Atmospheric Pollution Research xxx (xxxx) xxx-xxx



Contents lists available at ScienceDirect

Atmospheric Pollution Research



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

Impact assessment of vehicular exhaust emissions by microscale simulation using automatic traffic flow measurements

Grazia Ghermandi^a, Sara Fabbi^a, Alessandro Bigi^{a,*}, Giorgio Veratti^a, Francesca Despini^a, Sergio Teggi^a, Carla Barbieri^b, Luca Torreggiani^b

^a Dept. of Engineering "Enzo Ferrari", University of Modena and Reggio Emilia, Modena, Italy
^b Arpae Emilia Romagna, Italy

ARTICLE INFO	A B S T R A C T					
Keywords: Microspray Vehicular emissions Radar traffic counter Emission factors NO _x CO	In order to assess the impact of traffic on local air quality a microscale simulation of pollutant concentration fields was produced for two busy intersections, in Reggio Emilia and in Modena, Italy. The simulation was performed by the model suite Micro-Swift-Spray, a Lagrangian particle dispersion model accounting for buildings. Direct measurements of traffic flow were continuously collected in Reggio Emilia over the period January 13–24, 2014 by a two channel radar traffic counter and in Modena from October 28 to November 8, 2016 by four single channel radar traffic counters and used for the hourly modulation of vehicular emissions. Combining radar counts with vehicular fleet composition for each municipality, specific emission factors were obtained. For both cities, simulated concentration fields were compared to local air quality measurements at the nearest urban traffic and urban background sites. The simulated NO_x showed large correlation with the observations, notwithstanding some underestimation. The results proved the reliability of the procedure and provided a fair estimate of the NO_2 mass fraction of total NO_x (primary NO_2) due to vehicular emissions in the investigated traffic sites.					

1. Introduction

Road traffic is a notoriously significant source of air pollution. The pollutants emitted by vehicles are among the main causes of the degradation of air quality in urban areas. In regions where meteorological condition are unfavourable to atmospheric dispersion of the emissions, high level of pollution due to traffic emissions are detected even away from busy streets. The atmospheric monitoring of NO_{xy} main tracer of combustion emissions along with CO, provided by the local Environmental Agencies with stationary monitoring stations, clearly shows the impact of the daily traffic pattern both at kerbside sites on main urban streets and also in urban background sites.

The urban background monitoring stations should be located so that their pollution level is influenced by the integrated contribution from all sources upwind of the station. Those sampling points shall, as a general rule, be representative for several square kilometres, and the pollution level should not be dominated by a single source unless such a situation is typical for a larger urban area (Directive 2008/50/CE (Council of Europe, 2008), from which for Italy: D.Lgs. 155 - 13/08/ 2010). The vehicular emissions, however, can characterize so relevantly the air quality in the cities that peaks in traffic pollutants are detected during rush hours even in urban background stations. At urban traffic air quality stations the impact of the traffic of the adjacent street can be assumed to be superimposed on the urban background (Lenschow et al., 2001), producing higher NO_x and CO concentration values compared to urban background stations.

Within the same rationale, the regional background concentration can be attributed to all sources outside the agglomeration, i.e. natural sources and long range transport at local and global scale, with negligible influence of the sources within the agglomeration. As a consequence, a standard approach to identify the impact on urban air quality by local and distant emission sources is the comparison of the atmospheric concentration at regional, urban background and urban traffic sites (Lenschow et al., 2001).

To gain a deeper insight on the contribution by different emission sources to urban air pollution and to support environmental impact assessment studies several methods have been used: these include receptor models, Eulerian chemistry-transport models and atmospheric dispersion models. Receptor and Eulerian chemistry-transport models have been most successfully applied in source apportionment studies

https://doi.org/10.1016/j.apr.2019.04.004

Received 24 December 2018; Received in revised form 5 April 2019; Accepted 10 April 2019

1309-1042/ © 2019 Turkish National Committee for Air Pollution Research and Control. Production and hosting by Elsevier B.V.

Peer review under responsibility of Turkish National Committee for Air Pollution Research and Control.

^{*} Corresponding author.

E-mail address: alessandro.bigi@unimore.it (A. Bigi).

(Bove et al., 2014; Viana et al., 2008; Pirovano et al., 2015). Atmospheric dispersion models have been more commonly devoted to environmental impact assessments (Borrego et al., 2003; Ghermandi et al., 2014; Gariazzo et al., 2007) and their use in source apportionment is more commonly applied to long range transport episodes (Yttri et al., 2011). Nonetheless, the ability of dispersion models to employ also obstacle-resolving domains with a fine spatial resolution (e.g. featured by cells smaller than main dispersion obstacles, i.e. smaller than few metres) and to describe with good approximation the fate of atmospheric emissions makes them an effective tool for the apportionment of intra-urban emissions and for the definitions of urban air pollution control strategies in general.

These obstacle-resolving micro-scale dispersion models have been shown to provide more reliable results than local scale dispersion models for urban domains (Blocken et al., 2008) and two main mathematical approaches exist for this type of models: particle lagrangian dispersion models coupled either with a suitable meteorological model or with a 3-dimensional Computational Fluid Dynamic models. Both the former and the latter approach have been successfully applied in the literature for the investigation of fate of stack emissions (Ghermandi et al., 2015; Toja-Silva et al., 2017) and of traffic emissions (Ghermandi, Fabbi, et al., 2014; Santiago et al., 2017) within the urban environment. Main details of stack emissions may be directly measured and available at the source, while for the estimate of vehicle exhausts models need to be used (e.g. COPERT), along with details on vehicle fluxes and fleet composition. These latter data present large spatial and temporal variability over the urban area (Batterman et al., 2015), making the simulation of the dispersion of traffic emissions in urban areas a challenging tasks.

Studies employing direct measurements of vehicle number, speed and length to estimate traffic emissions successfully estimated the traffic impact on local air quality (Birmili et al., 2009; Oettl et al., 2001), however more research is needed for a better understanding of the capability and limitations of this type of dispersion models when dealing with direct traffic data, particularly for their use in intra-urban sources apportionment. This holds even more true for simulations on urban areas featured by high background concentrations and large traffic emissions.

In this study we used the micro-scale obstacle-resolving dispersion modelling chain Micro-Swift-Spray (Aria Technologies, 2010) to simulate the fate of vehicular emissions from two main intersections in two different Italian cities by coupling direct measurements of traffic count, composition and speed and local air quality monitoring. The cities are within the Po Valley (Northern Italy), a European hotspot for NO_x, characterised by recurrent wind calm episodes (Ghermandi et al., 2012) and high-pressure conditions leading to long-lasting high concentrations also at remote rural sites. Notwithstanding a long term improvement in air quality across the valley (Bigi and Ghermandi 2014, 2016), the strong anthropogenic pressure in the area, along with the characteristic climatic conditions, enhancing persistence and homogenization of air masses on a regional scale, allow emission sources to significantly impact valley-wide, including distant metropolitan areas and rural remote sites (Bigi et al., 2017; Masiol et al., 2015; Tositti et al., 2014). Within this context the first goal is to verify the suitability of the procedure used for investigating the fate of vehicular traffic emissions in urban areas, the second goal is the assessment of traffic contribution to local atmospheric pollution in terms of NO_x.

Finally, since computing resources continue to increase their performance at an accelerating pace and meanwhile are becoming much more accessible than a few years ago, there is an increasing number of studies simulating the pollutants dispersion over an entire city with two-three meters resolution. In this regard, the traffic measurement campaigns described in this study could be exploited to validate or calibrate modelled activity data used to represent traffic flows over a city road network. Indeed, assuming to use a traffic model to estimate passenger car and duty vehicle fluxes on urban roads, it's necessary to be able to rely on detailed traffic data (i.e. type and number of vehicles at each simulated hour) at specific sections of the network. Traffic measurement data collected in these two campaigns can certainly meet these needs and may lead to improved city-wide simulation, which can be a valuable tool for sound environmental policy and a reliable support for population exposure studies.

2. Experimental and methods

In the present work we performed microscale simulations of the NO_x and CO concentration fields due to exhaust emissions from road transport in two cities in central Po valley, Italy, Reggio Emilia and Modena; in both these case studies we used traffic fluxes obtained from automatic survey.

2.1. Investigated sites

Direct traffic flow measurements were carried out continuously in two cities of the Po valley, Reggio Emilia and Modena, with doppler radar traffic counters (Easy Data SDR).

In Reggio Emilia, a 171 000 inhabitants city about 60 km West of Bologna, we investigated the vicinity of a junction within the inner ring road, among the busiest road for that urban area (Baranzoni, 2017; Ghermandi et al., 2017). From January 13 to 24, 2014, a two-channel radar was positioned along this busy three-lane road, 150 hundred meters from a main junction within the inner ring road. Each radar channel detected the vehicle flow on one road lane, i.e. only two lanes were directly monitored: the vehicle flow for the third non-monitored lane was assumed equal to that measured by the radar on the adjacent lane with same running direction. This represents a reasonable assumption since both these two lanes are headed south and there are no intersections nearby forcing the vehicles to choose one of the two lanes.

In Modena, a 185 000 inhabitants city about 40 km West of Bologna, we investigated the area along the urban stretch of a main road, near a busy crossroads of the city ring road southwest of the city centre. This very busy four-lane road, 200 m from of the intersection with the urban ring road, was monitored from October 28 to November 8, 2016. Four radars were used, one for each road lane.

Hourly concentration of atmospheric NO, NO_x and CO at the two sites were provided by urban traffic stations within the regional air quality monitoring network: these stations are placed at kerbside of the roads monitored by traffic counters, both in Reggio Emilia and Modena and operated by the regional environmental agency (Arpae) (Fig. 1). Hourly background concentrations of atmospheric NO and NO_x for Reggio Emilia and Modena were also provided by Arpae urban background stations (Fig. 1). At all sites NO and NO_x are measured by Nitrogen Oxide Analyzer 200E (Teledyne-API, USA) using chemiluminescence detection principle, CO is measured by Carbon Monoxide Analyzer 300A (Teledyne-API, USA) using Non-Dispersive Infra-Red detection principle. The simulation results were compared to local air quality measurements at the above mentioned urban traffic and urban background sites.

For both cities the simulation domain of the dispersion model was sized in order to include, besides the directly monitored road, the nearby busier streets. The traffic counters directly monitored the street lanes next to the radar site: road section 1 in Reggio Emilia case, 286 m long (Fig. 1a) and road sections 1 and 2 in Modena, 355 m and 132 m long respectively (Fig. 2b). The road sections considered in the simulations as a set of linear emission sources (Fig. 1) are those directly monitored and also other sections of the main busy traffic axes included in the studied domains: in Reggio Emilia, road sections 2 and 3in Fig. 1a and 240 m and 160 m long respectively, in Modena road sections 3 (86 m), 4 (183 m) and 5 (383 m). The contribution by minor streets was then neglected.

The traffic fluxes for the street sections not directly monitored by the radars, but considered as sources in the simulations, derive from



Fig. 1. Maps of the investigation domain (UTM32-WGS84), for Reggio Emilia (a) and Modena (b) case studies. Sites of the radar traffic counter are reported by the yellow dots. Sites of the urban air quality monitoring stations are reported by the light blue (traffic stations) and green (background stations) dots respectively. The road sections (white lines) 1, 2, 3 in Reggio Emilia and 1, 2, 3, 4, 5 in Modena were considered in the simulations as a set of linear emission sources.

modelled data for rush hours provided by the Municipality of Reggio Emilia and Modena, since no direct survey was available: the hourly traffic modulations evaluated from radar measurements was applied to

Atmospheric Pollution Research xxx (xxxx) xxx-xxx

these traffic fluxes. The resulting vehicle flow budget at the nodes (the crossings) was verified.

2.2. Meteorological condition during traffic flow measurements

Concerning the Reggio Emilia case study, the measurement campaign experienced unusual weather conditions for winter in the Central Po Valley, with strong atmospheric instability and an exceptional storm rainfall on January 18 and 19. Mean wind speed during the simulation period was lower than 1 m s^{-1} , the daily average air temperature ranged from 3 to 10 °C, with larger daytime excursion (up to 9 °C) over the last four days of the period. The whole of January 2014 was characterised by heavy rainfall events and exceptionally high temperatures, the largest recording for this month over the period 2010–2017. These conditions were favorable to pollutant dispersion in the atmosphere, with pollution levels lower than usual for winter (Arpae, 2015).

In Modena the measurement campaign period was characterised by typical weather conditions for autumn in the Central Po Valley, with generally low rainfall, November 5 and 6 apart. Mean wind speed during the simulation period was lower than 2 m s^{-1} with 20% of calms (i.e. wind speed < 1 m s^{-1}), and mixing height was generally lower than 300 m; the daily average air temperature ranges from 7.4 to 13.6 °C, with large daytime excursion (maximum value 8.4 °C) and an overall decreasing trend during the measurement campaign period.

Nothing to report on traffic conditions during the measurement campaigns: in Modena the traffic was lower than usual from October 29th to November 1st, 2016 since October 29–30 were respectively Saturday and Sunday, and November 1st was National holiday.

2.3. Emission factors

The radar traffic counters recorded the time, the length and the



Fig. 2. Hourly observed concentration of NO_x at the urban traffic (red) and the urban background sites (green) along with hourly simulated concentration of NO_x (blue) by MSS from January 13 to 24, 2014 in Reggio Emilia (a) and from October 28 to November 8, 2016 in Modena (b).

speed for each passing vehicle. The radar is unable to count stationary vehicles and the minimum detected speed is 3 km/h, i.e. vehicles with speed < 3 km/h were not counted, potentially leading to an underestimation of the overall vehicular emissions. The recorded vehicles were divided in five groups according to the length L: motorcycles $(1 \text{ m} \le L \le 2.5 \text{ m})$, cars $(2.5 \text{ m} < L \le 6 \text{ m})$, light commercial vehicles $(6 \text{ m} < L \le 8 \text{ m})$, heavy vehicles $(8 \text{ m} < L \le 12 \text{ m})$ and buses $(12 \text{ m} < \text{L} \le 15 \text{ m})$. The vehicles were further sorted, according to local vehicle fleet composition, depending on the type of fuel (diesel, gasoline, LPG, methane) and the emission standard. Recorded vehicle speed was divided in 14 classes: the speed value distribution into each class was estimated, with the median value taken as representative of the corresponding class. Class medians were used to obtain emission factors (EF) for NO_x (i.e. NO and primary NO₂ (AQEG, 2007)) and CO as a function of vehicle speed, following the Tier 3 methodology defined in the European guidelines EMEP/EEA (Ntziachristos and Samaras, 2013) for the estimate of exhaust emissions from road transport. In the Tier 3 approach, total exhaust emissions from road transport are calculated as the sum of the hot emissions (when the engine is at normal operating temperature) and the emissions during the operation of the transient thermal engine (called "cold" emissions). In this study only hot emissions were considered. The Tier 3 emission factors were derived from several studies depending on the fuel, emission reduction technology and vehicle class (André, 2004; Eggleston et al., 1993; Ntziachristos and Samaras, 2000).

The NO_x EF from European guidelines is provided as NO₂ equivalent (NO_{2eq}): consequently, both simulated and measured NO_x concentrations in the present study are given as NO_{2eq}.

Finally EF values were mathematically weighted to obtain a single EF for each group of vehicles and for each pollutant (Table 1); the accuracy of the calculation of the weighted EF values depends on the availability of supporting data.

The calculation was most accurate for passenger cars (corresponding to about 81% of all recorded vehicles in Reggio Emilia and about 61% in Modena case studies), given the availability of detailed vehicle fleet composition data provided by Italian Automobile Club for the year 2013 in Reggio Emilia (ACI, 2013) and for the year 2015 in Modena (UCER, 2016), including also fuel type and emission standards. For motorcycles and light commercial vehicles, the average value of EF for the different emission standard and for the fuel type (mainly diesel for light commercial vehicles) were used.

The EF for heavy vehicles and for buses (that together amount to about 1% of all the recorded vehicles) were evaluated using the "Rigid 20–26 t" and the "Urban Buses Standard 15–18 t" guideline formulas respectively (assuming diesel as fuel type and the local fleet compositions for EURO emission standard), which are less detailed for heavy vehicles and buses than for passenger cars.

For mopeds (motorcycles with engine capacity $< 50 \, \text{cm}^3$) the guidelines directly provide the EFs.

Coupling the hourly radar records with the EF value for each counted vehicle, the hourly mass flows of CO and NO_x emitted for the whole road length were estimated: the modulated traffic emissions according to the hourly variation of traffic fluxes were thus obtained for each day of the measurement campaigns.

Atmospheric Pollution Research xxx (xxxx) xxx-xxx

2.4. Models

The Micro-Swift-Spray (MSS) simulation domain is $500 \text{ m} \times 500 \text{ m}$ large (Fig. 1) with grid step of 2 m (square cells) for both case studies. The vertical grid consists of 5 layers, 2 m deep each, with the domain top of 10 m high above ground level: simulation with higher domain tops were performed and showed negligible differences therefore the domain top was to 10 m to reduce computation time. The first layer for concentration computing is 2 m high above ground. The second layer concentrations were used in the comparison with observations, because the inlet of air quality monitoring instruments by ARPAE was at about 4 m height above ground level. Building volumes and road geometry were outlined from a high resolution 3D vectorial cartography (UVL_GPG) of the studied domains (E.R. 2013).

Following the model by Berkowicz et al. (1997), for the studied case the height of the traffic induced turbulence ranges between 5 and 9 m above ground level, while its width ranges between 14 and 27 m around the road axis, depending on the case study, i.e. on average vehicle size and speed, vehicular flux, on road width (higher limits for Modena); these values were used for MSS simulation and applied to all the road sections considered as linear emission sources (Fig. 1).

The simulations were run at an hourly time step, consistently with the meteorological data. The concentration values were all standardized for atmospheric temperature and pressure and all NO_x data in text are provided as NO_{2eq} . MSS results consist in hourly 3D concentration fields for atmospheric NO_x and CO, due to vehicular emission contribution only. The time series of hourly simulated concentrations for the duration of the measurement campaign was compared with measured concentration values collected at the respective air quality monitoring stations in urban traffic and urban background conditions (Fig. 1).

2.5. Meteorological data and modelling

The hourly meteorological data, mixing height values and turbulence parameters (i.e. friction velocity, convective velocity scale and Monin-Obukhov length) used, were derived from meteorological model simulations and from ground observations. The latter were collected at the urban meteorological stations of Arpae for Reggio Emilia and Modena and these are placed outside from the respective MSS simulation domains.

The simulation data, provided by the local Environmental Agency, proceed from two independent simulations, at a horizontal grid step of 5 km and 7 km respectively: CALMET-SIM (Deserti et al., 2001), based on the CALMET model, which was used for Reggio Emilia, and LAMA (Limited Area Meteorological Analysis), based on the COSMO model (Marsigli et al., 2005; Montani et al., 2011), which was used for Modena. It is worth noting that the urban meteorological stations for the two towns were not used among input data for the meteorological simulations by CALMET and COSMO.

3. Results and discussion

According to the rationale of Lenschow et al. (2001), the concentration measured at the traffic site can be assumed to be the sum of local traffic emissions and the emissions within the whole

Table 1

Emission Factors EF (g/km) weighted values. LDV stands for Light Duty Vehicles, HDV stands for Heavy Duty Vehicles.

Pollutant	Passenger cars		Motorcycles		LDV		HDV		Buses	
	RE	МО	RE	МО	RE	МО	RE	МО	RE	MO
NO _x CO	0.40 0.75	0.36 0.67	0.09 8.61	0.09 8.74	1.00 0.79	0.84 0.54	11.48 2.58	8.63 1.94	8.19 2.33	7.77 1.83

agglomeration, with the latter corresponding to the urban background. This assumption is reasonable for NO_x and CO in the two domains since the main emission source next to the urban traffic sites is only vehicular traffic and there is no other significant emission source nearby, i.e. these monitoring stations are compliant with 2008/50/CE (Council of Europe, 2008). Therefore the MMS simulated concentrations represent only the contribution by primary traffic emission to atmospheric concentrations.

In the recent literature an alternative approach to quantify the different contributions to air pollution in the city was proposed by Thunis (2017), and this is based on Chemical Transport Models (CTMs). Unlike the Lenschow incremental approach, this methodology suggests to create different emission scenarios on multi spatial scale, from regional to local, that can be exploited to estimate background concentrations keeping city emissions set to zero. Despite the employment of this type of modelling system can theoretically lead to a more accurate discrimination between city and background contribution on air quality, in the two case studies presented can raise complexities and more uncertainties than a combined measured-modelled approach.

The extension of the PMSS domains is particularly limited ($500 \text{ m} \times 500 \text{ m}$) and challenging for CTMs and for zeroing traffic emissions in a domain of this size (Kuik et al., 2016).

Besides that having CTMs with such a fine grid would require several nested domains, and the obtainable benefits from this procedure are not always straightforward, especially when model resolution is less than 12-10 km (Mass et al., 2002), e.g. some local meteorological phenomena might not be well described affecting the performance of the chemical mechanism.

Finally, the development of a hybrid modelling system composed of a CTM and a micro-scale model may not provide remarkable improvements with respect a CTM stand-alone due to the lack of ability of this type of system to correctly take into account some multi-scale features (e.g. local meteorological phenomena, high resolution emissions and detailed land cover data) (Pepe et al., 2016).

MMS Model performance was estimated by comparing observed concentrations at the urban traffic site and an interpolation of the simulated values in the 4 cells of domain closest to the inlet of the monitoring instruments. These latter instruments measured atmospheric CO, NO and NO_x concentrations, for which the main regulatory limits are: $200 \,\mu g \, m^{-3}$ for hourly maximum NO₂ and $10 \, m g \, m^{-3}$ for 8-h CO running average. Atmospheric NO₂ concentration was estimated from direct measurements of NO and NO_x.

The hourly time series of observed NO_x concentrations at the urban traffic and urban background stations, along with the simulated NO_x levels for both cities, are presented in Fig. 2.

The comparison between measured and simulated concentrations is better for NO_x than for CO (Fig. 3). This outcome depends on the low sensitivity of the CO monitoring instrument, and on the lack of CO observations at urban background sites, preventing the possibility to repeat the same data processing performed for NO_x and described below.

The NO_x traffic and urban background measured concentrations show a very similar pattern (Pearson coefficient r = 0.80 for Reggio Emilia and r = 0.84 for Modena case). This mostly depends on the Po Valley meteorological regime, mainly influenced by the valley morphological conformation and characterised by recurrent wind calm episodes, occurring also during the measurement campaigns. This condition determinates accumulation and persistence of the pollutant load, therefore also at the urban background site the air quality is clearly affected by the diurnal variability of the main pollutant sources. It is mainly evident for the Modena case, because in this city the measurement campaign was featured by a longer period of atmospheric stability compared to Reggio Emilia, occasionally leading to higher concentrations at the urban background than at the urban traffic site.

For both Modena and Reggio Emilia the simulated NO_x remains constantly lower than the measured concentrations, both at the urban

traffic and urban background stations.

The correlation between simulated and measured CO at the traffic site is moderate for Reggio Emilia (r = 0.42) and quite low for Modena (r = 0.12), mainly due to the low sensitivity of CO measuring instruments, which are most suitable for controlling the exceedance of the regulatory limits for CO concentration in the atmosphere. CO simulated data remain always largely lower than the measured concentrations (Fig. 3).

Given that CO data are not collected at urban background stations, the following data processing was performed for NO_x only.

The difference between NO_x observations at urban traffic and at urban background stations for the same city (hereafter Δ NO_x) can be attributed to the influence of local traffic, and this contribution was estimated by MSS simulation. Outliers in Δ NO_x series were considered not representative of episodic peak concentration, but of the statistical fluctuations intrinsic in the simulation model, therefore outliers were removed by applying a running median with a 3-h window. Goodness of fit indexes results better in Reggio Emilia than Modena: linear correlation coefficient (*r*) and Mean Absolute Error (MAE) between Δ NO_x and MSS simulated values result *r* = 0.56, MAE = 23.5 µg m⁻³ and *r* = 0.43, MAE = 20.7 µg m⁻³ for Reggio Emilia and Modena respectively. The MAE is 54% of mean Δ NO_x for Reggio Emilia and up to 86% of mean Δ NO_x for Modena. This not surprising outcome is partly due to the difference between the smooth variability of the simulated concentration and the large variability observed at the monitoring sites.

In spite of that, in Fig. 4 is reported the more effective comparison between the hourly NO_x concentrations measured at traffic stations and the sum of NO_x hourly simulated concentrations with urban background observations. These two time series result highly correlated (r = 0.84 for Reggio Emilia and r = 0.89 for Modena). This outcome highlights both the relevant contribution of urban background levels to the traffic-dominated environments, as previously mentioned, both the benefit of using direct measurements of traffic in the EF estimate.

However the vehicular emission contribution to air quality at the traffic site, as evaluated by MMS simulation, is generally underestimated: in fact urban traffic measured concentrations result mainly higher than the sum of MMS simulated concentration and urban background concentrations (Fig. 4).

MMS simulated NO_x concentration, indicating the contribution of traffic emissions to atmospheric levels, is lower of about 30% for Reggio Emilia and 56% for Modena case than ΔNO_x . Given that the NO_x traffic emissions used in MSS include only (primary) NO and primary NO₂ (Air Quality Expert Group, 2007), it was investigated whether the difference between simulated and observed levels are due to secondary NO₂ only, which are not described in the MSS simulation, or by an underestimation of the primary NOx by MSS.

In investigating the cause of this difference, it was preliminary assumed that it was originated only by an underestimation of primary NO_x by MSS. The correctness of this assumption was verified by comparing the $\Delta NO_2/\Delta NO_x$ ratio (with ΔNO_2 being the difference in NO₂ observations at the urban traffic and urban background sites for the same city and ΔNO_x defined as above), with the primary NO₂/NO_x ratio in the total vehicular emissions (Grice et al., 2007; Smit et al., 2010) for the local vehicular fleet (Ntziachristos and Samaras, 2013), i.e. the same fleet used in the estimate of the EF. This latter NO₂/NO_x ratio resulted 16.5% and 15.0% for Reggio Emilia and Modena respectively; the ratios are largely similar because of the similarity in their fleet composition. The $\Delta NO_2/\Delta NO_x$ ratios were 15.7% and 41.7% for Reggio Emilia and Modena respectively.

The comparability of these ratios in the emissions and in the atmospheric concentrations in Reggio Emilia hints to the occurrence of two conditions: 1. There is no extra secondary NO₂ at the urban traffic compared to urban background 2. The higher NO_x concentration observed at the traffic site are due mainly to traffic, i.e. the immediate impact of traffic at the urban background site is low.

On the contrary, for the Modena case the larger $\Delta NO_2/\Delta NO_x$



Fig. 3. Hourly observed concentration of CO at the urban traffic site (red) along with hourly simulated concentration of CO (blue) by MSS from January 13 to 24, 2014 in Reggio Emilia (a) and from October 28 to November 8, 2016 in Modena (b).

compared to the emission data suggests similar pollution conditions between the urban traffic and urban background site of this town, with the larger concentration observed at the urban traffic site sharing the same source of the concentration observed at the urban background site. It is noteworthy that NO₂/NO_x ratio for the observations at the Modena urban background site is 41.8%, i.e. the same of Δ NO₂/ Δ NO_x. This latter point supports the hypothesis that the urban traffic sites in

Modena experiences the same mix of emission sources of the urban background site, although with a stronger absolute emission, since the local meteorology, favoring air masses stagnation and homogenization, determines in the whole urban area the same pollution *facies*.

A summary statistical analysis was performed on the residuals, i.e. the differences between the MSS simulated NO_x concentrations and ΔNO_x . For both the case studies, the frequency density of the residuals



Fig. 4. Hourly observed concentration of NOx at the urban traffic (red) along with the sum of urban background observations of NOx with simulated concentration by MMS (black) from January 13 to 24, 2014 in Reggio Emilia (a) and from October 28 to November 8, 2016 in Modena (b). Note: the black curve in this figure corresponds to the sum of green and blue curves of Fig. 2.

Atmospheric Pollution Research xxx (xxxx) xxx-xxx



Fig. 5. Frequency density of the residuals, i.e. the differences between the MSS simulated concentrations and ΔNO_x , for Reggio Emilia (a) and Modena (c). Hourly box plots of the residuals for Reggio Emilia (b) and Modena (d).

shows that 90% are in the range \pm 50 µg m⁻³, with a larger fraction of negative values (Fig. 5); a relatively larger number of positive residuals occur for the Modena case. The hourly box plot of the residuals (Fig. 5b) indicates a fairly stable median for both the case studies, with minimum values most frequently occurring during atmospheric stability (i.e. at nighttime) and maximum atmospheric mixing (i.e. between 12:00–16:00). Larger variability, with occasional outliers is more commonly observed in other periods of the day. Within the limits of the residuals population tested, it can be assumed that the performance of the model does not vary systematically on a diurnal basis. The residual analysis support the hypothesis of a larger effectiveness of the model in simulating during maximum mixing height or night stability conditions than during stages of mixed layer erosion or development.

Uncertainties of the whole described procedure apart, the underestimation of MMS concentrations can be also attributed to the following main causes:

- only emissions by traffic along main roads were considered, while the contribution by minor streets was neglected
- the radar inability to count stationary vehicles, for example in case of vehicle queues on the street lanes (that may occur not too distant from the traffic lights) or bus pulling out to the bus stop, as in the case of Modena;
- underestimation of the emissions due to an overestimation of vehicle speed: given the radar inability to count stationary and very low speed-vehicles, as mentioned above, some lowest speed values

G. Ghermandi, et al.

- weather conditions, typical for autumn season in the Central Po Valley, featured by low rainfall and accumulation of the pollutant load, causing the large contribution of secondary NO_2 at the urban traffic site in Modena case

Occasionally, local and short meteorological events were not simulated by the meteorological model, leading to an additional mismatch between simulated and observed concentrations. This is the case of an anomalous night-time peak in NO_x and CO measured on January 16, 2014 at 01:00, at the urban traffic and urban background stations both in Reggio Emilia (Figs. 2 and 3) and in Modena (not shown). The small scale of this occurrence is supported by the concurrently low values of NO_x and CO at the neighboring urban sites of Bologna (35 km East of Modena) and Parma (38 km West of Reggio Emilia).

MMS simulated concentrations in Reggio Emilia for January 16, 2014 at 01:00 and nearest hours are low, consistently with the low traffic at that time of the night, as recorded by the radar (Figs. 2 and 3).

This pollution episode was probably generated by a meteorological event constricting low and polluted air masses towards the ground, as for the local evolution of a cold air front (Li et al., 2015). The model failed in the simulation of this episode either because CALMET (from which the hourly meteorological data set used for Reggio Emilia case is derived) is not able to simulate this type of meteorological events, featured by that small space-time scale, or because none of the meteorological stations from Reggio Emilia and Modena were used as input for CALMET.

4. Conclusions

The outcomes of the study support the reliability of the simulations by the micro-scale obstacle-resolving lagrangian dispersion model Micro-Swift-Spray when coupled with tailored emitting sources, i.e. the direct measurements of traffic flows and the accurate evaluation of pollutant Emission Factors, leading to a consistent estimate of the contribution to air quality by vehicular traffic. This represents a methodology useful tool for the validation and calibration of traffic emissions in large-domain models (e.g. city-wide). This latter type of studies, becoming increasingly common, is often used for planning municipal environmental policies and assessing personal exposure and therefore requires a reliable estimate of emissions.

Simulated and observed NO_x hourly concentrations exhibit a large agreement: a similar pattern is shown by NO_x observations under urban traffic and urban background conditions, however this high correlation is further improved by superimposing the simulated concentrations over the urban background observations, confirming the effectiveness of the methods applied in this study.

The results also highlight the effect of homogenization of the air across the urban areas for this part of the Po valley. This leads to relevant contributions by urban background NO_x to the urban traffic, and, *vice-versa*, to a significant impact by primary sources also at the urban background.

The homogenization of the air is mostly evident for Modena: similar pollution conditions occurred between the urban traffic and urban background, with the measurements at the latter being sometime higher than at the former. The urban traffic and urban background sites of Modena shared similar NO_x sources and have similar NO₂/NO_x ratios, confirming the large contribution of secondary NO₂ also at the traffic dominated site.

The cause for the underestimation of simulated traffic contribution to atmospheric NO_x is threefold: the presence of only the main roads as source emissions, neglecting minor streets; the undercount by the radars, which consider only moving vehicles, neglecting traffic jams conditions; the effect of the local meteorological condition effects, especially for Modena.

An analysis of the NO₂/NO_x ratio showed how in the Reggio Emilia

case the higher levels observed at the urban traffic site were mainly due to nearby traffic emissions. This latter outcome outlines the representativeness of the air quality stations for Reggio Emilia in characterizing the two sites for their pollution level and dominant impacting sources.

The results obtained for CO are less significant, as CO monitoring is performed only at traffic site and also due to the low sensitivity of CO measuring instruments.

Finally the results outline also the effects of meteorological input data at a local scale on the microscale simulation of the dispersion and they highlight how local and short meteorological events were not simulated by the meteorological model. This latter shortcoming might become a critical issue, particularly at temperate latitudes, in case of an increase in frequency of intense and local weather events, as expected in climate change (Ciscar et al., 2018).

References

- ACI. Available at: http://www.aci.it/laci/studi-e-ricerche/dati-e-statistiche/veicoli-emobilita.html.
- Air Quality Expert Group, 2007. Trends in Primary Nitrogen Dioxide in the UK. Department for the Environment, Food and Rural Affairs.
- André, M., 2004. The ARTEMIS European driving cycles for measuring car pollutant emissions. Sci. Total Environ. 334–335, 73–84.
- Aria Technologies, 2010. SWIFT Wind Field Model. General Design Manual.
- Arpae, 2015. Monthly Synthetic Report on Air Qualiy (In Italian) ARPAE Available at: https://www.Arpae.it/cms3/documenti/_cerca_doc/aria/aria_re/201401.pdf, Accessed date: 17 December 2018.
- Baranzoni, G., 2017. Microscale Atmospheric Dispersion of Vehicular Emissions: Automatic Count of Traffic Flow and Comparison with Air Quality Measurements. Dept. of Engineering "Enzo Ferrari", University of Modena and Reggio Emilia (in Italian).
- Batterman, S., Cook, R., Justin, T., 2015. Temporal variation of traffic on highways and the development of accurate temporal allocation factors for air pollution analyses. Atmos. Environ. 107, 351–363.
- Berkowicz, R., et al., 1997. Modelling Traffic Pollution in Streets. National Environmental Research Institute, Roskilde.
- Bigi, A., et al., 2017. Hourly composition of gas and particle phase pollutants at a central urban background site in Milan, Italy. Atmos. Res. 186, 83–94.
- Bigi, A., Ghermandi, G., 2014. Long-term trend and variability of atmospheric PM₁₀ concentration in the Po Valley. Atmos. Chem. Phys. 14 (10), 4895–4907.
- Bigi, A., Ghermandi, G., 2016. Trends and variability of atmospheric PM_{2.5} and PM_{10-2.5} concentration in the Po Valley, Italy. Atmos. Chem. Phys. 16 (24), 15777–15788.
- Birmili, W., et al., 2009. Dispersion of traffic-related exhaust particles near the Berlin urban motorway – estimation of fleet emission factors. Atmos. Chem. Phys. 9 (7), 2355–2374.
- Blocken, B., et al., 2008. Numerical evaluation of pollutant dispersion in the built environment: comparisons between models and experiments. J. Wind Eng. Ind. Aerod. 96 (10), 1817–1831.
- Borrego, C., et al., 2003. Emission and dispersion modelling of Lisbon air quality at local scale. Atmos. Environ. 37 (37), 5197–5205.
- Bove, M.C., et al., 2014. An integrated PM2.5 source apportionment study: positive Matrix Factorisation vs. the chemical transport model CAMx. Atmos. Environ. 94, 274–286.
- Ciscar, J.C., Ibarreta, D., Soria, A., Dosio, A., Toreti, A., Ceglar, A., Fumagalli, D., Dentener, F., Lecerf, R., Zucchini, A., Panarello, L., Niemeyer, S., Pérez-Domínguez, I., Fellmann, T., Kitous, A., Després, J., Christodoulou, A., Demirel, H., Alfieri, L., Dottori, F., Vousdoukas, M.I., Mentaschi, L., Voukouvalas, E., Cammalleri, C., Barbosa, P., Micale, F., Vogt, J.V., Barredo, J.I., Caudullo, G., Mauri, A., de Rigo, D., Libertà, G., Durrant, T.H., Vivancos, T.A., Miguel-Ayanz, J.S., Gosling, S.N., Zaherpour, J., Roo, A.D., Bisselink, B., Bernhard, J., Bianchi, L., Rozsai, M., Szewczyk, W., Mongelli, I., Feyen, L., 2018. Climate Impacts in Europe: Final Report of the JRC PESETA III Project. https://doi.org/10.2760/93257, JRC112769.
- Council of Europe, 2008. Directive 2008/50/EC of the European Parliament and of the Council of 21 May 2008 on Ambient Air Quality and Cleaner Air for Europe. Official Journal of the European Union, Official Journal of the European Union, pp. L152/ 1–L152/144.
- Deserti, M., et al., 2001. Operational meteorological pre-processing at Emilia- Romagna ARPA Meteorological Service as a part of a decision support system for air quality management. Int. J. Environ. Pollut. 16 (1–6), 571–582.
- Eggleston, H., Gaudioso, D., Gorissen, N., Joumard, R., Rijkeboer, R., Samaras, Z., Zierock, K.-H., 1993. CORINAIR Working Group on Emission Factors for Calculating 1990 Emissions from Road Traffic: Volume 1 : Methodology and Emission Factors.
- E.R, 2013. Geoportale Emilia-Romagna (Database Topografico 2013). Available at: url: https://geoportale.regione.emilia-romagna.it/it/download/databasetopografico, Accessed date: 17 December 2018.
- Gariazzo, C., et al., 2007. Application of a Lagrangian particle model to assess the impact of harbour, industrial and urban activities on air quality in the Taranto area, Italy. Atmos. Environ. 41 (30), 6432–6444.
- Ghermandi, G., et al., 2012. Model comparison in simulating the atmospheric dispersion

of a pollutant plume in low wind conditions. Int. J. Environ. Pollut. 48 (1–4), 69–77. Ghermandi, G., et al., 2015. Micro–scale simulation of atmospheric emissions from power–plant stacks in the Po Valley. Atmos. Pollut. Res. 6 (3), 382–388.

- Ghermandi, G., et al., 2017. Vehicular exhaust impact simulated at microscale from traffic flow automatic surveys and emission factor evaluation. In: HARMO 2017: 18th Int. Con. on Harmonization within Atmospheric Dispersion Modelling for Regulatory Purposes. Bologna, Italy, pp. 475–479.
- Ghermandi, G., Teggi, S., et al., 2014. Tri-generation power plant and conventional boilers: pollutant flow rate and atmospheric impact of stack emissions. Int. J. Environ. Sci. Technol. 12 (2), 693–704.
- Ghermandi, G., Fabbi, S., et al., 2014. Urban micro-scale investigation of NOx and CO emissions from vehicular traffic and comparison with air quality data. In: HARMO16: 16th Int. Con. on Harmonization within Atmospheric Dispersion Modelling for Regulatory Purposes. Varna, Bulgaria.
- Grice, S., et al., 2007. The Impact of Changes in Vehicle Fleet Composition and Exhaust Treatment Technology on the Attainment of the Ambient Air Quality Limit Value for Nitrogen Dioxide in 2010. AEA Energy & Environment.
- Kuik, F., et al., 2016. Air quality modelling in the Berlin–Brandenburg region using WRF-Chem v3.7.1: sensitivity to resolution of model grid and input data. Geosci. Model Dev. (GMD) 9 (12), 4339–4363.
- Lenschow, P., et al., 2001. Some ideas about the sources of PM10. Atmos. Environ. 35, S23–S33.
- Li, X., et al., 2015. The role of foehn in the formation of heavy air pollution events in Urumqi, China. J. Geophys. Res.: Atmos. 120 (11), 5371–5384.
- Marsigli, C., et al., 2005. The COSMO-LEPS mesoscale ensemble system: validation of the methodology and verification. Nonlinear Process Geophys. 12 (4), 527–536.
- Masiol, M., et al., 2015. Spatial, seasonal trends and transboundary transport of PM2.5 inorganic ions in the Veneto region (Northeastern Italy). Atmos. Environ. 117, 19–31.
- Mass, C.F., et al., 2002. Does increasing horizontal resolution produce more skillful forecasts? The results of two years of real-time numerical weather prediction over the Pacific Northwest. Bull. Am. Meteorol. Soc. 83 (3), 407–430+341.
- Montani, A., et al., 2011. Seven years of activity in the field of mesoscale ensemble forecasting by the COSMO-LEPS system: main achievements and open challenges. Tellus 63 (3), 605–624.

Atmospheric Pollution Research xxx (xxxx) xxx-xxx

Ntziachristos, L., Samaras, Z., 2000. Speed-dependent representative emission factors for catalyst passenger cars and influencing parameters. Atmos. Environ. 34, 4611–4619.

- Ntziachristos, L., Samaras, Z., 2013. EMEP/EEA Emission Inventory Guidebook 2013: Exhaust Emissions from Road Transport. [Internet] EEA, Copenhagen updated 2014. Available at: https://www.eea.europa.eu/publications/emep-eea-guidebook-2013/ part-b-sectoral-guidance-chapters/1-energy/1-a-combustion/1-a-3-b-road-transport/ view.
- Oettl, D., et al., 2001. Evaluation of a Gaussian and a Lagrangian model against a roadside data set, with emphasis on low wind speed conditions. Atmos. Environ. 35 (12), 2123–2132.
- Pepe, N., et al., 2016. Development and application of a high resolution hybrid modelling system for the evaluation of urban air quality. Atmos. Environ. 141, 297–311.
- Pirovano, G., et al., 2015. PM2.5 source apportionment in Lombardy (Italy): comparison of receptor and chemistry-transport modelling results. Atmos. Environ. 106, 56–70.
- Santiago, J.-L., et al., 2017. The impact of planting trees on NOx concentrations: the case of the plaza de la Cruz neighborhood in pamplona (Spain). Atmosphere 8 (7). Smit, R., Ntziachristos, L., Boulter, P., 2010. Validation of road vehicle and traffic
- emission models a review and meta-analysis. Atmos. Environ. 44 (25), 2943–2953. Thunis, P., 2017. On the validity of the incremental approach to estimate the impact of
- cities on air quality. Atmos. Environ. 173, 210–222. Toja-Silva, F., et al., 2017. CFD simulation of CO2 dispersion from urban thermal power plant: analysis of turbulent Schmidt number and comparison with Gaussian plume
- model and measurements. J. Wind Eng. Ind. Aerod. 169, 177-193. Tositti, L., et al., 2014. Source apportionment of particulate matter in a large city of
- southeastern Po Valley (Bologna, Italy). Environ. Sci. Pollut. Control Ser. 21 (2), 872–890.
- UCER, 2016. Report by the Union of Regional Chambres of Commerce. Available at: https://www.ucer.camcom.it/studi-ricerche/dati/bd/appendice-dati-comunali/ trasporti-stradali.-consistenza-dei-veicoli, Accessed date: 17 December 2018.
- Viana, M., et al., 2008. Source apportionment of particulate matter in Europe: a review of methods and results. J. Aerosol Sci. 39 (10), 827–849.
- Yttri, K.E., et al., 2011. Source apportionment of the summer time carbonaceous aerosol at Nordic rural background sites. Atmos. Chem. Phys. 11 (24), 13339–13357.