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From emissions to source allocation: Synergies and trade-offs between top-down and bottom-up information

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ABSTRACT

This study investigates the dispersion of atmospheric pollutants over a coastal region of north-western Italy by means of modelling techniques. A series of annual air quality model simulations corresponding to different emission reduction scenarios has been performed with a three-dimensional chemical transport modelling chain running at 3 km resolution. The emission reduction scenarios were used to develop bottom-up (locally produced) source-receptor relationships to perform a local source allocation analysis of the main atmospheric pollutants in a few polluted cities within the domain of interest. Results were compared with default top-down (EU-wide) source-receptor relationships, at roughly 7 km resolution. The results show the benefit of using the two sources of information in an integrated way. The analysis of the impacts of local emission reductions on concentrations and of the source allocation results reveals that nitrogen oxides concentrations are mostly affected by local emission sources, especially road transport and harbour related activities while the contribution of non-local sources is important for particulate matter (especially from industry and agriculture sources). For PM, larger scale modelling approaches (top-down) are necessary. Ideally, both a bottom-up approach for the characterisation of the local emission sources and a top-down larger scale approach to capture the impact of non-local sources would be necessary to perform an accurate source allocation, and provide support to the design of local air quality plans.

1. Introduction

In recent years, the study of the dispersion of air pollutants became of primary relevance to understanding the processes affecting air quality and their impact on human health and on the environment (Wakefield et al., 2001; Hu et al., 2015; Xue et al., 2018; Haines and Ebi, 2019). Within this framework, the extension of air quality monitoring networks (Harkat et al., 2018) as well as the adoption of innovative low cost (Kumar et al., 2015; Popoola et al., 2018) and portable sensors (Sun et al., 2016; Mueller et al., 2017; Shindler, 2019) become key tools for studying urban air quality. However, such sensors have intrinsic limitations, e.g. measurement uncertainty, that do not fully allow for the capturing the spatial and temporal variability exhibited by air pollutants. Satellite data can provide complementary information from this perspective, as they can provide spatial variability even if with lower temporal resolution and precision (Nicolantonio et al., 2007).Together with measurements, Eulerian Chemical Transport Models (CTM) are widely used to assess atmospheric physics, as they fully describe the transport, diffusion and chemical transformation processes involved in the formation of air pollutants (Pernigotti et al., 2013; Mailler et al., 2017; Ciarelli et al., 2019; Manders et al., 2017). On top of this, models represent the only option available to investigate potential scenarios (e.g. impact of a given emission reduction on concentration). However, a key component in modelling activities is related to the preparation of the required input data, consisting of the meteorological forcing, initial and boundary conditions and emissions that need to be estimated for several activity sectors. Since the quality of the modelling results will depend on the quality of this input data, a great deal of effort is devoted to this task. Regarding the emissions, a bottom–up approach (compiled with detailed local information) is generally more accurate than a top-down one

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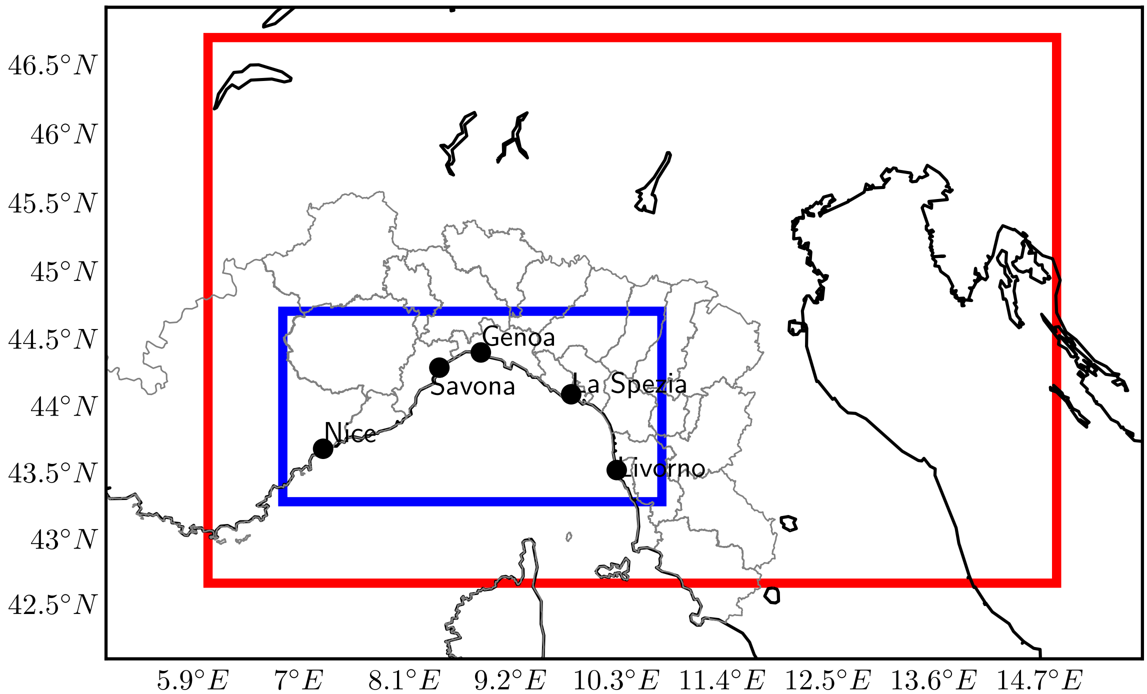


Fig. 1. Computational domains of the NINFA (in red) and LINEA (in blue) modelling systems. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

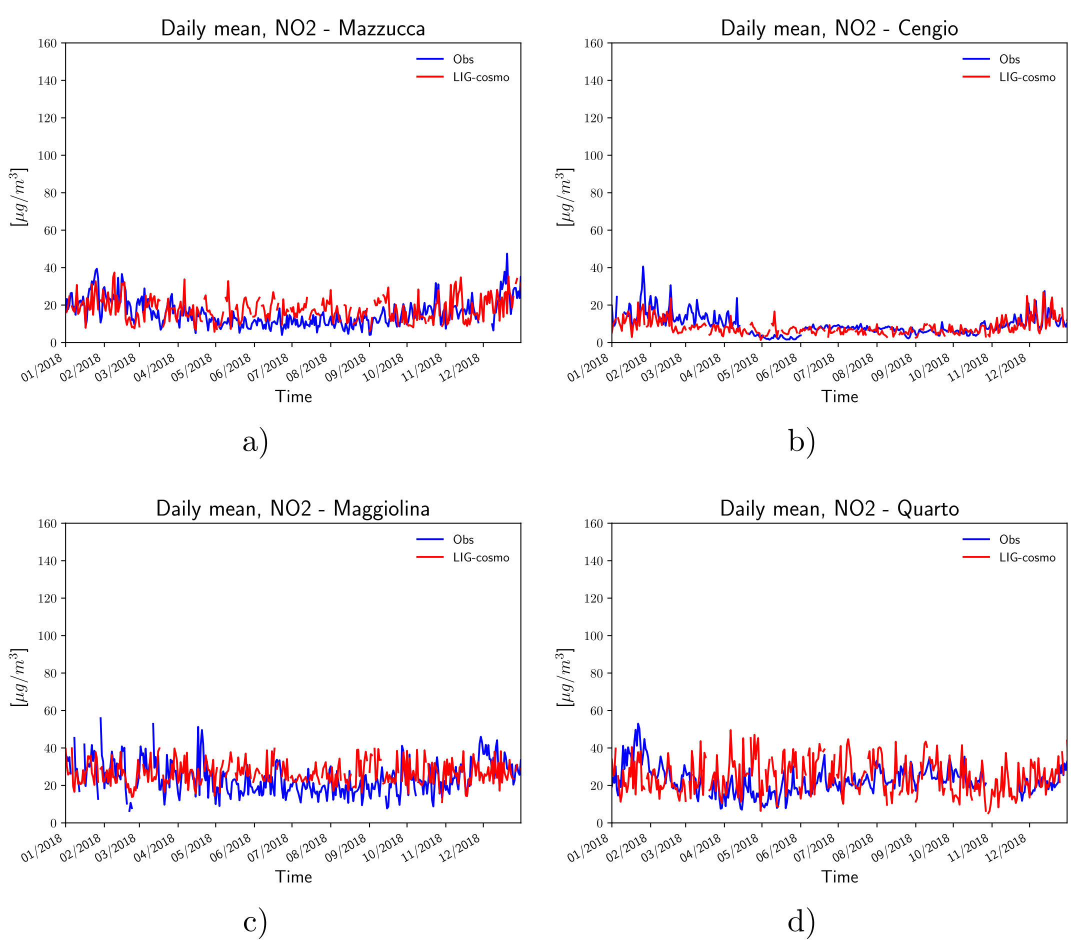


Fig. 2. Time series of NO_2 daily mean at four control stations. In red: simulated data; in blue: observations. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

Table 1
Summary of statistical indicators values for five control stations, classified as industrial (IS), rural (RS) and urban (US) stations.

Station name	Coordinates (Lat Lon)	$HH(NO_2)$	$NBI(NO_2)$	$HH(PM_{10})$	$NBI(PM_{10})$	$HH\ (O_3)$	$NBI(O_3)$
Mazzucca (IS)	44.39°N, 8.29°E	0.13	0.12	–	–	–	–
Cengio (RS)	44.40°N, 8.21°E	0.51	0.26	0.56	0.35	0.41	0.15
Maggiolina (US)	44.12°N, 9.84°E	0.81	0.33	0.62	0.32	0.51	0.10
Quarto (US)	44.40°N, 8.99°E	0.83	0.43	0.47	0.31	0.34	0.07
Varaldo (US)	44.32°N, 8.49°E	–	–	0.75	0.34	0.47	0.08

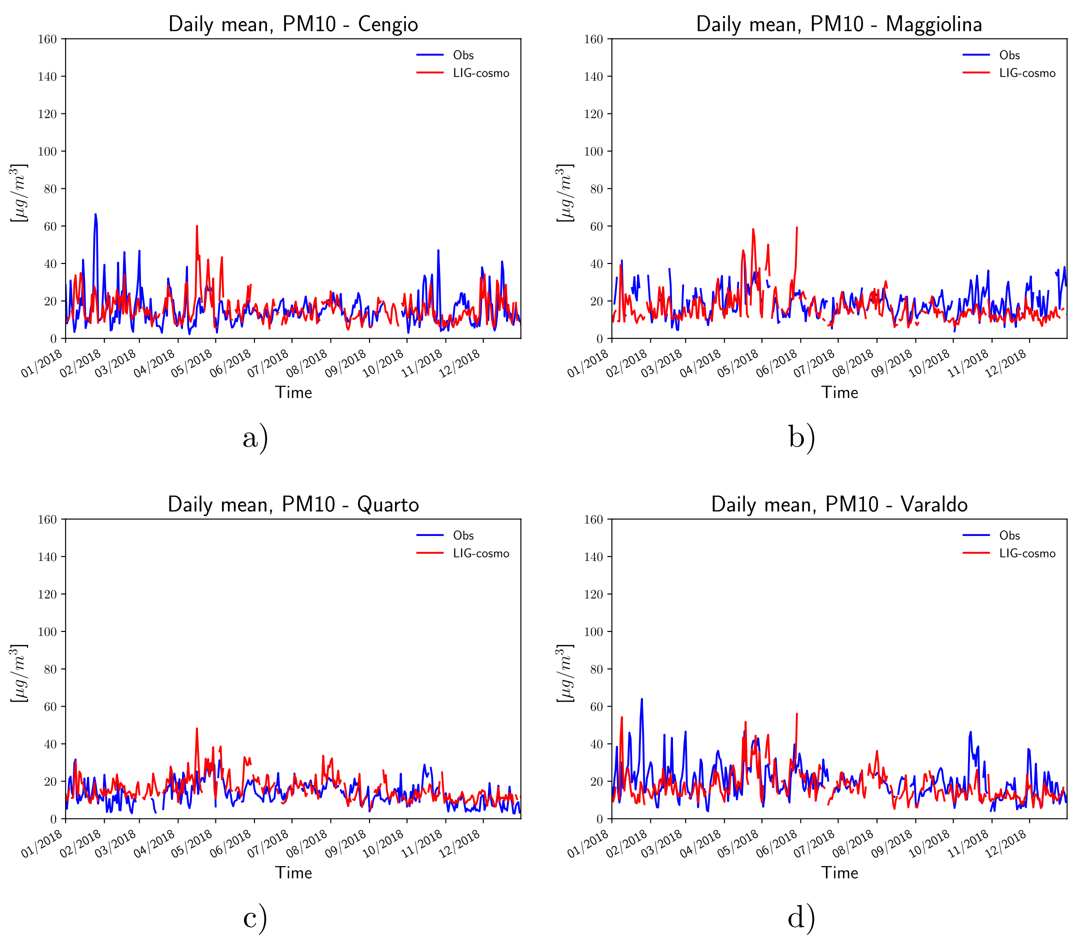


Fig. 3. Time series of PM_{10} daily mean concentrations at four control stations. In red: simulated data; in blue: observations. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

(produced with general EU-wide information) as it relies on local knowledge. But top-down methodologies based on spatial disaggregation techniques (using proxies such as population), down to the municipal level or to the smallest functional units are commonly used due to the lack of detailed data (Miranda et al., 2015). As mentioned above, one of the main skills of the CTM is to provide information on the potential outcome of "what if" scenarios since they are able to evaluate the impacts of emissions changes on concentration levels. But this skill comes at the expense of resources as the computational time required for performing scenarios (in general lengthy!) can easily become prohibitive. To cope with this limitation, simplified approaches based on the development of source-receptors relationships (SRR) (Pisoni et al., 2010; Seibert and Frank, 2004; Vedrenne et al., 2014) are available. These SRR mimic the behaviour of a full CTM model when used to predict the link

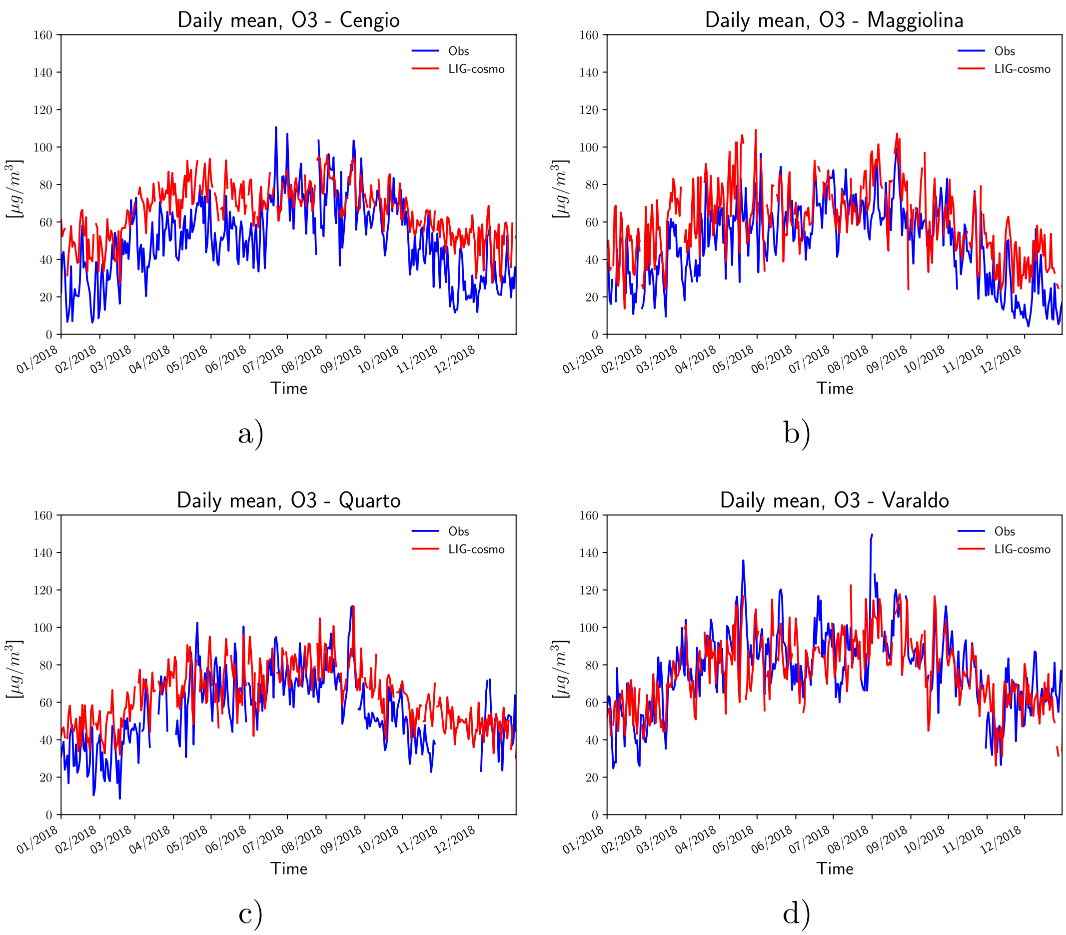


Fig. 4. Time series of O_3daily mean at four control stations. In red: simulated data; in blue: observations. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

between emission and concentration changes. Among these SRR, the SHERPA (Screening for High Emission Reduction Potentials on Air quality) modelling tool was recently developed. SHERPA consists of simple emission-concentration relationships/equations that are derived from a limited set of full CTM emissions scenario. The SRR, after validation, can then be used to evaluate the impact of policy scenarios (Clappier et al., 2015; Thunis et al., 2016; Pisoni et al., 2017). The gain in CPU requirement obtained with SRR, as compared to the full CTM, allows to increase the number of application scenarios. It then becomes feasible to test the impact of many different sources and perform a source allocation study whereby the contribution of different emission categories (e.g. industrial, transport, agricultural sectors) and different spatial scales (e.g. local, urban, metropolitan areas) on air pollution is quantified. The present study aims to improve our understanding of the processes governing air pollution over the region of northwestern Italy. To this aim, a set of CTM simulations is created to develop source-receptor relationships. We first analyse the results of this limited number of CTM scenario simulations and then use the SRR to deliver a source allocation for the main atmospheric pollutants. The strengths and weaknesses of using bottom-up information, rather than top-down, when developing source-receptors models are also addressed.The manuscript is organized as follows: first, a description of the CTM and source-receptor model are provided in Section 2. The results of the CTM scenarios and those of the SRR related source allocation analysis are then discussed in Section 3 for some control locations, by means of both a bottom-up and top-down SRR. Finally, conclusions are given and discussed.

2. Models and data

2.1. CTM model and inventory emissions

LINEA (Ligurian Network to Evaluate Aerosol and photochemical pollution) is the numerical system implemented and managed by ARPA Liguria to forecast the concentration of photochemical pollutants. Its domain covers the entire Ligurian Region at a 3-km horizontal resolution (blue rectangle in Fig. 1). Initial and boundary conditions are retrieved from the modelling chain NINFA (Northern Italy Network to Forecast Aerosol and photochemical pollution) that covers the entire region of Northern Italy at a 5-km resolution (red rectangle in Fig. 1). NINFA itself is nested within the larger scale CHIMERE-based PREV'AIR model running at a 50-km spatial resolution. PREV'AIR is managed by INERIS (Institut National de l'Environnement Industriel et des Risques) and IPSL (Institut Pierre Simon Laplace des Sciences de l'environment).

The CTM (Chemical Transport Model) model used in LINEA is CHIMERE (version 2014b, Menut et al. (2013)). The meteorological forcing is provided by two parallel configurations (respectively at 5 and 2.2 km resolution) of the modelling chain system COSMO (Consortium for Small ScaleModeling, Steppeler et al. (2003) and Baldauf et al. (2011)). Since the computational domain extends over different regions and nations, the emission inventory (reference year: 2016) is compiled on the basis of three different input data, as detailed below:

- The regional inventory E2GOV for emissions located within the Liguria region. This contains both the natural and anthropogenic sources for the main pollutants, greenhouse gases and metals;

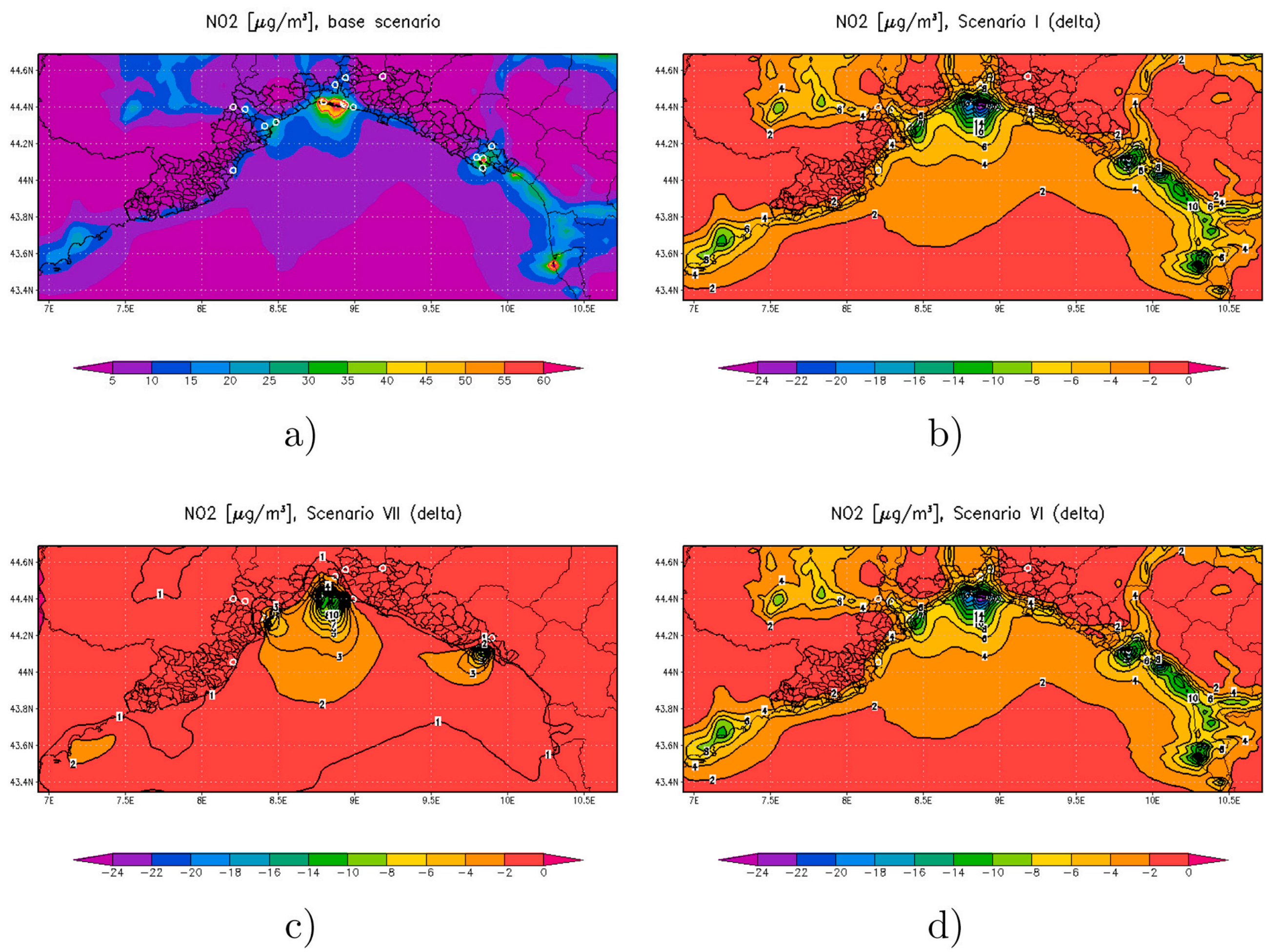


Fig. 5. Maps of annual mean and delta NO_2 concentrations (base case and scenarios).

- The national inventory ISPRA for the main natural and anthropogenic emissions originating from all Italian regions (other than Liguria);
  The European inventory MACC (Kuenen et al. 2014) for the emissions within the neighbouring region PACA (Provence-Alpes-Cote--d'Azur)Emissions are then spatially disaggregated by means of proxy surrogate variables in order to reproduce an accurate spatial variability over the domain.

The evaluation of the modelling results was performed by direct comparison with observed data from the stations of the Liguria air quality regional network,to be representative of the modelling domain covered by LINEA system. The agreement is good for the NO_2 daily mean for all industrial and rural stations (Fig. 2a) and 2b)).

Model performance are evaluated by means of statistical indicators; for the sake of brevity only the Normalized Bias *NBI* and the symmetrically normalized root mean square error *HH* are used. These indicators have been introduced by Hanna and Heinold (1986) and are defined as:

- $NBI = \sum(S_i - O_i)/\sum O_i$, where S_i and O_i are respectively simulations and observations. This indicator provides insight on the average error level (the closer this indicator is to zero, the better the simulation is);
- $HH = \sqrt{\sum(S_i - O_i)^2/\sum S_i O_i}$, provides insight on the average and scatter components of the unbiased error.

Values are reported in Table 1, together with the names and coordinates of each control station.

For NO_2, the worst performances occur at urban stations (Fig. 2c) and d)), with a tendency to overestimate daily mean values, even though

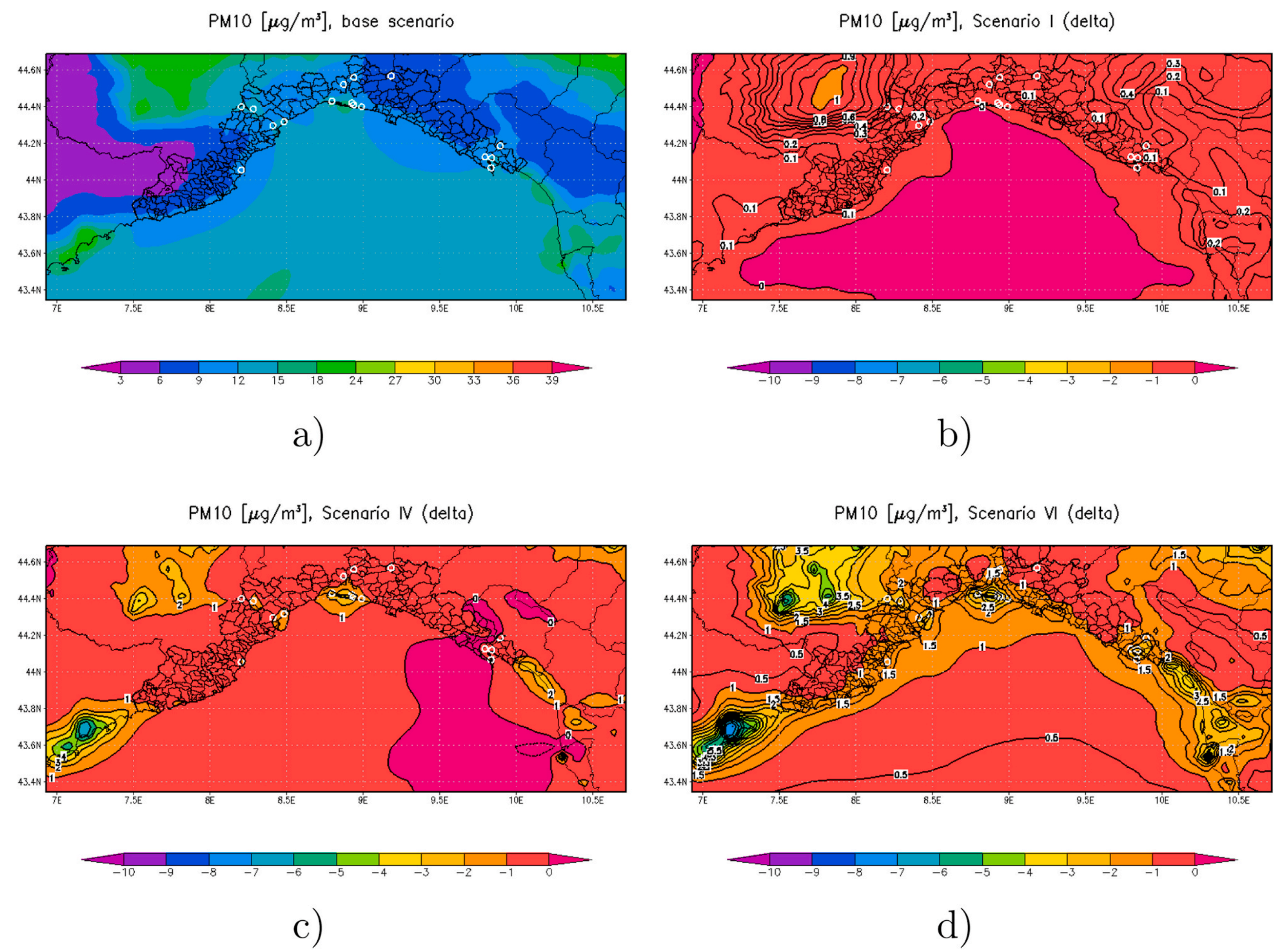


Fig. 6. Maps of annual mean and delta PM_{10} concentrations (base case and scenarios).

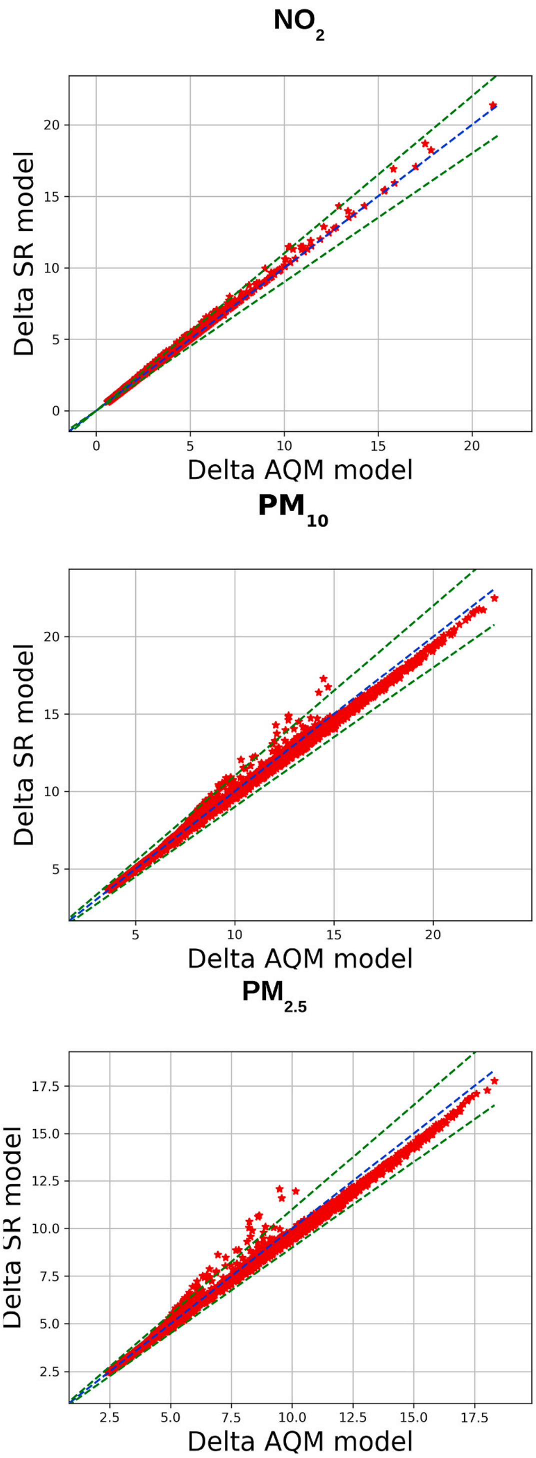
monthly means are quite similar. For PM_{10}, the time series of modelled daily mean concentrations shows a good match with observed data (Fig. 3a), 3b), 3c) and 3d)), with a slight tendency to underestimate daily mean values at the urban stations (Fig. 3b) and d)). This is also suggested by the higher values of the statistical indicators.

For O_3, all stations exhibit a good agreement between observed and simulated data (Fig. 4a), 4b), 4c) and 4d)), with a tendency to overestimate daily concentrations especially at Maggiolina station (Table 1).

2.2. Source-receptor modelling

As discussed in the previous sections, a CTM allows for simulation of the complex phenomena involving pollutants in the atmosphere. But one drawback of a CTM is the time required to perform a simulation, which then becomes an issue when several simulations are necessary to answer a specific question as source allocation. To hasten the response time, one way forward is to substitute the CTM by its associated simplified representation. SHERPA implements this concept. SHERPA mimics the behaviour of a given CTM (in this case CHIMERE) via a statistical approach. A limited number of full CTM simulations are produced, which are then used to identify the parameters needed to build the source-receptor relationships. After identification and validation of these simplified statistical relations, SHERPA can then be used to perform scenario analysis or source allocation studies, in a limited amount of time, in comparison to the full CHIMERE model. Here we recall only the main features of the SHERPA methodology. For more details, we refer to (Thunis et al., 2016; Pisoni et al., 2017, 2018).

The SHERPA concept is a data-driven approach, starting from the CTM input and output (defined as emissions and concentrations changes with respect to the basecase). The concentration change (CHIMERE concentration delta) in receptor cell j is defined as the sum of the concentration changes resulting from the changes in precursor emissions p



(caption on next column)

Fig. 7. Validation of the SRR model (as compared to the CTM reference) for NO_2, PM_{10} and $PM_{2.5}$, for scenario VII (shipping emissions).The SRR are then used to perform Source Allocation (SA) modelling. For this purpose, we selected three "control" areas (urban areas definition) that include the cities of Genoa, La Spezia and Savona, respectively. The exact control areas for the three cities correspond to the NUTS3 (province) levels as per the European Nomenclature of Territorial Units for statistics. In addition to these three areas, we also assess the contribution from emissions originating from outside the Liguria region. Results are compared with another version of the SHERPA model (hereafter SHERPA $_7$) developed by the JRC on the basis of CHIMERE 7 km resolution simulations over whole Europe (emission inventory reference year: 2010). While the SHERPA$_7$ results are based on the same underlying CTM, it should be noted that the spatial resolution, the meteorology and the emissions differ. The purpose of this comparison is therefore to evaluate the robustness of our responses. The maps of the α coefficients (these coefficients show the relative importance of a given emission precursors to pollution concentrations) e.g. for primary PM10 emissions (PPM_{10}) obtained by SHERPA$_3$ and SHERPA$_7$ are reported in Fig. 8. They are in good agreement in terms of values and spatial variability.

from any source cell i within the domain (CHIMERE emission deltas). The concentration delta in a receptor cell j can therefore be computed as follows:

$$\Delta C_j = \sum_{p}^{N_{prec}} \sum_{i}^{N_{grid}} \alpha_{ij}^{p} \Delta E_i^{p} \tag{1}$$

where N_{grid} is scenarios is required to create the SHERPA n the domain, N_{prec} is the number of precursors, ΔE_i^p [kton/year] and ΔC_j [μg/m3] are the emission and concentration deltas, and α_{ij}^p are the unknown transfer coefficients between each source cell i and receptor cell j. Finally precursor p refers to the precursor emissions changes causing the specific concentration changes for the pollutant under consideration (i.e. for NOx concentrations, the relevant emission is NOx. For PM2.5 concentrations, the relevant emissions are NOx, VOC, NH3, PPM, SO2).

The main assumption in SHERPA is that the unknown transfer coefficients α_{ij}^p can be expressed as a bell-shaped function of distance as follows:

$$\alpha_{ij}^{p} = \alpha_j^{p}\left(1 + d_{ij}\right)^{-\omega_j^{p}} \tag{2}$$

where d_{ij} is the distance between a receptor cell j and a source cell i.

The final formulation implemented in SHERPA is therefore as follows:

$$\Delta C_j = \sum_{p}^{N_{prec}} \sum_{i}^{N_{grid}} \alpha_j^{p}\left(1 + d_{ij}\right)^{-\omega_j^{p}} \Delta E_i^{p} = \sum_{p}^{N_{prec}} \alpha_j^{p}\left[\sum_{i}^{N_{grid}} \left(1 + d_{ij}\right)^{-\omega_j^{p}} \Delta E_i^{p}\right] \tag{3}$$

Additional details on the overall SHERPA methodology, and on the estimation of the coefficients (α_j^p and ω_j^p) , can be found in Clappier et al. (2015); Thunis et al. (2016); Pisoni et al. (2017).

With this formulation, various simulations can then be performed to analyse how concentrations change (in comparison to a base case) due to emission reduction scenarios. This approach is used in the next sections, to perform source allocation for the domain under study. We will compare the results obtained with a dedicated version of SHERPA, based on bottom-up (local) data with those obtained with the SHERPA top-down implementation (Pisoni et al., 2018) in which default EU-wide data are implemented.

3. Results and discussion

3.1. Scenario analysis

As mentioned above, a series of CTM scenarios is required to create the SHERPA surrogate model (training scenarios). In this work.

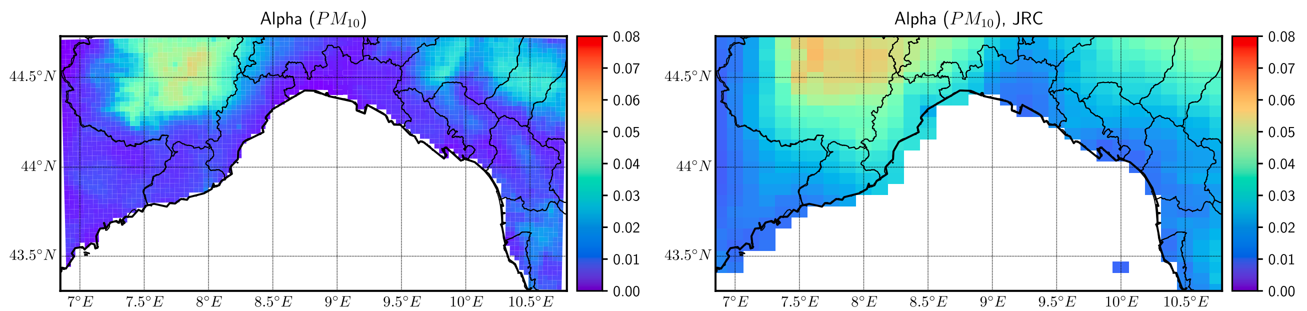


Fig. 8. Alpha values (SHERPA coefficient) for primary PM10 emissions PPM_{10}, estimated for SHERPA$_3$ (bottom-up model version) and SHERPA$_7$ (top-down model version) over the LINEA domain.

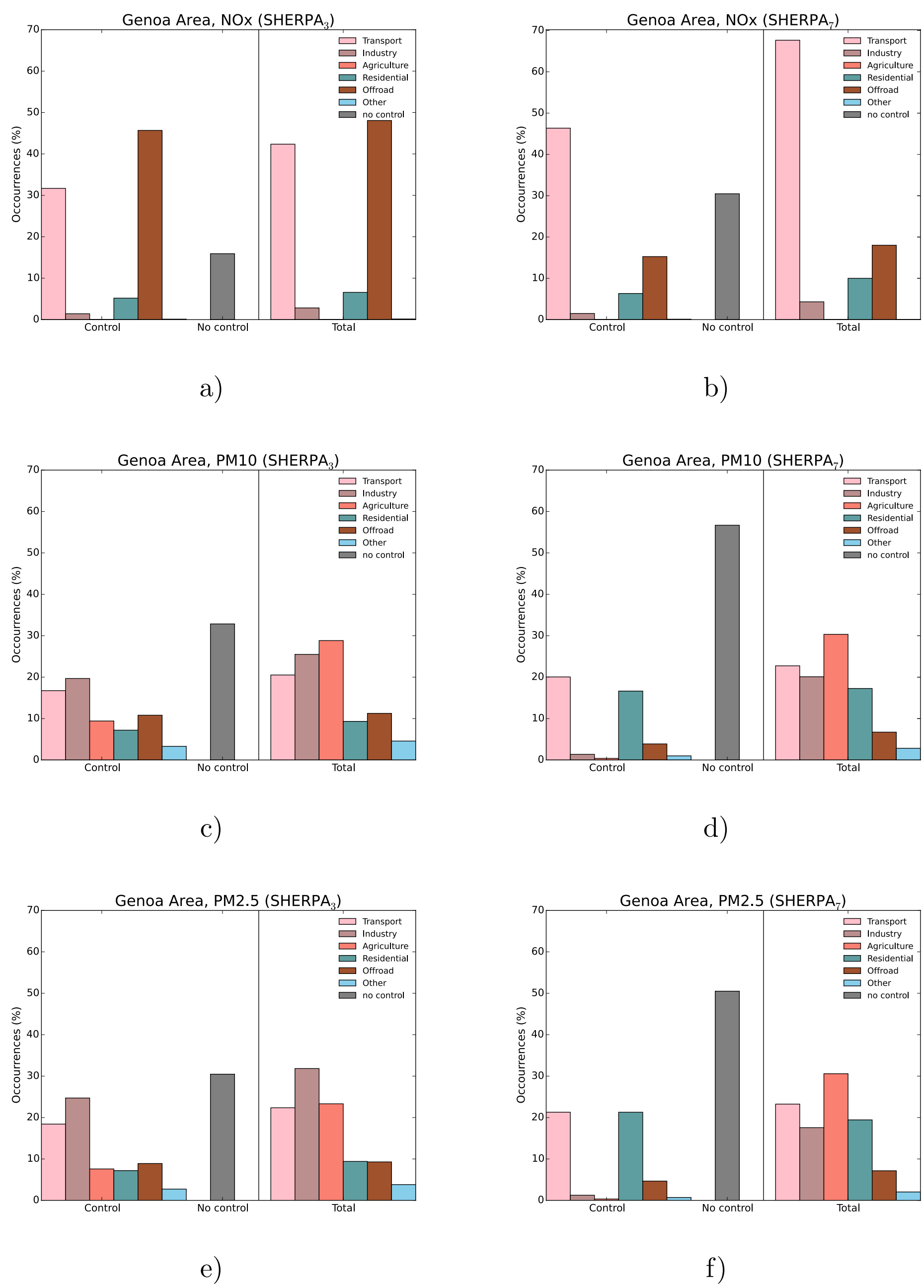


Fig. 9. SA (Source Allocation) at Genoa performed by SHERPA$_3$ (Fig. 9a), c) and 9e) and SHERPA$_7$ (Fig. 9b), d), 9f)). The "Total" entry represents the sum of the local and non-local contributions for each sector. The graphs show on the x-axis the various geographical ("control" = local emissions and "no control = linked to far-away emissions) or sectoral (Transport, Industry, etc …) sources of pollution, while the y-axis the values of relative contributions, from 0 to 100.

Table 2
Percentage contributions at Genoa, for NOx (top), PM10 (central) and PM2.5 (bottom).

	%Transp.	Indust.	Agric.	Resid.	Offroad	Other	No control
NOx							
SHERPA3	31.7	1.4	0.0	5.2	45.7	0.1	15.9
$SHERPA_3TOT$	47.6	4.3	0.0	9.9	45.0	0.0	–
$SHERPA_7$	46.2	4.4	0.0	6.3	15.2	0.1	30.5
$SHERPA_7TOT$	67.6	4.3	0.0	9.9	18.0	0.0	–
PM10							
$SHERPA_3$	16.7	19.7	9.4	7.2	10.8	3.3	32.9
$SHERPA_3TOT$	20.5	25.5	28.8	9.3	11.2	4.5	–
$SHERPA_7$	20.0	1.3	0.3	16.6	3.8	0.9	56.7
$SHERPA_7TOT$	22.7	20.1	30.3	17.2	6.7	2.8	–
PM2.5							
$SHERPA_3$	18.4	24.7	7.5	7.2	8.9	2.7	30.5
$SHERPA_3TOT$	22.3	31.8	23.3	9.4	9.2	3.8	–
$SHERPA_7$	21.3	1.2	0.3	21.3	4.6	0.7	50.6
$SHERPA_7TOT$	23.2	17.5	30.5	19.4	7.1	2.0	–

- Scenario I (training): NOx;
- Scenario II (training): NMVOC
- Scenario III (training): NH_3;
- Scenario IV (training): PPM;
- Scenario V (training): SO_2
- Scenario VI (training): All pollutants (NOx, NMVOC, NH_3, PPM1, PPM2, PPM3, CO, SO_2). Primary particulate is split in PPM1 ($\leq$ 10 mm), PPM2 ($\leq$ 2.5 and $\geq$10 mm) and PPM3 ($\geq$2.5 mm)
- Scenario VII (Validation): NOx but only SNAP8 (shipping emission).

Let's first analyse the output of these different scenarios.The map of the base case NO_2 annual mean concentrations (Fig. 5a)), shows, as expected, the highest concentrations along the main traffic lines and highways.

The highest values are located in the vicinity of the principal harbours, in particular Genoa and La Spezia where the daily limit value concentrations are frequently exceeded, followed to a lesser extent by Savona. Outside the Liguria Region, large concentrations are also modelled nearby Livorno and Nice, where significant industrial and maritime facilities are located.Scenario I (Fig. 5b)) mainly affects road transport, as exhibited by the marked concentration decrease in proximity of the western traffic lines. As expected, this is the reduction scenario that has the most impact on NO_2. The spatial distribution of the concentration deltas (difference between scenario and base case) shows a marked reduction (negative delta) on NO_2 values along the entire coastline as a result of the reductions of both the transport and off-road emissions.The impact of reducing shipping emissions (Scenario VII) is significant on harbour concentrations (Fig. 5c)), with a greater impact on the Genoa, La Spezia, and Savona locations and in a lesser measure on Nice. The Livorno area seems not to be affected by such emission reductions, suggesting that the main contributors to pollution in this area are the road transport and industrial activities. With the exception of the slightly larger values in the western part of the area (translating into a lower concentration delta), Scenario VI (Fig. 5d)) does not add any information, as compared to what was already confirmed in Scenario I. This corroborates the minor role of other than NO_x emissions on the formation of NO_2. For PM_{10}, the base case (Fig. 6a)) exhibits the largest concentrations outside of Liguria on the North-West and North-East side, as a consequence of the emissions related to the agricultural and industrial activities within the Po Valley (mostly Piedmont and Emilia-Romagna). Significant values are also found in France (Nice) and in the province of Livorno. Within the Liguria Region, the largest concentrations are modelled in proximity of the Genoa and Savona harbours, followed by La Spezia.

The impact of a NO_x emission reduction on PM_{10} (scenario I) is limited, as noticed in other CHIMERE studies. PM_{10} concentrations are little affected (Fig. 6b)), except for the decrease in the North-West area. The results of scenario IV (Fig. 6c)) show on average concentrations over the entire domain, especially in coastal areas; in particular, a substantial decrease of the PM_{10} concentrations is modelled at West (France) due probably to the reduced industrial emissions. Other reduction areas are modelled at the North-West and North-East as expected, in the proximity of Genoa, La Spezia and Livorno, and along the traffic line connecting West Liguria to France.A combined reduction of all pollutants (scenario VI) (Fig. 6d) shows a general decrease of the concentrations over the Po-Valley as well as over the Livorno province. Within the Liguria region, significant concentration reductions are modelled in proximity of the main harbours and along the whole stretch of coastline. $PM_{2.5}$ (not shown here) exhibits a spatial behaviour that is very similar to PM_{10}. Finally, the impact of emission reduction scenarios on O_3concentrations (not shown here due to lack of space) shows the usual titration effects of NOx emissions in urban areas.

3.2. Source allocation results

The 3-km resolution CHIMERE emission reduction scenarios described in Section 2.2 for the year 2016 were used together with the base case for the training of the SHERPA model (hereafter $SHERPA_3$) to produce Source Receptor Relationships (SRR). Not used for training, scenario VII which tests the impact of shipping emissions is used for the validation (see validation results in Fig. 7). In this scatter each point represents the yearly average concentrations (for the various considered pollutants) in each domain grid-cell, for the full CHIMERE model (AQM) and its the source-receptor approximation (SR).

Finally, we use both models in source allocation mode, to evaluate the impact of local (defined as local in the Figures) emissions on concentrations and the remaining concentration fraction (defined as non-local). Figures from 9a) onward show on the x-axis the various geographical or sectoral (Transport, Industry, etc …) sources of pollution ("control" means due to local emissions and "no control means linked to far-away emissions). The SHERPA $_3$SA of the NO_x emissions on NO_x concentrations[1] for the Genoa area (Fig. 9a)) reveals that the largest contributions are the offroad (harbour emissions, 45.7%) and transport (31.7%) emissions (Table 2), as already shown by the analysis of the scenario.

The remaining but minor contributions arise from the residential and industrial sectors (less than 10%). The external contribution is relatively small (less than 20%) and is mainly due to the transport sector, a sector that obtains an overall contribution of 47.6% when all transport

[1] Note that the NO_2SHERPA source allocation is done for NO_xconcentrations, later on to be converted to NO_2. In this manuscript we stick to NO_xsource allocation, to simplify the analysis; as the transformation to NO2 would also consider the application of chemical mechanisms and would make the analysis of the results more complex.

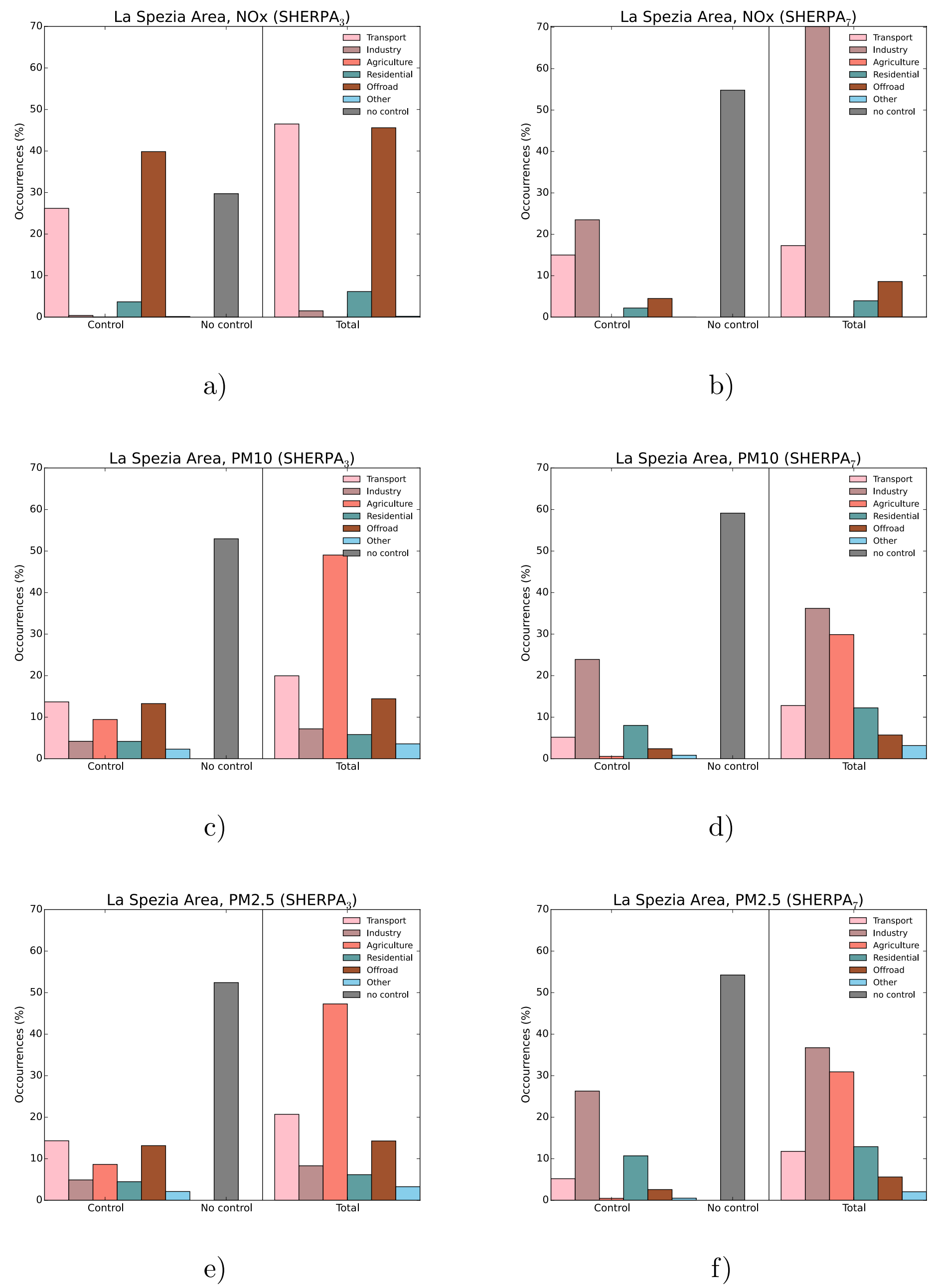


Fig. 10. SA for La Spezia location performed by $SHERPA_3$ (Fig. 10a), c) and 10e) and $SHERPA_7$ (Fig. 10b), d) and 10f)). The "Total" entry represents the sum of the local and non-local contributions from each sector.

contributions within the domain are summed up.With SHERPA $_7$, the local transport contribution (Fig. 9b)) is higher (46.2% vs 31.7%, Table 2), while the harbour traffic contribution is only 15.2% (vs. 45.7%). This difference could be linked to the different emission inventories used, as well as to the different model resolutions.

Although the differences in terms of resolution, emission inventory reference year or meteorology between $SHERPA_7$ and $SHERPA_3$ might explain some of the observed discrepancies between the two models, the

Table 3
Percentage contributions at La Spezia, for NOx (top), PM10 (central) and PM2.5 (bottom).

	%Transp.	Indust.	Agric.	Resid.	Offroad	Other	No control
NOx							
$SHERPA_3$	26.2	0.4	0.0	3.7	39.8	0.1	29.7
$SHERPA_3$TOT	46.5	1.5	0.0	6.2	45.6	0.2	–
$SHERPA_7$	15.0	23.5	0.0	2.2	4.5	0.0	54.5
$SHERPA_7$TOT	17.3	70.1	0.0	3.9	8.6	0.1	–
PM10							
$SHERPA_3$	13.7	4.2	9.4	4.1	13.2	2.3	52.9
$SHERPA_3$TOT	19.9	7.2	49.0	5.8	14.4	3.6	–
$SHERPA_7$	5.1	23.9	0.5	8.0	2.4	0.8	59.3
$SHERPA_7$TOT	12.8	36.2	29.9	12.2	5.7	3.2	–
PM2.5							
$SHERPA_3$	14.3	4.9	8.6	4.4	13.1	2.1	52.4
$SHERPA_3$TOT	20.7	8.3	47.3	6.2	14.3	3.2	–
$SHERPA_7$	5.2	26.3	0.4	10.7	2.6	0.5	54.3
$SHERPA_7$TOT	11.8	36.2	30.9	12.9	5.6	2.0	–

bottom-up and top-down approaches followed to derive the two emission inventories is likely one of the key factors explaining the varying results. It stresses the importance of using an inventory that is detailed at a regional level to capture the harbour contribution, a contribution that is of primary importance in this particular region for securing an accurate source allocation.The external Liguria contribution stands at 30.5% with SHERPA $_7$, mainly originating from the transport sector that thus reaches an overall contribution of 67.6%.According to SHERPA $_7$, the major local (e.g. from the Liguria province) contributions to PM_{10}(Fig. 9c)) in Genoa originate from the industrial (19.7%) and transport (16.7%) sectors; other contributions are lesser (e.g. off-road traffic (10.8%), agriculture and residential). The total of the local contributions reaches about 70%. The remaining 32.9% originate from outside the province and are mainly due to agricultural and industrial activities, that are transported and spread from the Po valley. On the other hand, $SHERPA_7$ identifies transport (20.0%) and residential (16.6%) activities as the prevailing sectors (Fig. 9d)), while the industrial contribution is marginal. However, the imported contribution is larger than with $SHERPA_3$ with a non-local contribution reaching 57%, of which a significant contribution comes from the agricultural emissions, followed by the transport and industrial contributions.Similar considerations can be made for $PM_{2.5}$(Fig. 9e)), with the difference that the industry contribution reaches 24.7% within the Liguria Region, and rise to 30.8% overall. $SHERPA_7$ provides a source allocation that is very similar to the one found for PM_{10}.

Generally speaking, it can be deduced that the implementation of a good SA of NO_xon a regional scale for the Liguria region requires a locally detailed (bottom-up) emissions inventory, to correctly identify the transport and harbours activities, while for particulate matter, larger scale models are necessary to capture the contributions from emissions outside the particular region of interest. Similarly to the results obtained for Genoa, the SA analysis for NO_xat the La Spezia location (Fig. 10a)) points to transport (26.2%, Table 3) and off-road (39.8%) as main contributors.

The contributions from external emissions (29.7%, higher than at Genoa) originate mainly from transport, increasing its overall contribution to 46.5%. On the contrary, $SHERPA_7$ (Fig. 10b)) indicates a significant contribution from the industrial sector (23.5%) which rises to 70.1% overall.This difference again confirms the need to use a detailed emission inventory for the correct quantification of the harbour and traffic contributions. In addition, larger scale models prove to be of prime importance to accurately quantify the non-local fraction. For PM_{10}(Fig. 10c)), the contributions are shared almost equally among transport (13.7%), agriculture (9.4%) and off-road (13.2%). The non-local contribution is actually higher than at Genoa (52.9%), mainly caused by the differences in the agricultural sector. $SHERPA_7$ (Fig. 10d)) estimates a non-local contribution of 59.3% and overall industrial and agriculture contributions of 36.2% and 29.9%, respectively. This confirms the significance of the industry sector.Similar considerations can be made for $PM_{2.5}$(Fig. 10e) and f)). The non-local contribution modelled by $SHERPA_7$ is slightly lower than for PM_{10}(54.3%). The harbour contribution on NO_Xat Savona (Fig. 11a)) is larger than at other locations with 51.8% (Table 4).

Another significant contibution arises from transport (23.1%).The non-local contribution amounts 20.8%, with off-road emissions rising up to 60% overall, more than that which was registered at other locations, probably due to other non-road machinery transport related to the agricultural-industrial activities, imported from the North-West area. Similarly to the area of La Spezia, $SHERPA_7$ (Fig. 11b)) models an important industrial contribution (23.5%), but lower contributions for

Table 4
Percentage contributions at Savona, for NOx (top), PM10 (central) and PM2.5 (bottom).

	%Transp.	Indust.	Agric.	Resid.	Offroad	Other	No control
NOx							
$SHERPA_3$	23.1	0.8	0.0	3.3	51.8	0.0	20.8
$SHERPA_3$TOT	32.5	2.6	0.0	4.7	60.0	0.1	–
$SHERPA_7$	14.1	23.5	0.0	1.3	9.9	0.0	50.9
$SHERPA_7$TOT	16.7	69.4	0.0	2.3	10.8	0.0	–
PM10							
$SHERPA_3$	16.7	14.2	11.7	16.8	7.5	1.2	31.7
$SHERPA_3$TOT	19.2	18.2	33.1	18.1	8.8	2.4	–
$SHERPA_7$	6.7	29.4	1.0	3.1	1.9	2.6	55.2
$SHERPA_7$TOT	14.2	31.9	37.7	6.5	5.0	4.6	–
PM2.5							
$SHERPA_3$	19.5	17.9	9.5	18.5	6.0	1.1	27.4
$SHERPA_3$TOT	22.1	22.8	25.8	19.8	7.1	2.2	–
$SHERPA_7$	7.1	31.0	0.9	4.0	2.1	2.9	51.8
$SHERPA_7$TOT	13.9	31.9	38.9	6.2	4.9	4.1	–

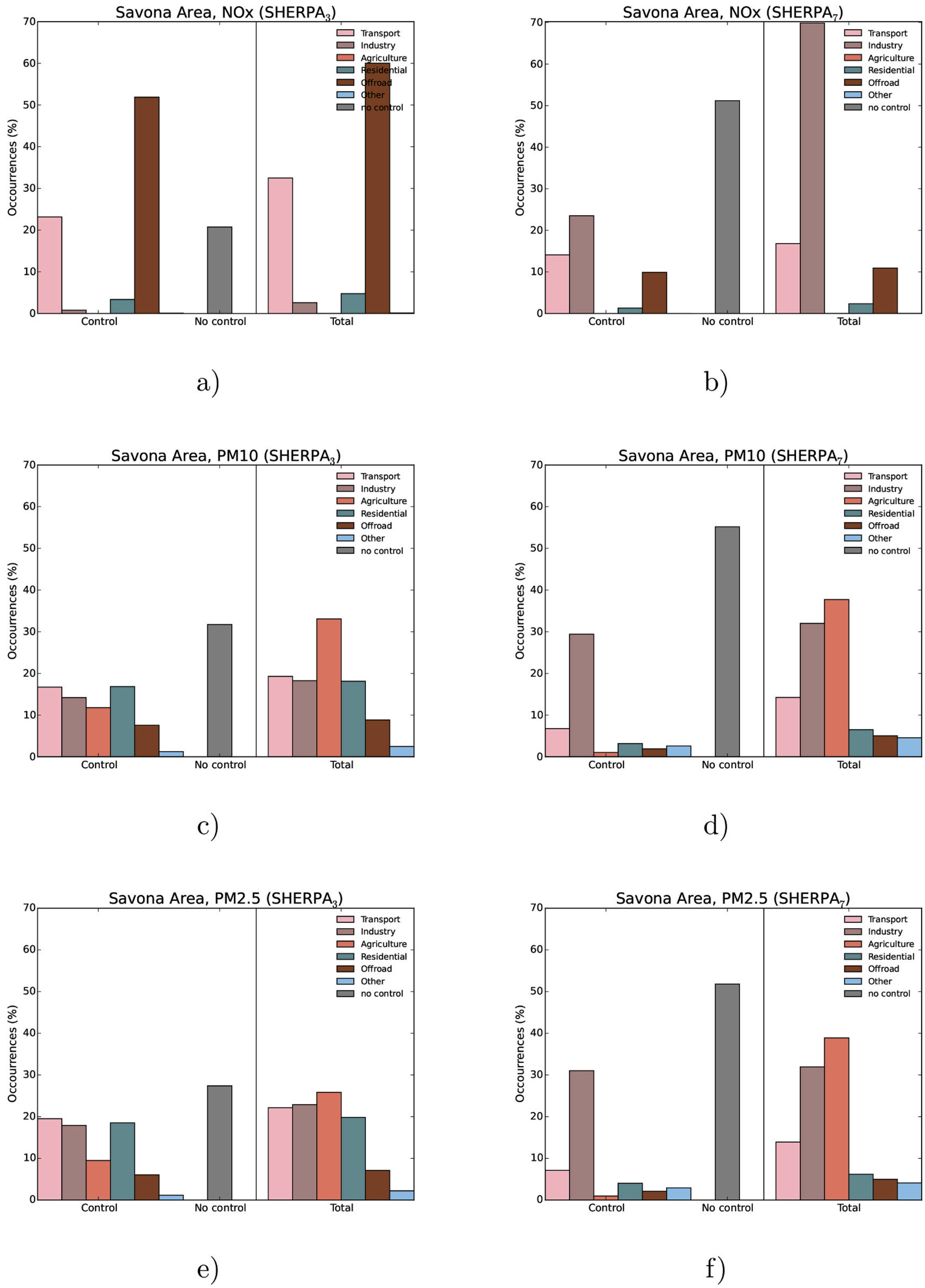


Fig. 11. SA for Savona location performed by $SHERPA_3$ (Fig. 11a), c) and 11e) and $SHERPA_7$ (Fig. 11b), d) and 11f). The "Total" entry represents the sum of the local and non-local contributions from each sector.

the off-road transport (9.9%). The contribution of the non-local emissions reaches 50.9%. This increases the overall industrial contribution to 69.4%, while off-road remains unchanged. We see from this analysis that both a detailed emission inventory and the use of large-scale models for the quantification of non-local emissions are of prime importance for local assessments.These points are well illustrated in the case of PM_{10}at Savona (Fig. 11a)). The regional model ($SHERPA_3$) estimates the industrial emissions to 14.2%, and similar values for the transport, agriculture and residential sector. The non-local contribution is 31.7% and is mainly related to agricultural emissions. With the EU-wide approach $SHERPA_7$ (Fig. 11d)), the local industrial contribution reaches 29.4%, while other local contributions remain marginal. The non-local contribution reaches 55.2%, with a high agricultural component (37%). Similar findings can be expressed for the SA of $PM_{2.5}$(Fig. 11e) and f)). The main difference with $PM_{10}$is that the non-local contribution is lower (27.4%), with a reduced relative contribution from the agriculture (25.8%). In contrast, $SHERPA_7$ models a similar agricultural contribution for both $PM_{10}$and $PM_{2.5}$.Generally speaking, La Spezia and Savona, located closer to the Piedmont and Emilia Romagna regions, respectively seem to be more affected by non-local emissions related to activities within the Po valley.

4. Discussion and conclusions

In this work, air quality modelling is performed with the purpose of assessing the impact of emission reduction scenarios on the concentrations of primary and secondary pollutants in the atmosphere, with a particular focus on the Ligurian coastal region in North western Italy. To assess a large number of potential scenarios but also to produce source allocation, it is necessary to simplify air quality models which generally require important CPU resources. To this aim, we performed 8 yearly emission scenario simulations with the multi-scale chemistry-transport CHIMERE model, at 3-km resolution over a northwestern Italy domain. We then developed a simplified set of emissions-concentrations relationships (source-receptors functions) to perform a quantitative source allocation.Results were analysed in vicinity of three major seaside cities but also within their associated provinces. The analysis of the scenarios revealed that NO_xemission reductions mostly impact NO_2concentrations locally, in proximity of traffic lines, especially in the western part of the domain and along the coastline with significant impacts close to harbours. While NO_Xemissions mostly impact NO_2locally (mainly concerning the inner transport and maritime activities), the non-local contribution is important for particulate matter. $PM_{10}$and $PM_{25}$are mostly influenced by contributions from the industrial and agricultural activities, especially from outside the Liguria territory.As we cannot compare source allocation results to observations, we also used a second set of source- receptor relationships (EU-wide) based on the CHIMERE model, but with a different set of emissions, meteorological drivers and for a different reference year. The purpose is to compare the results obtained with the two sets of SR but also to combine both results to increase the robustness of our source allocation estimates. Because of the different methodology used to build the emission inventory (emissions based on local vs. EU top-down), this second set of SR provides a slightly different signal. It is not possible to say which SR approach is the best or the most accurate, but the integrated use of the two sets of information can add value to our analysis. In this work, it is clear that the inclusion of regional scale information is key for an accurate quantification of the impacts of local emission reductions. On the other hand, a comprehensive characterization of the non-local contributions requires larger-scale models, especially when particulate matter is considered. The integration of local and non-local information is therefore important to assess the impact of emission reduction scenarios on concentrations but also for source allocation studies to support to the design of regional abatement strategies. Finally, it is important to stress that while simplified SR approaches are useful to screen different options, it is advisable to use the full air quality model to assess and confirm the impact of a designed air quality plan.

Author contribution

LS made the majority of the analysis and drafted the first version of the paper. EP contributed to the analysis and worked to finalize the manuscript. PT overviewed and supervised the manuscript finalization, and contributed to some of the key ideas implemented in the paper.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

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