## AIR QUALITY INFLUENCE OF AMMONIA AND NITROGEN OXIDES EMISSIONS REDUCTION OVER THE PO VALLEY

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Abstract: Ten different combinations of ammonia and nitrogen oxides emission reduction scenarios have been investigated. This analysis has been performed through the application of the chemical transport model (CTM) FARM on two monthly periods characterised by significant measured levels of ammonia and particulate matter, respectively during Spring and Autumn 2011. The simulation domain covers the whole Po Valley (hereafter P-V) and the Alpine Region, with a horizontal resolution of 4 km and 16 terrain-following vertical levels, irregularly spaced from the ground to 10000 m above surface level. The CTM was driven by meteorological input fields produced by the prognostic non-hydrostatic meteorological model RAMS; initial and boundary conditions (IC/BCs) have been derived from FARM national scale simulation at 12 km horizontal resolution. The base case emissions have been prepared merging high resolution bottom-up emission inventories developed by all the P-V administrative Regions applying the same methodology. Local inventories have been combined with the Italian national inventory and EMEP emission data for the portion of surrounding countries inside the simulation domain. Results obtained by the base case simulation (i.e. considering the actual emissions) have been compared with observed levels of PM<sub>2.5</sub>, PM<sub>10</sub>, ammonia, nitrogen oxides and ozone at different monitoring stations located in P-V. The amount and combination of emissions reduction needed to effectively reduce secondary PM levels has been estimated from the analysis of the ten different scenarios performed for the two investigated periods. The role of nitrogen oxides and ammonia on particle mass and composition is confirmed, claiming for the implementation of proper emission control strategies at regional level.

Key words: Air quality assessment and management, emission control strategies, AQ models.

# INTRODUCTION

P-V is one of the air pollution hot spots of major concern in Europe where the EC air quality standards are presently not attained. It is characterised by peculiar topographic features, being surrounded on three sides by the Alps/Apennines chain, and by one of the most densely populated area in Europe, with a global population of about 20 million clustered in different urban areas. Due to its anthropization level and to the presence of intense agricultural and breeding activities, this area is characterised by very high emissions and concentrations of ammonia, whose role as aerosol precursor together with NO<sub>X</sub> and SO<sub>2</sub> is well known. Since PM composition analysis showed that a large fraction of aerosol has secondary origin, and SO<sub>2</sub> emissions have significantly decreased during the last decades, leading to year-round low concentrations recordings, it is of major interest to understand the potential PM concentration reduction reachable through NO<sub>X</sub> and ammonia emission limitation policies. According to the Lombardia regional emission inventory (www.inemar.eu), 96% of NH<sub>3</sub> year emissions in the region comes from agriculture activities while the remaining portion (about 4%) is given, particularly in urban areas, by road traffic, from vehicles equipped with three-way catalytic converters.

## ATMOSPHERIC MODELLING SYSTEM

The atmospheric modelling system (AMS) used to simulate the chemical and physical processes involving the pollutants in the atmosphere is based on Flexible Air quality Regional Model (FARM, Silibello *et al.*, 2012). This model has been applied with the SAPRC-99 gas-phase chemical mechanism (Carter, 2000) and the AERO3 modal aerosol scheme implemented in CMAQ framework (Binkowski and Roselle, 2003). The AMS includes modules to reconstruct atmospheric flows and related turbulence parameters and to apportion data from the emission inventories to grid cells.

### **BASE CASE SCENARIO**

Meteorological input fields were produced with the prognostic, non-hydrostatic meteorological model RAMS (Cotton *et al.*, 2003) run in a 2-way nested grids configuration. The outer grid covers large part of Central Europe and the Mediterranean Sea (with a horizontal grid spacing of 16 km  $\times$  16 km) while the

inner grid dovetails with the target domain (Figure 1) that includes 146x96 cells with a spatial resolution of 4 km x 4 km and 16 terrain-following vertical levels irregularly spaced from the ground to 10000 m above surface level. Initial and boundary conditions have been based on ECMWF operational analyses with a horizontal space resolution of 0.25 degrees and a time frequency of 6 hours.

The meteorological fields together with land cover information (e.g. roughness length) and chemical species characteristics (gas reactivity), have been then used by the interface module SURFPro (Finardi et al., 2008) to produce dry deposition velocities for the modelled gas-phase species and turbulent diffusivity fields needed by FARM. This module also includes the Model of Emissions of Gases and Aerosols from Nature (MEGAN, Guenther et al., 2006) to estimate emissions from vegetation and algorithms to estimate natural seasalt and soil emissions driven by the surface wind (Zhang et al., 2005; Vautard et al., 2005). Boundary conditions for all modelled species on the target domain have been derived from the corresponding threedimensional fields obtained by the QualeAria<sup>1</sup> modelling system that simulates air pollution over the Italian peninsula. The emissions coming from major industrial facilities and the diffuse sources over the considered domain were obtained by merging data coming from regional and national inventories and from the EMEP Centre on Emission Inventories and Projections (CEIP) [http://www.ceip.at] for surrounding countries. Such input data was provided to the AMS and simulations were performed on two one-month periods during 2011, lasting respectively from March 17th to April 19<sup>th</sup> and from September 16<sup>th</sup> to October 16<sup>th</sup>. In the following Figure 2 an example of the performance of the AMS at an urban background station located in Milano urban area is given. The comparison between



Figure 1. Nested grids used by RAMS. The inner grid correspond to the target domain (see bottom map, the area in blue represents Lombardia region).

observed and predicted  $PM_{2.5}$ ,  $NH_3$ ,  $O_3$  and  $NO_2$  concentrations evidences the capability of the AMS to capture the seasonal variations shown by the observations. Underestimation of observed  $PM_{2.5}$  and  $NO_2$  concentrations is possibly due to uncertainties in the emissions and to the coarse horizontal resolution that does not permit to resolve local scale phenomena.

### **EMISSION SCENARIOS**

The ten studied scenarios (see Table 1) have been selected to study the influence on PM2.5 levels of possible ammonia and nitrogen oxides emission reduction strategies over Lombardia Region (blue area in Figure 1) or over the whole domain. Figure 3 shows the average variations [%] of  $PM_{2.5}$ ,  $PM_{2.5}$  components and OH radical concentrations obtained by the different scenarios with respect to the base case over Lombardia region. The variations have been computed according to the following formula

$$\Delta C^{x} = \frac{1}{N} \sum_{ij \in L} \left( \frac{C_{ij}^{x} - C_{ij}^{0}}{C_{ij}^{0}} \right)$$
(1)

where  $\Delta C^x$  is the percentage variation of species *C* due to scenario *x* with respect to Base Case (Scenario *0*); *N* represents the number of grid cells within Lombardia region (*L*);  $C_{ij}^0$  and  $C_{ij}^x$  are concentrations of the grid cell *i,j* within *L* computed respectively for Base Case and Scenario *x*.

<sup>&</sup>lt;sup>1</sup> http://www.aria-net.eu/QualeAria/index\_en.html



Figure 2. Comparison between observed and predicted  $PM_{2.5}$ ,  $NH_3$  and  $NO_2$  concentrations at an urban background station located in Milano urban area (Pascal-Città Study site).

Table 1. Emission scenarios (L	L means Lombardia Region, D	means whole domain, see inner	grid in Figure 1	)
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#	Scenario	#	Scenario
1	25% NH <sub>3</sub> reduction over D	6	50.0% NO <sub>x</sub> reduction over L
2	25% NH <sub>3</sub> reduction over L	7	25% NH3 and NO <sub>x</sub> reduction over L
3	50% NH <sub>3</sub> reduction over L	8	25.0% NH3 and NOx reduction over D
4	12.5% NO <sub>x</sub> reduction over L	9	25% NH3 and 50% NO <sub>x</sub> reduction over L
5	25% NO <sub>x</sub> reduction over L	10	50.0% NH3 and 25.0 NO <sub>x</sub> reduction over L



Figure 3. Emission scenarios analysis over Lombardia region: average variations with respect to Base Case scenario [%] of  $PM_{2.5}$ ,  $PM_{2.5}$  components ( $NO_3^-$ ,  $NH_4^+$ ,  $SO_4^-$  and ORG) and OH radical concentrations.

As for  $PM_{2.5}$ , the analysis of Figure 3 shows that:

- equal rate reductions of NH<sub>3</sub> (Scenarios 2,3) and NO<sub>x</sub> emissions (Scenarios 5,6) determine similar decrease of PM<sub>2.5</sub> concentrations. The NO<sub>x</sub> reduction is more effective during the autumnal period;
- combined reduction of NH<sub>3</sub> and NO<sub>x</sub> emissions (Scenario 7) is as effective as reducing at a higher rate the emissions of single species (Scenarios 3 and 6) in decreasing PM<sub>2.5</sub> concentrations;
- combined reduction of NH<sub>3</sub> and NO<sub>x</sub> emissions (Scenarios 7 and 9) is more effective than reducing the emissions of single species (see Scenarios 2, 3 for ammonia and 4,5 for Nox). As an example a reduction of NH<sub>3</sub> emissions by 25% over Lombardia lead to a an average decrease of PM<sub>2.5</sub> concentrations of 2.5% and 3.2% respectively during Spring and Autumn while the combined reduction of NOx emissions by 25% (Scenario 7) and by 50% (Scenario 9) determines a decrease of PM<sub>2.5</sub> concentrations respectively of 5.1% and 8.5% during Spring and 7.8 and 13% during Autumn;
- the effectiveness of NO<sub>X</sub> emission reduction is lower during Spring, because of higher availability of OH radical that determines the formation of sulfate and organic components compensating and even exceeding nitrate diminutions;
- the emission reduction over the whole domain (Scenarios 1 and 8) is more effective than the same reduction over Lombardia region (Scenarios 2 and 7); the effectiveness is higher when combined reductions are considered.

The analysis of the spatial distribution of pollutant levels corresponding to the different emission scenarios (not shown here) has evidenced non homogeneities due to differences of NH3 and NOx emission ratios in different areas of the investigated domain.

#### CONCLUSIONS

Results obtained by the application of an AMS over P-V, considering ten different emission scenarios characterised by different levels of ammonia and nitrogen oxides reductions, allowed to estimate the more effective strategies to reduce particulate matter levels over Lombardia region. Combined reductions appear to be more effective than higher reductions of individual species. Moreover, local Administrations strategies need to take into account the contribution from the whole P-V basin emissions. This result can help policy makers to set up more realistic emission control strategies to reduce the exposure of P-V inhabitants to particulate matter.

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