Atmospheric Environment 382 (2026) 122183

Contents lists available at ScienceDirect

Atmospheric Environment

journal homepage: www.elsevier.com/locate/atmosenv

High-resolution Atmospheric Transfer Matrices for policy-oriented air quality assessment in Italy: the new GAINS-Italy model

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HIGHLIGHTS

- High-resolution second-order Atmospheric Transfer Matrix developed for GAINS-Italy.
- Non-linear atmospheric responses are selectively captured with quadratic terms.
- High-resolution ATMs reproduce CTM results with errors within a few percent.
- Fast and reliable integrated modelling is enabled to support air quality planning.

ARTICLE INFO

Keywords:
Atmospheric transfer Matrices
Integrated assessment Modelling
Air quality Modelling
GAINS
Chemical transport Models
Air quality planning

ABSTRACT

Atmospheric Transfer Matrices (ATMs) are source-receptor relationships, widely applied in integrated assessment modelling to rapidly estimate air quality responses to emission changes, supporting policy design and evaluation. They enable fast policy-oriented analyses that would otherwise require computationally demanding Chemical Transport Model (CTM) simulations. However, the traditional linear formulation of ATMs may be insufficient for indicators influenced by non-linear atmospheric chemistry, especially at high spatial resolution and for secondary pollutants, or to manage air quality responses to large variations of input emissions.

In this study, we present an extended ATM framework for Italy at 4 km resolution incorporating selected second-order terms to account for non-linear responses. The matrices are developed using CTM simulations and implemented within the GAINS-Italy model. Their performance is evaluated against full CTM runs for multiple air quality indicators relevant to national air quality plans. Results show that first-order ATMs are adequate for quasi-linear processes, while the inclusion of second-order terms improve agreement with CTM simulations for non-linear indicators, reducing discrepancies within few percent. The proposed approach enables near-real time scenario screening, while preserving the spatial structure and magnitude of CTM responses providing a practical tool to support national and regional air quality planning.

1. Introduction

Integrated Assessment Models (IAMs) are essential tools in air quality and climate policy analysis, allowing systematic exploration of a wide range of emission scenarios and cost-effectiveness evaluation of mitigation strategies under complex constraints (Clappier et al., 2015; Colette et al., 2022; Pisoni et al., 2017). A core challenge in IAMs is reliably representing the relationship between changes in emissions and resulting impacts on air quality indicators (e.g., pollutant concentrations and deposition). While Chemical Transport Models (CTMs) provide detailed process-based simulations of atmospheric chemistry and transport, their high computational costs limit their direct use in IAMs, especially when high number of scenarios must be evaluated for policy (Van Dingenen et al., 2018).

To reconcile the needs for accuracy and for efficiency, reduced-form modelling approaches have been developed. Among these, Atmospheric Transfer Matrices (ATMs) and other source–receptor representations enable fast calculation of air quality responses to emission changes by

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https://doi.org/10.1016/j.atmosenv.2026.122183
Received 24 February 2026; Received in revised form 15 May 2026; Accepted 14 June 2026
Available online 15 June 2026

linearizing or approximating the CTM response around a baseline scenario (Amann et al., 2011; Pisoni et al., 2019). Unlike purely data-driven machine learning surrogates, ATMs retain a physical interpretation of source–receptor sensitivities, which is valuable for policy analysis, transparency, and scenario attribution (Yang et al., 2024).

The GAINS (Greenhouse Gas and Air Pollution Interactions and Synergies) model, developed at IIASA (International Institute for Applied Systems Analysis), is a widely used integrated assessment framework (https://gains.iiasa.ac.at/models/gains_models3.html) that evaluates cost-effective strategies for simultaneously reducing air pollution and greenhouse gas emissions across multiple sectors and regions (Amann et al., 2011, 2020). It integrates emission inventories, atmospheric source-receptor relationships, and impact assessment modules for human health, ecosystem and climate into a single modelling chain, enabling the identification of efficient emission control pathways under multi-pollutant, multi-effect constraints. It has been extensively applied to support major international and European policy processes, including the revision of the Gothenburg Protocol under the UNECE Convention on Long-range Transboundary Air Pollution (CLRTAP) and the development of EU emission reduction targets under the National Emission Ceilings (NEC) Directive.

GAINS-Italy is the national adaptation of this framework, developed jointly by ENEA (Italian National Agency for New Technologies, Energy and Sustainable Development) and IIASA.

It extends the GAINS system with an Italian specific modelling chain that includes: (i) national emission disaggregated at regional and sectoral level; and (ii) atmospheric transfer matrices derived from high resolution Chemical Transport Model. GAINS-Italy has been applied to support the developments of the Italian National Air Pollution Control Programme (NAPCP) under the NEC Directive (Ciucci et al., 2016; D'Elia et al., 2009; Piersanti et al., 2021). The model is available online at the following link https://gains-italy.enea.it/gains4/IT4/index.login.

Within the GAINS modelling suite, ATMs have been widely applied at regional and continental scales to support integrated assessment (Liu et al., 2025; Purohit et al., 2026). However, existing GAINS ATM implementations typically rely on coarse spatial resolutions and linear approximations, which may be insufficient when higher resolution or non-linear chemical responses (e.g., ozone and secondary particulate matter formation) significantly influence outcome estimates (Briganti et al., 2021; de Meij et al., 2024).

Recent advances in surrogate modelling techniques, including the use of sparse regression and physics-informed emulators, highlight opportunities for capturing non-linear dynamics with reduced computational cost (Guo et al., 2024; Yang et al., 2024). Nonetheless, systematic methodologies for constructing ATMs that balance computational feasibility, spatial detail, and chemical realism remain under-developed, particularly for national-scale domains.

In this paper, we present a new formulation of Atmospheric Transfer Matrices for GAINS-Italy (Ciucci et al., 2016; D'Elia et al., 2018; Piersanti et al., 2021), developed at 4 km horizontal resolution and calibrated using the state-of-the-art CTM within the MINNI atmospheric modelling framework (Italian National Integrated Model to support the international negotiation on atmospheric pollution, Ciucci et al., 2016; D'Elia et al., 2021, 2009; Mircea et al., 2014).

The focus of the present manuscript is on the systematic construction of the ATMs, the evaluation of approximation strategies (linear vs. second-order) and a sensitivity analysis of model configuration choices relevant to IAM implementations. While national-scale ATM implementations have been developed for several European countries and regions (e.g., the SHERPA tool based on EMEP MSC-W, (Pisoni et al., 2019); TM5-FASST at global scale, (Van Dingenen et al., 2018); the ACT surrogate model, (Colette et al., 2022), GAINS-Italy offers a distinctive combination of: (i) a 4 km horizontal resolution, significantly finer than most continental-scale implementations; (ii) a systematic evaluation of first- and second-order terms across a comprehensive set of air quality indicators; and (iii) direct integration with the Italian national regulatory framework (National Emission Ceilings Directive, Air Quality Plans). The methodology described here is general in its nature and, in principle, transferable to other national domains where high-resolution CTM simulations are available, providing a structure for similar national-scale ATM developments.

In the context of the development, assessment and implementation of the National Air Pollution Control Plan (for the National Emission Ceilings, Directive, NEC (EC, 2016), and Air Quality Plans (EC, 2024), there is an increasing demand for high-resolution tools capable of exploring several emission control scenarios. By explicitly analysing the trade-off between linear and second-order approximations at 4 km resolution, this work provides a practical guidance for constructing transfer matrices suitable for national-scale integrated assessment applications, bridging detailed CTM simulations and decision-support tools.

2. Methods

2.1. Conceptual framework of the atmospheric transfer matrices

Atmospheric Transfer Matrices (ATMs) are reduced-form models designed to approximate the response of air quality indicators to changes in precursor emissions. Within integrated assessment modelling frameworks, ATMs enable fast-response calculations that would otherwise require computationally expensive Chemical Transport Model (CTM) simulations.

In their standard formulation, ATMs assume a linear relationship between emission perturbations and air quality responses:

$$\Delta y = M \cdot \Delta E \tag{1}$$

where Δy represents changes in air quality indicators and ΔE represents emission changes relative to a baseline configuration (Amann et al., 2011), while M represents the matrix coefficients. This formulation assumes that the CTM response near the baseline scenario is approximately linear. However, chemical non-linearities — particularly for ozone and secondary PM formation — can reduce the accuracy of linear ATMs for when emission perturbations are large (de Meij et al., 2024).

To address these limitations, the present study adopts both first-order (linear) and second-order (quadratic) ATM formulations, explicitly assessing the relevance of non-linear terms for different pollutant–precursor combinations at high spatial resolution.

2.2. Mathematical formulation of first- and second-order ATMs

Air quality indicators (I) are expressed as functions of emission variations (ΔE) of GAINS precursors around a reference scenario. The basic idea is to evaluate the GAINS indicators (I) at the grid cell level (x), using a Taylor expansion centred on a baseline scenario, defining them as functions of emission variations of five GAINS precursors (k) on selected geographic areas (j, twenty Italian regions), as follows:

$$I(x,i) = I_{base}(x,i) + \sum_{j=1}^{20}\sum_{k=1}^{5} \Delta E(j,k) \cdot T^{(1)}(x,i,j,k) + +\frac{1}{2}\sum_{j'=1}^{20}\sum_{k'=1}^{5}\sum_{j=1}^{20} \times \sum_{k=1}^{5} T^{(2)}(x,i,j,k,j',k') \cdot \Delta E(j,k) \cdot \Delta E(j',k') + O(\Delta E^3) \tag{2}$$

where I_{base} denotes the indicator computed for the base scenario, x is the grid point, $T^{(1)}$ and $T^{(2)}$ are first- and second-order ATM coefficients, respectively, and the summations span emission precursors and source regions. Emission variations refer to the main GAINS precursors: sulphur oxides (SO_X); nitrogen oxides (NO_X); particulate matter with a diameter under 10 (PM_{10}) and 2.5 ($PM_{2.5}$) μm; non methanic volatile organic compounds (NMVOC), and ammonia (NH_3). The air quality indicators considered include sulphur (TS), nitrogen (TN) and reduced nitrogen (TNH) yearly deposition; yearly average concentrations of particulate matter (PM_{10} and $PM_{2.5}$) and nitrogen dioxide (NO_2); ozone (O_3) metrics for impact on human health (annual sum of maximum daily 8-h ozone means over 35 ppb and 0 ppb, SOMO35 and SOMO00, respectively) and vegetation (accumulated exposure over an hourly threshold of 40 ppb for forest and crops, AOT40F and AOT40C, respectively). Details on the method of calculation of $T^{(1)}$ and $T^{(2)}$ are presented in section A of the SM.

All GAINS indicators were calculated on annual basis. To assess the potential need for the second-order term, $T^{(2)}$, a preliminary set of full chemical test simulations was performed. The results of these linearity tests, referred to annual averaging period, are summarized on Table 1. Bold text is to underline leading dependencies. Sulphur depositions (TS) are depending only on SO_X emissions and show a perfect linearity. Total nitrogen deposition (TN) is the sum of oxidised and reduced nitrogen and depends on NO_X and NH_3 emissions. A sort of anticorrelation is seen between NO_X and NH_3, due to an antagonistic effect: abatement of ammonia emissions leaves more HNO_3 in the gas phase, which is deposited effectively in the form of nitrates, especially in areas with higher emission density, corresponding to the points with the greatest deposition variations. Reduced nitrogen deposition (TNH) depends on NH_3 emissions only. Ozone tests showed a non-linear correlation between SOMO35, AOT40F, AOT40C and NO_X that cannot be neglected in the ATMs. PM concentrations depend on emission from all precursors (PM_{10}, SO_X, NO_X, NH_3 and NMVOC). Non-linear relations have been found for NO_X and NH_3, while PM10 and PM2.5 concentrations depend mainly on primary PM. NO_2 concentrations principally depend on NO_X emissions, to a lesser extent on NMVOC emissions in a non-linear way, and they tend to decrease by reducing its emission precursors.

All analyses were confirmed by seasonal tests, with no major differences found.

Looking at Tables 1 and it is clear that the main contributions from precursor variations are all linear, with NMVOCs appearing to have a less pronounced impact compared to other precursors. This is an essential consideration in the perspective of defining the ATMs structure, because it allows neglecting the second order cross terms in the Taylor expansion around base case scenario, while maintaining sufficient reliability. The reason for deciding to consider second-order terms is then to achieve a more "robust" formulation of the transfer matrices, especially in cases where scenarios are characterized by substantial emission reductions and are therefore significantly distant from the base case. In such instances, the resulting errors could even be comparable to the typical uncertainties found in MINNI chemical transport model on an annual basis (Colette et al., 2025, https://regional-evaluation.atmosphere.copernicus.eu/pages/evaluation/?project=cams2-83&model=MINNI). Such 2nd order terms allow the ATM formulation to capture non-linear responses while preserving computational efficiency. Cross terms involving different precursor species and source regions were neglected, assuming that second-order interactions are predominantly local at the adopted spatial resolution. This assumption was explicitly tested and found to have negligible impact on ATM performance (Briganti et al., 2021). More details are provided in Section 3.7. For further details and graphical representation see also Section B and Figure B1, respectively, of the SM.

Sensitivity experiments were conducted to (i) optimize model configuration parameters and (ii) assess the validity of linear and second-order approximations. Emission perturbations of −25% and −50% were applied independently to each GAINS precursor for selected regions representative of different Italian climatic and emission conditions.

Linearity was evaluated by comparing indicator responses to proportional emission reductions, allowing identification of non-linear behaviours and guiding the inclusion of second-order terms. These analyses demonstrated that most indicator-precursor relationships are predominantly linear, while non-linear responses are relevant for ozone indicators, NO_2 concentrations, and secondary PM formation.

Based on these results, second-order terms were selectively retained only where they significantly improved agreement with CTM simulations. It is important to clarify that the sensitivity analyses described in the present section were conducted as part of this study, building upon and extending preliminary investigations reported in Briganti et al. (2021). Specifically, the validation of the cross-term neglect assumption and the systematic identification of non-linear precursor–indicator pairs across all GAINS-Italy indicators are original contributions of the present work, while the earlier study provided a proof of concept at coarser spatial resolution focused on computational feasibility.

Detailed sensitivity plots and extended analyses are reported in the SM, while in (Briganti et al., 2021) detailed information on the computational feasibility could be found.

In summary, ATM coefficients were estimated from a set of CTM sensitivity runs in which each precursor emission is perturbed independently by −25% and −50% relative to the baseline scenario, for each of the 20 Italian administrative regions and each of the 5 GAINS precursors (SO_X, NO_X, primary PM, NH_3, NMVOC). This required a total of 201 annual CTM simulations per meteorological year (1 base case + 200 perturbation runs, two runs for each precursor and region). First-order ($T^{(1)}$) coefficients were derived as finite-difference approximations of the first derivative of each air quality indicator with respect to each emission precursor. Second-order ($T^{(2)}$) coefficients were estimated from the curvature of the indicator response between the two perturbation levels: $T^{(2)} = (\Delta I\,(2\alpha) - 2\Delta I(\alpha))/(\alpha \cdot \Delta E)^2$, where α denotes the fractional perturbation and ΔE the emission change.

Full mathematical derivations and the complete set of sensitivity figures are provided in SM, sections A and B.

2.3. The MINNI integrated assessment modelling

ATMs were derived from a set of CTM simulations performed with

Table 1
– Qualitative summary of linearity tests results for annual averaged period. Bold text underlines the leading dependencies.

		Precursors				
		SO_X	NO_X	PM10	NH_3	NMVOCs
GAINS	TS	**linear**	absent	negligible	absent	absent
Indicators	TN	absent	**linear**	absent	anti-correlated, secondary, slightly non-linear	negligible
	TNH	linear, secondary	quasi-linear, secondary	absent	**linear**	absent
	O_3 as SOMO35, SOMO00 and AOT40F	absent	**quasi-linear**	absent	absent	linear, secondary
	PM2.5	linear, secondary	quasi-linear, secondary	**linear**	quasi-linear, secondary	linear, secondary
	PM10	linear, secondary	quasi-linear, secondary	**linear**	quasi-linear, secondary	linear, secondary
	NO_2	absent	**linear**	absent	absent	linear, secondary

the Atmospheric Modelling System (AMS) of the MINNI model (see Fig. 1), an Integrated Modelling System covering Italy, which links atmospheric science with the economics of emission abatement measures and policy analysis. MINNI has been extensively applied to national air quality assessments (D'Elia et al., 2018; D'Isidoro et al., 2022; Piersanti et al., 2021; Russo et al., 2021).

AMS is based on the FARM (Flexible Regional Atmospheric Model, Gariazzo et al., 2007; Silibello et al., 2008)) chemical transport model, driven by meteorological fields from the WRF (Weather Research and Forecasting, Skamarock et al., 2019) model and surface-atmosphere exchange parameters from SURFPRO (SURFace-atmosphere interface PROcessor).

Simulations were performed over the Italian domain at a horizontal resolution of 4 km, using the 2015 meteorological year and the 2030 "With Measures" emission scenario developed within GAINS-Italy (Piersanti et al., 2021). This configuration represents a compromise between spatial detail and computational feasibility, consistent with the objectives of integrated assessment applications.

The 2030 "With Measures" (WM) scenario includes the full implementation of currently adopted national and EU-level emission reduction policies, comprising targets from the NEC Directive and sector-specific measures for transport, energy, agriculture, and industry as described in the Italian National Air Pollution Control Programme (Piersanti et al., 2021). This scenario was selected as it represents a realistic near-future policy pathway and provides a challenging test case for ATM calibration. The simulations were performed using offline nesting. Boundary conditions were extracted from a coarse-resolution simulation (20 km) covering the study domain, which in turn was driven by CAMS global boundary conditions. The emission inputs are derived from the GAINS-Italy national emission inventory, spatially disaggregated at 4 km resolution using activity data and sector-specific spatial proxies. The meteorological driver for the FARM code is provided by the WRF model, utilizing CAMS ERA5 reanalysis fields for initial and boundary conditions. Furthermore, WMO (World Meteorological Organization) observation spectral nudging was applied to filter out small-wavelength features. Following the WRF output, the SURFPRO meteorological post-processor calculates the PBL (Planetary Boundary Layer) height, the dry deposition velocities for gas species and micrometeorological parameters, as well as natural emissions (sea salt and dust), which complete the FARM input dataset. More details about model formulation and features can be found in D'Elia et al. (2021). The ATM performance is evaluated by comparing ATM-based reconstructions against full CTM sensitivity simulations for each precursor perturbation (−25% and −50%) and further validated against a full CTM run for an independent emission scenario (Section 3.8). Performance is quantified using relative differences (%) computed grid-cell by grid-cell across the Italian domain.

AMS-MINNI takes advantage of the computing resources provided by CRESCO/ENEAGRID High Performance Computing infrastructure (Iannone et al., 2019).

3. Results

This section presents the performances of the ATMs developed for GAINS-Italy by comparing ATM-based reconstructions with full CTM simulations (AMS run in the following figures) based on the linearity sensitivity analyses explained in the previous section. ATMs were elaborated for the meteorological years 2004, 2005, 2015. In addition, an "average" matrix was constructed by computing the mean coefficients across the three selected annual datasets, providing a representative synthetic matrix suitable for subsequent modelling and comparative analyses. These three years were selected to represent contrasting atmospheric conditions relevant for Italian air quality, so to capture distinct climatic profiles: 2015 is characterized by high-

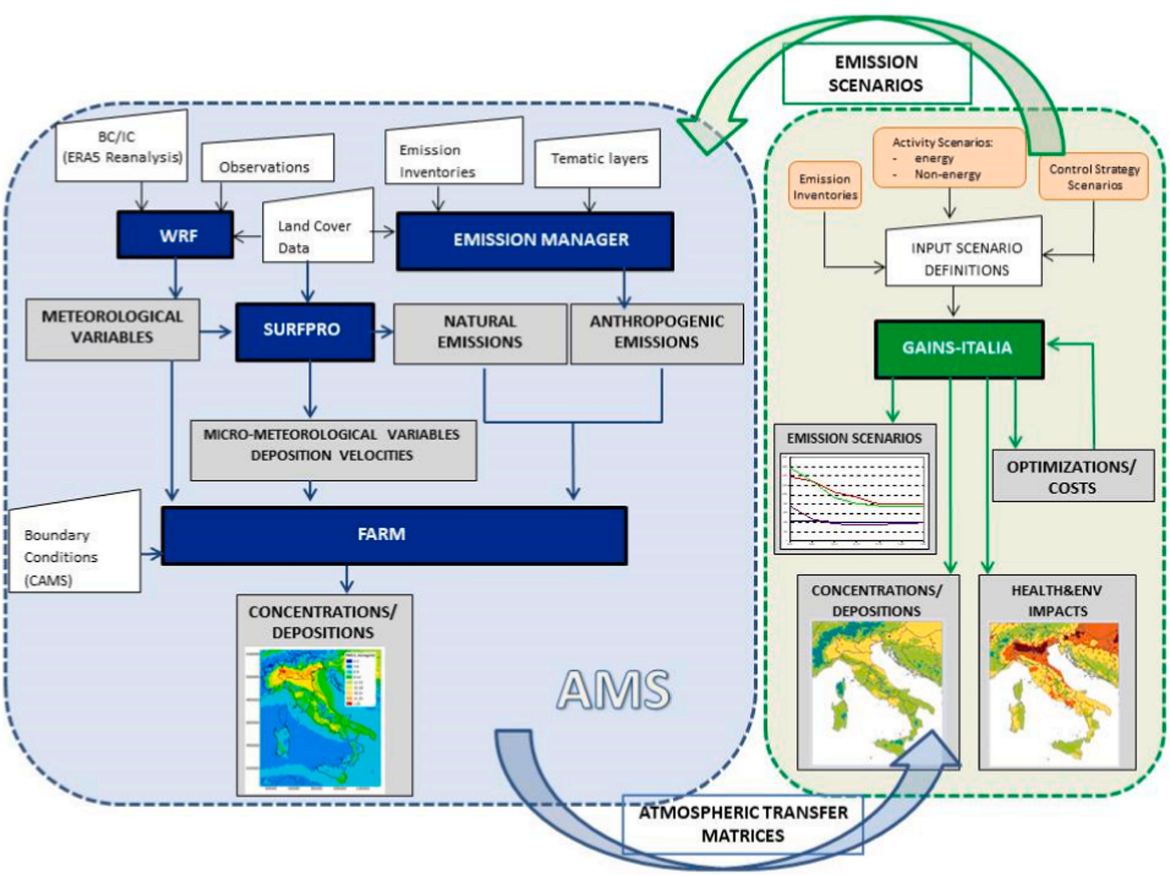

Fig. 1. – The MINNI integrated assessment modelling.

temperature, while maintaining high rainfall levels across the study area, considered as a more representative year for future climate change; 2005 represent cooler, high-precipitation year; 2004 serves as a representative average. This selection is based on a preliminary statistical analysis of the 2003-2015 period and on their representativeness of interannual variability in precipitation patterns and summer temperature extremes, ensuring that the ATMs are evaluated across a range of meteorological regimes (see SM, Section C), and it is consistent with prior AMS-MINNI modelling studies (D'Elia et al., 2021; Mircea et al., 2014).

The results demonstrate that the Atmospheric Transfer Matrices exhibit robust and internally consistent sensitivity to meteorological forcings. Specifically, enhanced precipitation levels lead to a dampened increase in sulphur and nitrogen deposition, reflecting the expected dilution and scavenging dynamics associated with wet removal processes. Conversely, higher air temperatures are associated with elevated ozone concentrations and a concurrent reduction in oxidised nitrogen deposition. This pattern is driven by the antagonistic photochemical mechanism through which NO_2 is preferentially consumed to sustain ozone production, thereby decreasing the atmospheric abundance of nitrate and nitric acid available for deposition. All the meteorological years are discussed in detail in the SM, while in the current section the results relative to the meteorological year 2015 and to the test with a 25% reduction of all precursors across all regions are presented.

The following results are organized by air quality indicator, highlighting the role of linear and second-order terms and identifying the conditions under which non-linearities need to be explicitly accounted for.

3.1. Oxidised sulphur deposition

Annual average deposition of oxidised sulphur (TS) exhibits an exclusively linear dependence on SO_X emissions, consistently with atmospheric chemistry considerations. Consequently, TS represents a benchmark case for the ATM framework, where a first-order approximation is expected to be fully adequate. Therefore, the following TS formulation has been adopted:

$$d_{TS} = d_{TS,ref} + \sigma \bullet \Delta s$$

Where σ is the transfer matrix associated with SO_X emissions, $d_{TS,ref}$ denotes the base-case deposition, and Δs represents regional emission perturbations.

Fig. 2 shows an excellent agreement between ATM-based reconstructions and full CTM simulations, with relative differences below 0.1% over the entire domain. This result confirms that, for purely linear regimes, high-resolution ATMs can reproduce CTM responses with negligible loss of accuracy.

3.2. Total nitrogen deposition

Total nitrogen deposition (TN) depends on both NO_X and NH_3 emissions, with a linear response to NO_X and a weak non-linear response to NH_3, represented as:

$$d_{TN} = d_{TN,ref} + \alpha_1 \bullet \Delta h + 0.5\ \alpha_2 \bullet (\Delta h)^2 + \nu \bullet \Delta n$$

Where α_1 and α_2 describe, respectively, the first- and second-order dependence on ammonia emissions (h), and ν represents the linear dependence on NO_X emissions (n).

As shown in Fig. 3, the first-order approximation already provides a good representation of TN variations, with relative differences generally within 0.8%. The inclusion of the second-order term for NH_3 further reduces discrepancies, lowering the maximum relative error to approximately 0.5%. These results indicate that, for TN, non-linear effects are moderate and second-order terms act mainly as a refinement rather than a structural correction.

3.3. Reduced nitrogen deposition

Reduced nitrogen deposition (TNH) shows a more complex dependence on emission precursors. While TNH varies linearly with NH_3, SO_X and NMVOC emissions, its response to NO_X is non-linear. Accordingly, TNH is expressed as:

$$d_{TNH} = d_{TNH,ref} + \nu_1 \bullet \Delta n + 0.5\ \nu_2 \bullet (\Delta n)^2 + \alpha \bullet \Delta h + \omega \bullet \Delta o + \sigma \bullet \Delta s$$

Where ν_1 and ν_2 describe the first- and second-order dependence on NO_X emissions (n), α, ω and σ represents, respectively, the linear dependence on NH_3 (h), NMVOC (o) and SO_X (s) emissions.

Fig. 4 demonstrates that the inclusion of the second-order term for NOx improves ATM performance, reducing relative discrepancies to below 0.3% over the Italian domain. For TNH, second-order terms are therefore required to achieve sub-percent accuracy.

3.4. Ozone indicators

Ozone-related indicators exhibit markedly different behaviour compared to deposition variables, due to the intrinsically non-linear nature of tropospheric ozone chemistry. In the analysed scenarios, ozone indicators respond primarily to variations in NO_X and NMVOC emissions, with a pronounced non-linear dependence on NO_X reductions.

For all ozone-related indicators (O_3, SOMO35, SOMO00, AOT40F and AOT40C), the ATM formulation adopted is:

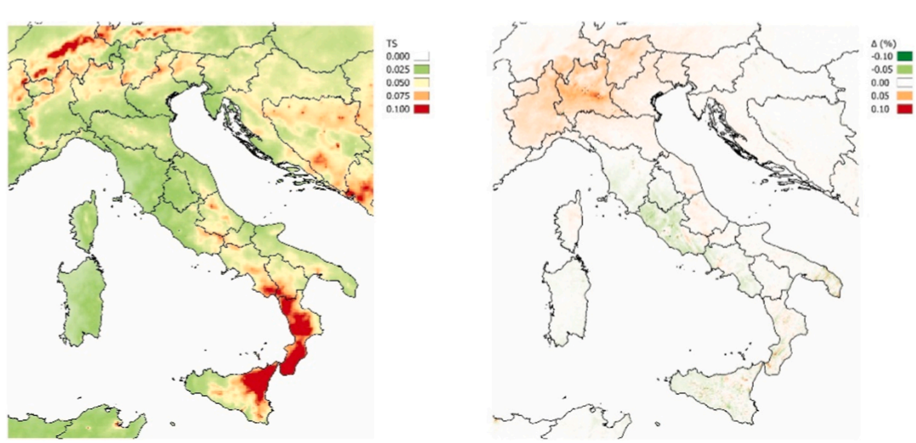

Fig. 2. – TS: annual averaged depositions for the base case (on the left, expressed in $mg \cdot m^{-2} \cdot h^{-1}$) and percentage differences (on the right) between ATMs and full AMS run.

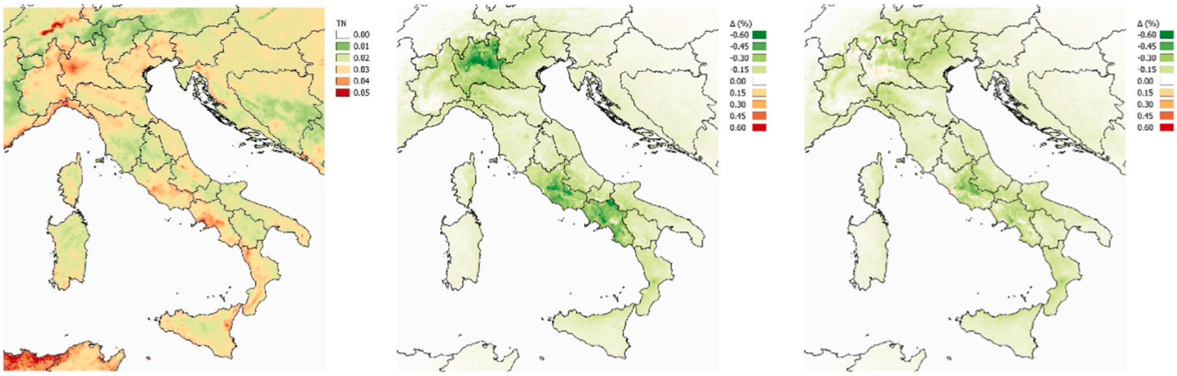

Fig. 3. – TN: annual averaged depositions for the base case (on the left, expressed in $mg \cdot m^{-2} \cdot h^{-1}$) and percentage differences with linear (centre) and 2nd order approximation (right) between ATMs and full AMS run.

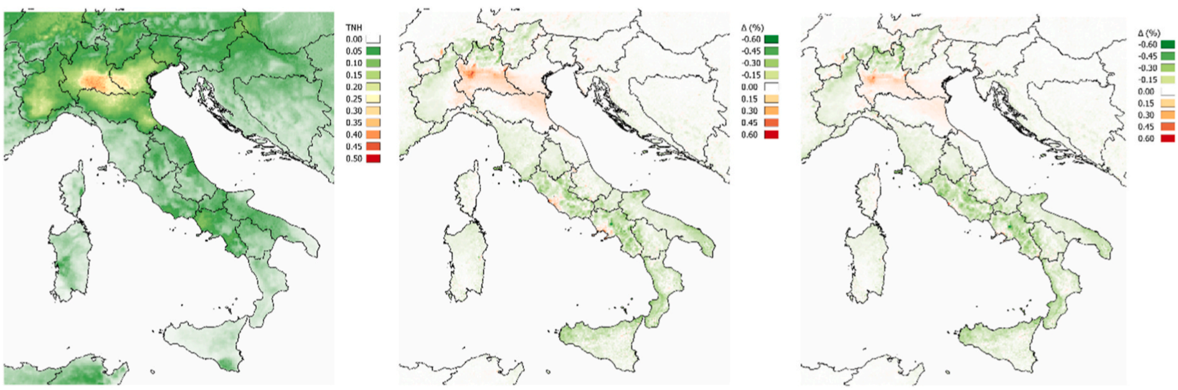

Fig. 4. – TNH: annual averaged depositions for the base case (on the left, expressed in $mg \cdot m^{-2} \cdot h^{-1}$) and percentage differences with linear (centre) and 2nd order approximation (right) between ATMs and full AMS run.

$$c_{O3} = c_{O3,ref} + \nu_1 \bullet \Delta n + 0.5\ \nu_2 \bullet (\Delta n)^2 + \omega \bullet \Delta o$$

Where ν_1 and ν_2 describe the first- and second-order dependence on NO_X emissions (n), and ω the linear dependence on NMVOC (o) emissions.

Figs. 5 and 6 show that first-order ATMs are insufficient to accurately reproduce CTM responses, particularly in regions characterized by elevated ozone levels. The introduction of the second-order term for NO_X substantially reduces absolute percentage differences and, in many areas, corrects the sign of the bias observed with the linear approximation. Overall, second-order ATMs are essential to represent ozone indicators under emission reduction scenarios relevant for policy analysis, and their performance is particularly robust for higher ozone values.

3.5. Concentrations of PM_{10} and $PM_{2.5}$

Particulate matter concentrations depend on all GAINS-Italy precursors. The response is linear with respect to primary PM, SO_X and NMVOC emissions, while non-linear behaviour is observed for NO_X and NH_3. The adopted formulation is therefore:

$$c_{PM} = c_{PM,ref} + \pi \bullet \Delta p + \nu_1 \bullet \Delta n + 0.5\ \nu_2 \bullet (\Delta n)^2 + \alpha_1 \bullet \Delta h + 0.5\ \alpha_2 \bullet (\Delta h)^2 + \sigma \bullet \Delta s + \omega \bullet \Delta o$$

Where ν_1 and ν_2 describe the first- and second-order dependence on NO_X

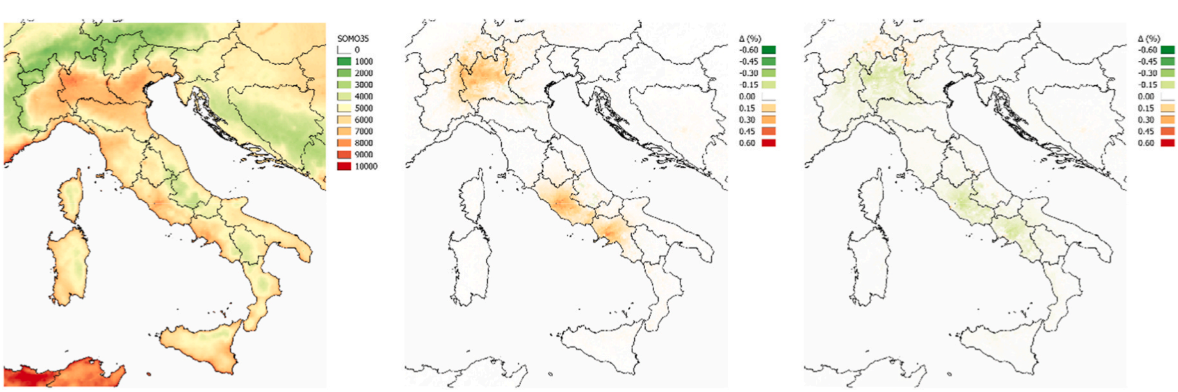

Fig. 5. – SOMO35: annual values for the base case (on the left, expressed in $\mu g \cdot m^{-3} \cdot day$) and percentage differences with linear (centre) and 2nd order approximation (right) between ATMs and full AMS run.

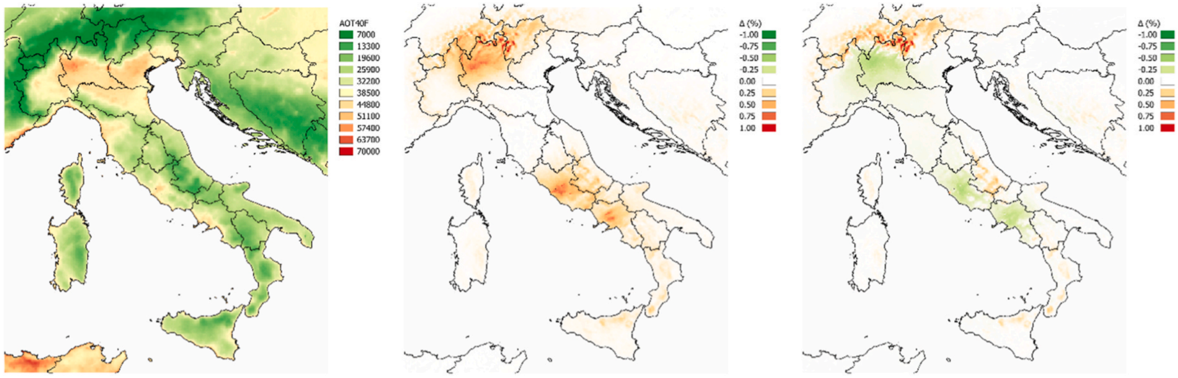

Fig. 6. – AOT40F: annual values for the base case (expressed in $\mu g \cdot m^{-3} \cdot h$) and percentage differences with linear (centre) and 2nd order approximation (right) between ATMs and full AMS run.

emissions (n), α_1 and α_2 the first- and second-order dependence on ammonia emissions (h), and π, ω and σ the linear dependence on, respectively, PM (p), NMVOC (o) and SO_X (s) emissions.

Figs. 7 and 8 compare linear and second-order ATM reconstructions against CTM simulations for PM_{10} and $PM_{2.5}$. The inclusion of second-order terms for NO_X and NH_3 emissions leads to a substantial improvement, markedly reducing relative discrepancies across the domain, particularly on the Po Valley, a well-known hotspot for secondary particulate matter formation (Clappier et al., 2021; Colombo et al., 2025; Thunis et al., 2021). These results indicate that, for PM, non-linear effects associated with secondary aerosol formation cannot be neglected, and second-order single-precursor terms capture most of the relevant non-linear response.

3.6. Concentrations of NO_2

NO_2 concentrations depend non-linearly on NO_X emissions and linearly on NMVOC emissions, similarly to ozone. The corresponding ATM formulation is:

$$c_{NO2} = c_{NO2,ref} + \nu_1 \bullet \Delta n + 0.5\ \nu_2 \bullet (\Delta n)^2 + \omega \bullet \Delta o$$

Where ν_1 and ν_2 describe the first- and second-order dependence on NO_X emissions (n), and ω the linear dependence on NMVOC (o) emissions.

As shown in Fig. 9, the introduction of the second-order term for NO_X significantly improves the agreement with CTM simulations. The results confirm that non-linearities play a relevant role also for NO_2, due to the non-linear photochemical equilibrium between NO and NO_2, and that their explicit representation is required for accurate ATM-based reconstructions.

3.7. Contribution of second-order cross terms

Different GAINS indicators exhibit different response structures. TN, ozone indicators and NO_2 depend linearly on NMVOC emissions and non-linearly on NO_X ones; therefore, second-order cross terms involving NO_X and NMVOC vanish by construction. This was confirmed by explicit calculations, which showed no differences between second-order formulations with and without cross terms.

TNH depends on multiple precursors but is non-linear only with respect to NO_X emissions; consequently, all second-order cross terms are negligible. In contrast, PM concentrations exhibit non-linear responses to both NO_X and NH_3 emissions, implying that a cross term ($\partial^2/\partial n \partial h$) could, in principle, be relevant. To assess its contribution, additional simulations were performed with simultaneous reductions of NO_X and NH_3 emissions.

Figs. 10 and 11 show that including the cross term does not improve ATM performance and, in some cases, worsens the reconstruction, likely due to the antagonistic interaction between NO_X and NH_3. Given the limited benefit and the additional computational cost, cross terms were deliberately excluded from the final ATM formulation. This choice represents a pragmatic compromise that enhances robustness and computational efficiency without degrading accuracy.

3.8. Comparison with a full CTM simulation for an independent scenario

To further assess the robustness of the proposed ATMs, a comparison was performed between ATM-based estimates and a full CTM simulation for an independent 2030 emission scenario (described in ISPRA, 2023) not used in the construction of the matrices. Both ATMs and the CTM simulation use the 2015 meteorological forcing. The comparison focuses

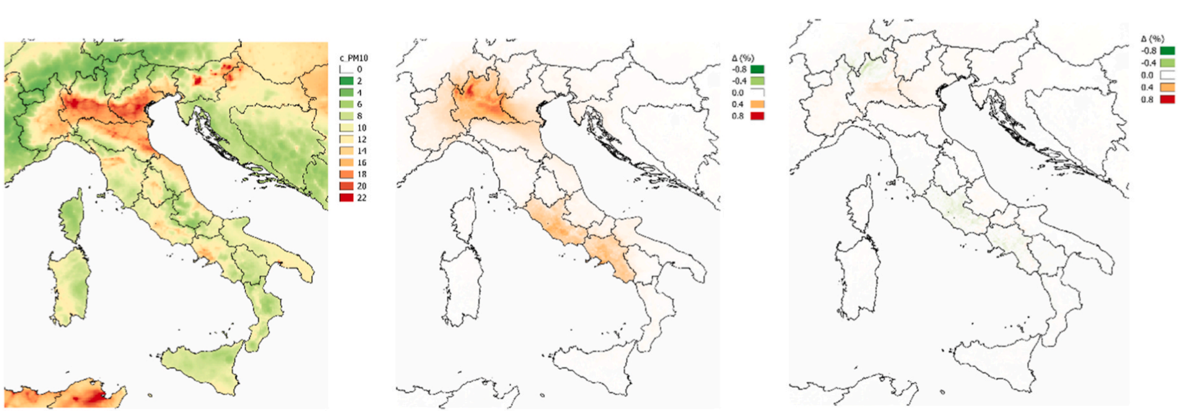

Fig. 7. – PM_{10}: annual averaged concentrations for the base case (on the left, expressed in $\mu g \cdot m^{-3}$) and percentage differences with linear (centre) and 2nd order approximation (right) between ATMs and full AMS run.

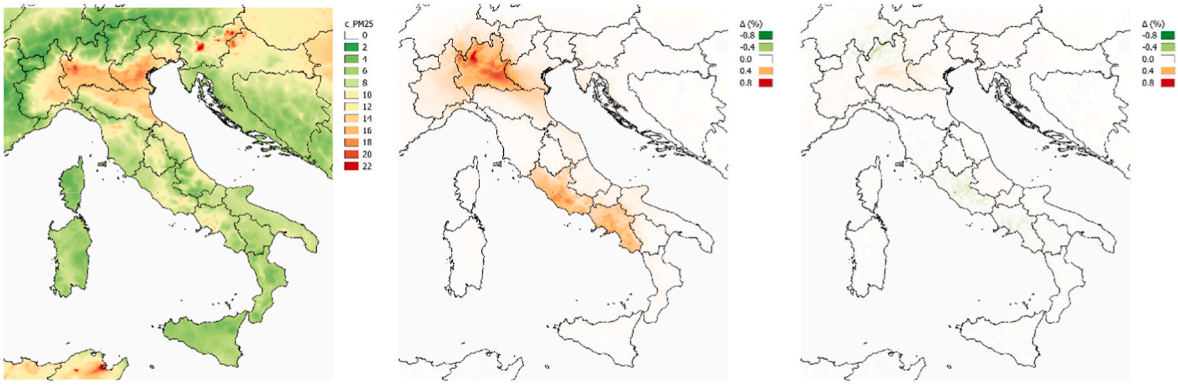

Fig. 8. – $PM_{2.5}$: annual averaged concentrations for the base case (on the left, expressed in $\mu g \cdot m^{-3}$) and percentage differences with linear (centre) and 2nd order approximation (right) between ATMs and full AMS run.

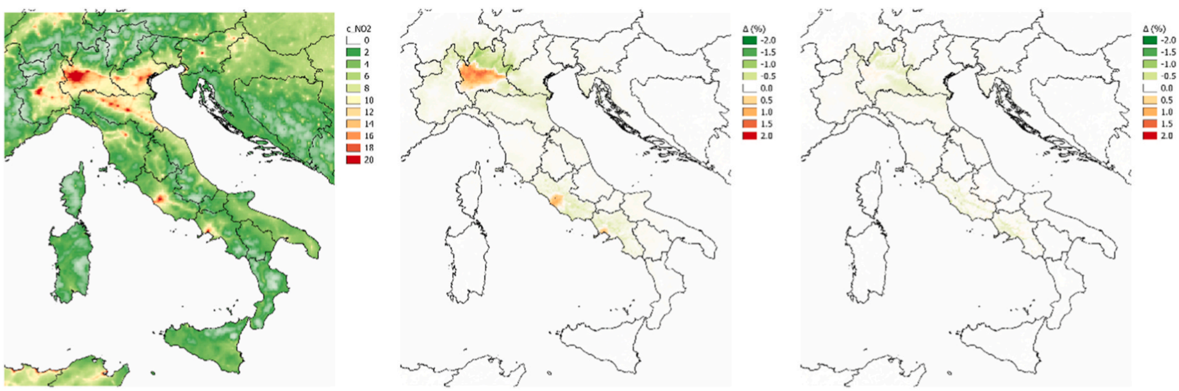

Fig. 9. – NO_2: annual averaged concentrations for the base case (on the left, expressed in $\mu g \cdot m^{-3}$) and percentage differences with linear (centre) and 2nd order approximation (right) between ATMs and full AMS run.

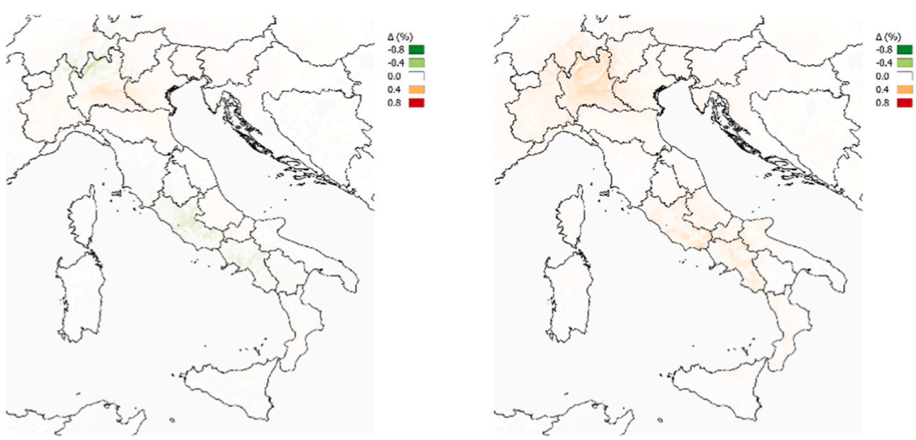

Fig. 10. – $PM_{2.5}$: comparison of matrices without cross term contribution (left) and the complete one.

on annual average concentrations of $PM_{2.5}$ and SOMO35, selected as representative indicators of secondary aerosol formation and non-linear photochemical processes.

Figs. 12 and 13 show the spatial distribution of $PM_{2.5}$ and SOMO35 concentrations, respectively, obtained from the full CTM simulation and the corresponding ATM-based reconstruction.

Overall, the ATM approach reproduces the main spatial patterns and gradients of both pollutants, correctly identifying high-concentration areas and regional-scale features relevant for air quality assessment.

Differences between ATM and CTM results are generally limited and spatially coherent, although local discrepancies may reach up to approximately 30% in specific areas for PM2.5 concentrations. These differences are primarily associated with regions characterized by strong emission gradients and secondary formation processes, where higher-order meteorological-chemical interactions play a relevant role. Notably, the largest discrepancies are not uniformly distributed and do not alter the relative ranking of regions or the identification of major pollution hotspots.

Despite these local deviations, the ATM-based results remain consistent in terms of order of magnitude and spatial structure, supporting the suitability of the proposed matrices as surrogate models for integrated assessment applications. The reached level of agreement

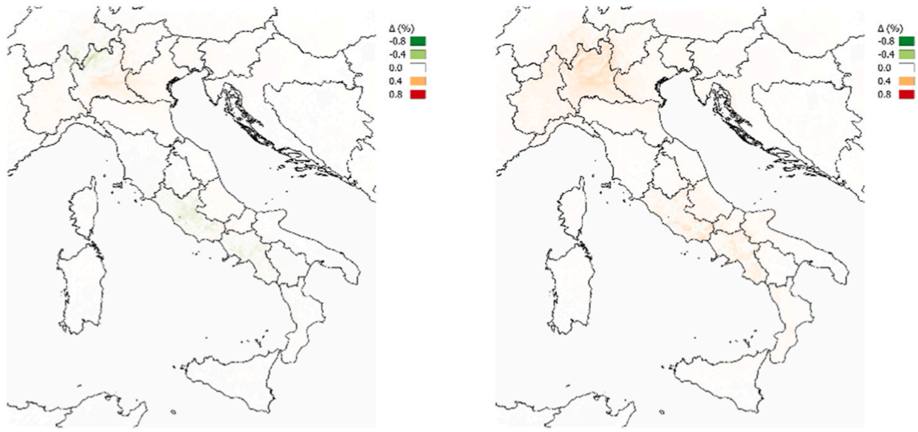

Fig. 11. PM_{10}: comparison of matrices without cross term contribution (left) and the complete one.

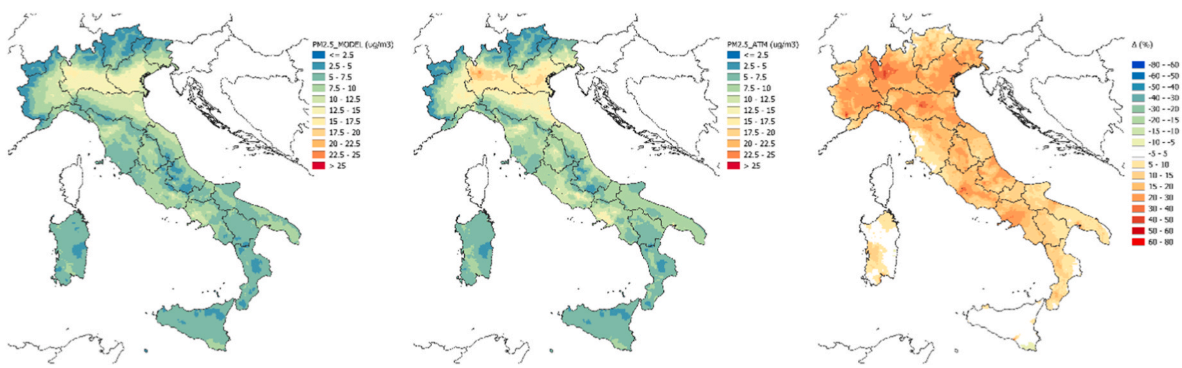

Fig. 12. Annual average concentrations of $PM_{2.5}$: 2030 CTM simulation (on the left, concentration values in μg/m^3), ATM simulation (centre, concentration values in μg/m^3), and percentage differences between ATM and full AMS run (on the right).

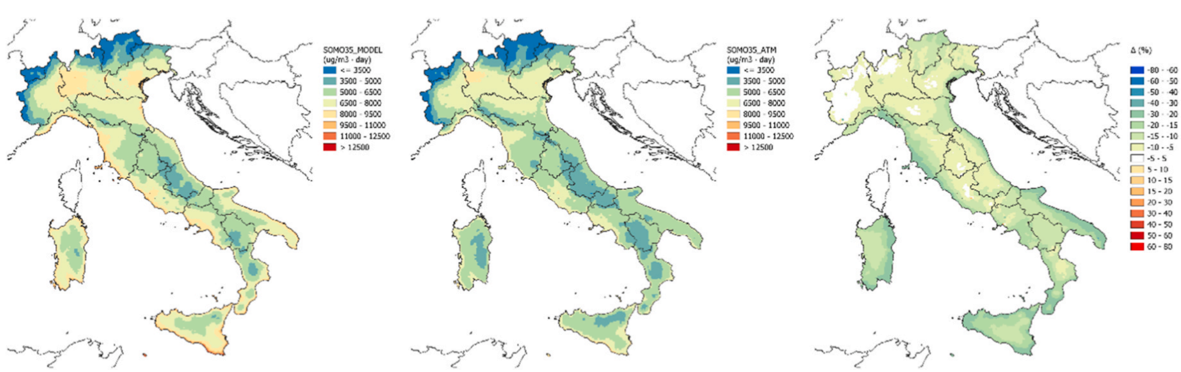

Fig. 13. Annual average concentrations of SOMO35: 2030 CTM simulation (on the left, concentration values in μg/m^3·day), ATM simulation (centre, concentration values in μg/m^3·day), and percentage differences between ATM and full AMS run (on the right).

indicates that ATMs can be reliably used for comparative policy analysis, without the need to repeat full CTM simulations. In this context, ATMs are so intended to support rapid scenario screening, rather than replace full CTM runs.

4. Discussion and conclusion

In this study, we present new high-resolution Atmospheric Transfer Matrices developed for Italy and implemented in the GAINS-Italy model.

Results demonstrate that the new developed ATMs can reproduce the response of a full chemical transport model on an annual average basis with high accuracy for a wide range of air quality indicators, relevant for national policy assessment. For indicators governed by essentially linear chemistry and deposition processes, such as oxidised sulphur deposition, first-order ATMs already provide an excellent approximation even at 4 km resolution, with discrepancies well below 1%. This confirms earlier findings on the robustness of linear source-receptor approaches for quasi-linear atmospheric processes (Amann et al., 2020; Van Dingenen et al., 2018). For pollutants influenced by non-linear atmospheric chemistry, the introduction of selected second-order terms leads to a systematic improvement of ATM performance. This is particularly evident for nitrogen deposition, ozone exposure indicators, particulate matter and nitrogen dioxide concentrations, where quadratic terms significantly reduce residual errors and improve spatial coherence with full CTM simulations. The relevance of non-linear terms varies markedly across indicators and precursors, reflecting the diversity of chemical regimes involved. Ozone-related indicators exhibit pronounced non-linear responses to NO_X emission reductions, especially in regions characterized by high precursor emissions and complex photochemical interactions. In these cases, second-order ATMs not only reduce absolute discrepancies but also improve the representation of high-concentration regimes, which are of primary relevance for health-related and vegetation-related metrics. For particulate matter, non-linearities associated with NO_X and NH_3 emissions are consistent with the chemistry of secondary inorganic aerosol formation. While second-order terms substantially improve the reconstruction of $PM_{2.5}$ and PM_{10} concentrations, the inclusion of cross terms between precursors was found to provide no additional benefit and, in some cases, to deteriorate model performance. This suggests that the dominant non-linear effects are adequately captured by single-precursor quadratic terms, and that higher-order interactions play a minor role within the explored emission reduction range. Overall, these results support a selective inclusion of non-linear terms, allowing ATMs to balance accuracy and computational efficiency.

A key requirement for policy-oriented applications is ATMs robustness under emission scenarios different from those used for calibration. To assess this aspect, the ATMs were applied to an independent 2030 emission scenario.

The comparison between ATM-based estimates and full CTM simulations under this independent scenario shows that the accuracy observed for the calibration scenario is largely preserved. Spatial patterns, concentration gradients, and depositions are consistently reproduced. Indicators characterized by stronger non-linear behaviour continue to benefit from the inclusion of second-order terms, confirming that the quadratic approximation captures intrinsic atmospheric responses rather than artefacts of a specific emission configuration.

From a policy perspective, the consistent performance obtained suggests that the matrices retain key features of CTM behaviour, that can be reliably applied to alternative policy-relevant emission pathways, within the range of the emission perturbations investigated, with the advantage that the ATM framework reduces computational time from days to near-instantaneous evaluation and enabling iterative scenario screening and optimisation analyses.

By preserving the main spatial and chemical features of CTM responses while drastically reducing the computational time, the new ATM framework provides an operational bridge between detailed atmospheric modelling and policy-oriented integrated assessment.

The performance of the ATMs developed in this study can be compared with analogous tools applied at different scales. TM5-FASST (Van Dingenen et al., 2018) achieves good agreement with full CTM simulations for $PM_{2.5}$ and O_3 at global and continental resolution, but does not systematically include second-order terms, and operates at much coarser spatial scales. The SHERPA tool (Pisoni et al., 2019), based on EMEP MSC-W, applies linear source-receptor relationships at European scale, while the ACT (Air Control Toolbox) surrogate model (Colette et al., 2022) explicitly includes non-linear terms up to second order for sector-level emission reductions, and reports relative errors below 2% for most pollutants and conditions, consistent with our findings. The present results suggest that, at 4 km resolution, second-order terms become increasingly important for secondary pollutants, consistent with the findings of (Thunis et al., 2021) for the Po Valley, which demonstrated marked non-linear responses of $PM_{2.5}$ to combined NO_X and NH_3 reductions. Compared to purely data-driven surrogate models (Guo et al., 2024; Yang et al., 2024), the ATM approach retains physical interpretability and explicit attribution to emission sources and regions, which is a key asset for policy transparency and regulatory applications.

It is important to contextualise the magnitude of the improvements achieved by second-order ATMs within the broader uncertainty framework of air quality modelling. CTM simulations themselves carry substantial uncertainties that can range from 20 to 40% for secondary pollutants, depending on the domain, species, and emission inventory used (de Meij et al., 2024). For quasi-linear indicators such as oxidised sulphur deposition, the improvement introduced by second-order terms is below 0.1% and could be neglected. However, for strongly non-linear indicators — particularly ozone and NO_2 — the linear ATM can introduce systematic biases, especially for large perturbations, that affect not only the magnitude but also the sign of predicted concentration changes in response to emission reductions. In these cases, the second-order correction provides a physically meaningful improvement that is relevant for policy conclusions, independently of the overall CTM uncertainty level. We therefore maintain that the selective inclusion of second-order terms is justified on physical and diagnostic grounds, while acknowledging that the practical impact on policy conclusions is primarily significant for non-linear indicators, rather than for the quasi-linear ones where improvements are numerically small.

Further developments may investigate the behaviour of the matrices under larger emission perturbations or more complex atmospheric regimes, while preserving transparency and interpretability.

Funding

This research was financed in the frame of the Collaboration Agreement between the Italian Ministry of the Environment and Energy Security, ENEA and other national bodies for the implementation of Directive 2016/2284 of the European Parliament and of the Council of 14 December 2016, regarding the reduction of national emissions of certain atmospheric pollutants ("NEC Directive"). The work was supported also by the European Union under the healthRiskADAPT project, "User-driven health Risk Assessment Services and Innovative ADAPTation options against Threats from Heatwaves, Air Pollution, Wildfire Emission and Pollen" (Grant ID: 101157458).

CRediT authorship contribution statement

I. D'Elia: Conceptualization, Data curation, Formal analysis, Funding acquisition, Methodology, Validation, Writing – original draft. **G. Briganti:** Conceptualization, Data curation, Formal analysis, Methodology, Software, Validation, Visualization, Writing – original draft. **G. Kiesewetter:** Conceptualization, Data curation, Formal analysis, Methodology, Software, Validation, Writing – original draft. **A. Cappelletti:** Data curation, Formal analysis, Methodology, Software, Validation, Writing – review & editing. **R. Sander:** Data curation, Formal analysis, Methodology, Software, Writing – review & editing. **M. D'Isidoro:** Data curation, Formal analysis, Software, Validation, Writing – review & editing. **W. Schöpp:** Data curation, Methodology, Software, Validation, Writing – review & editing. **A. Piersanti:** Conceptualization, Data curation, Funding acquisition, Validation, Writing – original draft.

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.

Acknowledgements

The computing resources and the related technical support used for this work have been provided by CRESCO/ENEAGRID High Performance Computing infrastructure and its staff. The infrastructure is funded by ENEA, the Italian National Agency for New Technologies, Energy and Sustainable Economic Development, and by Italian and European research programmes, see http://www.cresco.enea.it/english for information.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.atmosenv.2026.122183.

Data availability

Data will be made available on request.

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