

**L. VITALI, M. ADANI, L. CIANCARELLA, G. CREMONA
M. D'ISIDORO, M. MIRCEA, A. PIERSANTI, F. RUSSO**

Department for Sustainability
Division of Models and Technologies for Risks Reduction
Atmospheric Pollution Laboratory
Bologna Research Centre

G. BRIGANTI, A. CAPPELLETTI

Department for Sustainability
Division of Models and Technologies for Risks Reduction
Atmospheric Pollution Laboratory
Pisa Research Centre

I. D'ELIA

Department for Sustainability
Division of Models and Technologies for Risks Reduction
Atmospheric Pollution Laboratory
Rome Headquarters

M.G. VILLANI

Department for Sustainability
Division of Models and Technologies for Risks Reduction
Atmospheric Pollution Laboratory
Ispra Research Centre

G. RIGHINI, G. ZANINI

Department for Sustainability
Division of Models and Technologies for Risks Reduction
Bologna Research Centre

G. GUARNIERI

Department of Energy Technology
Division of system development for ICT
High Performance Computing Laboratory
Portici Research Centre

AMS-MINNI NATIONAL AIR QUALITY SIMULATION ON ITALY FOR THE CALENDAR YEAR 2015

Annual air quality simulation of MINNI Atmospheric Modelling System:
results for the calendar year 2015 and comparison with observed data

RT/2019/15/ENEA



ITALIAN NATIONAL AGENCY FOR NEW TECHNOLOGIES,
ENERGY AND SUSTAINABLE ECONOMIC DEVELOPMENT

L. VITALI, M. ADANI, L. CIANCARELLA, G. CREMONA
M. D'ISIDORO, M. MIRCEA, A. PIERSANTI, F. RUSSO

Department for Sustainability
Division of Models and Technologies for Risks Reduction
Atmospheric Pollution Laboratory
Bologna Research Centre

G. BRIGANTI, A. CAPPELLETTI

Department for Sustainability
Division of Models and Technologies for Risks Reduction
Atmospheric Pollution Laboratory
Pisa Research Centre

I. D'ELIA

Department for Sustainability
Division of Models and Technologies for Risks Reduction
Atmospheric Pollution Laboratory
Rome Headquarters

M.G. VILLANI

Department for Sustainability
Division of Models and Technologies for Risks Reduction
Atmospheric Pollution Laboratory
Ispra Research Centre

G. RIGHINI, G. ZANINI

Department for Sustainability
Division of Models and Technologies for Risks Reduction
Bologna Research Centre

G. GUARNIERI

Department of Energy Technology
Division of system development for ICT
High Performance Computing Laboratory
Portici Research Centre

AMS-MINNI NATIONAL AIR QUALITY SIMULATION ON ITALY FOR THE CALENDAR YEAR 2015

Annual air quality simulation of MINNI Atmospheric Modelling System:
results for the calendar year 2015 and comparison with observed data

RT/2019/15/ENEA



ITALIAN NATIONAL AGENCY FOR NEW TECHNOLOGIES,
ENERGY AND SUSTAINABLE ECONOMIC DEVELOPMENT

ACKNOWLEDGMENTS

The atmospheric modelling system AMS-MINNI used for the 2015 air quality simulation was developed in the framework of the MINNI project funded by the Italian Ministry for the Environment, Land and Sea (IMELS) and coordinated by ENEA since 2002.

The authors would like to thank all those who provided, in various ways, support and data for the present study.

The National Emission Inventory and its spatial disaggregation at provincial level were provided by the Italian Institute for Environmental Protection and Research (ISPRA). The authors are particularly grateful to Riccardo De Lauretis, Ernesto Taurino and Antonio Caputo.

ISTAT (Italian National Institute of Statistics) provided the data from the last survey on energy consumption of families, which was used for improving the spatial disaggregation of emissions from biomass burning attributed to the residential sector.

Observational data for meteorological fields evaluation were provided by the following Regional Environmental Protection Agencies: ARPA Piemonte, ARPA Lombardia, ARPA Emilia Romagna, ARPA Veneto, ARPA Friuli Venezia Giulia, ARPA Toscana, ARPA Lazio and ARPA Puglia.

The computing resources and the related technical support were provided by CRESCO/ENEAGRID High Performance Computing infrastructure and its staff. CRESCO/ENEAGRID High Performance Computing infrastructure is funded by ENEA and by Italian and European research programmes (<http://www.cresco.enea.it/english>).

I rapporti tecnici sono scaricabili in formato pdf dal sito web ENEA alla pagina www.enea.it

I contenuti tecnico-scientifici dei rapporti tecnici dell'ENEA rispecchiano l'opinione degli autori e non necessariamente quella dell'Agenzia

The technical and scientific contents of these reports express the opinion of the authors but not necessarily the opinion of ENEA.

AMS-MINNI NATIONAL AIR QUALITY SIMULATION ON ITALY FOR THE CALENDAR YEAR 2015

Annual air quality simulation of MINNI Atmospheric Modelling System: results for the calendar year 2015 and comparison with observed data

L. Vitali, M. Adani, G. Briganti, A. Cappelletti, L. Ciancarella, G. Cremona, I. D'elia, M. D'isidoro, G. Guarnieri, M. Mircea, A. Piersanti, G. Righini, F. Russo, M.G. Villani, G. Zanini

Riassunto

Il sistema modellistico atmosferico (AMS) del modello integrato nazionale MINNI, sviluppato da ENEA nell'ambito dell'omonimo progetto finanziato dal Ministero dell'Ambiente e della Tutela del Territorio e del Mare (MATTM), fornisce su tutto il territorio nazionale e su lungo periodo, tipicamente un anno, dati meteorologici e di qualità dell'aria con risoluzione temporale oraria e con risoluzione spaziale orizzontale di 4 km. Dal 2010, in adempimento del D.Lgs. 155/2010, ENEA è tenuta a elaborare ogni 5 anni, e per la prima volta con riferimento all'anno 2010, simulazioni modellistiche della qualità dell'aria su base nazionale e a rendere disponibili i risultati di tali elaborazioni.

In questo rapporto sono presentati i risultati della simulazione nazionale di AMS-MINNI relativa all'anno 2015, insieme alla loro validazione tramite il confronto con i dati di misura disponibili sul territorio nazionale. Allo scopo di fornire una valutazione oggettiva della qualità dei risultati della simulazione, basata su criteri condivisi dalla comunità scientifica, la validazione è stata effettuata seguendo l'approccio e la metodologia proposti da FAIRMODE e, in dettaglio, utilizzando il software DELTA Tool.

L'analisi dei risultati ha mostrato complessivamente la buona qualità dei campi di concentrazioni simulati. In particolare, tutti i Criteri di Qualità delle performances sono risultati soddisfatti per O₃ e PM_{2.5}. Sia punti di forza sia margini di miglioramento sono invece emersi dalla validazione di NO₂ e PM₁₀ che tendono ad essere globalmente sottostimati, come del resto accade comunemente nello stato dell'arte della modellistica a scala regionale.

In riferimento all'assessment di qualità dell'aria dell'anno meteorologico 2015 nell'ambito del D.Lgs. 155/2010, in accordo con i dati di misura raccolti da ISPRA, diverse criticità sono emerse per quanto riguarda il rispetto dei limiti di legge, in particolare in riferimento alla media giornaliera di PM₁₀, al massimo delle medie mobili su 8 ore di O₃ e alle medie annuali di NO₂ e PM_{2.5}.

I campi prodotti sono ora disponibili sia come condizioni iniziali e al contorno per studi a scala locale (come da D.Lgs. 155/2010), sia come dati in ingresso per analisi d'impatto a scala nazionale.

Parole chiave: qualità dell'aria, modellistica atmosferica, validazione.

Abstract

The Atmospheric Modelling System (AMS) of the Italian National Integrated Assessment Model MINNI, developed by ENEA and funded by the Italian Ministry for the Environment, Land and Sea (IMELS), may compute anthropogenic, biogenic and other natural emissions, meteorological parameters and air quality concentrations with hourly time resolution at a spatial resolution of 4 km over the Italian territory.

Since 2010 and every five years, ENEA is required to carry out national air quality simulations and to make the simulation results available for supporting air quality policy, in fulfillment of Legislative Decree 155/2010 (implementing EU Directive 2008/50/EC).

This report presents the AMS-MINNI national simulation for the year 2015, along with its validation by comparisons with available measurement data. In order to provide an objective assessment of the quality of AMS-MINNI results, with respect to criteria currently adopted and applied by the scientific community, FAIRMODE evaluation methodology and criteria, consolidated in the DELTA Tool software, were used for the validation.

An overall good quality of simulated concentrations fields was obtained. More in detail, all the Model Performance Criteria turned out to be satisfied for O₃ and PM_{2.5}. Concerning NO₂ and PM₁₀, both strengths and weaknesses are highlighted; in particular AMS-MINNI tends to underestimate both of them, as shown by other state of the art regional air quality models.

Concerning 2015 air quality assessment in the framework of Legislative Decree 155/2010, AMS-MINNI simulation, in agreement with monitoring data collected by ISPRA, pointed out the non-compliance with EU requirements, particularly for daily PM₁₀, 8h daily maximum O₃ and yearly NO₂ and PM_{2.5}. The simulated concentration fields are now available to be used as initial and boundary conditions for local scale air quality assessments (according to Legislative Decree 155/2010) and as input data for impact studies at the national level.

Key words: air quality, atmospheric modelling, validation.

INDEX

1 INTRODUCTION	7
2 DESCRIPTION OF AMS-MINNI AND SIMULATION SET UP	9
2.1 Meteorological data	10
2.1.1 RAMS model	10
2.1.2 Meteorological simulation set-up	11
2.2 Emission data	12
2.2.1 Anthropogenic emissions	12
2.2.1.1 Emission Manager	12
2.2.1.2 ISPRA Emission Inventory (Year 2015)	13
2.2.1.3 Updates of emission gridded disaggregation	15
2.2.1.4 ISPRA emission inventory updates of the year 2015: a comparison between the submissions 2017 and 2019	16
2.2.1.5 EMEP Emission Inventory (Year 2015)	18
2.2.2 Natural emissions	18
2.3 Pollutants transport, dispersion and chemical reactions	18
2.3.1 FARM model	18
2.3.2 Micro-meteorological input parameters for FARM	20
2.3.3 FARM simulation set-up	21
3 RESULTS	22
3.1 Annual statistics of concentration fields	22
3.2 Fields of hourly and daily reference concentration percentiles	27
4 EVALUATION OF MODEL PERFORMANCES AGAINST OBSERVED DATA	32
4.1 Methods for the evaluation: the DELTA tool approach	32
4.1.1 MQI for hourly and daily time series	33
4.1.2 MQI for yearly averages	34
4.1.3 MPC for high percentiles	34
4.1.4 MPC for spatial correlation and standard deviation	35
4.2 Results of the evaluation	35
4.2.1 Nitrogen dioxide (NO ₂)	38
4.2.2 Ozone (O ₃)	41
4.2.3 Particulate matter PM ₁₀	44
4.2.4 Particulate matter PM _{2.5}	47
4.3 Evaluation summary	50
5 CONCLUSION	51
REFERENCES	52
APPENDIX: STATISTICAL EVALUATION OF METEOROLOGICAL SIMULATION	62

1 INTRODUCTION

The European Air Quality Directive (hereafter AQD, EC, 2008) clearly stated the importance of using modelling approaches in addition to monitoring data in order to better carry out air quality assessment and planning. In Italy, in the framework of the MINNI (Italian National Integrated Model to support the international negotiation on atmospheric pollution) project, launched in 2002 and coordinated by ENEA, the Italian Ministry for the Environment, Land and Sea (IMELS) funded the development of the Italian Integrated Assessment Model MINNI (Ciancarella et al., 2013; Mircea et al., 2011, 2014, 2016), which takes advantages of the computing resources provided by CRESCO/ENEAGRID High Performance Computing infrastructure (Ponti et al., 2014). The integrated system MINNI, developed for supporting the international negotiation process on air pollution (D'Elia et al., 2013) and for assessing air quality policies at national/regional level (D'Elia et al., 2009; Ciucci et al., 2016), is currently in use in Italy for national policy and regulatory purposes (D'Elia et al., 2018).

Since 2010 and every five years, ENEA is required to carry out national air quality simulations and to make the simulation results available, in fulfilment of Legislative Decree 155/2010 (D.Lgs., 2010). After the AMS-MINNI simulation for the calendar year 2010 (Ciancarella et al., 2016), the simulation presented here, referring to the calendar year 2015, is the second one made available in the framework of Legislative Decree 155/2010 (art. 22, paragraph 5). In addition, several different calendar years were simulated by AMS-MINNI for national and international projects. Currently AMS-MINNI data base includes meteorological and air quality data for 1999, for the whole period from 2003 to 2010, and for 2015, which was recently added.

Moreover AMS-MINNI, and in particular the chemical transport model, integrated into the system, has been continuously upgraded to take into account the more recent findings in atmospheric pollution research (Silibello et al., 2012; Ciucci et al., 2014; Adani et al., 2015).

In recent years, MINNI data base has been widely used both for providing initial and boundary conditions for local scale air quality assessments and as input data for impact studies assessing air pollution effects on cultural heritage (De Marco et al., 2017), ecosystems (Manes et al., 2016) and human health both at local (Piersanti et al., 2018) and national level (De Marco et al., 2019). In particular, concerning health impact, multi-year MINNI modelled data were used within the MED HISS (Mediterranean Health Interview Surveys Studies) Life+ project (Cadum et al., 2016; Ghigo et al., 2017; Gandini et al., 2018, 2019), aiming at assessing health effects due to long-term exposure to main atmospheric pollutants in four European countries (Italy, Spain, France, Slovenia).

The expertise gained at national scale supported the extension of AMS-MINNI model implementation in other contexts. On one side, the same modelling system has been frequently applied at local scale with a higher spatial resolution. As an example, it was recently applied over the Campania Region at 1 km spatial resolution, in support to an intensive monitoring campaign carried out in the framework of CAMPANIA TRASPARENTE project (<http://www.campaniatrasparente.it/>). On the other side, the computational domain

was extended on European scale within the EURODELTA III model inter-comparison exercise (Bessagnet et al., 2016; Vivanco et al., 2017, 2018; Ciarelli et al., 2019; Theobald et al., 2019; Mircea et al., 2019).

Moreover, in the recent years, the same models composing the AMS-MINNI modelling suite were used for the development of the forecast operational modelling system FORAIR (https://www.afs.enea.it/project/ha_forecast/index.html), which currently provides, on a daily basis, three days air quality forecast data over Italy at 4 km resolution and over Europe at 20 km resolution. This expertise gave to MINNI the opportunity to join the Copernicus Services Data Providers (<https://www.copernicus.eu/en>) as one of the candidate models for the CAMS Regional Service (CAMS_50). The purpose of this service is to provide atmospheric composition data (daily near-real time forecasts and analyses, and annual re-analyses) at European scale, making them freely and easily available to wide user communities (<http://macc-raq-op.meteo.fr/index.php>).

This report presents the results of the national AMS-MINNI air quality simulation, for the calendar year 2015, carried out by ENEA, in fulfilment of Legislative Decree 155/2010. The AMS-MINNI modelling system and the main features of the simulation set up are described in Chapter 2. In Chapter 3 the results of AMS-MINNI simulation are presented for all the pollutants regulated by the AQD, in terms of both annual mean concentrations and hourly or daily reference concentration percentiles. The evaluation of modelled atmospheric composition fields, by means of comparisons against observational data, is provided in Chapter 4, whereas the evaluation of the meteorological simulation is described in the Appendix. Chapter 5 outlines the main findings and the conclusions.

2 DESCRIPTION OF AMS-MINNI AND SIMULATION SET UP

AMS-MINNI calculates concentrations by means of a Chemical Transport Model implementing an integrated and multi-pollutant approach. In Figure 1 the main components of AMS-MINNI are presented. The core of the modelling system is the three-dimensional Eulerian model **FARM** (**F**lexible **A**ir **Q**uality **R**egional **M**odel; Gariazzo et al., 2007; Silibello et al., 2008; Kukkonen et al., 2012) that includes transport, turbulent dispersion and multiphase chemistry of pollutants in the atmosphere. Meteorological fields are produced by mean of the three-dimensional non-hydrostatic meteorological model **RAMS** (**R**egional **A**tmospheric **M**odelling **S**ystem; Cotton et al., 2003). The hourly gridded emissions used by FARM are prepared by the emission processor **Emission Manager** (Arianet, 2014) that breaks down annual data from emission inventories by applying daily, weekly and seasonal activity profiles, gridded spatial proxies and activity-related chemical speciation profiles. The modelling system is completed by the diagnostic module **SURFPRO** (**SURF**ace-atmosphere interface **PRO**cessor; Arianet, 2011), providing both meteorological and emission variables: from meteorological fields and from orographic and land use data, SURFPRO computes Planetary Boundary Layer (PBL) scale parameters, horizontal and vertical diffusivity coefficients, deposition velocities for different chemical compounds and natural emissions.

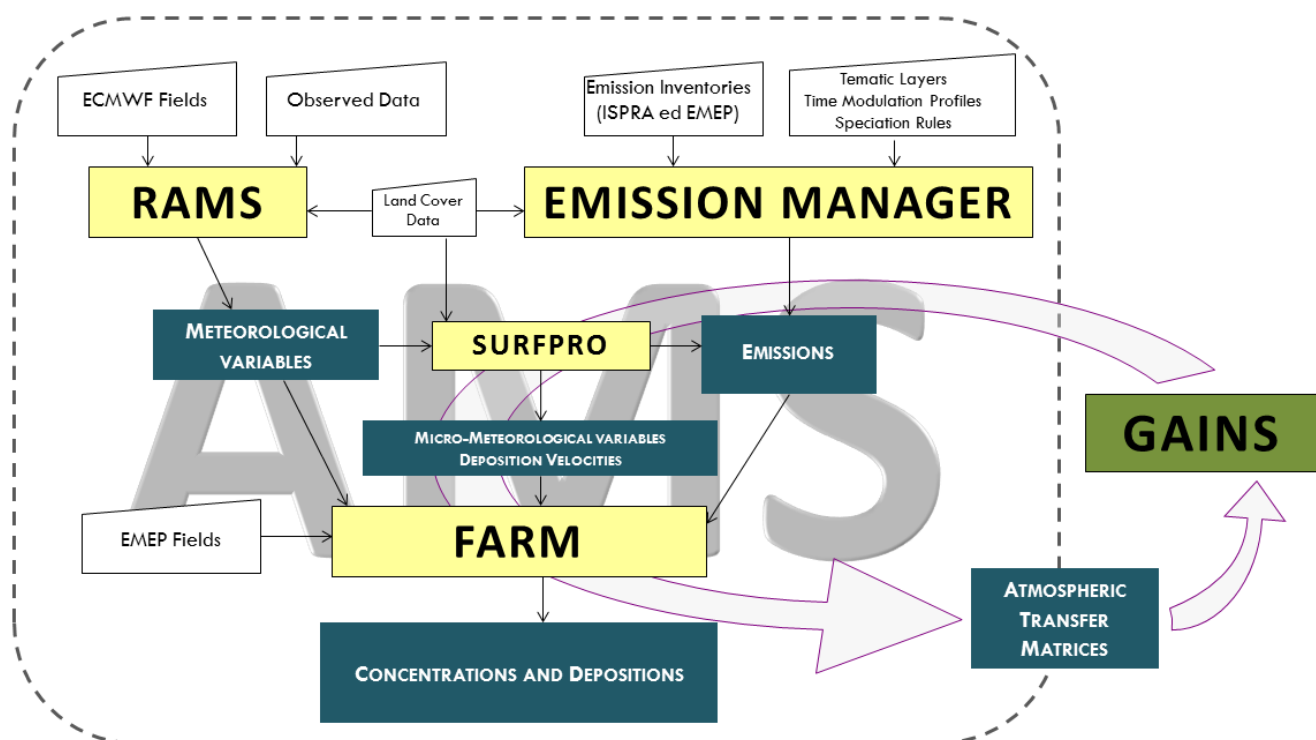


FIGURE 1. AMS-MINNI MODELLING SYSTEM.

AMS-MINNI calculates 3D fields of hourly concentrations of all pollutants regulated by the Air Quality Directives. Concentration fields are provided at 4 km resolution over the whole Italian territory. The computational domain is shown in Figure 2.

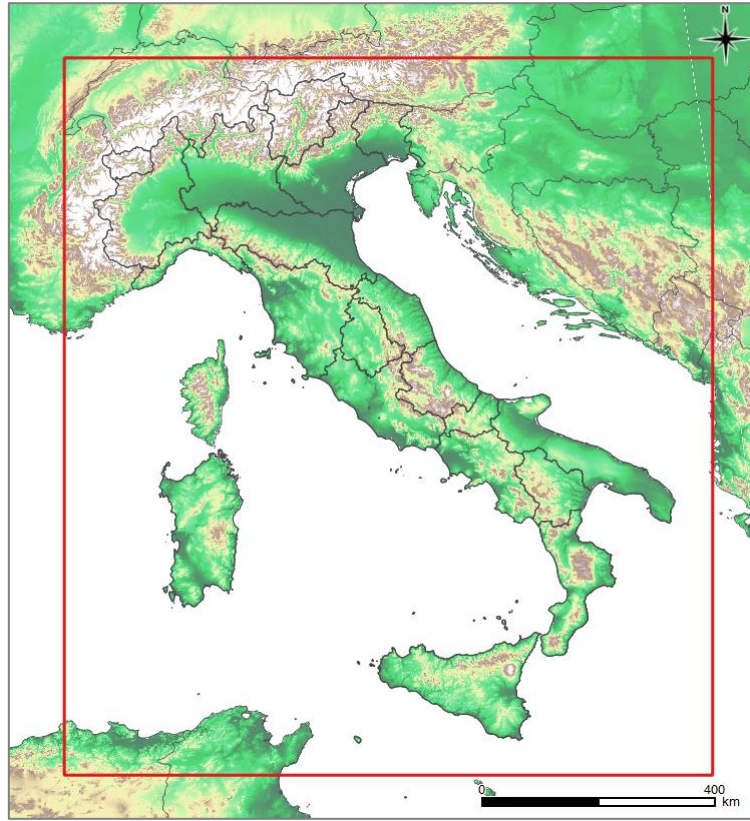


FIGURE 2. AMS-MINNI SIMULATION DOMAIN.

2.1 Meteorological data

2.1.1 RAMS model

RAMS is a prognostic non-hydrostatic meteorological model, based on terrain following vertical coordinate system and rotated polar stereographic projections. RAMS is highly customizable for a wide range of applications, specifically for different spatial scales. In particular, a multiple grid nesting scheme allows for the solution of model equations simultaneously on any number of interacting computational meshes at differing spatial resolution. Both one-way and two-way communication of all prognostic variables between any nested grids (at fine resolution) to its parent grid (at coarse resolution) can be alternatively activated. All these features make RAMS suitable for high-resolution simulations, especially in case of complex terrain environments.

The model implements a full set of non-hydrostatic, compressible Reynolds-averaged primitive equations, together with conservation equations for scalar quantities. Several parameterisation schemes are available to describe turbulent diffusion, solar and terrestrial radiation, moist processes, kinematic effects of terrain, cumulus convection, and sensible and latent heat exchange between the atmosphere and the surface. In particular the latter one is described through the integration within RAMS of the **LEAF-2** module (**L**and **E**cosystem–**A**tmosphere **F**eedback model; Walko et al., 2000), a prognostic model for the representation of temperature and water content status and evolution within soil, snow cover, vegetation, and canopy air, including turbulent and radiative exchanges between these components and the atmosphere.

2.1.2 Meteorological simulation set-up

RAMS runs for the national 2015 MINNI simulation were carried out on two-way nested domains, in nudging mode, i.e., assimilating analysis fields during model runs. A forcing term is added to the dynamical equations, driving the model to more closely follow the meteorological observations. Analysis fields for the assimilation were produced by means of the RAMS pre-processor **ISAN** (**IS**entropic **AN**alysis), that implements an optimal interpolation method based on Barnes algorithm (Barnes, 1964). ECMWF (European Centre For Medium-Range Weather Forecast) analyses fields, available every 6 hours, and surface synoptic data (temperature, pressure, and wind) from the WMO (World Meteorological Organization) network were used as input data for ISAN. In Figure 3 the positions of all the available WMO monitoring sites are shown.

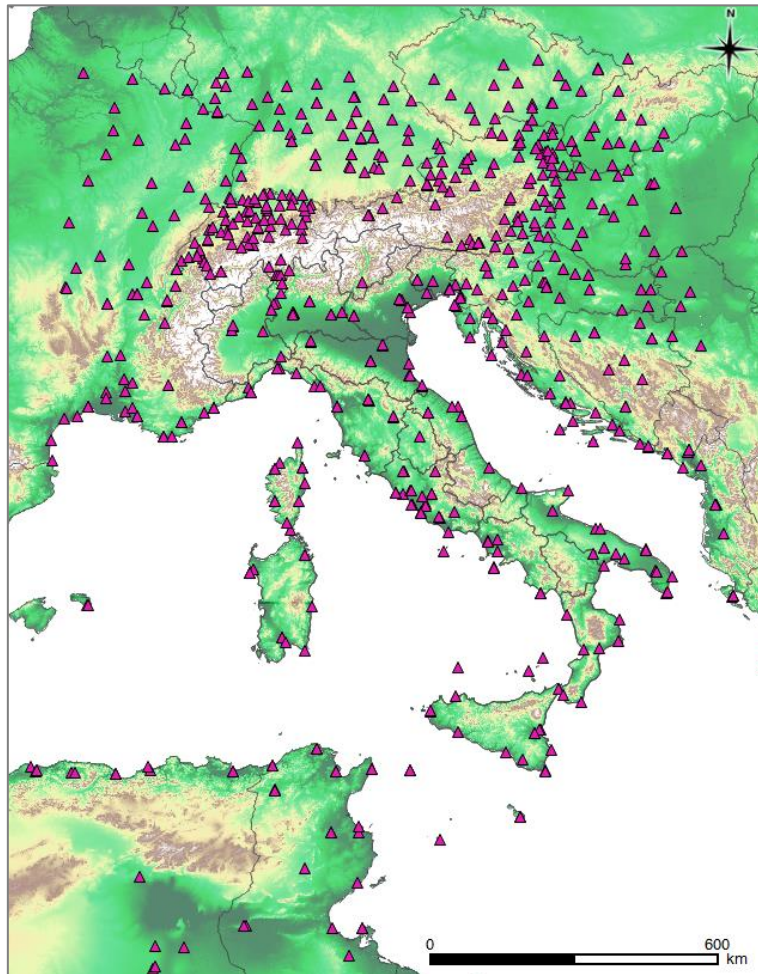


FIGURE 3. WMO METEOROLOGICAL STATIONS FOR DATA ASSIMILATION.

Main features of meteorological simulation set-up are described in Table 1.

<i>Radiation</i>	<i>Chen and Cotton (1983) long/shortwave model – cloud processes considering all condensate as liquid</i>
<i>Convection</i>	<i>Modified Kuo scheme (Tremback,1990)</i>
<i>Lower Boundary</i>	<i>LEAF-2, Land Ecosystem–Atmosphere Feedback model (Walko et al, 2000)</i>
<i>Turbulence Closure</i>	<i>Mellor-Yamada level 2.5 scheme – ensemble–averaged TKE (Mellor and Yamada, 1982)</i>
<i>Cloud Microphysics</i>	<i>Bulk microphysics parameterization: cloud water, rain, pristine ice, snow, aggregates, graupel, and hail, or certain subsets of these (Walko et al ,1995)</i>
<i>Data Assimilation</i>	<i>Nudging on pre-analysed fields</i>

TABLE 1. MAIN FEATURES OF METEOROLOGICAL SIMULATION SET-UP.

Among hourly meteorological data provided by RAMS, temperature, wind speed, relative humidity and precipitation have got a leading role in the chemical and physical processes that take place into the atmosphere and determine the levels of pollutant concentrations. Their quality was evaluated, through the comparison with observed data, and the outcomes of the validation are described in the Appendix.

2.2 Emission data

2.2.1 Anthropogenic emissions

2.2.1.1 EMISSION MANAGER

In the framework of AMS-MINNI, input anthropogenic emissions are prepared by means of the Emission Manager (Arianet, 2014), a modular pre-processing system allowing to compute model-ready emission inputs (hourly, gridded, chemical speciated) starting from one or multiple emission data bases. It can manage sources with different types of geometry: point (e.g. industrial facilities stacks, energy production plants, etc.), area (e.g. residential heating, agriculture, etc.) and line (e.g. traffic, shipping lanes, etc.).

The main input data and information used by the Emission Manager are:

- *emission data* associated to point, line and area sources; such emission data can come from an inventory (e.g. on a yearly basis), from automatic monitoring system, or from other modelling systems (e.g. a traffic emissions module) relative to different pollutants, different administrative levels and emission categories where a classification scheme is adopted (e.g. the SNAP (Selective Nomenclature for Air Pollution) nomenclature by the EEA);
- *geographic information* in vector or raster format, describing the geometry of complex sources (polygons for area sources and networks of line sources) or to be used when disaggregating in space the emission data for area sources (e.g. thematic layer on land use, population data, etc.);

- *time modulation data*, to describe the typical temporal profiles in months, day of the week and hours for each emitting sources when the emissions are coming from an inventory on e.g. yearly basis;
- *species split/speciation data*, speciation profiles for each emission activities and aggregated chemical species (e.g. “total NMVOC”, particulate matter) to separate emissions into individual species and required granulometry needed by the target model.

Point sources are individually characterized with geographical coordinates and physical and thermodynamic parameters (stack height and diameter, gas exit speed and temperature) so that plume rise effect can be taken into account.

Figure 4 shows the work flow of the Emission Manager for the preparation of an emission input for the AMS-MINNI.

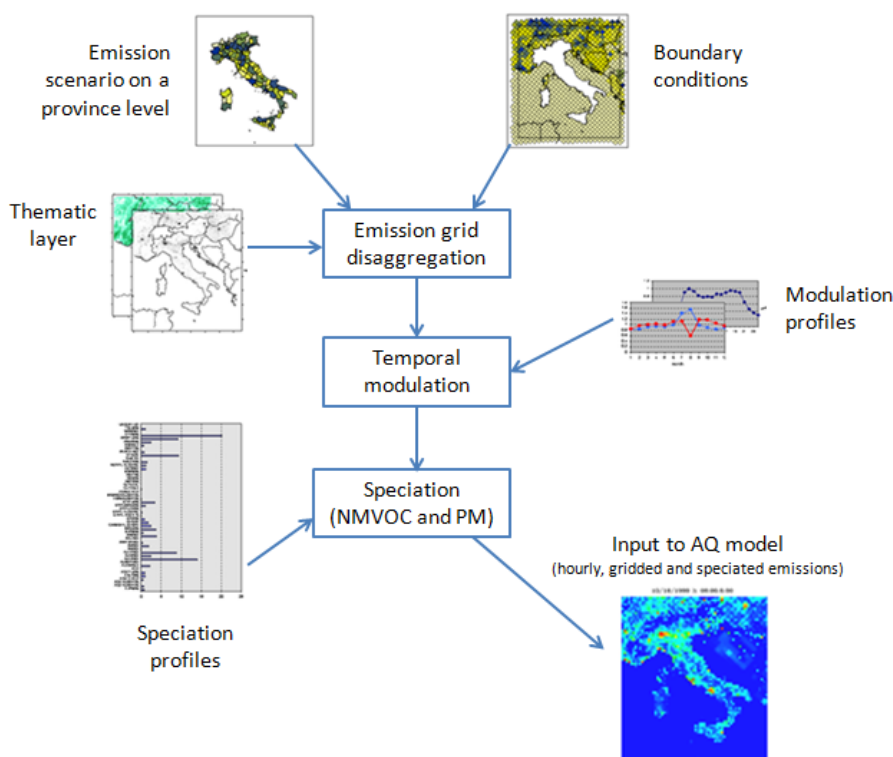


FIGURE 4. EMISSION MANAGER WORKFLOW TO BUILD AN EMISSION INPUT FOR MODELLING SIMULATIONS.

2.2.1.2 ISPRA EMISSION INVENTORY (YEAR 2015)

The emission input for the Emission Manager is elaborated starting from the national emission inventory prepared by ISPRA (Italian Institute for Environmental Protection and Research) both on a national level (NUTS1) and on a provincial level (NUTS3) (where NUTS stands for Nomenclature of territorial units for statistics, the hierarchical system for dividing up the territory of the European Union).

As stated by the Legislative Decree 155/2010 and then by the Legislative Decree 81/2018 (D.Lgs., 2018), ISPRA is responsible for: i) the national emission inventory, which has to be updated every year; ii) the

spatially disaggregated national emission inventory; and iii) the large point sources inventories that have both to be prepared and updated every four year. ENEA is responsible for the national emission projections that have to be updated and prepared every two years. The annual emission data have to be reported by 15 February while the Informative Inventory Report (IIR) by 15 March. The date when emissions are reported is defined as the national submission for the inventory in that year. Re-submissions due to errors are allowed but shall be provided within four weeks at the latest and include a clear explanation of the changes made. The spatial disaggregation of national emissions on a provincial level is described in ISPRA (2009) where details on methodological issues and proxies used for each sector are provided using the SNAP (Selective Nomenclature for Air Pollution; EEA, 2000) classification by activity level. In addition, since 2017, ISPRA has been providing spatial disaggregation on the EMEP model grid at 0.1° spatial resolution (about 10 km), as required by international protocols on air quality (see paragraph 2.2.1.5 for details on EMEP model and data) and by the new National Emission Ceilings Directive (EC, 2016).

The present simulation was realized using the ISPRA emission inventory disaggregated on a provincial level and downloaded by the official website in April 2019, <http://www.sinanet.isprambiente.it/it/sia-ispra/inventaria/disaggregazione-dellinventario-nazionale-2015/view>. As stated by the Legislative Decree 155/2010 (art. 22, c3 and 5), ISPRA has to provide the national emission inventory disaggregated on a provincial level every 5 years and for the first time in the year 2010; thereafter, ENEA in cooperation with ISPRA has to disaggregate emissions on a grid level fit for the AMS-MINNI model within 6 months from the official release of the provincial inventory. The provincial inventory was released by ISPRA in March 2018. The ENEA air quality simulations for the year 2015 were produced in October 2019 with the present report in December 2019.

A comparison on national level between the submission 2017, officially used by ISPRA for the provincial emission inventory, and the more recent submission, 2019, is presented in paragraph 2.2.1.4.

Total national annual emissions used for the present simulation are reported in Table 2 with the SNAP classification on macro-sector level.

SNAP - macrosector		EMISSIONS 2015 (ton/year)					
SNAP code	Description	SO ₂	NO _x	NMVOC	NH ₃	PM10	PM2.5
01	Combustion in energy and transformation industries	29677	51633	3694	194	1471	1095
02	Non-industrial combustion plants	9642	86846	199445	1664	111702	110473
03	Combustion in manufacturing industry	26870	64626	6603	663	6734	5636
04	Production processes	30393	9435	54799	452	11241	7580
05	Extraction and distribution of fossil fuels and geothermal energy	0	0	37533	0	613	61
06	Solvent use and other product use	0	0	352801	0	11	11
07	Road transport	375	394259	143734	6177	21858	17981
08	Other mobile sources and machinery	25050	180392	32610	25	10385	10358
09	Waste treatment and disposal	4437	2425	11023	6203	2557	2195
10	Agriculture	0	24662	1203	377937	12792	4875
	TOTAL	126443	814278	843444	393316	179364	160265

TABLE 2. ANNUAL ANTHROPOGENIC EMISSIONS (TON/YEAR) OF THE MAIN POLLUTANTS FOR SNAP CATEGORIES (ISPRA, 2018).

Furthermore, dust emissions from road transport resuspension were included (20290 tons PM10), obtained from emission factors measured in Italy and Spain (Amato et al., 2012; Padoan et al., 2018) together with the EPA algorithm AP-42, (<http://www.epa.gov/ttn/chief/ap42/ch13/bgdocs/b13s0201.pdf>). Dust emissions from agricultural activities (harvest, threshing and ploughing; 5733 tons PM10) were added as well, relying on the emission factors measured in Italy by Telloli et al., 2017.

2.2.1.3 UPDATES OF EMISSION GRIDDED DISAGGREGATION

In the present simulation, particular attention was devoted to the spatial disaggregation of emissions from the SNAP sector '02' – Non industrial combustion plants, and in particular, of the sector 02020201 where emissions from biomass-fuelled heat generators in the residential sector have been computed. In the national emission inventory, emissions from this sector are disaggregated on a provincial level using population as proxy (ISPRA, 2009). The last ISTAT (Italian National Institute of Statistics) survey on energy consumption in families, relative to the year 2013 (ISTAT, 2014) updated not only the energy consumptions on national level but also its regional distribution dividing the Italian municipalities in seven areas, depending on population and altimetry. In particular, the municipalities were divided in the following zones:

- centre municipalities of metropolitan areas;
- external municipalities of metropolitan areas;
- municipalities with more than 50000 inhabitants;
- mountain/hill municipalities between 10000 and 50000 inhabitants;
- mountain/hill municipalities up to 10000 inhabitants;

- plain municipalities between 10000 and 50000 inhabitants;
- plain municipalities up to 10000 inhabitants.

The differences on a provincial level of this new disaggregation for the PM2.5 emissions in the SNAP sector 02020201 are mostly due to a more realistic allocation of biomass consumption, which is higher in areas of wood production (mountain forests) and lower in populated urban areas where natural gas networks largely cover residential consumption.

The subsequent disaggregation on the 4 km grid for the 02020201 sector was realized elaborating a new layer based on the distribution of low buildings (1-2 floors, defined in the following as “E2P layer”) instead of population as in previous simulations. This assumption is based on the empirical evidence that solid biomass is mainly used for heating small private households. The resulting spatially gridded disaggregation for PM2.5 emissions in the 02020201 sector is shown in Figure 5.

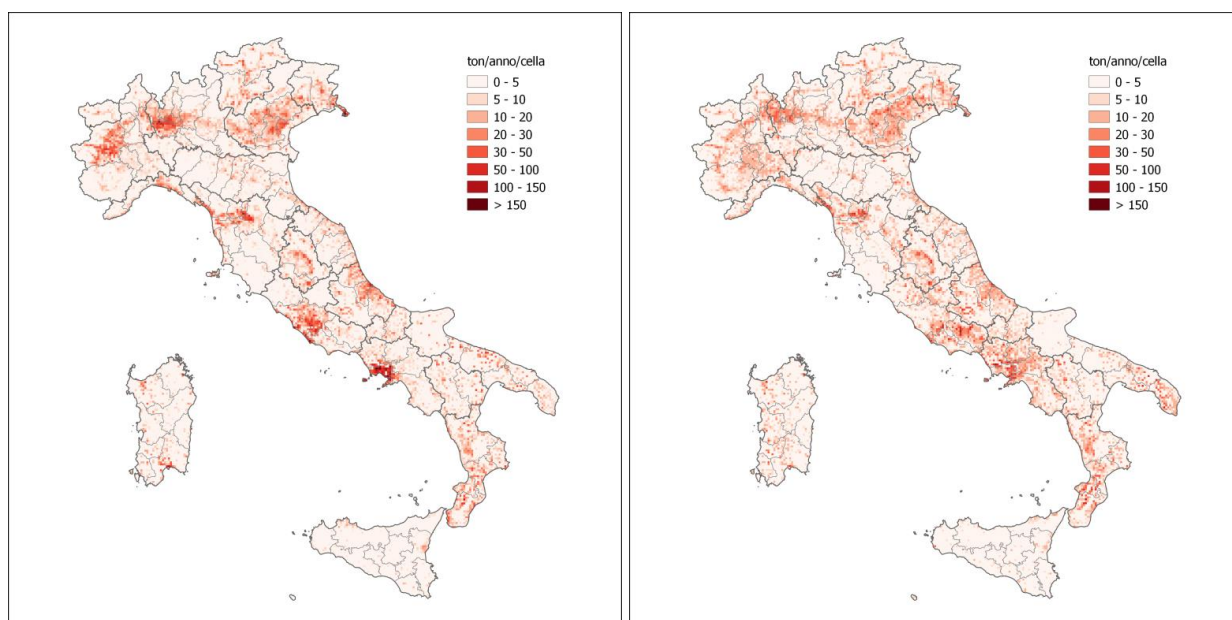


FIGURE 5. GRIDDED PM2.5 EMISSIONS OF THE 02020201 SECTOR (4 km HORIZONTAL RESOLUTION) USING THE E2P LAYER AND ON A PROVINCIAL LEVEL THE ISPRA DISAGGREGATION BASED ON POPULATION (ON THE LEFT) AND THE ISTAT DISAGGREGATION IN 7 MUNICIPALITY ZONES (ON THE RIGHT).

2.2.1.4 ISPRA EMISSION INVENTORY UPDATES OF THE YEAR 2015: A COMPARISON BETWEEN THE SUBMISSIONS 2017 AND 2019

As stated in paragraph 2.2.1.2, the emissions for the 2015 simulation come from the latest ISPRA emission inventory disaggregated on a provincial level, updated to the submission 2017. In the meantime, on a national level, more updated submissions were made available. The following Figure shows the differences on 2015 emissions on a national level and by SNAP sector for the pollutants reported in Table 2.

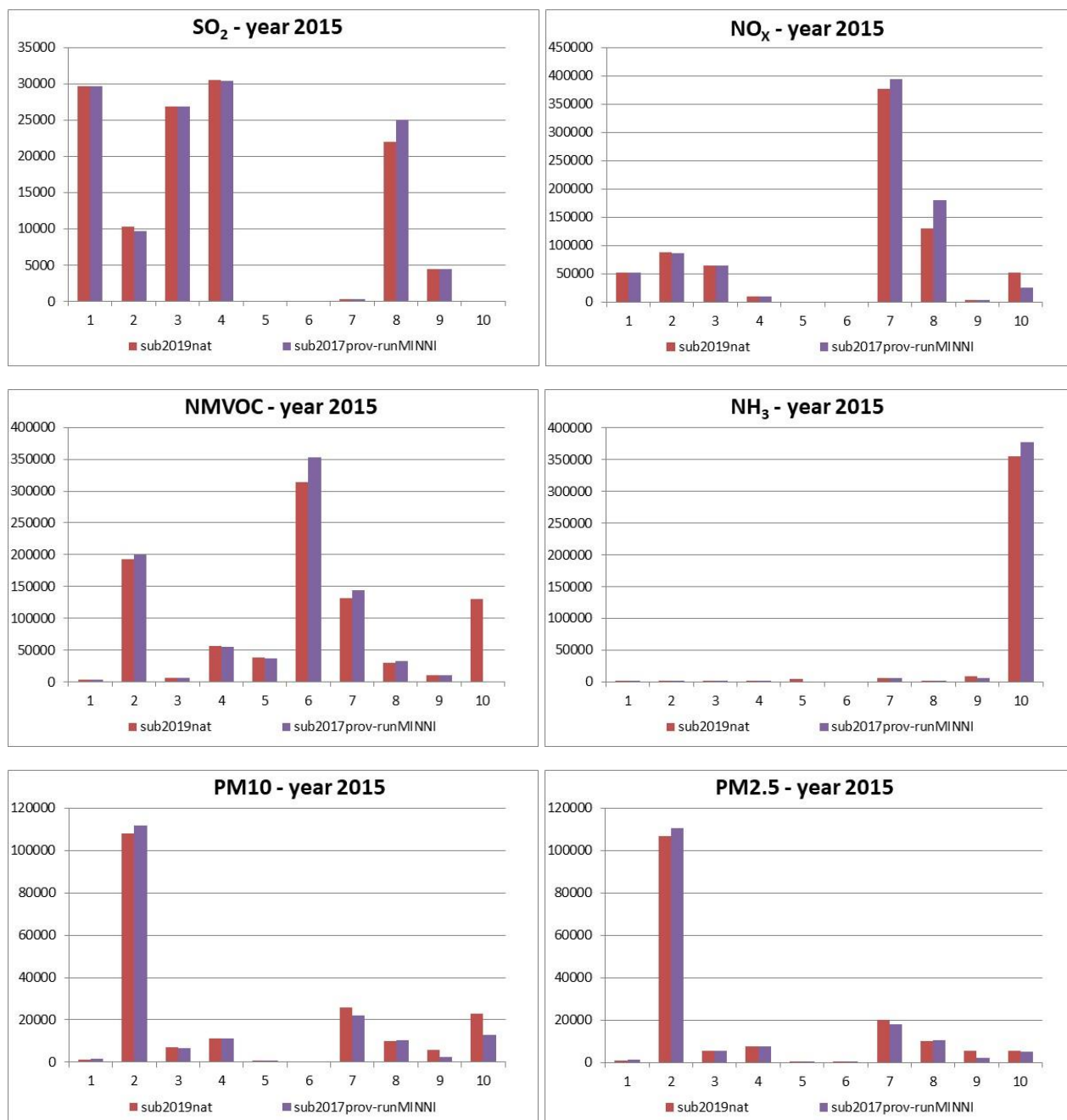


FIGURE 6. 2015 EMISSIONS: COMPARISON BETWEEN THE NATIONAL 2017 SUBMISSION (USED TO DISAGGREGATED EMISSIONS ON A PROVINCIAL LEVEL, IN PURPLE) AND THE 2019 SUBMISSION (IN RED).

The previous Figure shows slight changes in the maritime sector (SNAP 08) for SO₂ and NO_x, in the civil sector (SNAP 02) for PM₁₀ and PM_{2.5}, in the solvent sector (SNAP 06) for NMVOC, in the waste sector (SNAP 09) for PM₁₀ and PM_{2.5}, in the agricultural sector (SNAP 10) for NH₃, PM₁₀ and PM_{2.5}. The only significant update was introduced in the agricultural sector for NMVOC, due to the full implementation of the 2016 Guidebook EMEP/EEA emission factors on manure management. The comparison between 2017 and 2019 submissions shows that ISPRA emission estimates used in the present study are reasonably up-to-date and thus fit for feeding an air quality simulation of 2015.

2.2.1.5 EMEP EMISSION INVENTORY (YEAR 2015)

Concerning the anthropogenic sources of the other countries included in the simulation domain, the 2015 EMEP European emission inventory was used. EMEP (European Monitoring and Evaluation Programme) is the co-operative programme for monitoring and evaluation of the long-range transmission of air pollutants in Europe, scientifically based and policy driven under the Convention on Long-range Transboundary Air Pollution (CLRTAP) for international co-operation to solve transboundary air pollution problems. This inventory is compiled by CEIP (EMEP Centre on Emission Inventories and Projections, http://www.ceip.at/ms/ceip_home1/ceip_home/ceip_intro/) and contains the emission data officially submitted by the Parties to the CLRTAP. From 2017 the reporting of gridded emissions is based on a spatial grid with a horizontal resolution of $0.1^\circ \times 0.1^\circ$ (about 10 km x 10 km) and emissions are classified following the GNFR14 (Nomenclature for Reporting of gridded data and Large point sources) system.

2.2.2 Natural emissions

Natural aerosol and biogenic volatile organic compounds (BVOC) emissions were computed by means of SURFPRO using RAMS meteorological outcomes and CORINE Land Cover (Heymann et al., 1994) data.

More in detail, MEGAN model (Guenther et al., 2006) was integrated into SURPRO in order to produce biogenic VOC emissions.

Sea salt emissions were estimated according to Gong-Monahan (Monahan et al., 1986; Gong et al., 2003) and De Leeuw approaches (De Leeuw et al., 2000) for accumulation and coarse fractions, respectively. In both cases they were parameterized in function of relative humidity and wind speed, following Zhang et al. (2005).

Dust emissions take into account erosion and resuspension processes and they were parameterized according to Zender et al. (2003).

2.3 Pollutants transport, dispersion and chemical reactions

2.3.1 FARM model

FARM is a three-dimensional Eulerian model that includes transport and multiphase chemistry of pollutants in the atmosphere.

The code was derived from STEM (Carmichael et al., 1998) and, by means of the chemical pre-processor KPP (Kinetic Pre-Processor, Damian et al, 2002), it can be configured with different gas-phase chemical schemes, according to user requirements.

Several gas phase chemistry schemes are currently available:

1. an updated version of the chemical mechanism implemented in the EMEP Lagrangian Acid Deposition Model (Hov et al., 1988), which can be used to calculate concentrations and depositions of acidifying compounds, Persistent Organic Pollutants (POPs) and mercury;

2. SAPRC-99 (Carter, 2000), a chemical mechanism for the treatment of gas-phase atmospheric reactions of Volatile Organic Compounds (VOCs) and Nitrogen Oxides (NO_x) and, in particular, photochemical processes leading to the formation of ozone and secondary organic aerosols;
3. an updated version of SAPRC-99 that includes also Polycyclic Aromatic Hydrocarbons (PAHs), Metals and Hg chemistry (Silibello et al., 2012; Adani et al., 2015);
4. a simplified gas-phase chemical mechanism, which was derived from <https://www.shodor.org/master/environmental/air/photochem/smogapplication.html>.

In the framework of AMS-MINNI simulations the updated version of SAPRC-99 (option 3) is commonly used in order to fulfil the indication of EU Directive 2008/50/EC on ambient air quality and cleaner air for Europe (EC, 2008) that explicitly refers also to Metals and PAHs in ambient air.

Concerning aerosol dynamical and chemical processes, two modules are available to be associated with gas-phase mechanism: AERO0 (EMEP, 2003) and AERO3 (Binkowski and Roselle, 2003).

AERO0 accounts only for primary anthropogenic aerosols (subdivided in fine and coarse fractions) and secondary inorganic aerosols (SO₄, NO₃ and NH₄). Neither secondary organic contribution nor aerosol dynamic is accounted for. All aerosol species are anyway subjected to dry and wet removal.

In AERO3 the particles size distribution is represented through a lognormal distribution characterized by three modes: the Aitken mode (median diameter < 0.1 μm), the accumulation mode (0.1 μm < median diameter < 2.5 μm) and the coarse mode (median diameter > 2.5 μm). The aerosol dynamics takes into account nucleation, condensation and coagulation processes. The gas/particle mass transfer is implemented by means of ISORROPIA (Fountoukis et al., 2007) and SORGAM (Schell et al., 2001) for secondary inorganic and organic aerosol, respectively.

An upgraded version of AERO3 module was implemented to be used associated with the updated version of SAPRC-99. In this upgraded version also the absorption process of PAHs into aerosol water is considered, following Aulinger et al. (2007) approach.

To simulate the chemical processes involving Mercury in both gas and aqueous phases a chemical mechanism derived from CAMx (Yarwood et al., 2003) is used. The formation of condensable species from the gas-phase and the adsorption of bivalent Mercury on the particles within clouds is also considered according to Seigneur et al. (1998).

Dry depositions are evaluated by means of the well-known resistance model. The diagnostic module SURFPRO provides the velocities scales for each gas and the two AERO0 species (fine and coarse aerosol fractions). Aerosol settling velocities are dynamically computed within the AERO3 module, since they depend on aerosol bin size.

Wet depositions of gases and aerosols are computed according to EMEP model approach, distinguishing between in-cloud and below-cloud removal.

Main features of FARM model are summarized in Figure 7.

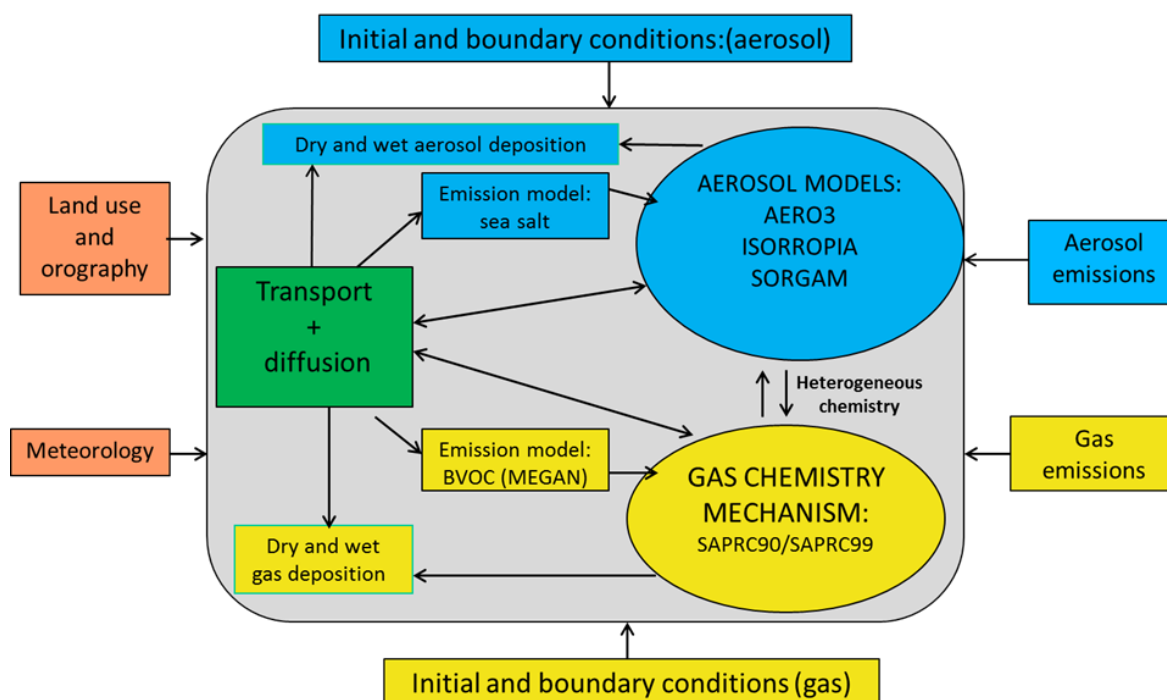


FIGURE 7. MAIN FEATURES OF FARM MODEL.

An updated chemical mechanism, SAPRC07 (Carter, 2010), has been recently made available.

Moreover, the CMAQ wet deposition scheme (Byun and Schere, 2006) taking into account the vertical distribution of precipitation and their physical phases (liquid, graupel, snow, ice) was added in addition to EMEP simplified scheme.

CMAQ in-cloud chemistry, AQCHEM (Schwartz, 1986), was added too, and it can be used in place of the current simplified in-cloud sulphate chemistry, describing S(IV) to S(VI) formation according to Seinfeld and Pandis (1998).

These new developments will be used and validated in the next AMS-MINNI simulations.

2.3.2 Micro-meteorological input parameters for FARM

Micro-meteorological parameters, describing atmospheric turbulence, were computed by means of the diagnostic model SURPRO. For the calculation of horizontal and vertical diffusion coefficients, Smagorinsky (1963) and Lange (1989) parameterizations were used, respectively. To take into account strong horizontal non-homogeneity and temporal non-stationarity of the boundary layer over terrain characterized by abrupt changes of surface (e.g. complex terrain, coastal areas, urban environment, etc.), mixing height was evaluated using prognostic schemes: in near neutral and unstable atmospheric conditions, the Gryning and Batchvarova (1996) approach was used; for stable conditions, the prognostic formulation proposed by Zilitinkevich and Baklanov (2002) was chosen.

2.3.3 FARM simulation set-up

The air quality simulation for 2015 calendar year was carried out running FARM with a horizontal resolution of 4 km and using 16 terrain-following levels irregularly spaced from the ground to 10000 m above surface level (20, 75, 150, 250, 380, 560, 800, 1130, 1570, 2160, 2970, 4050, 5500, 7000, 8500, 10000 m).

Initial and boundary conditions for chemical species were derived from the 2015 simulation of EMEP model at European scale: data provided by both EMEP Meteorological Synthesizing Centre-West (EMEP/MSC-W: http://www.emep.int/mscw/index_mscw.html) and EMEP Meteorological Synthesizing Centre-East (EMEP/MSC-E: <http://www.msceast.org/>) were used for conventional air pollutants and for POPs and heavy metals, respectively.

It worth noting that Saharan dust contributions coming from boundary conditions were not taken into account in this simulation. Indeed, a preliminary test, carried out including Saharan dust contributions in boundary conditions, showed significantly reduced PM10 annual mean bias. Anyway the simulated desert dust events turned out to be both overestimated and shifted in time, compared to the observations, thus leading to poor correlations skills.

Main features of FARM simulation set-up are described in Table 3.

<i>Gas phase Chemistry</i>	<i>updated version of SAPRC-99</i>
<i>Aerosol Module</i>	<i>upgraded version of AERO3</i>
<i>Inorganic Aerosols</i>	<i>ISORROPIA</i>
<i>Organic Aerosols</i>	<i>SORGAM</i>
<i>Dry and Wet Deposition</i>	<i>yes</i>
<i>Boundary Conditions</i>	<i>EMEP/MSC-W & EMEP/MSC-E</i>
<i>In-cloud sulphate chemistry</i>	<i>Simplified S(IV) to S(VI) formation</i>

TABLE 3. MAIN FEATURES OF FARM SIMULATION SET-UP.

3 RESULTS

In this section, outcomes of AMS-MINNI simulation for 2015 are presented in terms of several statistical syntheses. For each pollutant to which the AQD refers, the proper statistical synthesis was calculated in order to compare concentration values with EU critical limits recommended by the AQD.

More in details, in paragraph 3.1 maps of annual mean concentrations are presented for those pollutants for which a yearly limit value is recommended by the Legislative Decree 155/2010, which enforces the AQD in the Italian legislation.

In paragraph 3.2, fields of hourly or daily reference concentration percentiles are shown for those pollutants for which an hourly or daily limit value is recommended by the Legislative Decree 155/2010, together with a maximum number of allowed exceedances. More in detail, if N is the maximum number of allowed exceedances, the percentile corresponding to the highest (N+1)th value is shown and compared with the limit value.

3.1 Annual statistics of concentration fields

Table 4 shows the list of pollutants for which Annex XI of Italian Legislative Decree 155/2010 (implementing Annexes XI and XIV of Directive 2008/50/EC) recommends yearly limit values for the protection of human health.

<i>Pollutant</i>	<i>Averaging Period</i>	<i>Limit Value</i>
NO₂	CALENDAR YEAR	40 µg/m³
C₆H₆	CALENDAR YEAR	5 µg/m³
Pb	CALENDAR YEAR	0.5 µg/m³
PM₁₀	CALENDAR YEAR	40 µg/m³
PM_{2.5}	CALENDAR YEAR	25 µg/m³

TABLE 4. POLLUTANTS WITH YEARLY LIMIT VALUES IN THE FRAMEWORK OF ITALIAN LEGISLATIVE DECREE 155/2010 (IMPLEMENTING AQD 2008/50/EC) .

Actually, in Figure 8, Figure 9 and Figure 10 annual mean concentrations fields are presented only for NO₂ and PM. Indeed the reduction of both Benzene (C₆H₆) and Lead (Pb) concentration levels to values lower than the respective limit ones, was recently observed both in Italy and in the rest of Europe.

In particular, concerning C₆H₆, no exceedances were measured in Italy in 2015 (ISPRA, 2016b). Moreover, according to EEA (European Environment Agency) Report (EEA, 2017), among the 586 stations in 29 European countries (including Italy) reporting C₆H₆ measurements in 2015, only two industrial urban stations, located in Croatia and France, measured concentrations above the annual limit value.

Concerning Lead, no Pb measurement values for 2015 are reported in ISPRA (2016b). On European scale, EEA (2017) collected Pb data from 584 stations distributed in 24 European countries (including Italy). Only one urban background station, located in Belgium, reported Pb concentrations above the $0.5 \mu\text{g}/\text{m}^3$ limit value and about 99 % of the stations reported Pb concentrations below $0.25 \mu\text{g}/\text{m}^3$.

In the following maps, in order to highlight the exceedances, the same type of chromatic scale is proposed for the representation and the classification of both NO_2 and PM concentration values. Hence, every critical area with exceedances of the limit values, set by Legislative Decree 155/2010 and AQD, turns out to be coloured in red.

In addition to NO_2 and PM outcomes, SOMO35 is presented in Figure 11 as a statistical synthesis on annual base of O_3 concentrations. SOMO35 was chosen since this indicator is recommended by WHO for health impact assessment (UNECE, 2004). It is defined as the yearly sum of the daily maximum of 8-hour running average¹ over 35 ppb. For each day, the maximum of the running 8-hours average for O_3 is selected and the values over 35 ppb are summed over the whole year. In this case, chromatic scale and values classification were chosen according to EEA SOMO35 official map for 2015 (<https://www.eea.europa.eu/data-and-maps/figures/o3-indicator-somo35-in>).

¹ The daily maximum of 8-hour running average is selected by examining eight hour running averages, calculated from hourly data and updated each hour. Each eight hour average so calculated will be assigned to the day on which it ends i.e. the first calculation period for any one day will be the period from 17:00 on the previous day to 01:00 on that day; the last calculation period for any one day will be the period from 16:00 to 24:00 on that day.

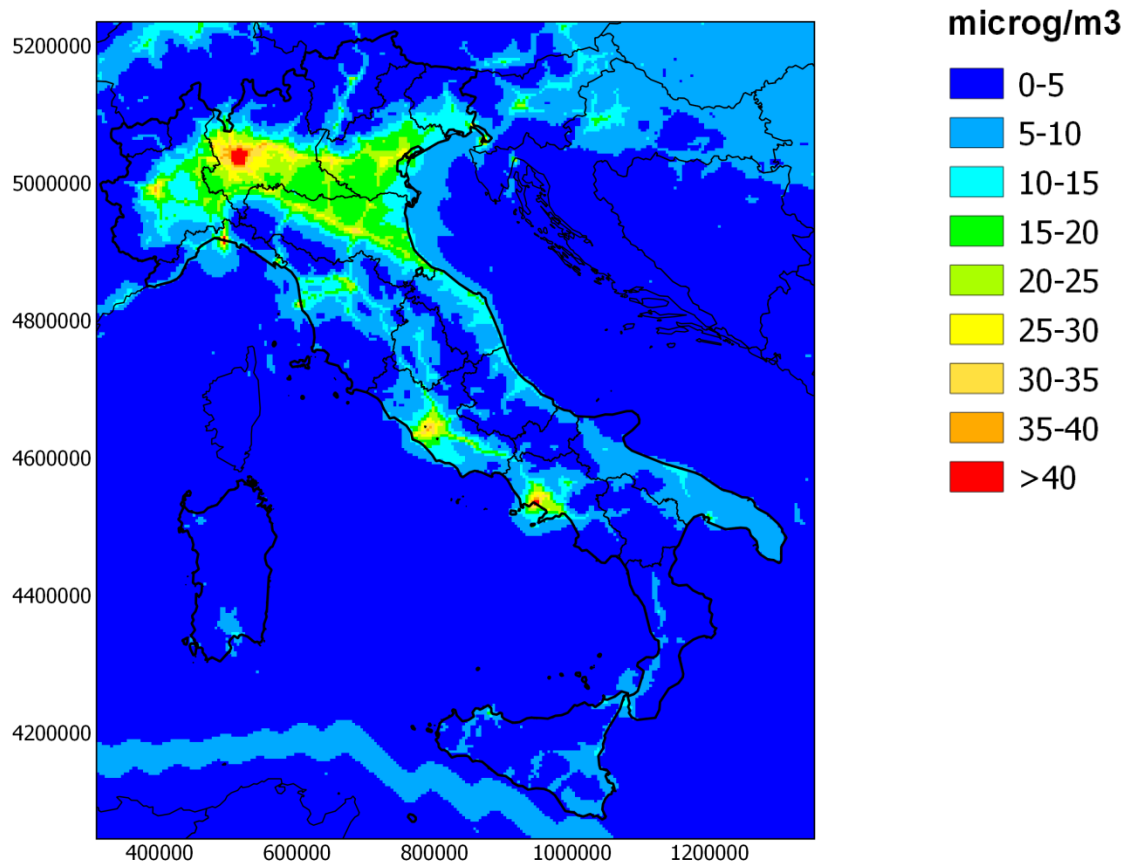


FIGURE 8. ANNUAL MEAN CONCENTRATIONS OF NO₂.

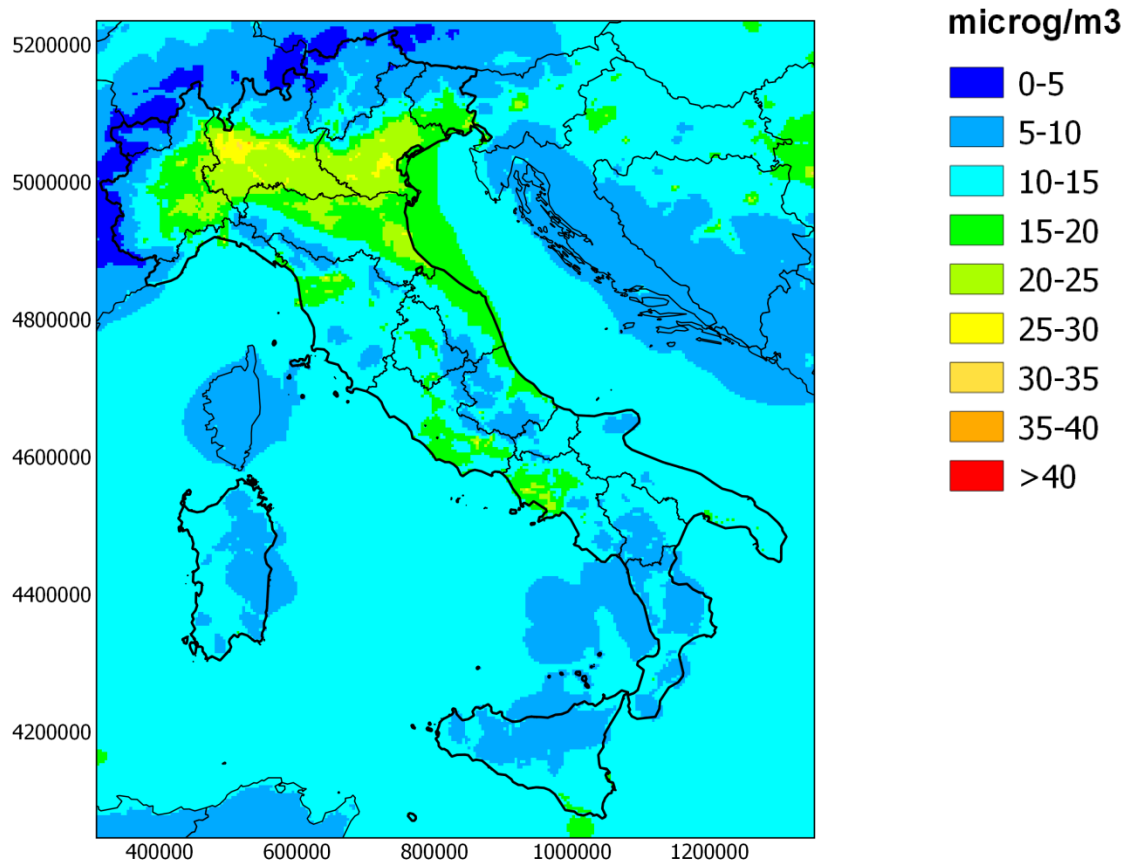


FIGURE 9. ANNUAL MEAN CONCENTRATIONS OF PM₁₀.

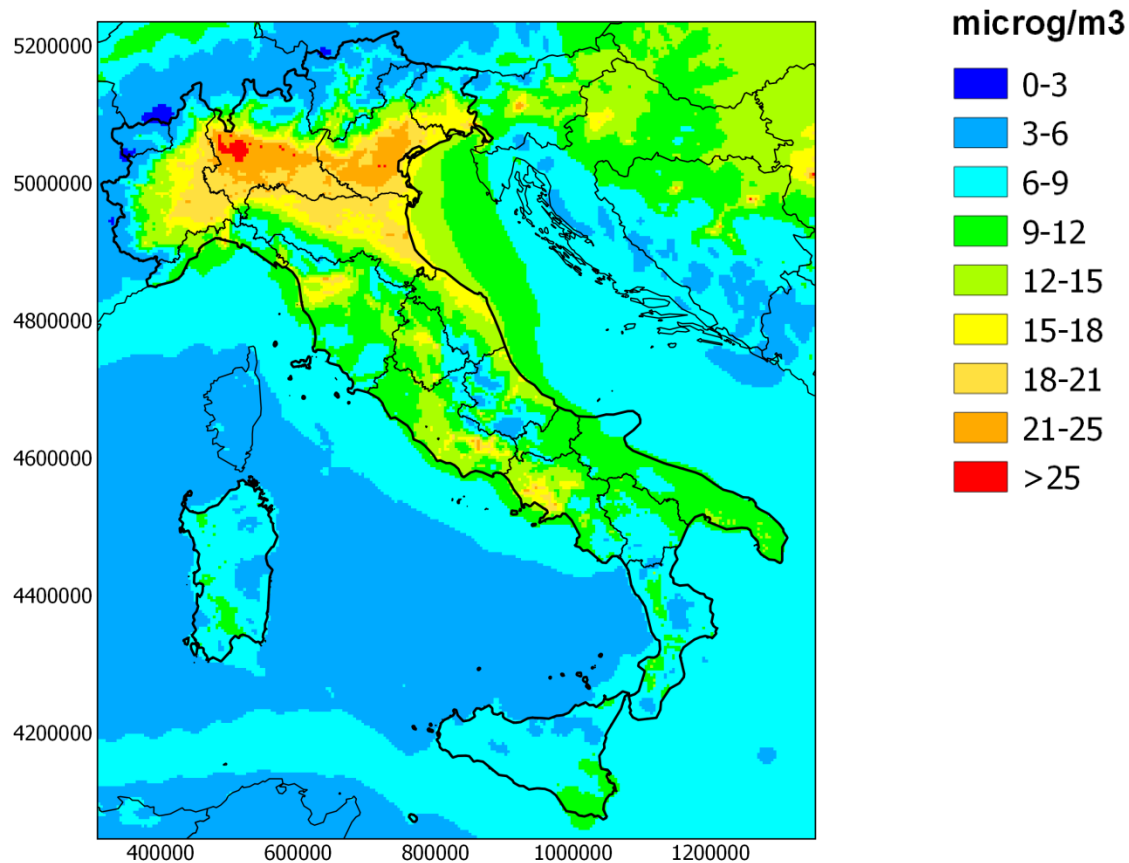


FIGURE 10. ANNUAL MEAN CONCENTRATIONS OF PM2.5.

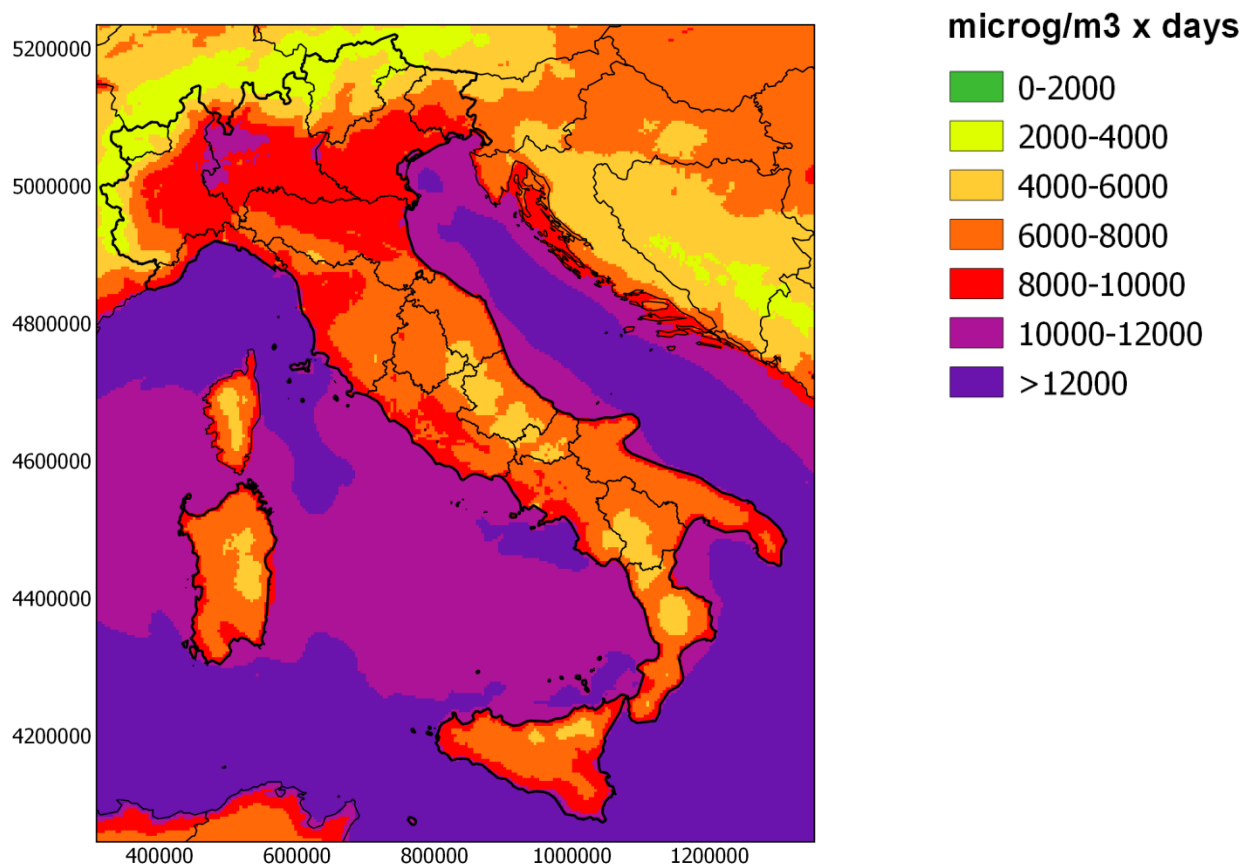


FIGURE 11. O₃ SOMO35.

Annual statistics of concentration fields, presented in Figure 8, Figure 9 and Figure 10, highlight some typical features. In particular, for all the pollutants the highest values are observed in the Po Valley.

Moreover, NO₂ (Figure 8) and PM (Figure 9 and Figure 10) are generally characterized by higher values in the urban areas where some exceedances of the AQD limit values are recorded for NO₂ and PM_{2.5} (Figure 8 and Figure 10, respectively). In particular, in the urban area of Milan, both pollutants exceed their respective annual limit values. NO₂ exceeds the limit value of 40 µg/m³ also in the area of Napoli whereas high PM_{2.5} concentrations are observed also in the provinces of Brescia and Padua. In addition, the pattern of NO₂ concentration field reflects the distribution of the road network and the main shipping lanes. In general, high NO₂ levels occur in areas of high emission density, where O₃ titration process has got a leading role.

Concerning O₃ (Figure 11), the highest values are found over the sea, as already observed and discussed in Ciancarella et al. (2016) in reference to 2010 AMS-MINNI simulation: the absence of NO emissions almost everywhere over the sea and the presence of NO₂, transported here from far high-polluted areas, could explain the main reason of the transformation of NO₂ into O₃. High Ozone levels over the sea are in agreement with EMEP MSC-W model simulation for 2015, which predicts the highest levels of all O₃ metrics over the Mediterranean Sea (Fagerli et al., 2017). Over the land, the highest values are found in the Po Valley (in particular in Lombardia) and in the coastal areas, where the transport of O₃ from the sea is probably caused by sea breeze circulation (Monteiro et al., 2015).

3.2 Fields of hourly and daily reference concentration percentiles

Table 5 shows the list of pollutants for which an hourly or daily limit value is recommended by the Annex XI of Italian Legislative Decree 155/2010 (implementing Annex XI of Directive 2008/50/EC), together with a maximum number of allowed exceedances. O₃ target value for the protection of human health was added in Table 5, according to Annex VII requirements.

<i>Pollutant</i>	<i>Averaging Period</i>	<i>Limit Value</i>
SO₂	ONE HOUR	350 µg/m³ , not to be exceeded more than 24 times a calendar year
	ONE DAY	125 µg/m³ , not to be exceeded more than 3 times a calendar year
NO₂	ONE HOUR	200 µg/m³ , not to be exceeded more than 18 times a calendar year
CO	MAXIMUM DAILY OF 8-HOUR RUNNING MEAN	10 mg/m³
PM10	ONE DAY	50 µg/m³ , not to be exceeded more than 35 times a calendar year
O₃	MAXIMUM DAILY OF 8-HOUR RUNNING MEAN	120 µg/m³ not to be exceeded on more than 25 days per calendar year averaged over three years

TABLE 5. POLLUTANTS WITH HOURLY OR DAILY LIMIT (OR TARGET) VALUES IN THE FRAMEWORK OF ITALIAN LEGISLATIVE DECREE 155/2010 (IMPLEMENTING AQD 2008/50/EC) .

In Figure 12, Figure 13, Figure 14, Figure 15 and Figure 16, for all the pollutants listed in Table 5, the highest (N+1)th values of the hourly or daily time series are shown and compared with the limit value, being N the maximum number of allowed exceedances.

Only Carbon monoxide (CO) outcomes are not shown since CO air concentration values are now very low and exposure of the European population to CO concentrations above the AQD limit value is quite localised and sporadic (Guerreiro et al., 2014). Moreover, concerning 2015, no CO measurement values are reported in ISPRA (2016b). On European scale, EEA (2017) reported that in 2015 the highest CO levels are found in urban areas, typically during rush hour, or downwind from large industrial emission sources. Of the 776 operational stations with more than 75 % of valid data in 36 EEA member countries (including also Italy), only four stations, one suburban background station in Albania and three urban background stations in Germany, Montenegro and Serbia, registered concentrations above the CO limit value.

As for annual statistics (paragraph 3.1), in the following maps, the areas of non-compliance with the requirements set by Legislative Decree 155/2010 and AQD are highlighted in red.

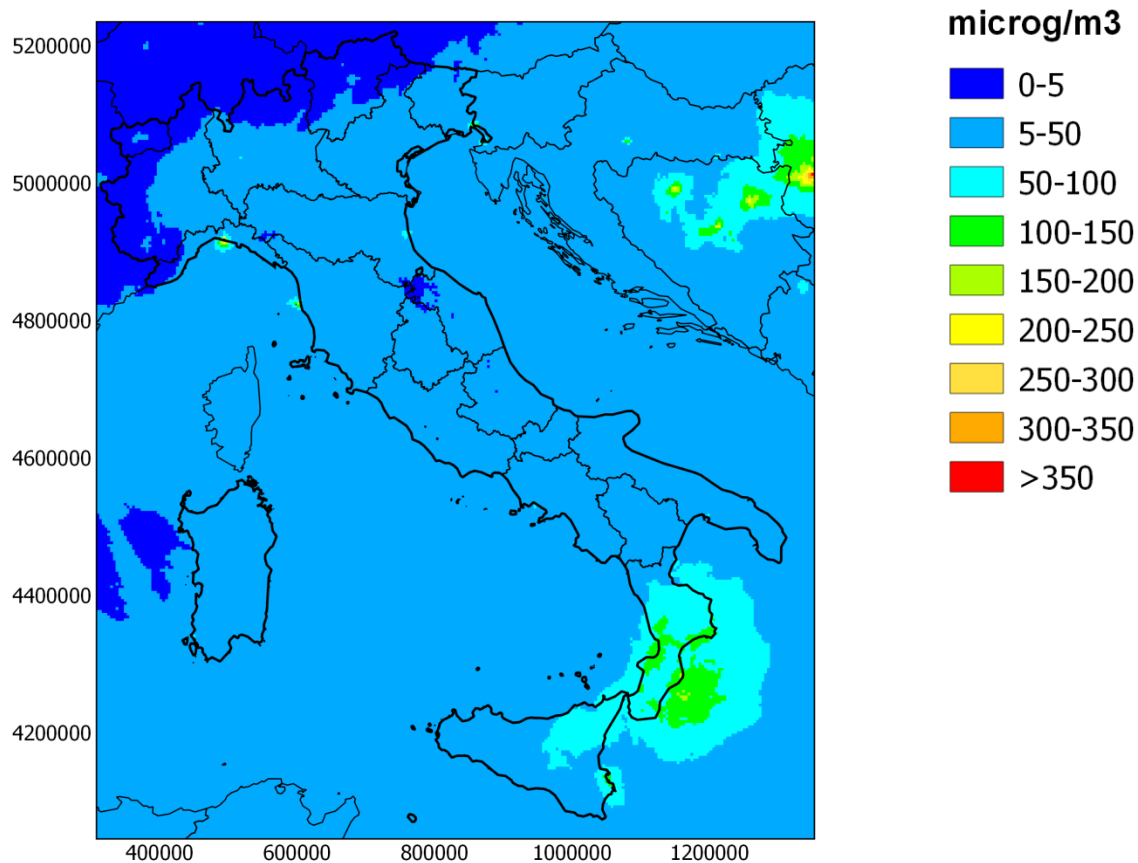


FIGURE 12. 25th HIGHEST VALUE OF THE HOURLY TIME SERIES OF SO₂.

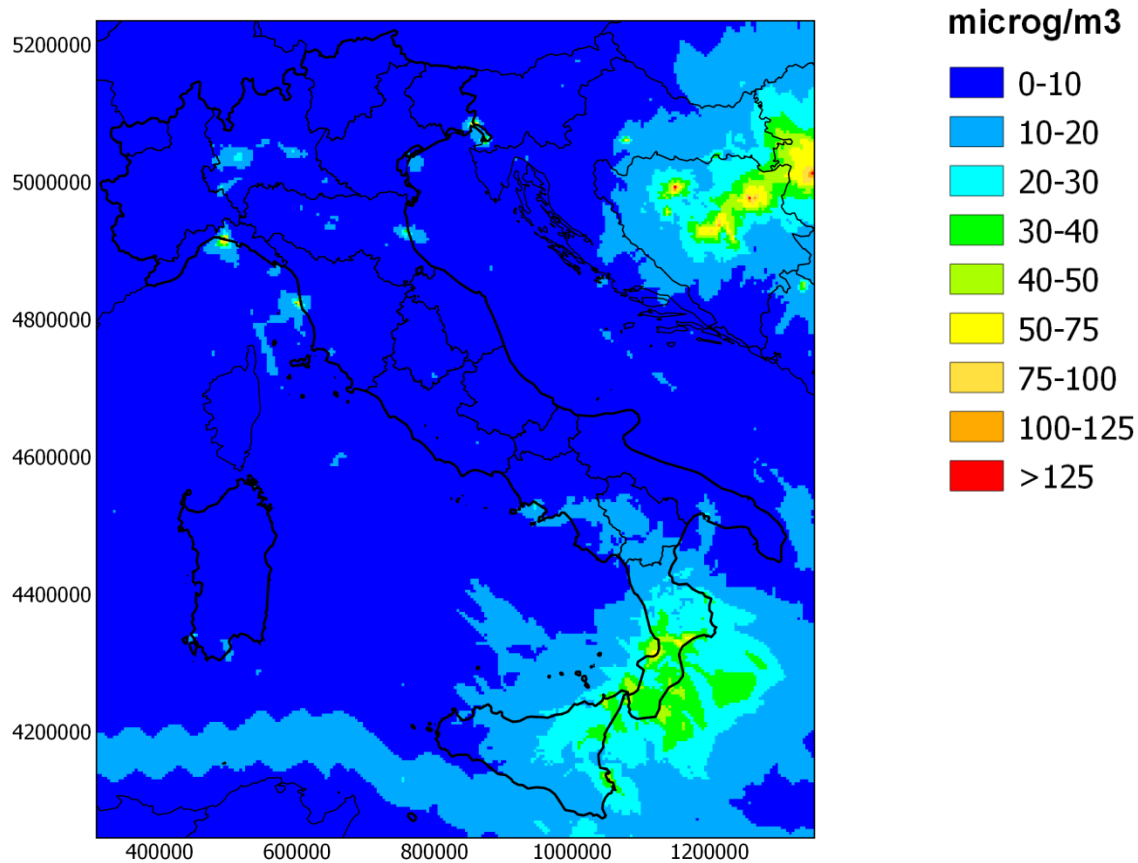


FIGURE 13. 4th HIGHEST VALUE OF THE DAILY TIME SERIES OF SO₂.

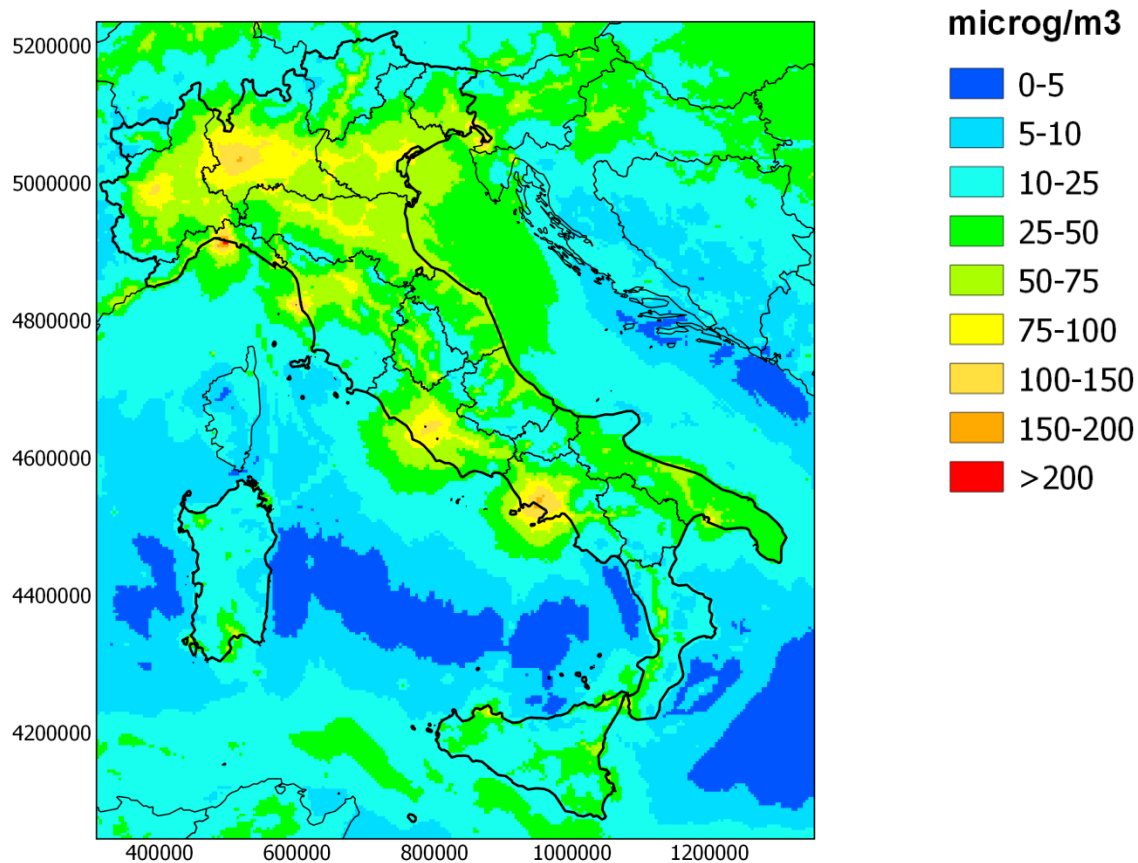


FIGURE 14. 19th HIGHEST VALUE OF THE HOURLY TIME SERIES OF NO_2 .

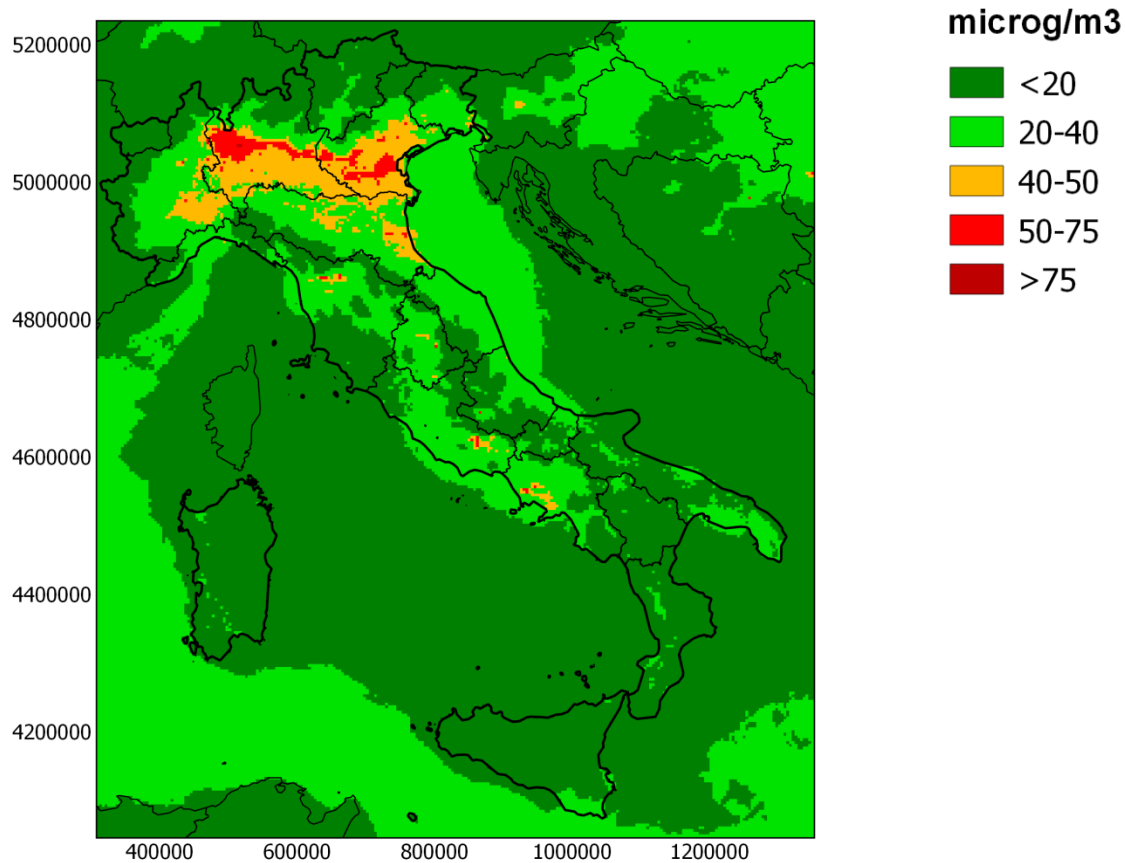


FIGURE 15. 36th HIGHEST VALUE OF THE DAILY TIME SERIES OF PM_{10} .

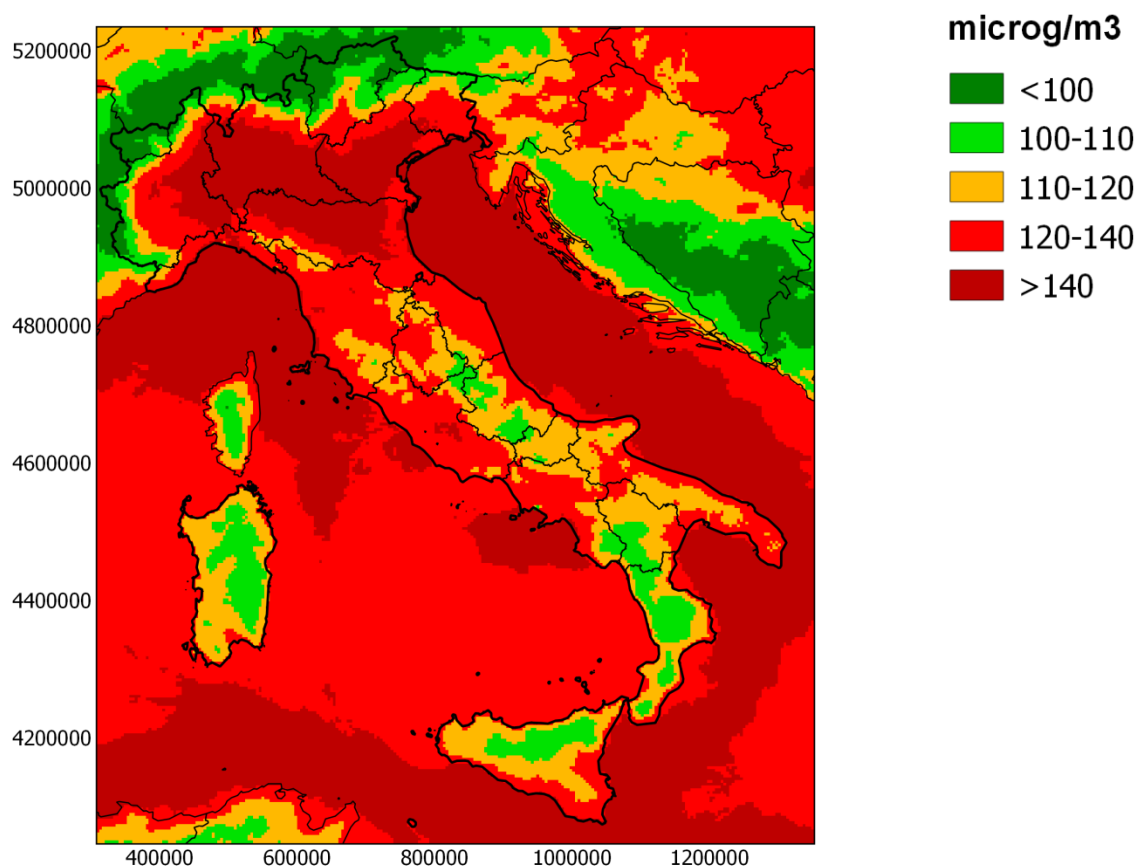


FIGURE 16. 26th HIGHEST VALUE OF THE TIME SERIES OF O₃ MAXIMUM DAILY OF 8-HOUR RUNNING MEAN VALUES.

Fields of hourly and daily reference concentration percentiles, in the above Figures, show that low SO₂ concentration values are generally observed all over the Italian territory (Figure 12 and Figure 13). The highest values of NO₂ (Figure 14) are observed both in urban areas and in correspondence of road networks. In particular, only in the urban area of Genova, the modelled exceedances of the hourly limit value of 200 µg/m³ are recorded for more than 18 times, since the 19th highest value is higher than 200 µg/m³, probably due to the combined effect of road traffic and port activities in a rather small area.

The most critical issues arise from PM₁₀ and O₃ outcomes. Concerning PM₁₀ (Figure 15), more than 35 exceedances of the daily limit value of 50 µg/m³ are simulated in wide areas of Lombardia and Veneto and in several other Regions: Piemonte, Liguria, Emilia Romagna, Friuli Venezia Giulia, Toscana, Umbria, Lazio, Abruzzo, Campania. Globally, most of the Regions involved in monitored PM₁₀ daily exceedances for the year 2015 (ISPRA, 2016b) are adequately captured by AMS-MINNI simulation; the only exceptions are Marche and Sicilia Regions; for the latter case, it is worth noting that in the present AMS-MINNI simulation the Saharan dust contribution (likely to play a significant role in exceedances in Centre-Southern regions) was not taken into account.

Concerning O₃ (Figure 16), almost the whole Italian territory is characterized by the non-compliance with the requirements set by Legislative Decree 155/2010 and AQD. This outcome is in agreement with observed data: ISPRA (2016b) pointed out that, concerning O₃ 2015 data, most of the monitoring stations turned out to be not in compliance with the target requirements. This condition of extreme photochemical pollution finds

its explanation in the particular 2015 meteorological conditions. The calendar year 2015 was among the warmest historically year recorded in Europe (Fagerli et al., 2017); in particular, in Italy, it turned out to be the third driest year since 1961 and the one with the highest average temperature (ISPRA, 2016a).

4 EVALUATION OF MODEL PERFORMANCES AGAINST OBSERVED DATA

Models applied for regulatory air quality assessment are commonly evaluated on the basis of comparisons against observations data. A statistical analysis is carried out and multiple performance indicators (bias, correlation coefficient, standard deviation differences, fraction of modelled data within a given factor of the observations, etc.) are calculated in order to describe model skills for a given simulation from different point of views.

Statistical indicators provide a quantitative insight on model performances but they do not tell whether or not model results are good enough for a given application, e.g. for policy support. This is the reason why it can be very useful to define Model Performance Criteria (MPC), i.e. the minimum level of quality to be achieved by a model. Over recent years, this issue has been extensively addressed by the FAIRMODE (Forum for Air Quality Modelling in Europe, <http://fairmode.jrc.ec.europa.eu/>) community, a European network of air quality model developers and users, whose aim is the exchange of experiences, competence and good practices on model applications, especially under the implementation of AQD by EU Member States.

In the framework of FAIRMODE activities, a procedure for air quality models benchmarking was suggested along with the recommendation of Modelling Performance Criteria (Thunis et al., 2012a, 2013; Pernigotti et al., 2013). The goal is the harmonisation of the diagnostics and reporting of air quality model application performances, focusing on the pollutants mentioned in the most recent EU AQD (EC, 2008).

FAIRMODE evaluation methodology and criteria, consolidated in the DELTA Tool software (Thunis et al., 2012b), have been widely used and tested during the last years (Monteiro et al., 2018). Hereinafter its application to validate MINNI simulation outputs is shown. The purpose is providing an objective assessment of the quality of MINNI results, with respect to criteria currently adopted and applied by the European scientific community.

4.1 Methods for the evaluation: the DELTA tool approach

In the framework of the DELTA Tool approach, in order to cover all the aspects of the model performances in terms of amplitude, phase and bias, the following core set of statistical indicators is proposed: the Root Mean Square Error (*RMSE*), the *BIAS*, the correlation coefficient (*CORR* or *R*) and the Normalized Mean Standard Deviation (*NMSD*), defined as follows:

$$RMSE = \sqrt{\frac{1}{N} \sum_{i=1}^N (M_i - O_i)^2} \quad (1)$$

$$BIAS = \frac{1}{N} \sum_{i=1}^N (M_i - O_i) = \bar{M} - \bar{O} \quad (2)$$

$$CORR = R = \frac{\frac{1}{N} \sum_{i=1}^N (O_i - \bar{O})(M_i - \bar{M})}{\sigma_O \sigma_M} \quad (3)$$

$$NMSD = \frac{\sigma_M - \sigma_O}{\sigma_O} \quad (4)$$

where the letters O and M stand for observations and model results, respectively, the subscript i indicates the time step, the overbars indicate the time average over N time intervals, while the symbol σ indicates the standard deviation.

4.1.1 MQI for hourly and daily time series

Within the DELTA Tool, a Modelling Quality Indicator (MQI) is defined taking into account both the comparison between modelled and measured data and the measurement uncertainty. More in detail, the MQI_{HD} for the comparison between modelled and measured time series and its related Modelling Quality Objective (MQO_{HD}) are defined as follows:

$$MQI_{HD} = \frac{RMSE}{\beta RMS_U} \quad MQO_{HD}: MQI_{HD} \leq 1 \quad (5)$$

According to this formulation, the $RMSE$ between observed and modelled values is compared to a value representative of the maximum allowed measurement uncertainty (RMS_U). The value of β determines the stringency of the MQO_{HD} : in the current DELTA formulation, β is set to 2, thus allowing the deviation between modelled and measured concentrations to be twice the measurement uncertainty. According to Thunis et al. (2013), the root mean square of the measurement uncertainty, RMS_U , can be expressed as:

$$RMS_U = U_{95,r}^{RV} \sqrt{(1 - \alpha^2) (\bar{O}^2 + \sigma_O^2) + \alpha^2 RV^2} \quad (6)$$

where $\bar{O}$ e σ_O are the mean and the standard deviation of the measured time series, respectively; $U_{95,r}^{RV}$ is the standard measurement uncertainty around the reference value (RV) for a reference time interval (e.g. the daily/hourly limit value, as defined in EU AQD); α is the non-proportional fraction (between 0 and 1) of the measurement uncertainty around that reference value. All the parameters (β , $U_{95,r}^{RV}$, RV , α) involved in equations (5) and (6) are pollutant-dependent and they were set according to Thunis et al. (2013) e Pernigotti et al. (2013); please refer to these publications for further details.

Further insight into modelling performance is provided by defining supplementary Modelling Performance Indicators (MPI) and their related Model Performance Criteria (MPC). More in details, $MPIs$ and $MPCs$ for $BIAS$, R and $NMSD$, are derived by applying the condition $MQI_{HD} \leq 1$ and substituting in equation (5) the mathematical relationship (Murphy, 1988) linking statistical indicators among themselves²:

² In equation (7) $CRMSE$ stands for Centered Root Mean Square Error, defined as follows:

$$CRMSE = \sqrt{\frac{1}{N} \sum_{i=1}^N ((M_i - \bar{M}) - (O_i - \bar{O}))^2} \quad (7.1)$$

$$RMSE^2 = BIAS^2 + CRMSE^2 = BIAS^2 + (\sigma_M - \sigma_O)^2 + 2\sigma_M\sigma_O(1 - R) \quad (7)$$

Substituting equation (7) in equation (5) and considering ideal cases where two out of three indicators perform perfectly, separate *MPCs* can be derived for each of these three statistics:

$$|BIAS| \leq \beta RMS_U \quad (8)$$

$$|NMSD| \leq \beta \frac{RMS_U}{\sigma_O} \quad (9)$$

$$R \geq 1 - \beta^2 \frac{RMS_U^2}{2\sigma_O\sigma_M} \quad (10)$$

4.1.2 MQI for yearly averages

Concerning yearly averaged concentrations assessment, the *MQO* is modified into a criterion in which the mean bias between modelled and measured concentrations is normalized by the expanded uncertainty of the mean observed concentration:

$$MQI_YR = \frac{|\bar{M} - \bar{O}|}{\beta U_{95}(\bar{O})} \quad MQO_YR: MQI_YR \leq 1 \quad (11)$$

See Pernigotti et al. (2013) for details, and in particular for the expression of the uncertainty of the yearly averaged observations.

4.1.3 MPC for high percentiles

In order to describe model capability to reproduce extreme events (e.g. exceedances), a specific *MPI* indicator was proposed within the DELTA Tool. It is defined as follows:

$$MPI_{perc} = \frac{|M_{perc} - O_{perc}|}{\beta U_{95}(O_{perc})} \quad MPC_{perc}: MPI_{perc} \leq 1 \quad (12)$$

where *perc* is a selected percentile and M_{perc} and O_{perc} are the corresponding modelled and observed values. The denominator is given as a function of the measurement uncertainty characterizing the O_{perc} value. The default percentile value is set to 95th excepted for hourly NO₂, 8h daily maximum O₃ and daily PM10 and PM2.5. In these cases *perc* is set according to the maximum number of exceedances of the limit values allowed by the 2008/50/EC AQD for these pollutants. More in details:

- for hourly NO₂: *perc*=99.8th, i.e. the highest 19th value among 8760 hours, being 18 the maximum allowed number of exceedances of the hourly limit value of 200 µg/m³;
- for 8h daily maximum O₃: *perc*=92.9th, i.e. the highest 26th value among 365 days, being 25 the maximum allowed number of exceedances of the 8h daily maximum limit value of 120 µg/m³;
- for daily PM10 and PM2.5: *perc*=90.1th, i.e. the highest 36th value among 365 days, being 35 the maximum allowed number of exceedances of the daily limit value of 50 µg/m³ (only as far as PM10 is concerned).

4.1.4 MPC for spatial correlation and standard deviation

In the framework of DELTA Tool approach, spatial statistics are calculated too. The model results are first yearly averaged at each station. Correlation and standard deviation indicators are then calculated for this set of averaged values; finally, *MPIs*, together with their related *MPCs*, are consistently defined. See Delta User's guide (https://aqm.jrc.ec.europa.eu/public/data/DELTA_UserGuide.pdf) for details.

4.2 Results of the evaluation

The benchmarking of MINNI results for the national 2015 simulation was carried out according to DELTA Tool approach and requirements, in particular with regards to FAIRMODE recommendation for the assessment of a model application in the framework of the AQD. More in detail, hourly NO₂, 8h daily maximum O₃ and daily PM₁₀ and PM_{2.5} were taken into account for the validation, being the DELTA Tool evaluation in benchmarking assessment mode currently limited to these pollutants.

Data collected by EEA, available on the Eionet website (European Environment Information and Observation Network, <http://cdr.eionet.europa.eu/en/eu/aqd/e1a/>), were used. Given the AMS-MINNI spatial resolution used in this simulation, only background stations were considered to have an adequate spatial representativeness for the comparison with modelled data. In addition to that, according to the AQD (EC, 2008), a minimum data availability (75%) was required for considering a monitoring station suitable for validation. Figure 17 shows the positions of all the considered monitoring sites for each pollutant. The colour of dots, blue, orange and red, identify the zone of the station: rural, suburban and urban respectively.

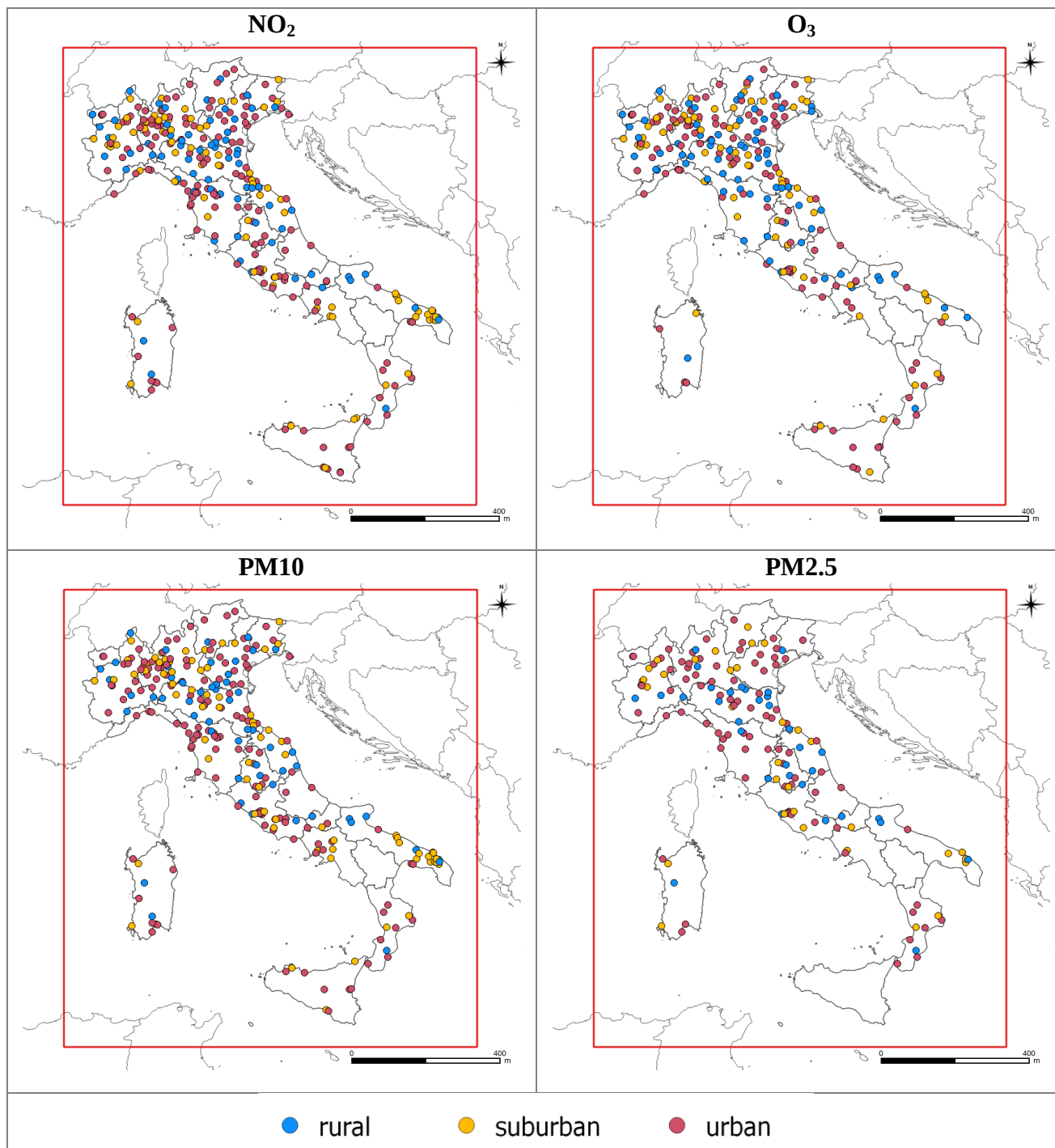


FIGURE 17. BACKGROUND MONITORING STATIONS USED FOR THE VALIDATION OF NO_2 (TOP LEFT), O_3 (TOP RIGHT), PM_{10} (BOTTOM LEFT) AND $\text{PM}_{2.5}$ (BOTTOM RIGHT) MODELLED CONCENTRATION. BLUE, ORANGE AND RED DOTS IDENTIFY RURAL, SUBURBAN AND URBAN STATIONS, RESPECTIVELY.

The density of the stations is higher in the North than in the South of the country. However, monitoring sites appear well distributed throughout all the national territory. The only exceptions, at the moment of this study, are the Basilicata region, which turns out to be without any observation point for all the four pollutants taken into account, and Sicily where no $\text{PM}_{2.5}$ measurement is available.

For each of the four pollutants, *MQO* and *MPCs* fulfilment was checked according to the criteria formulations, described in the paragraph 4.1. Results are presented in the Figures shown below.

In particular, Figure 18, Figure 21, Figure 24 and Figure 27 show MQI_HDs (computed according to equation (5)) for hourly NO_2 , 8h daily maximum O_3 , daily PM_{10} and daily $PM_{2.5}$, respectively. Results are presented for each of the monitoring stations, both in the Target Plots (top of the panels) and in their spatial distribution (bottom of the panels).

In the Target Plots, MQI_HD (i.e. $RMSE/\beta RMS_U$) represents the distance between the origin and a given point. For each point (representing one station), the ordinate is $BIAS/\beta RMS_U$, the abscissa is $CRMSE/\beta RMS_U$. The green area identifies the fulfilment of the criterion of equation (5). Top and bottom quadrants are used to identify positive and negative $BIAS$, respectively. Because $CRMSE$ is always positive by definition, only the right part of the diagram would be needed in the Target plot. Anyhow, the negative X axis section is used to provide additional information, i.e. to distinguish if $CRMSE$ is dominated by SD (standard deviation differences, second term in the right side of equation (7)) or by R (third term in the right side of equation (7)). Conventionally, right and left sides of the diagram are used if $CRMSE$ is dominated by SD and R, respectively.

The MQI_HD (equation (5)) and MQI_YR (equation (11)) associated to the 90th percentile worst station (hereafter MQI_HD_{90th} and MQI_YR_{90th}) are calculated and indicated in the upper left corner. These outcomes are used as the main indicators in the benchmarking procedure, for time series and annual averaged evaluation, respectively; they should be less or equal to 1 for the fulfilment of the benchmarking requirements.

The uncertainty parameters (β , $U_{95,R}^{RV}$, RV , α), involved in equations (5) and (6) and used to produce the diagram, are listed on the top right-hand side of the diagram. In blue colour, the resulting model uncertainty is provided (see Delta User's guide, https://aqm.jrc.ec.europa.eu/public/data/DELTA_UserGuide.pdf, for details).

In the bottom of the panels of Figure 18, Figure 21, Figure 24 and Figure 27 locations of all the monitoring points are shown. The colour of dots indicates whether or not MQO of equation (5) is satisfied for that monitoring station; and, if not, which part of the $RMSE$ is dominating ($BIAS > 0$, $BIAS < 0$, poor correlation or $NMSD$).

The fulfilment of Modelling Performance Criteria (MPC) for each of the three statistics ($BIAS$, R , $NMSD$) is shown in Figure 19, Figure 22, Figure 25 and Figure 28 for each pollutant, respectively, taken into account in DELTA Tool benchmarking assessment mode. In every Figure, a panel with three plots is provided: the top left plot is the scatter plot of modelled annual averaged concentrations versus observed ones; $NMSD$ and R values are shown in the top right and in the bottom left plots, respectively, as function of $\beta RMS_U/\sigma_O$. In each of these three plots, coloured areas indicate the fulfilment of the MPC for $BIAS$, $NMSD$ and R , respectively. More in detail, the whole coloured area between the two solid lines (no matter the colour, orange or green) is the area where the MPC criterion is satisfied, according to equations (8), (9) and (10); within this area, two inner areas are highlighted (the green one and, inside it, the one delimited by dashed lines), indicating the fulfilment of more stringent criteria, obtained by substituting in equations (8), (9) and (10) the value of β with $\sqrt{\beta^2/2}$ and 1, respectively. In particular, the last case, where β is replaced by 1,

indicates the area where the error between the modelled and the observed values lies within the measurement uncertainty range.

A summary of model performances can be found in Figure 20, Figure 23, Figure 26 and Figure 29 where benchmarking Summary Reports for each of the four pollutants are presented.

Here, in the section OBS, the main observed statistics are provided for each monitoring stations, i.e. the annual averaged values and the number of the exceedances of the hourly/daily limit value (reported in the box).

In the section TIME, MPI outcomes for $BIAS$, $NMSD$ and R , together with MPI_{perc} (according to equation (12)), are shown. Each point represents a monitoring station and green and orange areas represent the fulfilment zone, as described above.

The section SPACE provides an overview of spatial statistics for correlation and standard deviation, as described in paragraph 4.1.4.

For all the indicators, the third column provides information on the overall fulfilment of the performance criteria: green means the criterion is fulfilled, red means that it is not. In particular, in the section TIME, the overall criterion is satisfied if more than 90% of the stations fulfil it.

4.2.1 Nitrogen dioxide (NO_2)

Outcomes of hourly NO_2 validation are presented in Figure 18, Figure 19 and Figure 20.

Figure 18 (top of the panel) shows that MQO_{HD} is fulfilled for more than 90% of the stations, since $MQI_{HD_{90th}}$ value is lower than one. This is not true with regard to MQO_{YR} evaluation, even if $MQI_{YR_{90th}}$ value is not so far from one. Moreover, the Target Plot shows that negative $BIAS$ are more frequent than positive ones, indicating a general underestimation of NO_2 concentration. Concerning $CRMSE$, it turns out to be dominated by poor R more than by SD differences. Globally, from the bottom of the panel of Figure 18 it can be inferred that negative $BIAS$ are generally the dominant fraction of $RMSE$ values. In the same plot, spatial distribution of MQI_{HD} shows that the area between Lazio and Campania regions seems to be the most critical one for the MQO_{HD} fulfilment; other points where MQO_{HD} is not fulfilled are mainly located in the Northern Italy, in complex orographic contexts in alpine and pre-alpine areas, where the 4 km spatial resolution of the simulation is not sufficient to describe the phenomena.

Figure 19 highlights good model performances with respect to all the statistical indicators. MPC is fulfilled for all the stations concerning $NMSD$ and for most of them concerning both $BIAS$ and R ; more in detail, in all cases $MPCs$ are fulfilled for more than 90% of the stations, since the overall criterion turns out to be satisfied for all the fraction of the error, i.e. $BIAS$, R , $NMSD$ (green dots in Figure 20).

From Figure 20 it can be noticed that also spatial criteria are satisfied for both correlation and standard deviation. Overall only MPC_{perc} is not fulfilled indicating poorer performances with regard to model capability in predicting high hourly concentrations.

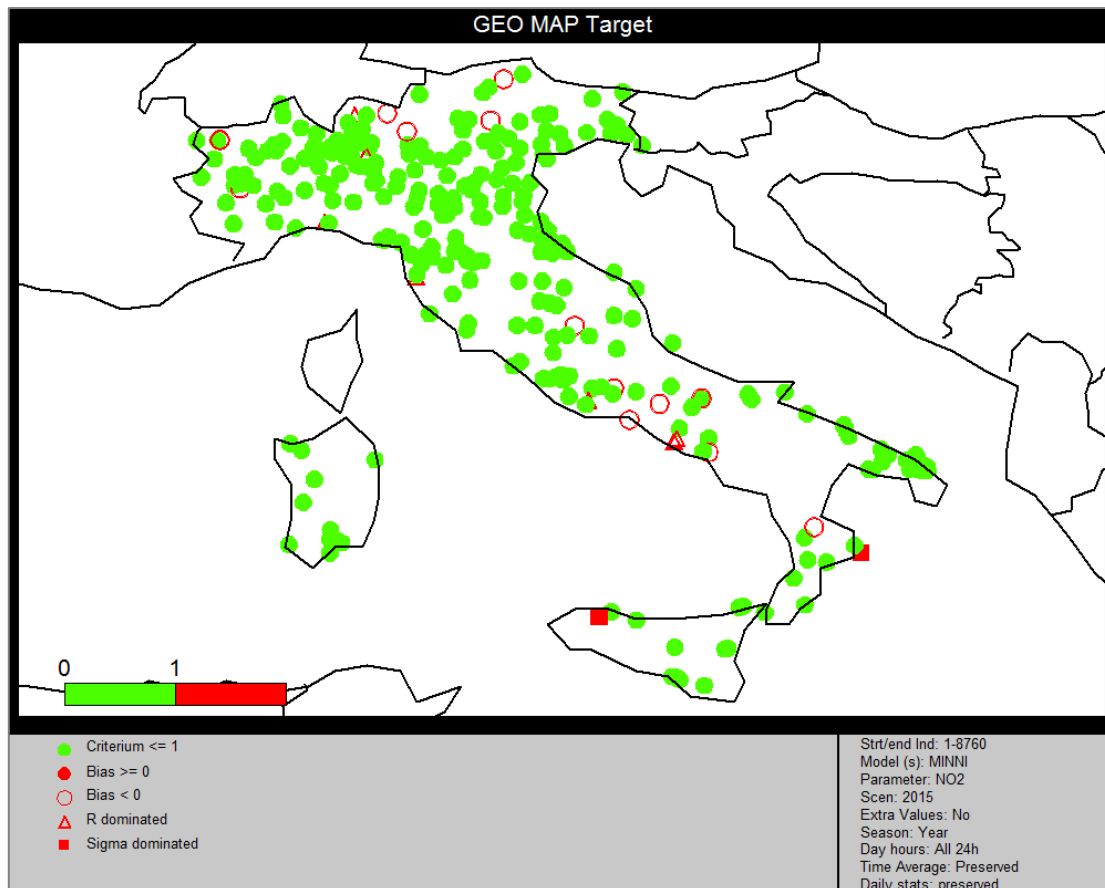
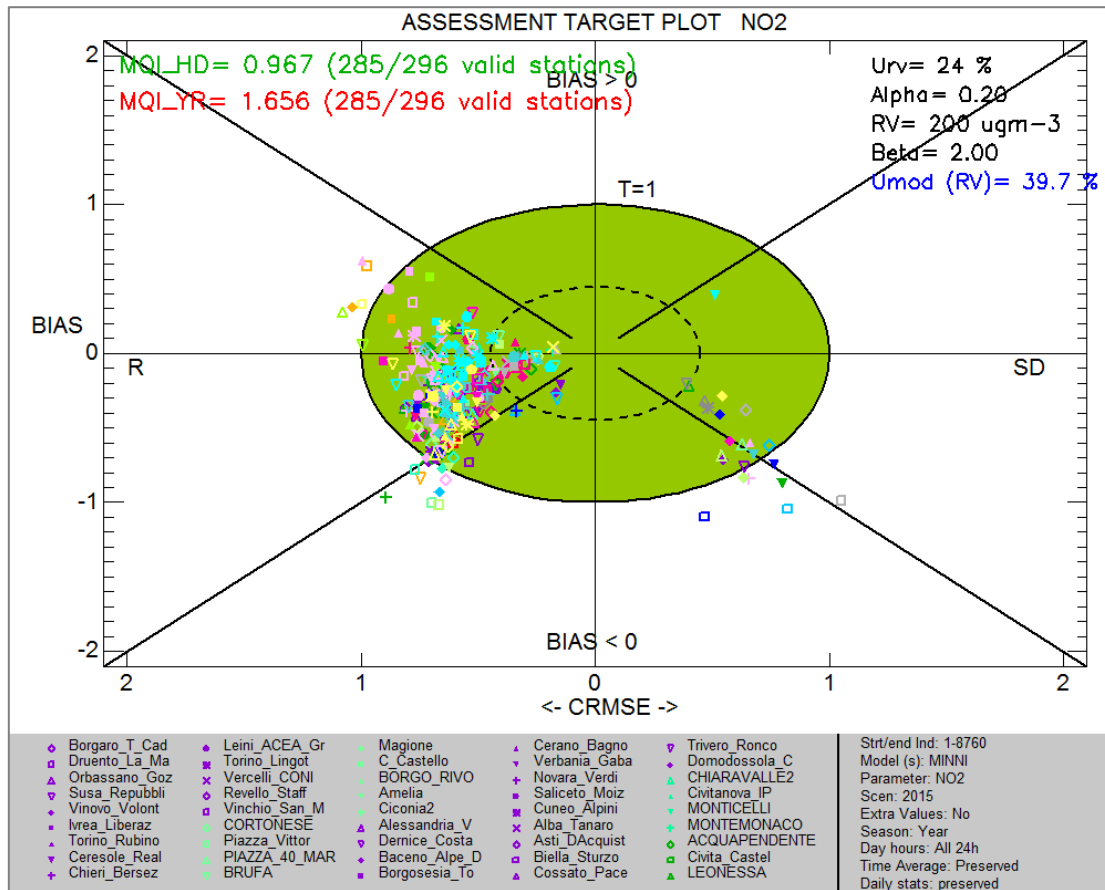


FIGURE 18. MQO FOR HOURLY NO₂ CONCENTRATIONS: ASSESSMENT TARGET PLOT (TOP) AND SPATIAL DISTRIBUTIONS OF CRITERION FULFILMENT OUTCOMES (BOTTOM).

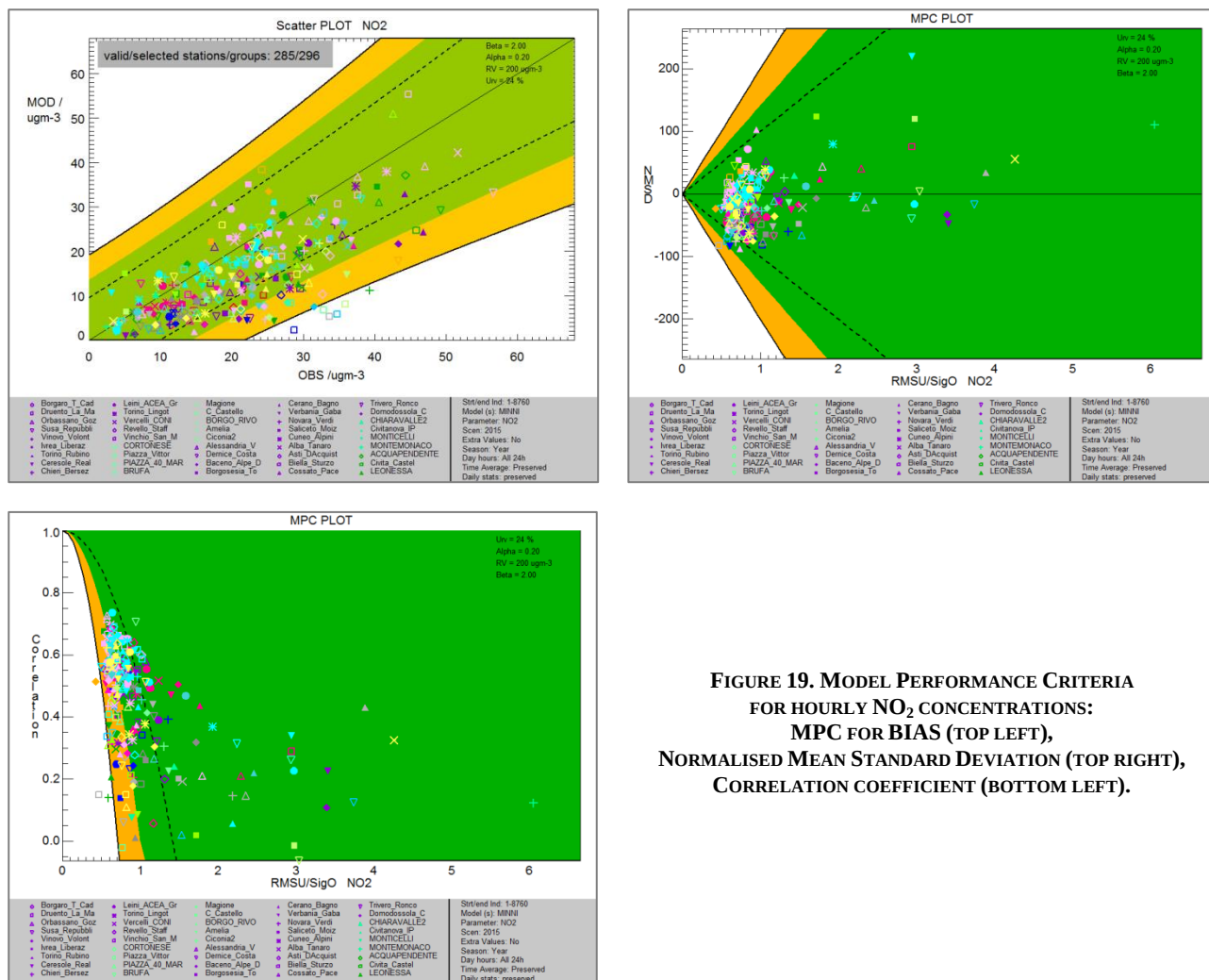


FIGURE 19. MODEL PERFORMANCE CRITERIA FOR HOURLY NO₂ CONCENTRATIONS: MPC FOR BIAS (TOP LEFT), NORMALISED MEAN STANDARD DEVIATION (TOP RIGHT), CORRELATION COEFFICIENT (BOTTOM LEFT).

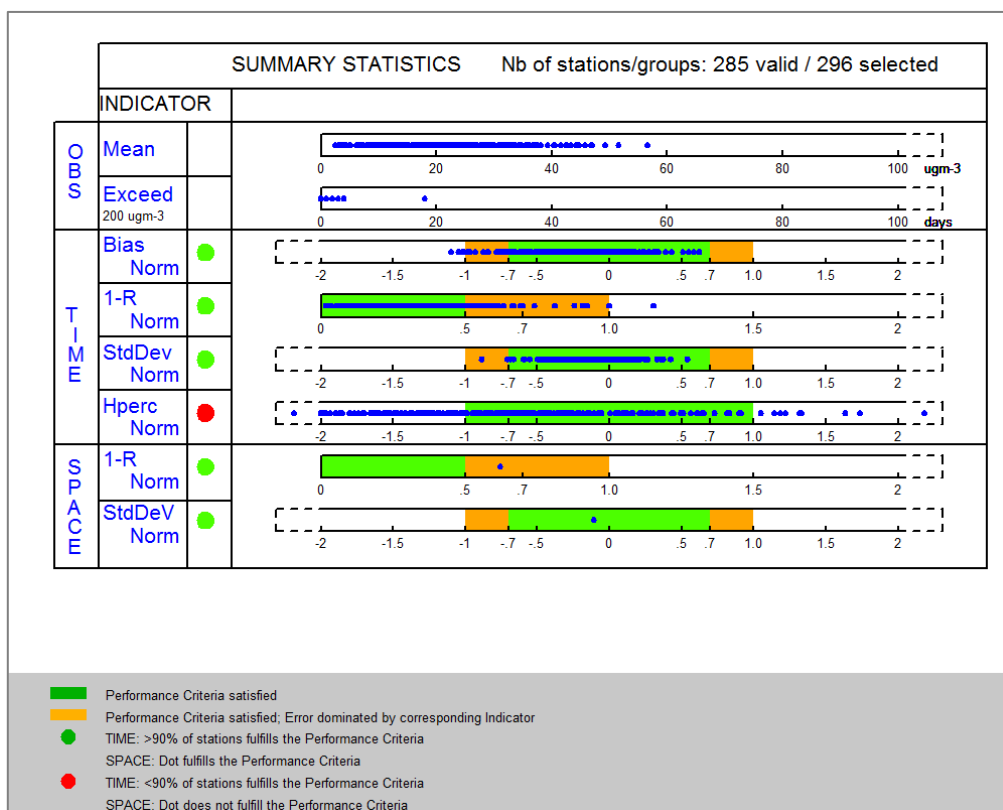


FIGURE 20. BENCHMARKING PERFORMANCE SUMMARY REPORT FOR HOURLY NO₂ CONCENTRATIONS.

4.2.2 Ozone (O_3)

AMS-MINNI shows excellent performances in simulating 8h daily maximum O_3 , as it can be inferred from all $MQOs$ and $MPCs$ outcomes in Figure 21, Figure 22 and Figure 23.

In particular, Figure 21 highlights the fulfilment of the overall benchmarking requirements, since both $MQI_{HD_{90th}}$ and $MQI_{YR_{90th}}$ are lower than one. More in detail MQO_{HD} is fulfilled for all the 234 valid stations, except for only one, an urban background station named “Corso Firenze Genova”, located in the urban area of Genova city. Here MQO_{HD} is not satisfied, with error dominated by negative $BIAS$. Since another urban background station, named “Quarto Genova”, located not so far from this monitoring point (less than 6 km, i.e. in an adjacent model grid cell), shows good performances together with the fulfilment of the criterion, the poor performances at “Corso Firenze Genova” may be explained by very local features of chemical and meteorological conditions not captured by the present simulation.

The Target Plot shows that both negative and positive $BIAS$ were obtained, being the latter one somewhat more frequent, indicating a general slight overestimation of O_3 concentration. Concerning $CRMSE$, it turns out to be dominated by R in most of the cases.

Figure 22 confirms the good model performances: MPC is fulfilled for all the stations concerning both $NMSD$ and R and for all of them (except for “Corso Firenze Genova”) concerning $BIAS$. Globally, the scatter plot in the top left of Figure 22 highlights that modelled values exhibit less variability than observed ones: this outcome is confirmed by the prevalence of negative $NMSD$ values in the top right of Figure 22.

Figure 23 summarizes all this outcomes and points out the fulfilment of MPO_{perc} and spatial criteria too.

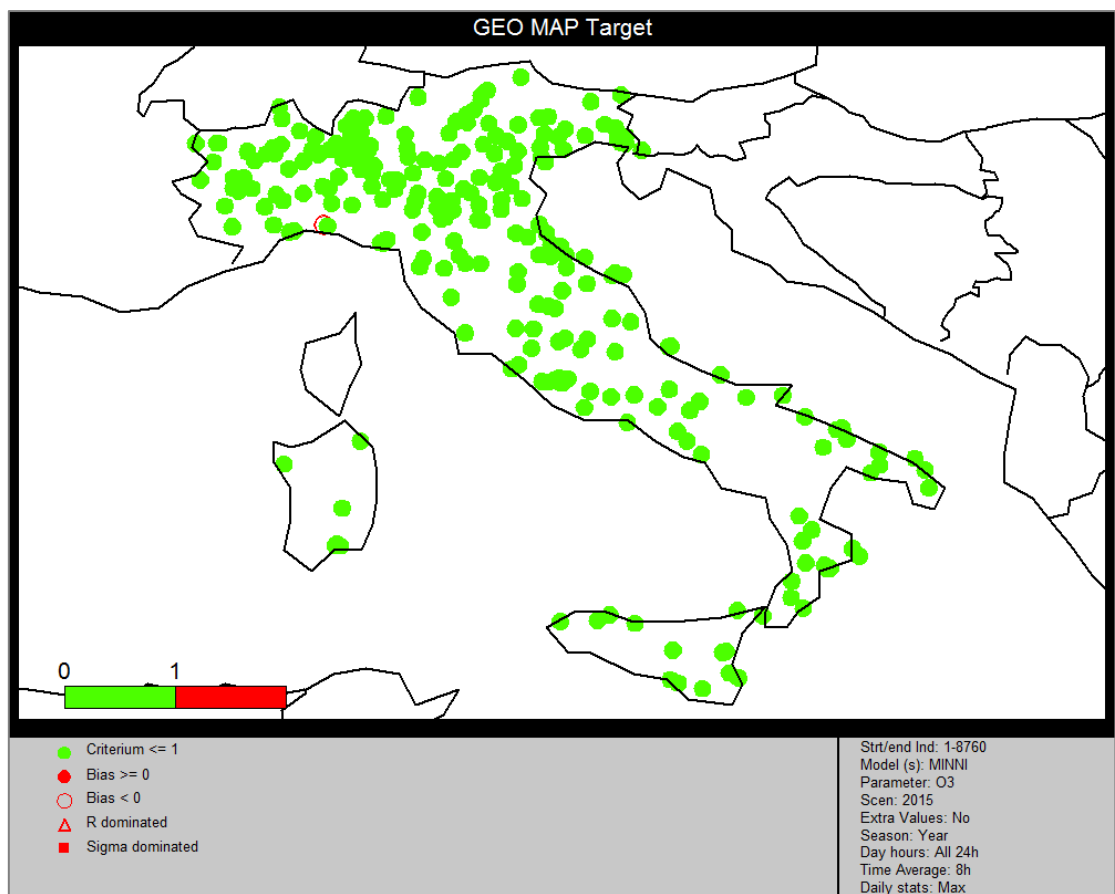
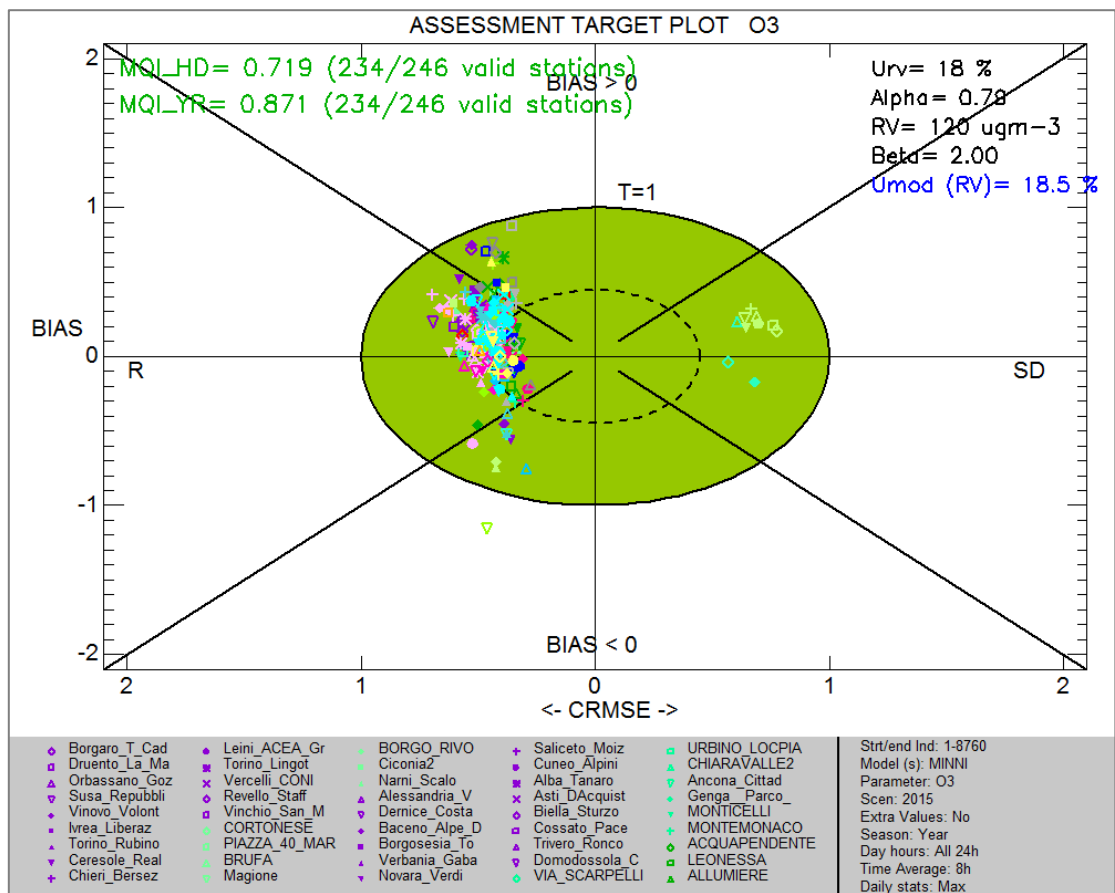


FIGURE 21. MQO FOR DAILY 8h MAXIMUM O₃ CONCENTRATIONS: ASSESSMENT TARGET PLOT (TOP) AND SPATIAL DISTRIBUTIONS OF CRITERION FULFILMENT OUTCOMES (BOTTOM).

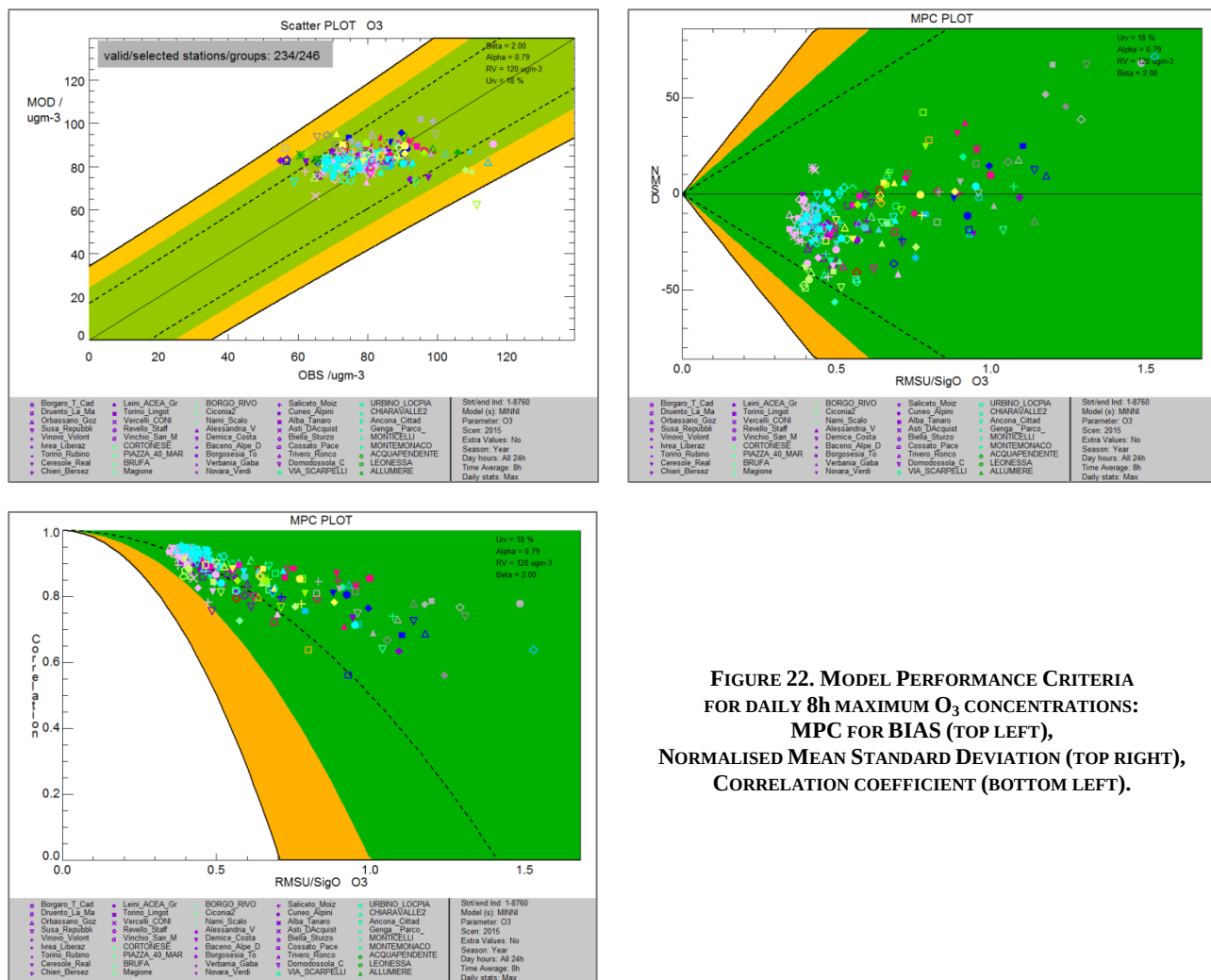


FIGURE 22. MODEL PERFORMANCE CRITERIA FOR DAILY 8h MAXIMUM O₃ CONCENTRATIONS: MPC FOR BIAS (TOP LEFT), NORMALISED MEAN STANDARD DEVIATION (TOP RIGHT), CORRELATION COEFFICIENT (BOTTOM LEFT).

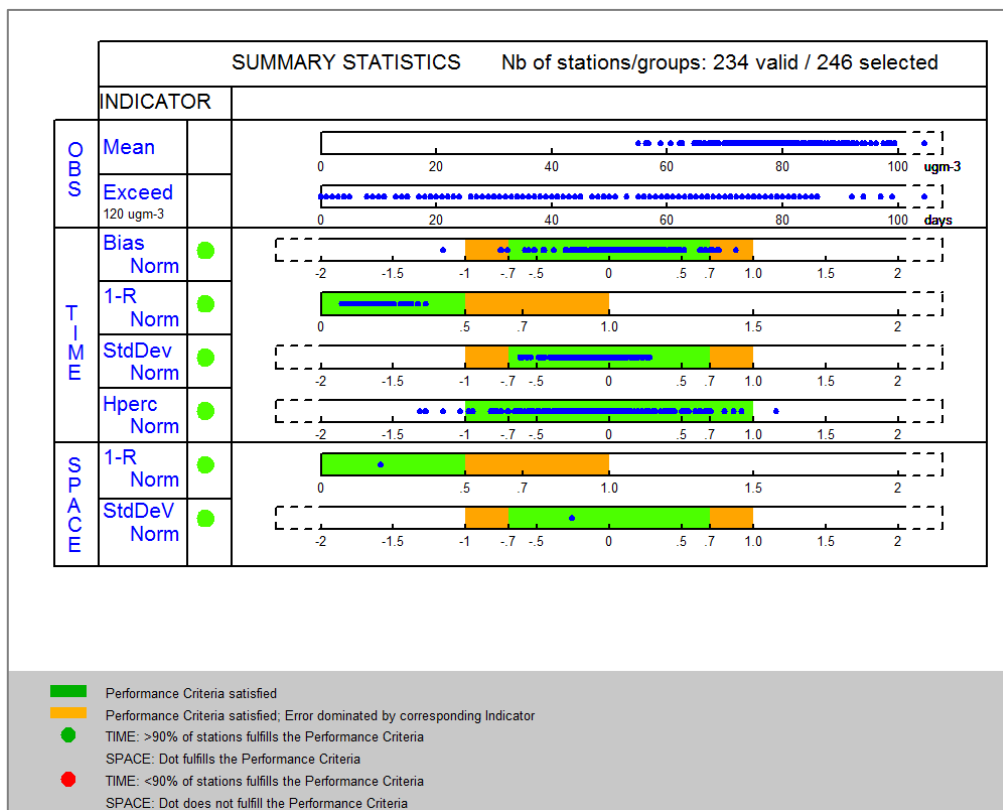


FIGURE 23. BENCHMARKING PERFORMANCE SUMMARY REPORT FOR DAILY 8h MAXIMUM O₃ CONCENTRATIONS.

4.2.3 Particulate matter PM10

Figure 24, Figure 25 and Figure 26 present daily PM10 validation results.

Globally, Figure 24 points out that the overall benchmarking requirements are not satisfied, since both $MQI_{HD_{90th}}$ and $MQI_{YR_{90th}}$ are higher than one; anyway both the quality indicators are not so far from the threshold, especially as far as $MQI_{HD_{90th}}$ is concerned. More in detail, the Target Plot shows that $CRMSE$ is generally dominated by poor R more than by SD differences and that negative $BIAS$ are much more frequent than positive ones, indicating a general PM10 underestimation; this is attributable to the well-known difficulties of air quality models in taking into account all the contributions to PM10 concentration, especially as far as primary components are concerned. It is worth noting that in the present AMS-MINNI simulation Saharan dust contribution was not included.

The plot in the bottom of Figure 24 confirms that negative $BIAS$ and poor R are generally the prevailing contributions of $RMSE$ values. In the same plot, spatial distribution of MQI_{HD} shows that the most critical areas for the MQO_{HD} fulfilment are mainly located in the Northern Italy, in complex mountain orographic contexts, and in the Southern regions where Saharan dust contribution could be more relevant. On the contrary, it is worth noting that the simulation exhibits good performances in the Po-valley region, where, as it is well known, high-polluted events are more frequent.

Even if $MQI_{HD_{90th}}$ is not satisfied, individual $MPCs$ (for all the fraction of the error, i.e. $BIAS$, R , $NMSD$) are fulfilled for more than 90% of the stations, since the overall criterion turns out to be satisfied (green dots in Figure 26). More in detail, concerning $NMSD$, Figure 25 shows that, in the framework of a general underestimation of standard deviations, MPC is fulfilled for all the valid stations, except for only one. Also with regard to $BIAS$ the criterion is satisfied for most of the stations. Only correlation features turn out to be more critical for a not negligible fraction of stations (anyway lower than 10%).

Concerning MPO_{perc} and spatial features, Figure 26 points out that also spatial criteria are satisfied for both correlation and standard deviation. On the contrary MPC_{perc} is not fulfilled indicating poorer performances with regard to model application capability in predicting high concentration values.

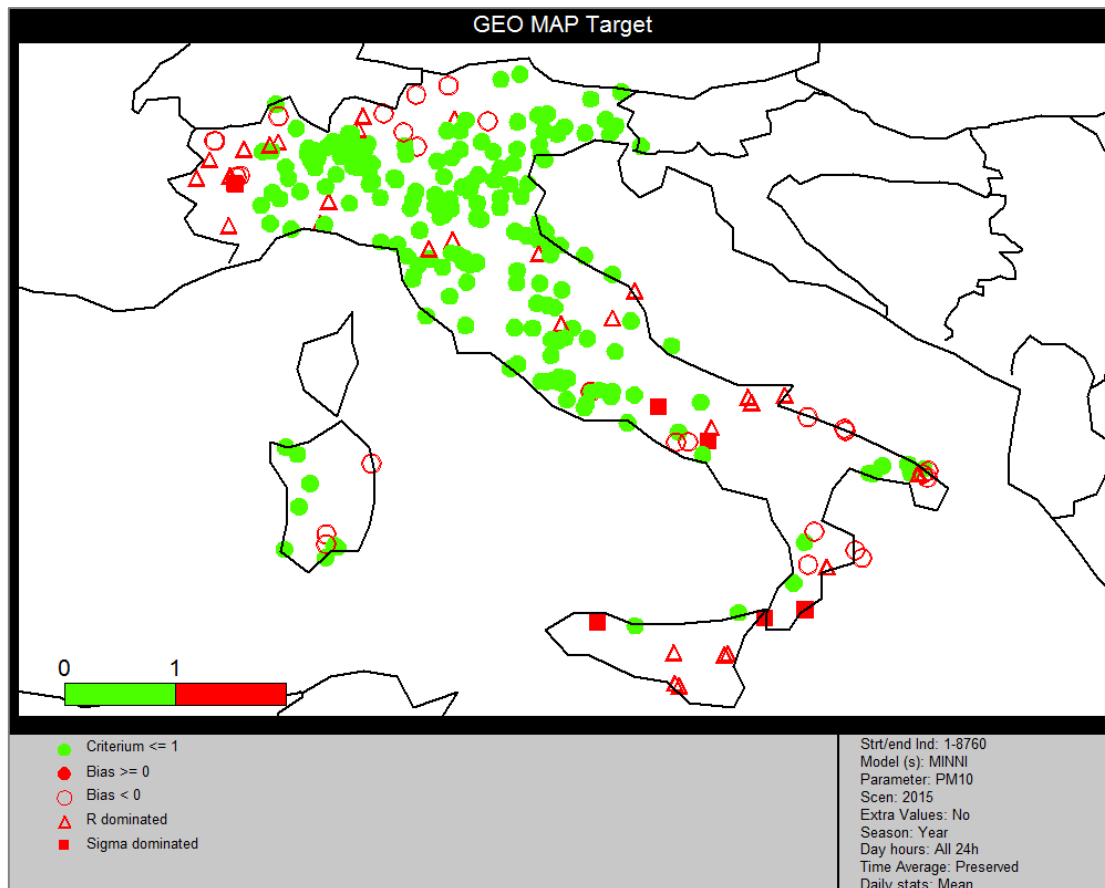
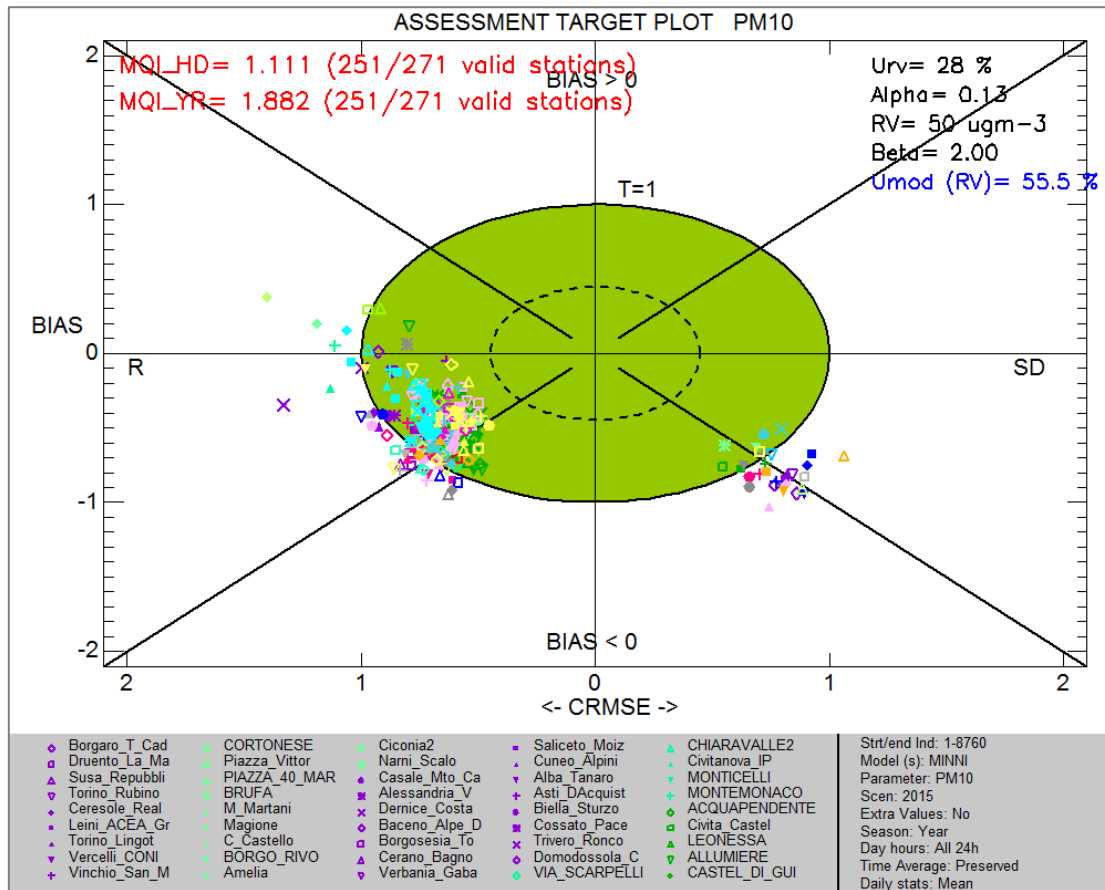


FIGURE 24. MQO FOR DAILY MEAN PM10 CONCENTRATIONS: ASSESSMENT TARGET PLOT (TOP) AND SPATIAL DISTRIBUTIONS OF CRITERION FULFILMENT OUTCOMES (BOTTOM).

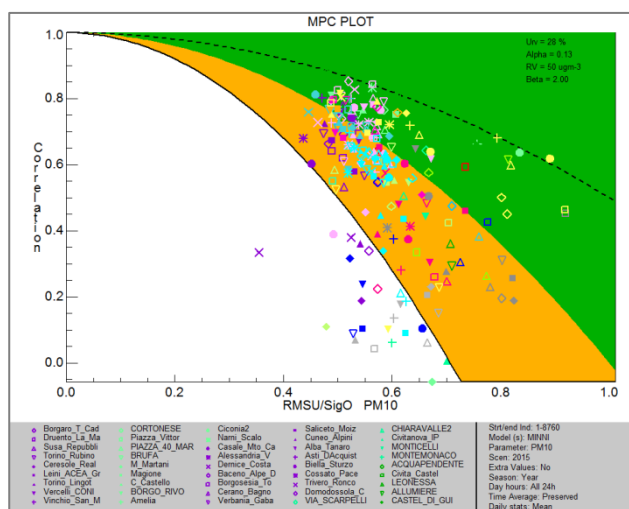
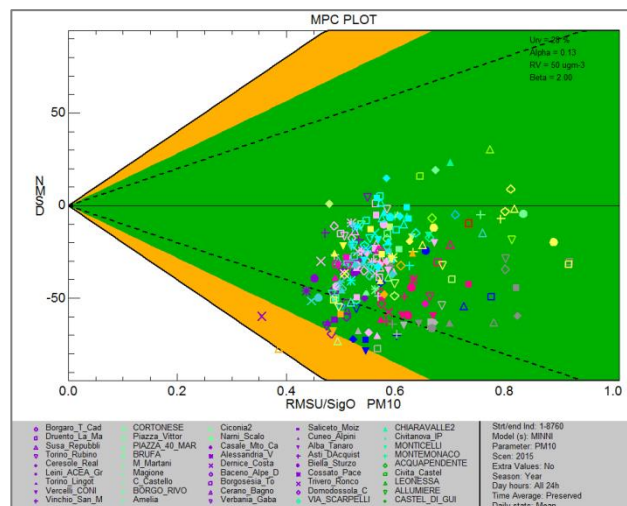
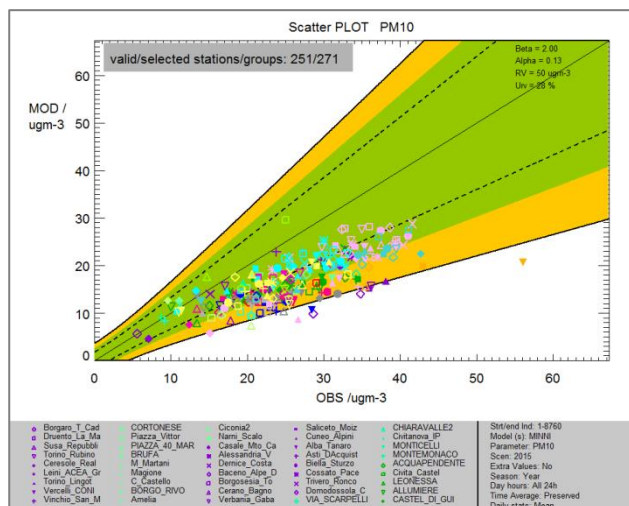


FIGURE 25. MODEL PERFORMANCE CRITERIA FOR DAILY MEAN PM10 CONCENTRATIONS: MPC FOR BIAS (TOP LEFT), NORMALISED MEAN STANDARD DEVIATION (TOP RIGHT), CORRELATION COEFFICIENT (BOTTOM LEFT).

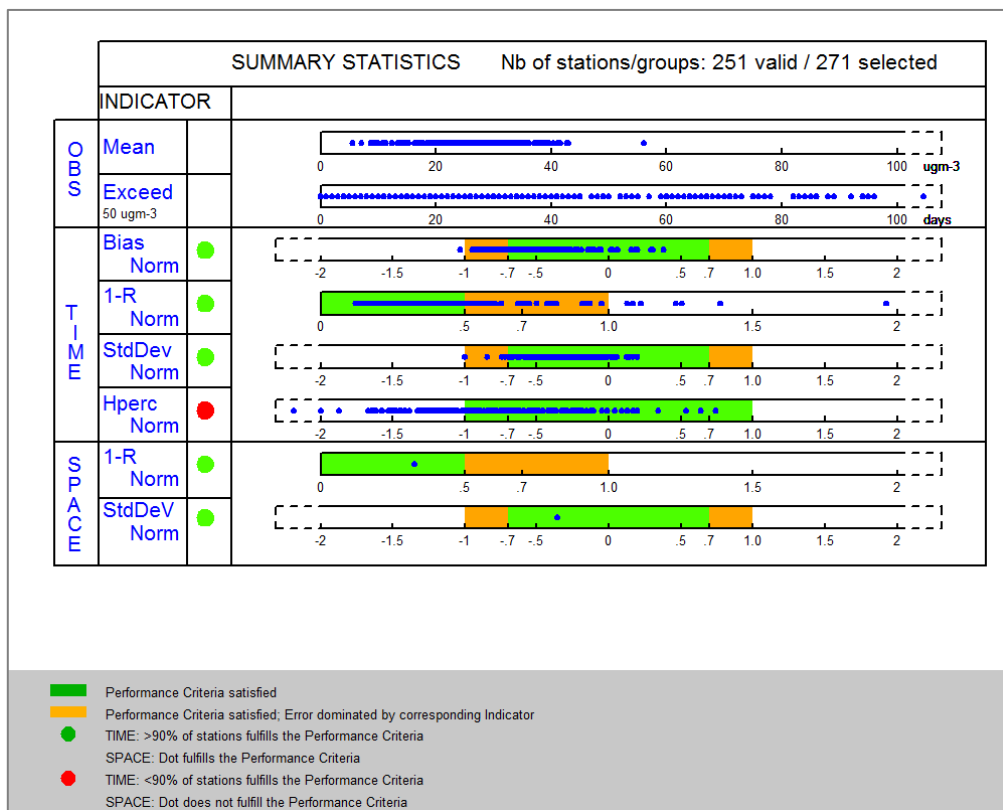


FIGURE 26. BENCHMARKING PERFORMANCE SUMMARY REPORT FOR DAILY MEAN PM10 CONCENTRATIONS.

4.2.4 Particulate matter PM_{2.5}

Concerning daily PM_{2.5} validation, very good performances were achieved, as it can be inferred from all *MQOs* and *MPCs* outcomes in Figure 27, Figure 28 and Figure 29.

In particular, Figure 27 shows that the overall benchmarking requirements are fulfilled, since both *MQI_HD_{90th}* and *MQI_YR_{90th}* are lower than one. Indeed, *MQO_HD* is fulfilled for all the 135 valid stations, except for two: one located in Piemonte region (Biella province) and the other one (with *MQI_HD* very close to one) located in Umbria region (Perugia province). For both of them poor correlation is the main reason for the *MQO_HD* non-compliance. In general, the Target Plot points out that *CRMSE* is dominated by poor *R* in most of the cases. Concerning *BIAS*, both positive and negative values were obtained, being the latter ones more frequent, indicating a general underestimation of PM_{2.5} concentration as previously observed for PM₁₀.

Figure 28 confirms the good model performances: *MPC* is fulfilled for all the stations concerning both *BIAS* and *NMSD* and for all of them (except for the station located in Piemonte) concerning *R*.

Figure 29 summarizes all this outcomes and points out the fulfilment of spatial criteria too.

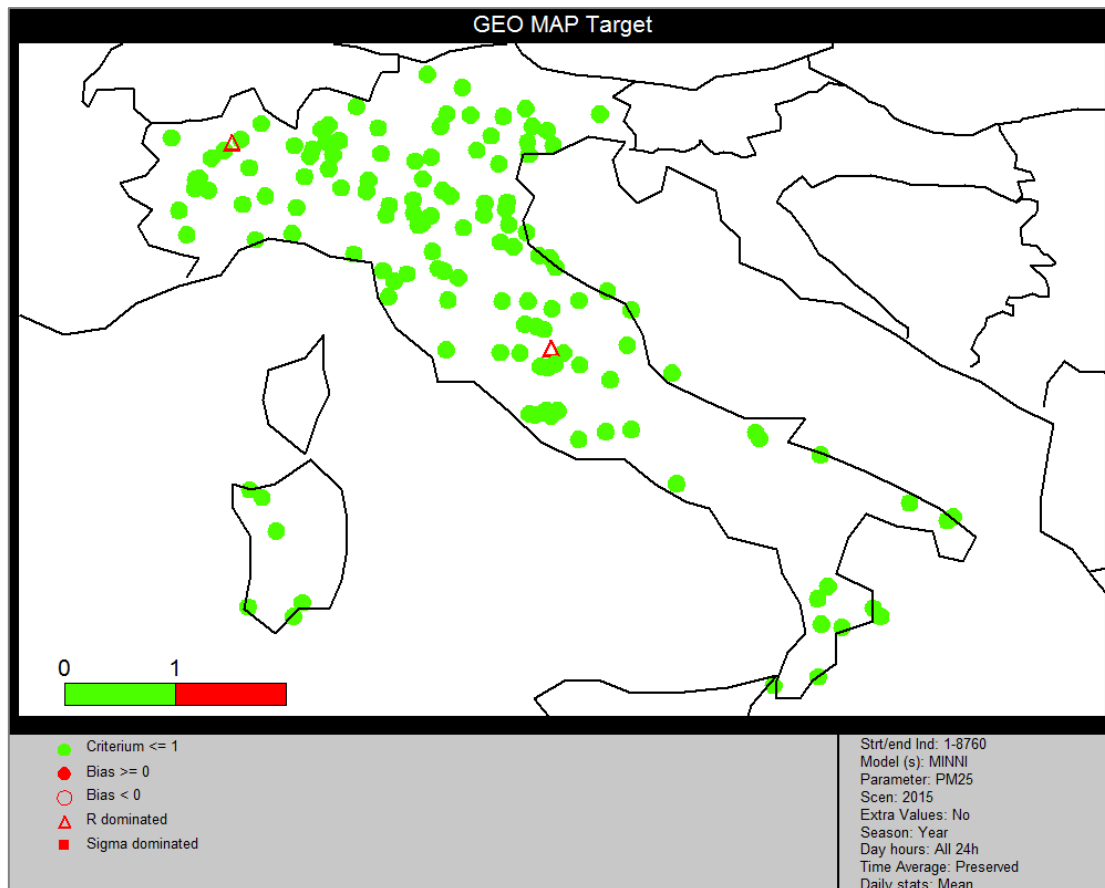
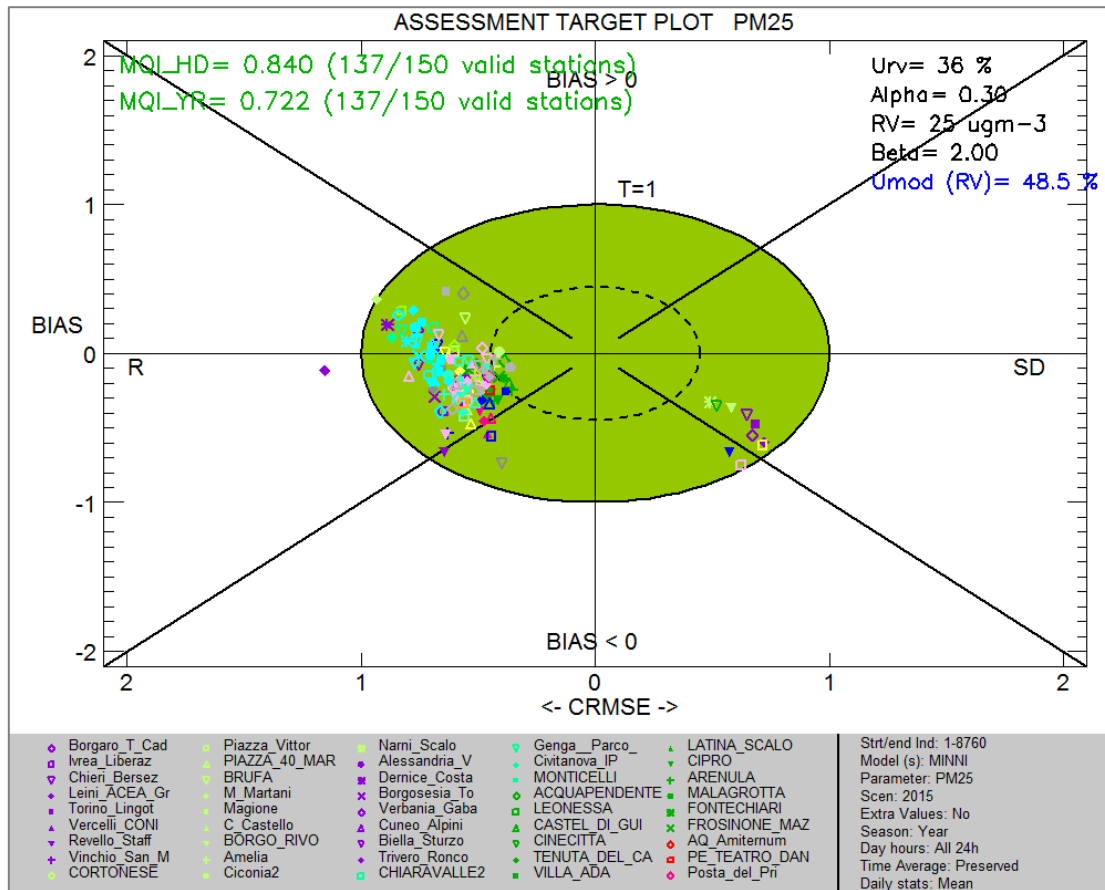


FIGURE 27. MQO FOR DAILY MEAN PM2.5 CONCENTRATIONS: ASSESSMENT TARGET PLOT (TOP) AND SPATIAL DISTRIBUTIONS OF CRITERION FULFILMENT OUTCOMES (BOTTOM).

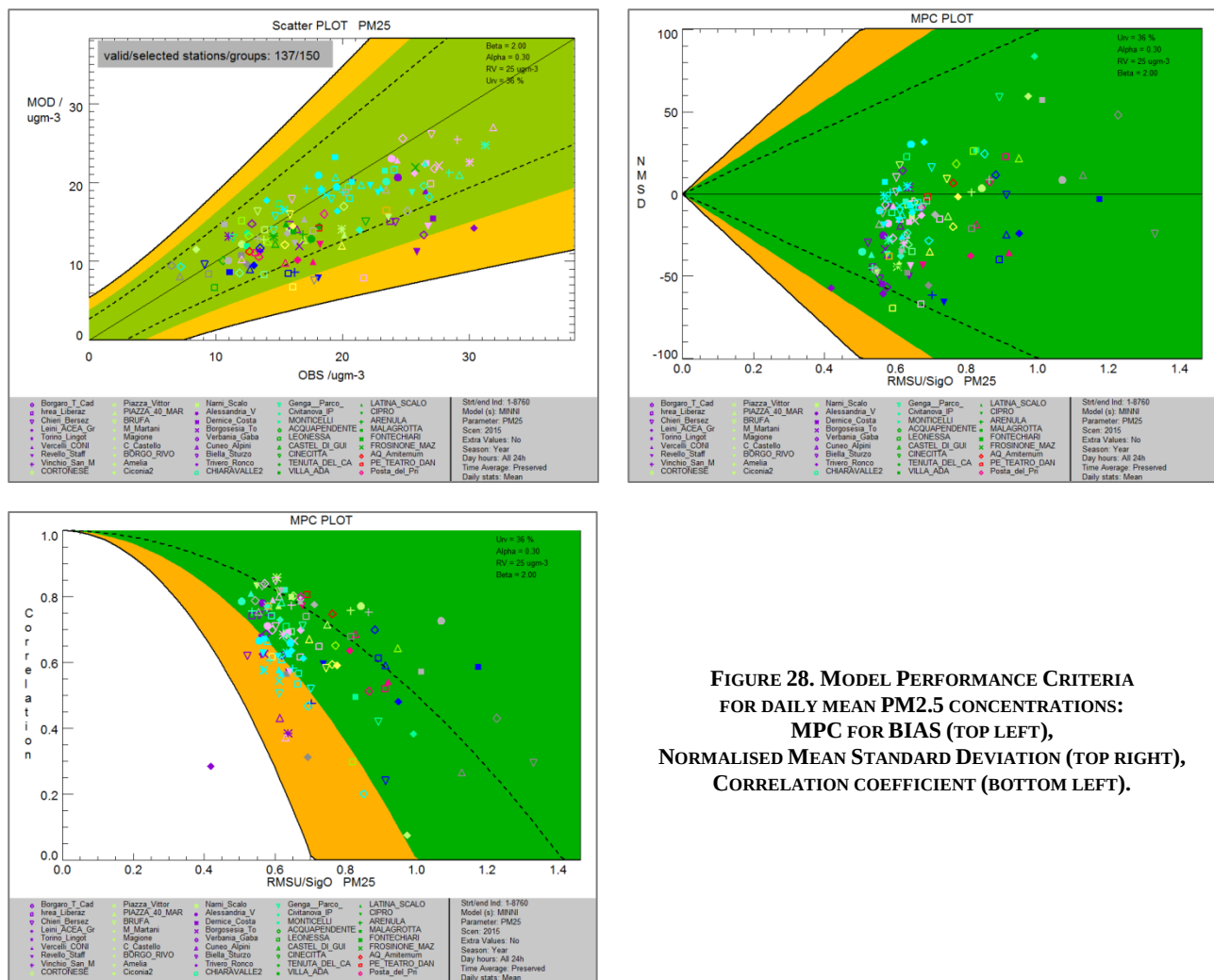


FIGURE 28. MODEL PERFORMANCE CRITERIA FOR DAILY MEAN PM2.5 CONCENTRATIONS: MPC FOR BIAS (TOP LEFT), NORMALISED MEAN STANDARD DEVIATION (TOP RIGHT), CORRELATION COEFFICIENT (BOTTOM LEFT).

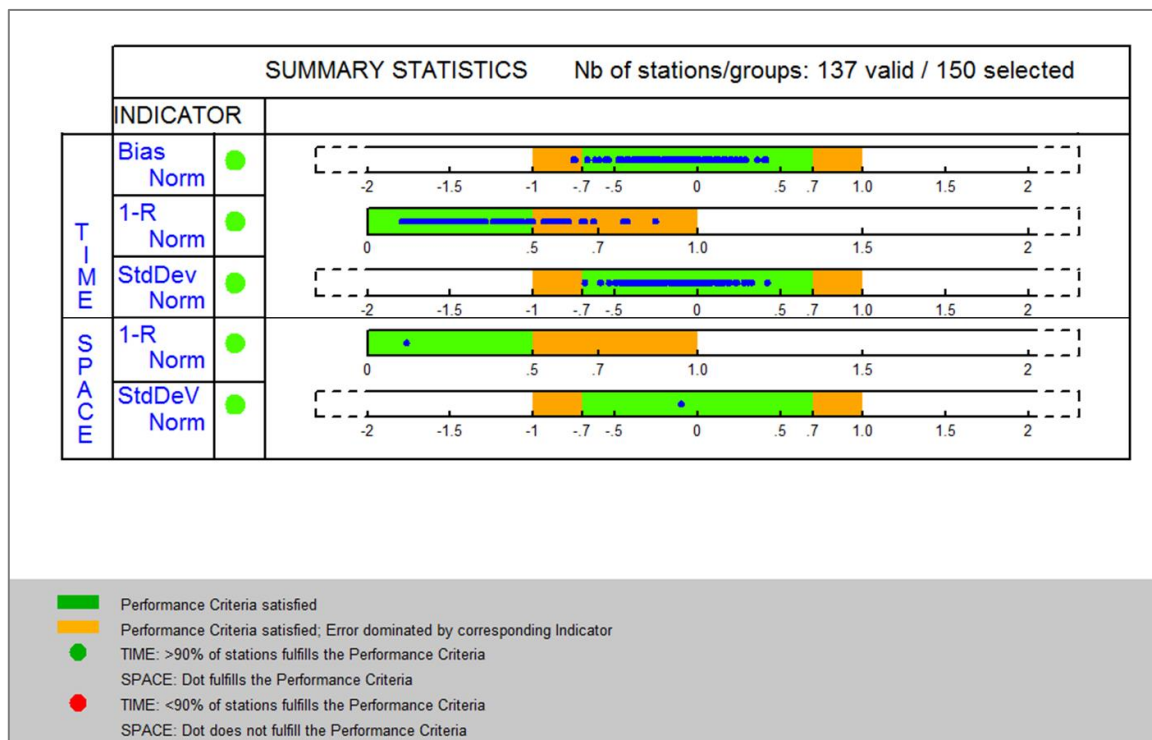


FIGURE 29. BENCHMARKING PERFORMANCE SUMMARY REPORT FOR DAILY MEAN PM2.5 CONCENTRATIONS.

4.3 Evaluation summary

Very good performances were achieved in the benchmarking of the MINNI results for the calendar year 2015, according to FAIRMODE approach and recommendation for model application assessment in the framework of the AQD.

In particular, as far as 8h daily maximum O_3 and daily $PM_{2.5}$ prediction is concerned, all the Model Performance Criteria turn out to be satisfied. Concerning hourly NO_2 and daily PM_{10} both strengths and weaknesses are highlighted: in general individual *MPCs* (for the fraction of the error, i.e. *BIAS*, *R*, *NMSD*) are fulfilled for more than 90% of the stations but it is not always the case for *MQI_XX_{90th}*.

Globally AMS-MINNI tends to overestimate O_3 and to underestimate NO_2 , PM_{10} and $PM_{2.5}$; in some cases, this underestimation can lead to poorer performances, concerning model capability in predicting high concentration values.

Anyway AMS-MINNI behaviour, concerning in particular NO_2 and PM_{10} underestimation, is in line with the state of the art of model performances, when operating at the regional scale. Moreover, according to Legislative Decree 155/2010, ENEA is required to use, for the national simulation, the national emission inventory disaggregated on a provincial level; indeed, the observed underestimations are consistent with the intrinsic spatio-temporal uncertainty of the national top-down emission inventory.

5 CONCLUSION

This report describes the AMS-MINNI national air quality simulation, for the calendar year 2015. It was carried out and made available by ENEA, in fulfilment of Legislative Decree 155/2010 (D.Lgs., 2010).

Two main aspects were outlined:

- On one side, the quality performances of the simulation results were described, checked by means of the state of the art evaluation methodology proposed by FAIRMODE community.
- On the other side, in fulfilment of Legislative Decree 155/2010, the air quality assessment was presented through the discussion of modelled concentration outcomes for the main pollutants regulated by AQD.

Concerning the model validation, it turned out that very good performances were achieved in the benchmarking of 2015 MINNI results. More in details, both O₃ and PM_{2.5} showed excellent skills, being all the Model Performance Criteria satisfied. Slightly poorer performances were found for NO₂ and PM₁₀. In particular AMS-MINNI tends to underestimate both of them, similarly to other model applications operating at the regional scale. According to the article 22 of Legislative Decree 155/2010, for the national simulation, ENEA is required to use the national emission inventory scaled appropriately, so higher resolutions more suitable to capture local emissions and topographic features cannot be produced with a top-down approach. At the same time a bottom-up approach does not fit with a vast domain. Moreover, the PM₁₀ underestimation may be explained by the fact that the coarse PM sources are still missing from emission inventories as previously found in other studies (Pirovano et al, 2012).

Concerning air quality assessment in the framework of Legislative Decree 155/2010, the outcomes of the AMS-MINNI simulation for the calendar year 2015 were presented in terms of both annual mean concentrations and hourly or daily reference concentration percentiles. Modelling results, in agreement with monitoring data collected by ISPRA, highlighted the non-compliance with legislation requirements for several pollutants indicators, in particular for daily PM₁₀, 8h daily maximum O₃ and yearly NO₂ and PM_{2.5}. The meteorological peculiarity of the calendar year 2015, which resulted as one of the driest and the warmest years among the recent ones, could explain the main reasons of the observed and modelled high air pollution levels, in particular as far as photochemical pollution is concerned.

The concentration fields are now available, both as initial and boundary conditions for local scale air quality assessments (according to Legislative Decree 155/2010) and as input data for impact studies at the national level.

REFERENCES

- Adani, M., Mircea, M., D'Isidoro, M., Costa, M.P., Silibello, C., 2015. Heavy Metal Modelling Study over Italy: Effects of Grid Resolution, Lateral Boundary Conditions and Foreign Emissions on Air Concentrations. *Water, Air, & Soil Pollution*, 226:46 <http://dx.doi.org/10.1007/s11270-015-2324-7>.
- Amato, F., Karanasiou, A., Moreno, T., Alastuey, A., Orza, J., Lumbreras, J., Borge, R., Boldo, E., Linares, C., Querol, X., 2012. Emission factors from road dust resuspension in a Mediterranean freeway. *Atmospheric Environment*, 61, 580-587, <http://dx.doi.org/10.1016/j.atmosenv.2012.07.065>.
- Arianet, 2011. SURFPRO3 User's guide (SURFace-atmosphere interface PROcessor, Version 3). Software manual. Arianet R2011.31.
- Arianet, 2014. Emission Manager. Modular processing system for model-ready emission input Preparation. Software Manual.
- Aulinger, A., Matthias, V., Quante, M., 2007. Introducing a partitioning mechanism for PAHs into the community multiscale air quality modeling system and its application to simulating the transport of benzo(a) pyrene over Europe. *Journal of Applied Meteorology and Climatology* 46, 1718-1730, <http://dx.doi.org/10.1175/2007JAMC1395.1>.
- Barnes, S.L., 1964. A Technique for maximizing details in numerical weather map analysis. *Journal of Applied Meteorology*, 3, 396-409, [https://doi.org/10.1175/1520-0450\(1964\)003<0396:ATFMDI>2.0.CO;2](https://doi.org/10.1175/1520-0450(1964)003<0396:ATFMDI>2.0.CO;2).
- Bessagnet, B., Pirovano G., Mircea Mihaela, Cuvelier C., Aulinger A., Calori G., Ciarelli G., Manders A., Stern R., Tsyro S., García Vivanco, M., Thunis, P., Pay, M.-T., Colette, A., Couvidat, F., Meleux, F., Rouïl, L., Ung, A., Aksoyoglu, S., Baldasano, J.M., Bieser, J., Briganti, G., Cappelletti, A., D'Isidoro, M., Finardi, S., Kranenburg, R., Silibello, C., Carnevale, C., Aas, W., Dupont, J.-C., Fagerli, H., Gonzalez, L., Menut, L., Prévôt, A.S.H., Roberts, P., White, L., 2016. Presentation of the EURODELTA III intercomparison exercise-evaluation of the chemistry transport models' performance on criteria pollutants and joint analysis with meteorology. *Atmospheric Chemistry and Physics*, 16 (19), 12667-12701, <https://doi.org/10.5194/acp-16-12667-2016>.
- Binkowski, F.S., Roselle, S.J., 2003. Models-3 community multiscale air quality (CMAQ) model aerosol component 1. Model description. *Journal of Geophysical Research*, 108, 4183, <http://dx.doi.org/10.1029/2001JD001409>, D6.
- Byun, D., Schere, K.L., 2006. Review of the Governing Equations, Computational Algorithms, and Other Components of the Models-3 Community Multiscale Air Quality (CMAQ) Modeling System. *Applied Mechanics Reviews*, 59 (2), 51-77, <https://doi.org/10.1115/1.2128636>.
- Cadum, E., Rowinski, M., Berti, G., Basagaña, X., Ciancarella, L., Spadea, T., Annesi-Maesano, I., Otorepec, P., Zanini, G., Costa, G., 2016. LIFE MED HISS ENV/it/000834: an 'health surveillance' pilot

project on long term effects exposure to air pollution to implement a european system. In: Abstracts of the 2016 meeting of the International Society for Environmental Epidemiology (ISEE), pp P2–310, <https://doi.org/10.1289/ehp.isee2016>.

Carmichael, G.R., Uno, I., Padnis, M.J., Zhang, Y., Sunwoo, Y., 1998. Tropospheric ozone production and transport in the springtime in east Asia. *Journal of Geophysical Research*, 103, 10649-10671, <http://dx.doi.org/10.1029/97JD03740>.

Carter, W.P.L., 2000. Documentation of the SAPRC-99 chemical mechanism for VOC reactivity assessment. Final Report to California Air Resources Board, Contract No. 92-329, and (in part) 95-308. May 8, 2000.

Carter, W.P.L., 2010. Development of the SAPRC-07 chemical mechanism. *Atmospheric Environment*, 44 (40), 5324-5335, ISSN 1352-2310, <https://doi.org/10.1016/j.atmosenv.2010.01.026>.

Chen, C., Cotton, W.R., 1983. A one-dimensional simulation of the stratocumulus-capped mixed layer. *Boundary-Layer Meteorology*, 25, 289–321, ISSN 0006-8314, <http://dx.doi.org/10.1007/BF00119541>.

Ciancarella L. e Zanini G. Progetto Minni - Sistema modellistico per le politiche di qualità dell'aria a supporto di Governo e Regioni. Edizioni ENEA, 2013. ISBN 978-88-8286-287-9, <http://www.enea.it/it/pubblicazioni/pdf-volumi/V2013ProgettoMinni.pdf>.

Ciancarella, L., Adani, M., Ciucci, A., Cremona, G., D'Isidoro, M., Mircea, M., Piersanti, A., Russo, F., Vitali, L., Righini, G., Zanini, G., Briganti, G., Cappelletti, A., D'Elia, I., 2016. La simulazione nazionale di AMS-MINNI relativa all'anno 2010. Simulazione annuale del Sistema Modellistico Atmosferico di MINNI e validazione dei risultati tramite confronto con i dati osservati. ENEA Technical Report, RT/2016/12/ENEA, <http://openarchive.enea.it/bitstream/handle/10840/7628/RT-2016-12-ENEA.pdf>.

Ciarelli, G., Colette, A., Schucht, S., Beekmann, M., Andersson, C., Manders-Groot, A., Mircea, M., Tsyro, S., Fagerli, H., Ortiz, A.G., Adani, M., Briganti, G., Cappelletti, A., D'Isidoro, M., Cuvelier, C., Couvidat, F., Meleux, F., Bessagnet, B., 2019. Long-term health impact assessment of total PM_{2.5} in Europe during the 1990–2015 period, *Atmospheric Environment*: X, 3, 100032, ISSN 2590-1621, <https://doi.org/10.1016/j.aeaoa.2019.100032>.

Ciucci, A., D'Elia, I., Ciancarella, L., Zanini, G., 2014. The Italian optimization tool for the GAINS-Italy model. Rapporto Tecnico RT/2014/10/ENEA, ENEA, ISSN 0393-3016, <http://hdl.handle.net/10840/4937>.

Ciucci, A., D'Elia, I., Wagner, F., Sander, R., Ciancarella, L., Zanini, G., Schöpp, W., 2016. Cost-effective reductions of PM_{2.5} concentrations and exposure in Italy. *Atmospheric Environment*, 140, 84-93, <https://doi.org/10.1016/j.atmosenv.2016.05.049>.

Cotton, W. R., Pielke Sr., R. A., Walko, R. L., Liston, G. E., Tremback, C. J., Jiang, H., McAnelly, R. L., Harrington, J. Y., Nicholls, M. E., Carrio, G. G., McFadden, J. P., 2003. RAMS 2001: Current status and future directions. *Meteorology and Atmospheric Physics*, 82, 5-29, ISSN 0177-7971, <http://dx.doi.org/10.1007/s00703-001-0584-9>.

Damian, V., Sandu, A., Damian, M., Potra, F., Carmichael, G. R., 2002. The kinetic preprocessor KPP-a software environment for solving chemical kinetics. *Computers & Chemical Engineering*, 26, 1567-1579, ISSN 0098-1354, [http://dx.doi.org/10.1016/S0098-1354\(02\)00128-X](http://dx.doi.org/10.1016/S0098-1354(02)00128-X).

D.Lgs., 2010. Decreto Legislativo 13 agosto 2010, n. 155. Attuazione della direttiva 2008/50/CE relativa alla qualità dell'aria ambiente e per un'aria più pulita in Europa (GU n.216 del 15-9-2010 - Suppl. Ordinario n. 217).

D.Lgs., 2018. Decreto Legislativo 30 maggio 2018, n. 81. Attuazione della direttiva (UE) 2016/2284 del Parlamento europeo e del Consiglio, del 14 dicembre 2016, concernente la riduzione delle emissioni nazionali di determinati inquinanti atmosferici, che modifica la direttiva 2003/35/CE e abroga la direttiva 2001/81/CE (GU n. 151 del 2-7-2018).

De Leeuw, G., Neele, F.P., Hill, M., Smith, M.H., Vignati, E., 2000. Production of sea spray aerosol in the surf zone. *Journal of Geophysical Research*, 105(D24), 29397–29409, <http://dx.doi.org/10.1029/2000JD900549>.

D'Elia, I., Bencardino, M., Ciancarella, L., Contaldi, M., Vialletto, G., 2009. Technical and Non-Technical Measures for air pollution emission reduction: The integrated assessment of the regional Air Quality Management Plans through the Italian national model. *Atmospheric Environment*, 43, 6182-6189, ISSN 1352-2310, <http://dx.doi.org/10.1016/j.atmosenv.2009.09.003>.

D'Elia, I., Peschi, E., 2013. Lo scenario emissivo nazionale nella negoziazione internazionale. ENEA Technical Report, RT/2013/10/ENEA, ISSN 0393-3016, <http://openarchive.enea.it/handle/10840/4505>.

D'Elia, I., Piersanti, A., Briganti, G., Cappelletti, A., Ciancarella, L., Peschi, E., 2018. Evaluation of mitigation measures for air quality in Italy in 2020 and 2030. *Atmospheric Pollution Research*, 9, 977-988, <http://dx.doi.org/10.1016/j.apr.2018.03.002>.

De Marco, A., Screpanti, A., Mircea, M., Piersanti, A., Proietti, C., Fornasier, M. F., 2017. High resolution estimates of the corrosion risk for cultural heritage in Italy. *Environmental Pollution*, 226, 260-267, ISSN 0269-7491, <https://doi.org/10.1016/j.envpol.2017.03.066>.

De Marco, A., Proietti, C., Anav, A., Ciancarella, L., D'Elia, I., Fares, S., Fornasier, M.F., Fusaro, L., Gualtieri, M., Manes, F., Marchetto, A., Mircea, M., Paoletti, E., Piersanti, A., Rogora, M., Salvati, L., Salvatori, E., Screpanti, A., Vialletto, G., Vitale, M., Leonardi, C., 2019. Impacts of air pollution on human and ecosystem health, and implications for the National Emission Ceilings Directive: Insights from Italy. *Environment International*, 125, 320-333, ISSN 0160-4120, <https://doi.org/10.1016/j.envint.2019.01.064>.

EEA, 2000. EMEP/CORINAIR Atmospheric emission inventory guidebook - Second edition 1999. Technical report No 30/2000.

EEA, 2017. Air quality in Europe — 2017 report. EEA Report No 13/2017. ISSN 1977-8449, <http://dx.doi.org/10.2800/850018>.

- EMEP, 2003. Transboundary acidification, eutrophication and ground level ozone in Europe. EMEP Status Report 2003, Norwegian Meteorological Institute, August 2003.
- European Commission (EC), 11.06.2008. Directive 2008/50/EC of the European Parliament and of the Council of 21 May 2008 on ambient air quality and cleaner air for Europe (The Framework Directive). Official Journal European Union En. Series, L152/51.
- European Commission (EC), 17.12.2016. Directive (EU) 2016/2284 of the European Parliament and of the Council of 14 December 2016 on the reduction of national emissions of certain atmospheric pollutants, amending Directive 2003/35/EC and repealing Directive 2001/81/EC. Official Journal of the European Union, L. 344/1.
- Fagerli, H., Tsyro, S., Rolstad Denby, B., Nyíri, Á., Gauss, M., Simpson, D., et al., 2017. Transboundary particulate matter, photo-oxidants, acidifying and eutrophying components. EMEP Report 1/2017, <http://smhi.diva-portal.org/smash/get/diva2:1369098/FULLTEXT01.pdf>.
- Fountoukis, C., Nenes, A., 2007. ISORROPIA II: A Computationally Efficient Aerosol Thermodynamic Equilibrium Model for K⁺, Ca²⁺, Mg²⁺, NH₄⁺, Na⁺, SO₄²⁻, NO₃⁻, Cl⁻, H₂O Aerosols. *Atmospheric Chemistry and Physics*, 7, 4639–4659, <https://doi.org/10.5194/acp-7-4639-2007>.
- Gandini, M., Scarinzi, C., Bande, S., Berti, G., Carnà, P., Ciancarella, L., Costa, G., Demaria, M., Ghigo, S., Piersanti, A., Rowinski, M., Spadea, T., Stroschia, M., Cadum, E., 2018. Long term effect of air pollution on incident hospital admissions: Results from the Italian Longitudinal Study within LIFE MED HISS project. *Environment International*, 121 (2), 1087-1097, <https://doi.org/10.1016/j.envint.2018.10.020>.
- Gandini, M., Scarinzi, C., Bande, S., Berti, G., Ciancarella, L., Costa, G., Demaria, M., Ghigo, S., Marinacci, C., Piersanti, A., Sebastiani, G., Cadum, E., 2019. LIFE Med Hiss: An innovative cohort design for public health. *MethodsX*, 6, 82-91, ISSN 2215-0161, <https://doi.org/10.1016/j.mex.2018.12.007>.
- Gariazzo, C., Silibello, C., Finardi, S., Radice, P., Piersanti, A., Calori, G., Cecinato, A., Perrino, C., Nussio, F., Cagnoli, M., Pelliccioni, A., Gobbi, G. P., Di Filippo, P., 2007. A gas/aerosol air pollutants study over the urban area of Rome using a comprehensive chemical transport model. *Atmospheric Environment*, 41, 7286-7303, ISSN 1352-2310, <http://dx.doi.org/10.1016/j.atmosenv.2007.05.018>.
- Ghigo, S., Bande, S., Ciancarella, L., Mircea, M., Piersanti, A., Righini, G., Baldasano, J. M., Basagaña, X., Cadum, E., 2017. Mapping air pollutants at municipality level in Italy and Spain in support to health impact evaluations. *Air Quality, Atmosphere & Health*, 11 (1) (2017), 69-82, <http://dx.doi.org/10.1007/s11869-017-0520-x>.
- Gong, S.L., Barrie, L.A., Blanchet, J.P., von Salzen, K., Lohmann, U., Lesins, G., Spacek, L., Zhang, L.M., Girard, E., Lin, H., Leaitch, R., Leighton, H., Chylek, P., Huang, P., 2003. Canadian Aerosol Module: A size-segregated simulation of atmospheric aerosol processes for climate and air quality models 1. Module development. *Journal of Geophysical Research*, 108(D1), 4007, <http://dx.doi.org/10.1029/2001JD002002>.

- Gryning, S.E, Batchvarova, E., 1996. A model for the height of the internal boundary layer over an area with irregular coastline. *Boundary-Layer Meteorology*, 78, 405–413, ISSN 0006-8314, <http://dx.doi.org/10.1007/BF00120944>.
- Guenther, A., Karl, T., Harley, P., Wiedinmyer, C., Palmer, P. I., Geron, C., 2006. Estimates of global terrestrial isoprene emissions using MEGAN (Model of Emissions of Gases and Aerosols from Nature). *Atmospheric Chemistry and Physics*, 6, 3181-3210, <http://dx.doi.org/10.5194/acp-6-3181-2006>.
- Guerreiro, C.B.B., Foltescu, V., de Leeuw, F., 2014. Air quality status and trends in Europe. *Atmospheric Environment*, 98, 376-384, ISSN 1352-2310, <https://doi.org/10.1016/j.atmosenv.2014.09.017>.
- Heymann, Y., Steenmans, Ch., Croissille, G., Bossard, M., 1994. CORINE Land Cover. Technical Guide. EUR12585 Luxembourg, Office for Official Publications of the European Communities.
- Hov, Ø., Eliassen, A., Simpson, D., 1988. Calculation of the distribution of NO_x compounds in Europe. In Isaksen I.S.A. (ed.) *Tropospheric ozone. Regional and global scale interactions*, 239-262, Dordrecht D. Reidel.
- ISPRA, 2009. La disaggregazione a livello provinciale dell'inventario nazionale delle emissioni. Anni 1990-1995-2000-2005. Rapporti 92/2009 ISPRA. ISBN 978-88-448-0392-6.
- ISPRA, 2016a. Gli indicatori del clima in Italia nel 2015, Anno XI. *Stato dell'Ambiente* 65/2016. <http://www.isprambiente.gov.it/it/pubblicazioni/stato-dellambiente/gli-indicatori-del-clima-in-italia-nel-2015.-anno-xi>.
- ISPRA, 2016b. Qualità dell'ambiente urbano – XII Rapporto (2016) ISPRA *Stato dell'Ambiente* 67/16 ISBN 978-88-448-0793-1.
- ISPRA, 2018. Disaggregazione dell'inventario nazionale 2015. Inventario provinciale delle emissioni in atmosfera. Banca dati delle emissioni atmosferiche per gli anni 1990, 1995, 2000, 2005, 2010, 2015, classificate per livello di attività CORINAIR (SNAP), disaggregate dall'inventario nazionale (Submission 2017), <http://www.sinanet.isprambiente.it/it/sia-ispra/inventaria/disaggregazione-dellinventario-nazionale-2015/view>.
- ISTAT, 2014. I consumi energetici delle famiglie. Anno 2013. <https://www.istat.it/it/archivio/142173>.
- Kukkonen, J., Olsson, T., Schultz, D. M., Baklanov, A., Klein, T., Miranda, A. I., Monteiro, A., Hirtl, M., Tarvainen, V., Boy, M., Peuch, V.-H., Poupkou, A., Kioutsioukis, I., Finardi, S., Sofiev, M., Sokhi, R., Lehtinen, K. E. J., Karatzas, K., San José, R., Astitha, M., Kallos, G., Schaap, M., Reimer, E., Jakobs, H., Eben, K., 2012. A review of operational, regional-scale, chemical weather forecasting models in Europe. *Atmospheric Chemistry and Physics* 12, 1-87, <https://doi.org/10.5194/acp-12-1-2012>.

- Lange, R., 1989. Transferability of a three-dimensional air quality model between two different sites in complex terrain. *Journal of Applied Meteorology*, 28 (7), 665-679, [http://dx.doi.org/10.1175/1520-0450\(1989\)028<0665:TOATDA>2.0.CO;2](http://dx.doi.org/10.1175/1520-0450(1989)028<0665:TOATDA>2.0.CO;2).
- Manes, F., Marando, F., Capotorti, G., Blasi, C., Salvatori, E., Fusaro, L., Ciancarella, L., Mircea, M., Marchetti, M., Chirici, G., Munafò, M., 2016. Regulating Ecosystem Services of forests in ten Italian Metropolitan Cities: Air quality improvement by PM₁₀ and O₃ removal. *Ecological Indicators*, 67, 425-440, ISSN 1470-160X, <https://doi.org/10.1016/j.ecolind.2016.03.009>.
- Mellor, G.L., Yamada, T., 1982. Development of a turbulence closure model for geophysical fluid problems. *Reviews of Geophysics*, 20, 851–875, <http://dx.doi.org/10.1029/RG020i004p00851>.
- Mircea, M., Zanini, G., Briganti, G., Cappelletti, A., Pederzoli, A., Vitali, L., Pace, G., Marri, P., Silibello, C., Finardi, S., Calori G., 2011. Modelling Air Quality over Italy with MINNI Atmospheric Modelling System: from regional to local scale. In: Steyn DG and Trini Castelli S, editors. *Air Pollution Modeling and its Application XXI*, 491-498, ISSN 1874-6519, http://dx.doi.org/10.1007/978-94-007-1359-8_82.
- Mircea, M., Ciancarella, L., Briganti, G., Calori, G., Cappelletti, A., Cionni, I., Costa, M., Cremona, G., D'Isidoro, M., Finardi, S., Pace, G., Piersanti, A., Righini, G., Silibello, C., Vitali, L., Zanini, G., 2014. Assessment of the AMS-MINNI system capabilities to predict air quality over Italy for the calendar year 2005. *Atmospheric Environment*, 84, 178–188, ISSN 1352-2310, <http://dx.doi.org/10.1016/j.atmosenv.2013.11.006>.
- Mircea, M., Grigoros, G., D'Isidoro, M., Righini, G., Adani, M., Briganti, G., Ciancarella, L., Cappelletti, A., Calori, G., Cionni, I., Finardi, S., Larsen, B.R., Pace, G., Perrino, C., Piersanti, A., Silibello, C., Zanini, G., 2016. Impact of grid resolution on aerosol predictions: a case study over Italy. *Aerosol and Air Quality Research*, 16, 1253–1267, <https://doi.org/10.4209/aaqr.2015.02.0058>.
- Mircea, M., Bessagnet, B., D'Isidoro, M., Pirovano, G., Aksoyoglu, S., Ciarelli, G., Tsyro, S., Manders, A., Bieser, J., Stern, R., Vivanco, M.G., Cuvelier, C., Aas, W., Prévôt, A.S.H., Aulinger, A., Briganti, G., Calori, G., Cappelletti, A., Colette, A., Couvidat, F., Fagerli, H., Finardi, S., Kranenburg, R., Rouil, L., Silibello, C., Spindler, G., Poulain, L., Herrmann, H., Jimenez, J.L., Day, D.A., Tiitta, P., Carbone, S., 2019. EURODELTA III exercise: An evaluation of air quality models' capacity to reproduce the carbonaceous aerosol. *Atmospheric Environment*: X, 2, 100018, ISSN 2590-1621, <https://doi.org/10.1016/j.aeaoa.2019.100018>.
- Monahan, E.C., Spiel, D.E., Davidson, K.L., 1986. In: Monahan, E.C., Mac Niocaill, G. (Eds.), *Oceanic Whitecaps*. Riedel, Norwell, MA, pp. 167-174.
- Monteiro, A., Gama, C., Candido, M., Ribeiro, I., Carvalho, D., Lopes, M., 2015. Investigating ozone high levels and the role of sea breeze on its transport. *Atmospheric Pollution Research*, Available online 3 November 2015, ISSN 1309-1042, <http://dx.doi.org/10.1016/j.apr.2015.10.013>.

Monteiro, A., Durka, P., Flandorfer, C., Georgieva, E., Guerreiro, C., Kushta, J., Malherbe, L., Maiheu, B., Miranda, A.I., Santos, G., Stocker, J., Trimpeneers, E., Tognet, F., Stortini, M., Wesseling, J., Janssen, S., Thunis, P., 2018. Strengths and weaknesses of the FAIRMODE benchmarking methodology for the evaluation of air quality models. *Air Quality, Atmosphere & Health*, 11, 373, <https://doi.org/10.1007/s11869-018-0554-8>.

Murphy, A.H. 1988. Skill Scores Based on the Mean Square Error and Their Relationships to the Correlation Coefficient. *Monthly Weather Review*, 116 (12), 2417-2424, [https://doi.org/10.1175/1520-0493\(1988\)116<2417:SSBOTM>2.0.CO](https://doi.org/10.1175/1520-0493(1988)116<2417:SSBOTM>2.0.CO).

Padoan, E., Ajmone-Marsan, F., Querol, X., Amato, F., 2018. An empirical model to predict road dust emissions based on pavement and traffic characteristics. *Environmental Pollution*, 237, 713-720, <https://doi.org/10.1016/j.envpol.2017.10.115>.

Pernigotti, D., Gerboles, M., Belis, C., Thunis, P., 2013. Model quality objectives based on measurement uncertainty. Part II: NO₂ and PM₁₀. *Atmospheric Environment*, 79, 869-878, ISSN 1352-2310, <https://doi.org/10.1016/j.atmosenv.2013.07.045>.

Piersanti, A., Adani, M., Briganti, G., Cappelletti, A., Ciancarella, L., Cremona, G., D'Isidoro, M., Lombardi, C., Pacchierotti, F., Russo, F., Spanò, M., Uccelli, R., Vitali, L., 2018. Air quality modeling and inhalation health risk assessment for a new generation coal-fired power plant in Central Italy. *Science of The Total Environment*, 644, 884-898, ISSN 0048-9697, <https://doi.org/10.1016/j.scitotenv.2018.06.393>.

Pirovano, G., Balzarini, A., Bessagnet, B., Emery, C., Kallos, G., Meleux, F., Mitsakou, C., Nopmongkol, U., Riva, G.M., Yarwood, G., 2012. Investigating impacts of chemistry and transport model formulation on model performance at European scale. *Atmospheric Environment* 53, 93-109, <https://doi.org/10.1016/j.atmosenv.2011.12.052>.

Ponti, G., Palombi, F., Abate, D., Ambrosino, F., Aprea, G., Bastianelli, T., Beone, F., Bertini, R., Bracco, G., Caporicci, M., Calosso, B., Chinnici, M., Colavincenzo, A., Cucurullo, A., D'Angelo, P., De Rosa, M., De Michele, P., Funel, A., Furini, G., Giammattei, D., Giusepponi, S., Guadagni, R., Guarnieri, G., Italiano, A., Magagnino, S., Mariano, A., Mencuccini, G., Mercuri, C., Migliori, S., Ornelli, P., Pecoraro, S., Perozziello, A., Pierattini, S., Podda, S., Poggi, F., Quintiliani, A., Rocchi, A., Scio, C., Simoni, F., Vita, A., 2014. The role of medium size facilities in the HPC ecosystem: the case of the new CRESCO4 cluster integrated in the ENEAGRID infrastructure. In: *Proceedings of the 2014 international conference on high performance computing and simulation, HPCS 2014*, art. no. 6903807, 1030–1033.

Schell, B., Ackermann, I. J., Hass, H., Binkowski, F. S., Ebel, A., 2001. Modeling the formation of secondary organic aerosol within a comprehensive air quality modeling system. *Journal of Geophysical Research*, 106, D22, 28275-28293, <https://doi.org/10.1029/2001JD000384>.

Schwartz, S.E., 1986. Mass transport considerations pertinent to aqueous-phase reactions of gases in liquid water clouds. In *Chemistry of multiphase atmospheric systems*, NATO ASI Series, G6, 415-471.

- Seigneur, C., Abeck, H., Chia, G., Reinhard, M., Bloom, N.S., Prestbo, E., Saxena, P., 1998. Mercury adsorption to elemental carbon (soot) particles and atmospheric particulate matter. *Atmospheric Environment*, 32, 2649-2657, ISSN 1352-2310, [http://dx.doi.org/10.1016/S1352-2310\(97\)00415-9](http://dx.doi.org/10.1016/S1352-2310(97)00415-9).
- Seinfeld, J.H., Pandis, S. N., 1998. *Atmospheric Chemistry and Physics*. John Wiley&Sons, Inc.
- Silibello, C., Calori, G., Brusasca, G., Giudici, A., Angelino, E., Fossati, G., Peroni, E., Buganza, E., 2008. Modelling of PM10 concentrations over Milano urban area using two aerosol modules. *Environmental Modelling & Software*, 23, 333-343, ISSN 1364-8152, <http://dx.doi.org/10.1016/j.envsoft.2007.04.002>.
- Silibello, C., Calori, G., Costa, M.P., Dirodi, M.G., Mircea, M., Radice, P., Vitali, L., Zanini, G., 2012. Benzo[a]pyrene modelling over Italy: comparison with experimental data and source apportionment. *Atmospheric Pollution Research*, 3, 399-407, ISSN 1309-1042, <http://dx.doi.org/10.5094/APR.2012.046>.
- Smagorinsky, J., 1963. General circulation experiments with the primitive equations: 1. The basic experiment. *Monthly Weather Review*, 91, 99-164, [https://doi.org/10.1175/1520-0493\(1963\)091<0099:GCEWTP>2.3.CO;2](https://doi.org/10.1175/1520-0493(1963)091<0099:GCEWTP>2.3.CO;2).
- Telloli, C., Malaguti, A., Mircea, M., Berico, M., Piersanti, A., Ciancarella, L., 2017. Aerosol emission factors estimated for harvest threshing and ploughing in Po Valley. ENEA Technical Report, RT/2017/13/ENEA, ISSN 0393-3016, <http://openarchive.enea.it/handle/10840/8666>.
- Theobald, M.R., Vivanco, M.G., Aas, W., Andersson, C., Ciarelli, G., Couvidat, F., Cuvelier, K., Manders, A., Mircea, M., Pay, M.-T., Tsyro, S., Adani, M., Bergstrom, R., Bessagnet, B., Briganti, G., Cappelletti, A., D'Isidoro, M., Fagerli, H., Mar, K., Otero, N., Raffort, V., Roustan, Y., Schaap, M., Wind, P., Colette, A., 2019. An evaluation of European nitrogen and sulfur wet deposition and their trends estimated by six chemistry transport models for the period 1990–2010. *Atmospheric Chemistry and Physics*, 19, 379–405, <https://doi.org/10.5194/acp-19-379-2019>.
- Thunis, P., Pederzoli, A., Pernigotti, D., 2012a. Performance criteria to evaluate air quality modeling applications. *Atmospheric Environment*, 59, 476–482, ISSN 1352-2310, <https://doi.org/10.1016/j.atmosenv.2012.05.043>.
- Thunis, P., Georgieva, E., Pederzoli, A., 2012b. A tool to evaluate air quality model performances in regulatory applications. *Environmental Modelling & Software*, 38, 220-230, ISSN 1364-8152, <https://doi.org/10.1016/j.envsoft.2012.06.005>.
- Thunis, P., Pernigotti, D., Gerboles, M., 2013. Model quality objectives based on measurement uncertainty. Part I: Ozone. *Atmospheric Environment*, 79, 861-868, ISSN 1352-2310, <https://doi.org/10.1016/j.atmosenv.2013.05.018>.
- Tremback, C.J., 1990. Numerical simulation of a mesoscale convective complex: Model development and numerical results. PhD Diss., Colorado State University, Dissertation Abstracts International, 51-06, B, 2941.

UNECE, 2004. Modelling and assessment of the health impact of particulate matter and ozone. Economic commission for Europe, Executive body for the convention on long-range transboundary air pollution. Working group on effects, twenty-third session; Geneva. 1–3 September 2004. EB.AIR/WG.1/2004/11. <https://www.unece.org/fileadmin/DAM/env/documents/2004/eb/wg1/eb.air.wg1.2004.11.e.pdf>.

Vivanco, M.G., Bessagnet, B., Cuvelier, C., Theobald, M.R., Tsyro, S., Pirovano, G., Aulinger, A., Bieser, J., Calori, G., Ciarelli, G., Manders, A., Mircea, Mihaela, Aksoyoglu, S., Briganti, G., Cappelletti, A., Colette, A., Couvidat, F., D'Isidoro, M., Kranenburg, R., Meleux, F., Menut, L., Pay, M.T., Rouïl, L., Silibello, C., Thunis, P., Ung, A., 2017. Joint analysis of deposition fluxes and atmospheric concentrations of inorganic nitrogen and sulphur compounds predicted by six chemistry transport models in the frame of the EURODELTAIII project. *Atmospheric Environment*, 151, 152–175, <http://dx.doi.org/10.1016/j.atmosenv.2016.11.042>.

Vivanco, M.G., Theobald, M.R., García-Gómez, H., Garrido, J.L., Prank, M., Aas, W., Adani, M., Alyuz, U., Andersson, C., Bellasio, R., Bessagnet, B., Bianconi, R., Bieser, J., Brandt, J., Briganti, G., Cappelletti, A., Curci, G., Christensen, J.H., Colette, A., Couvidat, F., Cuvelier, C., D'Isidoro, M., Flemming, J., Fraser, A., Geels, C., Hansen, K.M., Hogrefe, C., Im, U., Jorba, O., Kitwiroon, N., Manders, A., Mircea, M., Otero, N., Pay, M.-T., Pozzoli, L., Solazzo, E., Tsyro, S., Unal, A., Wind, P., Galmarini, S., 2018. Modeled deposition of nitrogen and sulfur in Europe estimated by 14 air quality model systems: evaluation, effects of changes in emissions and implications for habitat protection. *Atmospheric Chemistry and Physics*, 18, 10199–10218, <https://doi.org/10.5194/acp-18-10199-2018>.

Walko, R.L., Tremback, C.J., Pielke, R.A., Cotton, W.R., 1995. An interactive nesting algorithm for stretched grids and variable nesting ratios. *Journal of Applied Meteorology*, 34, 994–999, [http://dx.doi.org/10.1175/1520-0450\(1995\)034<0994:AINAFS>2.0.CO;2](http://dx.doi.org/10.1175/1520-0450(1995)034<0994:AINAFS>2.0.CO;2).

Walko, R.L., Band, L.E., Baron, J., Kittel, T.G. F., Lammers, R., Lee, T.J., Ojima, D., Pielke, R.A., Taylor, C., Tague, C., Tremback, C. J., Vidale, P. L., 2000. Coupled Atmosphere–Biophysics–Hydrology Models for Environmental Modeling. *Journal of Applied Meteorology*, 39, 931–944, [http://dx.doi.org/10.1175/1520-0450\(2000\)039<0931:CABHMF>2.0.CO;2](http://dx.doi.org/10.1175/1520-0450(2000)039<0931:CABHMF>2.0.CO;2).

Yarwood, G., Lau, S., Jia, Y., Karamchandani, P., Vijayaraghavan, K., 2003. Modeling Atmospheric Mercury Chemistry and Deposition with CAMx for a 2002 Annual Simulation. Final Report., ENVIRON, 2003.

Zender, C. S., Bian, H., Newman, D., 2003. Mineral Dust Entrainment and Deposition (DEAD) model: Description and 1990s dust climatology, *Journal of Geophysical Research*, 108, 4416, <http://dx.doi.org/10.1029/2002JD002775>, D14.

Zhang, K.M., Knipping, E.M., Wexler, A.S., Bhave, P.V., Tonnesen, G.S., 2005. Size distribution of sea-salt emissions as a function of relative humidity. *Atmospheric Environment*, 39, 3373–3379, ISSN 1352-2310, <http://dx.doi.org/10.1016/j.atmosenv.2005.02.032>.

Zilitinkevich, S., Baklanov, A., 2001. Calculation of the Height of Stable Boundary Layers in Operational Models. Danish Meteorological Institute scientific report ISBN 978-87-7478-444-9.

APPENDIX: STATISTICAL EVALUATION OF METEOROLOGICAL SIMULATION

Meteorological simulation, conducted for year 2015 with RAMS model over Italy at 4 km x 4 km horizontal resolution, was compared with observational data made available from the environmental agencies of the following administrative regions: ARPA Piemonte, ARPA Lombardia, ARPA Emilia Romagna, ARPA Veneto, ARPA Friuli Venezia Giulia, ARPA Toscana, ARPA Lazio and ARPA Puglia. In particular hourly temperature (T), Wind Speed (WS), Relative Humidity (RH) and precipitation (PREC) observations were considered to perform comparisons with modelled quantities. As an example, Figure A1 depicts the spatial distribution of measurement sites for T. Even if data coverage is not uniform over the country, Northern Italy and Po Valley, the most affected areas by air pollution problems, are well covered.

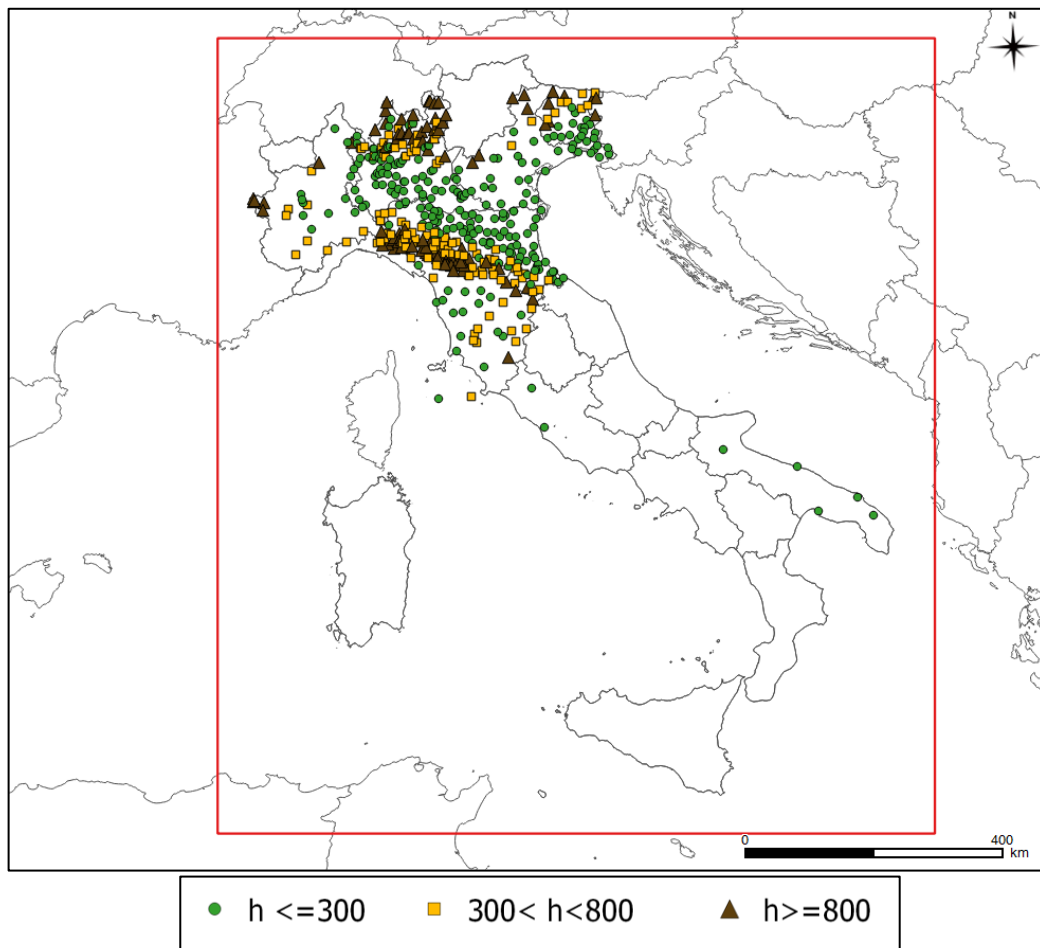


FIGURE A1. GEOGRAPHICAL POSITION OF SITES MEASURING TEMPERATURE: GREEN CIRCLES INDICATE LOW ELEVATION STATIONS (<300m A.M.S.L.); ORANGE SQUARES MID ALTITUDE STATIONS (BETWEEN 300m AND 800m), WHILE BROWN TRIANGLES REPRESENT MOUNTAIN SITES (ABOVE 800m).

Analyses were conducted grouping stations following their elevation above mean sea level to roughly represent locations at plain, hill and mountain altitudes. In Figure A1, green circles are used for stations having altitude (h) below or equal to 300m (i.e. plain); orange squares for stations between 300m and 800m (i.e. hill), while brown triangles indicate stations having altitude above or equal 800m (i.e. mountain).

In order to compare simulated with observed quantities, statistical indicators such as correlation coefficient (*CORR*), bias (*BIAS*), Root Mean Square Error (*RMSE*) and Index Of Agreement (*IOA*)³ were computed considering hourly time series for T, RH and WS, while specific indicators for PREC were calculated over daily accumulations.

The above indicators are largely used in meteorological studies and are among those recommended by FAIRMODE (Forum in air quality modelling in Europe, <https://fairmode.jrc.ec.europa.eu/index.html>). Moreover, Emery et al. (2001) and Tesche et al. (2002) reviewed literature data on the benchmark values for the statistical indicators of meteorological parameters when used for air quality applications: for temperature they suggest *IOA* to be equal or above 0.8 and *BIAS* between -0.5K and 0.5K; for wind speed, *RMSE* less than 2m/s, *IOA* above 0.6 and *BIAS* between -0.5 m/s and 0.5 m/s are suggested; for humidity, *IOA* above 0.6 is recommended. Elaborations are presented using boxplots. This kind of graphical representation contains information about the data distribution such as the median, 1st and 3rd quartiles and outliers, as schematically shown in Figure A2.

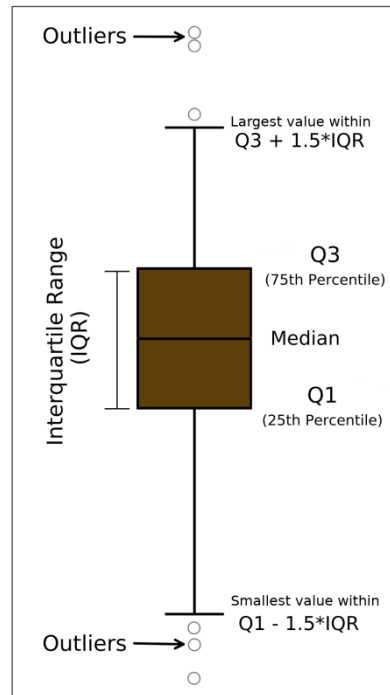


FIGURE A2. SCHEMATIC DESCRIPTION OF BOXPLOTS REPRESENTATION.

³ For the definitions of correlation coefficient (*CORR* or *R*), bias (*BIAS*) and Root Mean Square Error (*RMSE*) see equations (3), (2) and (1), respectively. Index Of Agreement (*IOA*) is defined as follows:

$$IOA = 1 - \frac{\sum_{i=1}^N (M_i - O_i)^2}{\sum_{i=1}^N [|M_i - \bar{O}| + |O_i - \bar{O}|]^2} \quad (A1)$$

Figures A3, A4 and A5 show boxplots for statistical scores computed over annual hourly time series at the measurement sites for temperature, relative humidity and wind speed, respectively. Plots are presented as a function of station altitude intervals.

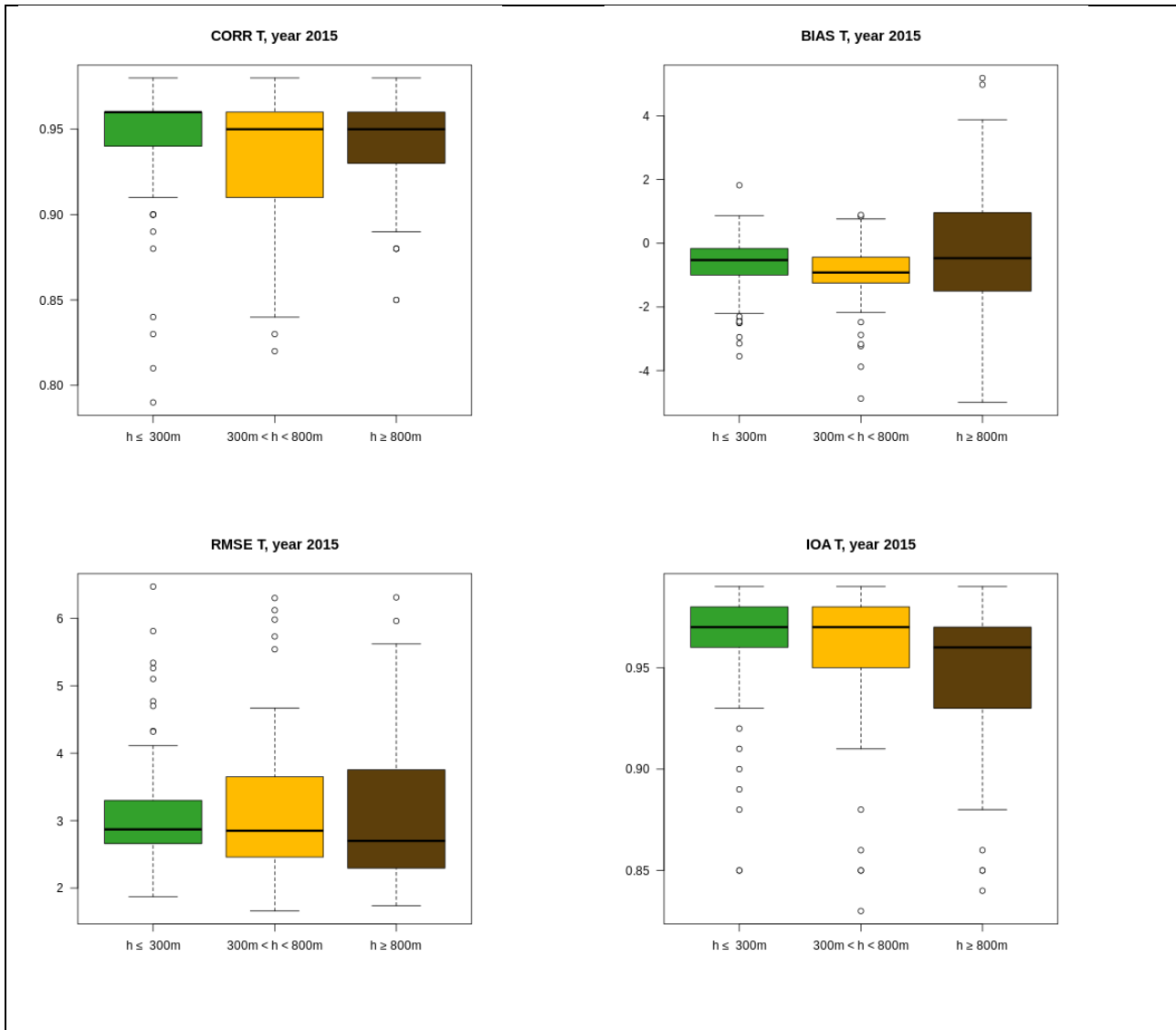


FIGURE A3. STATISTICAL SCORES COMPUTED FOR TEMPERATURE. FROM TOP LEFT CORNER, CLOCKWISE: *CORR*, *BIAS*, *IOA* AND *RMSE*. STATIONS ARE GROUPED FOLLOWING ALTITUDE ABOVE MEAN SEA LEVEL: 300m OR BELOW (GREEN), BETWEEN 300m AND 800m (ORANGE), 800m OR ABOVE (BROWN).

Statistics for temperature were computed having available 246 sites below 300m, 129 at mid-level and 97 over 800m. In general RAMS performs very well for all the adopted metrics. In Figure A3, top left, median of correlation distribution is shown to be near 0.95 or more at all altitudes and, except very few cases, sites have values higher than 0.9. Temperature median *BIAS* (top, right) is around -0.5K, with a higher spread at mountain sites, probably due to model deficiency in simulating temperature daily cycle at narrow valleys, not well captured at the adopted grid resolution. Median *RMSE* (bottom, left) is below 3K at all levels with the spread among sites increasing with altitude. Finally, median of *IOA* for temperature sites distribution (bottom, right) has values above 0.95, with best results for plain and hill.

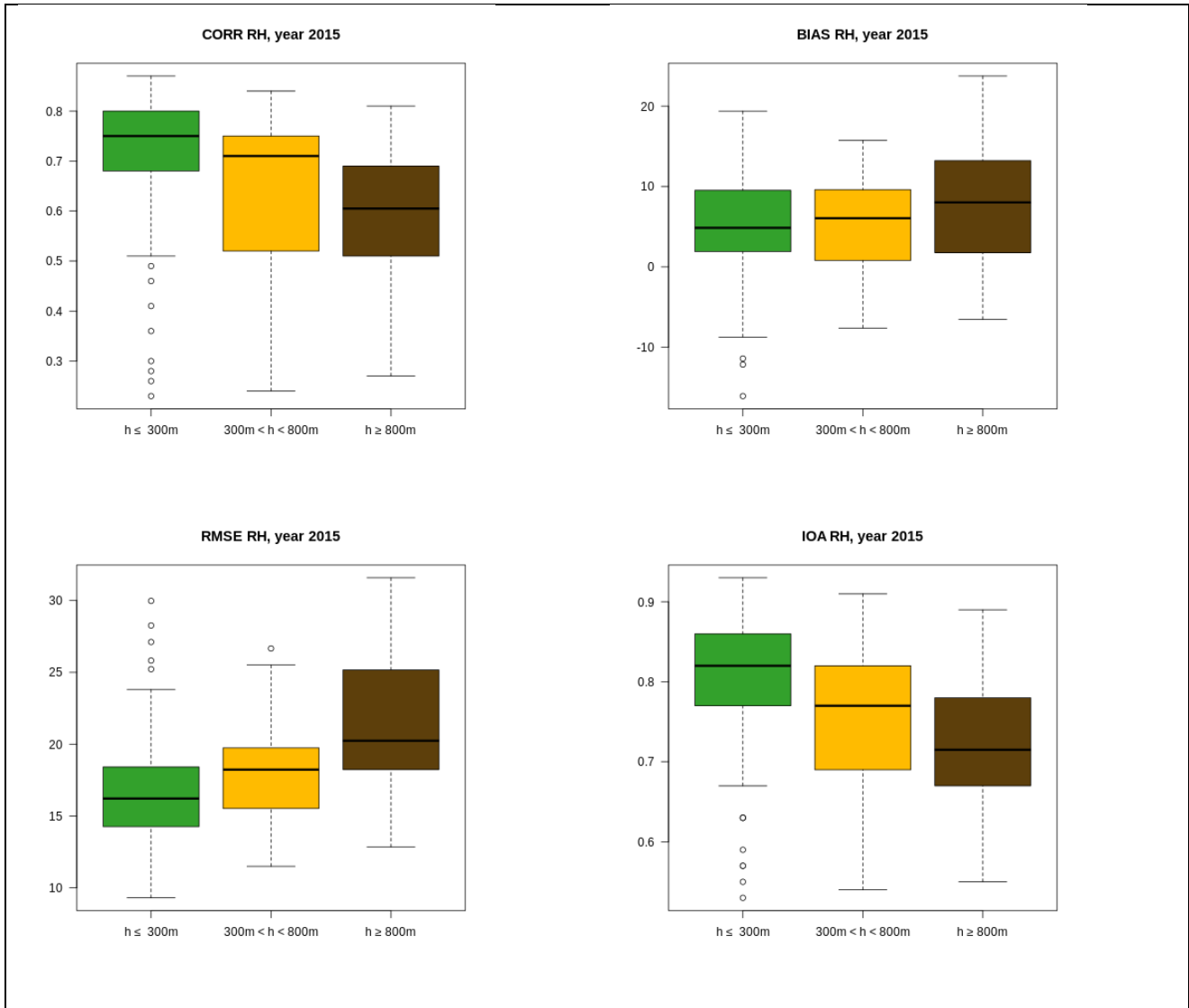


FIGURE A4. STATISTICAL SCORES COMPUTED FOR RH. FROM TOP LEFT CORNER, CLOCKWISE: *CORR*, *BIAS*, *IOA* AND *RMSE*. STATIONS ARE GROUPED FOLLOWING ALTITUDE ABOVE MEAN SEA LEVEL: 300m OR BELOW (GREEN), BETWEEN 300m AND 800m (ORANGE), 800m OR ABOVE (BROWN).

Relative humidity statistics were computed having available 198 sites below 300m, 61 at mid-level and 52 over 800m. Even in this case RAMS shows satisfactory skills, in agreement with the benchmarks suggested in Emery et al. (2001) and Tesche et al. (2002). In Figure A4, top left, the median of correlation distribution for RH decreases from 0.75 at plain to around 0.6 at mountain sites, while the median *BIAS* (top, right) is found between 5% (plain) and 8% (mountain) with a higher spread at mountain sites. The tendency to worsen the skill with altitude is confirmed looking at *RMSE* (bottom, left) which median ranges from about 15% to 20% passing from plain to mountain stations. The median of *IOA* (bottom, right) is between 0.7 and 0.8; best results were obtained for locations at altitude below 300m.

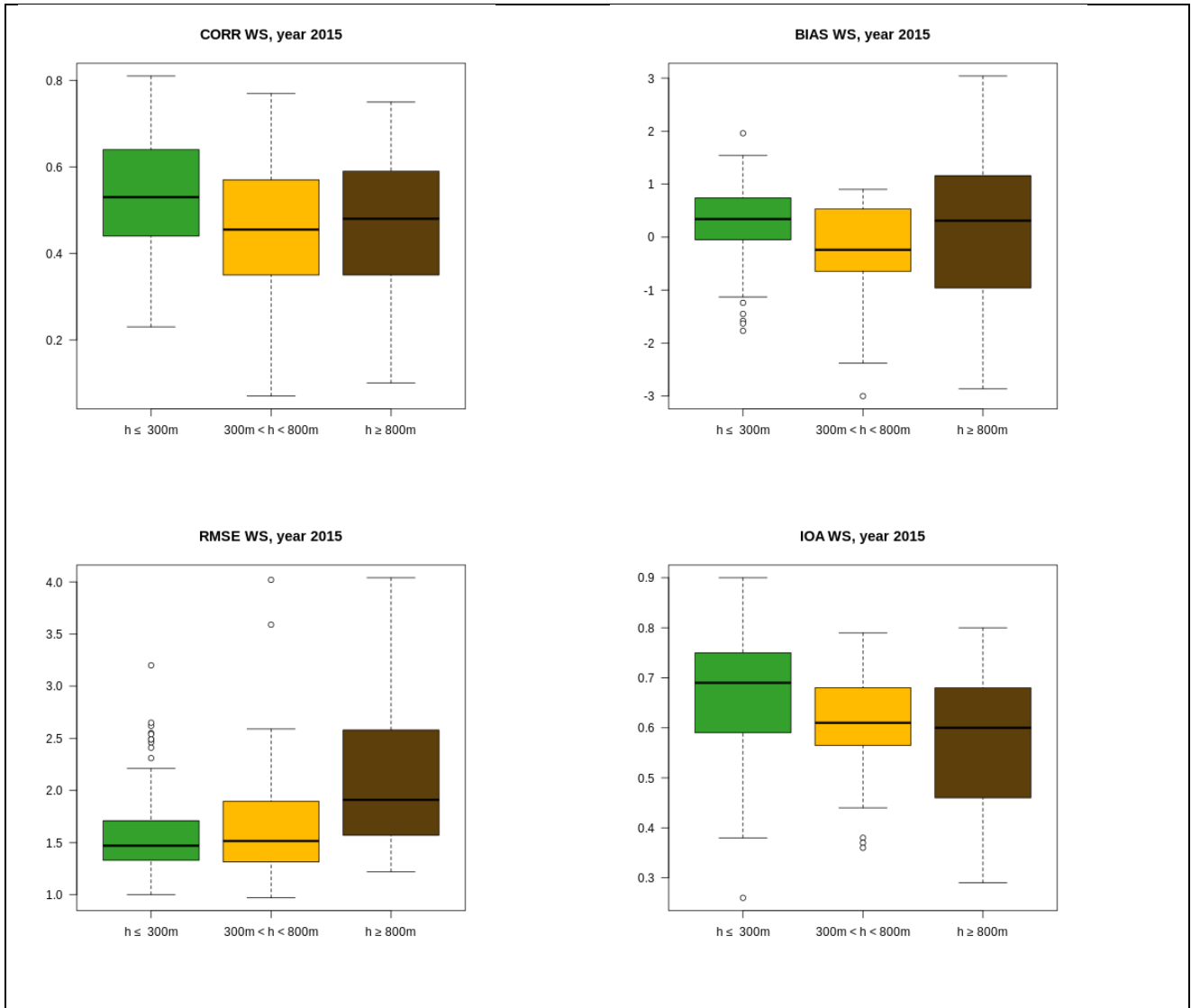


FIGURE A5. STATISTICAL SCORES COMPUTED FOR WS. FROM TOP LEFT CORNER, CLOCKWISE: *CORR*, *BIAS*, *IOA* AND *RMSE*. STATIONS ARE GROUPED FOLLOWING ALTITUDE ABOVE MEAN SEA LEVEL: 300m OR BELOW (GREEN), BETWEEN 300m AND 800m (ORANGE), 800m OR ABOVE (BROWN).

Statistics for wind speed were computed having available 149 sites below 300m, 41 at mid-level and 44 over 800m. Model results are in agreement with the benchmarks suggested in Emery et al. (2001) and Tesche et al. (2002). In Figure A5, *CORR* is around 0.5 for median (top, left), with the distributions having a similar spread for the different altitude ranges; *BIAS* (top, right) has median values between -0.5m/s and 0.5m/s and data spread increases with altitude. In most cases *RMSE* (bottom, left) is below the value of 2m/s suggested in Emery et al. (2001) and Tesche et al. (2002) as for *IOA* (bottom, right) which is almost 0.7 at plain sites and around 0.6 elsewhere. Also in this case, mountain locations show the largest spread in the results.

Finally, specific skill scores for precipitation were computed and summarised in the performance diagram presented in Figure A6, considering four different accumulation thresholds: 1mm, 5mm, 10mm and 20mm, i.e. the statistics were calculated for daily accumulations above the chosen threshold. The diagram, described in detail in Roebber (2009), contains in a single graphical representation the Probability of Detection (*POD*), the Success Ratio (*SR*), the *bias* and the Critical Success Index (*CSI*; also known as the threat score).

Typically these skills measure, from different points of view, the ability of the model in reproducing an event. Actually the “event” consists in daily-accumulated precipitation above the threshold.

Contingency Table		Observed Event	
		yes	no
Modelled Event	yes	A	B
	no	C	D

TABLE A1. CONTINGENCY TABLE.

Given the chances described in Table A1 (the so called contingency table), *POD*, *SR*, *bias* and *CSI* are defined as follows:

$$POD = \frac{A}{A+C} \quad (A2)$$

$$SR = \frac{A}{A+B} \quad (A3)$$

$$bias = \frac{A+B}{A+C} \quad (A4)$$

$$CSI = \frac{A}{A+B+C} \quad (A5)$$

For good forecasts, *POD*, *SR*, *bias* and *CSI* approach unity, such that a perfect forecast lies in the upper right part of the performance diagram.

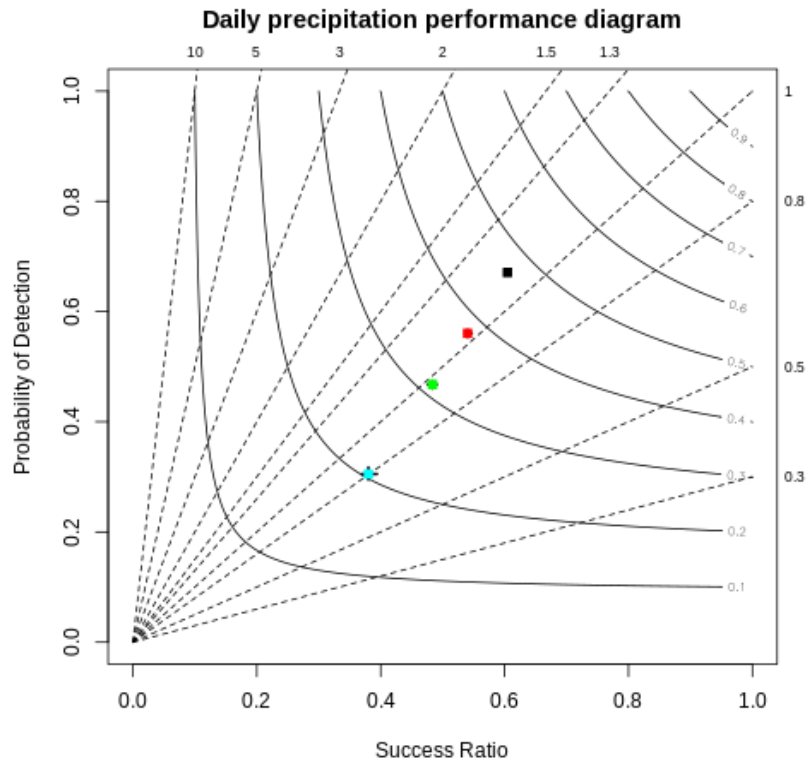


FIGURE A6. PERFORMANCE DIAGRAM SUMMARIZING *SR* (X AXIS), *POD* (Y AXIS), *bias* (DASHED LINES), AND *CSI* (SOLID CURVES). SCORES ARE COMPUTED OVER 564 RAIN GAUGE STATIONS FOR YEAR 2015 DAILY CUMULATED PRECIPITATION WITH DIFFERENT THRESHOLDS: 1mm/DAY (BLACK), 5mm/DAY (RED), 10mm/DAY (GREEN) AND 20mm/D (CYAN).

Simulated precipitation can be considered satisfactory, in line with RAMS performances found in literature: the results can be compared with Federico (2016) who performed RAMS simulations adopting different model configurations over Italy at 4 km resolution, even if for a period less than 2 months. Looking at Figure A6, *bias* values are found between 0.8 and 1 and *CSI* ones between 0.2 and 0.5; similar values as in Federico (2016) are obtained for *POD*, which decreases from 0.7 to 0.3 as threshold increases, while *SR* ranges between 0.6 (1mm threshold) and 0.4 (20mm threshold).

This model comparison was performed having the limitation that only a part of the Italian territory, mainly North (Po valley, Alps), was covered by available observations. However, it is worth note that this part of the country is widely more affected by air pollution than the rest of territory. Nevertheless, given the typical meteorological features occurring in the covered area (complex orography, weak flow with frequent stagnant conditions, coastal circulation) we consider that the results can be generalised at the rest of the Italian Peninsula.

References

Emery, C., Tai, E., Yarwood, G. 2001. Enhanced Meteorological Modeling and Performance Evaluation for Two Texas Ozone Episodes. Report to the Texas Natural Resources Conservation Commission, prepared by ENVIRON, International Corp, Novato, CA.

Federico, S., 2016. Implementation of the WSM5 and WSM6 Single Moment Microphysics Scheme into the RAMS Model: Verification for the HyMeX-SOP1. *Advances in Meteorology*, 2016, Article ID 5094126, <https://doi.org/10.1155/2016/5094126>.

Roebber, P.J., 2009. Visualizing multiple measures of forecast quality. *Weather and Forecasting*, 24, 601-608, <https://doi.org/10.1175/2008WAF2222159.1>.

Tesche, T.W., McNally, D.E., Tremback, C., 2002. Operational evaluation of the MM5 meteorological model over the continental United States: Protocol for Annual and Episodic Evaluation. Prepared for US EPA by Alpine Geophysics, LLC, Ft. Wright, KY, and ATMET, Inc., Boulder, CO. http://www.epa.gov/scram001/reports/tesche_2002_evaluation_protocol.pdf.

ENEA
Servizio Promozione e Comunicazione
www.enea.it

Stampa: Laboratorio Tecnografico ENEA - C.R. Frascati
gennaio 2020