Measured and modelled air quality trends in Italy over the period 2003-2010

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Abstract.

Air pollution harms human health and the environment. Several regulatory efforts and different actions have been taken in the last decades by authorities. Air quality trend analysis represents a valid tool in assessing the impact of these actions taken both at national and local levels. This paper presents for the first time the capability of the Italian national chemical transport model, AMS-MINNI, in capturing the observed concentration trends of three air pollutants, NO_2, inhalable particles having diameter less than 10 micrometres (PM10) and O_3, in Italy over the period 2003-2010. We firstly analyse the model performance finding it in line with the state of the art of regional air quality modelling. The modelled trends result in a general significant downward trend for the three pollutants and, in comparison with observations, the values of the simulated trends were of a similar magnitude for NO_2 (in the range -3.0 - -0.5 μg m^{-3} yr^{-1}), while a smaller range of trends was found than those observed for PM10 (-1.5 - -0.5 μg m^{-3} yr^{-1}) and O_3-maximum daily 8-hour average concentration (-2.0 - -0.5 μg m^{-3} yr^{-1}). As a general result, we find a good agreement between modelled and observed trends; moreover, the model provides a greater spatial coverage and statistical significance of pollutant concentration trends with respect to observations, in particular for NO_2. We also conduct a qualitative attempt to correlate the temporal concentration trends to meteorological and emission variability. Since no clear tendency in yearly meteorological anomalies (temperature, precipitation, geopotential height) was observed for the period investigated, we focus the discussion of concentration trends on emission variations. We point out that, due to the complex links between precursors emissions and air pollutant concentrations, emission reductions do not always result in a corresponding decrease in atmospheric concentrations, especially for those pollutants that are formed in the atmosphere such as O_3 and the major fraction of PM10. These complex phenomena are still uncertain and their understanding is of the utmost importance in planning future policies for reducing air pollution and its impacts on health and ecosystems.

1 Introduction

Air pollution represents one of the main environmental challenges of modern society. Numerous studies have already demonstrated the adverse effects on health (Pope III et al., 2020; WHO, 2019; Pope III and Dockery, 2006; Cohen et al., 2017) and environment (EEA, 2020; Feng et al., 2019), as well as on climate (Watts et al., 2019; Fuzzi et al., 2015), society and economy (Lanzi and Dellink, 2019; OECD, 2016). The adverse impact on health of fine particulate matter results in premature deaths due to ischemic heart disease, strokes, lung cancer, chronic obstructive pulmonary disease and respiratory infections (Apte et al., 2018; Rajagopalan et al., 2018).

Efforts aimed at reducing air pollution have been ongoing for decades, namely in the framework of the Convention on Long-Range Transboundary Air Pollution drawn up under the United Nations Economic Commission for Europe, leading to a general decrease of air pollutant concentrations in Europe (Maas and Grennfelt, 2016). The trends in concentrations are useful to verify if and to what degree environmental regulations establishing limits for pollutant emissions, e.g. the Gothenburg protocol (UNECE, 1979) and the European Directive on National Emission Ceilings (EC, 2016), have been effective and efficient in improving air quality at national and local level. Several European studies addressed this topic, focussing on the entire continent (Colette et al., 2011, 2016, 2017a; Wilson et al., 2012; Guerreiro et al., 2014; Yan et al., 2018) and on single countries (Sicard et al, 2009; Cattani et al., 2014; Querol et al., 2014; Carnell et al., 2019; Velders et al., 2020). The studies were carried out using observed and/or modelled concentrations. The best approach should be the one which integrates both of these sources of information. Indeed, the observed concentrations provide an actual air quality evaluation, though at sparse locations and sometimes with poor temporal coverage, while the modelled concentrations offer a comprehensive spatial and temporal coverage, although they have intrinsic uncertainties in describing the complex processes of atmospheric chemistry and physics (Iversen, 1993).

For Europe, Colette et al. (2011) performed an assessment of nitrogen dioxide (NO_2), particulate matter with diameter of 10 µm or less (PM10) and ozone (O_3) concentrations trends over the 1998-2007 decade, using 6 regional and global chemical transport models (CTMs). The simulated trends were evaluated against observed ones at background monitoring stations located in major anthropogenic emission hotspots. This comparison showed that the primary pollutants trends were generally well reproduced by simulations, with lower performance for O_3 which is a secondary pollutant produced in the atmosphere. Wilson et al. (2012) also investigated the O_3 trends over Europe using the CHIMERE model between 1996 and 2005. The data collected in 158 rural background stations showed that the model reproduces well the European-averaged O_3 trend of the annual 5th percentiles but failed to reproduce the positive trend in the observed 95th percentiles. Another European-wide study was conducted by Yan et al. (2018) for the period 1995-2012 using the global chemical transport model EMAC. The results showed that the model successfully captured the observed temporal variability in O_3 mean concentrations at EMEP background stations, as well as the contrast in the trends of 95th percentile (decreasing) and 5th percentile (increasing). Solberg et al. (2015) and Colette et al. (2017b) provided reviews of scientific papers which compare modelled to observed trends in Europe. In the EURODELTA-Trends multi-model exercise at European scale, Colette et al. (2017a) investigated the period 1990-2010 with 8 chemical transport models (including the AMS-MINNI, Atmospheric Modelling System of the Italian National Integrated Model to support the international negotiation on atmospheric pollution, Mircea et al., 2014; Vitali et al., 2019). The authors showed the time variability of PM10, PM2.5 (Tsyro et al., 2017), organic aerosols and precursor gases (Ciarelli et al., 2019) and O_3 (Mar et al., 2016; Colette et al., 2017b). In particular, the EURODELTA-Trends study, by analysing emissions, intercontinental inflow and meteorological variability, confirmed that the reduction of European anthropogenic emissions plays a fundamental role in the modelled net reduction of ambient air pollution.

Italy is affected by air pollution at the highest levels recorded in Europe (EEA, 2020). Despite this evidence, even if the above mentioned studies over the European area include Italy in their investigations of long-term air quality trends, few analyses focussing on the Italian territory are available. Most of the available trend analyses rely on measured concentrations of single pollutants at single monitoring stations (Casale et al., 2000; Cristofanelli et al., 2015; Gilardoni et al., 2020) or in distinct urban areas (Cadum et al., 1999; Cattani et al., 2010; Gualtieri et al., 2014; Pozzer et al., 2019) and administrative regions (Carugno et al., 2017; Masiol et al., 2017; Lonati and Cernuschi, 2020). Some works cover the whole Po Valley, in Northern Italy, which is a well-known regional hot-spot for air pollution (Putaud et al., 2014; Bigi and Ghermandi, 2016). Currently, the studies by Cattani et al. (2014; 2018) are the only Italian-wide analyses and they are based on measured concentrations available from the National Air Quality database (BRACE, 2013). In particular, Cattani et al. (2014) show significant reduction trends in concentrations of carbon monoxide (CO) and benzene (C_6H_6), linearly related with emission reductions, a large number of stations measuring PM10 and NO_2 decreasing trends and low statistical significance in O_3 trends, which indicates that no clear trend exists in measured ozone concentrations. So far, to the authors' knowledge, there is not a modelling study exploring concentration trends and their relations with emission changes over time covering the whole Italian territory.

This paper evaluates the trends of three air pollutants (NO_2, PM10, O_3) in Italy, over the period 2003-2010, using the AMS-MINNI air quality model. The evaluation of CTM capabilities to reproduce the trends of pollutants increases the reliability of their application in assessing air quality and supporting air quality plans, especially for models regularly used in national regulatory assessments, as requested by Air Quality (EC, 2008) and National Emission Ceilings (EC, 2016) directives but also for other scientific studies. The analysis is based on statistical methods widely used in literature, for the sake of comparability with other investigations on air quality trends. The ability of the model to reproduce the concentration trends is evaluated through the comparison with independent data available from the National Air Quality database (BRACE).

Moreover, in order to identify the potential efficacy of mitigation policies in reducing air pollution, concentration trends were qualitatively compared with variations in meteorology and anthropogenic emissions.

2 Data and methods

2.1 Air quality measurements

The air quality monitoring data considered in the present work derive from BRACE in which data from regional/local monitoring networks were collected for the formal submission to the European Environment Agency (EEA), in the framework of the reciprocal exchange of information and data from networks and individual stations measuring ambient air pollution within the Member States (EC, 1997). BRACE fed the European database Airbase (Airbase, 2020) with data from 2002 to 2012, thus covering the period investigated in this study.

Several processing steps were applied to the raw BRACE database in order to adapt the database to model validation requirements and to verify station reported metadata, in particular concerning geographical coordinates (Piersanti et al., 2012).

In the present work, in order to analyse the concentration trends, we selected only stations covering the 100% of the investigated years with at least 75% of valid data per year. The two thresholds for time coverage were chosen according to the legal requirements on yearly time series stated in the Air Quality Directive (EC, 2008) and also widely adopted in scientific literature (Colette et al., 2011; Colette et al., 2016), for a robust analysis. The threshold of 100% of the investigated years is a more stringent criterion with respect to other studies, generally adopting a less stringent criterion (e.g. 75% is set in Colette et al. (2011), corresponding in same cases to 8 years). Our choice guarantees that the trend analysis is always based on an 8-year period, which can be considered quite robust. Indeed, several studies are available in literature, presenting trend analysis over similar or shorter periods (Zhai et al., 2019; Dufour et al., 2018; Sheng et al., 2018). Of course, data covering a longer period would strengthen our findings. Anyway, in this first study over Italy, the choice of the period to investigate was determined by the availability of coherent model results that have the same model setup for the years 2003 to 2010. More specifically, in the following years, AMS-MINNI simulations adopted a different setup (spatial domain, chemical mechanism, boundary conditions), that clearly affects time series homogeneity. The pollutants considered are NO_2, PM10 and O_3 due to their large monitoring coverage in the period of interest. Particulate matter with diameter less than 2.5 µm (PM2.5) could not be included in the analysis, as the data coverage from BRACE started in 2007 (Uccelli et al., 2017). Time resolution is given in hours (for NO_2 and O_3) and days (for PM10).

The number of the air quality monitoring stations that satisfied the chosen criteria is reported in Table 2. In Appendix S1 of the Supplementary Material (SM), Figure S1 represents the 20 Italian administrative regions and Figures S2-S4 the locations of all sites that passed the selection criteria, by station type (background - BKG, traffic - TRA, industrial - IND) and the background sites by zone type (rural, suburban, urban). The model spatial resolution of 4 km is not sufficient to describe TRA and IND stations with the same skills of BKG stations, nevertheless for the sake of completeness we chose to include them in the validation.

2.2 Model simulations

The air quality modelling system used for our simulations is AMS-MINNI (Mircea et al., 2014, 2016; D'Elia et al., 2009, 2018; Ciucci et al., 2016) which includes a meteorological prognostic model (RAMS), a chemical transport model (FARM), an emission processor model (EMMA) and a meteorological diagnostic processor (SURFPRO).

The three-dimensional Eulerian chemical transport model FARM (Flexible Air Quality Regional Model, http://www.farm-model.org; Gariazzo et al., 2007; Silibello et al., 2008; Kukkonen et al., 2012) describes the transport, turbulent dispersion, formation and destruction of the pollutants in the atmosphere. The mesoscale non-hydrostatic meteorological model RAMS (Regional Atmospheric Modelling System; Cotton et al., 2003) generates the required input meteorological fields. Another fundamental AMS-MINNI component is the emission processor, the Emission Manager EMMA (Arianet, 2014), which prepares the hourly gridded emissions by breaking down annual data from emission inventories in space and time. Moreover, the diagnostic module SURFPRO (Arianet, 2011), computes the Planetary Boundary Layer (PBL) scale parameters, horizontal and vertical diffusivity coefficients, deposition velocities for different chemical compounds and natural emissions, using meteorological fields from RAMS and orographic and land use data.

The main features of the AMS-MINNI simulation setup used to carry out the simulations are synthetized in Table 1.

Table 1. Main features of the AMS-MINNI simulation setup.

Chemical Transport Model Simulation	
Model and version	FARM version 4.7
Horizontal resolution	4 km
Vertical layers	16 terrain-following layers
Vertical extent	10000 m
First layer depth	40 m
Gas-phase chemistry	SAPRC99 (Carter, 2000)
SIA module	ISORROPIA v1.7 (Fountoukis et al., 2007)
SOA module	SORGAM module (Schell et al., 2001)
Aerosol model	AERO3 (Binkowski and Roselle, 2003)
In-cloud sulphate chemistry	Simplified S(IV) to S(VI) formation (Seinfeld and Pandis, 1998)
Boundary Conditions	Eurodelta (Colette et al., 2017a)
Meteorological Simulation	
Model and version	RAMS version 6.0
Horizontal resolution	12 km and 4 km (two way nesting)
Vertical	32 levels (sigma coordinate) from 30 m above ground level to lower stratosphere
Radiation	Chen and Cotton (1983) long/shortwave model – cloud processes considering all condensate as liquid
Convection	Modified Kuo scheme (Tremback,1990)
Lower Boundary	LEAF-2, Land Ecosystem–Atmosphere Feedback model (Walko et al., 2000)
Turbulence Closure	Mellor-Yamada level 2.5 scheme – ensemble–averaged TKE (Mellor and Yamada, 1982)

Cloud Microphysics	Bulk microphysics parameterization: cloud water, rain, pristine ice, snow, aggregates, graupel, and hail, or certain subsets of these (Walko et al., 1995)
Boundary conditions	GFS analyses at 0.5° horizontal resolution (https://wwdata/model-datasets/global-forcastw.ncdc.noaa.gov/data-access/model--system-gfs)
Data Assimilation	Nudging on pre-analysed fields
Emission Processing	
Anthropogenic Emissions Software and version	EMMA version 6.0
Anthropogenic emissions Inventories	National Emission Inventories of Italy and neighbouring countries reported to the European Monitoring and Evaluation Programme of the UNECE Convention on Long-range Transboundary Air Pollution
Biogenic model e Soil-NO	MEGAN v2.04 (Guenther et al., 2006)
Saharan dust	None
Sea salt	Zhang et al. (2005)
Windblown dust	Vautard et al. (2005)
Dust traffic suspension	Amato et al. (2012); Padoan et al. (2018)

More details about the anthropogenic emissions and the meteorological data are reported in paragraph 2.3 and 2.4, respectively.

A complete description of the standard configuration of the modelling system can be found in Vitali et al. (2019).

2.3 Anthropogenic emissions

Emission data used as input for AMS-MINNI simulations derive from the national emission inventories covering the period from 1990 to 2015, elaborated by ISPRA (Italian Institute for Environmental Protection and Research, Taurino et al., 2017) available in 2017. Figure 1 shows the emission variation for SO_X (sulphur oxides), NO_X (nitrogen oxides), PM2.5, PM10, NMVOC (non-methane volatile organic compounds) and NH_3 (ammonia) for the period 2003-2010 considered in the present work. The variation over the whole period, 1990-2015, by SNAP nomenclature (Selective Nomenclature for Air Pollution, see Table S1 of Appendix S2 in the SM) for the selected pollutants is reported in the SM (Appendix S2, Figs. S5-S7).

SO_X emissions show the highest reduction, -58% in the period 2003-2010, followed by NO_X (-29%) due to a large decrease in combustion from energy and road transport sectors, respectively. NMVOC emission reduction is driven by the road transport and solvent use sectors, while NH_3 emissions show a very slight decrease. PM2.5 and PM10 emissions increase from 2005 to 2008 due to an increase in biomass combustion in the residential sector (SNAP code 02) (IIR, 2021).

The estimated emissions at national level need to be further disaggregated in space, before being assigned to the AMS-MINNI grid at 4 km spatial resolution. A provincial distribution (NUTS3 level, where NUTS stands for Nomenclature of territorial units for statistics, the hierarchical system for dividing up the territory of the European Union, https://ec.europa.eu/eurostat/web/nuts/background) is provided by ISPRA every 5 years; hence it was available for both the years 2005 and 2010. For the purposes of this work, the 2005 NUTS3 disaggregation was used for the years 2003, 2004, 2005, 2006 and 2007, while the 2010 NUTS3 disaggregation for 2008, 2009 and 2010. Finally, hourly and speciated gridded emissions on the AMS-MINNI grid were produced by means of EMMA processor. The spatial allocation of NUTS3 emissions to the 4 km grid of the MINNI model relied for point sources on geographic coordinates of each facility (for example, large combustion plants) and for diffuse/linear sources on spatial layers used as proxy variables, like population density (for residential heating and urban traffic), georeferenced road networks (for rural and highway traffic), land-use (for agriculture).

2.4 Meteorological simulations

The meteorological simulations required by AMS-MINNI were elaborated making use of the RAMS model whose main features are summarized in Table 1. The hourly meteorological fields produced by RAMS, such as temperature, wind speed, relative humidity and precipitation play an important role in determining the level of air pollution concentrations. In trend analysis, it is important to establish the role of the emissions and the meteorology in influencing air pollutant concentration trends. It is out of the scope of the present paper to attribute a relative weight to these factors in determining the analysed concentration trends, but, as a first approximation, we can consider that it could be reasonably attributed to emission trends rather than to a clear tendency in meteorology. In fact, looking at the anomalies (referred to 1981-2010 climatology) of some meteorological fields for the considered years (2003-2010) computed from NCEP/NCAR reanalyses (National Centers for Environmental Prediction/National Center for Atmospheric Research, Kalnay et al., 1996), it is worth noting that no clear tendency is shown. In Appendix S3 of the SM, yearly maps for temperature at 850hPa (T850), precipitation and 500hPa geopotential height (Z500) anomalies are reported, together with the near surface temperature trend computed from the Copernicus Climate Data Store (CDS, http://climate.copernicus.eu/climate-data-store).

2.5 Trend methodology

The detection and calculation of trends in measured and simulated concentrations were performed using the “openair” package (Carslaw and Ropkins, 2012), specifically designed for air pollution data analysis developed for the open source R software (version used v.3.6.1, http://www.R-project.org). The presence of a monotonic increasing or decreasing trend was estimated using the non-parametric Mann-Kendall trend test together with the Theil-Sen’s method for estimating the slope of a linear trend (as a concentration variation per year) (Mann, 1945; Theil, 1950; Sen, 1968; Kendall, 1975), adopting the deseasonalisation option. The calculated trends were considered as statistically significant if the significance level (i.e., the p-value of the Mann-Kendall test) is lower than 0.05 ($p<0.05$). This method does not require assumptions about the data distribution, it is not sensitive to outliers and it has been used in several studies, for example in the EMEP Task Force on Measurements and Modelling during the Eurodelta experiment (Colette et al., 2016) and in the EEA air quality trend reports (EEA, 2009; 2020). Temporal trends were calculated considering monthly averages of the pollutant concentrations at each monitoring stations. Table 2 summarizes the number of stations, grouped per type, with significant and non-significant trends both for observations and modelled estimates.

Table 2. Number of stations considered in the trend analysis for the period 2003-2010 separated into background (BKG), traffic (TRA), industrial (IND) and classified as statistically significant (p<0.05) for observed, simulated and both observed and simulated trends.

	Number of stations				Observations: number of stations with $p<0.05$				Simulations: number of stations with $p<0.05$				Number of stations where both obs and sim with $p<0.05$			
Pollutant	BKG	TRA	IND	***Tot***	BKG	TRA	IND	***Tot***	BKG	TRA	IND	***Tot***	BKG	TRA	IND	***Tot***
NO_2	36	33	4	***73***	26	19	2	***47***	32	33	3	***68***	22	19	1	***42***
PM10	14	16	2	***32***	12	13	2	***27***	7	6	1	***14***	5	5	1	***11***
O_3 – conc: All year	53	8	4	***65***	23	3	4	***30***	19	3	1	***23***	6	1	1	***8***
O_3 – conc: Apr-Sep					30	7	3	***40***	21	3	2	***26***	11	3	2	***16***
O_3 – MDA8: All year					26	4	4	***34***	31	5	4	***40***	15	3	4	***22***
O_3 – MDA8: Apr-Sep					33	6	4	***43***	35	5	4	***44***	22	4	4	***30***
O_3 – AOT40: Apr-Sep					32	6	3	***41***	32	4	4	***40***	20	4	3	***27***
O_3 – SOMO35					21	2	3	***26***	8	1	1	***10***	3	0	1	***4***

3 Results and discussion

3.1 Model validation results

Before inspecting the capability of AMS-MINNI to capture the trends of pollutant concentration, a comprehensive evaluation of the model results was carried out.

Comparisons between time series of observed and modelled values were performed on the same set of monitoring stations satisfying the selection criteria used for the trends analysis (i.e. with at least 75% of valid data per year covering all the 8 years from 2003 to 2010, see Table 2).

For all the pollutants included in the trend analysis, annual time series of daily values were used for the comparison, this metric being considered the most appropriate one for model performances assessment (Colette et al., 2011). For O_3, in addition to daily values, the MDA8 metric (maximum daily 8-hour average concentration), calculated for the period from April to September, was considered as well, since it turned out to be the most suitable metric for O_3 trends analysis within the context of this study (see Section 3.2.3).

As recommended by literature on model validation (Chang and Hanna, 2004), a comprehensive set of statistical indices was computed in order to quantify, from different points of view, the agreement between modelled and observed values.

Here, for the sake of brevity, only three out of all the computed statistical metrics are presented: ***M****ean* ***B****ias* (***MB***), ***R****oot* ***M****ean* ***S****quare* ***E****rror* (***RMSE***) and the ***corr****elation coefficient* (***corr***); see Appendix S4 in the SM for their formulations. These indices were chosen because they globally capture several features of model performance in terms of amplitude, phase and bias. Moreover, such indicators are frequently used in model evaluation studies (Simon et al., 2012), namely those previously cited on temporal trends. Indeed, in Colette et al. (2011), which we consider as a reference for the present evaluation, model validation is based on the same subset of these three statistical indices. Values of ***MB***, ***RMSE*** and ***corr*** for each pollutant are presented here as an average over the 8 years period and over all the available stations, classified according to their type (BKG, TRA, IND) and, for BKG stations, by zone type (rural, suburban, urban).

Results are shown in Fig. 2 for daily values of NO_2 (upper left panel), PM10 (upper right panel), O_3 (lower left panel) and for MDA8 of O_3 (lower right panel).

Overall, model performance is in line with the results obtained by analogous modelling systems (e.g. Solazzo et al., 2012; Pirovano et al., 2012; Badia and Jorba, 2015; Bessagnet et al., 2016), especially when applied at similar spatial resolution (e.g. Chemel et al., 2010; Pay et al., 2014). More specifically, in Table S2 of Appendix S4 in the SM, the statistical score values are reported together with the outcomes of Colette et al. (2011), used hereafter as a reference for an explicit comparison of the performances.

As far as NO_2 daily values are concerned (upper left panel of Fig. 2), ***RMSE*** and ***corr*** values, ranging from 10.8 to 28.6 µg m^{-3} and from 0.578 to 0.689, respectively, with BKG stations scoring best, are in line with Colette et al. (2011). According to ***MB***, negative values**,** between -22.4 and -4.2 µg m^{-3}, are obtained for all station types, stressing a general underestimation of NO_2 concentration values. Anyway, underestimation is generally lower than in Colette et al. (2011) at BKG stations, getting worse at TRA sites. This feature is commonly expected in chemical transport model applications at regional scale and it can be ascribed to the intrinsic difficulties of regional models in capturing, at their resolution, high gradients in spatial concentration variability (Schaap et al., 2015). This hypothesis is confirmed by the evidence that model performance (according to both ***RMSE*** and ***MB***) deteriorates with decreasing spatial representativeness of monitoring sites; in particular, absolute values of ***MB*** (i.e. underestimations) increase passing from rural to urban environments and even more at TRA stations.

AMS-MINNI tends to underestimate PM10 daily values too, which is common for regional models, as shown by negative values of ***MB*** in the upper right panel of Fig. 2. However, underestimation does not seem to increase with decreasing spatial representativeness of sites, and can be attributable to the well-known difficulties of air quality models to take into account all the contributions to PM10 concentration (Solazzo et al., 2012; Im et al., 2015). In particular, it is worth noting that, in the present AMS-MINNI simulations, the contribution of Saharan dust was not included and this could be the main reason for the underestimation at rural sites. As far as ***MB*** and ***corr*** are concerned, simulated PM10 concentrations are overall in agreement with observations, with values ranging from -12.8 to -3.9 µg m^{-3} and from 0.453 to 0.630, respectively.

AMS-MINNI O_3 daily values (lower left panel of Fig. 2) are in line with the findings of Colette et al. (2011) concerning both the general overestimation of O_3 concentration levels and the range of values of the statistical indices (21.7 - 25.7 µg m^{-3} for

RMSE, 2.2 - 18.6 μg m^{-3} for ***MB*** and 0.683 - 0.822 for ***corr***). More specifically, Table S2 shows that, for background stations, similar ***RMSE*** values are obtained, together with generally lower ***MB*** values and better correlation values. Similarly to the performance for NO_2, the ***MB*** of O_3 concentrations changes with the spatial representativeness of monitoring sites, i.e. as NO_2 underestimation increases passing from rural to urban environments, O_3 overestimation increases, since close to NO_2 sources the titration process acts as an O_3 sink (Seinfeld and Pandis, 1998).

Model performance in reproducing MDA8 of O_3 for the period from April to September (lower right panel of Fig. 2) is similar to that for daily concentrations, evaluated throughout the whole year, apart from the negative (albeit small) ***MB*** value obtained at rural stations. With respect to daily values, correlation for MDA8 (0.712 - 0.853) is generally better, as is ***MB*** (lower absolute values). With regards to ***RMSE*** (24.4 - 25.1 μg m^{-3}) the values are worse at BKG stations and slightly better at IND and TRA sites. Nevertheless, it is worth noting that, when assessing O_3 performance, higher biases in concentration estimates could be expected when using the MDA8 metric, instead of daily average, since concentration levels are higher too. Indeed, higher MDA8 concentration values are expected when compared with daily values for two reasons: i) maximum values are taken into account instead of average ones; ii) only the warm period (April-September) is considered here, when higher O_3 values are generally observed.

Globally, AMS-MINNI performs quite well, with the results being in line with the performances of state of the art of air quality models, when operating at the regional scale, when considering both the values of the statistical indices used for the comparison and the general tendency to overestimate O_3 and to underestimate NO_2 and PM10.

3.2 Trend analysis

From the concentration fields provided by AMS-MINNI simulations, data were extracted at each monitoring station to compare observed trends (OT) and simulated trends (ST).

In the following paragraphs, for each of the pollutants considered and for the whole set of stations described in Table 2, an analysis of observed and simulated trends is discussed examining different parameters. For each pollutant, we present:

- the overall distribution of stations with statistically significant/not significant trends, with their sign, for both observations (OT) and simulated (ST) values, in order to evaluate model performance in reproducing temporal trends in measured concentrations (Figs. 3, 7, 11);
- the time series of observed and simulated monthly average concentrations (averaged over all stations for each station type) (Figs. 4 (a), 5 (a), 8 (a), 9 (a), 12 (a), 13 (a));
- scatter plots of observed/simulated slopes, by station type (Figs. 4 (b), 5 (b), 8 (b), 9 (b), 12 (b), 13 (b));
- maps of simulated slopes at each grid point, in comparison with the spatial distribution of observed slopes by station type, in order to provide a more detailed description of the results, since observed and simulated slopes are presented according to their spatial distribution and geographical context. Moreover, simulated quantities are provided not only at monitoring sites but at every grid point of the computational domain, to fully exploit model capabilities at their best in terms of both spatial coverage and variability (Figs. 6, 10, 14).

In Appendix S5 of the SM the observed and simulated slopes (both in terms of µg m^{-3} yr^{-1} and % yr^{-1}) are reported for each pollutant and for each station with a significant trend ($p<0.05$).

3.2.1 NO_2

Out of 73 monitoring stations, 47(68) have a statistically significant OT(ST) whereas 42 result to have both significant observed and simulated trends. Figure 3 shows that all STs are negative (93% significant), whereas 79% of the OTs were negative (58% significant; 21% non-significant) and 21% were positive (7% significant; 14% non-significant). Figures 4 (a) and 5 (a) show that the model reproduces monthly values better at BKG sites than at TRA and IND stations, while the intra-annual variability is well reproduced for all types of stations. This result confirms the good model performance for daily values at BKG stations (Fig. 2).

The scatter plots of Figs. 4 (b) and 5 (b) show an overall good agreement for BKG sites with statistically significant trends, while, as expected, performance is worse at TRA sites where the absolute values of the simulated slopes are mostly lower than the observed ones.

Figure 6 shows that model simulations provide coverage and information in parts of the domain where observations are completely absent in the considered period, i.e. in the Southern part of Italy. Overall, at BKG stations the model captures both the sign and the variability of the slopes better, while it is worse at TRA stations. The map of the simulated slopes not only has a wider coverage but also shows a greater area with significant trends compared with observations.

3.2.2 PM10

The well-known underestimation of PM10 concentrations when simulated by regional models, already discussed in Section 3.1, and the poor quality of the observation network, shown by the low number of stations fulfilling the selection criteria, greatly influenced the trend estimates. Out of 32 monitoring stations, 27(14) have a statistically significant OT(ST) while 11 have both observed and modelled significant trends. The fraction of all the sites with statistically significant observed trends, shown in Fig. 7, is 84% compared with only 44% for the ST. The simulated monthly mean time series illustrated in panel (a) of both Figs. 8 and 9 for BKG and TRA/IND stations, respectively, show a general underestimation of observed concentrations, with performances improving slightly from 2007 onwards. Focussing on sites where both observed and simulated trends are statistically significant, Figures 8 (b) and 9 (b) show that the model succeeds in capturing not only the sign of all the observed trends, but also the slopes at many sites, even if the absolute values are underestimated, especially at the industrial site. This result is confirmed by the maps in Fig. 10, which show a general agreement at most of the available monitoring stations. Although to a lesser extent when compared with NO_2 and O_3, the simulated statistically significant trends for PM10 cover a wider area with respect to observations especially in the Northern area and in the Sardegna island where no observations are available at all. A poor coverage of significant trends in both model and observations occurs in some areas of Central and Southern Italy, where the model estimates larger areas of significant trends, especially for the Puglia region and Sicilia island.

3.2.3 O_3

As underlined in Lefohn et al. (2017; 2018) and Colette et al. (2017b), the choice of the O_3 metrics is of the utmost importance since each indicator could show a different trend. In our analysis, both effect-based indicators (AOT40 and SOMO35) and process-based indicators (MDA8) were computed and analysed. The different metrics explored are: the mean O_3 concentration (O_3 avg); the maximum daily 8-hour average concentration (MDA8); the accumulated amount of ozone over the threshold value of 40 ppb (AOT40) calculated from April to September (Apr-Sep) and the sum of the daily maxima of 8-hour running average over 35 ppb (SOMO35) for the whole year. Concerning O_3 avg and MDA8 metrics, analyses were carried out for both the entire year and from April to September. The number of stations with increasing and decreasing trends and their significance depends on the metric used (Table 2 and Fig. 11). The fraction of stations with significant trends also varies between the observed and modelled datasets. Annual metrics (O_3 avg, MDA8 and SOMO35) have a lower fraction of significant trends than the metrics calculated in Apr-Sep. For the purpose of our analysis, i.e. to show the capacity of the AMS-MINNI in capturing the air pollution trends through a comparison of observations and simulations, we preferred to focus on the MDA8 indicator calculated in the warm period (Apr-Sep), when higher O_3 values are generally recorded. Indeed, the MDA8 calculated in the period Apr-Sep has the highest number of stations with significant trend among all indicators.

The fraction of stations with significant trend is comparable between observations and simulations, 66% and 68% respectively, but when looking at the sign of the trend, we found out that all significant simulated trends are decreasing, while the 39% of significant observed trends are increasing. The monthly mean shows a good agreement for BKG stations (Fig. 12 (a)) and a slight overestimation for both IND and TRA stations (Fig. 13 (a)). The scatter plots (panel (b) of both Figs. 12 and 13) show a higher variability for observed trends than for those simulated.

When looking at the spatial distribution, Figure 14 shows a large area of significant simulated slopes, ranging from -2.0 $\mu g\ m^{-3}\ yr^{-1}$ to -0.5 $\mu g\ m^{-3}\ yr^{-1}$ with an area of non-significant ST in the North-Eastern area. The comparison with observations is particularly interesting for BKG stations, for which there are more stations available. As already pointed out, the model does not reproduce the observed positive trends, but the model has a good agreement with the significant decreasing OT, although with a lower variability. Moreover, there are some areas, especially in Central and Southern Italy, where the model shows a significant trend, whereas monitoring sites are not available at all or the OT is not significant.

3.3. Discussion

Our analysis shows that AMS-MINNI is capable of reproducing observed trends albeit with some differences between the pollutants studied. Although a quantitative analysis of the influence of variations in emissions and meteorology on concentration trends was not performed, we present a preliminary qualitative attempt to compare the temporal concentration trends to variation in emissions, having already observed (see Section 2.4) that there is no clear tendency in the meteorology.

The nitrogen oxides (NO_X) that are most relevant for air pollution (namely NO and NO_2) are mostly emitted during fossil fuel combustion processes, and in particular by road transport. In our analysis, the road transport sector represents almost the 50% of all the total emitted NO_X (see Fig. S5 of Appendix S2 of the SM). The decrease of NO_2 concentrations is almost consistent with the decrease in NO_X emissions, since NO_2 concentrations are directly linked to primary emissions (Colette et al., 2011; Henschel et al., 2015) and mainly driven, in our case, by a reduction in emissions from the road transport sector. Despite the underestimation of absolute values of background concentrations, AMS-MINNI adequately reproduces the observed trends at a national scale (Fig. 4 (b)), demonstrating its potential for supporting reduction policies of background pollution. On the other hand, besides underestimating concentrations at traffic stations like many state of the art CTMs, the decreasing concentrations trends observed at traffic stations is underestimated (Fig. 5 (b)). This indicates that the model is either misrepresenting the decrease of emissions, or the model is not responding correctly to the changes in emissions. Moreover, the spatial resolution can limit the model's ability to capture large concentration gradients, typical of the urban environment, and this may be the reason for failure of the model to capture the positive trends. As an interesting example, from Fig. 6 (lower right panel) it turns out that the traffic station with the highest positive observed slope (as showed in Fig. 5 (b)) is located in Florence, Toscana region. The comparison of lower right and upper left panels of Fig. 6 shows that this traffic monitoring site, (airbase code IT0861A, see Table S3 of Appendix S5 in the SM), is located between two urban BKG sites that have non-significant OT. The three monitoring points are located within about 4 km (i.e. in the same cell of the computational domain). This is a feature that the model is not able to capture; indeed, in this area simulated trends are not significant or decreasing. Something similar occurs in most of the cases with positive OT. Most of these points are very close to other monitoring sites where the opposite behaviour (negative slopes) is observed; see for example the couple of BKG sites in Lombardia surrounded by other BKG sites where the opposite sign is found, or the IND site located in Eastern Liguria very close to a TRA site with a decreasing trend. Therefore, when designing mitigation scenarios at local urban scale, these results suggest that a regional scale CTM like AMS-MINNI needs to be integrated with high resolution models.

Concerning PM and O_3, given their secondary nature, a direct link between emissions and atmospheric concentrations is not expected (Guerreiro et al., 2014).

PM10 is both primarily emitted and secondarily generated in the atmosphere from reactions of chemical precursors (NO_X, SO_X, NH_3, NMVOC). Therefore, observed concentrations reflect these and other contributions, like long-range transport, including Saharan dust, in variable fractions depending on the site. The national emissions of primary PM10 (Fig. 1) are stable for the first four years (apart from 2004), then grow for four years and diminish in the last period, resulting in a final increase of 13% from 2003 to 2010. On the other hand, the emissions of all four mentioned precursors decrease at different rates. These contrasting trends in emissions could partly explain the large areas of non-significant trends shown in Fig. 10, whereas the areas with the higher simulated decrease correspond mainly to industrial and traffic areas, underlying the significant efforts in reducing emissions from the industrial and road transport sectors. The few stations available for the comparison show a nice model skill in reproducing observed negative trends, even on TRA stations. This could be a preliminary confirmation of the fitness of AMS-MINNI for the purpose of supporting emission reduction planning, even though further evaluations of model trends are needed, especially for more recent time intervals. Moreover, the observed biases in concentrations, especially at the beginning of the time series (Figs. 8 (a) and 9 (a)), suggest that further insights are needed to investigate how the change of model performances could affect the trend estimates.

O_3 is a secondary pollutant produced in the troposphere by the chemical reactions of its precursors, such as NO_X and NMVOC, while CH_4 and CO become more important at a wider scale (Guerreiro et al., 2014).

The number of cited studies focussing on O_3 indicates how critical this pollutant is when exploring relations between temporal trends of emissions and concentrations, given the complex photochemistry, showing sometimes a discrepancy between the emission decrease of O_3 precursors and the variation of O_3 concentrations (Colette et al., 2011; Guerreiro et al., 2014; Querol et al., 2014). This is particularly important in Mediterranean areas, which are susceptible to ozone-related impacts (De Marco et al., 2019) due to climatological conditions that are more favourable for O_3 formation. The national emissions of the main ozone precursors follow a similar descending trend in the 8 years considered, thus there has been little change in the ratio between them, which is the main driver of the chemical equilibrium for O_3 formation (Seinfeld and Pandis, 1998; Sillman, 1999). This could partly explain (Fig. 14) why the model gives not significant or close to zero trends in Northern Italy, especially in the Po Valley, a well-known air pollution hot spot, densely populated and with high anthropogenic emissions. In the same region, where most of the monitoring stations are concentrated, different behaviours of OT are observed: negative slopes (especially in the Western part), not significant trends and positive slopes in some stations, mainly located in complex orographic contexts or near to the coastline, where the transport of O_3 from the sea, caused by sea breeze circulation (Monteiro et al., 2016), together with precursor emission by nearby harbours, could lead to local peculiar features. Similar findings can be found in the literature, as for example in Guerreiro et al. (2014) or in Colette et al. (2011), who noticed in particular that different models had different behaviours. On a national level, Cattani et al. (2014), focussed on observations throughout the Italian territory in the period 2003-2012, showed that it is not possible to estimate a general significant statistical trend (although with a different reference metric, i.e. SOMO0 calculated from April to September), regardless of the type or the area of the stations, and that there are discrepancies in significant trend between adjacent stations. Moreover, as already mentioned, the choice of the O_3 metrics can influence the trend estimate. Overall, in our analysis, AMS-MINNI underestimates the absolute value of the descending OT at background stations. This result is driven by North-Western monitoring locations, where further work is needed to analyse the quality of local emission estimates and external contributions to ozone concentrations.

4 Conclusions

The present work aims to assess for the first time the capability of the Italian chemical transport model AMS-MINNI of capturing the trends of three pollutants, namely NO_2, PM10 and O_3. The analysis for O_3 was carried out using different metrics, both for observations and simulations. We firstly conducted a thorough analysis of the model skill considering some statistical score parameters most commonly used in the literature. This analysis confirms that the model performance is in line with the state of the art for regional model applications. Statistical indicators are as good as other CTMs in literature and a similar behaviour to that of most regional models was observed concerning the general tendency to overestimate O_3 and to underestimate NO_2 and PM10. The trend evaluation was performed using the non-parametric Mann-Kendall trend test together with the Theil-Sen's method for the estimation of the slopes and an in-depth comparison between observed and AMS-MINNI modelled trends was carried out. Comparing the sign of modelled and observed trends we found a good agreement for almost all sites. Our main result is a general downward simulated trend for the three pollutants. With respect to observations, modelled slopes show the same magnitude for NO_2 (in the range -3.0 - -0.5 μg m^{-3} yr^{-1}), while a smaller variability is detected for PM10 (-1.5 - -0.5 μg m^{-3} yr^{-1}) and O_3-MDA8 (-2.0 - -0.5 μg m^{-3} yr^{-1}). The reason for the discrepancy for PM10 could be attributed to the well-known underestimation of modelled PM10 concentrations. The results for O_3 could be influenced by the poor quality of the monitoring network data in the period we considered, together with the well-documented difficulties of models in capturing O_3 concentration trends, given its non-linear dependence on precursors emissions.

Model capabilities in terms of both spatial coverage and variability are illustrated by the maps for the three pollutants, showing larger area for significant simulated trends compared with those observed, with a larger coverage for NO_2 and O_3-MDA8 and a smaller one for PM10. For all pollutants, almost the entire domain of Northern Italy has significant simulated trends. Even for Southern Italy, where in general a low coverage of significant modelled PM10 trends is obtained, there are areas with significant simulated trends where there are no observations. It is also worth noting that in the major islands, Sardegna and Sicilia, the simulated trends give useful information, filling the gap due to a sparse or absent monitoring network.

Moreover, a qualitative comparison between the temporal concentration trends and the meteorological and emission variations was carried out too. Since we do not observe a clear tendency in meteorological anomalies, concentrations trends were discussed in connection with emission variations. Indeed, it was pointed out that, due to the complex links between precursor emissions and air pollutant concentrations, emission reductions do not always result in a corresponding decrease in atmospheric concentrations, especially for secondary pollutants like PM10 and O_3. Studies on air pollutant trends are relevant to evaluate the impact of the actions taken to reduce emissions in different environmental policies both at national and local levels. The evaluation of the AMS-MINNI capability to reproduce the trends of pollutants increases the reliability of its application in assessing air quality and supporting air quality plans, especially for its use in national regulatory assessments. Indeed, our analysis demonstrates the good agreement between modelled and observed trends and the added value of the model in increasing both the coverage and the significance of air concentration trends with respect to observations. Model performance is best for NO_2, while for the others, especially O_3, the issue is more challenging. Moreover, the capability to interpret past air quality trends is fundamental in understanding the efficacy of already applied air quality policies and measures and in planning further actions. As demonstrated, the understanding of complex interactions is still uncertain and represents a gap to be filled since it is of the utmost importance in planning future policies aimed at reducing air pollution and its impacts on health and ecosystems.

The present analysis may be applied to other pollutants, especially substances of potential concerns for health (e.g. PM2.5). Moreover, it can be considered a reference for other studies in complex geographical conditions such as the Italian territory, that represents an interesting environmental framework, due to its complex orography, resulting in peculiar meteorological conditions, the great variety of natural and anthropogenic contexts, and the presence of the Po Valley, a well-known air pollution hot spot.

Appendix A: list of acronyms

AMS = Atmospheric Modelling System

BKG = Background

BRACE = Banca Dati e Metadati di Qualità dell’aria (National Air Quality database)

corr = correlation coefficient

CTM = Chemical Transport Model

ECMWF = European Centre For Medium-Range Weather Forecast

EEA = European Environmental Agency

EMAC = ECHAM/MESSy Atmospheric Chemistry

EMEP = European Monitoring and Evaluation Programme

FARM = Flexible Air Quality Regional Model

IND = Industrial

ISPRA = Istituto Superiore per la Protezione e Ricerca Ambientale (Italian Institute for Environmental Protection and Research)

MB = Mean Bias

MDA8 = Maximum Daily 8-hour Average

MINNI = Modello Integrato Nazionale a supporto della Negoziazione Internazionale sui temi dell’ Inquinamento atmosferico (Italian National Integrated Model to support the international negotiation on atmospheric pollution)

NO_2 = nitrogen dioxide

NUTS = Nomenclature of territorial units for statistics

O_3 = ozone

OECD = Organization for Economic Co-operation and Development

OT = Observed Trends

PBL = Planetary Boundary Layer

PM10 = particulate matter with diameter of 10 μm or less

RAMS = Regional Atmospheric Modelling System

RMSE = Root Mean Square Error

SIA = Secondary Inorganic Aerosol

SM = Supplementary Material

SNAP = Selective Nomenclature for Air Pollution

SOA = Secondary Organic Aerosol

ST = Simulated Trends

SURFPRO = SURFace-atmosphere interface PROcessor

TRA = Traffic

WHO = World Health Organization

WMO = World Meteorological Organization

Code and data availability

The meteorological model RAMS v6.0 is freely available at http://www.atmet.com/software/rams_soft.shtml. The chemical transport model FARM v4.7.0 is freely available at https://hpc-forge.cineca.it upon request to ARIANET s.r.l. (http://www.aria-net.it). The emission software emma6 is available on charge upon request to ARIANET s.r.l. All the codes can be provided confidentially for the editor and reviewers in order to enable peer review. All the modelled data (gridded emissions, meteorological and concentrations fields at 4 km resolution) and the trend analysis calculations are available upon request to the authors. Observation data are publicly available from the BRACE website (http://www.brace.sinanet.apat.it/web/struttura.html).

Authors contribution:

ID, LV, GR, AP put original effort into: conceptualization, methodology, study and interpretation of the trend analysis. MD, GB, AC performed the model simulations; ID and GB carried out the data processing; ID, LV, GR, AP, MD wrote the original draft, including visualization, and performed the review and editing; AC, GB, MA, MM, GZ, LC contributed to the writing review and editing; GZ and LC accomplished the acquisition of funds.

Competing interests:

The authors declare that they have no conflict of interest.

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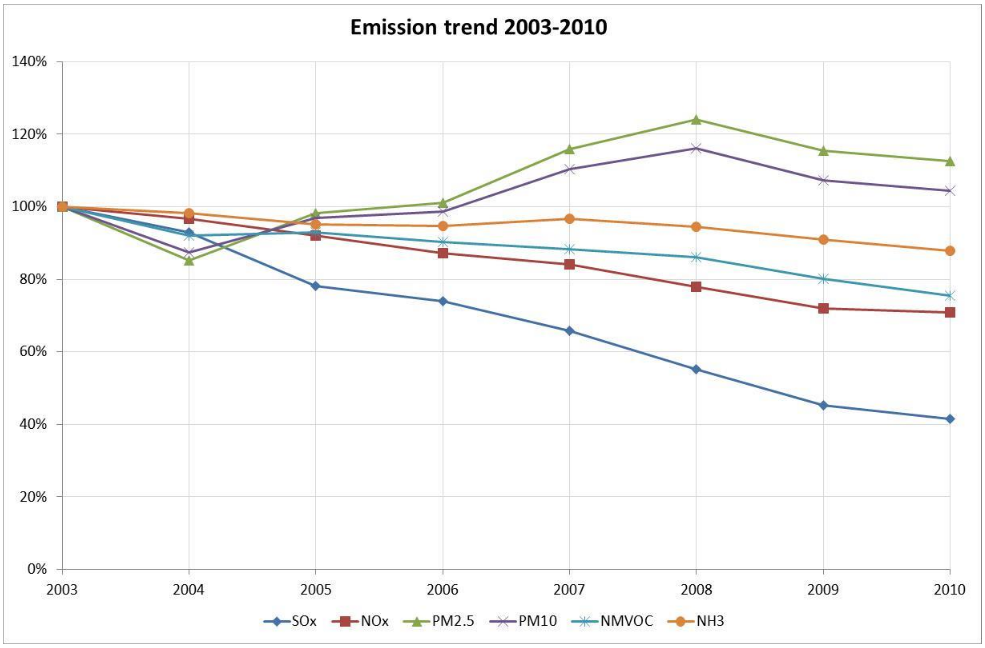

830 **Figure 1: Italian anthropogenic emissions, from 2003 to 2010 relative to 2003, , elaborated from ISPRA emission data set described in Taurino et al. (2017).**

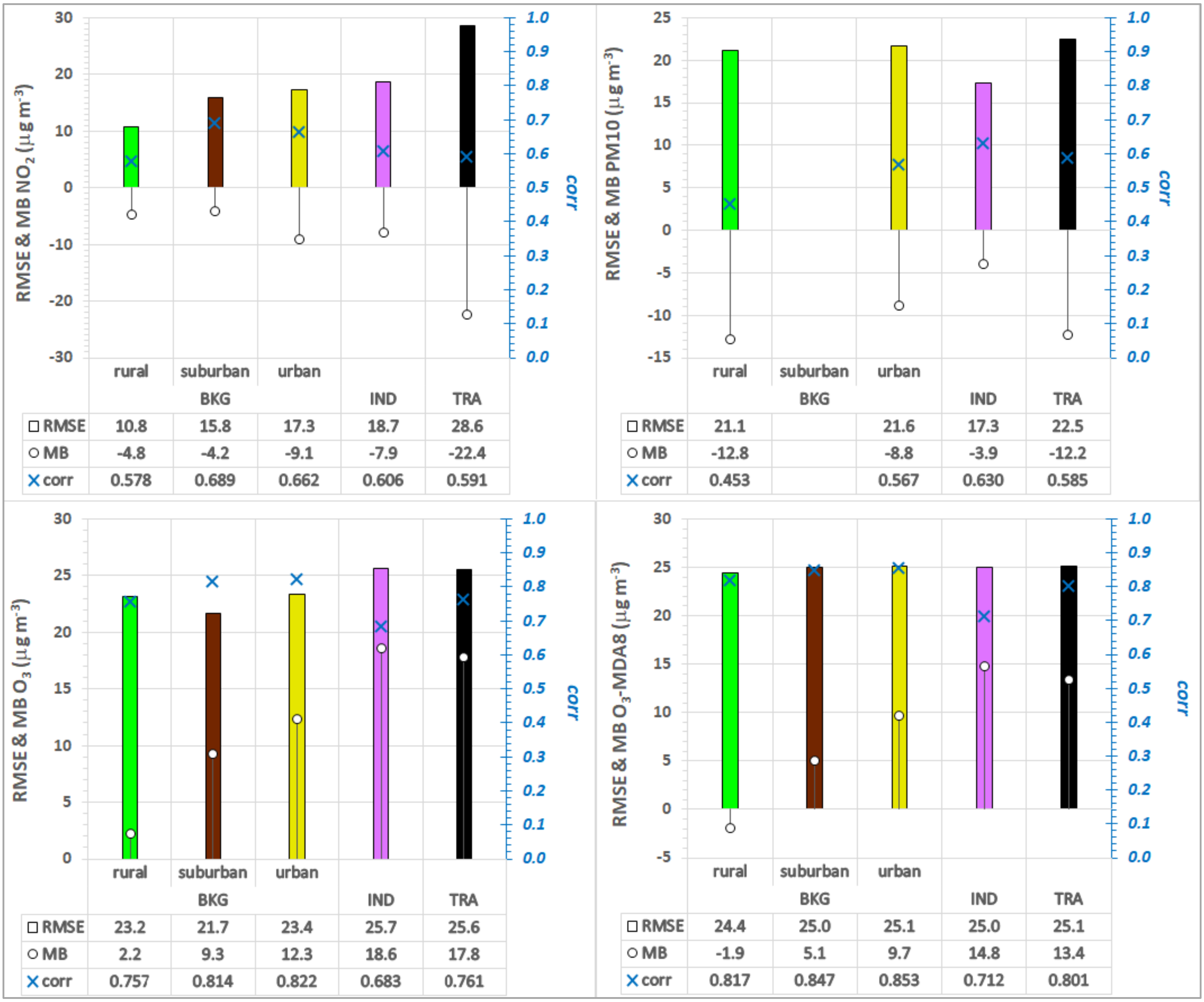

Figure 2: Summary of model performance evaluated at all valid Italian monitoring stations during 2003-2010. The statistical scores are based on annual time series of daily average values of NO_2 (upper left panel), PM10 (upper right panel) and O_3 (lower left panel) and on MDA8 of O_3, calculated for the period Apr-Sep (lower right panel).

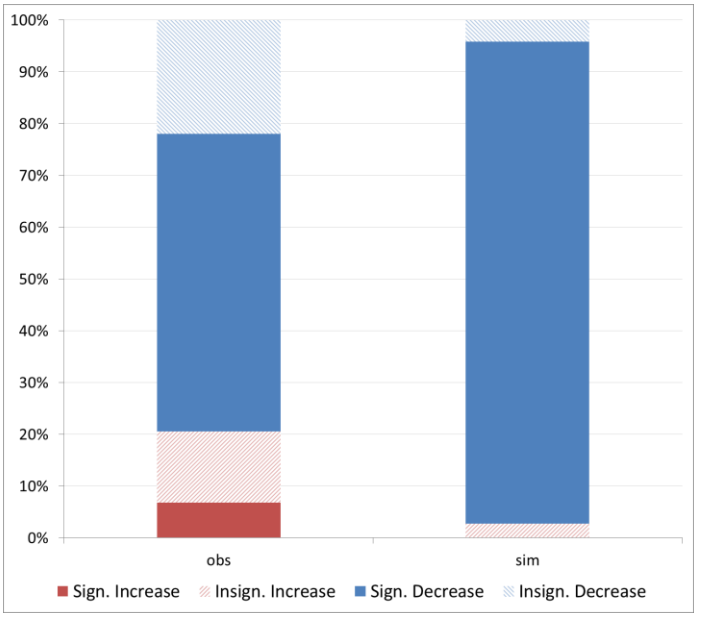

Figure 3: Percentage of sites where statistically significant upward trends (dark red), not significant upward trends (hashed dark
840 **red), significant downward trends (dark blue) and not significant downward trends (hashed dark blue) were obtained for NO_2 observations and simulated data.**

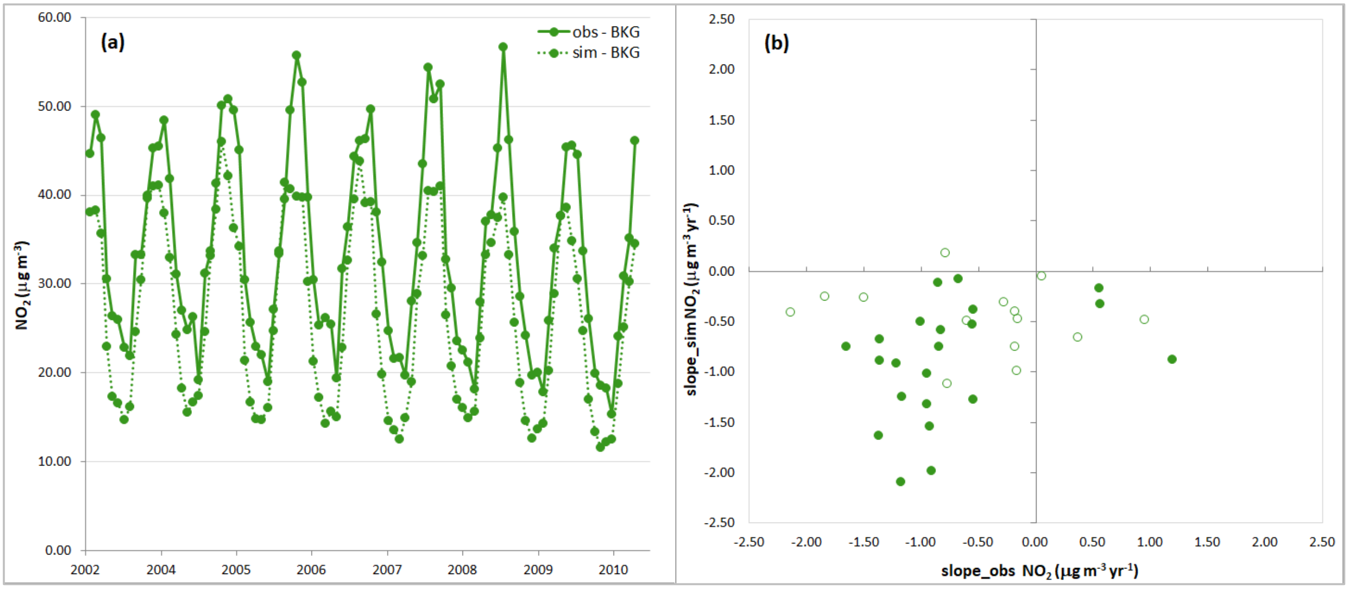

Figure 4: (a) Observed (solid line) and simulated (dashed line) monthly means of NO_2 concentrations (in μg m^{-3}) for all the
845 **background monitoring stations. (b) Scatter plot of observed and simulated slopes (in μg m^{-3} yr^{-1}) at each individual station. Sites where significant slopes are estimated for both observations and simulated data are indicated with a filled symbol.**

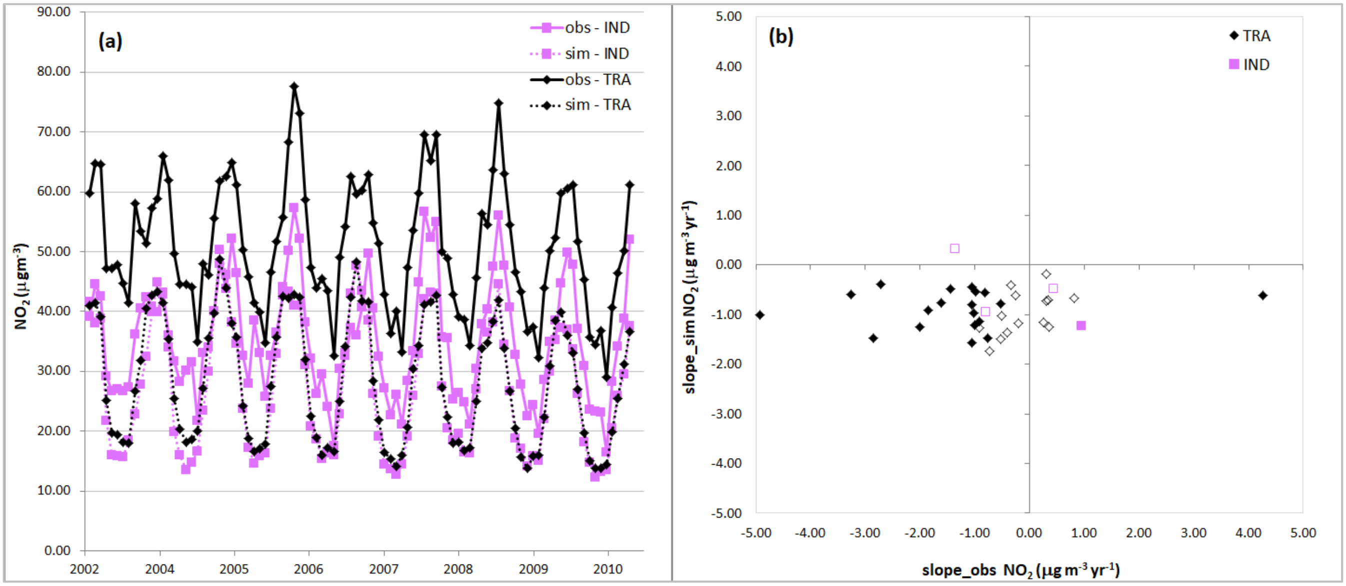

Figure 5: (a) Observed (solid line) and simulated (dashed line) monthly means of NO_2 concentrations (in µg m^{-3}) for all the traffic (black diamond) and industrial (pink square) monitoring stations. (b) Scatter plot of observed and simulated slopes (in µg m^{-3} yr^{-1}) at each individual station. Sites where significant slopes are estimated for both observations and simulated data are indicated with a filled symbol.

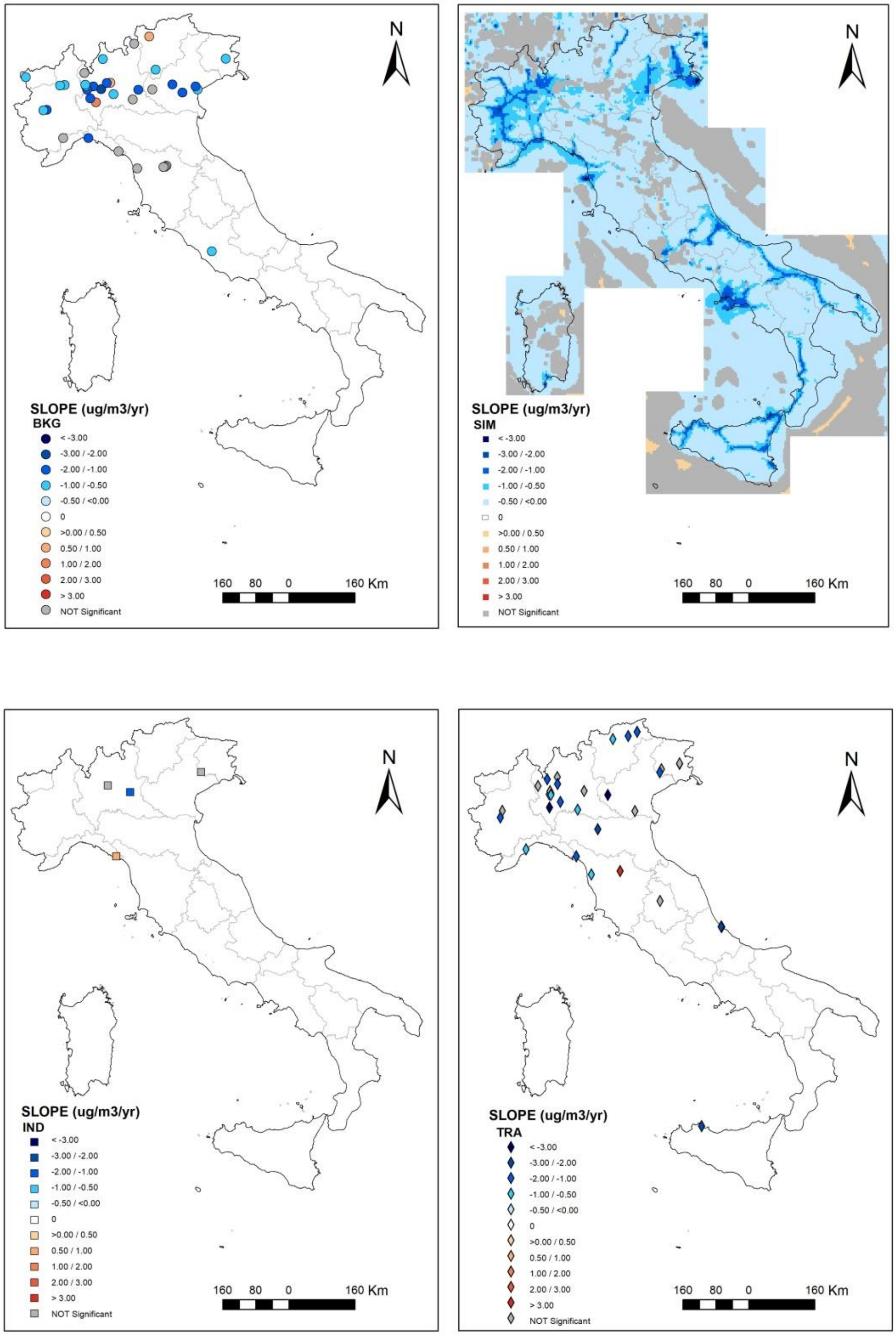

Figure 6: Slopes of NO_2 (µg m^{-3} yr^{-1}) observed at background (BKG – upper left panel), industrial (IND – lower left panel) and traffic (TRA – lower right panel) stations and simulated (upper right panel) slopes at each grid point. The grey symbols refer to not significant trends for both the observations and the simulated data.

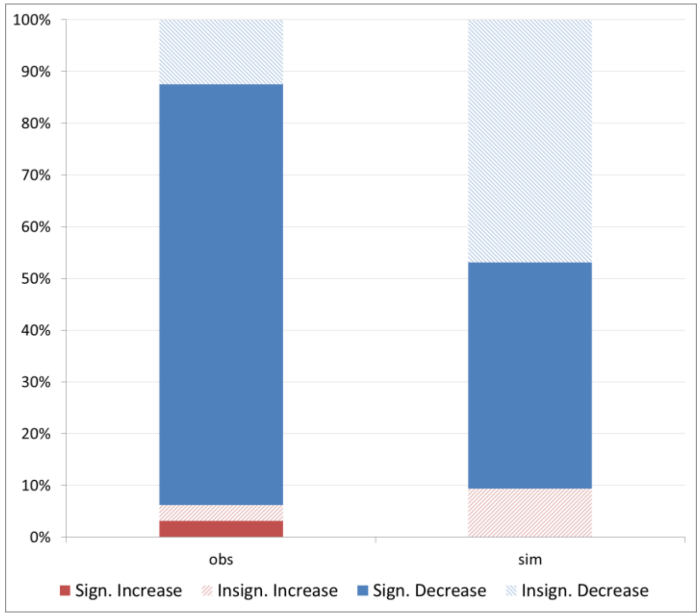

Figure 7: Percentage of sites where statistically significant upward trends (dark red), not significant upward trends (hashed dark red), significant downward trends (dark blue) and not significant downward trends (hashed dark blue) were obtained for PM10 observations and simulated data.

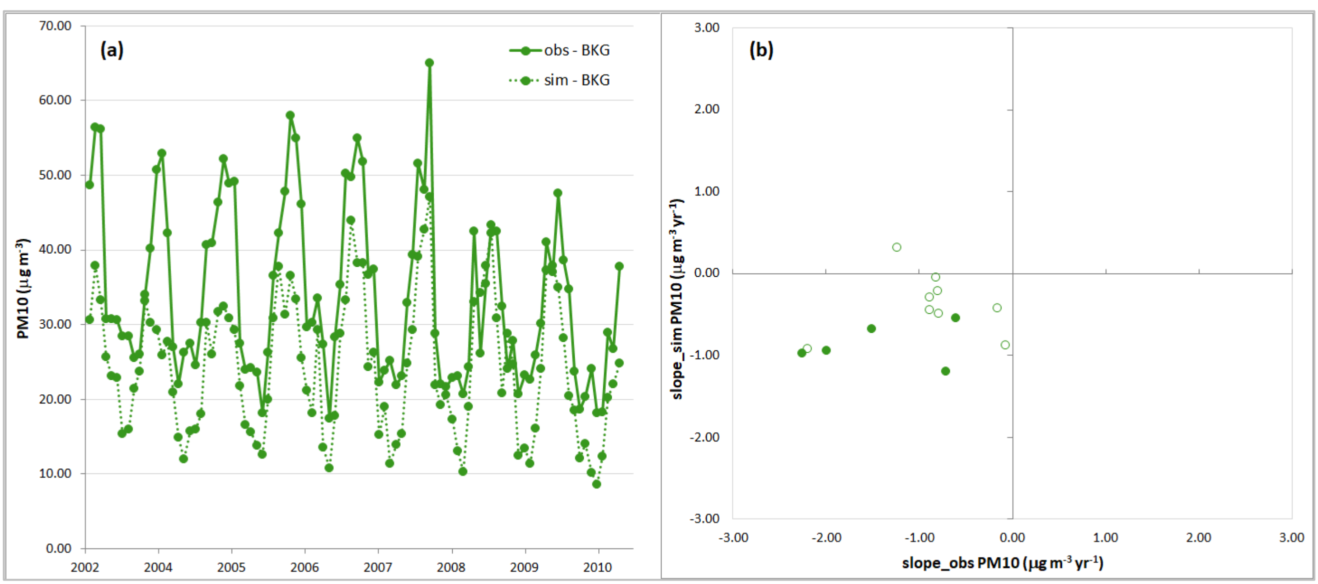

Figure 8: (a) Observed (solid line) and simulated (dashed line) monthly means of PM10 concentrations (in μg m^{-3}) for all the background monitoring stations. (b) Scatter plot of observed and simulated slopes (in μg m^{-3} yr^{-1}) at each individual station. Sites where significant slopes are estimated for both observations and simulated data are indicated with a filled symbol.

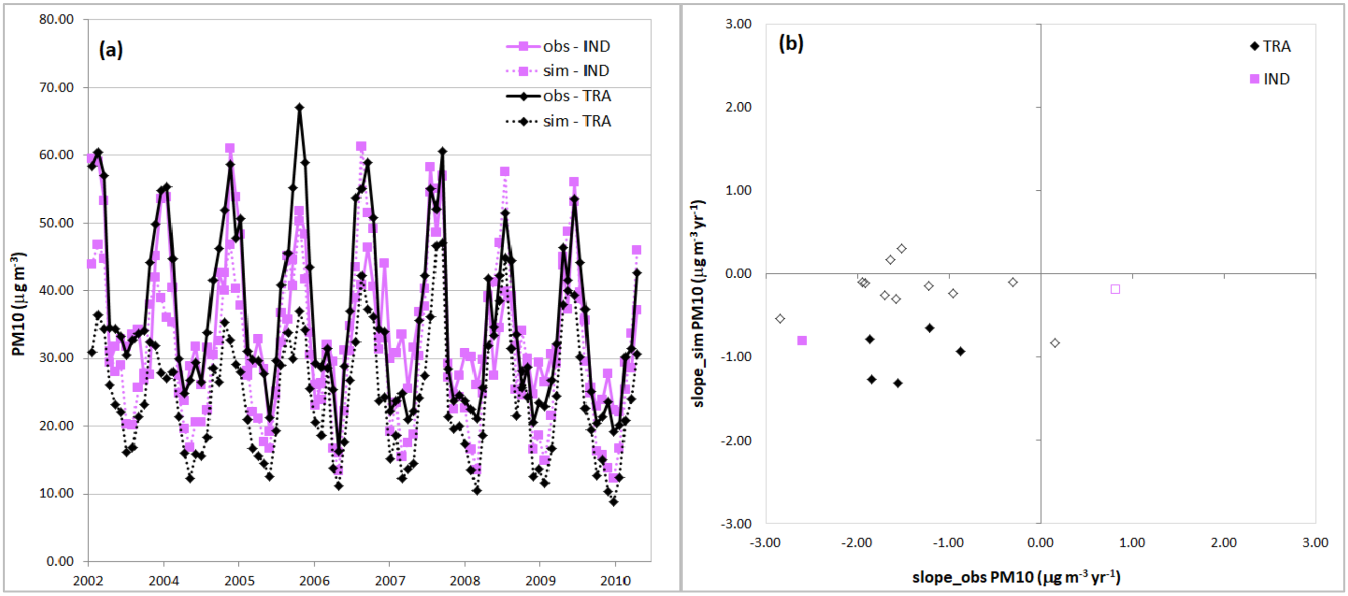

Figure 9: (a) Observed (solid line) and simulated (dashed line) monthly means of PM10 concentrations (in µg m^{-3}) for all the traffic (black diamond) and industrial (pink square) monitoring stations. (b) Scatter plot of observed and simulated slopes (in µg m^{-3} yr^{-1}) at each individual station. Sites where significant slopes are estimated for both observations and simulated data are indicated with a filled symbol.

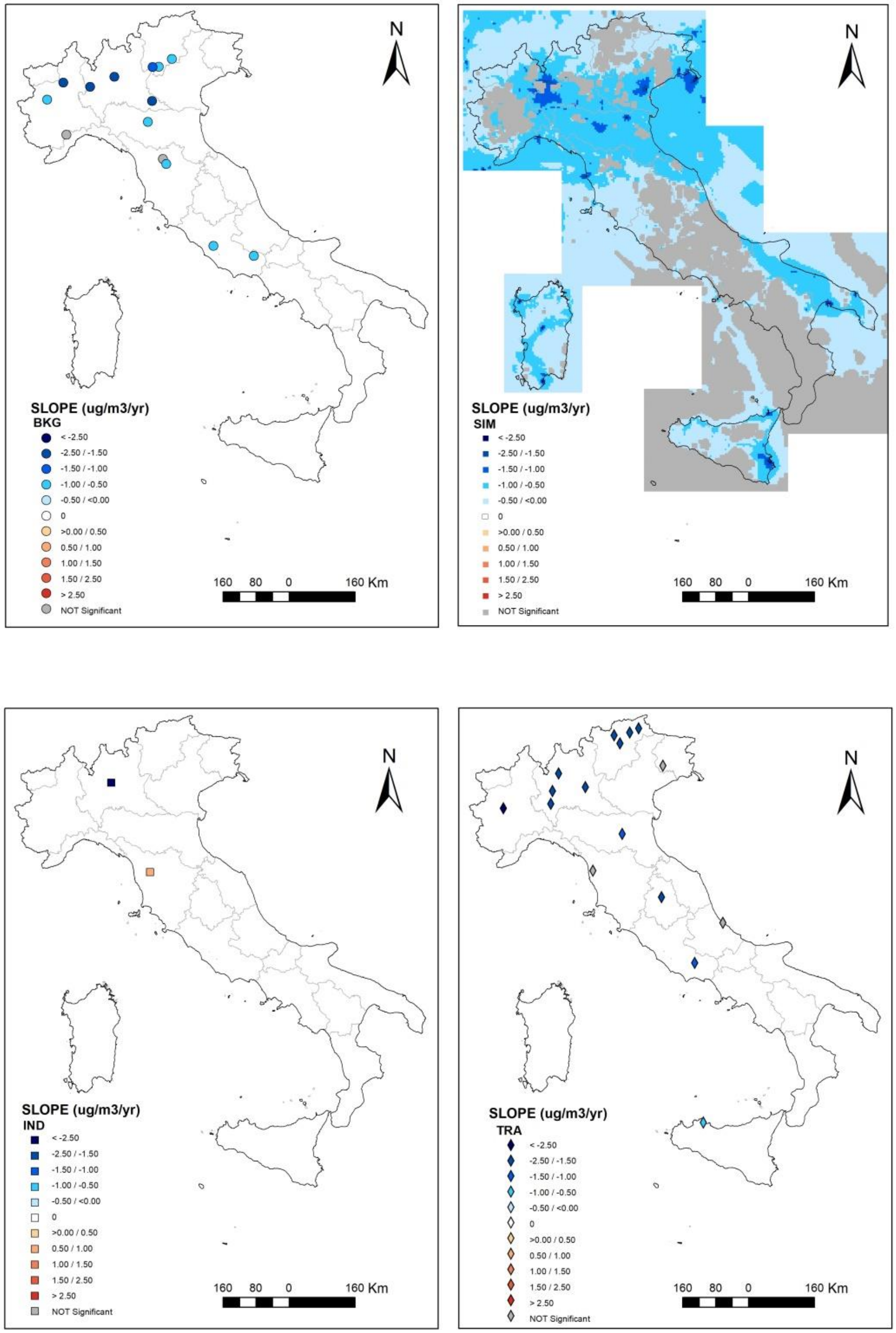

Figure 10: Slopes of PM10 ($\mu g\ m^{-3}\ yr^{-1}$) observed at background (BKG – upper left panel), industrial (IND – lower left panel) and traffic (TRA – lower right panel) stations and simulated (upper right panel) slopes at each grid point. The grey symbols refer to not significant trends for both the observations and the simulated data.

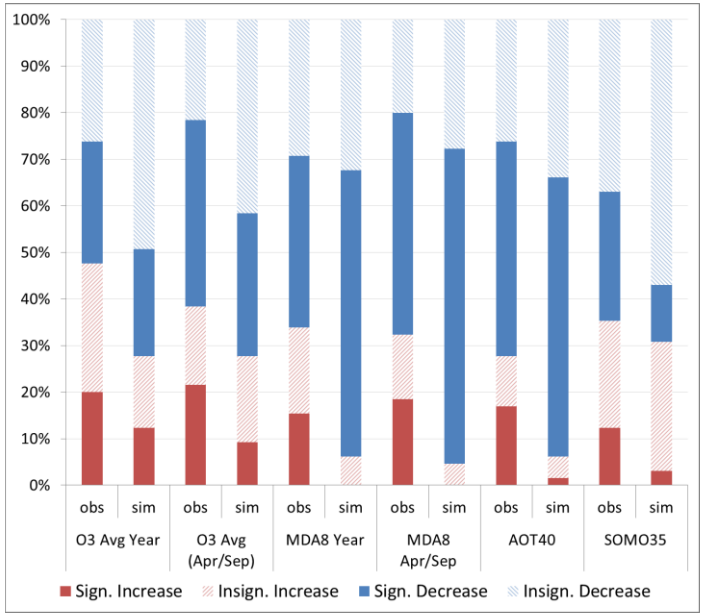

Figure 11: Percentage of sites where statistically significant upward trends (dark red), not significant upward trends (hashed dark red), significant downward trends (dark blue) and not significant downward trends (hashed dark blue) were obtained for different observed and simulated metrics for O_3.

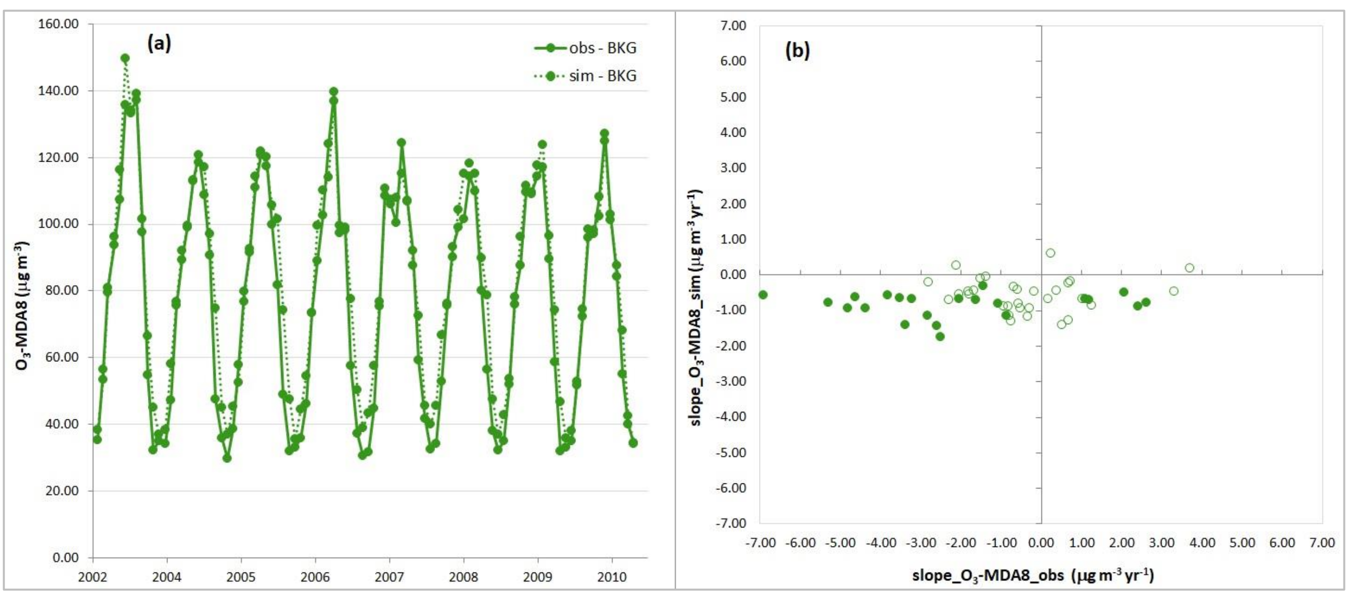

Figure 12: (a) Observed (solid line) and simulated (dashed line) monthly means of O_3-MDA8 concentrations (in µg m^{-3}) for all the background monitoring stations. (b) Scatter plot of observed and simulated slopes (in µg m^{-3} yr^{-1}) for O_3-MDA8 in the period April/September at each individual station. Sites where significant slopes are estimated for both observations and simulated data are indicated with a filled symbol.

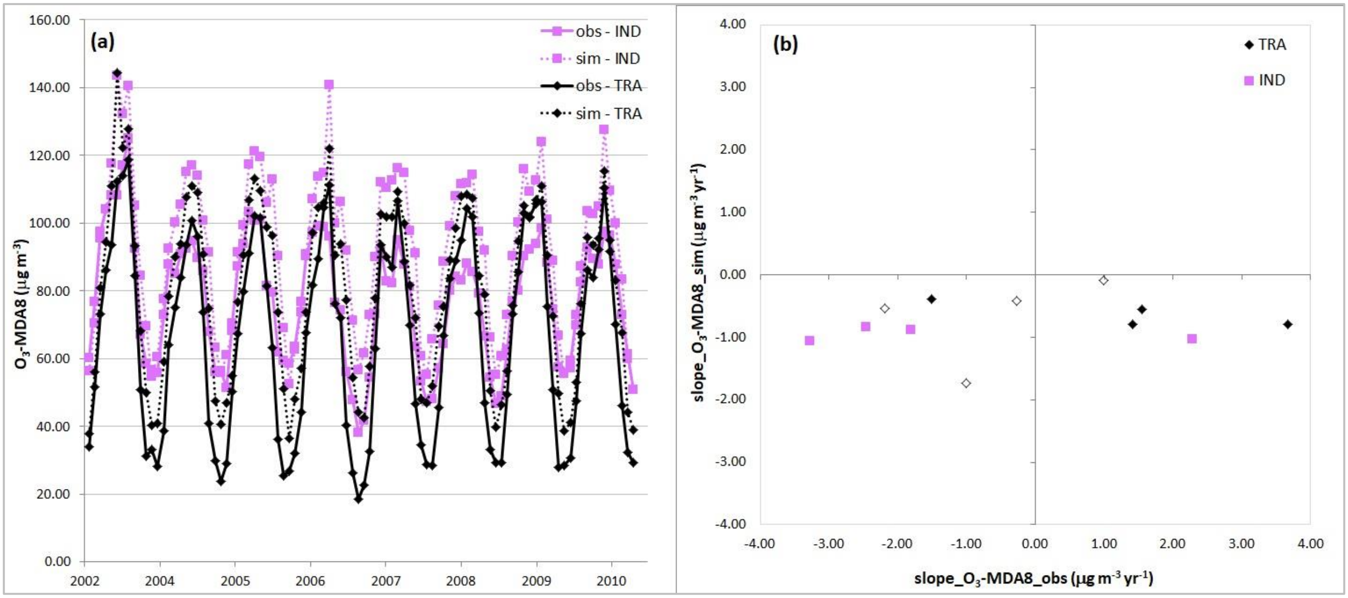

Figure 13: (a) Observed (solid line) and simulated (dashed line) monthly means of O_3-MDA8 concentrations (in µg m^{-3}) for all the traffic (black diamond) and industrial (pink square) monitoring stations. (b) Scatter plot of observed and simulated slopes (in µg m^{-3} yr^{-1}) for MDA8 in the period April/September at each individual station. Sites where significant slopes are estimated for both observations and simulated data are indicated with a filled symbol.

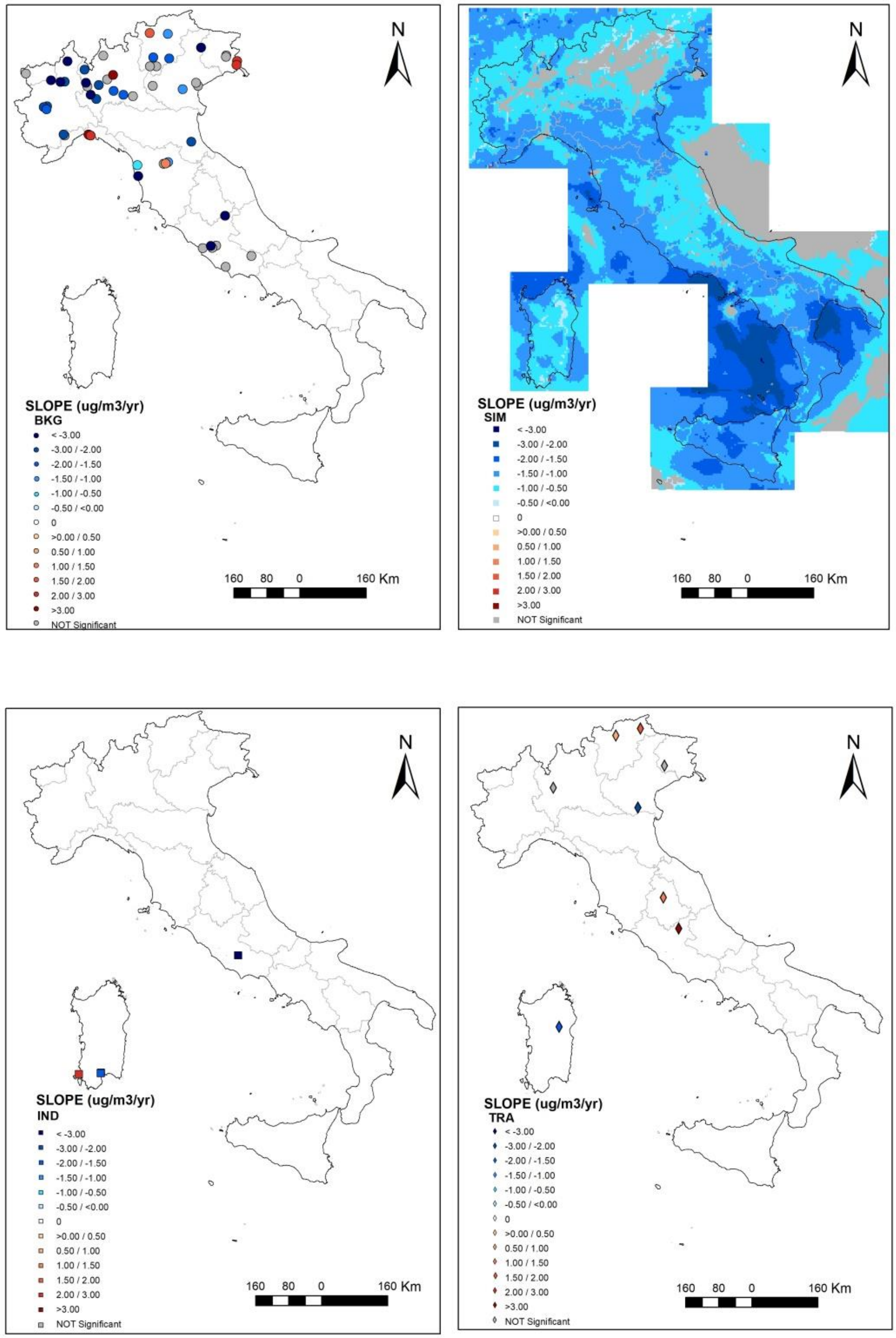

Figure 14: Slopes of O_3-MDA8 ($\mu g\ m^{-3}\ yr^{-1}$) observed at background (BKG – upper left panel), industrial (IND – lower left panel) and traffic (TRA – lower right panel) stations and simulated (upper right panel) slopes at each grid point. The grey symbols refer to not significant trends for both the observations and the simulated data.