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$PM_{2.5}$ WINTERTIME SENSITIVITY TO CHANGES IN NO_X, SO_2 AND NH_3 EMISSIONS IN LOMBARDY REGION

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Abstract: According to the results of the EU LIFE PREPAIR Project on primary emission estimates on Po-basin, the main source of primary PM_{10} is the non-industrial combustion, mainly due to biomass burning in the residential sectors. Moreover, SO_x are emitted by industrial activities and combustion, NO_x for quite a half of the total from the road transport and NH_3 from agriculture. The role of ammonia emissions is widely identified in $PM_{2.5}$ formation while the emission trend of this precursor in the last year is showing a slower slope compared to the emission estimates on NO_x and SO_2.
The aim of this work, that is part of a project required by Lombardy Region focused on the role of agricultural sector on air quality and on atmospheric ammonia emissions, is to evaluate the effects on $PM_{2.5}$ concentrations with a reduction of the emissions of NH_3, NO_x and SO_2 in the Po basin extended area. The simulation is focused on the wintertime period 1st January – 31st March 2019 based on a deeper analysis of NH_3, PM_{10}, $(NH_4)NO_3$, $(NH_4)_2SO_2$ concentrations and the daily ammonia emissions derived from slurry spreading. Based on the bulletins emitted by Regional Authority, research in scientific literature and the crops diffusion on the Lombardy Region, a reconstruction of the annual profile of slurry spreading for different crops has been improved.
The model simulations have been run by FARM-LO operational AQ model of the Environmental Agency of Lombardy Region (ARPA) over two nested domains: one including Lombardy region, the other the Po Valley and Slovenia. The model suite includes a Chemical Transport Model (FARM) and a meteorological model (WRF). Emissions have been derived for the two domains from the regional emission inventory INEMAR and from the dataset on emission inventories developed in the frame of the LIFE IP Project PREPAIR. The initial and boundary conditions are obtained by QualeAria (http://www.qualearia.it). Evaluations carried out interesting results for domain sub-areas with different dependence from NH_3 and NO_x reduction in relation to emission source types and intensities and could support AQ policy makers to better understand the impacts of precursors abatements on $PM_{2.5}$ concentration.

Key words: *Emission reduction, non-linearity, wintertime chemical regimes, PM formation*

INTRODUCTION

Ammonia (NH_3) is the atmospheric pollutants that contributes significantly, together with nitrogen oxides and sulfur, to the secondary inorganic $PM_{2.5}$ formation.
This pollutant reacts in the atmosphere both with nitric acid and sulfuric acid originating ammonium nitrate and ammonium sulfate which are the inorganic ions more present in the particulate matter.
In Itay (ISPRA, 2021) and in Lombardy Region, ammonia is emitted in atmosphere mainly (94%) by the agricultural sector while the SO_X are released by the energy production and industrial sectors (90%) (EDGAR, 2020). For NO_X, emissions are spread out from different sectors (i.e. transport, industry and agriculture). The INEMAR inventory emission (2017) has estimated that 97% of ammonia regional emission are linked both to fertilisers and zootechnical sector. The latter representes the 86% of the overall emission in atmosphere.
The aim of this work is to identify the chemical regimes where secondary inorganic PM is formed over the Po basin, which is a peculiar area where the chemical regimes distributions are the most complex, showing a non-linear processes expecially within NO_x and NH_3 (Clappier et al., 2021; Carnevale, 2020; Bessagnet, 2014). We focus the present analysis on the wintertime chemical regimes as it is the period of the year more affected by the exposure to fine particlaute matter ($PM_{2.5}$). We start describing the modelling set-up, the modelled base case concentrations and detail the indicators/statistics done to perform analysis. Then, we

analyse the sensitivity of $PM_{2.5}$ concentrations to NH_3, NO_X and SO_2 emissions and finally we provide an analysis of the chemical-regimes of $PM_{2.5}$ consequently to these emissions reduction.

MATERIAL AND METHODS

Modelling set-up

The modeling study is performed by the ARIA Regional System developed by AriaNET society and used by Lombardy Environmental Agency (Arianet, 2010). The emission input consists of gridded annual (2017) emissions referred to municipalities and classified by SNAP (Selected Nomenclature for Air Pollution) codes. Meteorological input data are based on forecasts from GFS model (https://www.nco.ncep.noaa.gov/pmb/products/gfs/). The modelling domain covers the entire Po basin with an extension of 836 x 416 km^2 (Figure 1) with a grid resolution of 4 km by 4 km and includes 16 vertical levels. The initial and background concentrations are from Qualearia (http://www.qualearia.it).
The base case simulations cover the entire meteorological year 2019 whereas the scenarios analyses have been focused only to wintertime.
The choice of the winter season has been derived by the analysis of ammonia emissions and their correlation with measured concentrations of PM_{10}, NH_3, $NH_4(NO_3)$ and $NH_4(SO_2)$ in 6 permanent stations.

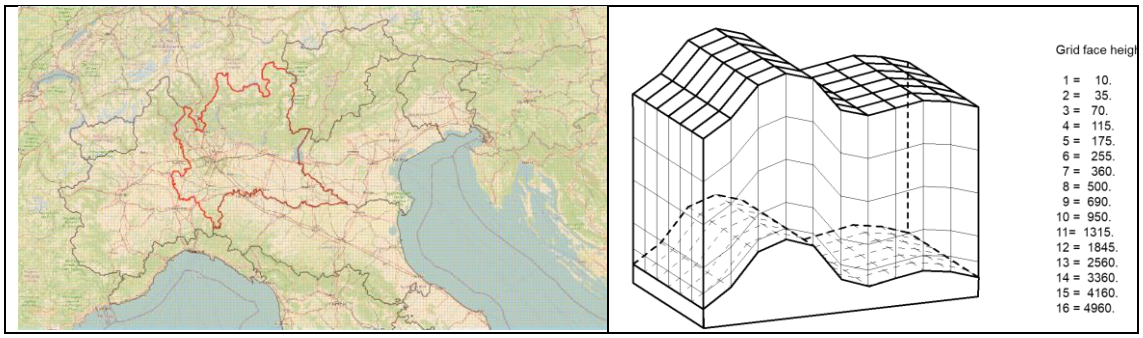

Figure 1. Horizontal and vertical discretization of the ARPA Lombardy modelling system. The grid (x,y: 210,105) has a discretization of 4 km x 4 km

In this work, we simulated a series of 16 scenarios where NO_X, NH_3 and SO_X emissions were reduced indipendently or simulaneously by 10, 25, 50, 75 % from the base case emission reference level.

Table 1. Scenarios reducing 100-α % of the emissions, one for each single precursor, for the reduction of two precursors and for the reduction of all precursors

Scenarios (name)	**NO_X (emissions 100-α)**	**NH_3 (emissions 100-α)**	**SO_X (emissions 100-α)**
Base case	100%	100%	100%
Sc_u_1a	90%	0	X
Sc_u_2a	0%	90%	X
Sc_u_1b	75%	0	X
Sc_u_2b	0	75%	X
Sc_m_1	75%	75%	X
Sc_u_1c	50%	0	X
Sc_u_2c	0	50%	X
Sc_m_2	50%	50%	X
Sc_m_2bis	25%	25%	X
Sc_u_3a	0	0	90%
Sc_u_3b	0	0	75%
Sc_m_3	75%	75%	75%
Sc_u_3c	0	0	50%
Sc_m_4	50%	50%	50%
Sc_u_f1	25%	0	x
Sc_u_f2	0	25%	x

The emission reductions were applied over the entire Po basin domain only for wintertime which covers the period from 1st January to 31st March.
The NH_3 annual emission, estimated by regional inventory INEMAR and originated by the management of nitrogen compound wastewater, have been disaggregated in space and in time to identify daily emissions on the regional territory. The contribution linked to the stabulation and warehousing is considered constant during the entire year, whereas the contribution linked to the practice of the spreading livestock manure is time-dependent following 1) Nitrate Directive 91/676/CE which regulates the distribution over different areas in Lombardy and 2) meteorological conditions (i.e. the distribution is strictly forbidden during the rainy day and over the frozen ground).

Representation of results and indicators

Results are presented in terms of average concentration maps of the wintertime period (1st January – 31st March). Moreover, we aggregate all the background stations located in the Po basin in order to show different behaviours in terms of response to emission changes based on different emission reductions. The impact of NO_X, NH_3 and SO_2 emission reductions on secondary organic PM2.5 has been discussed. To describe the interactions between emissions, we use the relationship proposed by Thunis and Clappier (2014). Potential impacts (P) are defined as the ratio between the concentration change and the emission reduction intensity:

$$P_{NH3}^{\alpha} = \frac{\Delta C_{NH3}^{\alpha}}{\alpha} \qquad P_{NOX}^{\alpha} = \frac{\Delta C_{NOX}^{\alpha}}{\alpha} \qquad P_{SO2}^{\alpha} = \frac{\Delta C_{SO2}^{\alpha}}{\alpha} \qquad (1)$$

Where ΔC_x^{α} is the PM concentration change resulting from a reduction of the NH_3, NO_X and SO_2 emissions respectively; α is the emission reduction intensity that varies from 0 (no reduction and then 100% emission) to 1 (i.e. 0% emission).

RESULTS

Base Case

In Figure 2 the wintertime averaged $PM_{2.5}$ concentration fields show a diffused pollution covering the most of the Po basin, extending from the metropolitan areaof Turin till the Veneto plain. The maximum modelled values reach 53 μg/m^3.This is due to the more stable atmospheric conditions and to the particular orographic setting that let to favour the accumulation of the particulate matter in the area. The spatial field for NO_X reflects generally the emission patterns locations resulting ”richest” in the higher urbanized area (Turin and Milan) and along the main regional and statal streets while NH_3 is more abundant in the agricultural land area between southern of Milan area and northern Emilia Romagna and in the Cuneo area. Finally, high SO_2 concentrations are located nearby big industrial sites located all over the Basin.

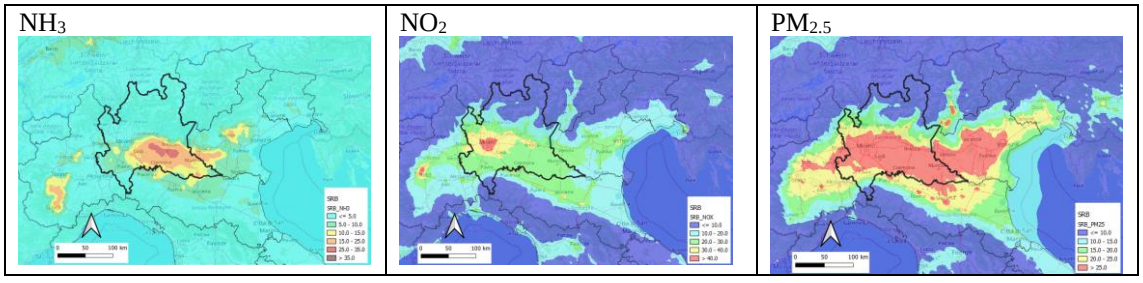

Figure 2. Spatial concentration (μg/m^3) based on the base case of the wintertime period of the NH_3, NO_2 PM precursors line and $PM_{2.5}$ concentrations

Figure 3 shows the boxplots of $PM_{2.5}$ observed values for 2019 at each monitoring station and the predicted concentration. It can be observed that the higher decreasing of precursors the higher decreasing of $PM_{2.5}$ concentrations is. Firstly, it is worth noting that the reduction is more consistent with coupled precursors rather than a singular reduction: this advantage increases more and more in function of emission reduction (but in principle with no linearity).

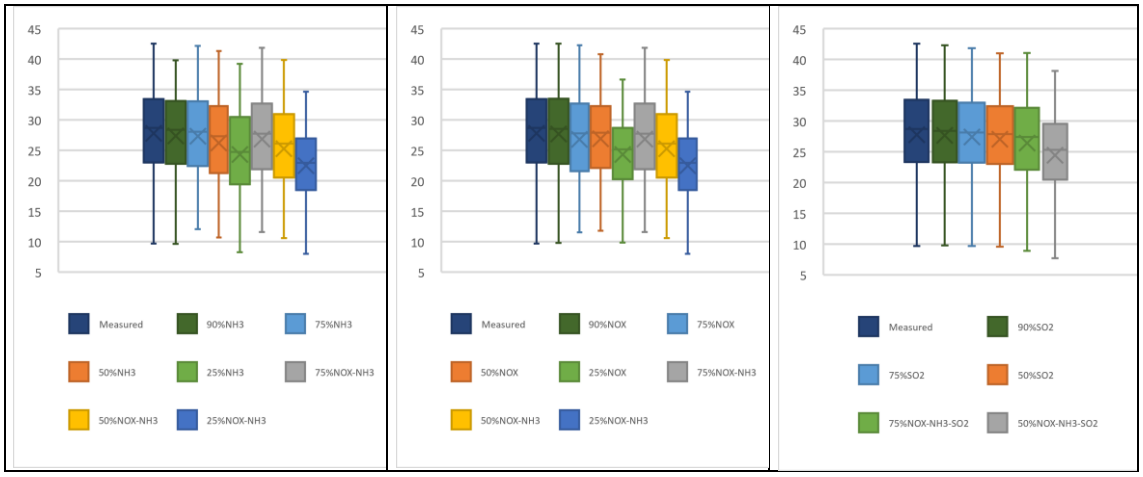

Figure 3. $PM_{2.5}$ wintertime averages concentrations (μg/m^3): boxplots of observed and predicted concentrations at each monitoring point station for all reduction scenarios of NH_3, NO_X and SO_2.

We provided the chemical regime maps for only the NO_X-NH_3 reduction also because during wintertime, the NH_3-sensitive regime area are more important with respect to SO_X (Clappier et al., 2021). By using (1) it is possible to detect the areas where a $PM_{2.5}$ stronger reduction is obtained with an identique emission reduction of precursors. By using a difference between P^{α}_{NH3} and P^{α}_{NO2} a chemical regime can be defined: if the difference will be positive a reduction of NH_3 is more effective whereas on the contrary the zones will be named NO_X- sensitive (negative difference).

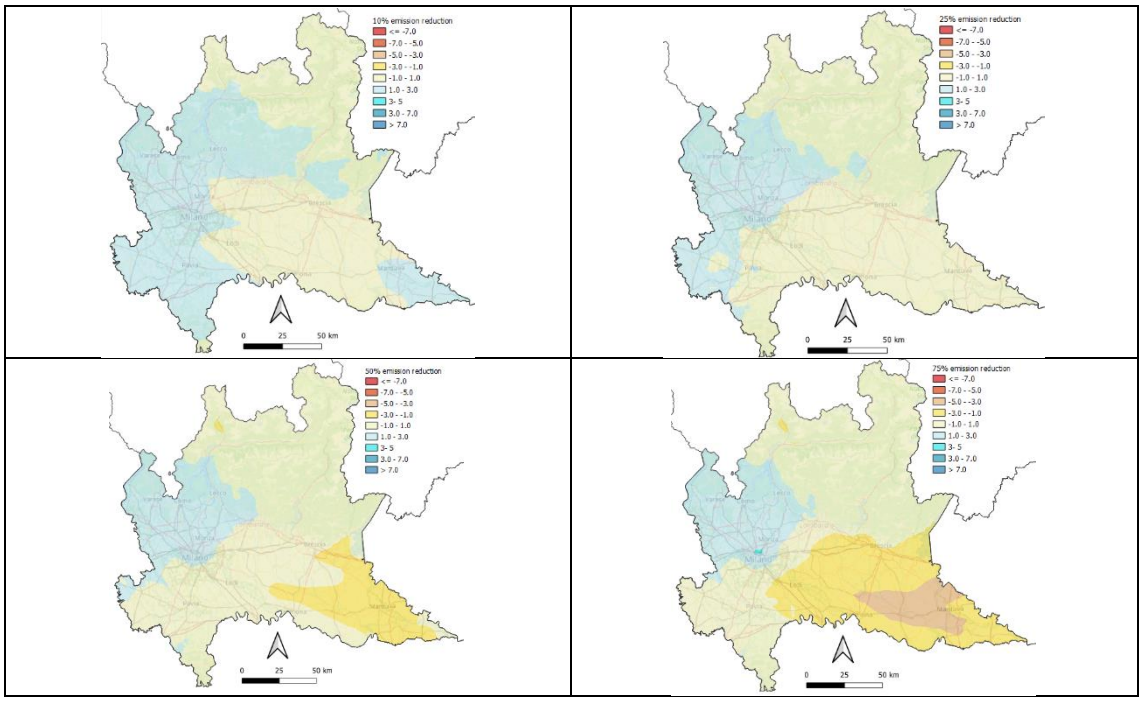

Figure 4. Wintertime chemical-regimes obtained at a reduction level of 10-25-50-75%. The maps represent the $P^{\alpha}_{NH3} - P^{\alpha}_{NO2}$ in μg/m^3 indicator that shows the NO_X- sensitive (from yellow to red) and NH_3- sensitive (from light to dark blue) areas. The light yellow represents areas sensitive to both precursors reduction.

DISCUSSION AND CONCLUSIONS

In this study we analysed the $PM_{2.5}$ sensitivities to NH_3, NO_X and SO_2 emission reductions. It can be observed that the decreasing of precursors favours a diminishing of $PM_{2.5}$ concentrations. Moreover, it is worth noting that the reduction is more consistent with coupled precursors rather than a singular reduction: this advantage increases more and more in function of emission reduction (but in principle with no linearity). It can be assessed that areas NH3-sensitive don't correspond to those where the ammonium release is higher (as presented in Figure 2 where the higher concentrations of NH_3 are in the border of Lombardy and Emilia Region). Furthermore, it can be noticed that the reduction of NH_3 is as important as of NO_X in the $PM_{2.5}$ concentration control. The chemical regimes have shown that for higher reduction of the precursors (i.e from 25% to 75%) there are as many areas NO_X-sensitive as NH_3-sensitive, but it is important to reduce coupling NO_X-NH_3. The latter behaviour is also evident in Baojing Gu et al., 2021.
The study confirms the results obtained with different hypothesis (i.e. different emission inventories, different chemical transport model, different meterological year, different boundary conditions in comparison to previous work of Clappier et al., 2021; Thunis et al., 2021) and leads robustness to the previous results present in literature; the results show that a combined reduction of the NO_X-NH_3 precursors are very large to be effective on PM abatement. An important finding which has an implication on air quality strategies is that in wintertime in urbanized areas the NH_3-sensitive areas are predominant with respect to the NO_X ones. The results obtained in this study will be used in the frame of the EU LIFE PREPAIR in order to compare with different chemical trasport models and to have the possibility to consider uncertainty in the results (for example by using ensemble method with different model).

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