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COMBINED USE OF EULERIAN AND LAGRANGIAN MODELLING FOR LOCAL SCALE SOURCE APPORTIONMENT

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Abstract: Air pollution apportionment studies focused on major industrial sources in Italy were carried out through an integrated approach, combining higher resolution contributions for primary components from a Lagrangian particle dispersion model (SPRAY) and wider-area contributions including secondary components from an Eulerian chemical transport model (FARM). To quantify the proportion of the contribution due to the industrial source of interest, the considered emissions included all relevant sources for the investigated area (vehicular traffic, heating of buildings, ports and airports, other industrial sources, agriculture, etc.). Weather variability and seasonal differences in emissions were taken into account through yearly model runs, leading to results comparable with legislation limits on annual basis. After successful comparison of base-case simulated concentrations against data from monitoring stations, the modelling system was used in source apportionment mode. Results appear to be consistent with observations and sources distributions, with case-by-case peculiarities.

Key words: *Source apportionment, Lagrangian model, Eulerian model*

INTRODUCTION

The presence of an important polluting source in a territory is often referred to by the population as the predominant driver of air pollution levels. Monitoring networks built to observe its impact can fulfill the purpose, but can also give misleading or inconclusive responses, since they measure pollution also from other sources. In these cases it is essential to be able to quantify the proportion of the contribution due to the source of interest with respect to other ones, either local or remote, usually referred as source apportionment. The quantitative evaluation of the relative contribution to ground level concentrations of pollutants due to different emission sources is in fact a key component of environmental impact assessment studies, as well as policy scenarios development.

AN INTEGRATED SOURCE APPORTIONMENT METHODOLOGY

Assessment studies were carried out for the evaluation of the relative contribution to ground level concentrations due to emissions from large industrial sources in Italy, with respect to the rest of the emitting sources in the surrounding territory. To better take into account the complex dynamics of dispersion and transformation of pollutants from multiple sources over local to regional domains, the combined use of a Lagrangian particle dispersion model (SPRAY, Tinarelli *et al.*, 2000) and an Eulerian chemical transport model (FARM, Silibello *et al.*, 2012; Mircea *et al.*, 2014; Bessagnet *et. al.*, 2016) was chosen. In the studies performed for this work, characterised by elevated sources with significant emissions, SPRAY is able to describe the detailed dispersion patterns of the primary pollutants plume and its footprint on the ground even at high temporal resolutions, since the plume is affected explicitly by the three-dimensionality of meteorology and turbulence at any point, and not only at its centre of gravity, as in simpler plume-oriented models. This allows to simulate local scale phenomena such as the vertical separation of portions of the plume in presence of wind direction changes with height (wind shear), breezes, orographic effects and calm winds with stagnation and accumulation phenomena. In addition, it allows to easily and naturally separate the effects of different sources, if necessary. On the other hand, FARM allows to complement the information produced by the Lagrangian model, accounting for the emissions related to all sectors, long-range transport from sources outside the computational domain, and chemical transformations of gases and particles that cause the formation of secondary pollution, a major fraction of the total concentrations of regulated pollutants (e.g. NO_2, O_3, PM10). In fact, a previous similar source apportionment study on a coal power plant in Monfalcone, northeast Italy, in which only SPRAY was used, actually showed that long-range contributions and secondary pollution formation could not be neglected. Base-case simulations were first carried out considering all the emission sources included in the computational domains, in order to compare the results against data from monitoring stations, describing the overall air quality levels over the investigated area. Weather variability and seasonal differences in emissions were taken into account by performing yearly model runs; the resulting sets of results are therefore comparable with limits defined by law on an annual basis. Sectorial source apportionment was then carried out with the so-called *brute force method* (Koo *et al.*, 2009; Burr and Zhang, 2011). A series of FARM simulations was run, separately varying the emissions from each sector (e.g. the plant; other industrial activities; heating; road transport; maritime and air transport; agriculture; and all other sources), and keeping constant all other model inputs. The primary component of pollutants concentrations associated to plant emissions, obtained from additional non-reactive FARM simulations, were then subtracted from the original fields and replaced with those calculated by SPRAY at higher resolution. Each concentration map resulting from the perturbed runs was then compared with the one of the reference run, to obtain a set of variation maps; at the end, the estimated contribution of each set of sources was calculated as the ratio between the variation obtained with the corresponding run and the sum of the variations from all runs.

BASE CASE RESULTS

The outlined source apportionment technique is being applied to two waste-to-energy plants, located in Acerra, 20 km northeast of Naples (CNR-ISAFOM Napoli, 2016), and Corteolona, 35 km southeast of Milan (work in progress). The extent and grid resolution of the simulation domains were chosen in order to allow a realistic representation of the phenomena relevant to the involved models and an accurate estimation of the contribution of both the emissions from local sources (including the plant under study) and from longer distances. SPRAY simulations were run over "local" domains centred on the plant, with 0.25 km grid spacing, 25x25 km^2 size for Acerra, and 0.2 km grid spacing, 22x22 km^2 size for Corteolona. In both studies, FARM simulations were run on two nested "intermediate" and "regional" domains, with 1 km and 4 km grid spacing respectively.

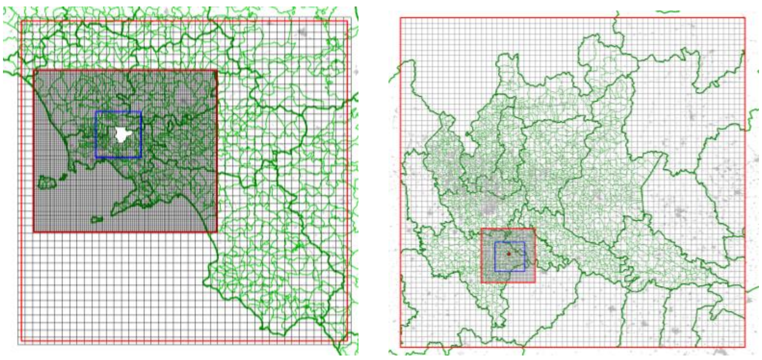


Figure 1. Simulation domains: SPRAY "local" domains in blue, FARM "intermediate" and "regional" domains in red

For each study, detailed plant emission data were taken from continuous emission monitoring systems, while emission data from all other sources were integrated starting from the most detailed inventory available. In the Acerra study, the national ISPRA 2010 emission inventory, available at provincial level, was taken as a starting point for a remarkable work to obtain the most accurate and updated information about the known sources included in the modelling domains: downscaling at municipal level by means of relevant proxy variables, identifying additional local sources, updating existing data with information taken from other public databases (ETS, E-PRTR, Integrated Environmental Authorisation - AIA). In the Corteolona study the starting point for emissions integration was the regional inventory from ARPA Lombardia, at municipal level. The meteorological fields needed to drive the dispersion models were derived from the QualeAria dataset (www.aria-net.it/qualearia), covering the whole Italian territory at 12 km resolution. The dataset contains 3D (wind, temperature and humidity) and 2D (precipitation and cloud cover) meteorological fields with hourly temporal resolution. Using topography and land use data at higher resolution, a downscaling of the fields to the final resolution was then performed through interpolation and objective analysis applying the SWIFT mass-consistent diagnostic model, allowing for realistic channelling of the flow due to orography and local land use. Comparison with ground observations at Capodichino and Grazzanise airports for Acerra, and at Sant’Angelo Lodigiano and Pavia for Corteolona, showed that the model chain was able to reproduce the general characteristics of the local wind, dominated by breeze phenomena in Acerra and by synoptic circulation in Corteolona.

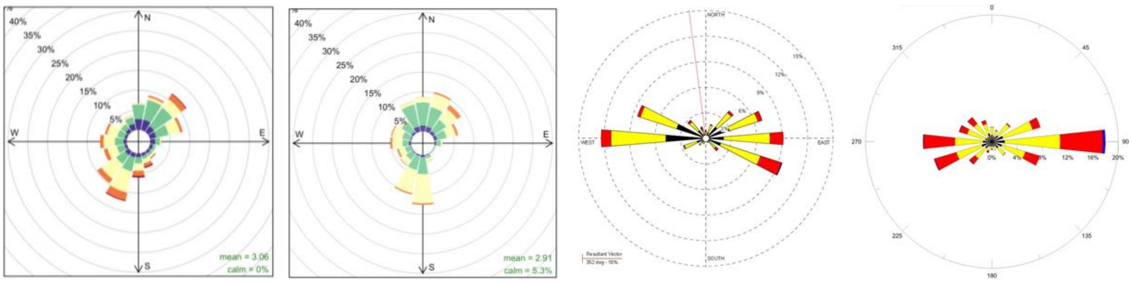


Figure 2. Wind roses at Naples Capodichino airport (left) and Sant'Angelo Lodigiano (right); for each location, modelled data are on the left, observed data on the right

The estimation of 2D and 3D scale parameters used to describe atmospheric turbulence, needed by both SPRAY and FARM to compute the contribution to concentration gradients due to dispersion in addition to advection along the main wind flow, was performed using the SurfPro pre-processor, taking into account the horizontal non-homogeneities induced by the presence of different land use characteristics. SurfPro is also able to estimate dry deposition rates for each chemical species considered by FARM. The influence of the sources outside the computational domains was taken into account through a set of time-varying boundary conditions for all the pollutants considered, derived from national scale air quality simulations of the QualeAria system, also based on FARM model. The FARM runs in reactive mode were carried out using the SAPRC99 chemical scheme, which considers 121 gas phase reactions and the AERO3 module for particulate matter transformations, thus allowing the formation of secondary compounds to be modelled.

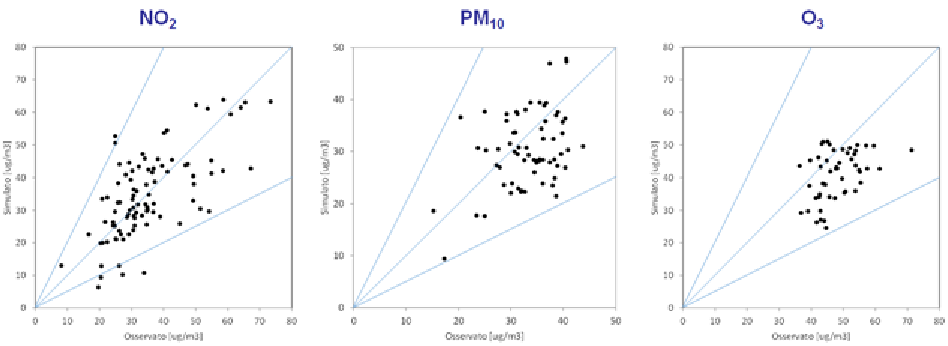


Figure 3. Annual average concentration values from the 4 km grid step simulation in the Corteolona study, compared to values measured in 2010 by ARPA Lombardia regional monitoring network

To validate FARM simulations against data from monitoring stations and compare the resulting indicators with limits defined by law on an annual basis, yearly base-case runs (June 2013 to May 2014 for Acerra, the year 2010 for Corteolona) were first carried out considering all the emission sources included in the computational domains, describing the overall air quality levels over the area of interest, taking into account weather variability and seasonal differences in emissions. The obtained performance for average concentration levels showed good agreement with the available data (examples in Figures 3-4), except for some underestimations in the Acerra study, probably due to emission sources or activities not registered in the national inventory, such as uncontrolled combustion of various kinds in the open air. To investigate this hypothesis, an additional FARM simulation was performed on the "regional" domain, adding daily

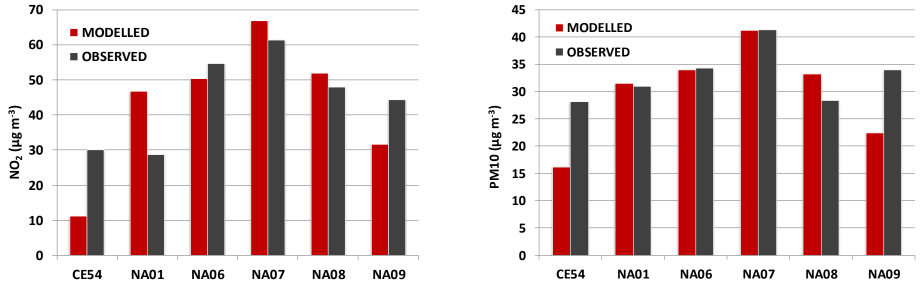


Figure 4. Annual average concentration values from the 1 km grid step simulation in the Acerra study, compared to values measured in 2013-2014 by ARPA Campania regional monitoring network

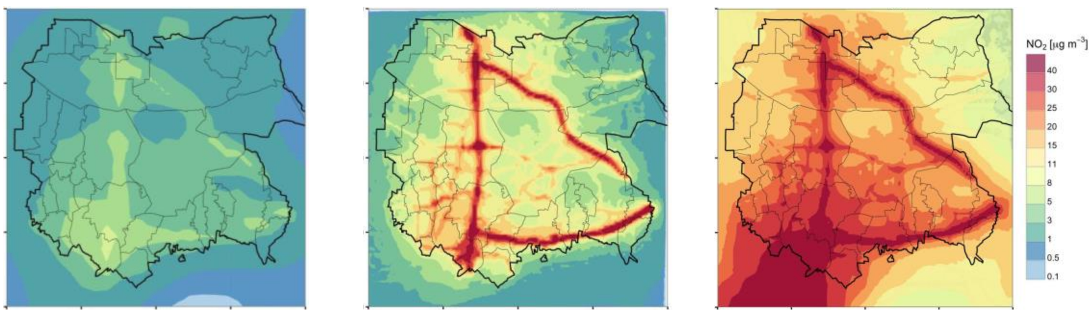
emission data from the "Fire inventory from NCAR" (FINN). From the obtained results it is reasonable to deduce that the contribution of open fires to PM concentrations could be significant.

SOURCE APPORTIONMENT RESULTS

Using the described *brute force method*, the FARM source apportionment runs were performed at 1 km resolution ("intermediate" domains). Results produced by FARM and SPRAY over the "local" domains were then combined to obtain a picture as complete and detailed as possible of the contributions of the different sectors – in terms of sources, spatial detail and pollutant species – with the following approach:

- the primary component coming from plant emissions (and also, in the Acerra study, from other relevant sources within the "local" domain: industries, heating and road traffic) was taken from SPRAY runs;
- the secondary component coming from "local" sources and the contribution from the remaining sources (primary and secondary component) was instead taken from FARM runs.

To separate the primary and secondary parts of the pollutant coming from "local" sources, two sets of FARM runs were performed, one with full chemistry, and one considering only the dispersion of the primary component of the emitted species. The primary part obtained from FARM was then substituted with the corresponding one at higher resolution from SPRAY. NO_2 concentrations from SPRAY runs were estimated from NOx applying the $NO_2 \cdot (NO+NO_2)^{-1}$ ratios derived from the corresponding reactive FARM run. An example of the resulting annual average concentration maps is shown in Figure 4.



NO_X NO_X NO_2

Figure 5. Overall annual average concentration values for nitrogen oxides on the "local" domain in the Acerra study. From left to right: NO_X from local sources modelled with FARM at 1 km resolution; NO_X from local sources modelled with SPRAY at 250 m resolution; NO_2 from all sources as sum of the contributions of both models.

The third map highlights the benefit arising from the combination of the two models, clearly showing both the footprint of the most relevant local sources (related to the more realistic treatment of line sources at higher spatial resolution offered by the Lagrangian model) and the contribution of sources outside the "local" domain, particularly in the south-western part where the metropolitan area of Naples is located. Considering the individual sector maps (Figure 6), in the area around Acerra traffic emissions were found to represent the greatest pressure factor; the port of Naples, heating emissions, and some industries follow, while the contribution due to the incinerator is definitely very small. Similar results were obtained in the Corteolona study, although the predominance of traffic is less pronounced and agriculture also gives a non negiglible contribution.

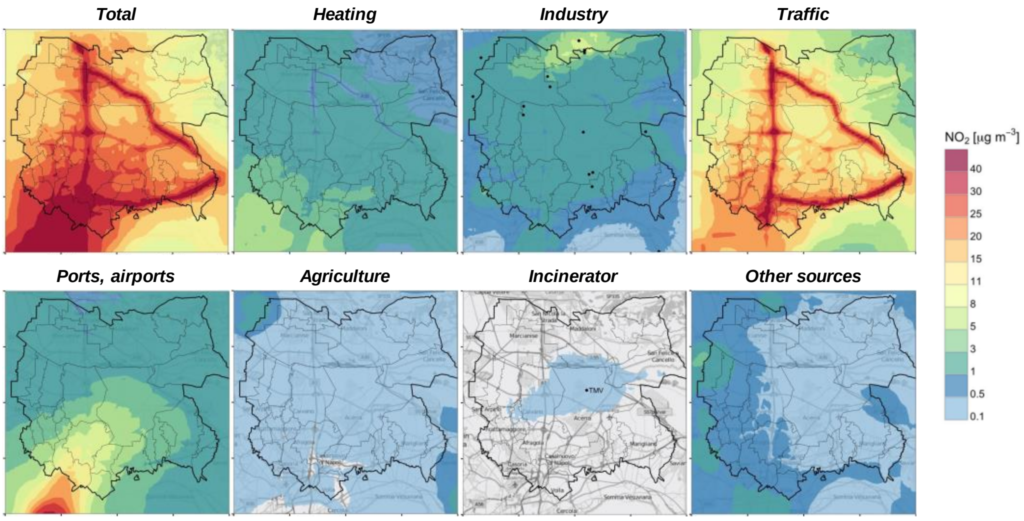


Figure 6. Acerra: average concentration due to all sources (top left) and contributions estimated for different sources.

CONCLUSIONS

Maps of annual average concentrations of pollutants showed the benefit that comes from the combined use of a Lagrangian model (SPRAY) and an Eulerian model (FARM) in source apportionment studies. Both the footprint of the most important local sources and the influence of sources outside the high resolution domain were clearly depicted, allowing to quantify their relative and absolute contribution in different parts of the area of interest, highlighting critical spots but also suggesting gaps in knowledge concerning some emission sources that have not been identified in the inventory of currently available emissions (e.g. uncontrolled open fires). This approach also allows, in principle, a comparative assessment of future scenarios that can be defined through a thorough and exhaustive selection of emission sources, and gives more detailed information to epidemiological studies investigating the possible impacts of each source set on the health of the population in the surrounding areas.

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