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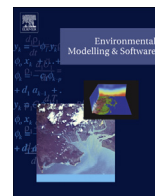
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# Adding spatial flexibility to source-receptor relationships for air quality modeling



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## ABSTRACT

To cope with computing power limitations, air quality models that are used in integrated assessment applications are generally approximated by simpler expressions referred to as “source-receptor relationships (SRR)”. In addition to speed, it is desirable for the SRR also to be spatially flexible (application over a wide range of situations) and to require a “light setup” (based on a limited number of full Air Quality Models - AQM simulations). But “speed”, “flexibility” and “light setup” do not naturally come together and a good compromise must be ensured that preserves “accuracy”, i.e. a good comparability between SRR results and AQM.

In this work we further develop a SRR methodology to better capture spatial flexibility. The updated methodology is based on a cell-to-cell relationship, in which a bell-shape function links emissions to concentrations. Maintaining a cell-to-cell relationship is shown to be the key element needed to ensure spatial flexibility, while at the same time the proposed approach to link emissions and concentrations guarantees a “light set-up” phase. Validation has been repeated on different areas and domain sizes (countries, regions, province throughout Europe) for precursors reduced independently or contemporarily. All runs showed a bias around 10% between the full AQM and the SRR.

This methodology allows assessing the impact on air quality of emission scenarios applied over any given area in Europe (regions, set of regions, countries), provided that a limited number of AQM simulations are performed for training.

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## 1. Introduction

Like in any other policy area, modeling tools are nowadays commonly used in the field of air pollution, to support policy makers in choosing the best options to improve air quality (Reis et al., 2005; Terrenoire et al., 2015). Air quality models (AQM) indeed represent the best (and only) instruments to screen and assess the impact of future policy options. But because these models include the current state of the art in terms of physical and chemical representation of the complex processes taking place in the atmosphere (captured through the numerical resolution of complex nonlinear differential equations) they generally run slow in terms of computer time and do not allow for the interactivity required by policy makers when testing various options in relation

to possible air quality plans.

This problem is exacerbated when AQMs are used in the frame of complex integrated assessment modeling (IAM) tools. IAMs have been extensively used in different policy related scales/contexts, as e.g. at the international level in support to preparation of the LRTAP (United Nation Economic Commission for Europe “Air Convention”) Gothenburg protocol (Amann et al., 2011), at European level in the frame of the National Emission Ceilings and Air Quality Directive (Kieseewetter et al., 2015), or at the national/local scales to elaborate plans and programs to improve air quality (Carnevale et al., 2014). But due to computing power limitations in IAM applications, AQM are generally approximated by simpler expressions that guarantee speed and interactivity. These expressions, often referred as “source-receptor relationships (SRR)” approximate the behavior of the complex air quality model with the objective of providing simple relationships between emissions and concentrations (Oxley et al., 2007; Pistocchi and Galmarini, 2010; Ratto et al., 2012). The first step to derive SRR consists in running the full AQM with

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different input data (i.e. emissions) that cover the desired range of future application. This step is referred to as training. In contrast, the validation phase consists in running a few AQM simulations to test the capacity of the SRR to mimic the AQM in different applications. For a meaningful evaluation, these simulations should be independent from the training simulations.

In addition to speed, it is desirable that the SRR also fulfill other characteristics, namely “spatial flexibility” and “light set-up”. By “spatial flexibility” we intend here the possibility of applying the SRR over a wide range of possible situations, in terms of the spatial design of the scenarios (i.e. having freedom in defining the areas where emission reductions will be applied). By “light set-up” we mean both that the number of full AQM simulations requested for the training of the SRR should be limited, and that the level of knowledge required for the analyst to train the SRR should be limited (i.e., using simple regression techniques, etc ...). Given the complexity of the AQM and the time required to perform simulations, it is important to keep the number of simulations in the training set under control, without compromising accuracy. Speed, flexibility and light setup do not naturally come together and a gain in spatial flexibility will most of the time be obtained at the expense of a heavier set-up or of a loss in terms of speed. The challenge therefore consists in ensuring a good compromise among these three characteristics, while preserving accuracy, i.e. a good comparability between SRR results and AQM.

According to their purpose, currently used SRR methodologies generally privilege one or two of the above mentioned characteristics in detriment of the others. The GAINS (“Greenhouse Gas - Air Pollution Interactions and Synergies”, Amann et al., 2011, Kieseewetter et al., 2015) integrated assessment tool relies on the EMEP (“European Monitoring and Evaluation Programme”) air quality model to build its SRR (Tarrasón et al., 2004). In this approach, emissions are aggregated in terms of countries, resulting in “country-to-grid” SRR. Being proportional to the number of countries and emission precursors considered, the number of simulations requested for the training is substantial. Given the way they are constructed, the country-to-grid EMEP SRR can only be applied to assess the impact of scenarios in which emissions have been changed over the countries considered during the training. This results in a lack of spatial flexibility, i.e. the impossibility to use SRR to evaluate subnational emission reduction scenarios. The GAINS-EMEP SRR, however, run fast as the number of operations is proportional to the number of countries and precursors involved.

In the AERIS (“Atmospheric Evaluation and Research Integrated system for Spain”) model emissions are not aggregated spatially but in specific sectors (Vedrenne et al., 2014). Full AQM simulations in which these sectors are reduced individually are then used in the training phase to construct the SRR. Because the number of requested simulations is proportional to the number of sectors considered, the setup can be quite light. Spatial flexibility is on the contrary absent because all emission reductions considered in the training are performed domain wide. Similarly to the EMEP SRR, this approach also runs fast.

Another methodology has been implemented in the RIAT + tool (Carnevale et al., 2012). Emissions are here aggregated in four large quadrants that are defined relatively to each grid cell of the domain (sliding quadrants). The quadrant emission values and their related grid cell concentrations are then used to feed a neural network that delivers the SRR (Carnevale et al., 2009). Although the approach requires a limited number of full AQM simulations (around 20), the set-up of the SRR remains complex due to the need of implementing neural networks. Neural networks also require that their application is limited to the range of situations covered during the training phase. From a speed point of view, the sliding quadrant-to-

cell approach performs very well.

Clappier et al. (2015) (referred as C2015 in the following) proposed a new methodology (referred to as “Multi-ring”) to derive SRR. Similarly to the quadrant-to-cell approach described above, these SRR make use of sliding emission aggregations (rings) but assume linearity in the emission-concentration relationships. The main consequence of this linearization is the simplification of the training phase.

In this work, we further elaborate on the approach of C2015 and show how it can be further developed to improve spatial flexibility. In Section 2, we briefly review the main elements of the C2015 work and discuss its limitations in terms of spatial flexibility. In Section 3 an improved methodology is proposed while Section 4 evaluates the results of this approach for a series of case-studies.

## 2. The “multi-ring” approach and its limitations

In this section we briefly review the C2015 methodology main features and limitations.

### 2.1. Methodology

As previously stated, the goal of the SRR is to mimic an AQM to calculate as quickly as possible the effect of emission reductions on concentration levels (Castelletti et al., 2012). In general, the SRR model consists in an algebraic relationship between gridded emissions and concentrations. Although concentrations and emissions are defined on the same grid cells, we make here a distinction between sources (emissions) and receptors (concentrations) grids for convenience.

A series of steps are detailed in C2015 in order to design the SRR, which are briefly summarized as follows:

- 1) The calculation of SRR algebraic relationships between emissions and concentrations expressed in absolute terms can lead to errors if not accounted for correctly. This problem disappears if emission and concentration are expressed in relative terms, i.e. as difference (delta) between a base case and a reduction scenario (Thunis et al., 2016).
- 2) For long term indicators (i.e. yearly average) which are the focus of this work, the relationship between emission and concentration deltas can be approximated accurately with a linear function (Thunis et al., 2015). Consequently and since the concentration change in a receptor cell “j” can result from the reduction of different emission precursors “p” coming from any source cell “i” within the domain, the concentration delta in a receptor cell “j” can therefore be computed as follows:

$$\Delta C_j = \sum_p^P \sum_i^N a_{ij}^p \Delta E_i^p \quad (1)$$

where  $N$  is the number of source grid cells within the domain,  $P$  is the number of precursors,  $\Delta E_i^p$  and  $\Delta C_j$  are the emission and concentration deltas,  $a_{ij}^p$  are unknown parameters to be identified.

- 3) The number of unknown parameters ( $a_{ij}^p$ ) which need to be identified in the case of a cell per cell relationship is prohibitive (equation (1)). Indeed for a  $N = 50 \times 50$  grid cells domain and  $P = 5$ , the identification of about 12,500 parameters is required to calculate  $\Delta C_j$ . 12,500 unknown parameters would need to be identified by solving an equations system that contains at least 12,500 equations, each of these relying on  $\Delta C_j$  and  $\Delta E_i^p$  provided by an independent CTM scenario run, which is materially unfeasible.

- 4) The number of unknown parameters can be reduced by aggregating source cells in entities called “source aggregations” (S-aggregations). The number of unknowns becomes then proportional to the number of S-aggregations and equation (1) becomes:

$$\Delta C_j = \sum_p^P \sum_k^{N_A} a_{kj}^p \Delta E_{kj}^p \quad (2)$$

where  $i$ , substituted by  $k$ , is now the index referring to the S-aggregation “ $k$ ”,  $N_A$  is the number of S-aggregations related with receptor “ $j$ ” and  $\Delta E_{kj}^p$  is the sum of the emission deltas of the source cells which have been aggregated into the S-aggregation “ $i$ ”.

The S-aggregations can be fixed “geographically” (e.g. countries, regions or a set of regions/countries) similarly to the GAINS-EU approach (Amann et al., 2011). These S-aggregations remain then unchanged for all receptor cells. C2015 showed that sliding S-aggregations can also be defined. In this case, their locations relative to the receptor cells always remain unchanged. After testing different sliding aggregations, configurations entities arranged in several rings increasing in size around a receptor cell were shown to better describe the spatial resolution of the emission impacts. Fig. 1 (left) shows an example of 25 entities distributed in 3 rings with dimensions increasing with distance from the receptor cell. The number of unknowns parameters to be identified in the case of sliding entities becomes  $N_A \times P = 25 \times 5 = 125$  per cell.

- 5) As per equation (2) the number of unknown parameters ( $a_{kj}^p$ ) that need to be identified for one receptor cell “ $j$ ” equals the number of emission aggregations ( $N_A$ ), if a single precursor is considered. As mentioned above, this system can only be solved if at least  $N_A$  equations are available as a result of a similar number of independent scenarios. C2015 showed that the number of equations available from a given scenario could be increased by opening a so-called “receptor window” (R-windows). This R-window is defined by assuming that the  $a_{kj}^p$  coefficients are the same for receptor cells belonging to a given zone defined around each receptor cell “ $j$ ”. With this assumption, additional equations can be created from the same set of available AQM scenarios as shown with the example below:

$$\Delta C_j^{SC(1)} = \sum_{p=1}^P \sum_{k=1}^{N_A} a_{kj}^p \Delta E_{kj}^{p,SC(1)}$$

$$\Delta C_{j+1}^{SC(1)} = \sum_{p=1}^P \sum_{k=1}^{N_A} a_{kj}^p \Delta E_{kj+1}^{p,SC(1)}$$

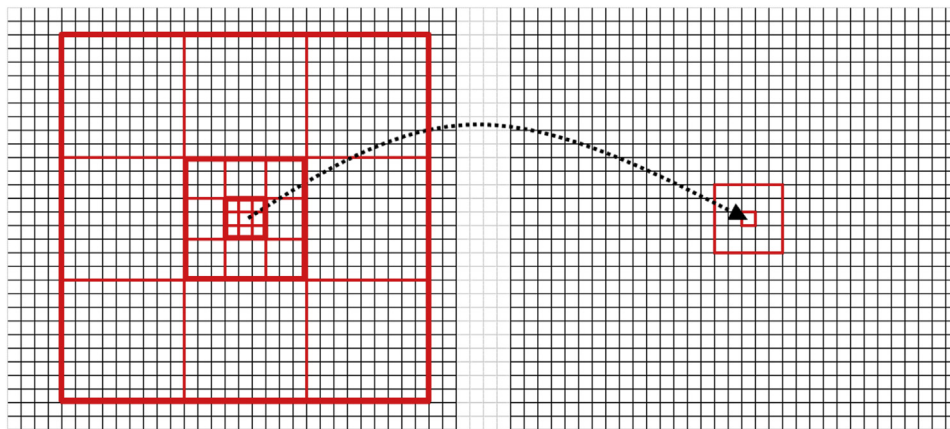
in which the same “ $a$ ” coefficients are used in both equations. Considering each receptor cell “ $j$ ”, the number of available equations is then equal to  $N_{sc} \times N_w$  where  $N_{sc}$  is the number of scenario runs and  $N_w$  is the number of receptor cells inside a R-window. For a R-window containing 25 cells Fig. 1, right), the number of available equations becomes 200 if 8 reduction scenarios are considered, which is more than the 125 unknowns to be identified.

- 6) On the one hand, a system containing more equations than unknowns (e.g. a large R-window combined with few S-aggregation entities) increases the robustness of the estimation of the unknowns. As a result the SRR coefficients will be less sensitive to the input (i.e. different set of scenario runs will always produce similar regression coefficients  $a_{kj}^p$ ). On the other hand, large R-windows and limited S-aggregations are not a good configuration to capture spatial variations in emissions and concentrations and the resulting accuracy might be lower. A compromise needs therefore to be found between accuracy and robustness when selecting the number of aggregations and the dimension of the R-window.

## 2.2. Limitations

In C2015 the “Multi-ring” approach (implemented as explained in the previous section) showed to perform very well for a test case over the Emilia Romagna region (Northern Italy). In this application a set of 8 emission reduction scenarios (training scenarios) performed with an AQM model has been used to calculate the SRR unknown parameters  $a_{kj}^p$ , while a second set of 4 scenarios (validation scenarios) was used to check the accuracy of the SRR.

The “Multi-ring” approach has been further tested over several other regions and always showed to perform well. However, in all the designed validation scenarios, emission reductions have been applied over the same entire domain as imposed in the training scenarios. In the current work, validation is extended to account for



**Fig. 1.** Receptor windows and source aggregations configurations, showing (left) an example of 25 emission aggregation entities (thin red lines) distributed in 3 rings (thick red lines), and (right) the assumption where the SR model coefficients are assumed to be equal over a given geographical area of 25 cells. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

the possibility of reducing emissions over a smaller area than the training area. To illustrate this issue, Fig. 2 shows the comparison between the SRR and the AQM model when the model is trained over a larger area (entire Europe) than the one where emission reductions are applied (France in our case). In such situation, the performances of the “Multi-ring” approach clearly deteriorate.

The SRR overestimates concentration deltas (Fig. 2, left) leading to an underestimation of concentrations levels over most of the domain (Fig. 2 right). The error reaches 16%, while it was kept below 5% for previous test cases (C2015), characterized by similar emission reduction areas in both the training and validation scenarios. We will show below that this poorer performance can be explained by a lack of robustness.

Generally, robustness can be enhanced by increasing the number of input parameters ( $\Delta E_{kj}^p$  and  $\Delta C_j$ ), which results in covering a larger range of their values and therefore provides a more comprehensive overview of their possible variations. Increasing the number of input parameters also allows increasing the number of equations used for the regression. However, this will result in an effective improvement of the robustness only if equations are independent from each other. Indeed, correlated input parameters will lead to a system in which equations can be linearly predicted from each-others, with a substantial degree of accuracy. This is referred as “co-linearity” in statistics, and was partly tackled in C2015 by using a PCR (Principal Component Regression) approach (Seber et al., 2003). In C2015 the PCR was computed in two steps. First, a Principal Component Analysis (PCA) was applied to the input data (i.e. to the  $\Delta E_{kj}^p$  of the multi-ring aggregated emissions) to calculate a new set of input data, called Principal Components, expressed as a linear combination of the original ones. These new inputs are independent from each other (i.e. linearly uncorrelated) and their components ranked in terms of variance from highest to lowest. If the inputs data are already independent or close to independency, the variance of the new components calculated by the PCA remains close to the variance of the input data, and the PCA is not efficient. On the contrary, if the input data are initially highly correlated, the PCA increases the discrepancy between the variance of the different components (i.e. the variance of the first new component is much higher than the variance of the original input, while the variance of the last component is much lower than the variance of the original input). In the second step, a multi-linear regression is applied only to the subset of components that

explains at least 95% of the total variance.

Although different tests have shown that the PCR approach generally improves robustness (C2015), it was still not sufficient to get accurate results with the “Multi-ring” approach in case of sub-domain reductions, as shown in Fig. 2.

Note that for the “Multi-ring” approach consisting of 25 S-aggregations and 5 precursors, the PCA first transformed the 125 inputs (for each receptor cell “j”) into 125 linearly independent new inputs. Application of the PCA in our case studies led to 95% of the total variance being explained by only the 5 highest ranked components in terms of variance. All other components (i.e. 120) did not show enough variance to be significant and were therefore not considered for the next step, the PCR. Moreover, each of these 5 remaining components corresponded to one of the 5 precursors, and therefore resulted from a linear combination among all multi-ring aggregation entities for that precursor. This clearly indicated that the original input data (i.e.  $\Delta E_{kj}^p$  of the S-aggregations provided by the training scenarios) were spatially highly correlated.

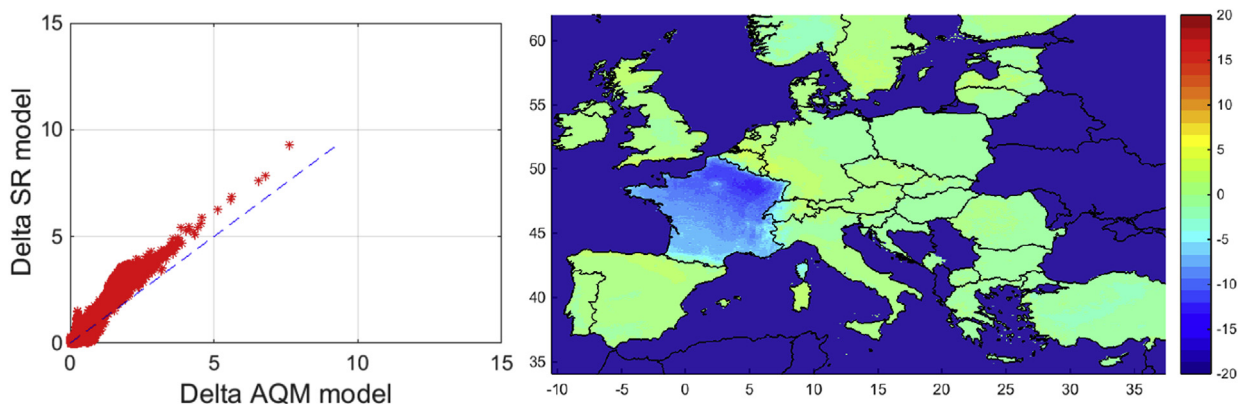
In summary, the PCR improved the robustness by removing non-significant components but did not solve the issue of the correlation among multi-ring aggregation entities. As a consequence, robustness was increased but only as long as validation and training scenarios were spatially well correlated. This is the case when training and validation are performed over the same areas. When validation is performed on smaller areas the spatial correlation between training and validation emissions is lower. This is why performance was reduced in such case. We present in the next section an approach to overcome this problem.

### 3. The “bell-shape” approach

#### 3.1. Methodology

The approach proposed in this section is based on the cell-per-cell relationships described by equation (1). It builds on the concept of “Geographically Weighted Regression “ (GWR, Fotheringham et al., 2002)” or “local modeling approaches” (Lloyd, 2010), a family of approaches that uses “bell-shaped” kernel functions to establish weighted, local regressions between input and output variables. To the knowledge of the Authors, these approaches have never been used for SRR in the field of air quality IAM.

As mentioned, equation (1) requires a very large number of scenario simulations to identify all the unknown parameters. If we



**Fig. 2.** Comparison of AQM and SRR results for  $PM_{25}$  concentrations [ $\mu g/m^3$ ] deltas (change in comparison to the basecase), when emissions are reduced only over France. Left: scatter plot representing, for each cell, the CTM vs SRR yearly averages of  $PM_{25}$  concentration deltas. Right: “percentage bias map” of the relative difference between SRR and CTM results (note the scale between  $-+20\%$ , stressing the high percentage error).

assume this large number of simulations to be available and each simulation to provide independent information, the unknown parameters could then easily be computed by means of a multi linear regression and be equal to:

$$a_{ij}^p = r_{ij}^p \cdot \frac{\sigma_j}{\sigma_i^p} \quad (3)$$

where  $r_{ij}^p$  is the correlation coefficient between the concentration and emission deltas ( $\Delta C_j$  and  $\Delta E_i^p$ ) of the different scenario runs,  $\sigma_j$  is the standard deviation of the concentration deltas ( $\Delta C_j$ ) and  $\sigma_i^p$  is the standard deviation of the emission deltas ( $\Delta E_i^p$ ).

Unfortunately, the number of scenario runs required to perform a multi-linear regression to solve equation (1) is prohibitive and therefore not feasible. Moreover, the scenario runs can never be fully independent (i.e. the  $\Delta E_i^p$  provided by the different scenarios are always more or less correlated to each other) so that expression (3) cannot be used to compute the unknown coefficients of equation (1).

The basic principle used in the approach proposed here is to link the  $a_{ij}^p$  coefficients (varying on a cell by cell basis) to the distance between receptor cells “j” and source cells “i”. We will assume that the relationship that links the  $a_{ij}^p$  coefficients to distance depends on the correlation coefficient  $r_{ij}^p$ , as expressed in (3).

To identify these relationships, we proceed as follows: for a given distance ( $d_{ij}$ ), we select all receptor cells “j” that are at a distance  $d_{ij}$  from a given source cell “i” and select the corresponding emission and concentration deltas. The operation is then repeated for all source cells in the domain and a correlation coefficient  $r_{ij}^p$  calculated. These steps are performed for a series of growing distances and for each precursor. Fig. 3 shows the progressive decrease of the precursors (NO<sub>x</sub>, NH<sub>3</sub>, PPM, SO<sub>2</sub>) correlation coefficient with distance. VOC is not shown as this precursor does not impact significantly PM concentrations.

If we assume the coefficients  $a_{ij}^p$  to be closely linked to the  $r_{ij}^p$ , the  $r_{ij}^p$  trend can be used as a model for the  $a_{ij}^p$  trend. Then, this trend can be reasonably well approximated by the following function:

$$a_{ij}^p = \alpha_j^p (1 + d_{ij})^{-\omega_j^p} \quad (4)$$

where  $\alpha_j^p$  and  $\omega_j^p$  are the amplitude and width of the function.  $\alpha_j^p$  is the value of  $a_{ij}^p$  when  $d_{ij} = 0$  (i.e. when  $j = i$ ), and can be interpreted as the relative importance of each precursor “p” in producing the pollutant concentration  $C_j$ . Fig. 3 shows that PPM contributes more to the PM<sub>2.5</sub> production than NH<sub>3</sub>, NO<sub>x</sub> and SO<sub>2</sub>.  $\omega_j^p$  represents the decay rate of  $a_{ij}^p$  with  $d_{ij}$  and indicates how the contribution of the precursor “p” emissions decreases with the distance. Fig. 3 shows that the influence of the PPM emissions decrease more rapidly with distance than for NH<sub>3</sub>, NO<sub>x</sub> and SO<sub>2</sub> emissions.

We assume that, even if the general trend is the same over the whole domain, parameters  $\alpha_j^p$  and  $\omega_j^p$  can vary spatially.

### 3.2. Computation procedure

Using equation (4) to calculate  $a_{ij}^p$  considerably reduces the number of unknowns, as only the two parameters  $\alpha_j^p$  and  $\omega_j^p$  need to be identified to solve equation (1) at a given receptor cell “j”:

$$\Delta C_j = \sum_p \sum_i \alpha_j^p (1 + d_{ij})^{-\omega_j^p} \Delta E_i^p = \sum_p \alpha_j^p \left[ \sum_i (1 + d_{ij})^{-\omega_j^p} \Delta E_i^p \right] \quad (5)$$

The parameters  $\alpha_j^p$  and  $\omega_j^p$  can be calculated using a methodology comparable to the “Multi-ring” approach described above. A least square estimation is performed using as input the concentration and emission deltas ( $\Delta C_j$  and  $\Delta E_i^p$ ) provided by the different scenario runs. As compared to the “Multi-ring” approach, the fit function is not anymore multi-linear but becomes exponential because  $\omega_j^p$  appears as an exponent in (5). Note also that the distances  $d_{ij}$  appear as input in addition to  $\Delta C_j$  and  $\Delta E_i^p$ . A R-window similar to the one defined for the “Multi-ring” approach can be used to generate additional equations. With this approach, each cell belonging to the R-window provides a different set of inputs ( $d_{ij}, \Delta C_j$  and  $\Delta E_i^p$ ) and all sets of inputs are then used to estimate a unique

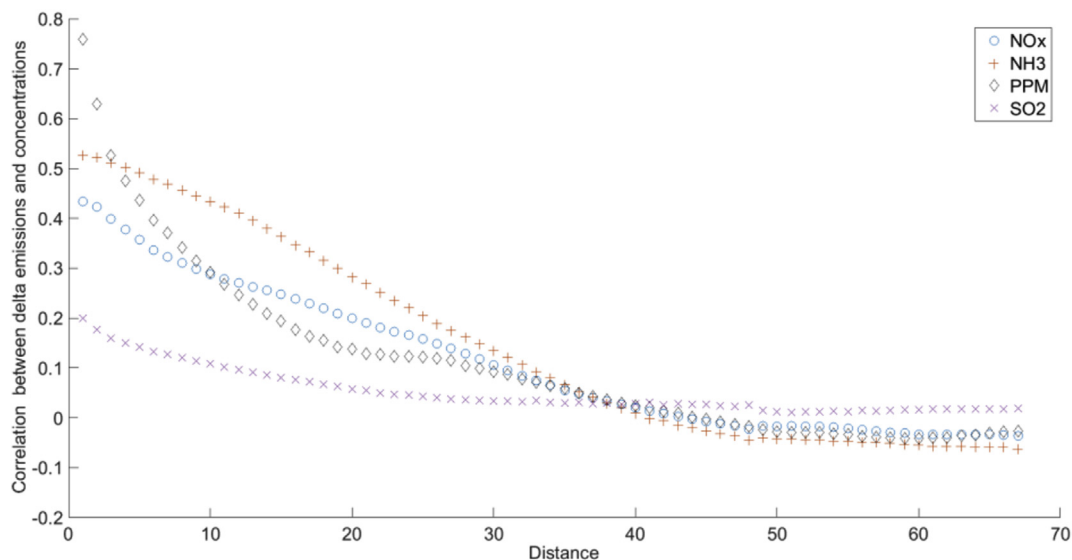


Fig. 3. Spatial average correlation between delta emissions and delta concentrations, for growing distance (in terms of number of cells) among cells (see text for more details).

set of outputs ( $\alpha_j^p$  and  $\omega_j^p$ ) for each receptor cell. Similarly to the “Multi-ring” approach, a large R-window provides a large range of input data and leads to a more robust estimation of the outputs  $\alpha_j^p$  and  $\omega_j^p$  while a small R-window better captures the spatial variability of the outputs and leads to a better accuracy. Sensitivity tests performed for different R-window sizes have shown that:

- The least square estimation converges with difficulty when applied to all precursors at the same time and works best when applied to scenarios in which emissions have been reduced one precursor at a time.
- Values of  $\omega_j^p$  differ significantly from one precursor to the other but show a low spatial variability whereas the  $\alpha_j^p$  values show a high spatial variability.

Consequently, a two-steps procedure has been designed to find the best compromise between robustness and accuracy for each of the 2 parameters.

Initially (Step 1), each precursor is treated independently and we therefore only consider scenarios in which emission reductions are applied independently to each precursor (i.e., we consider independently 4 scenarios, with reductions of the NO<sub>x</sub>, NH<sub>3</sub>, PPM, SO<sub>2</sub> emission precursors). As mentioned above, VOC is not used as it does not impact PM significantly. A least square estimation (between emission and concentration changes) is performed (for each precursor separately) to estimate  $\alpha_j^p$  and  $\omega_j^p$ , using all cells in the domain (i.e. a unique R-window covering the entire domain). Experience however showed that results improved when grid cells were split in groups. In the current approach two groups of cells differentiated in terms of wind speed intensity have been selected (cells with a wind speed  $\leq 0.5$  m/s and cells with a wind speed  $> 0.5$  m/s). This split leads, for the first step, to two values for  $\alpha_j^p$  and  $\omega_j^p$ .

In a second step (step 2) an emission weighted average delta is computed at each receptor cell “j” using function (4) with the  $\omega_j^p$  identified in step 1:

$$\Delta \bar{E}_j^p = \sum_i (1 + d_{ij})^{-\omega_j^p} \Delta E_i^p$$

During this step, all training scenarios are used at the same time (scenarios with reduction of one precursor at a time, and scenarios with all precursors reduced contemporarily) to calculate more

precise values for  $\alpha_j^p$ . To this purpose, a multi linear regression (between emission weighted average deltas and concentration deltas) is used at each receptor cell as follows:

$$\Delta C_j = \sum_p \alpha_j^p \Delta \bar{E}_j^p$$

This two-step approach leads to a good compromise between robustness and accuracy. Indeed while step 1 increases robustness by using a large number of equations (provided by one scenario per precursor but for a large number of cells) to estimate only two values per precursor, it is not very accurate. In fact the accuracy of  $\omega_j^p$  is quite satisfactory already as this parameter exhibits only minor spatial variability, but this is not the case for  $\alpha_j^p$ . This is why the approach includes a second step during which new values of  $\alpha_j^p$  are estimated using all scenarios on a grid by grid basis for each precursor. This of course generates much less equations than for Step 1, resulting in more accurate but less robust estimates for the  $\alpha_j^p$  parameters.

#### 4. Case study

In this section the proposed “Bell-shape” approach is tested on a real case study. More specifically, we will focus on the link between PM<sub>2.5</sub> concentrations and its emission precursors (PPM, NO<sub>x</sub>, SO<sub>2</sub> and NH<sub>3</sub>) in Europe. To this purpose, the CHIMERE air quality model was run over a domain covering the entire European territory (Fig. 4) to deliver the necessary emission precursor and related concentration fields (Terrenoire et al., 2015). Because the long-term effects of PM<sub>2.5</sub> high concentrations are the most significant, only annual mean concentrations are considered and thus all model input and output data (emissions and concentrations) are averaged yearly.

The simulations include a base case (2010 emissions, 2009 meteorology) and a series of emission reduction scenario to identify the parameters of the SRR.

In all training scenarios emission reductions are applied over the entire modeling domain (i.e. Europe). Another series of scenarios is dedicated to the validation of the approach. Given that our main objective resides in the possibility of applying emission reductions over any given area, the validation scenarios will focus on emission reductions imposed regionally and locally in different areas of the

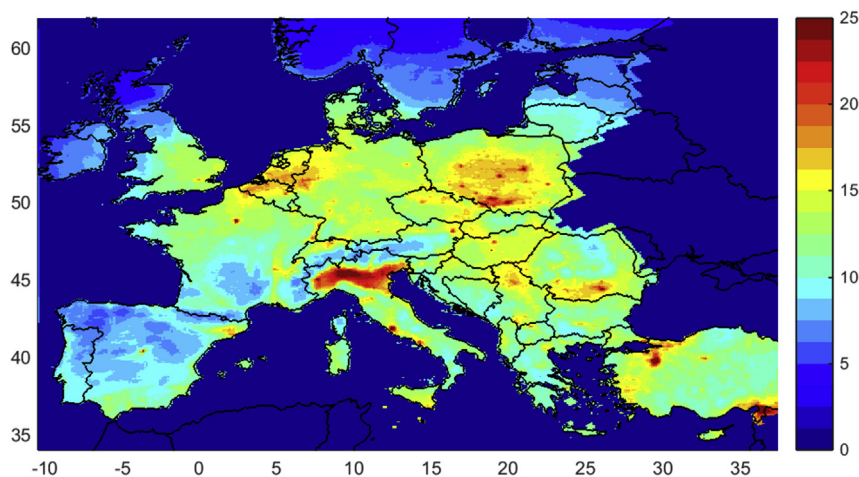


Fig. 4. Domain selected for the simulations with the AQM. The map shows yearly PM<sub>2.5</sub> concentrations [µg/m<sup>3</sup>] computed by the model.

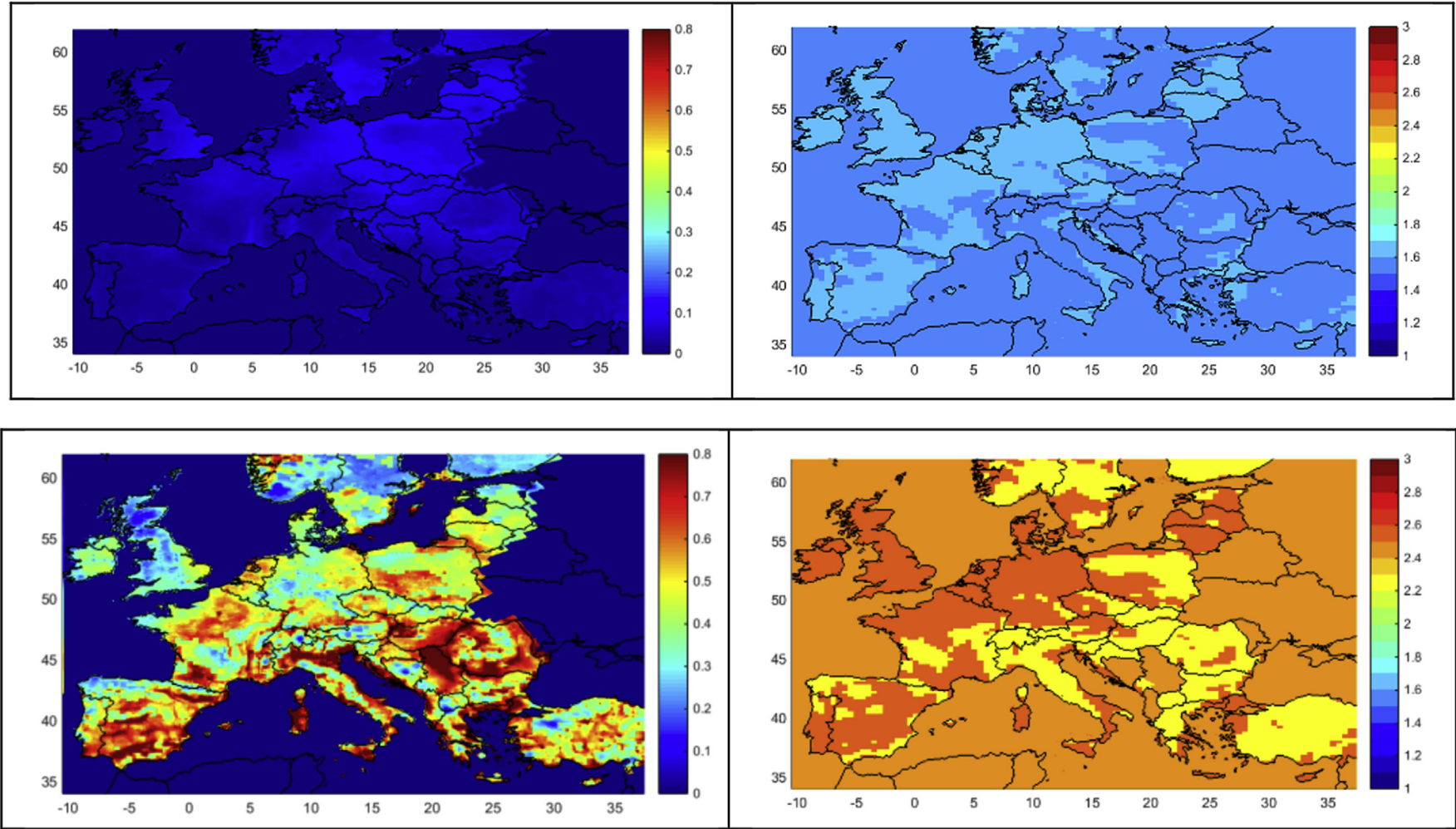
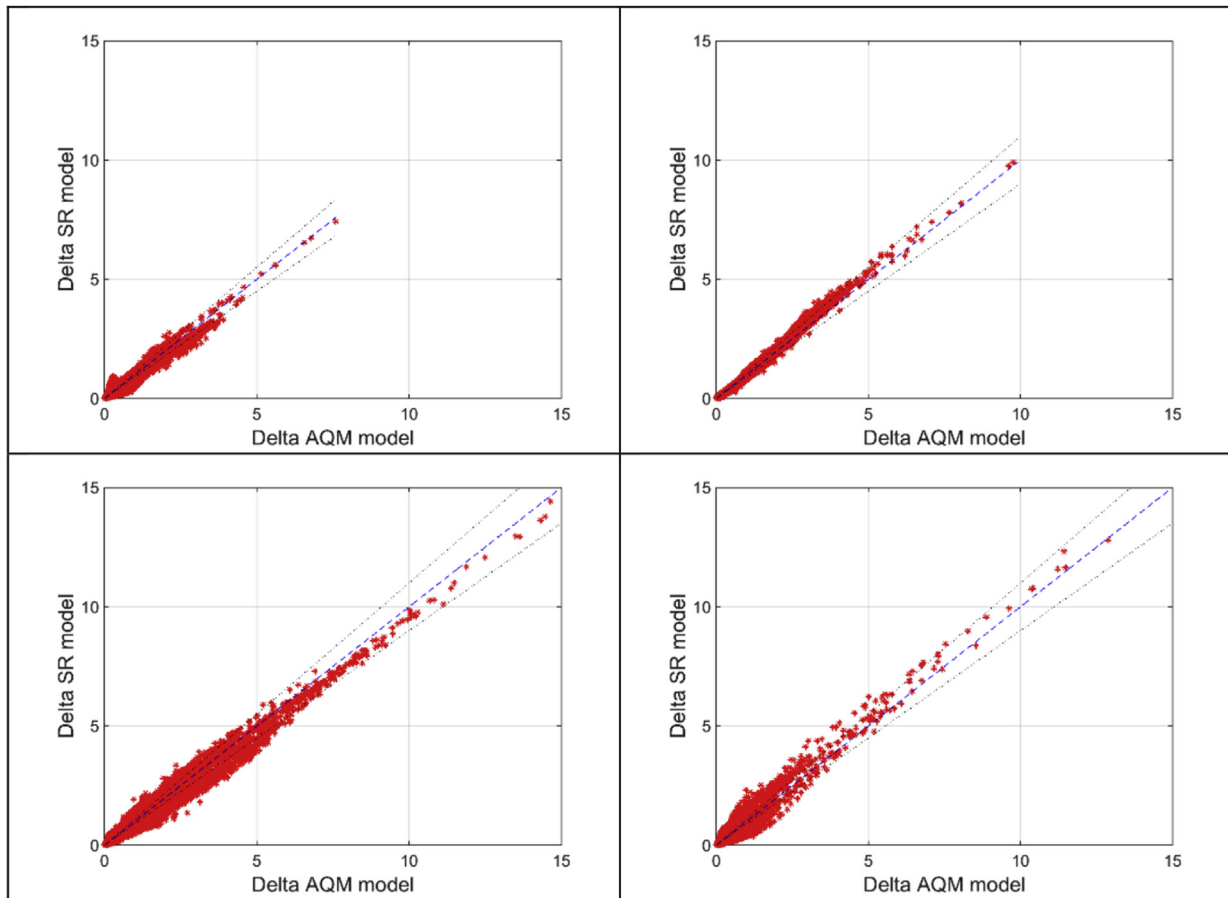


Fig. 5.  $\alpha$  (left) and  $\omega$  (right) coefficients for  $\text{NH}_3$  (top) and PPM (bottom) over all considered geographical domain.





**Fig. 6.** Validation scenarios for yearly  $PM_{25}$  [ $\mu\text{g}/\text{m}^3$ ], with emission reductions applied over France (top left), Poland (top right), for all “regional” reduction scenarios (bottom left) and all “local” reduction scenarios together (bottom right). Values are expressed in terms of concentration changes in  $\mu\text{g}/\text{m}^3$ , comparing the AQM (x-axis) and the source-receptor model (y-axis). Blue dotted lines represent the  $\pm 10\%$  error range. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

domain. The purpose of the validation will then be to assess whether the SRR are able to reproduce the full AQM results for these local reduction cases.

The training set (7 simulations) contains scenarios with reductions of one precursor at a time (to identify the  $\omega_i^p$  parameters in Equation (4)) and reductions for all precursor contemporarily (to identify the  $\alpha_i^p$  parameters in Equation (4)) The level of reductions for each precursor and scenario is set to 50% but this level can freely be selected as a consequence of the linearity assumption made in this work.

The validation set includes emission reductions applied at “country”, “regional” and “local” level, for different precursors (see next Sections for more details).

#### 4.1. Identification of the SRR parameters

Before assessing the performances of the SRR, the values of the SRR coefficients  $\alpha$  and  $\omega$ , resulting from the training phase, are discussed, as they provide useful information on the dependency of the distance (between emissions and concentrations) based SRR, in terms of geographical area and emission precursor.

The results of the training phase show that, for any given precursor, the geographical variability of  $\omega$  is limited (Fig. 5, right) whereas  $\omega$  shows larger differences from precursor to precursor (results are shown only for  $NH_3$  and PPM, but similar conclusions are valid for all other precursors). Higher values are found for PPM

stressing the more local influence of that precursor on concentrations (narrow bell-shape) while smaller values are found for  $NH_3$ , indicating the higher influence from far-away cells.

In comparison to  $\omega$ , the  $\alpha$  parameter exhibits a higher spatial variability (Fig. 5, left) indicating that the importance of one emission precursor with respect to the others can change significantly from cell to cell (this is mainly visible for PPM, but also for  $NH_3$ ). The  $\alpha$  parameter for PPM is the largest of all precursors, indicating the key importance of PPM in producing  $PM_{25}$  concentrations, in comparison to the other precursors.

#### 4.2. Validation of the SRR

The evaluation of the “Bell-shape” methodology is performed by comparing the results of the SRR with the validation scenario concentrations from the AQM model. Validation tests have been performed by reducing all precursors around 60% (between current legislation and maximum feasible reduction) over different European domains: two tests with country scale reductions (Poland and France) and two tests in which emissions have been reduced over smaller areas (considering reductions on six “regional” domains

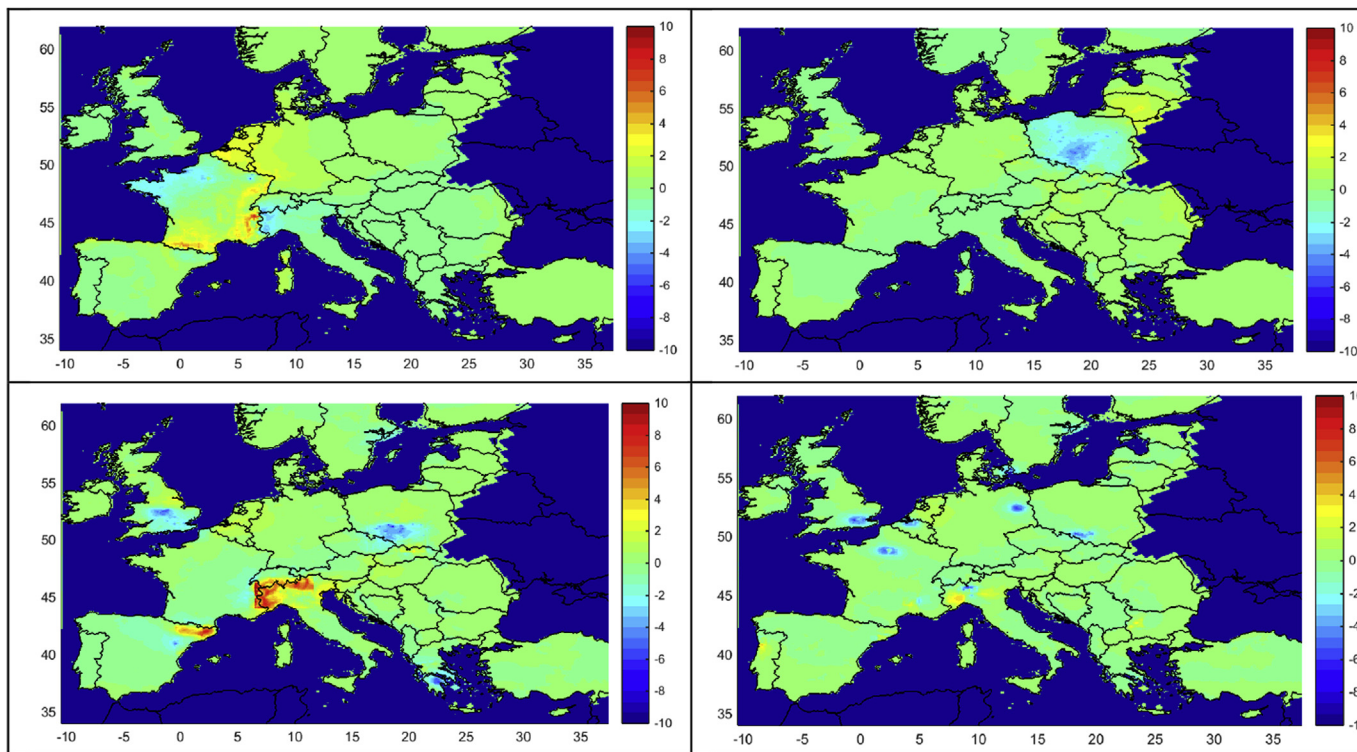


Fig. 7. Yearly  $PM_{25}$  concentration bias maps (SRR-AQM) for the 4 validation scenarios considered in Fig. 6, i.e. emission reductions applied over France (top left), Poland (top right), for all “regional” reduction scenarios (bottom left) and all “local” reduction scenarios together (bottom right).

and on thirteen “local” domains<sup>1</sup>).

Figs. 6 and 7 illustrate the comparison of the SRR and AQM results for these different validation scenarios.

As expected the main biases are localized in the areas where emission reductions are applied (Fig. 7). For the country reductions this bias is limited to  $\pm 9\%$  while it slightly increases for smaller areas (less than  $\pm 13\%$ ,  $\pm 8\%$  for the “regional” and “local reductions”, respectively). Note also that the largest percentage errors do not occur for the largest biases.

Fig. 6 (top-left) shows the validation of the new approach for the French emission reduction case (to be compared with Fig. 2 with the multi-ring approach) while results for the Poland emission reduction case are shown in Fig. 7 (top-left).

The multi-ring approach described in Section 2 accounts for the distance as well as the direction between sources and receptors as it attributes different weights ( $a_{kj}^p$ ) to S-aggregations which are distributed all around one receptor cell (Fig. 1). But this approach reduces the number of unknowns on the detriment of spatial resolution and ranges. Indeed, source cells are aggregated over a limited number of entities which become very large with distance and only cover a limited area around the receptor cell. In comparison, the “Bell-shape” approach does not account for direction between sources and receptors as it uses a symmetric function to calculate the coefficients  $a_{ij}^p$  (Equation (4)). On the other hand, spatial resolution is not degraded with distance as source cells are not aggregated. As previously mentioned, the “Bell-shape”

produces lower errors ( $\pm 9\%$ , Figs. 6–7) for the French test than the multi-ring ( $\pm 16\%$ , Fig. 2), highlighting the priority to be given to the quality of the discretization with distance rather than capturing directionality. Note that this conclusion does not hold for shorter term averages for which directionality may become more important.

All validation tests show the same level of performance for the “Bell-shape” SRR when compared to the full AQM. The main advantage of this approach resides in its spatial flexibility. Indeed emission reductions can be applied a posteriori on any geographical area, independently from the training simulations. In addition, the proposed method only requires a very limited number of simulations for training which makes it easy to set-up for any domain of interest.

## 5. Conclusions

In this work we further developed the SRR approach proposed in C2015. Already satisfactory from the point of view of “speed”, “set-up”, “accuracy” and “robustness”, the methodology has been shown to bear some limitations in terms of “spatial flexibility”, an aspect that has been improved in this work. The updated methodology is based on a cell-to-cell relationship, in which a bell-shape function links emissions to concentrations. Maintaining a cell-to-cell relationship was shown to be the key element needed to ensure spatial flexibility, while the sliding approach to link emissions and concentrations guarantees a “light” set-up phase (reduced number of simulations required for the training). This “light training” and gain in “spatial flexibility” is obtained at the expense of speed as cell-to-cell relationships imply a much larger number of operations in the SRR. This time is however limited to one minute on currently available computers. It is unfortunately not straightforward to compare the accuracy of the proposed

<sup>1</sup> “Regional” domains apply emission reduction over areas of  $140 \times 140 \text{ km}^2$  surrounding the cities of Katowice, Milan, London, Barcelona, Athens and Stockholm. “Local” domains apply emission reduction over areas of  $35 \times 35 \text{ km}^2$  surrounding the cities of Katowice, Milan, London, Barcelona, Athens, Stockholm, Antwerp, Porto, Paris, Clermont-Ferrand, Berlin, Copenhagen and Sofia.

approach with existing ones, such as RIAT+, GAINS, etc ... because these methodologies work with different input data (e.g. AQM simulations).

In this work we chose to compare the concentration delta obtained with the SRR to those obtained with the AQM (comparing delta is more challenging than comparing absolute values). Because spatial flexibility was the main focus, the validation has been repeated on different areas of different sizes (countries, regions, province throughout Europe) for precursors reduced independently or contemporarily. All runs showed the accuracy to be around 10%. The proposed methodology has also been shown to combine accuracy and robustness with a two-step approach for the estimation of the bell-shape coefficients.

In summary the “Bell-shape” SRR allow assessing the impact on air quality of emission scenarios applied over any given area in Europe (regions, set of regions, countries), provided that few AQM simulations are performed for training. Computation time for one scenario is around one minute (for a Europe wide domain) while accuracy is high.

While the approach has been developed with a specific model, a specific resolution and over a specific area, its application to other models and areas is straightforward.

The level of performance of the proposed methodology is very satisfying for annual average concentrations of PM<sub>2.5</sub>, PM<sub>10</sub> and NO<sub>2</sub> but still need to be improved for O<sub>3</sub>, especially if shorter time periods are considered (e.g. summer). Future efforts will consist in accounting for non-linearity (e.g. interactions among precursors) which have been shown to become more important in such cases (Thunis et al., 2015).

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