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INFLUENCE OF NO_2 - O_3 URBAN BACKGROUND ON NITROGEN DIOXIDE CONCENTRATION NEAR ROADWAY SOURCES IN BARCELONA CITY (SPAIN)

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1. INTRODUCTION

High NO_2 levels in ambient environments have been associated with adverse health impacts. The main contributors to NO_2 levels within urban areas are primary NO and NO_2 emissions from traffic, NO_2 from other sources and local NO-NO_2-O_3 chemistry, which strongly depends on the availability of O_3 supplied from outside the city. Hence, background levels of NO_2 and O_3 are key to estimate air NO_2 concentrations at street level.

Air pollutant observations measured in urban areas not directly influenced by local sources are commonly used as background concentrations for modelling the dispersion of traffic-related pollutants in urban streets (Vardoulakis et al,. 2003). However, these observations are not available in advance for forecasting purposes and wind direction changes during the day may decrease the background representativeness of a monitoring site falling out of air masses trajectory.

Mesoscale photochemical models (e.g. CMAQ) have been used to provide urban background. This approach can lead to double count traffic emissions in both local dispersion and photochemical models. To avoid this circumstance, re-executing the photochemical model with zero-out local emissions have been proposed (Arunachalam et al., 2014).

In this work, we introduce the upwind urban background scheme that provides background concentrations using CMAQ depending on the wind speed and direction avoiding double counting emissions and using CMAQ outputs without need to re-run the model.

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We combine observations and upwind urban background scheme results to analyse NO_2 and O_3 background influence on urban NO_2 in the Barcelona city. To estimate urban NO_2 we use CALIOPE-Urban, which couples CMAQ and R-LINE (Snyder et al., 2013).

2. METHODOLOGY

2.1 CALIOPE air quality system

CALIOPE (Pay et al., 2012) is a mesoscale air quality modelling system that provides 48 hour air quality forecasts at 12 km horizontal resolution over Europe, 4 km over Spain and 1 km over urban areas (e.g. Barcelona city). The mesoscale system is used to provide background concentrations and meteorological data. It integrates the Weather Research and Forecasting meteorological model (WRF), the High-Elective Resolution Modelling Emission System (HERMES), the Community Multiscale Air Quality Modeling System (CMAQ) and the mineral Dust REgional Atmospheric Model (BSC-DREAM8b). Vertically, CMAQ levels are collapsed from the 38 WRF levels to 15 layers up to 50 hPa with six layers falling within the PBL. CMAQ version 5.0.1 with CB05 chemical mechanism and AERO5 aerosol scheme is used.

2.2 Upwind urban background scheme

To avoid double counting traffic emissions in CMAQ and R-LINE and to take into account mesoscale meteorological patterns, a new method is under development. The upwind urban background scheme makes a selective choice of CMAQ cells as sketched in Figure 1. For each hour, a polygon covering upwind air masses (white) is created. In the figure, the average distance traversed by air masses during an hour

(10.8 km) is estimated for WRF bottom layer wind speed (3 m/s in the image). Squares falling within the scheme polygon represent CMAQ cells and their color refer to cell pollutant values (e.g. NO_2 at peak traffic hours may be higher within the city than over the Mediterranean sea). Grid cells values falling over the scheme polygon are averaged to produce the background estimate of the scheme. This method is inspired by Berkowicz (2000) who apply a similar concept based on air masses trajectory to develop a background model.

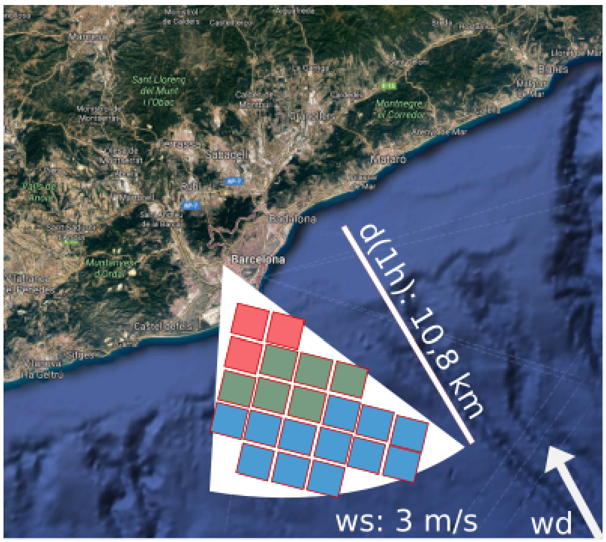

Figure 1. Upwind urban background scheme concept.

2.3 Observations

Ambient street-level pollutant and meteorological measurements collected during a field study in April 2013 within the structured grid of Eixample neighborhood of Barcelona city (Amato et al. 2014) are used to explore contributions to urban NO_2 in combination with observations from the official XVPCA network. We explore the period 15th to 18th of April 2013 to understand the influence of background NO_2 - O_3 on urban NO_2. This period is chosen because there was an air quality episode with exceptional patterns in background observations. Valencia Street No. 455 site from the experimental campaign is used as urban traffic site to compare the influence of background concentrations from different sources on NO_2 at street level. We use measurements from Ciutadella Park, Montseny and Eixample sites from XVPCA network: Ciutadella as urban background, Montseny as regional background and Eixample as urban traffic. In order to estimate regional contributions to urban O_x ($NO_2 + O_3$) in Barcelona, we apply Clapp and Jenkin (2001) method that describes regional and local contributions using NO, NO_2 and O_3 observations.

3. RESULTS

3.1 Contributions to urban NO_2 using observations

We estimate the local and regional contribution to oxidant ($NO_2 + O_3$) levels in Eixample station following Clapp and Jenkin (2001). Figure 2 compares mean daylight levels of O_x and NO_x at Eixample traffic station in Barcelona city from October to February for the years 2012 to 2016. To avoid greater scatter, days in which regional-scale photochemical events happened are not considered. These events are assumed to take place when O_3 concentrations are higher than average in Montseny regional background station. From Figure 2, Clapp and Jenkin (2001) define the intercept as the regional contribution, which is local NO_X independent, and the gradient of the regression line is interpreted as the local NO_x dependent contribution.

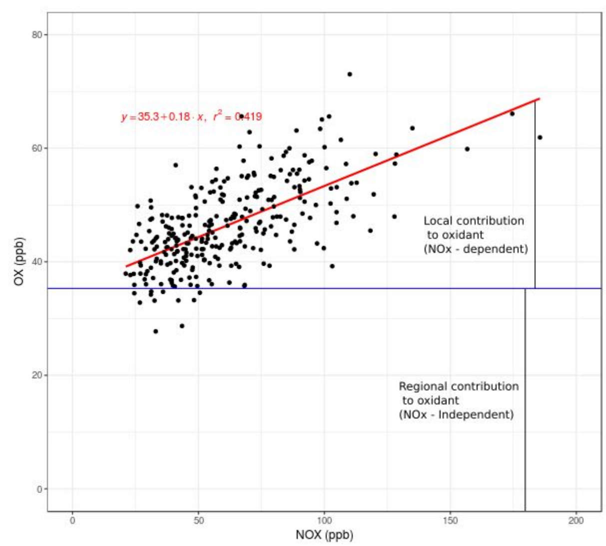

Figure 2. Daylight mean O_x vs. NOx concentrations at Eixample station from October to February from 2012 to 2016.

According to Carslaw and Beevers (2004), O_x ($NO_2 + O_3$) slope value of 18.9% can be considered an estimate of the potential primary NO_2 contribution from vehicles on Eixample traffic station. This value is consistent with studies conducted in other cities with as high diesel vehicle fleet as Barcelona (Carslaw et al., 2016).

3.2 Influence of background NO_2 - O_3 on urban NO_2 using model and observations

We start by analysing the urban background contribution using models and measurements. Ciutadella Park urban background station is chosen as urban background reference station because it is located upwind of the dominant wind direction in Barcelona, determined by sea breeze. With respect to models, the upwind urban background scheme is applied to characterize background concentrations obtained from CMAQ. Additionally, to have a model reference on city background levels, we use CMAQ values from a grid cell over Barcelona city.

The upwind urban background scheme selects CMAQ cells depending on wind conditions leaving out the cell over the estimated area, to avoid double counting. Figure 3 shows NO_2 and O_3 scheme estimates for April 17th (6h to 12h UTC) with triangular shapes representing scheme polygons for each hour, colors are background levels and numbers are time of the day. We can see north eastern winds entering from the river basin turning to sea breezes during the afternoon. Pollutant levels strongly depend on the area over which the scheme polygon falls, being in general higher the NO_2 in the city during peak traffic times and higher O_3 from the sea on the afternoon.

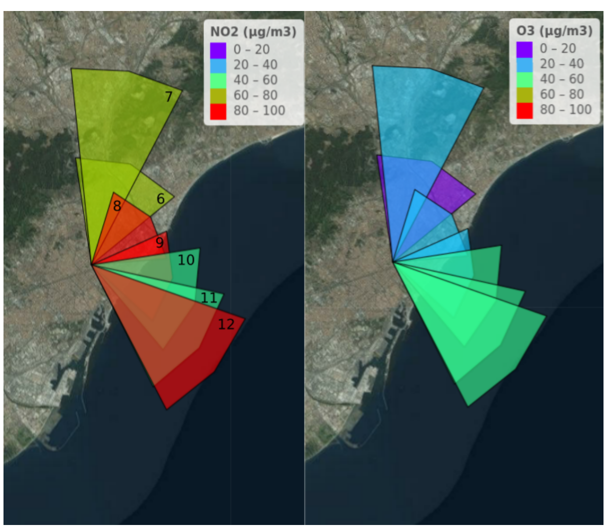

Figure 3. Background of estimated NO_2 and ozone concentrations for 17/4/2013 from 6 am to 12 pm (UTC) over Valencia Street No. 455 in Barcelona.

Figure 4 and Figure 5 show O_3 and NO_2 background levels measured from Monday 15th to Thursday 18th April 2013 at Ciutadella Park station (black line), those produced by the upwind background scheme for Valencia Street No. 455 (red) and CMAQ values over the city (blue), from a grid cell over Valencia Street. This period represents background levels during the air pollution episode. Figure 4 presents the ozone main daily pattern on the observations that is well reproduced by the models. The upwind background scheme overestimates O_3 levels at midday as expected due to its design dependent on wind conditions that brings to the city O_3 levels found on the trajectory of the air masses. A different pattern is present on the 17th, which may be due to precipitation.

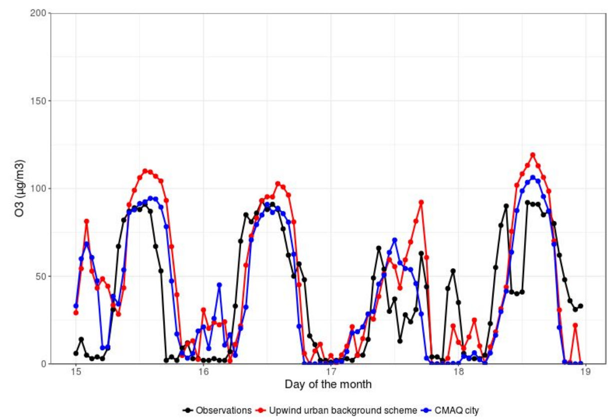

Figure 4. O_3 background concentrations in Barcelona from 15th to 18th April 2013. Observations shown in BLACK, R-LINE with upwind background in RED, and R-LINE with CMAQ from city grid cell in BLUE.

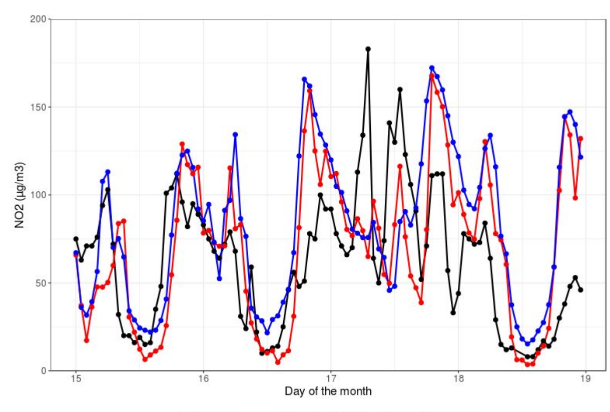

Figure 5. NO2 background concentrations in Barcelona from 15th to 18th April 2013. Observations shown in BLACK, R-LINE with upwind background in RED, and R-LINE with CMAQ from city grid cell in BLUE.

In Figure 5 a similar pattern shows that on 15th, 16th and 18th background NO_2 concentrations are higher at peak traffic times and lower on the afternoon. On the 17th NO_2 diurnal pattern changes and levels increased presenting exceptional spikes at 7h (183 μg/m^3) and 13h (160 μg/m^3).

Model results reproduce the main pattern and hourly levels but can not predict the urban background behaviour on the 17th. Furthermore, the upwind background scheme get closer to observations at the afternoon minimum compared to CMAQ over the city, meaning that background scheme is less influenced by local sources on the afternoon when faster winds are found. However, at morning and evening peaks the scheme pattern is very similar to CMAQ over the city. This result indicates that further improvements on the scheme need to be done in order to better represent urban background, but encouraging results are already obtained.

Finally in Figure 6, we compare hourly NO_2 levels produced by R-LINE using GRS chemistry (Valencia et al., Submitted) and the new local meteorology module within R-LINE, which is presented by Michelle Snyder in this conference with title *Adaptation of meteorology and R-LINE to street canyon micro-climates: Application in Barcelona city Spain*.

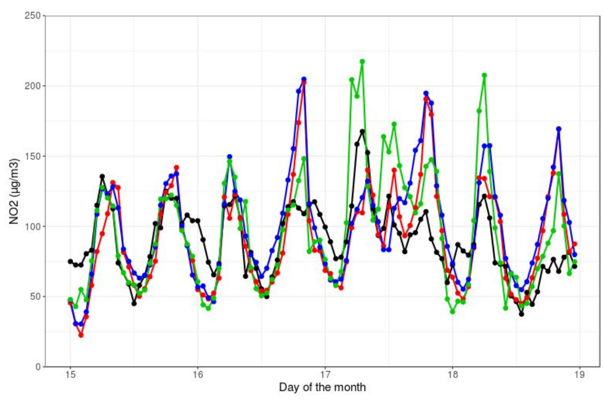

Figure 6. Estimated urban NO_2 concentrations at Valencia street No. 455 in Barcelona (15th to 18th April 2013). Observations shown in BLACK, R-LINE with upwind background in RED, R-LINE with CMAQ from city grid cell in BLUE and R-LINE with observations in GREEN.

R-LINE is executed given different options of background concentration input: upwind urban background scheme (red), CMAQ over city (blue) and Ciutadella Park background station observations (green). The simulation is run over Valencia street No. 455 site, a highly trafficked street, during the same period (15th to 18th). We can see from Figure 6 that NO_2 background has a marked influence in model results temporal pattern. For instance, on the 17th at 7h R-LINE using background observations (green) gives a morning peak overestimation directly related to the high peak found at the same time in Ciutadella Park observations in Figure 5. The afternoon minimums are better represented by R-LINE using upwind background scheme and this configuration slightly improves peak results compared to R-LINE using CMAQ city as background.

4. CONCLUSION

A method to estimate background concentrations using CMAQ, the upwind urban background scheme, has been introduced in an urban street model. CMAQ grid cells are chosen depending on wind conditions, tracing air masses back trajectory. The scheme avoids double counting emissions and provides background concentrations to urban models without re-running CMAQ.

Scheme results show its ability to inject O_3 from outside the city on the afternoon with moderate winds. In addition the scheme provides more precise estimates of afternoon background levels compared to using a CMAQ grid cell falling over the city as background. However, on its current state of development morning and evening peak values are still similar to CMAQ over the city.

The results show that in cities with a high vehicle density with a majority of vehicles being diesel as Barcelona, urban background NO_2 highly influences urban NO_2 concentrations at street level.

This methodology is implemented as part of the CALIOPE-URBAN project. Additional results will be presented in CALIOPE-URBAN: COUPLING R-LINE WITH CMAQ FOR URBAN AIR QUALITY FORECASTS OVER BARCELONA by Jaime Benavides at 9:10am on Wednesday 10/25/2017.

Disclaimer

The results presented here are part of ongoing research and should not be used or referenced until the research is complete and published. If you wish to use results from this extended abstract

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5. REFERENCES

Amato, F., Karanasiou, A., Cordoba, P., Alastuey, A., Moreno, T., Lucarelli, F., Nava, S., Calzolai, G. and X. Querol, 2014: Effects of Road Dust Suppressants on PM Levels in a Mediterranean Urban Area, *Environmental Science and Technology, 48, 8069−8077.*

Arunachalam, S., Valencia, A., Akita, Y., Serre, M., Omary, M., Garcia, V. and Isakov. V., 2014. Method for Estimating Urban Background Concentrations in Support of Hybrid Air Pollution Modeling for Environmental Health Studies. *International Journal of Environmental Research and Public Health, 11, 10518–10536.*

Berkowicz, R., 2000: A simple model for urban background pollution, *Environmental Monitoring and Assessment, 65, 259–267.*

Carslaw, D.C. and Beevers, S.D., 2004: Investigating the potential importance of primary NO_2 emissions in a street canyon, *Atmospheric Environment, 38, 3585–3594.*

Carslaw, D. C., Murrells, T. P., Andersson, J. and Keenan, M., 2016: Have vehicle emissions of primary NO_2 peaked?, Faraday Discuss., 189, 439–454.

Clapp, L.J. and Jenkin, M.E., 2001: Analysis of the relationship between ambient levels of O_3 , NO_2 and NO as a function of NO_x in the UK, *Atmospheric Environment, 35, 6391–6405*.

Pay, M.T., Jiménez-Guerrero, P., Jorba, O., Basart, S., Pandolfi, M., Querol, X. and Baldasano, J.M., 2012: Spatio-temporal variability of concentrations and speciation of particulate matter across Spain in the CALIOPE modeling system. *Atmospheric Environment, 46, 376–396.*

Snyder, M.G., Venkatram, A., Heist, D.K., Perry, S.G., Petersen, W.B. and Isakov, V., 2013. RLINE: A line source dispersion model for near-surface releases. *Atmospheric environment, 77, 748–756.*

Valencia, A., Arunachalam, S., Isakov, V., Heist, D., Carruthers, D., Venkatram, A, submitted. Development and evaluation of the R-LINE model algorithms to account for chemical transformation in the near-road environment.

Vardoulakis, S., Fisher, B.E.A., Pericleous, K. and Gonzalez-Flesca, N, 2003. Modelling air quality in street canyons: A review. Atmospheric Environment, 37, 155–182.