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ESTIMATE OF SECONDARY NO_2 LEVELS AT AN URBAN TRAFFIC SITE BY MICROSCALE SIMULATION OF TRAFFIC EMISSIONS

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Abstract: In this study we investigated the share of secondary NO_2 in intense traffic areas in Modena and Reggio Emilia (Northern Italy) by the analysis of regulatory air quality data in urban traffic and urban background conditions, and by the simulation of the dispersion of NO_x traffic emissions by the lagrangian particle dispersion models PMSS (Parallel-Micro-SWIFT-SPRAY). In both cities, the simulation focuses on a subset of the urban area having intense traffic and including a urban traffic air quality monitoring site, during the period 13 to 24 January 2014 in Reggio Emilia and 28 October – 8 November 2016 in Modena. An increment of pollutants from rural background to urban hotspots was assumed in order to estimate primary and secondary NO_2 at the urban traffic site. The mean ratio of the primary NO_2/NO_x was compared to the mean ratio of observed NO_2 and NO_x concentrations in order to assess the contribution of local secondary NO_2 to the atmospheric levels at the traffic site. Results show the large amount of secondary NO_2 mainly at the Modena traffic site.

Key words: *NO_2, NO_x, traffic emissions, dispersion models.*

INTRODUCTION

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 at large distance from busy streets, as clearly shown by the atmospheric concentration values of the main tracers of combustion emissions, like NO_x, monitored by the local Environmental Agencies at urban traffic and urban background sites.

The urban traffic stations, placed at kerbside sites on main urban streets, are directly influenced by local traffic, while at background monitoring stations the pollution level should be influenced by the integrated contribution from all sources upwind of the station (Directive 2008/50/CE, recieved in Italy by the D.Lgs. 155 - 13/08/2010). The vehicular emissions, however, can characterize so relevantly urban air quality that peaks in traffic pollutants are detected during rush hours even in urban background stations. At urban traffic air quality stations the local influence of traffic on the adjacent street can be assumed to be superimposed on the urban background (Lenschow et al. 2001), producing higher NO_x concentration values compared to urban background stations. Moreover, the regional background concentration can be attributed to natural sources and long range transport at local and global scale, with negligible influence of nearby sources. Therefore, according to the Lenschow et al. rationale (2001), to identify the impact on

urban air quality by local and distant emission sources, the atmospheric concentration at regional, urban background and urban traffic sites must be compared.
To investigate the contribution by different emission sources to urban air pollution several methods have been used among which the atmospheric dispersion models, more commonly devoted to environmental impact assessments (Ghermandi et al. 2012; Ghermandi et al. 2014; Gariazzo et al. 2007). The ability of dispersion models to employ also obstacle-resolving domains with a fine spatial resolution (at micro scale, i.e. cells smaller than few metres) (Ghermandi et al. 2015) makes them an effective tool to describe the fate of intra-urban atmospheric emissions and to support urban air pollution control strategies.
In this study a single micro scale model combined with observations was used (Ghermandi et al. 2017) to simulate the atmospheric dispersion of NO_x vehicular emissions (i.e. NO and primary NO_2) in urban areas having intense traffic, in Modena and Reggio Emilia (Northern Italy); 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 (Bigi and Ghermandi 2014; Bigi and Ghermandi 2016; Bigi et al. 2017).
The NO_x simulated concentrations and the difference in NO_2 and NO_x between the regulatory air quality data collected at urban traffic and urban background stations, in Modena and Reggio Emilia, are here used to estimate primary and secondary NO_2 at the urban traffic site.

EXPERIMENTAL SET UP AND METHODS

Direct traffic flow measurements were carried out continuously in two cities of the Po valley, Modena, 185 000 inhabitants, about 40 km West of Bologna, and Reggio Emilia, 171 000 inhabitants, about 60 km West of Bologna, with doppler radar traffic counters (Easy Data SDR).
In Modena the area along the urban stretch of a main road, near a busy crossroads of the city ring road southwest of the city centre was monitored from October 28 to November 8, 2016; in Reggio Emilia, the vicinity of a junction within the inner ring road, among the busiest road for that urban area had been monitored from January 13 to 24, 2014 (Ghermandi et al. 2017). During both measurement campaigns, hourly concentration of atmospheric NO and NO_x were provided by the local urban traffic station, placed at the kerbside of the roads monitored by traffic counters, and by the urban background station, all of them within the regional air quality monitoring network operated by the regional environmental agency (Arpae) (Figure 1). At all sites NO and NO_x are measured by Nitrogen Oxide Analyzer 200E (Teledyne-API, USA) using chemiluminescence detection principle.
For both case studies, the simulation domain of the dispersion model was sized in order to include the nearby busier streets (Figure. 1), besides the directly monitored road lanes. All these road sections were considered in the simulations as a set of linear emission sources. The contribution by minor streets was neglected. The traffic fluxes for the street sections not directly monitored by the radars, but considered in the simulation, derive from modelled data for rush hours provided by the Municipality of Modena and Reggio Emilia. The hourly traffic modulations evaluated from radar measurements was applied to these traffic fluxes.
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. Mean wind speed during the simulation period was lower than 1 $m \cdot s^{-1}$, the daily average air temperature ranged from 3 to 10 °C, with larger daytime excursion. The whole of January 2014 was characterized by heavy rainfall events and exceptionally high temperatures, the largest recording for this month over the period 2010 - 2017. These conditions were favourable to pollutant dispersion in atmosphere.
In Modena the measurement campaign period was characterized by typical weather conditions for autumn in the Central Po Valley, with generally low rainfall. Mean wind speed during the simulation period was lower than 2 $m \cdot s^{-1}$ with 20% of calms (i.e. wind speed < 1 $m \cdot s^{-1}$), and the mixing height was generally lower than 300 m; the daily average air temperature ranged from 7.4 to 13.6 °C. The traffic was less intense than usual from October 29th to November 1st, 2016 for a national holiday period.
The radar traffic counters recorded the time, the length and the speed for each passing vehicle (vehicles with speed $\leq$ 3$km \cdot h^{-1}$ were not counted). Emission Factors for NO_x (i.e. NO and primary NO_2) were evaluated as a function of vehicle speed, following the European guidelines EMEP/EEA (Ntziachristos & Samaras 2013) for the estimate of exhaust emissions from road transport. Coupling the hourly radar records with the EF value for each counted vehicle, the hourly mass flows of 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.

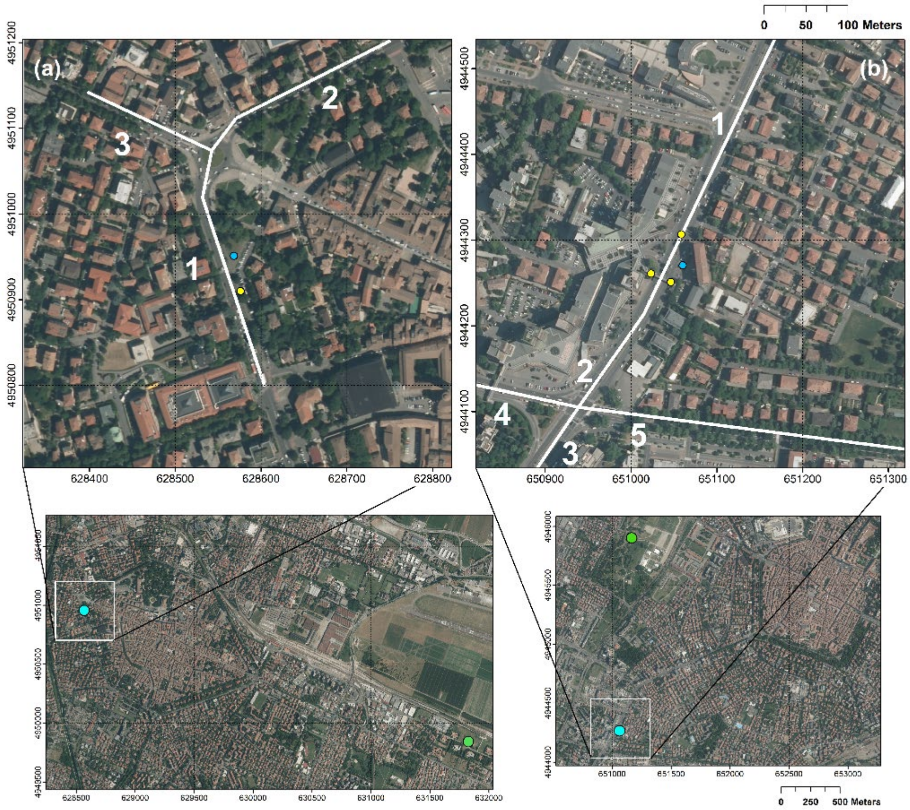

Figure 1. Maps of the investigation domain (UTM32-WGS84), for Reggio Emilia (a) and Modena (b) case studies. Sites of the radar traffic counter are reported by the yellow dots. Sites of the urban air quality monitoring stations are reported by the light blue (traffic stations) and green (background stations)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

The atmospheric emissions from the road sections were simulated using the Micro Swift Spray (MSS) model (Tinarelli et al. 2004) over 500 m x 500 m domains with grid step of 2 m (square cells). The simulation was run at hourly time step, consistently with the meteorological data. The hourly meteorological data were derived from CALMET and COSMO mesoscale model simulations by Arpae.

RESULTS AND DISCUSSION

For both case studies, series of hourly simulated concentration at the urban Arpae traffic site position, for the duration of the measurement campaign, was compared with measured concentrations collected at the urban traffic and urban background station (Ghermandi et al. 2017).

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. 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.

The difference between NO_x observations at urban traffic and at urban background stations (hereafter ΔNO_x) can be attributed to the influence of local traffic, and this contribution was simulated by MSS.

The vehicular emission contribution to air quality at the traffic site, as evaluated by MMS simulation, is generally underestimated: MMS simulated NO_x concentration is lower of about 30% for Reggio Emilia and 56% for Modena case than ΔNO_x. Uncertainties of the whole described procedure apart, the underestimation of MMS concentrations can be also attributed to the following main causes: 1. the contribution by minor streets was neglected 2. the stationary vehicles are not counted (vehicle queues on the street lanes or buses at the bus stop) 3. undercounting of vehicles, since very low speed-vehicles are not detected by the radar.

A further, relevant, cause of underestimation of MMS concentrations may be a large contribution of secondary NO_2 at the urban traffic sites. Given that the NO_x traffic emissions used in MSS include only (primary) NO and primary NO_2 (Air Quality Expert Group 2007), it was investigated whether the difference between simulated and observed levels are due to secondary NO_2 only, which are not described in the MSS simulation, or by an underestimation of the primary NO_x 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. This assumption was tested by comparing the $\Delta NO_2/\Delta NO_x$ ratio (ΔNO_2 being the difference in NO_2 observations at the urban traffic and background sites and ΔNO_x defined as above), with the primary NO_2/NO_x ratio in the total vehicular emissions derived from Grice et al. (2007) and Smit et al. (2010) for the local vehicular fleet. The test was performed using for each city the Arpae observations from urban traffic and background sites collected during both the measurement campaigns, because they were featured by very different weather conditions (Table 1).

Table 1. Comparison among primary NO_2/NO_x ratio in total vehicular emissions and ratios from observations

		primary NO_2/NO_x (%) in total vehicular emissions	$\Delta NO_2/\Delta NO_x$ (%)	NO_2/NO_x (%) urban background
Jan 2014	**Modena**	15.0	26.0	36,5
	Reggio Emilia	16.5	15.7	43.4
Oct -Nov 2016	**Modena**	15.0	41.7	41.8
	Reggio Emilia	16.5	28.7	50.3

The primary NO_2/NO_x ratio resulted very similar for Modena and Reggio Emilia, because of the similarity in the fleet composition of the two cities: but it was not the same for $\Delta NO_2/\Delta NO_x$ and urban background.

The 2014 campaign shows in Reggio Emilia great comparability of primary NO_2/NO_x and $\Delta NO_2/\Delta NO_x$ ratios, and both are in the order of one third of NO_2/NO_x urban background ratio. This may indicate that there is no extra secondary NO_2 at the urban traffic compared to urban background, and also that the direct impact of traffic at the urban background site is low. Conversely in Modena the $\Delta NO_2/\Delta NO_x$ ratio is about 60% larger than this ratio in vehicular emissions and only 40% lower than the NO_2/NO_x ratio at the urban background site. The uncommon weather conditions, favourable to pollutant dispersion, occurring during the campaign of January 2014, emphasize the differences between traffic and background sites.

During the 2016 campaign the $\Delta NO_2/\Delta NO_x$ ratio in Modena results much larger than emission data, while the NO_2/NO_x ratio at the urban background is the same of the $\Delta NO_2/\Delta NO_x$: this suggests similar pollution sources between the urban traffic and urban background site of Modena, although with difference intensities, i.e. the larger concentration observed at the urban traffic site originates from a similar source mix of the concentration observed at the urban background site. This occurrence is amplified by the concurrent weather conditions, which favour air masses stagnation and homogenization of *aged* emissions, determining similar pollution *facies* across the whole urban area. However these same weather conditions of November 2016 have a different effect in Reggio Emilia, where the $\Delta NO_2/\Delta NO_x$ ratio at the urban traffic site is again ca. 40% larger than the NO_2/NO_x ratio in emissions, and slightly lower than 50% of the NO_2/NO_x ratio at the urban background site.

The location of the monitoring sites of Reggio Emilia is better representative than for Modena of the different atmospheric pollution conditions for the city, also in case of weather conditions unfavourable to pollutant dispersion in atmosphere. A large contribution of secondary NO_2 at traffic site, as occurs mainly in Modena, may cause a considerable underestimation of MMS concentrations.

CONCLUSIONS

This work highlights the high reliability of the Micro Swift Spray model combined with observations to simulate the atmospheric dispersion of NO and primary NO_2 vehicular emissions in an urban area having intense traffic, in two cities in Central Po Valley (Northern Italy) and with different meteorological conditions. The results support the hypothesis that local weather conditions, typical for autumn season, cause large contribution of secondary NO_2 also at the urban traffic sites, so much to determine complete pollutant homogenization in the whole urban area. This secondary NO_2 contribution at traffic site may be a relevant cause of underestimation of MMS concentrations.

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