

## **Point by point responses to the Anonymous Referee #2**

We are very thankful to the reviewer for providing a detailed revision of our manuscript. The comments of the reviewer are indicated point-by-point in the following text. We explain how we have carefully addressed each of them (our answers in blue text). Modifications and new sections are highlighted with track changes in the manuscript and the Supporting Information.

### **General comments**

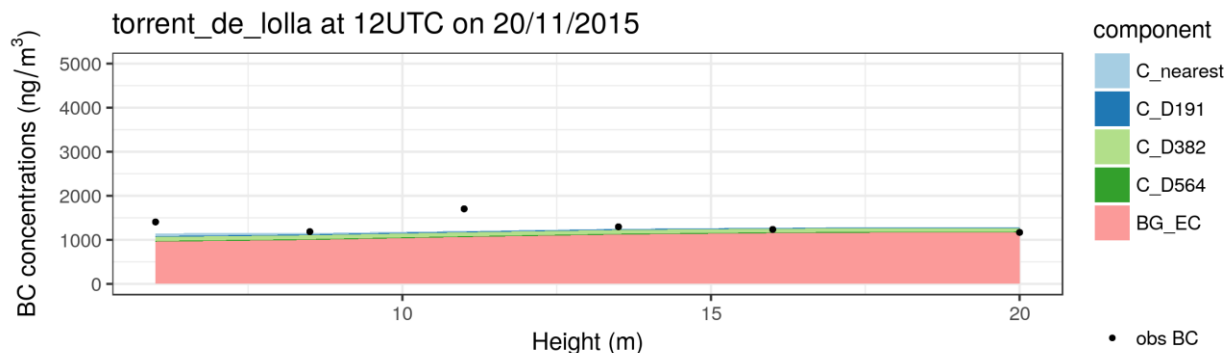
**1. Reviewer #2.** First, it is claimed that atmospheric stability influences the relation between rooftop concentrations and the concentrations at street level (e.g. p. 11, lines 8-11). While this well may be the case, no independent evaluation of the influence of atmospheric stability on the vertical concentration profile in street canyons is given in the paper. The authors should either refer to previous studies in Barcelona or show an evaluation based on own measurement data.

### **Authors:**

Several dispersion models integrate in their formulation the concept of street and over-roof concentrations exchange dependent on atmospheric stability (Hotchkiss and Harlow., 1973; Berkowicz et al., 2000; Soulhac et al., 2011; Kim et al., 2018). In addition, this influence has been demonstrated using wind tunnel experiments (Salizzoni et al., 2009). The influence of atmospheric stability on street wind speed and over roof winds has been shown using experimental measurements, too (Rotach., 1995). For these reasons, we consider scientifically robust to assume that atmospheric stability influences the relation between rooftop concentrations and the concentrations at street level.

The methodology in this manuscript is a first attempt at coupling the urban-scale model, R-LINE, with a mesoscale model, CMAQ, to obtain concentrations throughout the city. We do present multiple street level measurements of varying degrees of urban structural influence. The results indicate that using one consistent background methodology in all these instances provides validity in our approach. Our methodology is still under refinement and will need further evaluation based on additional datasets. We are currently working on the analysis of measured vertical profiles of Black Carbon (BC) within a few street canyons of the Barcelona city. Unfortunately, we don't have access to high frequency vertical profiles of NO<sub>2</sub> concentrations and wind conditions within street canyons in Barcelona. From BC vertical profiles results, it seems that our hypothesis is well-oriented as the reviewer can see in the figure below, where we show the modelled contributions compared with the hourly averaged observed BC at different heights in a very narrow Street in Barcelona. We show 12 UTC (13 hour local time), an

hour that it is expected to have a low contribution from local traffic (i.e. more signal from background). In addition, we expect a well mixed vertical BC column due to the convective conditions typically occurring at this period of the day. We see in the figure that the overall dynamic of the modelled vertical profile is in agreement with the observed profile.



**Figure 1. Local traffic and background contributions to BC vertical profile at Torrent de l’Olla Street at 12 UTC on 20th November 2015. Observations are depicted as dots and coloured levels represent each local traffic contribution: nearest roads is light blue, roads within 191 m (excluding the nearest roads) is dark blue, roads in 392 m (excl. roads within 191 m) is light green, dark green is for roads in 1 km<sup>2</sup> (excl. roads within 392 m) and pink is for background.**

Lastly, it is also important to note that there may not be datasets available that specifically address all possible aspects of our approach, and thus we must rely on the datasets available and on the results of our methodology within the modeling system.

We added a note in Section 2.3.3 page 11 lines 14-16 to support the incorporation of atmospheric stability influence on vertical mixing: “In the research literature, the influence of atmospheric stability on vertical mixing within a street canyon has been demonstrated using experimental measurements (Rotach, 1995), wind tunnel experiments (Salizzoni et al., 2009), and it has been implemented in some dispersion models (e.g. Soulhac et al., 2011; Kim et al., 2018)”.

**2. Reviewer #2.** Second, the model system shows poor skill when the observed wind speed is low. I would expect that the traffic-induced turbulence dominates the turbulence in street canyons at low wind speeds. However, it seems that turbulence generated by the moving traffic is not included in the parametrization. Calm winds potentially lead to highest concentrations and can cause severe pollution episodes. Hence, it would be crucial for a street canyon model to cope with low wind situations.

**Authors:**

In our approach, we have not considered the traffic-induced turbulence directly, however we do have an initial vertical dispersion of roadway emissions which somewhat models traffic-induced turbulence. In previous roadway studies (Snyder et al., 2013; Heist et al., 2013) this same approach has been used in the median of the roadway for Caltrans Highway 99 (Benson, 1989) and results are accurate when compared to near-road measurements, therefore not explicitly modeling vehicle-induced turbulence is not believed to have a large impact on the results.

### Specific Comments

**3. Reviewer #2.** 1.) P. 2 line 9-21: In this part of the Introduction, several systems coupling regional and urban scale models are described. It would be better to divide this presentation into (1) systems that apply nesting of an urban scale model within a regional scale model and (2) regional scale models that apply downscaling (using a dispersion kernel). The given examples from literature are not exhaustive. Also mention a second method for downscaling, by embedding Gaussian dispersion models within the grid.

#### Authors:

In the revised manuscript we present a more complete list of systems that couple off-line regional and urban scale models by downscaling the regional model using a dispersion kernel. We consider adequate to uniquely present downscaling methods because our system belongs to this category, which as far as we know is the most extended methodology to couple regional and urban scales. The revised paragraph in the manuscript is as follows (Section 1 Page 2 Line 9 to 27),

“In order to overcome these limitations, coupling off-line the regional and urban scales by downscaling the regional model using a dispersion kernel has been successfully applied in some cities (Beevers et al., 2012; Moussafir et al., 2014; Isakov et al., 2014; Jensen et al., 2017; Maiheu et al., 2017; Kim et al., 2018; Hood et al., 2018, Fagerli et al., 2019). For instance, Hood et al. (2018) coupled a regional climate-chemistry model with 5 km horizontal resolution (EMEP4UK) with the fine-scale model ADMS-URBAN to simulate air quality over London in 2012. They compared the coupled system results with the regional and the fine-scale models run separately. Authors found that both the fine-scale model and the coupled system performed better than the regional for NO<sub>2</sub> at both annual mean and hourly concentration levels due to the explicit treatment of traffic emissions within the city. In addition, Jensen et al. (2017) estimated annual NO<sub>2</sub> concentrations at 2.4 million addresses in Denmark using the street canyon model OSPM coupled with DEHM for regional background concentrations and UBM for urban background obtaining a good correlation in Copenhagen ( $r^2 = 0.70$ ) against 98 measurement sites for NO<sub>2</sub> in the year 2012. Maiheu et al. (2017) covered a broader

spatial context, estimating EU-wide NO<sub>2</sub> annual average levels at 100 meter resolution with a regional model coupled with a dispersion kernel-based method. The approach does not produce hourly concentration levels and approximates road-link level traffic emissions by distributing the regional grid cell traffic emissions to each road-link based on road capacity. Hence, it provides more spatial detail than previous EU scale NO<sub>2</sub> assessment studies, but more specific methods are required to resolve air quality in cities. In this sense, there is a lack of air quality urban forecasting methodologies that can be applied to a diverse range of cities and that consistently resolve at least some of the major challenges already identified by the community, i.e., 1) downscaling regional meteorology to street level as required to drive pollutant dispersion; 2) obtaining background concentrations from the mesoscale system avoiding the double counting of traffic emissions. Additionally, we consider vertical mixing with background air a key process to be resolved when coupling the regional and urban scales.”

**4. Reviewer #2.** 2.) P.4 line 1: How representative is this period (April and May 2013) for the season? Why was such a short period chosen?

**Authors:**

NO<sub>2</sub> exceedances in BCN are chronic along the year. April and May are months with a reduced amount of holidays and vehicular traffic behaviour is representative of the pulse of the city. We are aware that a longer period would be of interest to evaluate the skills of the model across different seasons. This will be presented in a future work. In this work, we focus on the experimental campaign with multiple simultaneous measurements along trafficked street canyons. We find this dataset relevant because we can evaluate CALIOPE-Urban close to road sources.

**5. Reviewer #2.** 3.) P.5 line 26-27: Why were the 38 vertical layers from WRF collapsed to 15 layers for the CMAQ computation? With only six layers in the PBL, this leads to a rather crude treatment of the near-ground chemistry and boundary layer mixing processes.

**Authors:**

In Europe, there are a wide range of mesoscale air quality models that work with low vertical resolution for computational reasons (e.g., LOTOS-EUROS, CHIMERE), and the skills of those models have been shown to be in the same order as other systems with higher vertical resolution. We use the default CALIOPE forecast configuration, which aims to reduce computational time to allow for rapid forecasting. CALIOPE skills are within the state-of-the-art forecasting systems (e.g. Pay et al., 2014). Since we are

most concerned with NO<sub>2</sub> here, which has a rapid chemical transition from emitted NO to ambient NO<sub>2</sub>, the most important chemistry is the near-road chemistry that is simulated in the fine-scale dispersion model.

**6. Reviewer #2.4.)** P.4 line 1: Please provide a list of the chemical reactions in the GRS here.

**Authors:**

We have included a list of the chemical reactions in the GRS in Section 2.2 page 6 Table 1 and a note referencing the table in page 6 lines 1-3: "In order to estimate NO<sub>2</sub> concentrations, R-LINE incorporates a chemistry module to resolve simple NO to NO<sub>2</sub> chemistry with the Generic Reaction Set (GRS; Valencia et al., 2018) considering the chemical reactions in Table 1."

**7. Reviewer #2. 5.)** P. 7 line 3 and P.12 line 4: Several of the empirical parametrizations in this paper have been calibrated with NO<sub>2</sub> measurements (parameters C and m). This raises the question about independence of the calibration data. Was the calibration done with an independent NO<sub>2</sub> dataset, not used in the presented model evaluation?

**Authors:**

The scarcity of measurements didn't allow us to separate the observations for an independent calibration and validation process. We have used the whole set of observations to calibrate and evaluate the model. As responded in reviewer comment 4, we are aware that a longer period with an independent NO<sub>2</sub> dataset would be of interest to evaluate the skills of the model across different seasons. This will be presented in a future work.

**8. Reviewer #2. 6.)** P.7 line 25: Wind channelling may not occur in streets that are relatively short. The validity of the channelling effect should be analysed for street network of Barcelona.

**Authors:**

We tried to apply a common simple approach for the entire city. We agree with the reviewer that a more refined implementation of the channelling will be needed in the future but it is out of the scope of the present paper. We didn't assess the effectiveness of the channelling effect formulation because we don't have access to a complete dataset of measured wind conditions within a diverse range of streets in the city.

**9. Reviewer #2.** 7.) Section 2.3.3: The large scale model grids are step wise in nature. This could lead to significant edge effects caused by the concentration steps between the CMAQ grid cells. How is this considered in the UBS when applying bilinear interpolation to provide background concentrations at the receptors? The error of the background concentrations at low wind speeds should be estimated.

**Authors:**

We estimate background concentrations at roof level using the urban background scheme in two steps. First, our method selects CMAQ cells as background concentration providers depending on the wind speed and direction. Second, for each receptor we apply a bilinear interpolation method to provide a background at very high resolution calculating weights at each receptor and computing weighted data.

With regard to the comment “The Error of background concentrations at low wind speeds should be estimated”, this has been discussed in Section 4.3 Figure 10 panel f) page 25 lines 8-12 as “background concentrations are underestimated at the beginning of the day (1-4 UTC) and are overestimated during nighttime (19-22 UTC) in days with calm conditions.”

**10. Reviewer #2.** 8.) P. 11 line 10: Does wind channelling affect the ratio  $ws\_sfc/ws\_bh$ ?

**Authors:**

No. Wind channelling does not affect the ratio  $ws\_sfc/ws\_bh$  because in the formulation we consider that channelling would affect equally winds at surface and rooftop level. Hence, dividing the channelling effect by itself would give 1 and it would be omitted.

We added a note to clarify this in the revised manuscript in Section 2.3.3 page 12 lines 2-4 as follows, “wind channelling does not affect the ratio  $ws\_sfc/ws\_bh$  because we assume that channelling affects equally winds at surface and rooftop level”.

**11. Reviewer #2.** 9.) P.12 line 5 - 10: Determination of the surface background concentrations needs more explanation. An illustration of the surface background concentration as function of building density would be helpful for understanding how it is derived from the rooftop background under different stability conditions.

**Authors:**

We added the illustration below to the revised manuscript showing the adimensional vertical mixing variable ( $fa_{cbg}$ ) that is multiplied to rooftop background to obtain surface background concentration as a function of building density and atmospheric stability.

Under low building density (i.e.  $bd$  below 0.1), background concentrations at surface level and over-roof level are assumed to be similar because there are almost no buildings acting as barriers. When building density increases, the difficulty of the overlying air masses to penetrate the street cavities (building height is 20 m in the illustration) is assumed to increase and more difference in concentrations is expected as a consequence. Under convective conditions, we expect more air mixing between street air and overlying air. Hence, for those conditions we assign a background within the street that is higher compared to stable atmospheric cases. Under stable atmosphere, we assume that a decrease of air mixing will increase air stratification bringing more difference in concentrations between over-roof and surface level concentrations. We include the image below in the revised manuscript in Section 2.3.3 Page 12 to visually support the explanation of the background decay method.

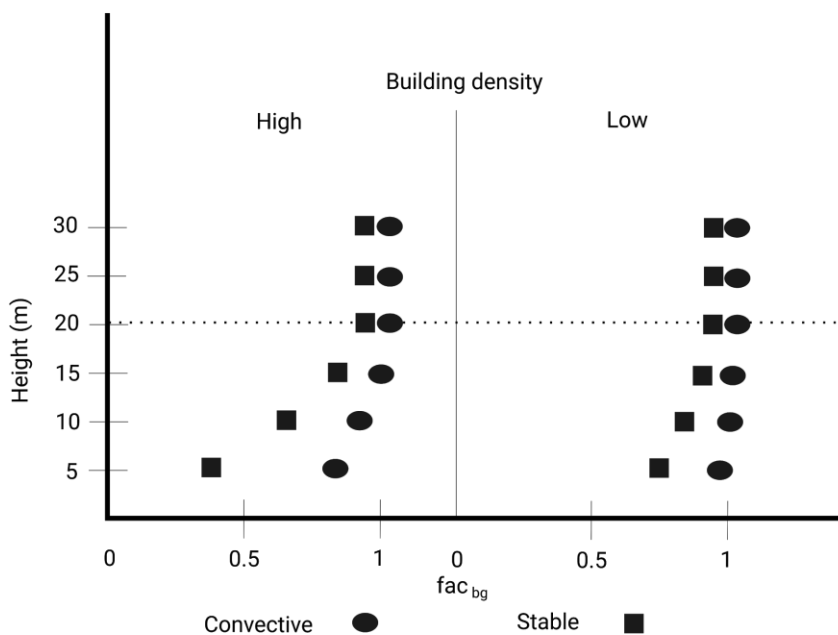


Figure 2. Illustration of the background decay method concept. Building height is approx. 20 m.

12. Reviewer #2. 10.) P.15 line 11 - 12: Which QA/QC procedure was in place for the monitoring with passive dosimeters?

Authors:

Duplicate dosimeters (reproducibility) were installed in some sites, and other dosimeters were installed in the permanent XVPCA network sites for comparison with reference

instrumentation.

The dosimeters were 7 cm diffusion tubes (Palm, GRADKO) that were sent to the laboratory once removed to obtain the concentrations at ambient conditions (nonstandard). Although the concentrations obtained with the dosimeters were ambient, the comparison with the data supplied by the XVPCA network permitted to correct the concentrations with the measures obtained using reference instrumentation at standard conditions. Therefore the concentrations corrected are equivalent to the standard conditions.

**13. Reviewer #2.** 11.) Table 3 and Figure 8: Measurements at station Gracia-Sant Gervasi are underestimated by all three model configurations in the daytime between morning and afternoon rush hours. Table 3 shows a positive bias for CALIOPE-urban-nl (marked as best performance for MB at this site), but this is deceiving since Figure 8 shows that overestimation at rush hours increased the bias. Obviously, the traffic increment is not correctly represented at this site. Could this be caused by the missing contribution from recirculation of traffic exhaust?

**Authors:**

The area of Gracia-Sant Gervasi site is a wide street area, which has a large street width compared to building height thus a low aspect ratio (i.e. approx. 0.38). According to Oke (1988), this kind of street is considered to be in the transition between isolated roughness and wake interference flows. The recirculation in that kind of geometrical settings is not as well documented as skimming flow cases (i.e. higher aspect ratio), where a stable recirculatory vortex is established in the canyon. Hence, we do not expect the missing contribution to be from traffic exhaust recirculation. In addition, in case of missing a relevant contribution from recirculation of traffic exhaust in this site we would expect to miss a similar contribution in all the other sites, specially in the street canyons. From the results analysis, we didn't miss that relevant contribution in all the other sites.

**14. Reviewer #2.** 12.) P.19 line 9 and Figure 8: Give the possible reason for the afternoon underestimation of NO<sub>2</sub> concentrations at sites with low traffic. The underestimation of NO<sub>2</sub> in the afternoons could also be linked to photochemical conversion. Therefore, I recommend to repeat the plots of Figure 8 for NO<sub>x</sub> concentrations.

**Authors:**

As suggested by the reviewer, we repeated the plots of Figure 8 for NO<sub>x</sub> concentrations below. We believe that the possible reason for the afternoon underestimation may be



the overestimated mixing from WRF that leads CMAQ to NO<sub>2</sub> underestimations over the city. We find very similar NO<sub>2</sub> and NO<sub>x</sub> afternoon underestimations, indicating that photochemical conversion may not be the principal reason.

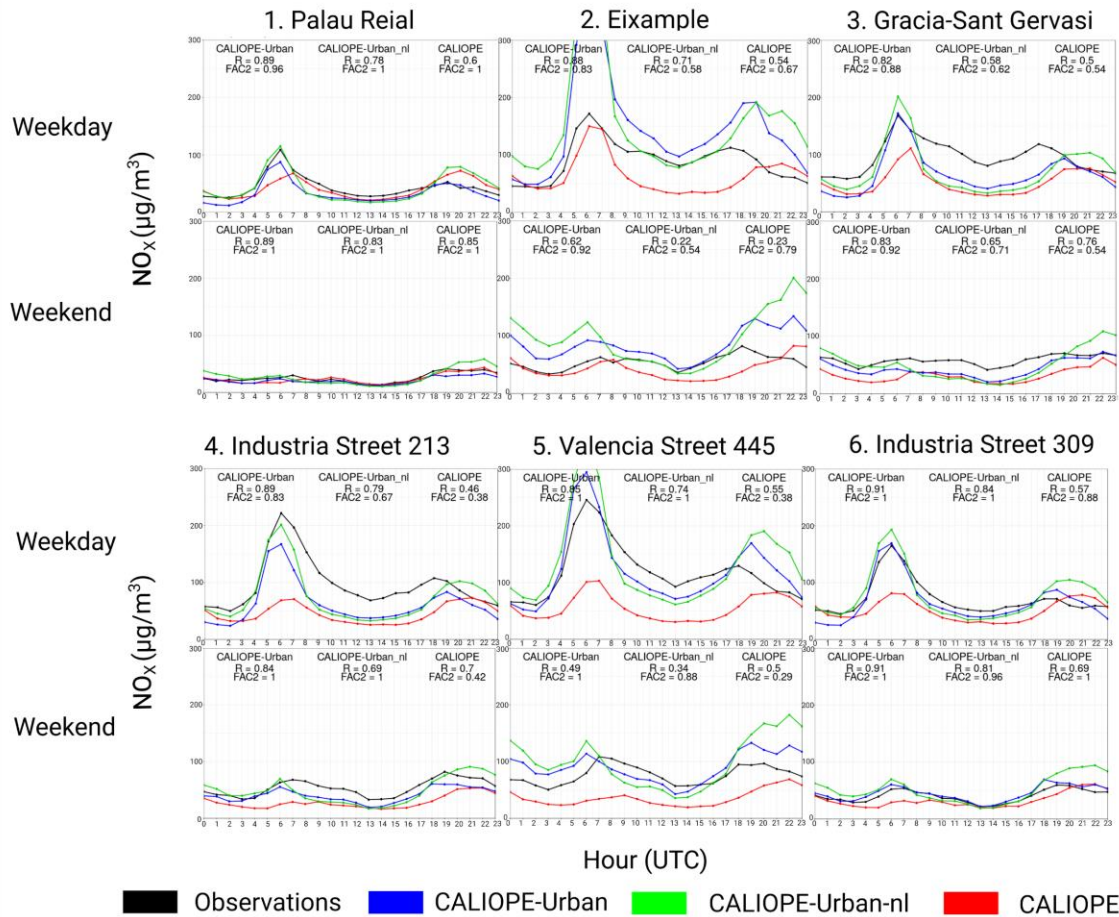


Figure 3. NO<sub>x</sub> average daily cycle at all sites described in Sect. 3.1 during April and May 2013 for weekday and weekend. Observations are represented in black coloured lines, red lines are CALIOPE, blue lines are CALIOPE-Urban and green lines represent CALIOPE-Urban without local developments (CALIOPE-Urban-nl).

## 15.Reviewer#2.Technical Corrections

P. 6 line 8: "This approach addresses" fits better.

Authors: Amended

P.15 line 12: In every km<sup>2</sup> grid cell?

Authors:

The measurements were taken independently from the model grid. Every km<sup>2</sup> in the manuscript refers to square kilometers of surface. A comment has been added in the manuscript in page 16 line 2 making it explicit: “In every km<sup>2</sup> of surface there were at least two dosimeters”.

Figure 11: It should be mentioned in the figure caption whether the resolution is 10m x 10m for the entire concentration map or only in the 250m buffers along streets.

**Authors:**

We added the following note in the figure caption (page 26, Figure 12 in revised manuscript): “The resolution is for the entire concentration map.”

Figure A1: What explains the zero values for the aspect ratio values in the scatterplot?

**Authors:**

The aspect ratio is assumed to tend to zero when there are no buildings on street segments sides within a distance of 100m. The algorithm to assign aspect ratio to a street segment follow this procedure:

- Build two rectangular buffers at each side of street segment given a rectangle side of 100 m (i.e. set as the maximum distance between road edge and buildings to be considered a street).
- Intersect the Barcelona buildings dataset with the two buffers
- If there are buildings at both sides:
  - Estimate the minimum distance between road edge (line) and buildings, which is assumed to represent the distance between road edge and buildings on the side of the street. Add distances at both sides of the road edge to obtain the street width.
  - Estimate the average building height of the buildings falling in both buffers
  - Estimate the aspect ratio by dividing average building height by street width.
- If there are no buildings at both sides: assign aspect ratio equal zero.

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