

This is the peer reviewed version of the following article:

Towards the coupling of a chemical transport model with a micro-scale Lagrangian modelling system for evaluation of urban NO_x levels in a European hotspot / Veratti, Giorgio; Fabbi, Sara; Bigi, Alessandro; Lupascu, Aurelia; Tinarelli, Gianni; Teggi, Sergio; Brusasca, Giuseppe; Butler, Tim M.; Ghermandi, Grazia. - In: ATMOSPHERIC ENVIRONMENT. - ISSN 1352-2310. - 223:(2020), pp. 1-19. [10.1016/j.atmosenv.2020.117285]

Terms of use:

The terms and conditions for the reuse of this version of the manuscript are specified in the publishing policy. For all terms of use and more information see the publisher's website.

19/12/2025 19:28

(Article begins on next page)

1 Towards the coupling of a chemical transport model with a micro- 2 scale Lagrangian modelling system for evaluation of urban NO_x 3 levels in a European hotspot

4 Giorgio Veratti¹, Sara Fabbi¹, Alessandro Bigi¹, Aurelia Lupascu², Gianni Tinarelli³, Sergio Teggi¹,
5 Giuseppe Brusasca³, Tim M. Butler² and Grazia Ghermandi¹

6 ¹Department of Engineering “Enzo Ferrari”, University of Modena and Reggio Emilia, via Pietro Vivarelli 10, 41125
7 Modena, Italy

8 ²Institute for Advanced Sustainability Studies, Helmholtzstraße 5, 14467 Potsdam, Germany

9 ³ARIANET S.r.l., via Gilino 9, 20128 Milano, Italy

10 *Correspondence to:* Giorgio Veratti (giorgio.veratti@unimore.it)

11 **Abstract.** A multi-scale modelling system was developed to provide hourly NO_x concentration fields at a building-resolving
12 scale in the urban area of Modena, a city in the middle of the Po Valley (Italy), one of the most polluted areas in Europe. The
13 WRF-Chem model was applied over three nested domains and employed with the aim of reproducing local background
14 concentrations, taking into account meteorological and chemical transformation at the regional scale with nested resolutions
15 of 15 km, 3 km and 1 km. Conversely, the PMSS modelling system was applied to simulate 3D air pollutant dispersion, due
16 to traffic emissions, with a very high-resolution (4 m) on a 6 km x 6 km domain covering the city of Modena.
17 The methodology employed to account for anthropogenic emissions relies on two different strategies. Traffic emissions were
18 based on a bottom-up approach using emission factors suggested by the European Environmental Agency with traffic fluxes
19 estimated by the PTV VISUM model in the urban area of Modena, combined with direct traffic flow measurements
20 performed between October 28 and November 8, 2016 which was used for the hourly vehicle modulation. Other
21 anthropogenic emissions were taken from the TNO-MACC III inventory at the scales resolved by the WRF-Chem model.
22 Simulations were performed for the same period whereby the traffic measurement campaign was carried out.
23 2 m temperature and 10 m wind speed were captured quite well by the WRF-Chem model with statistical metrics in line with
24 similar case studies related to the Northern Italy. The NO_x concentrations reproduced in the Po Valley area by WRF-Chem
25 were on average simulated reasonably well with a general negative bias in almost all the examined rural background
26 monitoring stations. Additionally, the deployment of an emission inventory at the original resolution (7 km) highlighted that
27 increasing resolution from 3 km to 1 km does not generally improve the model performance.
28 Nevertheless, simulated and observed NO_x hourly concentrations in the urban area of Modena exhibit a large agreement in
29 particular for urban traffic site where detailed traffic emission estimations proved to be very successful in reproducing the
30 observed NO_x trend. At urban background stations, despite a general underestimation of the observed concentrations, the
31 combination of WRF-Chem with PMSS provided daily pattern in line with observations. The analysis of the modelled NO_x
32 daily cycle pointed out also that at both traffic and background urban stations the morning NO_x peak concentration was on

33 average underestimated. This could be explained with an overestimation of mixing phenomena between 07:30 a.m. and
34 10:00 a.m. by WRF-Chem which leads to a greater dispersion of NO_x along the vertical and thus a morning underestimation.
35 The statistical analysis showed finally that PMSS combined with WRF-Chem at both the resolutions (3 km and 1 km) and at
36 both traffic and background sites fulfilled standard acceptance criteria for urban dispersion model evaluation, confirming that
37 the proposed multi-modelling system can be employed as a tool to support environmental policies, epidemiological studies
38 and urban mobility planning.

39 **Keywords:** WRF-Chem, PMSS modelling system, Multi-model approach, Traffic emissions, Modena.

40 **1 Introduction**

41 Atmospheric pollution is one of the main risk factors for a number of pollution-related diseases and health conditions: they
42 may occur through the appearance of harmful and carcinogenic effects on the respiratory system as well as the onset of other
43 cardiovascular, nervous and ocular pathologies (Loomis et al., 2013, Novaes et al., 2010, PopeIII et al., 2003). These critical
44 issues particularly affect urban areas with a higher population density: a complex mixture of pollutants is produced by
45 inefficient combustion of fuels in internal combustion engines, power generation and other human activities like domestic
46 heating and cooking.

47 One of the most critical air pollutants in terms of health effects is nitrogen dioxide (NO_2), whose levels in the last years
48 exceeded national and WHO standards in many urban areas across Europe (European Environment Agency, 2016). In Italy,
49 and more particularly in the Po Valley, despite an overall decrease in PM in the last 10 years (Bigi and Ghermandi, 2016),
50 the urban population is still exposed to harmful levels of NO_2 .

51 The present research has as its goal the estimation of the air quality in the urban area of Modena, a city in the central Po
52 Valley, in terms of NO_x atmospheric concentrations. More in detail, the aim of the project is to support environmental
53 policies, epidemiological studies and urban planning and management.

54 Current approaches to produce spatial maps of urban air pollution include the use of interpolation methods and land-use
55 regression (LUR) models. However, all these techniques need a large number of in-situ observations at strategic locations to
56 represent the full spatial and temporal pollutant variability and cannot be used to take into account turbulent atmospheric
57 dispersion. To meet this need, a variety of micro (Moussafir et al, 2004, Öttl, 2015) and local (Tinarelli et al, 1992, Bellasio
58 and Bianconi, 2012, Cimorelli et al, 2004) scale air dispersion models have been developed in the last few years, as they can
59 provide a high-resolution information on air pollution level within urban city area by taking into account space-time
60 emissions distribution and local meteorological characteristics (Ghermandi et al, 2014, Ghermandi et al, 2015).

61 A key issue is the quantitative estimation of the different contribution to air pollution level from emissions sources located
62 within the city urban environment and from countryside areas, also known as rural background. An approach that has been
63 used for several years to account for both urban and rural contribution is generally called “Lenschow” approach (Lenschow
64 et al., 2001), which envisages the influence of a city as the difference between the concentrations in the urban environment

65 and the concentrations at rural site. This methodology is generally applied in source apportionment studies in order to
66 estimate the primary and secondary component of PM by means of receptors models (Pirovano et al., 2015, Bove et al.,
67 2014), or in urban impact assessments through an approach combining measurements and modelling results. In this latter
68 case, the model is used to evaluate the contribution (effects) of the sources located within the city, while measured
69 background concentrations are added to the simulated concentrations to account for remote sources outside the simulation
70 domain (Ghermandi et al. 2019, Berchet et al., 2017).

71 Despite the flexibility in enforcing measured-based activity and modelling results, a number of criticisms related to the
72 applicability of this methodology were recently highlighted. Following Thunis (2018) the city urban impact can be defined
73 as the sum of three components: the Lenschow urban increment, which is the concentration difference between the city and
74 background locations, the “city spread” meaning the impact of the city at the rural background location, and the “background
75 deviation” that quantifies the concentration difference between city and background location when city emissions are set
76 equal to zero. According to this definition, the “Lenschow” approach can be correctly employed only when the “city spread”
77 and the “background deviation” are negligible or compensate each other, i.e. the urban impact in background area is close to
78 zero and when background levels are spatially homogenous.

79 An alternative approach to quantify the different contributions to air pollution in the city is based on Chemical Transport
80 Models (CTMs) which, unlike the Lenschow incremental approach, are able to generate different emission scenarios on
81 multi spatial scale, from regional to local, which can be exploited to estimate background concentrations keeping city
82 emissions set to zero.

83 Based on the advantages given by CTMs, the methodology employed in this study was a modelling activity relied on the
84 NO_x dispersion by combining two different models: the Weather Research and Forecasting (WRF) model coupled with
85 Chemistry (Grell et al., 2005), which is able to compute concentrations fields over regional domain by considering specific
86 emission scenarios, and the Parallel Micro SWIFT and SPRAY (PMSS, Moussafir et al., 2013, Oldrini et al., 2017)
87 modelling suite accounting for dispersion phenomena within the urban area.

88 In this project, the PMSS modelling suite was used to simulate at building-scale resolution the NO_x dispersion produced by
89 urban traffic flows in the city of Modena. Conversely, the WRF-Chem model simulations were performed to estimate the
90 NO_x background concentrations on multiple domains with a nesting technique, in order to take into account emissions both
91 at regional and local scale by excluding traffic emissions sources over the city of Modena. A similar approach was used also
92 by Tewari et al. (2010), Wyszogrodzki et al. (2012) and Kwak et al. (2015), which developed an integrated urban air quality
93 modelling system by coupling a CFD model with a chemical transport model in order to account not only the dispersion in
94 built-up areas but also to consider larger scale influences.

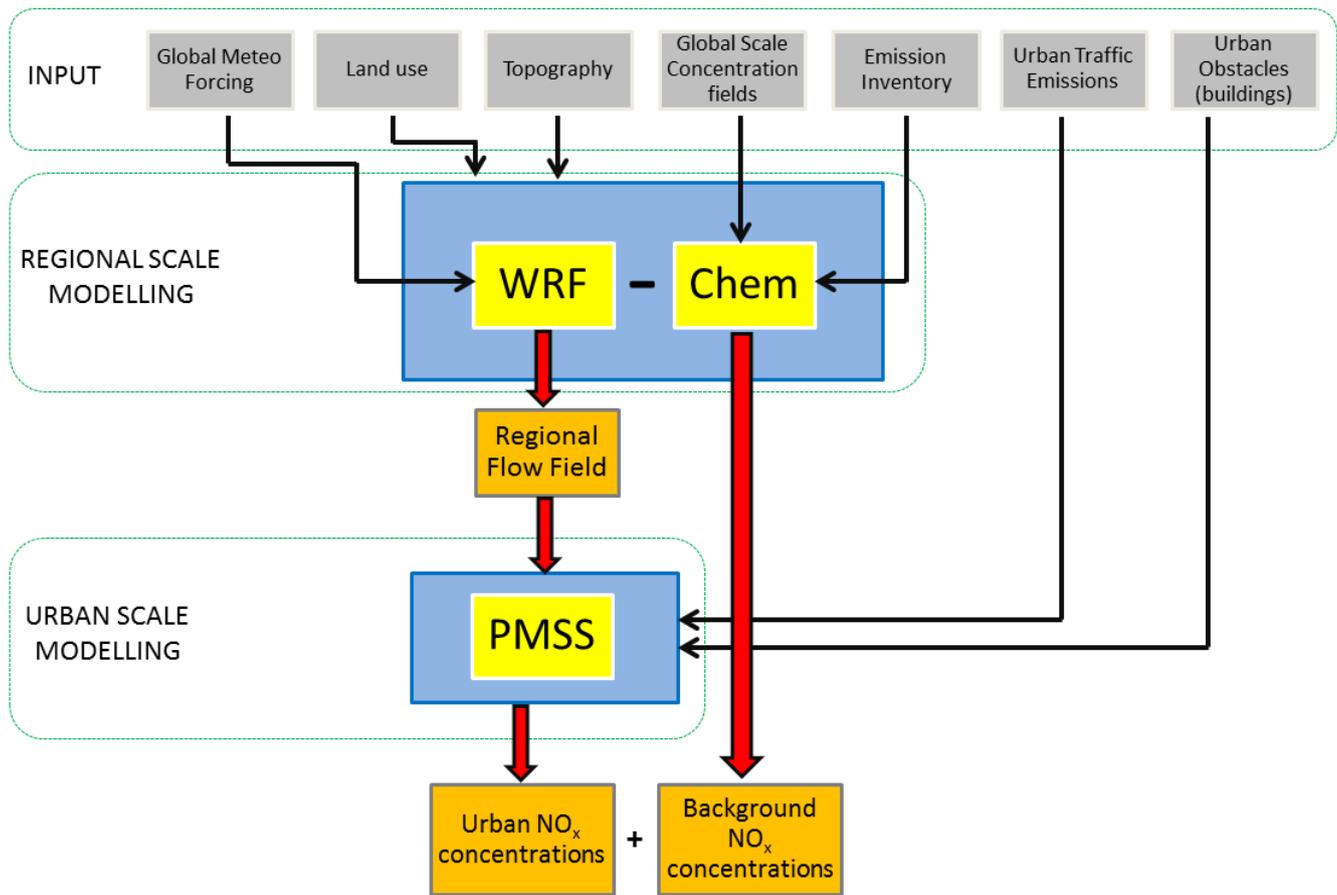
95 In the first part of the paper the modelling system which is applied to take into account both urban and background emissions
96 sources is described. The following section describes the set-up of the models employed in the simulation, along with the
97 method for estimation of emissions. Finally, the performance of the models in representing the meteorological mesoscale
98 reconstruction and the pollutant dispersion is evaluated.

99 2 Materials and method

100 2.1 Modelling chain description

101 With the aim of providing hourly NO_x concentration maps over the urban area of Modena, a hybrid modelling system
102 composed by WRF-Chem, an Eulerian model and PMSS, a Lagrangian particle dispersion model, was employed in this
103 study. The choice of this modelling chain was based on the WRF-Chem ability to simulate the emissions, transport and
104 chemical transformations simultaneously with meteorology at large scale, and on the PMSS capability to provide high
105 resolution air quality maps over an entire urban domain characterized by large spatial and temporal concentrations gradient,
106 with a reasonable computation time (Ghermandi et al., 2015).

107 Figure 1 illustrates the interplay between WRF-Chem and PMSS. The WRF-Chem model was used to estimate NO_x
108 concentrations on multiple domains at different grid resolutions spanning from the European domain to the Po Valley area
109 with a nesting technique, necessary to take into account emissions at regional scale that can affect urban air quality in
110 Modena. Then, wind streams within the city were determined by a cascade of scales from global to buildings level: synoptic
111 and local scale meteorological conditions in the region surrounding the city of Modena were simulated by WRF-Chem
112 taking into account the local topography and land-use data. Driven by these mesoscale flow patterns, high resolution winds
113 were computed in the city to account for buildings and street canyons by performing the diagnostic mass-consistent Parallel-
114 Micro-SWIFT model. Secondly, Lagrangian dispersion simulations driven by high resolution winds were carried out with
115 Parallel-Micro-SPRAY by estimating urban traffic emission flows. As a final step of the procedure, NO_x traffic urban
116 concentrations, simulated with Parallel-Micro-SPRAY, were added to NO_x background concentrations estimated with the
117 WRF-Chem model. Further explanations regarding the methodology used to avoid double counting of traffic emissions
118 within Modena urban domain are reported in section 2.5.



119

120 **Figure 1:** Outline of the multi-model approach implemented to generate hourly NO_x concentrations fields.

121

122 2.2 WRF-Chem Set-up

123 For this application, the Weather Research and Forecasting model with chemistry (WRF-Chem), version 3.9.1, was applied
 124 over three one-way nested domains, centred in the urban area of Modena. The outer domain (d01) covers most of Europe
 125 with 150 x 150 grid cells at 15 km horizontal resolution, the intermediate domain (d02) covers the North of Italy with a
 126 resolution of 3 km (150 x 150 grid cells) and the innermost domain (d03) focuses on the Po Valley area with a spatial
 127 resolution of 1 km with 175 x 175 grid cells (Figure 2). The model was configured with 35 vertical levels with the first layer
 128 approximately at 30 m and the model top set at 50 hPa.

129

149 **Table 1:** WRF-Chem model parameterisation.

Process	WRF-Chem option
Land-surface model	Noah Land Surface Model
Boundary layer scheme	YSU
Cumulus parameterization	Grell-Freitas (only for the outer domain)
Microphysics	Lin
Short-wave radiation	RRTM with MCICA method
Long-wave radiation	RRTM with MCICA method
Gas-phase mechanism	MOZART
Aerosol model	MOSAIC 4 bins

150

151 The MOZART gas-phase chemical mechanism developed by Emmons et al. (2010), and the MOSAIC aerosol model (Zaveri
 152 et al. 2008) were used to simulate airborne pollutants over the nested domains. The first one includes 85 chemical species,
 153 196 reactions and is consistent with the chemistry used in the global model that provides the chemical input and boundary
 154 conditions for the nested simulations. MOSAIC uses a sectional bin approach for the representation of the aerosol size
 155 distribution. The MOSAIC model predicts several aerosol species, such as sulfate, nitrate, ammonium, elemental carbon, and
 156 primary aerosols (POAs). Processes involving secondary organic aerosols (SOAs) formation were represented by the scheme
 157 based on Hodzic and Jimenez (2011).

158 Meteorological initial and boundary conditions were provided by the 6-hourly ECMWF analysis field (ERA5 dataset) with a
 159 horizontal resolution of $0.25^\circ \times 0.25^\circ$, interpolated to 37 pressure levels from 1000 to 1 hPa. Data included 3D fields of
 160 temperature, specific humidity and wind speed components. 2D surface parameters such as mean sea level pressure, sea
 161 surface temperature, soil temperature and volumetric soil water content were also considered. A grid nudging on temperature
 162 and wind field has been also performed within the boundary layer in all three model configurations using as input data the
 163 ECMWF analysis.

164 As to land use, the Corine Land Cover (CLC) dataset was adopted after reclassifying it into the 33 USGS classes to match
 165 the WRF land use tables. Chemical initial and boundary conditions were provided by the global Model for Ozone And
 166 Related chemical Tracers output (MOZART-4; Emmons et al., 2010). Biogenic emissions were calculated online by the
 167 Model of Emissions of Gases and Aerosols from Nature version 2.1 (MEGAN2.1) by Guenther et al. (2012). In addition, sea
 168 salt and dust emissions are calculated online.

169 **2.3 PMSS modelling system description**

170 Parallel Micro SWIFT SPRAY or PMSS (Oldrini et al. 2011, 2017) is the parallelized version of the MSS modelling suite
171 (Tinarelli et al. 2007, 2013) constituted by the individual models SWIFT and SPRAY, both used in small scale urban model
172 (a.k.a. Micro-SWIFT and Micro-SPRAY).

173 Micro-SWIFT is a 3D mass-consistent diagnostic model that uses terrain-following coordinates to provide diagnostic wind,
174 turbulence, temperature and humidity fields using data from a dispersed meteorological network.

175 The first step performed by Micro-SWIFT is the interpolation of the heterogeneous meteorological input data, such as
176 surface and vertical measurements profile or regional meteorological model output, to reconstruct the 3D wind field. Then,
177 the first computed wind field is modified in the zones around isolated or group of buildings following the approach
178 suggested by Röckle (1990) and Kaplan and Dinar (1996) adopting a parametrization of the recirculating flow regions
179 around, behind, over and between obstacles. Subsequently, the mass conservation constraint is imposed through the
180 impermeability conditions on the ground and at building surfaces. Finally, a RANS flow solver can be optionally used to
181 simulate more accurate velocity and pressure fields in built-up environments than obtained with the pure diagnostic flow
182 model configuration. With the RANS approach, the momentum equation is introduced in the computation and the turbulent
183 Reynolds stress tensor is modelled by a zero-order closure based on mixing-length theory and the momentum and pressure
184 equations are solved using the fractional time step technique (Gowardhan et al., 2011).

185 The estimation of the turbulence needed by Micro-SPRAY to drive pollutants dispersion is diagnosed by Micro-SWIFT
186 through the superimposition of the background turbulence, obtained by standard boundary layer parameterizations (Hanna et
187 al., 1982) and the turbulence inside the flow zones modified by the obstacles.

188 Micro-SPRAY is a 3D Lagrangian Particle Dispersion Model (LPDM) (Rodean, 1996) able to simulate the advection and the
189 diffusion of gaseous species or fine aerosol by accounting for the presence of obstacles. The dispersion of an airborne
190 contaminant is modelled by virtual particles that follow the turbulent motion of the air as passive tracers and their spatial
191 distribution at a certain time represents the concentration of an emitted substance.

192 The trajectories of the particles emitted by a source are obtained by integrating in time their velocity. This can be considered
193 as the sum of a transport component, defined by the local averaged wind, usually provided by Micro-SWIFT, and a
194 stochastic component, standing for the dispersion due to the atmospheric turbulence. The stochastic component is obtained
195 by solving a 3D form of the Langevin equation for the random velocity, following Thomson's approach (Thomson, 1987).

196 The PMSS modelling system has been validated (Trini Castelli et al. 2017 and 2018, Oldrini et al., 2017 and 2019) and
197 applied (Carlino et al., 2016, Moussafir et al., 2013) to several experiments and real cases. In this study, the performances of
198 the PMSS modelling system are exploited to estimate urban air quality in the city of Modena in a real case scenario.

199 **2.4 PMSS set-up**

200 A 3D wind and turbulence field and air pollution dispersion reconstruction was performed on a 6 km x 6 km square domain
 201 covering the city of Modena (Figure 2) with the PMSS modelling suite. Given the low altitude difference between different
 202 areas of the city, a flat domain was considered and a 3D reconstruction of buildings was made by using a pre-processor:
 203 25,600 polygons contained in the ESRI shapefile (provided by Geoportale Regione Emilia-Romagna) were transformed into
 204 approximately 146,000 triangular prisms directly usable by Micro-SWIFT.

205 In order to guarantee both flow and pollutant dispersion fields at a high resolution in each part of the domain, a horizontal
 206 grid step of 4 m (square cells) was chosen for both Micro-SWIFT and Micro-SPRAY models. To represent the flow entering
 207 the Micro-SWIFT computational domain, vertical profiles of temperature, humidity, wind speed and direction from the
 208 innermost domain (d03) of the WRF-Chem simulation were extracted on an hourly basis. In addition, mixing height values
 209 and main background turbulence parameters (i.e. friction velocity, Monin-Obukhov length and convective scale velocity)
 210 were estimated with the Mesoscale Model Interface Program v3.4 (MMIF, [https://www.epa.gov/scram/air-quality-](https://www.epa.gov/scram/air-quality-dispersion-modeling-related-model-support-programs)
 211 [dispersion-modeling-related-model-support-programs](https://www.epa.gov/scram/air-quality-dispersion-modeling-related-model-support-programs)), which converts prognostic meteorological WRF output fields into
 212 turbulence scale parameters.

213 3D fields of wind, temperature and turbulence were obtained for 20 vertical levels from 3 m up to 200 m above the ground
 214 using the Micro-SWIFT model with the RANS flow solver option activated. The 2D Cressman interpolation method wind
 215 field was also considered in the configuration.

216 Regarding the Micro-SPRAY model simulations the horizontal grid was chosen to be identical to that of the Micro-SWIFT
 217 model computation and the vertical grid structure consisted of 10 levels with a linear progression up to 200 m above the
 218 ground level with 3 m height for the first layer close to the soil. This arrangement leads to a configuration of 1504 x 1504 x
 219 10 nodes and a total number of $2.26 \cdot 10^7$ cells. The main SPRAY parameters also included an emissions time step of 5
 220 seconds and a synchronisation time step of 10 seconds. Concentrations are computed every hour and sampled every 10
 221 seconds. Since the cumulative precipitation for all the examined period was less than 10 mm, the wet deposition was not
 222 included in the set-up. The main PMSS set-up parameters are reported in Table 2.

223

224 **Table 2:** Micro-SWIFT and Micro-SPRAY set-up.

Micro-SWIFT		Micro-SPRAY	
Parameters	Value	Parameters	Value
Horizontal resolution	4 m	Horizontal resolution	4 m
Horizontal grid	1504 x 1504 points	Horizontal grid	1504 x 1504 points
Vertical grid	from 3 up to 200m 20 vertical levels	Vertical grid	from 3 up to 200m 10 vertical levels

Interpolation method	Cressman 2D	Emission time step	5 s
		Averaging period for concentrations	3600 s

225

226

227 **2.5 Anthropogenic Emissions**

228 Following the regional emissions inventory database produced by Arpae Emilia-Romagna, the local environmental agency
 229 (INEMAR 2013), the road traffic in Modena contributes up to the 60% of the total emissions in terms of NO_x, while the
 230 domestic heating and industrial combustion represent only the 15% and 14% of the total amount. Based on this percentage
 231 distribution, the methodology employed to account for anthropogenic emissions rely on two different strategies: a city-
 232 tailored emission estimate, to describe traffic emissions at micro-scale resolution in the urban area of Modena, and an
 233 emission inventory estimate, more suitable to account for emissions at large-scale area, used as an input for the chemical
 234 transport model in order to estimate the contribution of all the SNAP (Selected Nomenclature for Air Pollution) emission
 235 categories throughout Europe.

236 The anthropogenic emissions used for the parent and the nested WRF-Chem domains were taken from the TNO-MACC III
 237 inventory, available on a regular grid with a horizontal resolution of 0.125° x 0.0625°, which contains emissions for air
 238 pollutants such as NO_x, SO₂, NMVOC, NH₃, CO and primary particulate matter (PM_{2.5} and PM₁₀). The inventory is based on
 239 nationally reported emissions for specific sectors and spatially distributed with proxy data such as the population density for
 240 urban emissions or the road network for non-urban emissions. The main developments with respect the version II of the
 241 inventory (Kuenen et al. 2014) includes improved emissions and trends for the international sea shipping, improved wood
 242 consumption estimation and more detailed spatial distribution, as well as improved industrial emissions apportioning
 243 achieved through the use of CORINE land cover data instead of population density data as a default item.

244 TNO-MACC III emissions are provided as annual totals, therefore each SNAP category was scaled to take into account
 245 monthly, daily variation (weekend or weekday) and hour of the day (diurnal cycle), as suggested by Kuik et al. (2018). A
 246 vertical emissions distribution was also taken into account by distributing the emissions of industrial sources, airports,
 247 extraction and distribution of fossil fuel into seven vertical layers, up to 750 m.

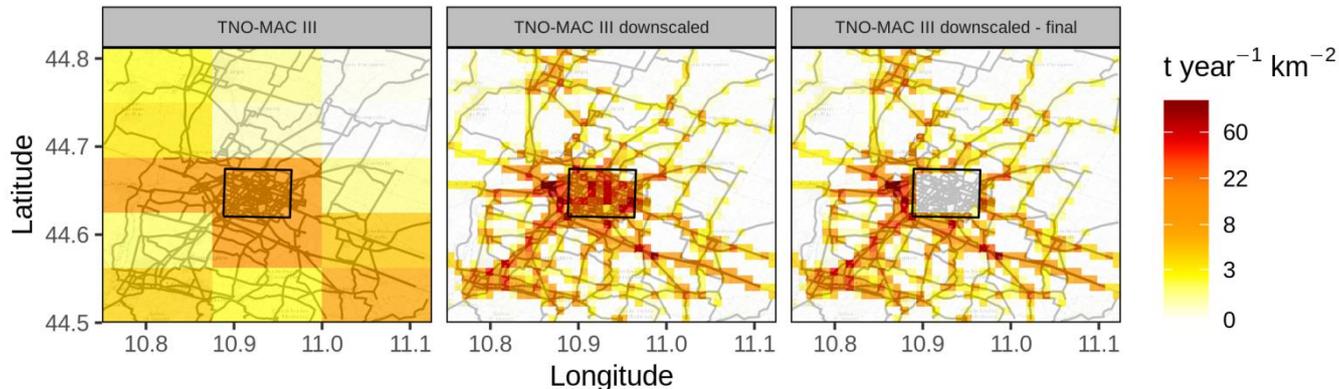
248 In order to avoid the double counting of the traffic emissions placed inside the urban area of Modena and to better represent
 249 the spatial distribution of traffic sources in the nearby territory, a downscaling procedure was conducted for the SNAP
 250 sectors 71-75. The original TNO dataset (resolution ca. 14 km x 7 km) covering the inner most WRF-Chem domain was
 251 subdivided to a finer grid with a horizontal resolution of 1 km x 1 km.

252 Traffic fluxes of light (passenger cars and L-category vehicles) and duty (light commercial vehicle and heavy-duty trucks)
 253 vehicles on the main roads of the province of Modena at morning rush hour (07:30 – 08:30, local time) for the year 2010
 254 were provided by the Municipality of Modena and proceed from a simulation study by means of the PTV VISUM model

255 (PTV Group, Karlsruhe, Germany <http://vision-traffic.ptvgroup.com/en-us/products/ptv-visum/>). These data were used as a
256 proxy variable to assign TNO-MAC III traffic emissions over the province of Modena to the portion of the land interested by
257 PTV VISUM road network: the more traffic fluxes were estimated for a specific road segment, the more emissions were
258 assigned to the corresponding grid (1 km x 1 km) (Figure 3).

259 Once the downscaled grid dataset was created, a spatial surrogate function was implemented to identify the TNO-MACC III
260 traffic emissions within the PMSS domain. This function returns zero if the territory cell is completely inside the PMSS
261 domain, one if the territory cell is completely outside and a value between zero and one (proportional to the area outside the
262 PMSS domain) if the territory cell crosses the domain boundaries. Finally, to exclude TNO-MACC III emissions from the
263 PMSS domain, the spatial mask created with the surrogate function was multiplied by the downscaled traffic emission
264 inventory. The same traffic simulation obtained with the PTV VISUM model includes also vehicle fluxes in the urban area of
265 Modena, providing the number and the average speed for light and duty vehicles at rush hour for each segment of the urban
266 road network, which encompasses about 1100 sections with a total length of 210 km (Figure 4).

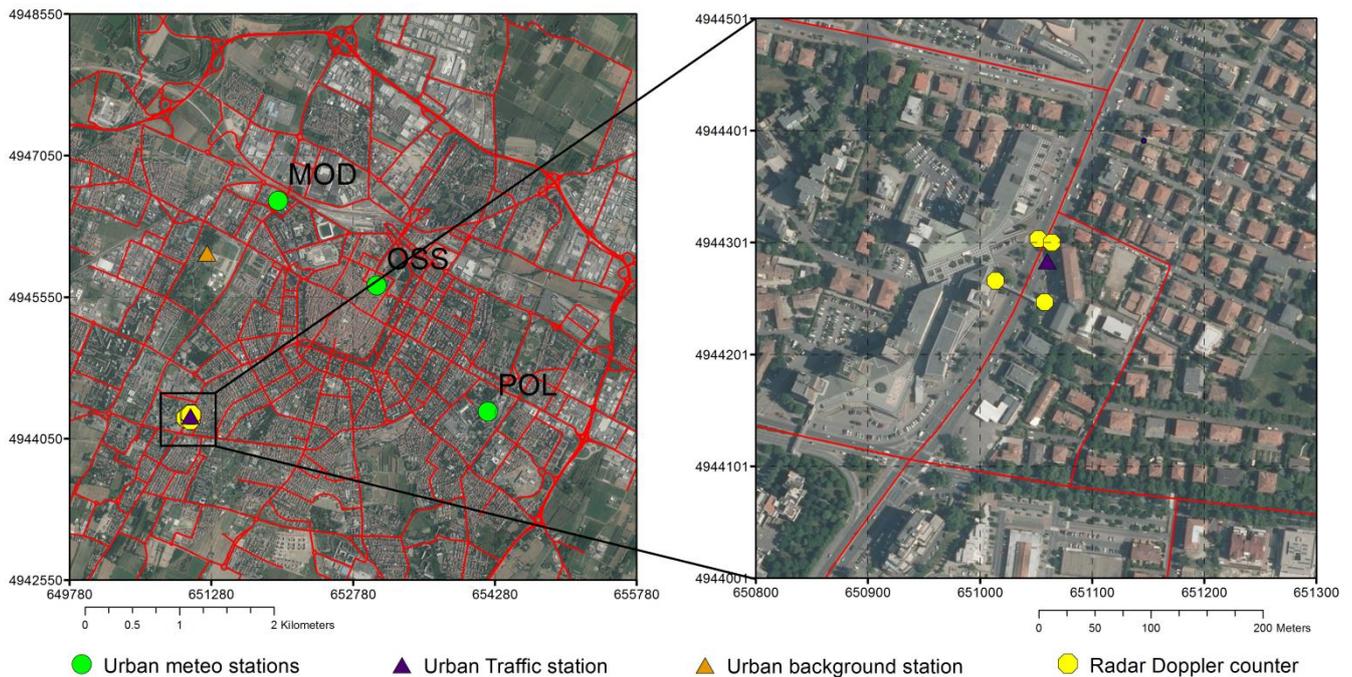
NO_x traffic emissions SNAP 71-75



267
268 **Figure 3:** Total NO_x traffic emissions (SNAP sector 71-75) in the province of Modena. On the left the original TNO-MACC III inventory
269 emissions (resolution ca. 14 km x 7 km). At the centre the TNO-MACC III inventory emissions downscaled to a resolution of 1 km x 1 km
270 and distributed according to the street traffic flow estimated by PTV VISUM. On the right the TNO-MACC III inventory emissions
271 downscaled to a resolution of 1 km x 1 km without the emissions by road traffic in the urban area of Modena.

272 The methodology chosen to estimate traffic emissions in the urban area of Modena was based on a bottom-up approach:
273 traffic flow data simulated by PTV VISUM model were employed as “activity factor” and specific Emission Factors (EF)
274 were used to estimate total emissions according to the fleet composition, vehicle type, fuel, engine capacity, load
275 displacement, slope of the road, Euro emission standard and average traveling speed.

276



296 (Ntziachristos and Samaras, 2016). Actual average annual mileage (AAAM) for gasoline and diesel cars in the Italian
 297 vehicle stock, respectively $AAAM_{gasol}$ and $AAAM_{diesel}$, were estimated with the formulas suggested by Caserini et al. (2013).
 298 Considering *age* as the number of years between the current year and the year of vehicle purchase, the formulas can be
 299 written as follow:

300

$$301 \quad AAAM_{gasol} = 20.817 \cdot age^2 - 1124.3 \cdot age + 15651 \quad \text{if } age \leq 30 \text{ years} \quad (1)$$

$$302 \quad AAAM_{gasol} = 20.817 \cdot 30^2 - 1124.3 \cdot 30 + 15651 \quad \text{if } age > 30 \text{ years} \quad (2)$$

$$303 \quad AAAM_{diesel} = 35.072 \cdot age^2 - 1811.2 \cdot age + 23942 \quad \text{if } age \leq 30 \text{ years} \quad (3)$$

$$304 \quad AAAM_{diesel} = 35.072 \cdot 30^2 - 1811.2 \cdot 30 + 23942 \quad \text{if } age > 30 \text{ years} \quad (4)$$

305

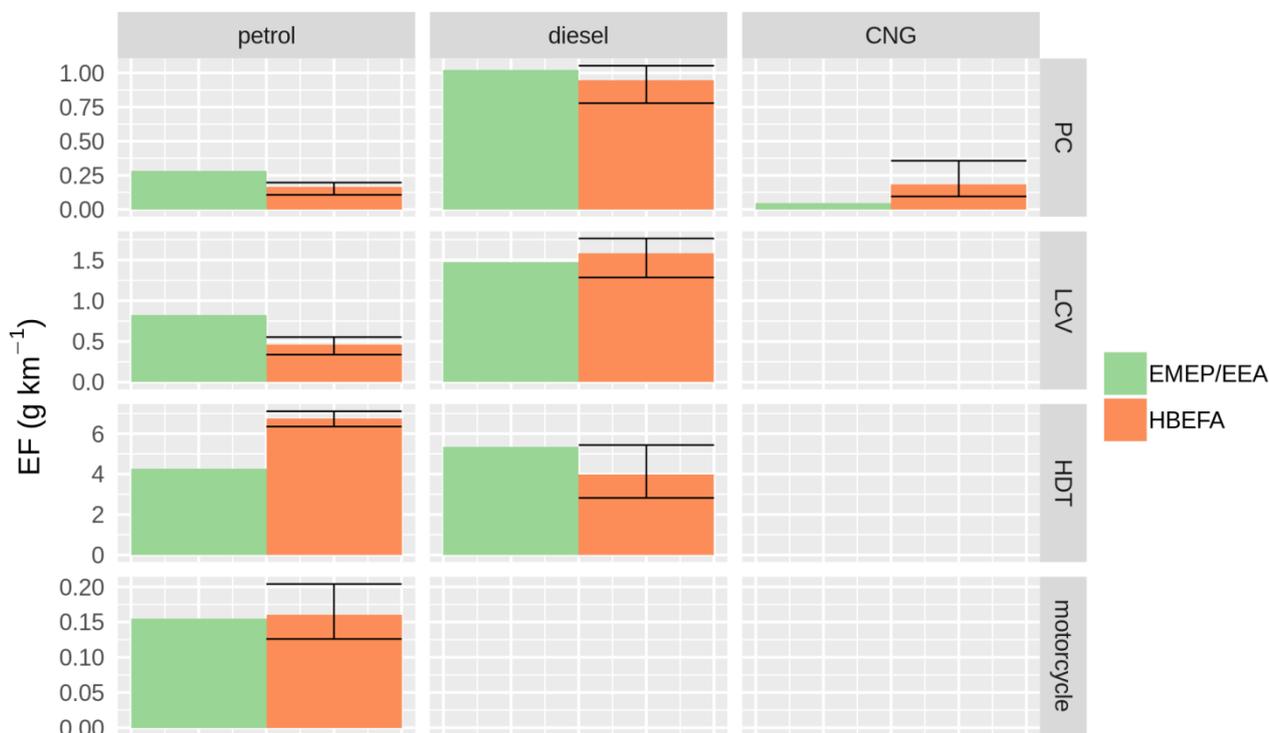
306 In addition to the traffic model data, a direct vehicle flow measurement campaign was carried out continuously over two
 307 weeks between October 28 and November 8, 2016, with 4 Doppler radar counters (one for each road lane) in a four-lane
 308 road in the proximity of the intersection with the urban ring road (Ghermandi et al. 2019) (Figure 4). The radar traffic
 309 counters recorded the time, the length and the velocity for each passing vehicle. The captured vehicles were subdivided into
 310 two different groups according to the vehicle classes modelled by PTV VISUM: light vehicles, with measured length less or
 311 equal to 6 m and duty vehicles, with measured length greater than 6 m. Finally, to appropriately describe NO_x emissions
 312 under typical vehicle flux conditions, recorded flow data were used to reproduce hourly modulation rates for the entire road
 313 network.

314

315 In order to test the reliability of the hot EFs computed with the methodology previously described and to check how far these
 316 EFs are with respect the one computed considering recently emission data, based on new PEMS tests (Hausberger et al,
 317 2019, Sjödin et al., 2018), the weighted EFs for Passenger Cars (PC), Light Commercial Vehicle (LCV), Heavy Duty Trucks
 318 (HDT) and Motorcycle in Modena were compared with the weighted average hot EFs calculated following the handbook of
 319 emission factors (HBEFA, 2019) for different European countries (Germany, Austria, Switzerland, France, Norway and
 320 Sweden).

321 Since the HBEFA desktop application is not available for free, an extensive analysis relied on the real vehicle fleet
 322 composition in Modena was not possible. Despite this limitation, an indicative comparison between the actual hot EFs
 323 considered in the emission computation for the city of Modena and the average hot EFs for the respective vehicles category
 324 between the 6 European countries mentioned before is shown in Figure 5. The green rectangles indicate the EFs used in
 325 study, while the red rectangles represent the average EFs computed between Germany, Austria, Switzerland, France, Norway
 326 and Sweden, when available. The black horizontal segments below and above the red rectangles upper limit indicate
 327 respectively the minimum and the maximum EF for each respective vehicle category.

328



329

330 **Figure 5:** Comparison between weighted NO_x hot EFs used in this study, computed following the EMEP/EEA guidelines (green
 331 rectangles) and the corresponding average NO_x hot EFs computed between Germany, Austria, Switzerland, France, Norway and Sweden
 332 following the handbook of emission factors (HBEFA) version 4.1 (red rectangles). With horizontal black segment are also
 333 indicated the minimum and maximum HBEFA EF for the same six European countries. Please note the difference in the scale on the y-
 334 axis in each panel.

335

336 The hot EFs obtained following the EMEP/EEA guidelines for the city of Modena are in general in line with the hot EFs
 337 computed using the HBEFA methodology (v4.1) for similar European countries. The main differences in absolute terms
 338 regard the Heavy Duty Truck category, where the HDT EFs estimated in Modena are 4.28 and 5.34 g km⁻¹ for petrol and
 339 diesel respectively, while the average hot EFs for the same vehicles category are 6.75 and 3.97 g km⁻¹. The variability
 340 between diesel HDT EF for the six European countries considered is also large and the maximum of these EFs is greater than
 341 the EF considered in Modena. On the other hand, diesel LDV EFs are very similar to each other and equal to 1.47 g km⁻¹ for
 342 Modena and 1.58 g km⁻¹ for the average EF between the six countries. By contrast, petrol LDV EFs differ of about 0.37 g
 343 km⁻¹ (0.83 g km⁻¹ for Modena and 0.46 g km⁻¹ for the average of the other countries). Finally, Motorcycle and Passenger Cars
 344 EFs estimated in Modena agree very well with the average EFs estimated following the HBEFA methodology: 0.28 (petrol),
 345 1.02 (diesel) and 0.05 (CNG) g km⁻¹ are the PC EFs estimated in Modena, while the HBEFA respective average is 0.16, 0.94
 346 and 0.18 g km⁻¹. Motorcycle EFs are almost the same, 0.15 g km⁻¹ for Modena and 0.16 g km⁻¹ for the HBEFA average.

347 3 Model Simulations

348 Following the setup described in the previous section, simulations were performed for the period between 28 October 2016
349 and 8 November 2016, the same period whereby the traffic measurement campaign was carried out. These days were
350 characterized by weather condition typical for autumn in the central Po Valley (Bigi et al., 2012, Thunis et al., 2009) with a
351 very little atmospheric circulation due to recurrent thermal inversions at low altitude, low mixing layer heights and persistent
352 foggy and hazy events which lasted also during day time. Recurrent wind calm episodes and high-pressure conditions
353 enhance persistence and homogenization of air masses on a regional scale: the characteristic climate conditions, along with
354 the strong anthropic pressure in the area, lead to long-lasting high concentrations of pollutants also at remote rural sites (Bigi
355 et al., 2017, Masiol et al., 2015, Tositti et al., 2014).

356 Low rainfall rate (cumulative precipitation lower than 10mm), mean wind speed lower than 2 m s⁻¹ and daily average
357 temperature from 7.4 to 13.6 °C characterized the meteorological condition in Modena during the investigated period.

358 WRF-Chem was performed using a 2 days spin-up period to ensure consistency with meteorological and chemical fields, as
359 well as PMSS was executed using 6 hours of spin-up at the beginning of the simulation.

360 4 Model Evaluation

361 The statistical performance analysis considered multiple statistical indicators regardless of the model's application since each
362 one has its advantages and disadvantages and it is not possible to identify a unique exhaustive index of quality.

363 The main statistical metrics employed in this study are Pearson correlation coefficient (r), Mean Bias (MB), Normalized
364 Mean Bias (NMB), Root Mean Square Error (RMSE), Mean Absolute Error (MAE) and the fraction of predicted values
365 within a factor of two of observations, also referred as Factor of two (FAC2). These statistical indicators are defined as

366 follows, with n the number of model–observation pairs, M the modelled values (with $\bar{M} = \frac{\sum_{i=1}^n M_i}{n}$ the averaged modelled
367 value) and O the observations (with $\bar{O} = \frac{\sum_{i=1}^n O_i}{n}$ the averaged observed value):

$$r = \frac{\sum_{i=1}^n (M_i - \bar{M})(O_i - \bar{O})}{\sqrt{\sum_{i=1}^n (M_i - \bar{M})^2} \sqrt{\sum_{i=1}^n (O_i - \bar{O})^2}}$$

$$MB = \frac{1}{n} \sum_{i=1}^n M_i - O_i$$

$$NMB = \frac{\sum_{i=1}^n M_i - O_i}{\sum_{i=1}^n O_i}$$

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

$$MAE = \frac{\sum_{i=1}^n DWD_i}{n}$$

where DWD (difference of the wind direction) is calculated from:

$$DWD_i = \begin{cases} \min(M - O, O - M + 360), & \text{for } M > O \\ \min(O - M, M - O + 360), & \text{for } M < O \end{cases}$$

$$FAC2 \text{ (fraction where } 0.5 < \frac{M}{O} < 2 \text{)}$$

368 In order to evaluate the performance of the models in urban environment, the Fractional Mean Bias (FB), Normalized
369 Absolute Difference (NAD) and Normalized Mean Square Error (NMSE) were also considered. They can be defined as
370 follow:

$$FB = 2 \frac{\overline{O - M}}{\overline{O + M}}$$

$$NAD = \frac{|\overline{O - M}|}{\overline{O + M}}$$

$$NMSE = \frac{\overline{(O - M)^2}}{\overline{O} \cdot \overline{M}}$$

371 5 Results and discussion

372 5.1 Meteorology

373 2 m temperature (T2), 10 m wind speed (ws10) and 10 m wind direction (wd10) meteorological fields predicted by WRF-
374 Chem were compared against corresponding surface observations of these variables provided by 33 stations within the d03
375 domain (see Table 3 and Figure 6): 18 stations (with 16 of them for T2, ws10 and wd10) belong to the RIRER (Rete idro-
376 meteo-pluviometrica integrata) Arpae-Simc network, 11 stations (with 7 for T2, ws10 and wd10) belong to the Archivio dati
377 idro-nivo-meteorologici ARPA Lombardia network, 4 stations belong to the Osservatorio Meteo Idrologico della Regione
378 Liguria (OMIRL) ARPAL network. Other two stations belonging to the Osservatorio Geofisico di Modena weather network
379 were considered for the micro-scale wind field evaluation. All the meteorological stations mentioned before are automated
380 and realized according to WMO (World Meteorological Organization) directive. On the other hand, NO_x concentrations at
381 air quality stations are measured using chemiluminescence. With this method all the NO_2 contained in an air sample is
382 converted to NO with a molybdenum converter, then the sample gas goes straight to the reaction chamber where NO (both
383 the original NO contained in the air sample and the NO_2 converted to NO) will react with O_3 to form NO_2 and O_2 while
384 emitting light. Finally, the measure of the emitted light will be proportional to the NO_x concentrations in the air sample.

385

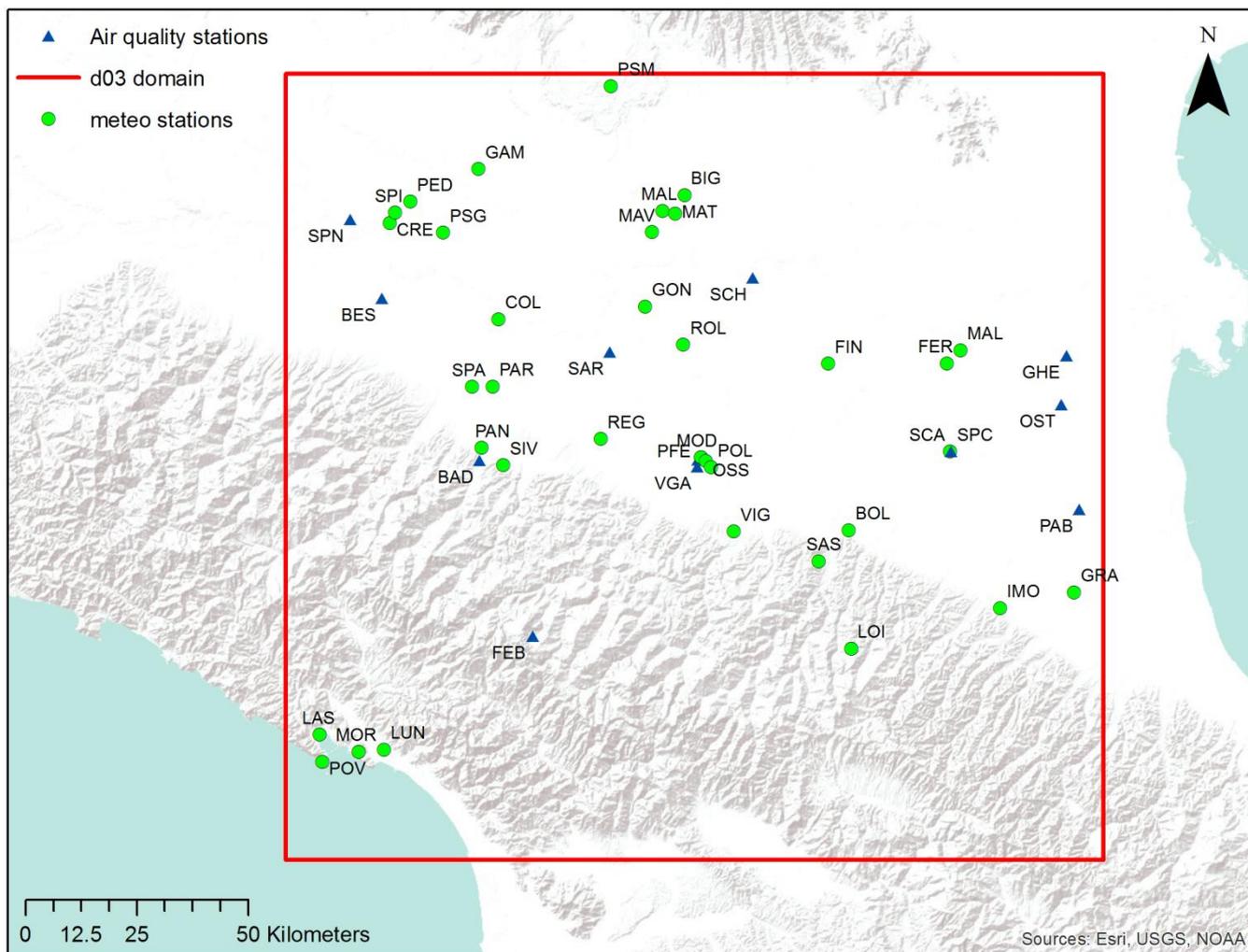
386 **Table 3:** Observation sites. Locations are provided in Geographic coordinates (WGS84). Available parameters are: “T2” for
387 temperature at 2 m height above the ground, “ws10” for wind speed and “wd10” for wind direction at 10 m above the ground
388 and “ NO_x ” for NO_x concentrations.

Station name	Label	Network	Longitude (°)	Latitude (°)	Type	Parameters
Bologna urbana	BOL	Arpae-Simc	11.32879	44.50075	Meteorology	T2, ws10, wd10
Colorno	COL	Arpae-Simc	10.34959	44.94378	Meteorology	T2, ws10, wd10
Ferrara urbana	FER	Arpae-Simc	11.621138	44.832498	Meteorology	T2
Finale Emilia	FIN	Arpae-Simc	11.284	44.839	Meteorology	T2
Granarolo Faentino	GRA	Arpae-Simc	11.95861	44.36013	Meteorology	T2, ws10, wd10
Imola	IMO	Arpae-Simc	11.74953	44.3332	Meteorology	T2, ws10, wd10
Loiano	LOI	Arpae-Simc	11.32646	44.26093	Meteorology	T2, ws10, wd10
Malborghetto di Boara	MAL	Arpae-Simc	11.66134	44.85799	Meteorology	T2, ws10, wd10
Modena urbana	MOD	Arpae-Simc	10.91699	44.65639	Meteorology	T2, ws10, wd10
Panocchia	PAN	Arpae-Simc	10.29584	44.6837	Meteorology	T2, ws10, wd10
Parma urbana	PAR	Arpae-Simc	10.33049	44.808	Meteorology	T2, ws10, wd10
Reggio Emilia urbana	REG	Arpae-Simc	10.6337	44.69781	Meteorology	T2, ws10, wd10
Rolo	ROL	Arpae-Simc	10.874	44.88481	Meteorology	T2, ws10, wd10
S.Pietro Capofiume	SPC	Arpae-Simc	11.62264	44.65378	Meteorology	T2, ws10, wd10
San Pancrazio	SPA	Arpae-Simc	10.27245	44.80806	Meteorology	T2, ws10, wd10
Sasso Marconi	SAS	Arpae-Simc	11.24125	44.43967	Meteorology	T2, ws10, wd10
Sivizzano	SIV	Arpae-Simc	10.35704	44.6482	Meteorology	T2, ws10, wd10
Vignola	VIG	Arpae-Simc	11.00414	44.50405	Meteorology	T2, ws10, wd10
Bigarello	BIG	ARPA Lombardia	10.8874	45.18783	Meteorology	T2, ws10, wd10
Cremona	CRE	ARPA Lombardia	10.04414	45.14188	Meteorology	T2, ws10, wd10
Gambara	GAM	ARPA Lombardia	10.29949	45.24903	Meteorology	T2, ws10, wd10
Gonzaga	GON	ARPA Lombardia	10.7678	44.96381	Meteorology	T2
Mantova Lunetta	MAL	ARPA Lombardia	10.82421	45.15733	Meteorology	T2, ws10, wd10
Mantova Tridolino	MAT	ARPA Lombardia	10.86007	45.15135	Meteorology	T2, ws10, wd10
Mantova Virgilio	MAV	ARPA Lombardia	10.79293	45.11438	Meteorology	T2
Persico Dosimo	PED	ARPA Lombardia	10.1041	45.1851	Meteorology	T2
Pieve S.Giacomo	PSG	ARPA Lombardia	10.19548	45.12107	Meteorology	T2, ws10, wd10
Ponti sul Mincio	PSM	ARPA Lombardia	10.68363	45.41211	Meteorology	T2, ws10, wd10
Spinadesco	SPI	ARPA Lombardia	10.05912	45.16297	Meteorology	T2
La Spezia	LAS	ARPAL	9.82819	44.10703	Meteorology	T2, ws10, wd10
Luni	LUN	ARPAL	10.00899	44.07491	Meteorology	T2, ws10, wd10
Monte Rocchetta	MOR	ARPAL	9.93842	44.07129	Meteorology	T2, ws10, wd10
Porto Venere	POV	ARPAL	9.83594	44.052	Meteorology	T2, ws10, wd10
Badia	BAD	Arpae Emilia-Romagna	10.28937	44.65823	Air Quality	NOx
Besenzone	BES	Arpae Emilia-Romagna	10.0192	44.9895	Air Quality	NOx

Febbio	FEB	Arpae Emilia-Romagna	10.43104	44.30071	Air Quality	NOx
Gherardi	GHE	Arpae Emilia-Romagna	11.96125	44.83975	Air Quality	NOx
Ostellato	OST	Arpae Emilia-Romagna	11.94194	44.7409	Air Quality	NOx
Parco Ballirana	PAB	Arpae Emilia-Romagna	11.98236	44.52743	Air Quality	NOx
San Pietro Capofiume	SCA	Arpae Emilia-Romagna	11.62482	44.65423	Air Quality	NOx
San Rocco	SAR	Arpae Emilia-Romagna	10.66478	44.87373	Air Quality	NOx
Schivenoglia	SCH	ARPA Lombardia	11.0761	45.01688	Air Quality	NOx
Spinadesco	SPN	ARPA Lombardia	9.930599	45.15047	Air Quality	NOx
Policlinico	POL	Osservatorio Geofisico MO	10.94429	44.63580	Meteorology	ws10, wd10
Osservatorio Geofisico	OSS	Osservatorio Geofisico MO	10.92981	44.64809	Meteorology	ws10, wd10
“Parco Ferrari”	VGA	Arpae Emilia-Romagna	10.90731	44.65157	Air Quality	NOx
“via Giardini”	PFE	Arpae Emilia-Romagna	10.90572	44.63699	Air Quality	NOx

389

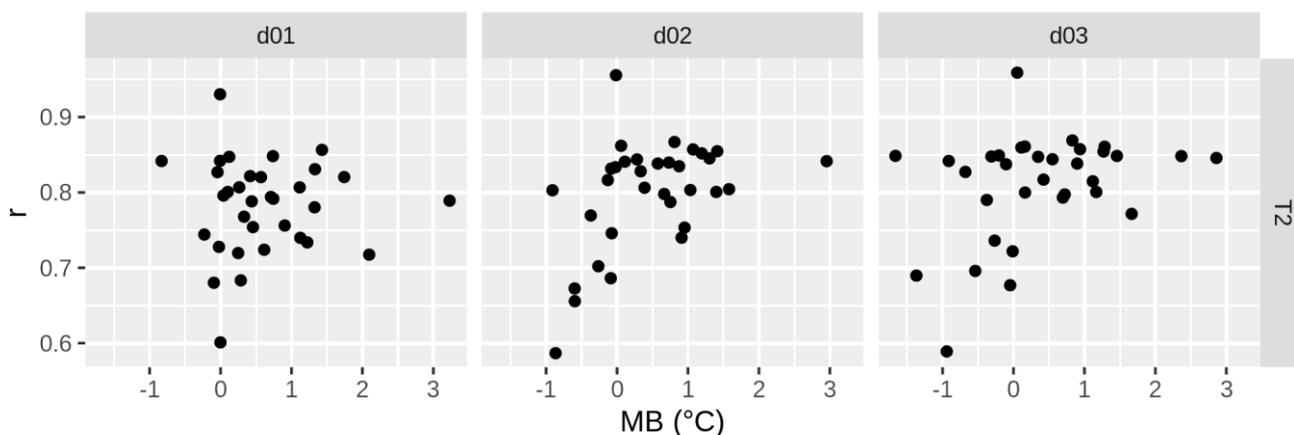
390



391
 392 **Figure 6:** Map of the stations inside the WRF-Chem d03 domain. Site of the meteorological stations are reported by the green dots and
 393 site of air quality station are reported by the blue triangles.

394
 395 Generally, modelled hourly 2 m temperatures reproduced by WRF-Chem at 1 km resolution (d03 domain) are consistent
 396 with observations at all the stations: r is between 0.68 and 0.87, except for two stations, Persico Dosimo and Bologna
 397 urbana, where the correlation is respectively very good (0.96) and not particularly high (0.59). The minimum value can be
 398 attributed to the difficulties of the model in representing urban meteorological dynamics where land use data for a large
 399 urban area may not be extremely accurate and some local phenomena can be missed. This could be the case of the Bologna
 400 city environment. By contrast, the maximum correlation observed at the Persico Dosimo station could in this case reached by
 401 a chance where hourly measurements were available only for one fourth of the investigated period. However, the good
 402 correlation between modelled 2 m temperature and observations for a large number of stations shows that the WRF-Chem
 403 model represented the observed meteorological variability quite well.

404 The model on average tends to be positively biased with a MB smaller than $+1^{\circ}\text{C}$ for most of the stations where only two of
 405 them exceed $+2^{\circ}\text{C}$ of MB, respectively at the Reggio Emilia urbana and Colorno stations. On the other hand, the minimum
 406 MB is -1.6°C , achieved at the Porto Venere station on to the Ligurian Sea shore. These results are in the same range as the
 407 MB that Gsella et al. (2014) found using MM5, WRF and TRAMPER meteorology models for the same area.
 408 Figure 7 shows the statistical performance of WRF-Chem in reproducing 2 m temperature. r is plotted as a function of MB
 409 for the three different model resolutions: 15 km (d01), 3 km (d02) and 1 km (d03). The variability of MB tends to increase by
 410 increasing the model resolution, conversely, the average r including all the stations is 0.78 for the d01 WRF-Chem domain,
 411 0.80 for the d02 domain and 0.81 for the d03 domain, showing that the increase of the model resolution from 15 km to 1 km
 412 generally leads to slightly improve the performance of the model in reproducing 2 m temperature.

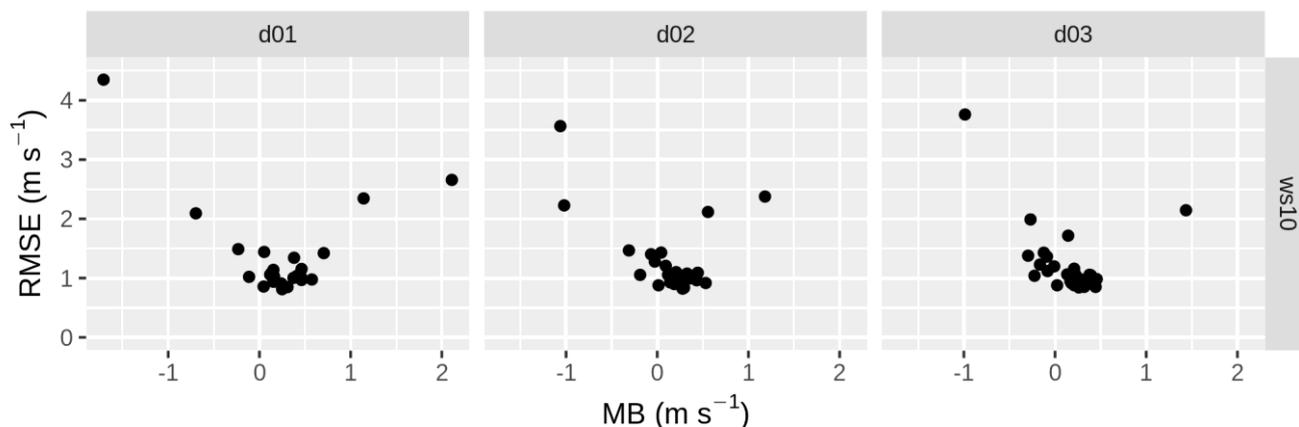


413
 414 **Figure 7:** Pearson correlation coefficient (r) reported as function of the Mean Bias (MB) between modelled hourly 2 m temperature (T2)
 415 and observation at 33 measurements sites for the three WRF-Chem resolutions: 15 km (d01), 3 km (d02) and 1 km (d03).

416
 417 Simulated hourly wind speed at the d03 domain generally express performance in line with similar case study in literature
 418 (Kuik et al., 2016, Mar et al., 2016, Gsella et al., 2014) since the great majority of the stations exhibits a MB between -0.5 m s^{-1}
 419 and $+0.5 \text{ m s}^{-1}$, range suggested by Malm et al. (2009) and by European Environmental Agency (EEA) guidelines (EEA,
 420 2011). Only the stations of Loiano and La Spezia are outside these limits with respectively a MB of -0.99 m s^{-1} and $+1.43 \text{ m s}^{-1}$:
 421 the former is located in the Tuscan-Emilian Apennines at about 700 m above the sea level and the latter is close the
 422 Ligurian sea and then characterized by a strong influence of land-sea breeze, similarly to the temperature monitoring station
 423 of Porto Venere. The large bias found at these two stations suggests that the model might have difficulties in simulating the
 424 wind field in mountainous areas and close to the sea where complex orography and local breeze characterize the territory,
 425 however for the rest of the stations the MB values are consistent with the reference benchmarks proposed in literature.
 426 Another statistical indicator suggested by the EEA guidelines (2011) and by Malm et al. (2009) is the Root Mean Square
 427 Error, for which the recommended benchmark for wind speed is less than 2 m s^{-1} . As for the MB, RMSE of modelled wind
 428 speed values are below 2 m s^{-1} at all stations besides Loiano and La Spezia, where RMSE are 3.76 m s^{-1} and 2.14 m s^{-1} ,

429 respectively. Nonetheless, modelled wind speed at the vast majority of the stations is in line with the benchmark for a
430 mesoscale meteorological reconstruction.

431 In Figure 8 the performance of WRF-Chem in reproducing wind speed for the three different resolutions is shown: RMSE is
432 plotted as a function of the MB. As the resolution is increased, the model tends to show lower MB in absolute terms.
433 Similarly, the RMSE generally tends to moderately decrease as well with increasing resolution. It is therefore possible to
434 conclude that in increasing model resolution from 15 km to 1 km there is a slight improvement of performance also for wind
435 speed reconstruction.



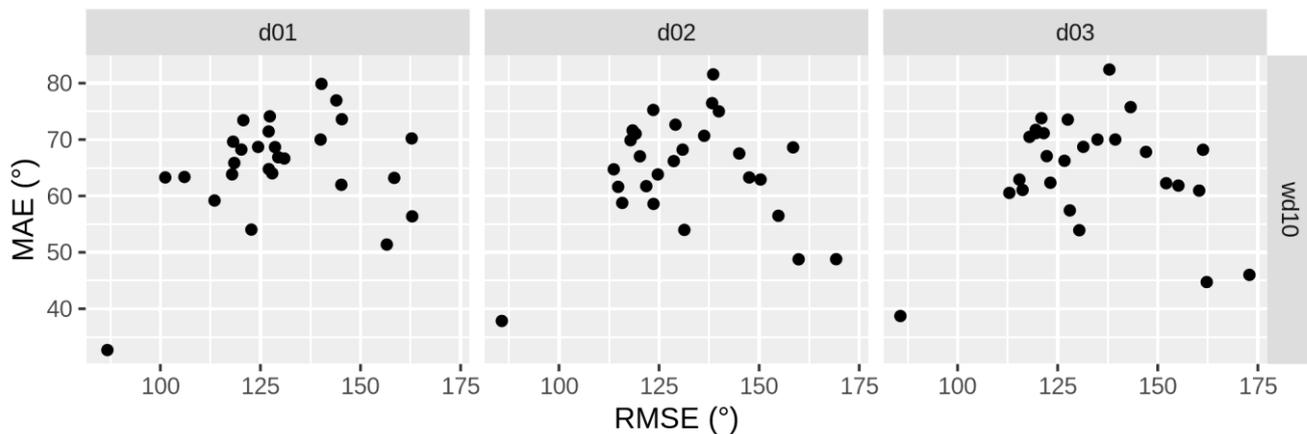
436

437 **Figure 8:** Root Mean Square Error (RMSE) reported as function of the Mean Bias (MB) between modelled hourly 10 m wind speed
438 (ws10) and observation at 27 measurements sites for the three WRF-Chem resolutions: 15 km (d01), 3 km (d02) and 1 km (d03).

439

440 Despite the WRF-Chem performance in reproducing observed wind speed were satisfactory, wind direction at the same
441 locations were in general poorly reproduced by the model in all the three domains. The MAE statistical indicator, modified
442 as indicated in section 4 to meet the special requirement for circular data, was used to quantify the model capability to
443 capture measured wind direction. In Figure 9 MAE is plotted in function of RMSE, where nevertheless this later one ignores
444 the particularities of circular data, it has been applied in several previous studies for wind direction evaluation. WRF-Chem
445 captured particularly well wind direction at Loiano station where RMSE is around 85° for all the three domains and MAE is
446 33° for domain d01 and 38° for domain d02 and d03. By contrast wind direction at Parma urbana and Reggio Emilia urbana
447 stations the model expressed the maximum MAE with values respectively equal to 80° (domain d01) and 82° (for both d02
448 and d03 domains), with RMSE about 140° at Parma urbana station for all the three domains and equal to 127° (domain d01)
449 and 137° (domain d02 and d03) at Reggio urbana.

450 Although the model performances in reproducing the wind directions are not outstanding, they are in line with other case
451 studies within the Po Valley, where MAE was between 42° and 93° (Gsella et al., 2014, de Meij et al., 2009) and RMSE was
452 between 127° and 148° (Gsella et al., 2014), confirming the difficulties in modelling the wind fields in this area
453 characterized during winter and fall time by stagnant conditions and low wind speed.



454

455 **Figure 9:** Mean Absolute Error (MAE) reported as function of the Root Mean Square Error (RMSE) between modelled hourly 10 m wind
 456 direction (wd10) and observation at 27 measurements sites for the three WRF-Chem resolutions: 15 km (d01), 3 km (d02) and 1 km (d03).

457

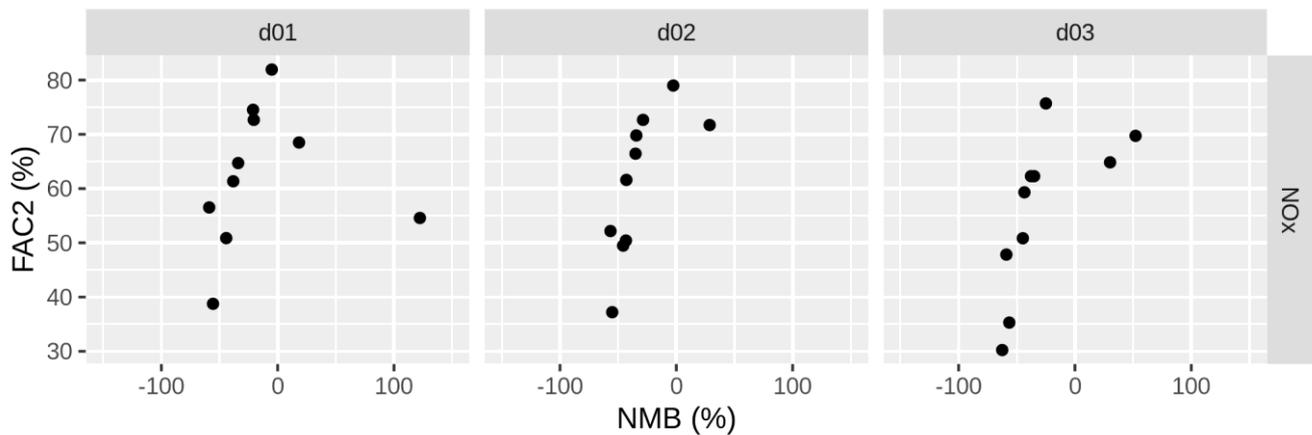
458 5.2 WRF-Chem nitrogen oxides

459 Since the role of WRF-Chem in this study was to estimate NO_x ($\text{NO} + \text{NO}_2$) concentrations due to emissions on the regional
 460 scale that may affect the background air quality in Modena, modelled hourly NO_x concentrations were compared with
 461 observations at 10 rural background sites (8 of them from the Arpa Emilia-Romagna network and 2 of them from Arpa
 462 Lombardia network, see Table 3 and Figure 6) within the d03 WRF-Chem domain.

463 Modelled NO_x concentration at 1 km resolution is biased negatively for 8 stations with a minimum MB equal to $-18.1 \mu\text{g m}^{-3}$
 464 (-62% of NMB) and biased positively for 2 stations with a maximum MB of $+7.8 \mu\text{g m}^{-3}$ ($+30\%$ of NMB). In addition, for
 465 each reference station, the fraction of predicted values within a factor of two of observations was computed, also referred as
 466 FAC2. The corresponding average value over all stations is 56% (minimum 30% at the station of Ostellato and maximum
 467 76% at the stations of Schivenoglia), in accordance with the reference value suggested by Chang and Hanna (2004), greater
 468 or equal to 50%.

469 In order to test which level of model spatial resolution gives better results, observed NO_x concentrations were compared with
 470 modelled concentrations at 15 km (d01) and 3 km (d02) resolutions in terms of FAC2 and NMB (Figure 10). The model at
 471 15 km resolution presents on average the highest FAC2 with respect other configuration, 62% (maximum equal to 82% at
 472 the stations of Schivenoglia and minimum equal to 39% at the station of Ostellato), conversely the NMB presents its higher
 473 variability, from -58.9% to 122.2% (respectively $-17.5 \mu\text{g m}^{-3}$ and $+5.7 \mu\text{g m}^{-3}$ of MB).

474 Among the other configurations, WRF-Chem at 3 km resolution shows better performance with an averaged FAC2 between
 475 all the stations equal to 61% (79% of maximum at Langhirano station and 37% of minimum at the station of Ostellato) and
 476 the smallest variability between all the stations in terms of NMB, from -56.6% to 28.7% (respectively $-16.8 \mu\text{g m}^{-3}$ and $+1.3$
 477 $\mu\text{g m}^{-3}$ of MB).



478

479 **Figure 10:** Factor of two (FAC2) reported in function of the Normalized Mean Bias (MB) between modelled hourly NO_x concentrations
 480 and observation at 10 rural background sites for the three WRF-Chem resolutions: 15 km (d01), 3 km (d02) and 1 km (d03).

481

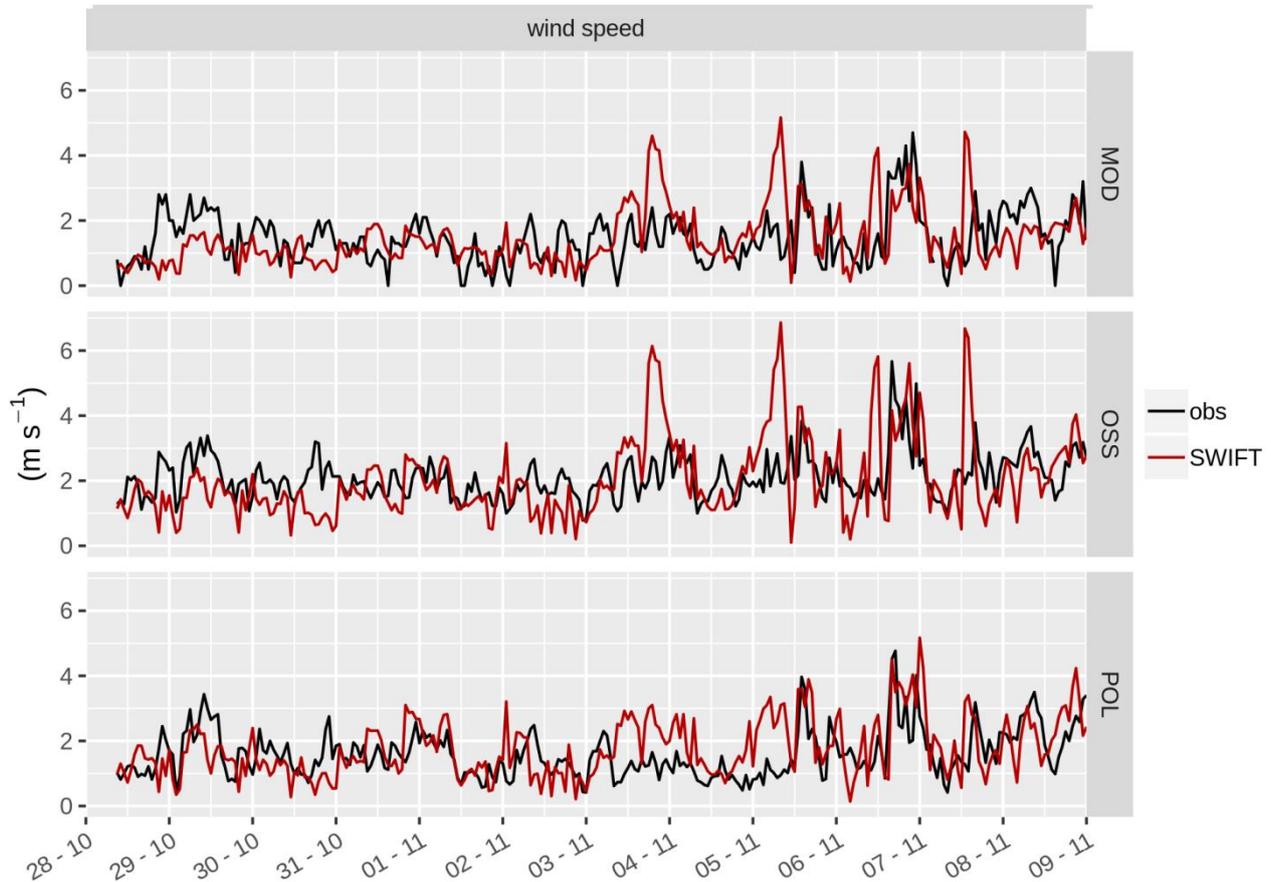
482 The comparison showed that generally the WRF-Chem model with a horizontal resolution of 3 km is better suited to
 483 reproduce NO_x concentrations at rural background sites considering an emission inventory of 14 km x 7 km resolution.
 484 Increasing resolution to 1 km, the model tends to decrease the number of predicted concentrations within a factor of two of
 485 observations, despite the variability of the NMB being approximately the same as the configuration at 3 km. It is also worth
 486 remarking that nonetheless the modelled NO_x concentrations are on average in agreement with the values suggested by
 487 Chang and Hanna (2004) for model performance evaluation in terms of NMB and FAC2, not all considered stations satisfy
 488 these statistical indicators, this might mean that a more detailed emissions estimation such as an improved emissions
 489 distribution in the area should be implemented to achieve better results.

490 5.3 WRF-Chem combined with PMSS

491 5.3.1 Micro-scale wind fields

492 High resolution winds in the urban area of Modena were estimated by the means of Micro-SWIFT (the 3D mass-consistent
 493 diagnostic model that composes PMSS), initialised with the meteorological fields reproduced by WRF-Chem at hourly time
 494 step. Simulated winds reproduced by Micro-SWIFT were compared with observation at three different meteorological sites
 495 located within the urban center of the city. These stations are respectively placed on top to the Geophysical Observatory
 496 tower, at 42 m height above the ground and placed in the historical part of the city (referred as OSS), above the public
 497 hospital to the Est of the historical city center at about 20 m height (referred as POL), and on top to the municipality building
 498 at 40 m height, to the West of the historical city center (MOD station), the latter used also to validate the WRF-Chem
 499 meteorology. Figure 4 depicts the position of these three stations.

500 The time series reported in Figure 11 show the comparison between modelled and measured hourly wind speed for the three
501 urban meteorological sites. Notwithstanding a few remarkable overestimations on November 3, 5 and 7, mostly visible at the
502 OSS and MOD stations, modelled data reproduced observed trend quite well for all the three locations. MOD and OSS
503 modelled time series show also a very similar behaviour (with a general less pronounced overestimation for MOD on
504 November 3, 5 and 7) due to the location of the sensor, both above 40 m and thus characterized by the same meteorological
505 input and not affected by the presence of buildings.



506
507 **Figure 11:** Hourly observed wind speed at MOD (on top), at OSS (in the middle) and at POL (on bottom) meteorological site along with
508 hourly simulated wind speed by Micro-SWIFT, from October 28 to November 8, 2016.

509
510 The satisfactory performances showed by the time series regarding wind speed are also confirmed by the statistical metrics.
511 The MB is less than 0.2 m s^{-1} for all the three stations and the RMSE is between 0.93 and 1.24 m s^{-1} . FB and FAC2 are also
512 in line with the values found during the validation of the PMSS modelling suite in urban environment (Oldrini et al., 2019).
513 Table 4 summarizes computed metrics.

514

515
516
517

Table 4: Statistics of hourly wind speed computed for the period between October 28 and November 8 at three urban meteorological stations.

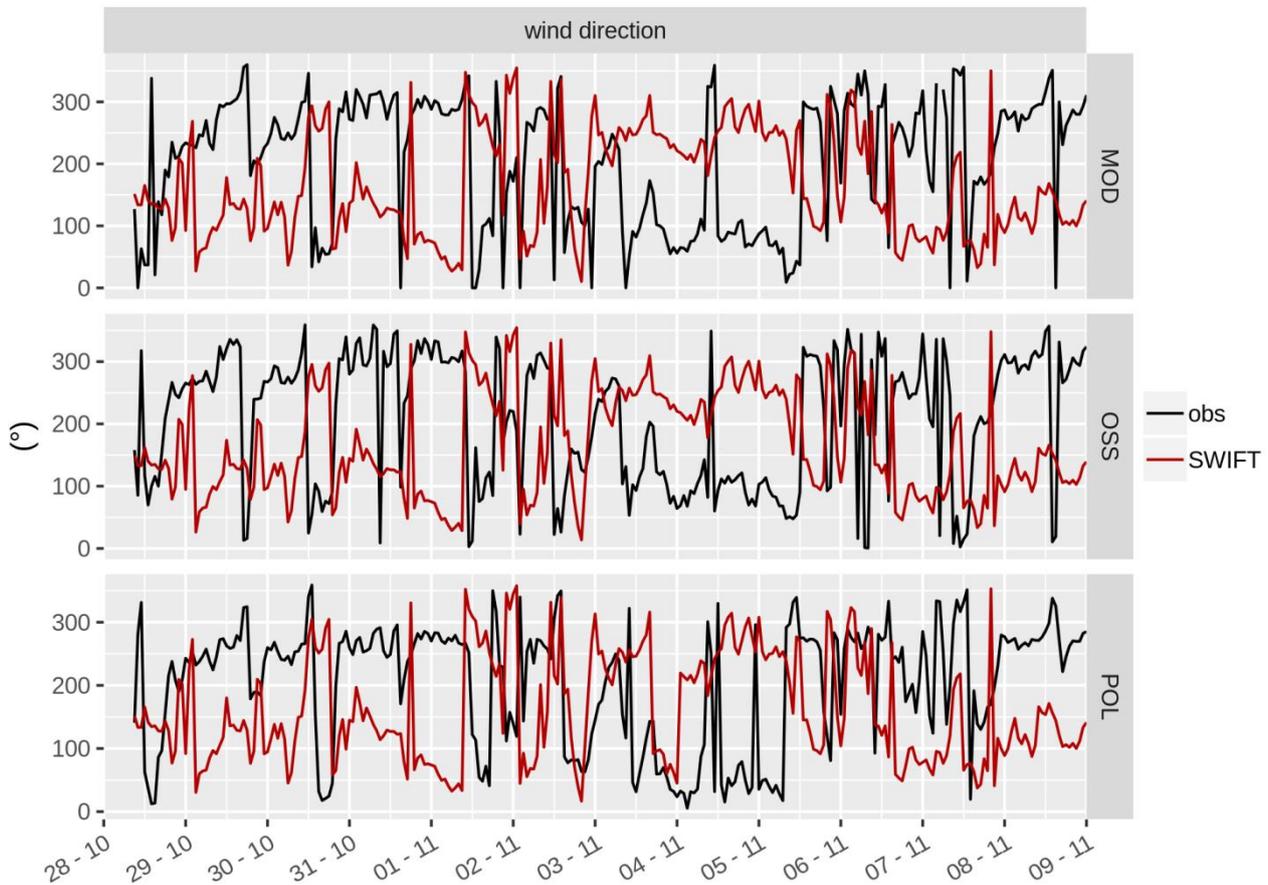
Station	FB	MB	NMB	RMSE	FAC2
MOD	0.003	-0.005	-0.3%	1.05	0.67
OSS	0.03	-0.05	2%	1.24	0.76
POL	-0.10	0.17	11%	0.93	0.76

518

519 In Figure 12 hourly simulated wind directions are compared with hourly observed wind for the same three locations. As for
520 regional wind field evaluation, wind directions were poorly reproduced by Micro-SWIFT, respectively with MAE equal to
521 125° for the MOD station, 124° for OSS and 114° for POL. By contrast the performance of Micro-SWIFT evaluated in terms
522 of FB and NMSE are similar to the results obtained in urban environment by Oldrini et al. (2019) using the same model: for
523 MOD site the FB and NMSE are respectively equal to 0.21 and 0.76, FB is 0.23 for both OSS and POL locations and NMSE
524 is 0.80 and 0.70 at OSS and POL stations.

525 This poor behaviour in wind direction reconstruction can be partly attributed to the input data used by Micro-SWIFT, which
526 reflects the bias given by the regional forecast model in wind direction estimation. This is mainly due to the difficulty of the
527 meteorological models in reproducing wind fields during the situations with very little atmospheric circulation (stagnant
528 condition) and low wind speed, as occur red in this case study (Gsella et al., 2014, de Meij et al., 2009).

529



530

531 **Figure 12:** Hourly observed wind direction at MOD (on top), at OSS (in the middle) and at POL (on bottom) meteorological sites along
 532 with hourly simulated wind direction by Micro-SWIFT, from October 28 to November 8, 2016.

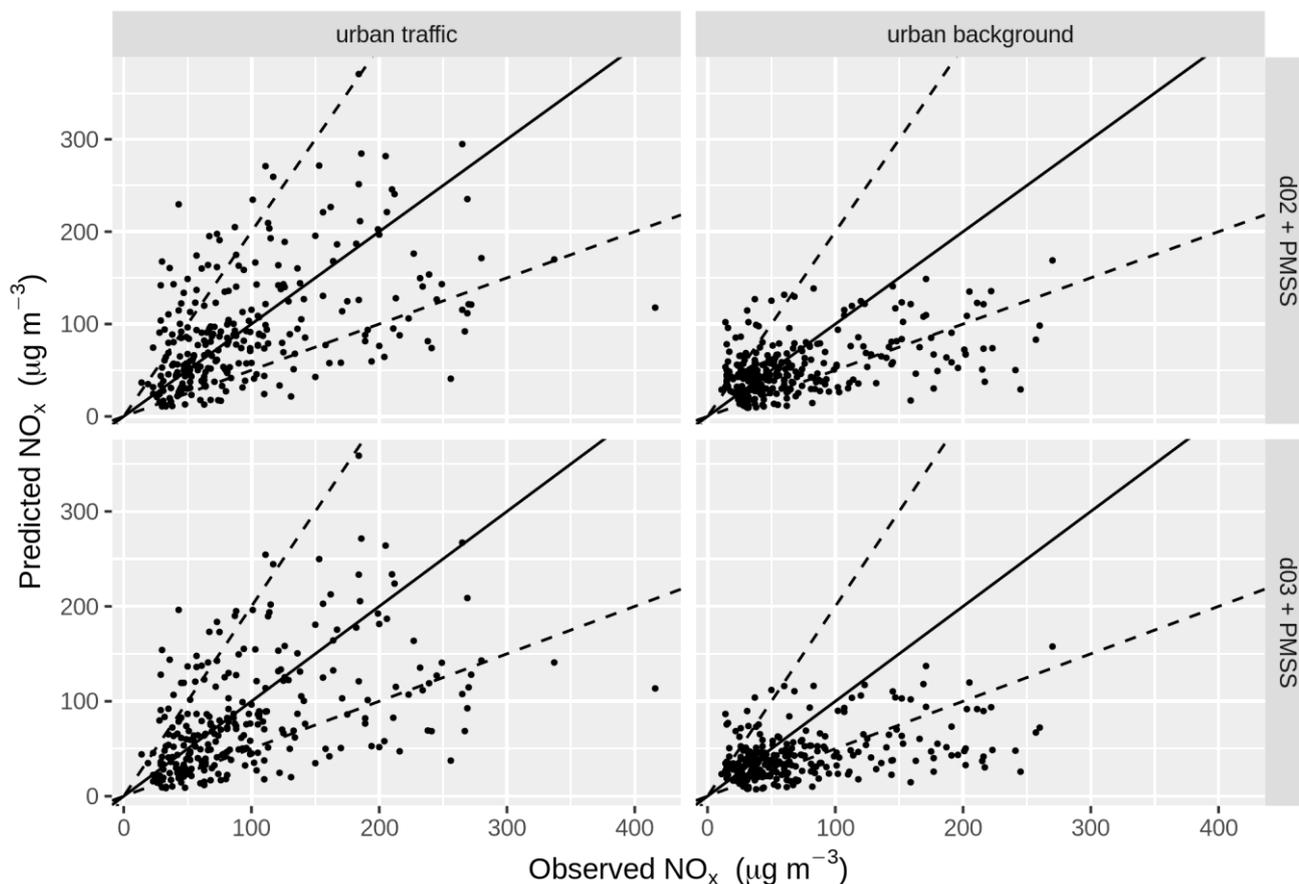
533

534 **5.3.2 Micro-scale NO_x concentrations**

535 NO_x background concentrations estimated with WRF-Chem in the urban area of Modena were added to the NO_x simulated
 536 concentrations reproduced with PMSS modelling system performed considering exclusively road traffic emissions. Modelled
 537 concentrations were compared with observations at two urban stations: the first one at a traffic site, located in the proximity
 538 of a busy street close to the urban ring road, named “via Giardini”, and the second one at background site, within a public
 539 park to the West of the historical city centre, named “parco Ferrari” (Figure 4).

540 In Figure 13, the hourly NO_x concentrations predicted by PMSS in combination with WRF-Chem at 3 km and 1 km
 541 resolutions (labelled as “d02+ PMSS” and as “d03+ PMSS”) are compared through scatter plots for both urban traffic and
 542 background stations. In this figure, the solid line represents perfect agreement with observations and within the dashed lines
 543 modelling results and observations agree with a factor of two.

544 Most of the modelled data are within a factor of two of observations, especially for the urban traffic site in both WRF-Chem
 545 configurations, whereas for the urban background station an under estimation is more noticeable. It is also worth noting that
 546 the results of PMSS combined with WRF-Chem at 3 km and at 1 km resolution depict similar behaviour and relative scatter
 547 plots are very comparable to each other, with a slightly less pronounced underestimation for WRF-Chem at 3 km resolution.
 548
 549
 550



551
 552 **Figure 13:** Scatter plots of predicted NO_x concentrations at urban traffic (“via Giardini”) and urban background (“parco Ferrari”) stations
 553 for different models configurations: PMSS combined with WRF-Chem at 3 km resolution (labelled as “d02+PMSS”) and PMSS combined
 554 with WRF-Chem at 1 km resolution (labelled as “d03+PMSS”).
 555

556 Modelled hourly NO_x concentrations are biased negatively in both urban stations: at the “via Giardini” traffic site the MB of
 557 simulated NO_x by PMSS and WRF-Chem at 1 km resolution is $-15 \mu\text{g m}^{-3}$, which corresponds to -15% of NMB, for the
 558 same model configuration the MB at the “parco Ferrari” background site is $-30 \mu\text{g m}^{-3}$ (-41% NMB). Modelled hourly

559 concentrations correlate reasonably well with observations in both sites, with r equal to 0.48 at the traffic station and 0.43 at
560 the background station.

561 The performance of the models generally increases when hourly NO_x concentrations reproduced by PMSS are combined
562 with the results of WRF-Chem at 3 km resolution, the MB at the “via Giardini” traffic station is $-4 \mu\text{g m}^{-3}$ (-4% NMB) and -
563 $18 \mu\text{g m}^{-3}$ (-25% NMB) at the “parco Ferrari” measurement station.

564 Despite an improvement in term of MB with WRF-Chem at 3 km, its combination with PMSS doesn’t particularly affect the
565 r between modelled and observed concentrations (0.47 at “via Giardini” traffic station and 0.44 at “parco Ferrari”
566 background site).

567

568 A quantitative estimation of the agreement between simulated and observed concentrations was also assessed following the
569 statistical metrics proposed by Hanna and Chang (2012) for urban dispersion model evaluation. FB, NMSE, FAC2 and NAD
570 were computed for both the urban stations located in Modena and for both the combination of WRF-Chem (3 km and 1 km
571 resolution) with PMSS. Table 5 summarizes all the computed statistics.

572 Following Hanna and Chang (2012) the reference acceptance criteria for the aforementioned metrics in urban dispersion
573 model evaluation are as follows:

- 574 • $|\text{FB}| < 0.67$, i.e. the relative mean bias is less than a factor of ~ 2
- 575 • $\text{NMSE} < 6$, i.e. the random scatter is less than 2.4 times the mean
- 576 • $\text{FAC2} > 0.30$, i.e. the fraction of predicted concentrations within a factor of two of observed concentrations exceeds
577 0.30
- 578 • $\text{NAD} < 0.50$, i.e. the fractional area for errors is less than 0.5

579

580 **Table 5:** Statistics of hourly NO_x concentrations computed for the period between October 28 and November 8, considering two different
581 model configurations.

Configuration	Station	FB	NMSE	FAC2	NAD
PMSS + d02	urban traffic	0.04	0.48	0.72	0.24
	urban background	0.29	0.75	0.62	0.29
PMSS + d03	urban traffic	0.16	0.54	0.68	0.26
	urban background	0.52	1.15	0.59	0.35

582

583

584 The statistical analysis shows that PMSS combined with WRF-Chem at both d02 and d03 domains fulfill the acceptance
585 criteria defined by Hanna and Chang (2012). Regarding the FB, the results are always less than the threshold of 0.67, in
586 particular at urban traffic site the outcomes of this metric are particularly good with values equal to 0.04 (PMSS + d02) and
587 0.16 (PMSS + d03). At urban background station the results are larger than the previous one (0.29 for PMSS + d02 and 0.52
588 for PMSS + d03) indicating that the models tend to underestimate more the mean concentrations but nevertheless in well

589 agreement with the reference benchmark. As far as the NMSE is concerned, the models show their best performances with
590 scores largely lower than the acceptance benchmark (6), with a maximum value of 1.15 at urban background station for
591 PMSS + d03 (minimum value at traffic station for PMSS + d03 equal to 0.48), meaning that predicted values very rarely
592 differ strongly from observations.

593 Regarding the FAC2 and the NAD there is a significant agreement between model results and relative acceptance criteria at
594 both urban stations and for both model configurations. Minimum and maximum FAC2 are equal to 0.59 and 0.72 achieved
595 respectively at the urban background station for PMSS + d03 and at urban traffic station for PMSS + d02 (the lower limit
596 proposed by Hanna and Chang is 0.30). For the same locations and model configuration the maximum and the minimum
597 NAD are 0.35 and 0.24 respectively (upper limit proposed by Hanna and Chang is 0.50).

598 The statistical analysis, despite supporting a moderately better behaviour when the resolution of WRF-Chem is 3 km, also
599 shows that the metrics of modelled NO_x concentrations are comparable between the two WRF-Chem resolutions (1 km and 3
600 km), without a clear difference of one of the two.

601

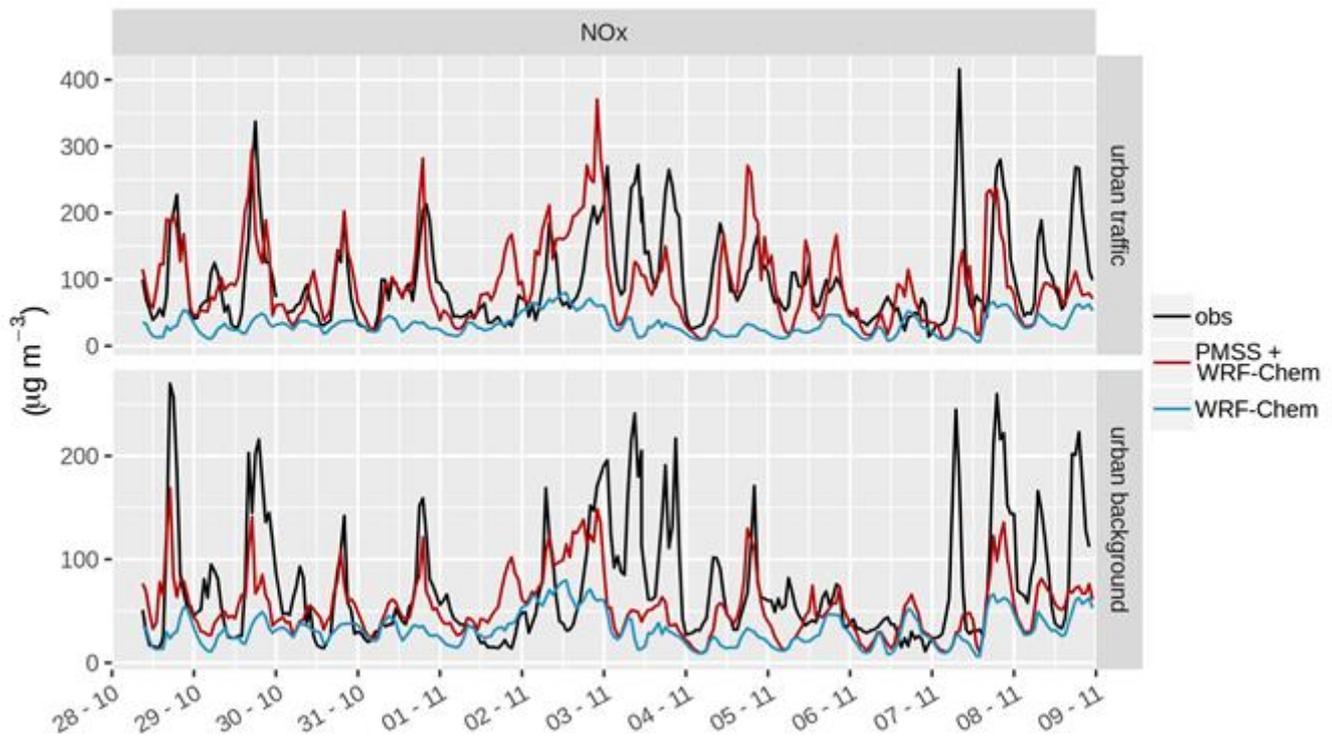
602 Time series analysis (Figure 14) shows that modelled NO_x concentrations agree quite well with observations. In particular
603 for the first five days of simulations (considering the combination with WRF-Chem at 3 km resolution as a reference),
604 between October 28 and November 1st observations are reproduced well and daily peaks are modelled with quite good
605 accuracy, especially for urban traffic station where the MB between October 28 and November 1 is $+7\mu\text{g m}^{-3}$ and NMB is
606 about 1% (at the background station the MB is $-14\mu\text{g m}^{-3}$ and NMB is -1%).

607 By contrast, between November 1 and 2 observed NO_x concentrations tend to be overestimated since the WRF-Chem
608 contribution during the central hour of days exceeds observed NO_x concentrations. This situation is particularly evident at the
609 “parco Ferrari” station. Furthermore, between November 2 and 3, PMSS failed to capture the diurnal cycle in observed
610 concentrations: on November 2 in the central hours of the day observations are largely overestimated with an increasing
611 trend up to the afternoon peak. A possible explanation to this episode can be addressed to an underestimation of the PBL
612 height during the central hours of the day, where its modelled value doesn't exceed 190 m (average daily maximum during
613 the whole simulation is 600 m) and during only 6 hours the modelled PBL height is greater than 100 m.

614 On the other hand, on November 3, when observed concentrations reach values over $200\mu\text{g m}^{-3}$, the models underestimate
615 the observed values. Between November 4 and November 6 observed NO_x concentrations were lower than in the previous
616 days (hourly maximum always lower than $200\mu\text{g m}^{-3}$) mostly due to the rainfall which occurred on November 5 (8 mm) and
617 6 (1 mm), where the combination of the two models was able to reproduce the hourly pattern well, with a slight
618 overestimation at the traffic station.

619 On November 7 and 8 extremely high peaks occurred with values that exceeded $400\mu\text{g m}^{-3}$ at urban traffic station and 250
620 $\mu\text{g m}^{-3}$ at urban background station. Despite an underestimation of absolute observed NO_x concentrations experienced on
621 these two days, the shape of the diurnal cycle was captured quite well by the models.

622



623

624 **Figure 14:** Hourly observed concentrations of NO_x at urban traffic (on top) and urban background (on bottom) measurements stations
 625 along with hourly simulated concentrations by WRF-Chem at 3 km resolution and PMSS combined with WRF-Chem at the same
 626 resolution, from October 28 to November 8, 2016. Please note the difference in the scale on the y-axis in each panel.

627

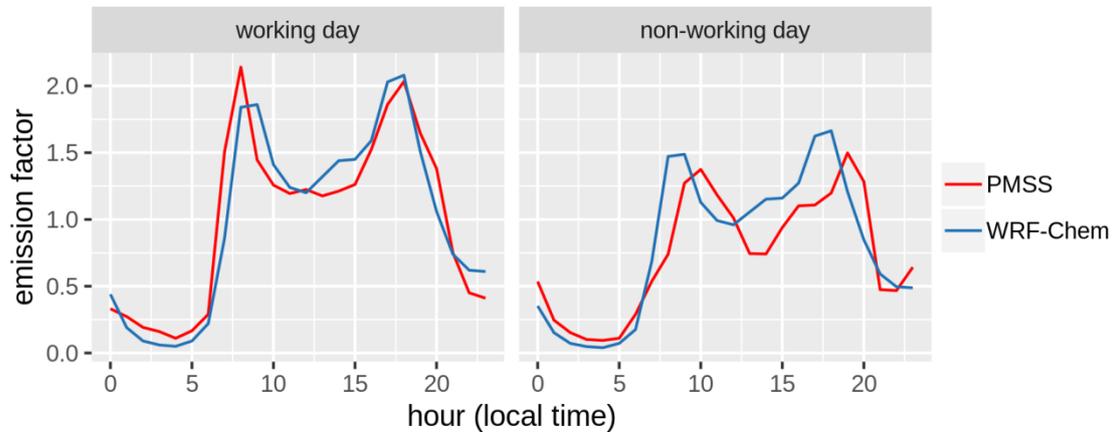
628

629 Finally, in order to study which part of day primarily affected the general underestimation of the models and to investigate
 630 the WRF-Chem contribution to the total NO_x concentrations during the day, the variation of observed and predicted NO_x
 631 concentrations by hour of the day was assessed. The comparison of mean observed NO_x daily cycle shows very similar
 632 behaviour between the “via Giardini” and “parco Ferrari” stations (Figure 16): two main peaks occur on average during the
 633 day, one between 08:00 and 09:00 a.m. (equal to 150 µg m⁻³ at traffic site and 105 µg m⁻³ at background site) and the second,
 634 on average greater of about 20-30 µg m⁻³ than the former, around 07:00 p.m. (about 170 µg m⁻³ at traffic site and 140 µg m⁻³
 635 at background site). At “parco Ferrari” station another peak occurs in the early morning (around 01:00 a.m.) less pronounced
 636 than the other two, with a mean concentration of about 70 µg m⁻³.

637

638 For the purpose of further investigate the modelled concentrations, in Figure 15 is shown the diurnal cycle of traffic
 639 emissions used for WRF-Chem and the average diurnal cycle used for PMSS in working and non-working days. There is a
 640 large correspondence between the traffic temporal cycle of WRF-Chem and PMSS in working days. For the former, the

641 afternoon peak (which occurs at 06.00 p.m.) is a little more pronounced than the morning maximum (08:00 and 09:00 a.m.),
642 while for PMSS is the opposite. During non-working days WRF-chem presents the same hourly behaviour of for working
643 days but absolute values are quite decreased. By contrast, as far PMSS is concerned, the morning peak is at 10:00 a.m. and
644 the afternoon maximum is at 07:00 p.m., both delayed by one hour compared to WRF-Chem.
645



646
647 **Figure 15:** Overview of the diurnal cycle of the traffic emission factor for WRF-Chem (light blue) and the average diurnal cycle for PMSS
648 (red) for working and non-working days.
649

650 Modelled daily NO_x concentrations present a good agreement with observed daily cycle for traffic station: the first peak of
651 the day on average tending to be underestimated and despite the second peak is anticipated of about two hours, its magnitude
652 is captured very well, especially when the WRF-Chem model is used at 3 km resolution. This confirms the advantages
653 obtained using a detailed traffic modulation recorded on a street in the proximity of the traffic station.

654 On the other hand, at urban background site, the modelled daily pattern is acceptable, but absolute concentrations are
655 generally underestimated in particular in the morning peak. This behaviour might be explained looking at the PBL height
656 simulated by WRF-Chem and used to drive the micro-scale dispersion (Figure 16): during the night the boundary layer is
657 stable due to surface longwave cooling and a shallow temperature inversion strong characterizing the area with a mean PBL
658 height during night time hours for the whole simulation around 100 m. After dawn, surface heating builds up a shallow
659 mixed layer, which deepens during the central hours of the day inducing a rising of the PBL height, on average up to 600 m.
660 During this situation, at the background site, where the traffic emissions are less pronounced compared to the traffic site,
661 modelled NO_x concentrations tend to be limited by the mixing phenomenon that occurs from around 07:30 a.m. onwards,
662 causing a strong underestimation of the morning observed peak.

663 In the late afternoon (between 04:00 and 05:00 p.m.), the solar heating is no longer sufficient to maintain an upward surface
664 buoyancy flux and the boundary layer becomes restricted to a shallow layer, around 100 m deep, as in the night. Under this
665 condition NO_x concentrations increase, generating the second daily peak more pronounced than the first one and anticipated

666 of about two hours. A more detailed description of the PBL evolution during the day, through for example ceilometer
667 observation, could help to improve the concentration estimation.

668 At urban traffic site the mechanism seems to be the same but large traffic emissions affected less the underestimation during
669 the morning peak.

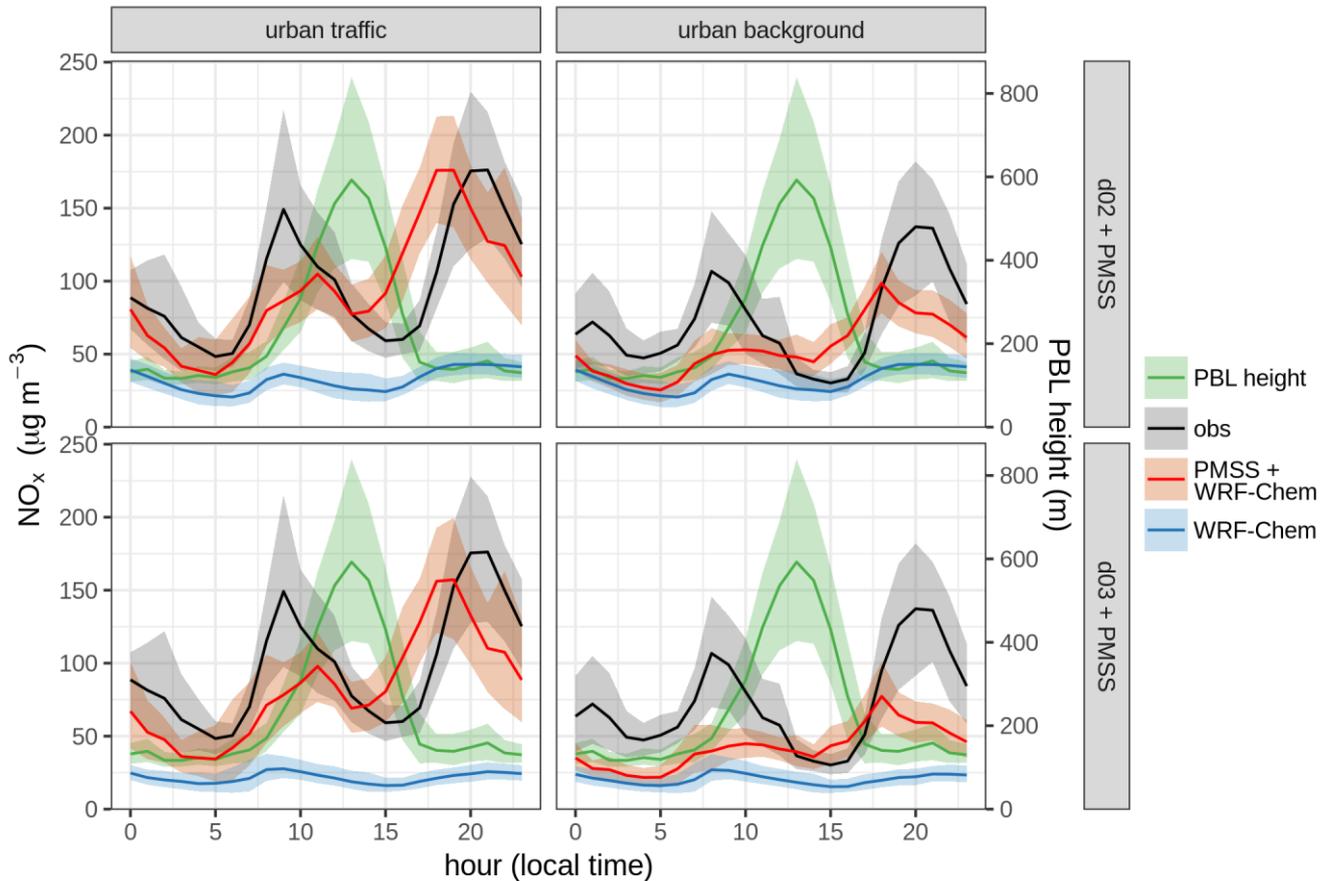
670 The NO_x underestimation could also be associated to the aged traffic fluxes estimated by the PTV VISUM model, which
671 simulation reference year is 2010 and then affected by economic recession whereby all western countries have fallen since
672 2007. The economic crisis caused a slower growth of gross national product and an increase in unemployment which, among
673 the other consequences, led to a reduction in traffic fluxes in the European business centres. For these reasons traffic fluxes
674 estimated by PTV VISUM in the city of Modena may be affected by an underestimation with respect the real traffic occurred
675 during the simulated period.

676 It is also worth noting that the PTV VISUM simulation doesn't include traffic fluxes of urban public transport, therefore the
677 vast majority of urban buses emissions are not taken into account in the simulation. Due to the presence of schools nearby
678 the "via Giardini" monitoring station, the morning rush hours (especially between 08:00 and 09:00 a.m.) are characterized by
679 an intense flow of public buses and the consequent omission of public transport emissions can influence and contribute to the
680 morning NO_x underestimation.

681 In addition, traffic fluxes in secondary streets seem to be modelled worse than busier roads and at the urban background
682 station the NO_x underestimation can also be attributed to a rough estimation of NO_x sources around the area where traffic
683 emissions are modulated according the measurement data collected at "via Giardini" street. Moreover, other NO_x emissions
684 sources are simulated by the WRF-Chem model through the use of the TNO-MACC inventory which level of detail is about
685 7 km and thus not as exhaustive as the traffic emissions, and these other NO_x sources are expected to have larger influence at
686 urban background than at urban traffic sites. Besides this, looking at the NO_x concentrations daily cycle, the contribution of
687 WRF-Chem to the total simulated concentrations seems to be modest compared to direct effect of traffic emissions. In
688 particular, when the resolution of WRF-Chem is 1 km, its share doesn't present a strong hourly trend but it is rather flat, with
689 an average contribution during each hour of simulation in the order of 20 µg m⁻³, at both traffic and background sites. On the
690 other hand, when the resolution of WRF-Chem is 3 km, its rate to the total concentrations is greater than the one a 1 km
691 resolution (average hourly contribution about 30 µg m⁻³), with an hourly trend more marked during the two daily peaks. This
692 difference in contribution estimate could be explained considering the position of the WRF-Chem computational cells. In the
693 case of 1 km resolution the computational cells over "parco Ferrari" and "via Giardini" stations, even if they present very
694 similar concentrations, are different and located both inside the PMSS domain. Conversely, for d02, both the stations are
695 within the same computational cell and the cell itself is no longer entirely contained in the PMSS domain but it extends to
696 the West, beyond the PMSS borders. This latter configuration leads to account within the PMSS domain part of the TNO
697 traffic emissions occurring outside the Modena urban area, originally excluded from the PMSS computation, causing a
698 homogenization and an increasing in NO_x concentrations over the entire WRF-Chem cell. The final outcome is that the

699 contribution of WRF-Chem using d02 to the urban concentrations is larger and also the daily trend during the two daily
700 peaks appears more pronounced.

701 More accurate traffic modulation across the city and a finer spatial resolution of non-traffic related exhaust emissions, as
702 well as a more accurate description of the planetary boundary layer height during the day, would better fit the 1km grid of
703 WRF-Chem used in d03 and can certainly contribute to achieve better results also at urban background station.



704

705 **Figure 16:** Mean daily cycle of observed NO_x concentrations (black), modelled by the combination of WRF-Chem and PMSS model (red)
706 and the only contribution of WRF-Chem (light blue), by station type (traffic or background) and by WRF-Chem resolution (3 km or 1 km).
707 Green lines show the mean daily cycle of planetary boundary layer height modelled by WRF-Chem and used in the micro-scale dispersion.
708 Solid lines represent the daily mean cycle, meanwhile shaded area show the variability between 25th and 75th percentiles.

709 6 Summary and conclusions

710 In this study the authors evaluated a hybrid modelling system consisting of the chemical transport model WRF-Chem and the
711 Parallel Micro SWIFT SPRAY (PMSS) modelling system for the urban area of Modena with the aim of providing NO_x

712 concentration maps at building-resolving scale and at hourly temporal resolution, suitable to resolve the variability of
713 emissions and atmospheric state. The WRF-Chem model was applied over three nested domains with an increasing
714 resolution from 15 km to 1 km passing by 3 km, in order to simulate the emissions, transport and chemical transformations at
715 the regional scale by accounting also at the same time for meteorological phenomena at the synoptic scale. Driven by these
716 meteorological fields, the PMSS modelling suite was run over a domain of 6 km x 6 km with 4 m grid step size, to
717 reconstruct micro-scale wind streams inside the urban area of Modena and then to simulate the dispersion of NO_x, coming
718 from traffic emissions, by accounting for the presence of buildings.

719 The 2 m temperature and 10 m wind speed were captured well by the WRF-Chem model with statistical metrics in line with
720 benchmark values suggested by the guidelines of the European Environmental Agency for meteorological mesoscale
721 reconstruction, and with similar case studies related to the same area. Only few exceptions were observed at particular
722 locations such as in mountainous area or close to the Ligurian sea, where complex orography and the local sea breeze
723 strongly influenced the model bias in reproducing the meteorology. Despite, 10 m wind direction was poorly reproduced by
724 WRF-Chem, its performance in term MAE and RMSE was in line with other cases studies focusing on the PO Valley,
725 suffering both from the models difficulties in reproducing wind field during the situations with very little atmospheric
726 circulation and low wind speed. Moreover, increasing WRF-Chem resolution from 15 km to 1 km resolution generally
727 tended to slightly improve the model performance in reproducing 2 m temperature and 10 m wind speed.

728 NO_x concentrations reproduced in the Po Valley area by WRF-Chem were on average simulated reasonably well, but
729 generally underestimated in almost all the rural background monitoring stations. The comparison with observations showed
730 also that with an emissions inventory of 7 km horizontal resolution, the 1 km model resolution does not generally improve
731 the results and the model configuration at 3 km resolution expressed the best performance in modelling NO_x concentrations.

732 Simulated and observed NO_x hourly concentrations in the urban area of Modena exhibit a large agreement, in particular for
733 urban traffic site (“via Giardini” measurement station), where detailed traffic emissions estimation (real traffic modulation
734 combined with a bottom-up approach) proved to be very successful in reproducing the observed NO_x pattern, confirming that
735 reasonable time modulation for traffic emissions are among the main parameters to trim for urban atmospheric dispersion.
736 Despite the morning rush-hour peak (between 08:00 a.m. and 09:00 a.m.) tending to be generally underestimated, the
737 magnitude of the afternoon peak around 07:00 p.m. was well captured by PMSS with an anticipation of about two hours. At
738 the urban background station, notwithstanding a general underestimation of the observed concentrations (more pronounced
739 than at the urban traffic site), the analysis of hourly daily modelled concentrations shows that PMSS combined with WRF-
740 Chem provided a daily pattern in line with observations.

741 These features highlight the strength of this modelling chain in representing urban air quality, in particular at traffic sites,
742 whose concentration levels make them the most critical area of the city; characteristics that chemical transport models alone
743 cannot express, due to the coarser resolution to which they operate and to their inability to reproduce street canyons and
744 urban structures.

745 Despite the inclusion of external emission sources for computational cells at PMSS boundaries, WRF-Chem at 3 km
746 resolution generally presented slightly better results than the 1 km resolution, demonstrating that the contribution of
747 background sources to urban pollution levels were generally underestimated and a better quantification of the emissions also
748 in the surrounding area of Modena could improve the final result. The background underestimation may depend also on
749 WRF-Chem perfectible effectiveness in reproducing the effects of the air mass homogenization on a regional scale that
750 characterizes the climate conditions in central Po Valley.

751

752 The statistical analysis showed finally that PMSS combined with WRF-Chem at both the resolutions (3 km and 1 km) and
753 both the urban measurement stations fulfil the acceptance criteria proposed by Hanna and Chang (2012) for urban dispersion
754 model evaluation. These outcomes confirmed and proved how this tool composed of two complex modelling systems (WRF-
755 Chem and PMSS) can be employed to support environmental policies, epidemiological studies and urban mobility planning.
756 Based on the performances expressed by modelled concentrations with respect observations, a further application of this tool
757 will concern the prediction of the air quality in Modena at building resolving scale based on meteorological and background
758 concentrations forecast provided by WRF-Chem and on local sources contribution estimated with the PMSS modelling suite.
759 Further work should focus also on other key species such as NO₂ and particulate matter (PM₁₀) and on integrating other in
760 situ observations to better characterize the emissions and the evolution of the PBL height during the day in the urban area of
761 Modena. For example, time-resolved and spatially-resolved CNG (compressed natural gas) consumption data can be used to
762 estimate domestic heating emissions, and also, traffic counts recorded by induction loops spires located in the main cross-
763 roads intersections could be exploited to better describe traffic emissions on the entire road network. As well as, urban
764 ceilometer observation would be necessary to correctly investigate the evolution of the mixing layer during the day.

765 **7 Acknowledgements**

766 The authors would like to thank the Municipality of Modena for providing traffic simulation data and Arpae Emilia-
767 Romagna for providing data of and information pertaining to the traffic measurement campaign.

768 **8 References**

- 769 Bigi, A., Ghermandi, G., Harrison, R. M., 2012: Analysis of the air pollution climate at a background site in the Po Valley.
770 *Journal of Environmental Monitoring*, 2012, 14, 552-563, <https://doi.org/10.1039/c1em10728c>.
- 771 Bigi, A. and Ghermandi, G., 2016: Trends and variability of atmospheric PM_{2.5} and PM_{10-2.5} concentration in the Po Valley,
772 Italy, *Atmos. Chem. Phys.*, 16, 15777-15788, <https://doi.org/10.5194/acp-16-15777-2016>.
- 773 Bigi, A., Bianchi, F., De Gennaro, G., Di Gilio, A., Fermo, P., Ghermandi, G., Prévôt, A.S.H., Urbani, M, Valli, G., Vecchi,
774 R., Piazzalunga, A., 2017: Hourly composition of gas and particle phase pollutants at a central urban background
775 site in Milan, Italy. *Atmospheric Research*, 186, 83-94, <https://doi.org/10.1016/j.atmosres.2016.10.025>.
- 776 Bellasio, R., Bianconi, R., 2012: The LAPMOD modelling system for simulating atmospheric pollution in complex
777 orography. *Ingegneria Ambientale*, vol. XLI, n. 6, 492-500.

778 Berchet, A., Zink, K., Oettl, D., Brunner, J., Emmenegger, L., and Brunner, D., 2017: Evaluation of high-resolution
779 GRAMM–GRAL (v15.12/v14.8) NO_x simulations over the city of Zürich, Switzerland. *Geosci. Model Dev.*, 10,
780 3441–3459, <https://doi.org/10.5194/gmd-10-3441-2017>.

781 Bove, M.C., Brotto, P., Cassola, F., Cuccia, E., Massabò, D., Mazzino, A., Piazzalunga, A., Prati, P., 2014: An integrated
782 PM_{2.5} source apportionment study: positive Matrix Factorisation vs. the chemical transport model CAMx. *Atmos.*
783 *Environ.* 94, 274–286, <https://doi.org/10.1016/j.atmosenv.2014.05.039>.

784 Carlino, G., Pallavidino, L., Prandi, R., Avidano, A., Matteucci, G., Ricchiuti, F., Bajardi, P., Bolognini, L., 2016: Micro-
785 scale modelling of urban air quality to forecast NO₂ critical levels in traffic hot-spots. 10th *International*
786 *Conference on Air Quality*, Milano, Italy.

787 Caserini, S., Pastorello, C., Gaifami, P., Ntziachristos, L., 2013: Impact of the dropping activity with vehicle age on air
788 pollutant emissions. *Atmospheric Pollution Research*, vol. 4, 282–289, <https://doi.org/10.5094/APR.2013.031>.

789 Chang, J. and Hanna, S., 2004: Air quality model performance evaluation. *S. Meteorol Atmos Phys*, 87: 167,
790 <https://doi.org/10.1007/s00703-003-0070-7>.

791 Chen, F., and Dudhia, J., 2001: Coupling an advanced land-surface/hydrology model with the Penn State/NCAR MM5
792 modeling system, Part I: Model description and implementation, *Mon. Weather Rev.*, 557129, 569–585,
793 [https://doi.org/10.1175/1520-0493\(2001\)129<0569:CAALSH>2.0.CO;2](https://doi.org/10.1175/1520-0493(2001)129<0569:CAALSH>2.0.CO;2).

794 Cimorelli, A.J., Perry, S. G., Venkatram, A., Weil, J. C., Paine, R. J., Wilson, R. B., Lee, R. F., Peters, W. D., and Brode, R.
795 W., 2004: AERMOD: A dispersion model for industrial source applications Part I: General model formulation and
796 boundary layer characterization. *J.Appl.Meteor.* <https://doi.org/10.1175/JAM2227.1>.

797 de Meij, A. and Gzella, A. and Cuvelier, C. and Thunis, P. and Bessagnet, B. and Vinuesa, J. F. and Menut, L. and Kelder,
798 H. M., 2009: The impact of MM5 and WRF meteorology over complex terrain on CHIMERE model calculations.
799 *Atmos. Chem. Phys.*, 9, 6611–6632, <https://doi.org/10.5194/acp-9-6611-2009>.

800 Emmons, L.K., Walters, S., Hess, P. G., Lamarque, J.-F., Pfister, G.G., Fillmore, D., Granier, C., Guenther, A., Kinnison,
801 D., Laepple, T., Orlando, J., Tie, X., Tyndall, G., Wiedinmyer, C., Baughcum, S. L., and Kloster, S., 2010:
802 Description and evaluation of the Model for Ozone and Related chemical Tracers, version 4(MOZART-4),
803 *Geosci. Model Dev.*, 3, 43–67, <https://doi.org/10.5194/gmd-3-43-2010>.

804 European Environment Agency, 2011: The application of models under the European Union's Air Quality Directive: A
805 technical reference guide, <https://doi.org/10.2800/80600> technical report 10/2011.

806 European Environment Agency, 2016. Air quality in Europe – 2016 report.

807 Ghermandi, G., Tegi, S., Fabbi, S., Bigi, A., Zaccanti, M.M., 2014: Trigeration power plant and conventional boilers:
808 pollutant flow rate and atmospheric impact of stack emissions. *Int. Journal Environ. Science and Technology*, 12
809 (2): 693-704, <https://doi.org/10.1007/s13762-013-0463-1>.

810 Ghermandi, G., Fabbi, S., Zaccanti, M. M., Bigi A., Teggi, S., 2015: Micro–scale simulation of atmospheric emissions from
811 power–plant stacks in the Po Valley. *Atmospheric Pollution Research*, 6, 382–388,
812 <https://doi.org/10.5094/APR.2015.042>.

813 Ghermandi, G., Fabbi, S., Bigi, A., Veratti, G., Despini, F., Teggi, S., Barbieri, C., Torreggiani, L., 2019: Impact assessment
814 of vehicular exhaust emissions by microscale simulation using automatic traffic flow measurements. *Atmospheric*
815 *Pollution Research*, <https://doi.org/10.1016/j.apr.2019.04.004>.

816 Gowardhan, A.A., Pardyjak, E.R., Senocak, I., Brown, M.J., 2011: A CFD based wind solver for an urban fast response
817 transport and dispersion model. *Environ. Fluid Mech.* 11, 439–464, <https://doi.org/10.1007/s10652-011-9211-6>.

818 Grell, G.A., Peckham, S.E., Schmitz, R., McKeen, S.A., Frost, G., Skamarock, W.C., and Eder, B., 2005: Fully coupled
819 “online” chemistry within the WRF model, *Atmos. Environ.*, 39, 6957–6975,
820 <https://doi.org/10.1016/j.atmosenv.2005.04.027>.

821 Grell, G.A. and Freitas, S.R., 2014: A scale and aerosol aware stochastic convective parameterization for weather and air
822 quality modeling, *Atmos. Chem. Phys.*, 14, 5233–5250, <https://doi.org/10.5194/acp-14-5233-2014>.

823 Gsella, A., de Meij, A., Kerschbaumer, A., Reimer, E., Thunis, P., Cuvelier, C., 2014: Evaluation of MM5, WRF and
824 TRAMPER meteorology over the complex terrain of the Po Valley, Italy. *Atmospheric Environment*, 89, 797–806,
825 <https://doi.org/10.1016/j.atmosenv.2014.03.019>.

826 Guenther, A.B., Jiang, X., Heald, C.L., Sakulyanontvittaya, T., Duhl, T., Emmons, L.K., and Wang, X., 2012: The Model of
827 Emissions of Gases and Aerosols from Nature version 2.1 (MEGAN2.1): an extended and updated framework for
828 modeling biogenic emissions, *Geosci. Model Dev.*, 5, 1471-1492, <https://doi.org/10.5194/gmd-5-1471-2012>.

829 Hanna, S.R., Briggs, G.A., Hosker, Jr R.P., 1982: Handbook on atmospheric diffusion. Atmospheric Turbulence and
830 Diffusion Lab, National Oceanic and Atmospheric Administration, Oak Ridge, Tennessee, USA. Tech Rep
831 DOE/TIC-1122

832 Hanna, S.R., Chang, J.C., 2012: Acceptance criteria for urban dispersion model evaluation. *Meteorological Atmospheric
833 Physics* 116, 133–146, <https://doi.org/10.1007/s00703-011-0177-1>.

834 Hausberger, S., Matzer, C., Lipp, S., Weller, K., Dippold, M., Röck, M., Rexeis, M. and Silberholz, G., 2019: Consistent
835 Emission Factors from PEMS and Chassis Dyno Tests for HBEFA 4.1; TAP Conference, Thessaloniki, Greece.
836 HBEFA, 2019, <https://www.hbefa.net/e/index.html> (last access 9 December 2019).

837 Hodzic, A. and Jimenez, J. L., 2011: Modeling anthropogenically controlled secondary organic aerosols in a megacity: a
838 simplified framework for global and climate models, *Geosci. Model Dev.*, 4, 901-917, <https://doi.org/10.5194/gmd-4-901-2011>.

840 Hong, S.-Y., 2010: A new stable boundary-layer mixing scheme and its impact on the simulated East Asia summer monsoon
841 Q. J. R. *Meteorol. Soc.*, 136 (651) (2010), pp. 1481-1496, <https://doi.org/10.1002/qj.665>.

842 Ibarra-Espinosa, S., Ynoue, R., O'Sullivan, S., Pebesma, E., Andrade, M.D.F., Osses, M., 2018: VEIN v0.2.2: an R package
843 for bottom-up vehicular emissions inventories, *Geosci. Model Dev.*, 11, 2209-2229, <https://doi.org/10.5194/gmd-11-2209-2018>.

844 INEMAR, 2013, https://www.arpae.it/dettaglio_generale.asp?id=4094&idlivello=1691(last access 2 September 2019).

846 Kaplan, H., Dinar, N., 1996: A Lagrangian dispersion model for calculating concentration distribution within a built-up
847 domain. *Atmos. Environ.* 30 (24), 4197–4207

848 Kuenen, J.J.P., Visschedijk, A.J.H., Jozwicka, M., Denier Van Der Gon, H.A.C., 2014: TNO-MACC-II emission inventory;
849 A multi-year (2003-2009) consistent high-resolution European emission inventory for air quality modelling, *Atmos.
850 Chem. Phys.*, 14, 10963-10976, <https://doi.org/10.5194/acp-14-10963-2014>.

851 Kuik, F., Lauer, A., Churkina, G., Denier van der Gon, H. A. C., Fenner, D., Mar, K. A., and Butler, T. M., 2016: Air quality
852 modelling in the Berlin–Brandenburg region using WRF-Chem v3.7.1: sensitivity to resolution of model grid and
853 input data, *Geosci. Model Dev.*, 9, 4339-4363, <https://doi.org/10.5194/gmd-9-4339-2016>.

854 Kuik, F., Kerschbaumer, A., Lauer, A., Lupascu, A., von Schneidmesser, E. and Butler, T. M., 2018: Top–down
855 quantification of NO_x emissions from traffic in an urban area using a high-resolution regional atmospheric
856 chemistry model. *Atmos. Chem. Phys.* 18, 8203-8225, <https://doi.org/10.5194/acp-18-8203-2018>.

857 Kwak, K. H., Baik, J. J., Ryu, Y. H., & Lee, S. H., 2015: Urban air quality simulation in a high-rise building area using a
858 CFD model coupled with mesoscale meteorological and chemistry-transport models. *Atmospheric Environment*,
859 100, 167-177. <https://doi.org/10.1016/j.atmosenv.2014.10.059>

860 Lenschow, P., Abraham, H., Kutzner, K., Lutz, M., Preub, J., Reichenbacher, W., 2001: Some ideas about the sources of
861 PM10. *Atmos Environ* 35, S23-S33, [https://doi.org/10.1016/S1352-2310\(01\)00122-4](https://doi.org/10.1016/S1352-2310(01)00122-4).

862 Lin, Y.-L., Farley, R. D., Orville, H. D., 1983: Bulk parameterization of the snow field in a cloud model, *J. Climate Appl.
863 Meteor.*, 22, 1065-1092, [https://doi.org/10.1175/1520-0450\(1983\)022<1065:BPOTSF>2.0.CO;2](https://doi.org/10.1175/1520-0450(1983)022<1065:BPOTSF>2.0.CO;2).

864 Loomis, D., Grosse, Y., Lauby-Secretan, B., El Ghissassi, F., Bouvard, V., Benbrahim-Tallaa, L., Guha, N., Baan, R.,
865 Mattock, H., Straif, K., 2013: The carcinogenicity of outdoor air pollution. 14, 1262-1263,
866 [https://doi.org/10.1016/S1470-2045\(13\)70487-X](https://doi.org/10.1016/S1470-2045(13)70487-X).

867 Masiol, M., Benetello, F., Harrison, R. M., Formenton, G., De Gaspari, F., Pavoni, B., 2015: Spatial, seasonal trends and
868 transboundary transport of PM2.5 inorganic ions in the Veneto region (Northeastern Italy). *Atmospheric
869 Environment*, 117, 19-31, <https://doi.org/10.1016/j.atmosenv.2015.06.044>.

870 Malm, W.C., Collett Jr., J.L., Barna, M.G., Gebhart, K.A., Schichtel, B.A., Beem, K., Carrico, C.M., Day, D.E., Hand, J.L.,
871 Kreidenweis, S.M., Lee, T., Levin, E., McDade, C., McMeeking, G.R., Molenaar, J.V., Raja, S., Rodriguez, M.A.,
872 Schwandner, F., Sullivan, A.P., Taylor, C., 2009: RoMANS Rocky Mountain Atmospheric Nitrogen and Sulfur
873 Study Report Appendix 2. MM5 Performance Evaluation and Quality Assurance Review for the White River Field
874 Office Ozone Assessment, URS Corporation, Denver.

875 Mar, K.A., Ojha, N., Pozzer, A., and Butler, T. M., 2016: Ozone air quality simulations with WRF-Chem (v3.5.1) over
876 Europe: model evaluation and chemical mechanism comparison, *Geosci. Model Dev.*, 9, 3699-3728,
877 <https://doi.org/10.5194/gmd-9-3699-2016>.

878 Mlawer, E. J., Taubaman, S. J., Brown, P. D., Iacono, M. J., and Clough, S. A., 1997: Radiative transfer for inhomogeneous
879 atmosphere: RRTM, a validate correlated-k model for the longwave, *J. Geophys. Res.*, 102(D14),16663-16682,
880 <https://doi.org/10.1029/97JD00237>.

881 Moussafir, J., Oldrini, O., Tinarelli, G., Sontowski, J., Dougherty, C., 2004: A new operational approach to deal with
882 dispersion around obstacles: the MSS (Micro-Swift-Spray) software suite. 9th International Conference on
883 Harmonisation within Atmospheric Dispersion Modelling for Regulatory Purposes Garmisch 1-4 June 2004.

884 Moussafir, J., Olry, C., Nibart, M., Albergel, A., Armand, P., Duchenne, C., Mahe, F., Thobois, L., Le Loaëc, S., Oldrini, O.,
885 2013: Aircity: a very high-resolution 3d atmospheric dispersion modeling system for Paris. Proc. 15th International
886 Conference on Harmonisation within Atmospheric Dispersion Modelling for Regulatory Purpose, Madrid, Spain,
887 May 6-9, 2013.

888 Novaes, P., Hilário, P., do Nascimento, S., Matsuda, M., Macchione, M., Peres Rangel, M., Kara-José, N., Berra, A., 2010:
889 The effects of chronic exposure to traffic derived air pollution on the ocular surface. 10, 372-374,
890 <https://doi.org/10.1016/j.envres.2010.03.003>.

891 Ntziachristos, L., Samaras, Z., 2016: EMEP/Eea Emission Inventory Guidebook; Road Transport: Passenger Cars, Light
892 Commercial Trucks, Heavy-Duty Vehicles Including Buses and Motorcycles. European Environment Agency,
893 Copenhagen.

894 Oldrini, O., Olry, C., Moussafir, J., Armand, P., Duchenne, C., 2011: Development of PMSS, the Parallel Version of Micro
895 SWIFT SPRAY. Proc. 14th Int. Conf. on Harmonisation within Atmospheric Dispersion Modelling for Regulatory
896 Purposes, 443-447.

897 Oldrini, O., Armand, P., Duchenne, C., Olry, C., Tinarelli, G., 2017. Description and preliminary validation of the PMSS fast
898 response parallel atmospheric flow and dispersion solver in complex built-up areas. *J. Environ. Fluid Mech.* 17 (3),
899 1–18, <https://doi.org/10.1007/s10652-017-9532-1>.

900 Oldrini, O., Armand, P., 2019: Validation and Sensitivity Study of the PMSS Modelling System for Puff Releases in the
901 Joint Urban 2003 Field Experiment, *P. Boundary-Layer Meteorol* 171: 513, <https://doi.org/10.1007/s10546-018-00424-1>.

903 Öttl, D., 2015: Evaluation of the revised Lagrangian particle model GRAL against wind-tunnel and field experiments in the
904 presence of obstacles. *Boundary-Layer Meteorol*, 155, 271-287, <https://doi.org/10.1007/s10546-014-9993-4>.

905 Pirovano, G., Colombi, C., Balzarini, A., Riva, G., Gianelle, V., Lonati, G., 2015: PM2.5 source apportionment in Lombardy
906 (Italy): comparison of receptor and chemistry-transport modelling results. *Atmos. Environ.* 106, 56–70,
907 <https://doi.org/10.1016/j.atmosenv.2015.01.073>.

908 PopeIII, C. A., Burnett, R.T., Thurston, G. D., Thun, M. J., Calle, E. E., Krewski, D., Godleski, J. J., 2003: Cardiovascular
909 Mortality and Long-Term Exposure to Particulate Air Pollution. *Epidemiological Evidence of General
910 Pathophysiological Pathways of Disease.* 109, 71–77, <https://doi.org/10.1161/01.CIR.0000108927.80044.7F>.

911 Röckle, R., 1990. Bestimmung der Strömungsverhältnisse im Bereich komplexer Bebauungsstrukturen. PhD dissertation,
912 (Darmstadt, Germany).

913 Rodean, H.C., 1996: Stochastic Lagrangian Models of Turbulent Diffusion, vol 45, American Meteorological Society,
914 Boston, USA.

915 Sjödin, A, Borken-Kleefeld, J., Carslaw, D., Tate, J., Alt, G.-M., De la Fuente, J., Bernard, Y., Tietge, U., McClintock, P.,
916 Gentala, R., Vescio, N., Hausberger, S., 2018: Real-driving emissions from diesel pas-senger cars measured by
917 remote sensing and as compared with PEMS and chassis dynamometer measurements -CONOX Task 2 report;
918 Commissioned by the Federal Office for the Environment (FOEN), Switzerland; ISBN: 978-91-88319-70-8.

919 Tewari, M, Kusaka, H., Chen, F., Coirier, W.J., Kim, S., Wyszogrodzki, A.A., Warner, T.T., 2010: Impact of coupling a
920 microscale computational fluid dynamics model with a mesoscale model on urban scale contaminant transport and
921 dispersion. *Atmos. Res.*, 96 , 656-664. <https://doi.org/10.1016/j.atmosres.2010.01.006>

922 Thomson, D.J., 1987. Criteria for the selection of stochastic models of particle trajectories in turbulent flows. *J. Fluid Mech.*
923 180, 529–556, <https://doi.org/10.1017/S0022112087001940>.

- 924 Thunis, P., Triacchini, G., White, L., Maffei, G., Volta, M., 2009: Air pollution and emission reductions over the Po-Valley:
925 Air quality modelling and integrated assessment. 18th World IMACS / MODSIM Congress, Cairns, Australia 13-17
926 July, p. 2009 2335-2341.
- 927 Thunis, P., 2018: On the validity of the incremental approach to estimate the impact of cities on air quality. *Atmos. Environ.*
928 173, 210-222, <https://doi.org/10.1016/j.atmosenv.2017.11.012>.
- 929 Tinarelli, G., Giostra, U., Ferrero, E., Tampieri, F., Anfossi, D., Brusasca, G., Trombetti, F., 1992: SPRAY, a 3-D particle
930 model for complex terrain dispersion", *Proc. of 10th Symposium on Turbulence and Diffusion*, American
931 Meteorological Society, Portland, Oregon (USA), 29-Sept. 2-Oct, P2.9, 147-150.
- 932 Tinarelli, G., Brusasca, G., Oldrini, O., Anfossi, D., Trini Castelli, S., Moussafir, J., 2007: Micro-SWIFT-SPRAY (MSS): a
933 new modelling system for the simulation of dispersion at microscale. General description and validation. In: *Air
934 Pollution Modeling and its Application XVII*. Springer International Publishing, pp. 449–458/449–458,
935 https://doi.org/10.1007/978-0-387-68854-1_49.
- 936 Tinarelli, G., Mortarini, L., Trini Castelli, S., Carlino, G., Moussafir, J., Olry, C., Armand, P., Anfossi, D., 2013: Review and
937 validation of Micro-Spray, a Lagrangian particle model of turbulent dispersion. *Geophysical monograph*, American
938 Geophysical Union (AGU), vol 200, pp 311–327, <https://doi.org/10.1029/2012GM001242>.
- 939 Tositti, L., Brattich, E., Masiol, M., Baldacci, D., Ceccato, D., Parmeggiani, S., Stracquadanio, M., Zappoli, S., 2014: Source
940 apportionment of particulate matter in a large city of southeastern Po Valley (Bologna, Italy). *Environ. Sci. Pollut.*
941 *Control Ser.* 21 (2), 872–890, <https://doi.org/10.1007/s11356-013-1911-7>.
- 942 Trini Castelli, S., Tinarelli, G., Reisin, T.G., 2017: Comparison of atmospheric modelling systems simulating the flow,
943 turbulence and dispersion at the microscale within obstacles. *Environ. Fluid Mech.* 17, 879–901,
944 <https://doi.org/10.1007/s10652-017-9520-5>.
- 945 Trini Castelli, S., Armand, P., Tinarelli, G., Duchenne, C., Nibart, M., 2018: Validation of a Lagrangian particle dispersion
946 model with wind tunnel and field experiments in urban environment. *Atmospheric Environment*, 193, 273-289,
947 <https://doi.org/10.1016/j.atmosenv.2018.08.045>.
- 948 Wyszogrodzki, A.A., Miao, S., Chen, F., 2012: Evaluation of the coupling between mesoscale-WRF and LES-EULAG
949 models for simulating fine-scale urban dispersion, *Atmos. Res.*, 118, 324-345.
950 <https://doi.org/10.1016/j.atmosres.2012.07.023>
- 951 Zaveri, R.A., Easter, R. C., Fast, J. D., and Peters, L. K., 2008: Model for Simulating Aerosol Interactions and Chemistry
952 (MOSAIC), *J. Geophys. Res.*, 113, D13204, <https://doi.org/10.1029/2007JD008782>.