Point by point responses to the Anonymous Referee #1

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 #1. The analysis only considers NO_2 concentrations. NO_2 is a pollutant of active current interest for regulation and health effects, however it is challenging to model due to the interaction of dispersion and chemistry. Hence it is valuable to analyse modelled and measured NO_X concentrations before considering NO_2, to assist with distinguishing between uncertainties in emissions, dispersion and chemistry. The consideration of chemistry effects should also be included in the associated discussions of NO_2 results.

Authors:

With regard to the chemistry used in the street-scale model, Valencia et al. (2018) evaluated multiple NO-NO_2-O_3 chemical mechanisms in R-LINE. They compared the GRS mechanism, used by ADMS-Urban (Malkin et al., 2016), and two other algorithms for NO_X chemistry with near-road data in Michigan. Their results indicate that the GRS mechanism was the most consistent in predicting NO_2 for near-roadway environments. We believe, based on the Valencia et. al. (2018) evaluation of GRS in R-LINE and the use of GRS in the ADMS-Urban model, that this is an appropriate way to model NO_2 chemistry without the need to also evaluate NO_X.

However, following the advice of the reviewer we show below the comparison of NO_X average daily cycle concentrations for each model (equivalent figure as Figure 8 of the initial submitted manuscript). We can see that the errors for NO_2 and NO_X are very similar for all the stations, indicating that the chemistry of GRS is performing well in our simulation experiments. Thus, we believe that it is reasonable to evaluate modelled NO_2 directly as suggested by Valencia et. al. (2018) and Malkin et al. (2016). Given that the paper is focused on NO_2 due to its interest for regulation and health effects we decided not to include the NO_X discussion in the manuscript and keep the focus on the NO_2 exclusively.

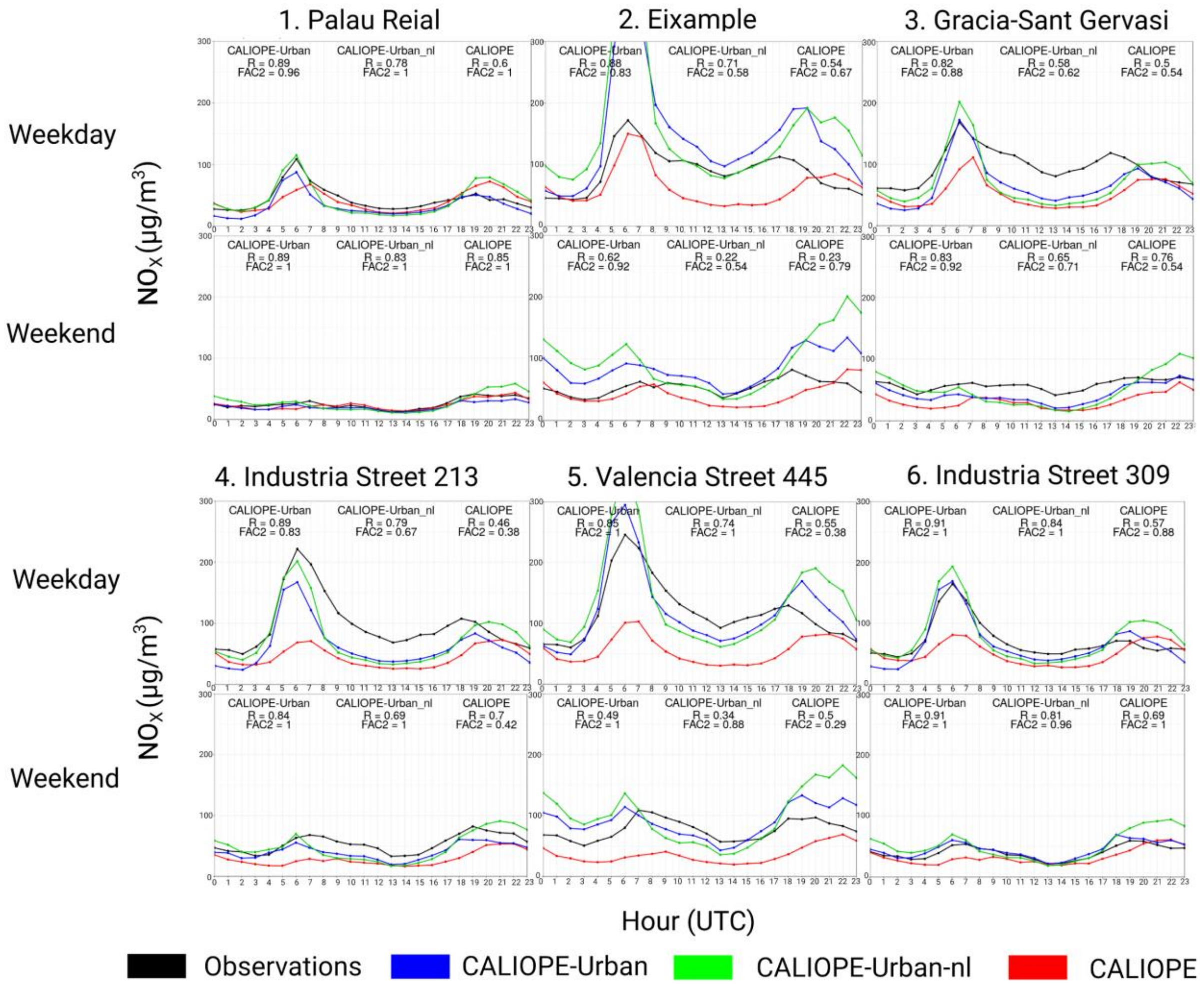

Figure 1. NO_X average daily cycle at all sites described in Sect. 3.1 during April and May 2013 for weekday and weekend. Observations are represented in black coloured lines, red lines are CALIOPE, blue lines are CALIOPE-Urban and green lines represent CALIOPE-Urban without local developments (CALIOPE-Urban-nl).

2. Reviewer #1. The method for taking into account the effects of a specific street canyon on dispersion described in Section 2.3.1 only considers flow channelling along the canyon. However, canyons are also known to cause recirculating flow across the canyon, which significantly alters the dispersion of road traffic emissions and hence the concentration variation with wind direction for receptors within the canyon. No analysis of the modelled or measured variation of concentrations with wind direction is presented, so it is difficult to assess the effectiveness of this formulation.

Authors:

We agree with the reviewer on the importance of modelling the recirculating flow across the canyon and its impact on pollutant dispersion. It is widely known that canyons with sufficient aspect ratio (i.e. more than 0.15) may cause recirculating flow across the canyon (Oke, 1988; Yamartino and Wiegand., 1986; Dobre et al., 2005) when over roof winds are close to perpendicular to street direction. This recirculation may affect significantly the dispersion of road traffic emissions. However, in this work we assume that recirculation process is negligible for multiple reasons: first, dispersion models, such as R-LINE, are not designed to model extremely detailed urban flows (e.g. CFD models), but rather are based on representative flows that are influenced by average urban attributes near the source location; second, dispersion models are designed to give accurate concentrations averaged over a long a time period (usually one hour) where variability in wind speeds occur and thus recirculation may not be longer consistent; third, there is a recirculation and a vehicle induced turbulence occurring within the street canyon, both are contributing to a well mixed more homogeneous air mass within the street canyon, especially over the long averaging time and variable wind conditions; and lastly evaluation of the potential impact (positive or negative) of including recirculating flows across the canyon is not possible without multiple simultaneous meteorological and pollutant measurements within a street canyon at a fine temporal scale, which was not part of the experimental design and thus are not available.

We have added an explicit note about this limitation in the revised manuscript in in Section 2.3.1, page 8 lines 18-22 as “In this work, we assume that recirculation flows within street canyons are negligible because R-LINE computes concentrations averaged over an hour, when recirculation and vehicle induced turbulence are assumed to contribute to a well mixed more homogeneous air mass driven by variable wind conditions. Additionally, evaluation of the potential impact of including recirculating flows across the canyon is not possible without multiple simultaneous meteorological and pollutant measurements at a fine temporal scale, which are not available.”.

Regarding flow channelling along canyon, we didn’t assess the effectiveness of this formulation because we don’t have access to a complete dataset of measured wind conditions within a diverse range of streets in the city. However, the positive results of the work indicate that our formulation is appropriate for the objective of the study.

Specific comments

3. Reviewer #1. Section 2.1: Please state explicitly the depth of the lowest model layer in WRF and CMAQ, which is alluded to in Section 2.3.1.

Authors:

We have added a comment in Section 2.3.1, page 6 lines 19-21 as “Most buildings in Barcelona have lower heights than the WRF bottom layer (40.6 m depth). WRF results are assumed to represent over roof wind and stability conditions because its mid-point height (20.3 m) is similar to average building height (bh) in a typical neighbourhood of Barcelona (e.g. Eixample district; 20.7 m).”

4. Reviewer #1. Section 2.3.2 and Figure 3: Please comment on the negative value of intercept, which may indicate that the Ciutadella site does not fully represent an appropriate urban background concentration for the Eixample traffic site.

Authors:

We believe that Ciutadella is a reasonable background site due to its upwind location in the predominant wind direction during the day and its location within the main park of the city (see Figure 2 in the manuscript). In addition, during the two-week period of the passive dosimeters campaign its mean NO_2 concentrations were 40.2 µg/m^3, which is very close to the observed mean concentrations of 42.1 µg/m^3 using the passive dosimeters, suggesting that it is a reasonable background site (Amato, personal communication, April 24, 2019). The value of the intercept is very close to zero (i.e. the remaining background influence), which means that the regional and urban background contribution have been taken out reasonably well.

5. Reviewer #1. Section 2.3.3: Is the background mixing correction applied uniformly to all pollutants? In particular, O_3 usually shows opposite behaviour to NOx and NO_2, so this formulation may distort background concentrations used for chemistry calculations. Please also clarify how the background concentration is used within R-LINE, especially in regard to the chemistry calculations.

Authors:

Yes, the background vertical mixing is applied uniformly as done in all split-operator models. The vertical distribution of pollutants are solved first with a vertical diffusion following similarity theory, applied uniformly to all pollutants, and then we solve the chemistry. With this approach, O_3 shows opposite profile to NO_X and NO_2 as noted by the reviewer. This is consistent with the chemical reaction of emitted NO with ambient O_3 to form NO_2. We have included a clarifying note in the revised manuscript in Section 2.3.3, page 11 lines 10-12 as “To calculate street-level NO_2 concentrations, the vertical distribution of pollutants are solved first using the background decay method, applied uniformly to all pollutants, and then the GRS chemical mechanism is solved.”

6. Reviewer #1. Section 2.4: Although the analytical model shows a significant reduction in execution time relative to the numerical local model, is 44 minutes execution time for 1 modelled hour realistic for use in an operational forecasting system?

Authors:

The current model design and methodology was explored as a potential way to forecast pollutant levels in the future, therefore we are using this initial evaluation to determine if this is possible and with what level of accuracy. It is important to explore both the analytical and numerical calculations in R-LINE to determine strengths and weaknesses of both. Here we present that the analytical solution is much faster, however the numerical solution is more accurate in some instances, so a final forecasting model would need to balance speed and accuracy. Once, we determine the validity and accuracy of our method we will begin the process of code optimization. For instance, the R-LINE code is not currently parallelized. Parallelization could be done at the road-segment level, which will speed-up the code by several orders of magnitude, making this an extremely cheap and valuable tool for a forecasting system.

7. Reviewer #1. Section 3.1: Please state the measurement height(s) for the official network sites.

Authors:

We added a note in Section 3.1 page 15 Table 3 to clarify this: “The measurement height of the official network sites and the mobile sites is 3 meters”.

8. Reviewer #1. Section 4: Please add an initial assessment of NO_X modelled and measured concentrations.

Authors:

As explained in response number 1, R-LINE has been evaluated for roadways in urban areas with inert pollutants (such as NO_X) and reactive pollutants (such as NO_2), therefore there is no value added to present a full evaluation of NO_X in this instance. In addition, chemical transport models such as CMAQ have been evaluated for use in urban areas and have been previously used to provide background concentrations (Beevers et al., 2012; Isakov et al., 2014). We are using these previous models as is, by coupling R-LINE and CMAQ, and making adjustments based on the data to evaluate the additions of the street canyon and background adjustments. The street canyon adjustments are evaluated using a variety of street canyons throughout Barcelona. The background adjustments are evaluated at background sites throughout the city.

Considering that the scope of the present manuscript is the modelling of NO_2 concentrations at street level, we prefer not to add the discussion of NO_X.

9. Reviewer #1. Section 4.1: The discussion relating to Figure 8 does not mention the varying influence of chemistry processes through the day, which can lead to inaccuracies in diurnal profiles.

Authors:

R-LINE first calculates the dispersion of pollutants from the road source, and then it resolves the parameterised equations of the chemical reactions for the pollutant transportation time interval. The GRS chemistry mechanism solves the photochemistry of NO_2 assuming clear-sky conditions. Hence, it does not consider cloud effects on photolysis, representing one of its major limitations.

Valencia et. al. (2018) also show that the GRS method in R-LINE has less than a 15% bias of the results even though they do not account for cloud cover. From our experience, the processes that may have a greater influence on the results in this modelling system are the correct wind and stability conditions and the accuracy of emission sources within the street canyons. These may have more influence on the concentrations than the photolysis in the chemistry scheme. For example, in Figure 2 we show the weekday average daily cycle for Valencia Street 445 to compare the effect of setting the GRS mechanism photolysis rates to zero (caliope_urban_no_photo) with the effect of setting atmospheric conditions to stable (caliope_urban_stable). The stable conditions are set using the following parameter values from Snyder et al. (2013): Lmon (Monin-Obukhov length) equals 11.1; ustar (friction velocity) equals 0.12; Wsrefh (wind speed at roof-top level) equals 2.0. The impact of neglecting completely the photochemical reaction of the GRS chemical mechanism results in an overestimation of NO2 concentrations during daytime. Although we see a negative impact of not using the photochemical reaction in the GRS chemical mechanism (purple line), it is clear that setting stable atmospheric conditions dramatically changes the modelled concentration levels, producing a greater overestimation (green line). These results confirm our initial hypothesis that atmospheric stability has a much greater influence on the NO_2 concentration than neglecting clouds in the calculation of the NO_2 photolysis rate applied in the GRS chemical mechanism.

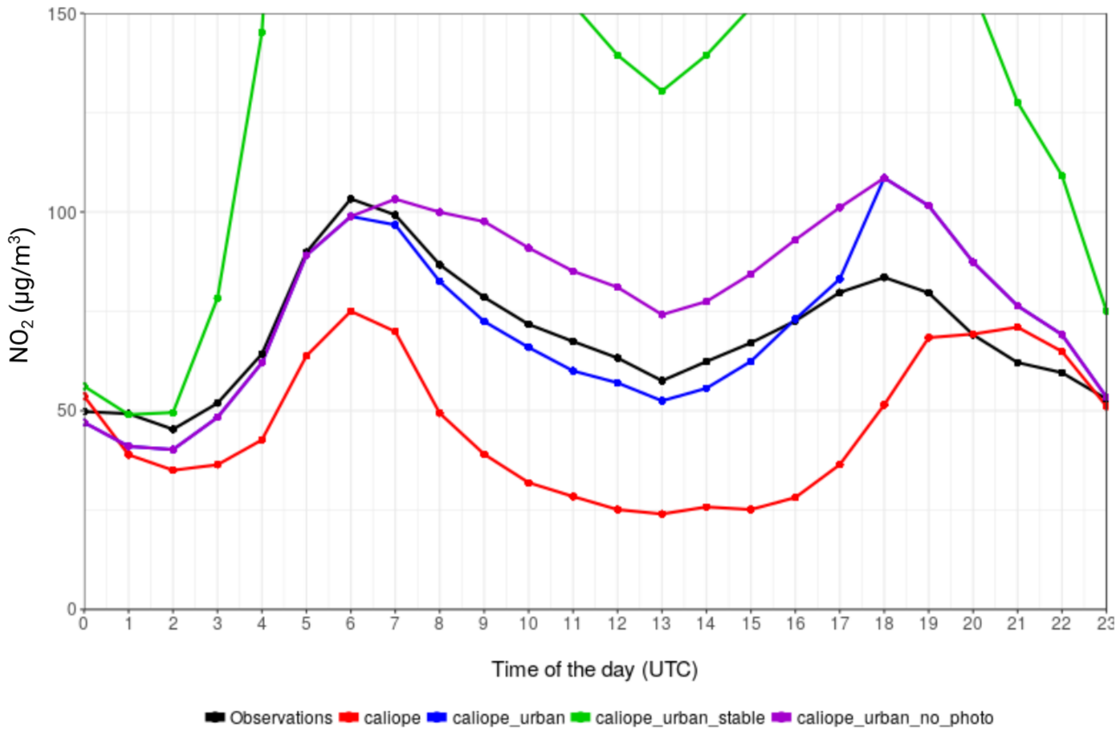

Figure 2. NO_2 average daily cycle at Valencia Street 445 site described in Sect. 3.1 during April and May 2013 for weekday. Observations are represented in black coloured lines, red lines are CALIOPE, blue lines are CALIOPE-Urban, green lines represent CALIOPE-Urban with stable atmospheric conditions (caliope_urban_stable) and purple lines depict CALIOPE-Urban with photolysis rates set to zero (caliope_urban_no_photo).

In the revised manuscript, we have added a note about neglecting the effect of clouds in the R-LINE photolysis rate (Section 2.2, page 6 lines 3-4): “GRS chemistry mechanism solves the photochemistry of NO_2 assuming clear-sky conditions. Thus, it does not consider cloud effects on the NO_2 photolysis rate, representing this one of its major limitations.”

10. Reviewer #1. Section 4.3: It is common for Gaussian-type models such as R-LINE to perform poorly in low wind speed conditions due to uncertainties about associated wind directions. They also do not take into account possible accumulation of pollutants between hours in low wind speed conditions, which is in contrast to the assumption in the background adjustment in this work of reduced mixing causing reduced surface background concentrations. Figure 10f) suggests that the unadjusted regional background could be more appropriate than the adjusted in the early morning hours, though not in the evening. Are there other differences (eg. wind direction) between these two periods?

Authors:

Regarding the accumulation of pollutants, it's true that we can't consider it from one hour to the next within the street canyon. As the reviewer correctly identifies, this is a limitation in dispersion models and in our implementation.

Regarding the reviewer comment on Figure 10 panel f) that the unadjusted regional background could be more appropriate than the adjusted in the early morning hours, we are aware that the result from the upwind background scheme (assumed as roof-level background concentration provider) gives a more precise result from 0 to 7 UTC under calm winds in Figure 10 panel f). In contrast, at night from 18 to 23 UTC the opposite happens, background concentrations at surface level are more accurate than at roof level in comparison with observations. We see this result as positive because it suggests that our method can reduce the overestimation of night surface level concentrations. The contradictory result at surface level between morning and night hours for stable hours with calm wind conditions is dependent on the results of the mesoscale system and we would need more observational data to further investigate this issue.

Concerning the uncertainties associated to wind direction under low wind conditions, we show in the following figure the difference in wind direction at the Barcelona airport for the two periods discussed in Section 4.3:

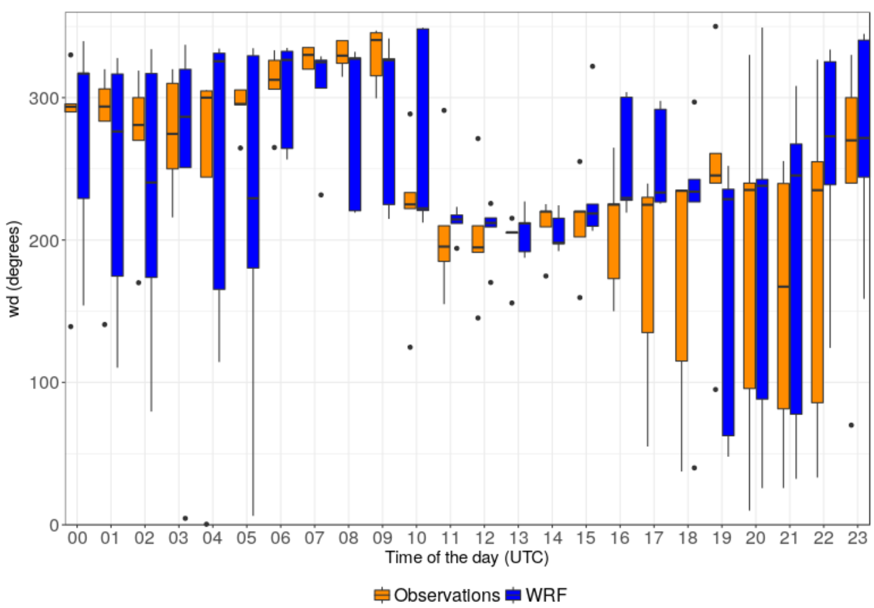

Figure 3. Boxplots by time of the day of good performance days for WRF (blue) and observed (orange) wind directions at Barcelona airport (10m height) with dots representing outliers.

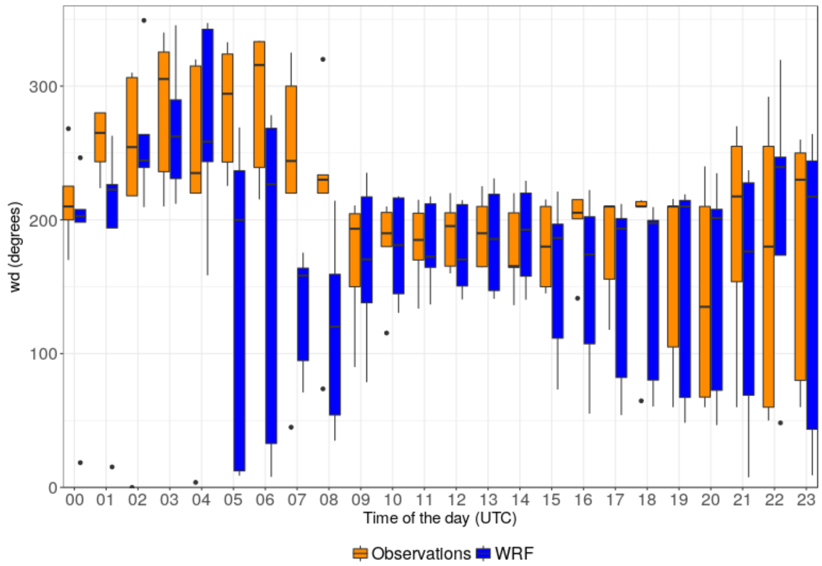

Figure 4. Boxplots by time of the day of bad performance days for WRF (blue) and observed (orange) wind directions at Barcelona airport (10m height) with dots representing outliers.

From the two figures, we see a similar pattern in both periods in general, being the exception the morning transition from stable to unstable atmospheric conditions (5-8 UTC) in bad performance days. The wind direction difference between WRF and observations is approximately 90 degrees on average. In contrast, looking at Figure 10 panel d) (surface NO_2 concentrations) and f) (background NO_2 concentrations), we do not see a clear impact of the mentioned error of wind direction from 5 to 8 UTC. In our system, wind direction may not play a crucial role when wind speed is very low (i.e. for bad performance days approx. 1.5 m/s) for two reasons: (1) the upwind background scheme will take CMAQ concentrations from nearby grid cells (i.e. tending to give similar concentration levels in different wind directions), typically within city boundaries, and (2) R-LINE meandering partial contribution, which disperses radially in all directions, is greater under low wind speeds reducing the potential impact of the wind direction error.

11. Reviewer #1. Figure 10: for panels e) and f) why is the model background concentration an average over six sites, not also taken from the Ciutadella site?

Authors:

As stated in response to reviewer comment 4, the Ciutadella Park monitoring station is the only reference site available for urban background in Barcelona. Our aim in Figure 10 for panels e) and f) is to compare the modelled background concentrations (i.e.

excluding local vehicular traffic contribution) with the most suitable observed urban background, which in our case is given by Ciutadella site. The model background concentration is taken to be an average over six sites because the interest is to represent a summary of the variability of the modelled background at the six sites in comparison with the observed background. We added a clarification in the revised manuscript in Section 4.3 page 25 lines 3-4: “We aimed to compare the modelled background concentrations (i.e. excluding local vehicular traffic contribution) with the most representative urban background observation, which in our case is the Ciutadella site.”

12. Reviewer #1. Section 5: Again, the uncertainty in NO_2 resulting from chemistry processes should form part of the discussion.

Authors:

We added a comment about the uncertainty of chemistry processes used in our solution in Section 5, page 28 lines 6-7: “Finally, we consider an additional source of uncertainty the assumption of clear-sky conditions in the photolysis rate calculation of the GRS chemistry mechanism.”

Technical corrections

13. Reviewer #1. Abstract, p1 line 15: In this case, the coupled system **also** shows

Authors: Amended

Section 1, p3 line 8: subtract its result **from** the mesoscale model

Authors: Amended

Section 1, p3 line 30: please re-phrase ‘over background roof-level concentrations’ as the meaning is unclear

Authors: Amended

Section 1, p3 line 35 – p4 line 1: 5 traffic site**s** and 1 background

Authors: Amended

Section 1, p4 line 2: campaign that **deployed** 182 NO2 passive dosimeters **across Barcelona** for two weeks..

Authors: Amended

Section 2.3.3, p12 line 8: please re-phrase ‘ends when the surface background gets over roof value for bd equals 0’ as the meaning is unclear

Authors: Amended

Section 3.1, p15 line 8: centred **on** the measurement site

Authors: Amended

Section 4.1, p18 lines 9 and 11: unnecessary **the** before Appendix B

Authors: Amended

Figure 7: these plots look vertically distorted, as the target area is usually viewed as circular.

We agree with the reviewer that the image looks vertically distorted but the current version of the Delta Tool for Windows is producing this kind of plot and as far as we know we can't do anything to change it. We downloaded the most updated version and it produced the same kind of plot. In the informational website of Delta Tools it is shown as vertically distorted, too: https://ec.europa.eu/jrc/en/scientific-tool/fairmode-delta-tool

Figure 8: The vertical and horizontal axis scale labels are too small to read.

Authors: Amended

Section 4.1, p20 line 3: higher modelled traffic emissions, **resulting** in higher local pollutant concentrations...

Authors: Amended

Section 4.3, p23 lines 15-18: The first sentence says ten days of highest RMSE and ten days of lowest RMSE, whereas the following sentences suggest five days of high RMSE and five days of low RMSE. Please clarify how many days were selected and analysed.

Authors: Amended

Section 5, p27 line 11: ... gives surface concentrations **by** applying a vertical…

Authors: Amended

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