EMISSION REDUCTION SCENARIOS OVER NORTHERN ITALY

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### INTRODUCTION

Air quality assessment and policies analysis show an increasing interest in long term simulations with Chemical Transport Models (CTMs). In this frame the CityDelta open model inter-comparison exercise (<http://rea.ei.jrc.it/netshare/thunis/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) Project (6<sup>th</sup> Framework Programme). The aim of CityDelta exercise was to compare the results of different photochemical dispersion models in order to estimate air quality response to local and global emissions variations. The exercise was carried out by twenty scientific groups working on eight European domains. Starting in 2002, the second phase of CityDelta (2003-2004) was focused on estimations of PM<sub>10</sub> and PM<sub>2.5</sub> concentrations fields, with a special attention devoted to the partitioning of their main inorganic components (nitrates, sulphates and ammonium) and to the response to emission reduction scenarios.

### DOMAIN AND METHODOLOGY

The computational domain (300 x 300 km<sup>2</sup>), centred on Milan (North of Italy), includes a large flat area (all the central and most of the western Po Valley) surrounded by mountains along three sides. 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 modelling system employed for the simulations was composed by a common set of pre-processors for input data (emissions, boundary conditions and meteorological fields) and two different numerical CTM models. To perform the simulations, the domain has been horizontally divided into 5x5 km<sup>2</sup> cells and vertically in 11 varying level ranging from 20 to 3900 m. In order to keep the results homogeneous as much as possible, the models shared the same input fields, derived from the same data provided by JRC to all participants. More details concerning the computational domain and the modelling system are reported on Angelino, E. *et al.* (2005).

The chemical and transport models used for the simulations were CAMx (*ENVIRON Corp.*, 2004) and TCAM (*Decanini, E. and M. Volta*, 2003). The Comprehensive Air quality Model with extensions (CAMx) provides the option of using two different chemical mechanisms: SAPRC99 (*Carter, W.P.L.*, 2000) and CBIV (*Gery, M. et al.*, 1989) 1999 version, modified to model ozone and fine/coarse PM, using RADM (*Chang, J.S. et al.*, 1987) mechanism for aqueous phase chemistry, ISORROPIA (*Nenes, A. et al.*, 1998) for inorganic sulphate-nitrate-ammonium chemistry, SOAP (*Strader, R. et al.*, 1999) semi-volatile scheme for secondary organic aerosols. CAMx user can choose the gas-phase chemistry solver as either IEH (Implicit-Explicit Hybrid) (*Sun, P. et al.*, 1994) or CMC (*ENVIRON Corp.*, 2004), based on an "adaptive-hybrid" approach. TCAM is a multi-phase Eulerian 3D model. It implements

different chemical mechanisms based both on lumped molecule (SAPRC90, SAPRC97, COCOH97) and on lumped structure (CB-IV) approach. The gas phase chemistry is solved by the IEH algorithm, which considers separately slow and fast reacting species. The TCAM model includes and harmonizes an aerosol module describing aerosols by means of a fixed-moving approach. The aerosol module is coupled with COCOH-97 and includes the most relevant aerosol processes: condensation and evaporation, the nucleation of H<sub>2</sub>SO<sub>4</sub> and the aqueous oxidation of SO<sub>2</sub> (Seinfeld, J.H. and S.M. Pandis, 1997).

## EMISSION SCENARIOS

Emissions dataset for the base case (1999) were developed by JRC employing two different inventories: a local one, covering the Lombardia region with a 5x5 km<sup>2</sup> resolution, and EMEP emission data following a resolution of 50x50 km<sup>2</sup>. The inventories include yearly emissions of SO<sub>2</sub>, NO<sub>x</sub>, VOC, CH<sub>4</sub>, CO, NH<sub>3</sub>, PM<sub>10</sub>. Profiles for temporal apportionment, speciation and diameter class distribution were provided by EMEP for all modelling groups. Within the frame of CityDelta exercise seven future scenarios have been simulated by the modelling groups, concerning different emission (and in a coherent way boundary conditions too) reduction patterns related to year 2010. The reductions were computed by IIASA as combinations of two main scenarios: the CLE (Current LEgislation) and the MFR (Maximum Feasible Reduction). This paper shows the results obtained from the two CTMs in relation to the base case, the CLE and the MFR scenarios (*tab. 1*).

Table 1. Total emissions (ton/year) and reduction percentages of the emissions in scenarios.

	Base	CLE		MFR		
	Emission	Emission	% vs. Base	Emission	% vs. CLE	% vs. Base
<b>NO<sub>x</sub></b>	485,556	339,194	-30%	248,827	-27%	-49%
<b>SO<sub>x</sub></b>	725,537	163,132	-78%	67,515	-59%	-91%
<b>VOC</b>	733,811	447,558	-39%	293,823	-34%	-60%
<b>PM<sub>10</sub></b>	92,312	55,683	-40%	31,309	-44%	-66%
<b>NH<sub>3</sub></b>	188,870	190,176	0%	122,486	-36%	-35%

## PM<sub>10</sub> RESULTS AND SCENARIOS IMPACT OVER SECONDARY PM

The model results have been analysed not only in relation to the computational domain but also to a smaller area where the majority of the population dwells. This sub-domain covers approximately an area located in the Po Valley between Varese and Parma cities (*fig. 1*).

Over both the domain and the sub-domain, PM<sub>10</sub> yearly means show some differences on absolute values and distribution in the base case and the CLE scenario (*tab. 2*), with TCAM producing almost everywhere higher average concentrations; however the two models agree within reasonable limits on the relative efficiency of the CLE future scenario (*central column of fig. 1, tab. 2*). When MFR scenario is simulated, instead, CAMx results show a greater reduction both with respect to the base case and the CLE scenario than TCAM (*right column of fig. 1, tab. 2*) does. The implementation of the future scenarios seems to lead to a strong reduction of the areas exceeding the target value established by European and Italian law (40 [µg/m<sup>3</sup>]), even if some areas with concentration above the proposed 2010 limit value (20 [µg/m<sup>3</sup>]) still appear to remain. Inside the sub-domain, as expected, mean concentrations are higher and the effect of emission reduction is stronger in term of absolute values. On the contrary, on relative terms, responses are very similar and for this reason SIA analysis has been performed only over the whole domain.

Table 2.  $PM_{10}$  simulated concentrations ( $\mu\text{g}/\text{m}^3$ ), yearly statistics over the entire computational domain and the sub-domain (between parentheses).

CAMx						
Parameter	Base	CLE	MFR	CLE - Base	MFR-Base	MFR-CLE
mean	19.9 (31.4)	14.4 (22.6)	8.9 (14.4)	-28% (-28%)	-55% (-54%)	-38% (-36%)
95th percentile	41.1	28.7	19.1	-30%	-53%	-33%
sigma	11.6	8.2	5.5	n/a	n/a	n/a
TCAM						
Parameter	Base	CLE	MFR	CLE - Base	MFR-Base	MFR-CLE
mean	29.2 (45.4)	21.9 (32.5)	17.8 (26.9)	-25% (-28%)	-39% (-41%)	-19% (-17%)
95th percentile	62.4	43.1	36.3	-31%	-42%	-16%
sigma	17.0	11.4	9.7	n/a	n/a	n/a

Table 3. SIA simulated concentrations ( $\mu\text{g}/\text{m}^3$ ), yearly statistics over the entire domain.

CAMx						
Specie	Base	CLE	MFR	CLE - Base	MFR-Base	MFR-CLE
Ammonium	3.1	2.6	1.6	-18%	-49%	-37%
Nitrates	5.7	5.5	3.7	-4%	-35%	-32%
Sulphates	4.8	2.7	1.4	-45%	-70%	-45%
Total	13.7	10.7	6.8	-22%	-50%	-37%
TCAM						
Specie	Base	CLE	MFR	CLE - Base	MFR-Base	MFR-CLE
Ammonium	8.1	6.3	3.6	-22%	-55%	-43%
Nitrates	8.0	6.3	5.5	-22%	-31%	-12%
Sulphates	4.3	1.9	1.3	-56%	-70%	-32%
Total	20.4	14.5	10.4	-29%	-49%	-28%

Table 4. Simulated speciation over the computational domain for all scenarios for SIA and other species.

Specie	Base		CLE		MFR	
	CAMx	TCAM	CAMx	TCAM	CAMx	TCAM
Ammonium	16%	28%	18%	29%	18%	20%
Nitrates	29%	27%	38%	29%	42%	31%
Sulphates	24%	15%	18%	9%	16%	7%
Other species	31%	30%	26%	34%	24%	42%

With respect to the Secondary Inorganic Aerosols (or SIA: sulphates, nitrates and ammonium), the differences between the two models widen (fig 2. tabs. 3 and 4). Both models agree very well on the relevance and the trends for sulphates which are clearly correlated with analogous reductions in  $\text{SO}_x$  emissions. The same can't be said for nitrates and ammonium, which show differences both in concentrations and trends in future scenarios. With regard to CAMx is worth noting that in CLE scenario nitrates concentration decreases of only 4%, even if  $\text{NO}_x$  emission reduction is about 30%. As  $\text{NH}_3$  emissions are unchanged in CLE scenario, ammonium availability remains more or less the same, thus inducing an increase in aerosol nitrate in order to compensate sulphate reduction. Differently in MFR scenario, when also  $\text{NH}_3$  decreases, nitrates reduction is noticeable. The complex relationship among SIA components is highlighted also by the total reduction of ammonium. Indeed both models forecast a decrease of about 50% of  $\text{NH}_4^+$  concentration, descending from a reduction of both emissions (36 %) and nitrates and sulphates concentrations.

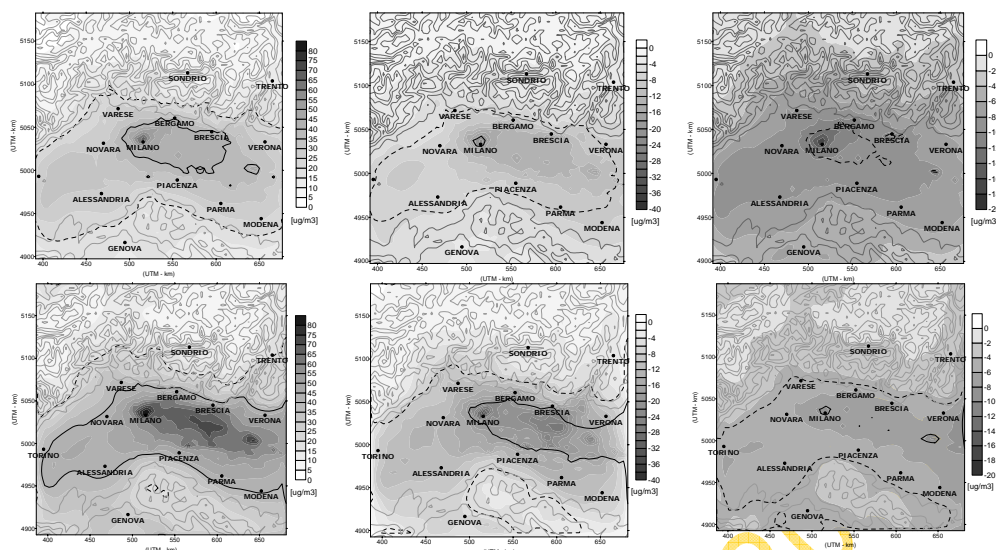


Fig 1. Left column: base case  $PM_{10}$  yearly means (CAMx on upper row; TCAM on bottom); central column: differences of  $PM_{10}$  yearly means between CLE scenario and base case; right column: differences of  $PM_{10}$  yearly means between MFR and CLE scenarios. The thick isoline shows the  $40 \mu\text{g}/\text{m}^3$  target value, the dotted line the  $20 \mu\text{g}/\text{m}^3$  limit value.

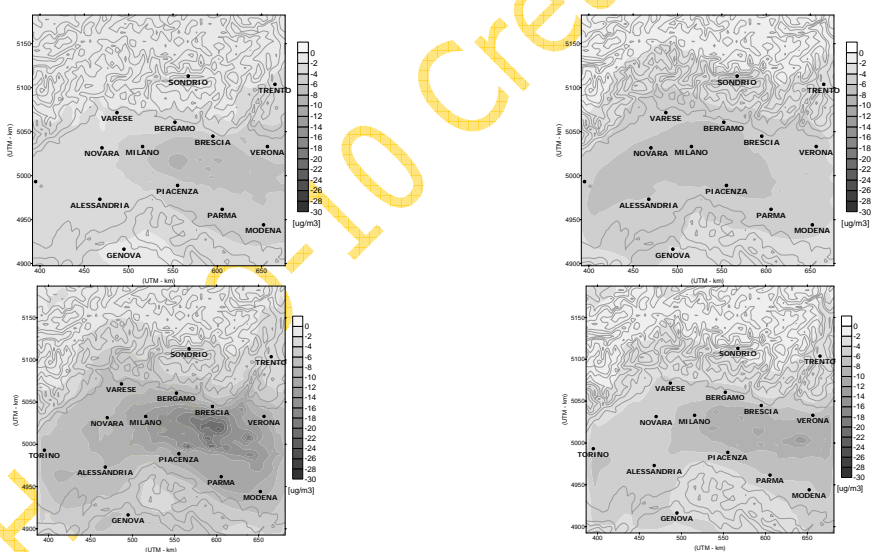


Fig 2. CAMx (upper left) and TCAM (bottom left) differences of SIA yearly means in secondary  $PM_{10}$  ( $\mu\text{g}/\text{m}^3$ ) between base case and CLE scenario. CAMx (upper right) and TCAM (bottom right) differences of SIA yearly means in secondary  $PM_{10}$  between CLE and MFR scenarios.

The figure 2 highlights the different response of the two models in relation to the MFR reproduced SIA: CAMx shows higher reductions in the west part of the domain, while TCAM simulates a deeper scenario impact in the east part. This could be explained by the different model sensitivity to the ammonia emissions, not reduced in the CLE scenario (see also in tab. 4 the deep decrease of the ammonium fraction simulated by TCAM in the MFR). Finally, in the base case the mass percentage of the SIA sum on total  $PM_{10}$  mass is quite similar for both models (69% CAMx vs. 70% TCAM), while in both scenarios this ratio diverges, increasing

with CAMx and decreasing with TCAM. This is due to a difference arising in concentration of *other species* (Primary PM and Secondary Organic Aerosol) and is probably linked to the different SOA mechanisms.

## CONCLUSIONS

The applied CTMs show good agreement both on the base case vs. CLE scenario average PM<sub>10</sub> trend and on total SIA mass over total PM<sub>10</sub> mass in the base case. Some differences are yet present in concentration levels and SIA composition (notably, ammonium and nitrates values, whereas there's very good agreement for sulphates share and trends). The differences between the two CTMs are emphasized in the MFR scenario, with the remarkable exception of sulphates. This may be a hint that, while aerosol sulphur chemistry is well established and shared, the two chemical mechanisms behave differently when nitrogen and other species (mostly carbon) are taken into account, especially when a strong perturbation is introduced in the system by means of relevant reductions of emissions and ratio between pollutants.

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## REFERENCES

- Angelino, E., M. Bedogni, C. Carnevale, G. Finzi, E. Minguzzi, E. Peroni, C. Pertot, G. Pirovano and M. Volta, 2005: PM<sub>10</sub> chemical model simulations over the Milan area in the frame of CityDelta exercise. *Proceedings of the 5<sup>th</sup> International Conference on Urban Air Quality*, Valencia, Spain.
- Carter, W.P.L., 2000: Documentation of the SAPRC-99 chemical mechanism for VOC reactivity assessment. *Report to California Air Resources Board*, Riverside CA, USA.
- 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**, 14,681-14,700.
- Decanini, E. and M. Volta, 2003: Application to Northern Italy of a new modelling system for air quality planning: a comparison between different chemical mechanisms. *International Journal of Environment and Pollution*, **20**, 85-95.
- ENVIRON Corporation, 2004: User's guide to the Comprehensive Air quality Model with extension (CAMx) ver 4.0. *Technical report*, Novato, USA., <http://www.camx.com>
- 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.
- 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. *Atmos. Environ.*, **33**, 4849-4863.
- Seinfeld, J.H. and S.M., Pandis, 1997. *Atmospheric chemistry and physics*. John Wiley & Sons.
- Sun, P., D.P. Chock and S.L. Winkler, 1994. An Implicit-Explicit Hybrid Solver for a System of Stiff Kinetic Equations. *Proc. 87th Air & Waste Management Association Annual Meeting*.