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

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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.

Keywords: 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. CAMs 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 5×5 km² 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).

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 OX annual mean (i.e. O3 + NO2) is considered instead of O3 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 μg m⁻³ modelled, 9.1 μg m⁻³ measured) with a better estimation in winter (13.2 μg m⁻³ modelled, 17.1 μg m⁻³ measured) than in summer when CAMx slightly overestimates (7.6 μg m⁻³ modelled, 6 μg m⁻³ measured). CAMx overestimates the sulphate annual mean (6.4 μg m⁻³ modelled, 3.9 μg m⁻³ observed). Such an overestimation is due to the winter values (10 μg m⁻³ modelled vs 4.9 μg m⁻³ measured), while in summer the model is quite skillful (2.9 μg m⁻³ modelled vs 3.5 μg m⁻³ measured). Ammonium is also well simulated (5.4 μg m⁻³ modelled annual mean vs 2.8 μg m⁻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 μg m⁻³ modelled, 1.4 μg m⁻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, O3 and Ox daily concentration values: statistics of CAMx performances.

<table>
<thead>
<tr>
<th>Station</th>
<th>Observed PM10 annual mean (μg m⁻³)</th>
<th>Modelled PM10 annual mean (μg m⁻³)</th>
<th>Corr.</th>
<th>NMB (%)</th>
<th>MFB (%)</th>
<th>MFE (%)</th>
<th>Modelled O3 annual mean of the daily max. (ppb)</th>
<th>Corr.</th>
<th>NMB (%)</th>
<th>Modell ed OX annual mean (ppb)</th>
<th>Corr.</th>
<th>NMB (%)</th>
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<td>0.55</td>
<td>-57</td>
<td>-79</td>
<td>82</td>
<td>49</td>
<td>0.84</td>
<td>40</td>
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<tr>
<td>Brescia</td>
<td>50</td>
<td>22</td>
<td>0.59</td>
<td>-55</td>
<td>-79</td>
<td>81</td>
<td>48</td>
<td>0.83</td>
<td>34</td>
<td>50</td>
<td>0.51</td>
<td>18</td>
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<td>-18</td>
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<td>46</td>
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<td>4</td>
<td>45</td>
<td>0.79</td>
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<td>-32</td>
<td>47</td>
<td>46</td>
<td>0.91</td>
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<td>0.63</td>
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<td>-30</td>
<td>53</td>
<td>46</td>
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<td><strong>-51</strong></td>
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<td><strong>0.86</strong></td>
<td><strong>23</strong></td>
<td><strong>48</strong></td>
<td><strong>0.65</strong></td>
<td><strong>10</strong></td>
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</tbody>
</table>

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 from the Milan receptor area, with a significant contribution from the city of Milan and its surrounding areas.
outside of Milan, thus testifying that particulate matter is a basin scale problem. It is worth noting that a relevant
collection 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 µg m\(^{-3}\),
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>
<thead>
<tr>
<th>P. Plants</th>
<th>Dom. Heat.</th>
<th>Industry</th>
<th>Road Tr.</th>
<th>Agric.</th>
<th>Other</th>
<th>BC_East</th>
<th>BC_North</th>
<th>BC_South</th>
<th>BC_West</th>
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<td>2.64</td>
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<td>1.58</td>
<td>2.58</td>
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<td>0.66</td>
<td>8.41</td>
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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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