Persistent Wintertime Thermal Inversion in the City. The case of Madrid. Part II. Dispersion

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Abstract.

A correct simulation of the dispersion during wintertime thermal inversion is important not only to improve the physical understanding of the phenomena but also to design and evaluate strategies to improve air quality. In Part II of this study the simulations performed in Part I using WRF with the multilayer urban canopy scheme BEP-BEM, are used to simulate the dispersion during one of those episodes over Madrid, Spain, using a passive tracer representing NO_x. The results are compared against NO_x measurements obtained at 24 ground stations, grouped in 5 zones. Coherently with Part I, the analysis shows that the best results for dispersion are obtained when BEP-BEM is used and the soil moisture is reduced to 25% of the value provided by the global model. The results also show a strong spatial variability of dispersive conditions, with a tendency of the pollutants to accumulate in the topographical depressions. Furthermore, a simple methodology is proposed to derive NO₂ from NO_x concentrations based on an empirical fitting of two months of observations. The statistical indicators for NO₂ computed in this way are similar to those obtained with photochemical models, making this approach a valid, and computationally efficient, alternative, in particular for forecasting purposes. In the last part of this contribution, the model is used to design and evaluate simple air pollution reduction strategies. In particular, it has been found that: 1) there is no pollutant accumulation from one day to the following one, so the air quality of one day is entirely determined by the emissions and meteorological conditions of that day, 2) displacing part of the emissions of the rush hours to the central hours of the day, when the dispersive conditions are more favorable, can significantly reduce the peaks, and to a lesser extent the daily mean, without changing the total amount of pollutants emitted.

Keywords:

Air quality during thermal inversions, NO_x-NO₂, Air quality mesoscale modelling, Air pollution reduction strategies.

1. Introduction

Air pollution levels in European cities have been continuously decreasing in the last decades in response to technological improvements that reduced emissions, in particular from traffic (Querol, et al, 2014, Colette and Rouil, 2020,). Despite this, threshold limits are still exceeded in many urban areas, especially during periods of strong atmospheric inversion. Being able to model these episodes is particularly important if we want to evaluate the efficiency of different air quality abatement strategies. However, it is well known that this type of conditions represents a challenge for meteorological models (Holstlag et al. 2013, Steeneveld, 2014, Svensson et al. 2011, Román-Cascón et al. 2016, Serafin et al. 2018), and that “new research is needed to develop a boundary layer and land surface scheme to capture multiday wintertime inversion episodes and enable more accurate simulations of wintertime air pollution episodes” (Baker et al. 2011). This is further complicated by the presence of complex terrain (Giovannini et al. 2020). The multilayer urban canopy scheme BEP-BEM (Martilli et al. 2002, Salamanca et al. 2010) was built to improve the representation of Urban Boundary Layer Structure in mesoscale models, and consequently, better simulate the dispersion of pollutants. However, the scheme has been mainly used for summertime conditions linked with photochemical models (Martilli et al. 2003, Liao et al. 2014, Fallman et al. 2016) or for long periods, as De la Paz et al. (2016), who used WRF-BEP-BEM coupled to CMAQ to simulate a full year over Madrid. To our knowledge, a study focusing on dispersion features during a persistent wintertime thermal inversion episode with WRF-BEP_BEM is still lacking. In Part I (Martilli et al., 2021), we assessed the strength and weaknesses of the model to simulate the meteorology during one of these episodes over Madrid, Spain (Borge et al. 2018a). We identified in soil moisture, and in the way in which the urban surface is treated (e.g. the use of the multilayer UCP) the two most important factors that affect the simulation of the thermo-dynamical evolution of the PBL in these conditions. Here we focus on the dispersion of pollutants, in particular those emitted in the city. Therefore, the research questions that motivate the present study are:

Can the model properly simulate the pollutant concentrations and capture the most important features of the pollutant dispersion during these situations? Is the inclusion of the UCP BEP-BEM beneficial?

What kind of policy-relevant information can be derived from model results?

The period chosen for the analysis is the same period simulated in Part I, corresponding to nine days from the 22nd to 30th of December 2016 in Madrid, Spain. This period is considered paradigmatic of the persistent winter thermal inversion episodes in the region because it caused 86 exceedances of the air quality standard for NO₂ (200 µg m⁻³ of hourly concentration) in 13 air quality stations in the city in only 5 days (Borge et al. 2018a). In this period, for the first time, the municipality of Madrid implemented level three of the NO₂ protocol, which consisted of restricting access to the city center of conventional private vehicles based on plate numbers.

This paper is structured as follows: in section 2 the model set-up is described. In section 3, the results of the simulations are compared to the concentrations of NO_x and NO_2 measured in Madrid during the episode. In section 4, the spatial distribution of the NO_x modeled is analyzed. In section 5, the model is used to derive relevant information from the policy point of view. Discussions and conclusions are presented in section 6.

2. Modelling approach.

The set-up of WRF is the one described in part I, with four nested domains and the inner one at 1 km resolution including the region of Madrid. In particular, we compared three different simulations:

- **25%SMOIS**, with the city represented by BEP-BEM and soil moisture in the rural areas equal to 25% of the value provided by the ERA5 global reanalysis model.
- **No city**, with the same soil moisture of **25%SMOIS**, but with the urban area replaced by the savannah landuse
- **100%SMOIS**, which is the original soil moisture provided by ERA5 global reanalysis model, and BEP-BEM.

The results in Part I show that the combination with the best skill for the simulated air temperature and the wind is **25%SMOIS**. However, here we consider also the other two simulations, to evaluate how they behave from the dispersion point of view.

In this work, the dispersion of pollutants is simulated using a passive tracer, which is implemented in WRF. This passive tracer is advected and diffused on the same grid of the meteorological model, and with the same turbulent diffusion coefficients, derived from Bougeault and Lacarrere (1989), used for scalars like temperature or moisture. The tracer is defined in the center of the grid cell, as any scalar in WRF, and the volume of the grid cell occupied by the buildings is taken into account in the vertical diffusion, in the same way as it is done for the temperature (Martilli et al. 2002).

The approach of using a passive tracer is similar to the one used by Saide et al. (2011) for Santiago de Chile, who also neglected the chemistry, with the difference that they used CO as a tracer. In our case, the choice of NO_x instead of CO, which can be more properly considered a passive tracer, is mainly because there are more measurements of NO_x than CO, and there is, therefore, more information to validate the simulations. The concentration of the tracer transported and diffused is expressed in $\mu\text{moles m}^{-3}$, considering that the total number of moles of NO and NO_2 is conserved. This is equivalent to neglect all the chemical reactions that imply a transformation of NO and NO_2 in other species, which is reasonable given the low temperatures and radiation intensity and the short daytime duration in this period of the year in Spain. The units of the emissions of the passive tracer are therefore $\mu\text{moles m}^{-2}$ of NO_x .

Emissions for each hour of the seven days of the week are derived from a top-down approach as described in Menut et al, (2013). Fig. 1 shows the daily profiles of the simulated emissions spatially averaged over the whole domain (in $\mu\text{moles m}^{-2} \text{s}^{-1}$) for week and weekend days (note the high and delayed peak of emissions in the evening of the weekend days, linked to Madrid's leisure activities), as well as the spatial distribution

(in the same units) at 0800 UTC and 1900 UTC for a weekday, the hours when the maximum concentrations are usually observed. As commented in Part I, given the longitude of Madrid, UTC corresponds to Local Solar Time.

Several differences are present in this modeling approach compared to the set-up used by Borge et al. (2018b) and De la Paz et al. (2016), who also used WRF-BEP over the same region. The most important is that they used the photochemical model CMAQ instead of a passive tracer to compute the pollutant concentrations. The biggest advantage of CMAQ is that considers the atmospheric chemical reactions, and so it can properly split between NO and NO₂ (the toxic component). The disadvantage is that it is more computationally expensive, it runs on a different grid than WRF, so the meteorological fields are interpolated, the meteorology is updated every hour, and the turbulent diffusion coefficients are recalculated, so there may be a lack of coherence between wind and temperature fields, and the turbulent diffusion. Moreover, the vertical resolution used is different, being much higher in our simulations: 57 vertical levels, of which 10 are equally spaced in the lowest 50 m, vs 30 vertical levels with the lowest level 7m thick used in Borge et al. (2018b) and De la Paz et al. (2016). On the other hand, the emissions of Borge et al. (2018b) and De la Paz et al. (2016) are based on a traffic model and a bottom-up approach, so they are much more reliable in terms of spatial distribution than those used in the present study. Finally, the changes mentioned in Part I (soil moisture adaptation, horizontal diffusion, and 6th order filter) are not implemented in Borge et al. (2018b) and de la Paz et al. (2016).

The simulations are validated against observational data from 24 air quality stations in the municipality of Madrid. At those stations, NO and NO₂ were measured separately in micrograms/m³. From those values, the number of moles of NO and NO₂ have been computed and then summed, so that they can be compared with the values computed by the model. Finally, the micromoles/m³ have been transformed in ppb (using a standard temperature of 25 Celsius), which is a more common unit. The stations are grouped in 5 zones, as indicated in Fig. 2: Zone 1 corresponds to the city center, predominantly Local Climate Zone (LCZ) 2, in large part located on the hill, while the zones 2, 3, and 5 are mainly LCZ5, and located in the topographical depressions induced by the two rivers, - Manzanares (zone 5) and Jarama (zones 2, and 3). Zone 4 includes big parks and its landuse type is mainly rural.

Zone number	Local Climate Zone	Description
Zone 1	LCZ 2	Hill, city center
Zone 2	LCZ 5	Topographical depression (Jarama river)
Zone 3	LCZ 5	Topographical depression (Jarama river)
Zone 4	Rural	Big Park
Zone 5	LCZ 5	Topographical depression (Manzanares river)

Table 1. Zones and description.

3. Evaluation and analysis of the spatial distribution

The top-down technique used for the emissions, while ensuring that the total amount of pollutant emitted is correct, does not guarantee a fine description of the spatial distribution, so we cannot expect a perfect agreement between the model and the measurements in each station. Moreover, it is well known that the primary pollutants in urban areas may have strong spatial gradients at scales of few meters that cannot be fully resolved with a mesoscale model with 1km x 1km resolution (Rivas et al. 2019, Sanchez et al. 2017, Santiago et al. 2021). Hence the model can only provide a general trend of the spatial distribution of pollutants in the city. Therefore, we decided to compare concentration values averaged over all the stations within the same zone against the concentrations produced by the model-averaged over all the grid points where the stations, that belong to the zone, fall.

The comparison of average NO_x for the 5 zones and the three simulations is presented in Fig.3 Overall, the model reproduces the time evolution of the concentrations, for example, the low values on the weekend of the 24th and 25th, and the increase in the days of the episode, or the higher concentration level in zone 5 than in the other zones (note zone 5 is in a topographic depression). There are, however, some discrepancies, in particular in the intensity of the peaks at certain hours. Part of these differences can be explained by the fact that this period of the year, around Christmas, introduces additional uncertainty in the emissions since several activities are not as usual (schools are closed, and a significant amount of people is on vacation). For example, the overestimation of the morning peak of concentration the Monday 26th, in most of the zones, can be explained by the fact that the emissions used are those of a typical working Monday, but that day was a public holiday in Madrid. Another source of uncertainty in the emissions is the implementation, by local authorities, of stage 2 of their emergency plan (days 28 and 30), which consists in forbidding the car parking in zone 1, and stage 3 the 29th, consisting in limiting the entry to zone 1 based on plate number. Other issues cannot be explained by the uncertainty in the emissions, like the underestimation of the evening peak of the 27th in zone 1,2, and 5, or the overestimation of the peaks of the 30th in zone 1, and indicate a deficiency of the model. Keeping in mind these uncertainties in the emissions, the following statistical indicators have been computed to quantify the performance of the three simulations and are represented in Table 2, separately for each Zone and all the stations together.

$$BIAS = \frac{1}{N} \sum_{i=1}^N (C_{mod_i} - C_{exp_i})$$

$$Root\ Mean\ Square\ Error = RMSE = \sqrt{\frac{1}{N} \sum_{i=1}^N (C_{mod_i} - C_{exp_i})^2}$$

$$Index\ Of\ Agreement = IOA = 1 - \frac{\sum_{i=1}^N (C_{mod_i} - C_{exp_i})^2}{\sum_{i=1}^N (|C_{mod_i} - \bar{C}_{exp}| + |C_{exp_i} - \bar{C}_{exp}|)^2}$$

and the correlation coefficient r . These indicators have been chosen because they are the most commonly used in the field, and they, therefore, allow a comparison with other studies.

<i>NO_x 25%SMOIS</i>	BIAS (ppb)	RMSE (ppb)	IOA	Correlation coefficient r
Zone 1	46.22	129.96	<i>0.75</i>	<i>0.67</i>
Zone 2	-23.87	<i>83.36</i>	<i>0.82</i>	<i>0.70</i>
Zone 3	<i>30.68</i>	<i>93.59</i>	<i>0.77</i>	0.67
Zone 4	<i>-26.97</i>	<i>34.96</i>	<i>0.62</i>	<i>0.63</i>
Zone 5	<i>-22.59</i>	<i>101.36</i>	<i>0.77</i>	0.61
All zones	18.90	<i>85.75</i>	<i>0.81</i>	<i>0.71</i>
<i>NO_x No city</i>				
Zone 1	58.56	145.17	0.73	<i>0.67</i>
Zone 2	<i>-5.75</i>	95.94	0.78	0.62
Zone 3	91.44	154.33	0.65	<i>0.69</i>
Zone 4	-27.22	38.25	0.57	0.45
Zone 5	76.47	178.30	0.69	<i>0.67</i>
All zones	54.07	117.66	0.76	0.73
<i>NO_x 100%SMOIS</i>				
Zone 1	<i>18.12</i>	<i>124.03</i>	0.72	0.57
Zone 2	-34.13	103.55	0.65	0.46
Zone 3	22.40	110.14	0.65	0.48
Zone 4	-30.51	43.75	0.46	0.13
Zone 5	-40.50	118.68	0.68	0.50
All zones	<i>1.24</i>	91.88	0.75	0.58

Table 2. Statistics for the three simulations and the different zones for NO_x. RMSE stands for Root Mean Square Error and IOA for Index Of Agreement (1 indicates a perfect agreement). IN bold-italic are the best scores

Overall, the best statistical indicators are for ***25%SMOIS***, while ***No city*** presents stronger overestimations, in particular in zone 5 and 3 (also clearly visible in Fig. 3), and ***100%SMOIS*** underestimations with a relatively worse index of agreement and correlation coefficient. This behavior is mainly linked with the fact that ***No city*** cools more and faster at night than ***25%SMOIS***, generating a more stable PBL and a lower mixing height, in particular in the topographical depressions. On the other hand, ***100%SMOIS***, having a larger heat capacity, cools slower and less than ***25%SMOIS***, generating a thicker PBL and therefore lower concentrations

3.1 NO₂ estimates

As mentioned above, the biggest drawback of using a passive tracer instead of a photochemical model is that it is not possible to derive the concentration of NO₂, because it is highly reactive. However, having even a rough estimate of NO₂ is important

because it is a toxic gas for which there are limit values for hourly and annual averages in the national and European legislation. With this in mind, here we propose an alternative and computationally cheaper way to get rough estimates of NO₂. A data binning procedure is followed to filter the two-months (November and December 2016) hourly measurements of NO_x and NO₂ recorded at the 24 air quality stations of Madrid. Based on the values of NO_x, the NO₂ data are grouped in bins. In the first bin, there are all the NO₂ data that correspond to NO_x values between 0 and 10 ppb. The second bin is for NO₂ data with NO_x concentrations between 10 and 20 ppb, and so on. Then, an average value of NO₂ is computed for each bin. The results are plotted in Fig. 4 in a log-log form. From the plot, it is clear that there is a simple relation between NO₂ and NO_x mixing ratios. With a simple linear fitting (on the log-log relation), we can derive the following empirical formula to estimate NO₂ from NO_x

$$NO_{2_FIT} = \alpha NO_x^\beta \quad (1)$$

Where $\alpha = 0.901325$, $\beta = 0.756209$ are derived using the data for bins between 200 and 500 ppb of NO_x, which is the zone of the log-log plot mostly linear (the region between vertical dotted lines in Fig. 4). For higher values of NO_x, the linearity is slightly broken, and the bins contain little data (less than 20), and therefore they are less reliable for the fitting. For values of NO_x lower than 200 ppb the linearity (in the log-log plot) is also broken, and those points are therefore excluded. Due to this, the linear fit underestimates the NO₂ for low values of NO_x, making this approach better suited for medium to elevated concentrations of NO₂, which are the most relevant concentrations from the health impact perspective.

Using this simple empirical relation, we derived NO₂ concentrations from the simulated NO_x, and we transformed them to micrograms/m³ (assuming a standard temperature of 25 °C), given that these are the most common units used for NO₂. The time evolution for the different zones is represented in Fig. 5, and the corresponding statistics are in Table 3.

NO₂ 25%SMOIS	BIAS (micrograms/m ³)	RMSE (micrograms/m ³)	IOA	Correlation coefficient <i>r</i>
Zone 1	1.83	41.64	0.75	0.61
Zone 2	-25.03	40.62	0.74	0.66
Zone 3	-2.79	32.30	0.78	0.64
Zone 4	-21.10	26.17	0.57	0.60
Zone 5	-17.08	41.78	0.70	0.54
All Zones	-6.92	32.39	0.79	0.65
NO₂ No city				
Zone 1	5.31	46.11	0.74	0.62
Zone 2	-19.29	41.78	0.74	0.60
Zone 3	18.23	47.83	0.70	0.64
Zone 4	-21.21	26.85	0.55	0.51
Zone 5	14.31	56.81	0.69	0.57

All Zones	4.49	38.41	0.77	0.65
<i>NO₂</i> <i>100%SMOIS</i>				
Zone 1	-6.73	44.76	0.68	0.48
Zone 2	-28.58	48.52	0.57	0.41
Zone 3	-5.93	39.60	0.66	0.45
Zone 4	-23.28	30.49	0.47	0.19
Zone 5	-23.90	47.44	0.63	0.45
All zones	-12.77	38.02	0.68	0.49

Table 3. Statistics for the three simulations and the different zones for NO₂. RMSE stands for Root Mean Square Error and IOA for Index Of Agreement. In bold italic are the best scores.

Again, **25%SMOIS** is the simulation with the best scores among the three, with **No city** that tends to overestimate the concentrations and **100%SMOIS** with the worse IOA and correlation coefficient, coherently with what has been seen for NO_x. Most interestingly, these statistics are in the same range as those obtained by De la Paz et al. (2016) in their annual simulations, using a full photochemical model and more reliable emissions, and the correlation coefficients are similar, and in some cases better, than those obtained with more complex, state of the art approaches (Benavides et al. 2019). The approach needs to be tested over a wider range of situations, and it can be improved in many aspects (e. g. introducing different relations for hours with solar radiation and without, or using a more complex function as a polynomial, etc.), but this is an indication that it may represent a valid alternative to full photochemical models to get quick and rough estimates of NO₂ levels in cities.

4. Spatial distribution.

The spatial distribution of the NO_x concentration produced by the **25%SMOIS** (considered as the best set-up) is analyzed focusing on the two hours when the peak of NO_x is usually observed (0800 UTC and 1900 UTC). The three representative days of different stages of the episode mentioned in Part I are used in the analysis: the 24th(pre-episode), the 27th(first part of the episode), and the 30th (the second part).

At 0800 UTC (Fig. 6), the wind fields are qualitatively similar in the three days, as already observed in Part I. The type of circulation is still typical of nighttime, with a wind from NE in the elevated part of the city, and a flow mainly from North in the two topographical depressions. As a consequence, the highest concentrations are at the downwind edge of the city center (zone 1) and in the western topographical depression (zone 5). This pattern is common to the three days, but the difference is in the concentration levels that are very high on the 27th and in particular the 30th. Figure 7 shows a vertical section of the concentration through an SW-NE transect through the city (see Fig. 1) for the NO_x concentration and the turbulent kinetic energy (TKE). The TKE is a key variable because the vertical diffusion coefficients are proportional to the square root of TKE multiplied by a length scale. The height where the TKE becomes very small indicates the height up to where the pollutants can be mixed, and it is, therefore, a measure of the PBL height.

In the figures, it is clear the reduction in PBL height between the period before the episode (24th) and during the episode (27th and 30th). Given that at 0800 UTC the atmosphere is still stable, and the surface heating is still too weak to break the nocturnal inversion, the TKE profile is determined by the balance between shear (and drag in the canopy) production and buoyancy destruction. Figures S1, S3, S5, S7 show the vertical profiles of the TKE budget, at 0800 UTC plotted for two points, one on the hill (S1, S3) and another in the topographical depression west of the city center (S5, S7). On the 30th, not only the atmosphere is more stable, but also the wind is much weaker and so the shear production is lower than on the 27th. It is also interesting to see that in the topographical depression West of the city (roughly zone 5), the concentration is much higher than in the depression East of the city (roughly zone 3), although the stability and turbulent regime are similar. This is mainly due to the advection of pollutants from the city center (zone 1). Moreover, in zone 1, the PBL is higher than in the two depressions, due to lower stability and stronger shear production.

At 1900 UTC (Fig. 8), the highest concentrations are still simulated in the same parts of the city where the maxima are in the morning, but elevated concentrations are also present in the Eastern part of the city, in particular the 27th and the 30th. On the 27th, the model has problems in simulating the position of the high NO_x concentrations in zone 1, something that may explain the underestimation shown in the time series (Fig. 3). The vertical sections indicate the importance of the TKE profile (Fig. 9). Also at that time, the atmosphere is already stable (no surface heating), and the height of the PBL is mainly given by the balance between shear production and buoyancy destruction (Fig. S2, S4, S6, S8). However, at this time, differently than what happened in the morning, there is NO_x concentration above the PBL height. This pollutant has been mixed up there during the central hours of the day when the PBL was thicker. In particular, on the 30th, characterized by very low winds, a large amount of the NO_x mixed up during the central hours of the day is still above the city in the evening.

In summary, model results are physically meaningful and broadly agree with the measurements, despite some local discrepancies. To perform a more detailed validation we would need an emission inventory with better spatial distribution, and a chemical module, both things beyond the scope of this article which is just to investigate the effect of the most important dispersion features in the region and these kinds of episodes.

5. Policy relevant analysis

In this section, we use the model to perform different types of analysis relevant both from the scientific and policy point of view. Note how we refer here to "pollutant" generically instead of NO_x because the analysis that will be done can be applied to any primary weakly reactive pollutant emitted from the same sources that emit NO_x (mainly traffic).

5.1 Impact of the previous day's emissions.

One of the most relevant questions in these types of episodes is to investigate whether the emissions of one day have an impact on the concentration levels of the following day. This is a key question to understand, for example, how many days in advance measures should be taken to avoid elevated levels of air pollution.

With this aim, we used seven different tracers, one per day of the week. Given that the tracers are considered non-reactive we can linearly add the tracer emitted in each day to get the total concentration. Figure 10 shows the percentage of the modeled concentration averaged at the measurements locations for zones 1,2,3, and 5 (Fig.2) due to previous and current-day emissions for the first 12 hours of the 24th, 27th, and 30th, respectively. As it can be seen, for all the days and all the zones, the impact of the emissions of the previous day decreases with time and becomes zero after 12 hours. This is because, even if weak, the wind blows all the time in the same direction, and this is enough to push all the pollutants emitted the day before several tens of kilometers away from the city. There are, nevertheless, differences between the periods and the zones, being the zone sin the depression (2 and 5) and the days of the episode (27th and in particular 30th) those where the pollutants emitted the previous day stay a few hours more indicating worse dispersion conditions. In any case, this is not enough to keep the pollutants more than 12 hours. This can be understood by looking at Fig. 11, where the concentrations at 0900 UTC of the 27th and the 30th of the pollutants emitted the previous day are plotted. Clearly, at that time the largest fraction of the pollutants emitted the previous day are already tens of kilometers away of the city on the morning of the following day.

The consequence of this is that even during an episode of strong atmospheric inversion, in Madrid, there is no accumulation of pollutants from one day to another. This is in line with the findings of Borge et al. (2018a) who show that the NO₂ concentration levels can be explained only with the PBL height of the same day measured in the sounding of Barajas airport, without the need to account for the concentration levels (or atmospheric conditions) of the previous days. As mentioned above, this is mainly because there is almost no reversal of the flow direction between day and night. In conditions of strong inversion, only katabatic or drainage flows can develop during the night, but the mountain ridge close to Madrid does not develop strong enough anabatic or valley flows during the daytime (Roman-Cascon et al. 2019). As a consequence, no recirculation is present in such conditions.

5. 2 Impact of the temporal distribution of emissions within the same day.

In the previous section, we have shown that there is no day-to-day pollutant accumulation. Here we analyze how the pollutant accumulation behaves within the same day. To do this we used three tracers. One tracer is emitted during the morning rush hours (three hours, from 0700 to 1000 UTC), and represents 18.5% of the total tracer emitted in the domain during a normal working day. The second tracer is emitted during the evening rush hours (three hours from 1800 to 2100 UTC) and represents 19.5% of the total tracer emitted during a typical working day. The third tracer is emitted in the remaining 18 hours and accounts for 62% of the daily total emissions. As explained

above, thanks to the linearity of the passive tracers, the sum of the three tracers is equivalent to a tracer emitted during 24h. In Fig. 12, the time evolution of the percentage of every single tracer to the total concentration is plotted for the four zones, and for the three days analyzed. As expected, the tracers emitted during the rush hours have a strong impact on the maxima, in all the zones and the three days. However, while during the period before the episode (24th) the impact of the rush hours morning emissions is mostly limited to the hours of the peak, and then the morning pollutants are quickly dissipated, during the episode, in particular the 30th, pollutants emitted during the morning accumulate, due to the worse dispersion conditions, and have an influence also in the central hours of the day. This influence is bigger for zones 2 and 5, which are in the topographical depressions. From these results, we can hypothesize that to reduce the peak it is important to reduce the emissions during the rush hours. To quantify this, five hypothetical scenarios have been investigated:

A) 25% of the emissions of the morning rush hour have been moved to the 1200-1500 UTC window

B) 50% of the emissions of the morning rush hour have been moved to the 1200-1500 UTC window

C) 25% of the emissions of the evening rush hour have been moved to the 1200-1500 UTC window

D) 50% of the emissions of the evening rush hour have been moved to the 1200-1500 UTC window

E) 25% of the emissions of the morning rush hour and 25% of the emissions of the evening rush hour have been moved to the 1200-1500 UTC window.

Note that the total amount of tracer emitted to the atmosphere is fixed for all the scenarios. As shown for the 30th in Fig. 13, moving the emissions to a period more favorable for the dispersion of the pollutants, like the central hours of the day, significantly reduces the peaks, and only marginally increases the concentrations in the central hours of the day. Importantly not only the peaks are reduced but also the mean daily concentration is reduced (Fig 15) both before the episode (22nd-25th, left bars) and during the episode (26th-30th, right bars). Here we can see that the daily mean concentration can be reduced up to 8-12%, depending on the zone, acting in particular on the evening emissions (Scenario D), while acting only on the morning ones leads to smaller reductions of the daily mean concentrations. For the peaks, on the other hand, the emission reductions in the evening rush hours translate to a reduction in the concentrations of 15-20 % (scenarios C and E), and of 30-35% for scenario D, with small differences between the period before and during the episode. On the other hand, for the morning peak, there is a strong variability between the period before and during the episode, with reductions of less than 10% before, and up to 40% during the episode when half of the morning rush hour emissions are moved to the central hours of the day. This is likely because the morning peak is during a period of the day when the PBL is rapidly growing, so a small difference in the rate of growth, induced, for example, by

different strength of the nocturnal inversion, may have a big impact on concentrations. On the other hand, at the time of the evening peak, the PBL has already collapsed, and it has a relatively constant-in-time behavior. Moreover, the percentage reduction shows a spatial variability, with the smallest reductions in Zone 5, located in the topographical depression where pollutants emitted before the rush hour can accumulate more easily, and the biggest reductions in zone 1, on the hill, where there is less accumulation of pollutants.

If we apply the simple empirical formula (1) to deduce NO_2 concentration from NO_x , the daily mean NO_2 concentration would be reduced by 6-7%, but the reduction of the peaks would still be significant with values up to 35% for the morning peak in Zone 1 for scenario B during the episode, and values close to 20-25% for the evening peak in all the zones (see Fig. S9). It is worth noting here, that a reduction of concentration by 10% in the peaks of NO_2 would have been enough to reduce the number of hourly exceedances of the $200 \mu\text{g m}^{-3}$ limit value of NO_2 by almost 60% during the episode, and reducing the peak by 20% would have caused a reduction of the exceedances by almost 90%.

5.3 Impact of the spatial distribution of emissions.

The relation between emission and pollutant concentrations depends on the structure of the PBL where the pollutants are emitted and the wind speed. As shown before, these conditions have a strong diurnal variability. However, the analysis of the meteorological fields presented in Part I, and the pollutant distribution shown in Section 3 of this paper, indicate that there is also a strong spatial variability of the PBL structure in the region of Madrid during this type of episode. Figure 15 shows the map of the ratio between the maximum concentration and the maximum amount of pollutant emitted during the first part of the day (e.g. including the morning peak), and the second part (evening peak). Mathematically, for the first part of the day this ratio $R_{i,j}$ for point i,j is computed as follows:

$$R_{i,j} = \frac{\max_{t=[0,12 \text{ UTC}]} (C_{i,j,t})}{\max_{t=[0,12 \text{ UTC}]} \left(\frac{E_{i,j,t} h}{\Delta z_{i,j} V_{i,j}} \right)}$$

Where $C_{i,j,t}$ is the pollutant concentration at the lowest model level in grid cell i,j at hour t , $E_{i,j,t}$ is the emission at grid point i,j and hour t , $\Delta z_{i,j}$ is the thickness of the lowest model level at grid point i,j , and $V_{i,j}$ is the fraction of air volume (e. g. excluding the buildings) of the lowest model level at grid point i,j , and h is one hour in seconds (3600s). A similar expression has been used for the second part of the day (12-24 UTC). A low value of this ratio indicates that most of the pollutants emitted in the grid cell left it, and so that there are favorable conditions for the dispersion. This is the case, for example of the urban area on the hill between the two topographic depressions, during most of the periods investigated. As shown before in that part of the city, the stability is relatively weaker than in the topographic depressions (see also Fig. S1-S8), in particular the 27th December, and the contribution of the shear in the production of the TKE is relatively large so to induce a PBL height of 100 m or more. The pollutants emitted in the urban

area on the hill are dispersed well. On the other hand, areas with a high value of this ratio indicate either that they have conditions not favorable for the dispersion, or that the local emissions are low and the pollutants levels are the result of advection from other regions. The latter is the case, for example, of the small urban areas south of the city. However, in the two topographical depressions in the urban area, one of them hosting a major highway, the high values of the ratio, in particular in the evening, indicate poor dispersive conditions, typically due to low wind and low PBL height.

6. Summary and conclusions.

We used a passive tracer implemented in WRF, with the emissions correspondent to NO_x , to investigate the most important dispersion features during a period of persistent wintertime inversion in Madrid. The comparison against NO_x measurements has shown that the model can reproduce the main dispersion patterns. However, there are also discrepancies, that can be partially attributed to the uncertainty in the emissions inventory, particularly because the episode studied was during the Christmas period when a large part of the citizens is on vacation, something not accounted for in the emissions used that are typical of a working period.

By comparing the results obtained with a simulation with the multilayer urban canopy parameterization BEP-BEM and one without, it has been possible to conclude that the presence of the city plays a relevant role in the modulation of the strength of the thermal inversion, and therefore the pollutant concentrations. However, the value of the soil moisture is equally important, because it directly affects the surface energy partitioning and the atmospheric conditions around the city that are then advected over the urban area.

We also introduced a new methodology to derive NO_2 from NO_x , based on an empirical relation fitted to two months of measurements. Statistics obtained by comparing model estimates obtained with this methodology with experimental data are similar to those obtained with photochemical models by other authors (over Madrid and other cities). This indicates that this methodology can represent a much cheaper alternative to full photochemical models to get rough estimates of the NO_2 concentration in cities, and also that a correct reproduction of the meteorology is the key element to correctly simulate the concentration even of a reactive pollutant like NO_2 . It is, therefore, worth to carefully represent the meteorology, if needed by tuning uncertain parameters like soil moisture, representing the city, and having a high vertical resolution to resolve at best the vertical structure of the PBL.

In summary, we consider that these results are meaningful and can be used to derive information that can help policymakers in the definition of which are the best actions to take to improve air quality during such types of episodes. In particular:

- The simulation results have shown that at least during the episode analyzed, there was no day-to-day accumulation of pollutants. The concentration levels reached in the city during one day are only induced by the pollutants emitted the same day. This is in contrast with what has been observed in other cities like

Santiago de Chile (Saide et al. 2011) where the impact of previous day emission is substantial. We identified the main reason for this in the fact that the wind, even if weak, blew almost always in the same direction during the whole period because during winter anabatic winds are usually weak (Roman-Cascon et al. 2019), something reinforced by the strong atmospheric inversion over the plain. Moreover, compared to Santiago de Chile or Salt Lake City, where there are mountain ranges on both sides of the city, Madrid has only a mountain range on one side (and farther) making it easier for the pollutants to evacuate the urban area. Therefore, the fact that very high pollutant concentrations were observed for several consecutive days, is not an indication of the accumulation of pollutants, but it is just because the atmospheric conditions (strong inversion) persisted. Pujadas et al. (2000) investigated another thermal inversion episode using measurements. Even if in that episode there was a clear change in wind direction between night and day, authors also concluded that local circulations can spread a significant volume of pollutants outside of the city also in winter stable conditions. This fact, if confirmed also for other periods, has important policy consequences, because *the pollutant concentration levels of one day are independent of the pollutant concentration levels of the previous day*. Therefore, *the decision about the type of actions to adopt to improve air quality should not be based on the observations of the previous day* (or on the number of consecutive days with high concentration levels, as it is currently done), *but, instead, on weather and air quality forecasts*. Most importantly, *pollutant levels of one day can largely be reduced controlling only the emissions of the same day*.

- Our results also show that pollutants emitted at different times have different impacts on concentration levels. In particular, the pollutants emitted during the rush hours are almost completely responsible for the morning and evening peaks, and have also a significant influence on the daily mean, despite representing less than 40% of the total and being emitted only 6 hours out of 24. *This is because, during winter, rush hours are at a time of the day when the PBL height is still very low, and the dispersion conditions are not favorable*. Simple experiments where a fraction of the morning and/or evening emissions were moved to the central hours of the day when the PBL is thicker, and so the pollutants can be dispersed better, without changing the total amount of the emission, resulted in a significant decrease of the peaks, and, in a smaller amount, of the daily mean. In other words, *“Flattening the curve”* of traffic, as suggested by Ratti (2020), may be used not only to reduce traffic jams, and the amount of pollutant emitted to the atmosphere, but *also to move part of the emissions to periods of the day with most favorable dispersion conditions*.
- Model results have also shown that not only the time of the day is important but also the specific location in the city. The central and elevated part of the city has a higher PBL and stronger wind during most of the days of the episode, and so more favorable dispersion conditions compared to the two topographical depressions (about 100 m lower) East and West of the city center. In the latter areas, there is an accumulation of cold air that leads to stronger atmospheric

stability, which prevents the dispersion of the pollutants emitted there. The most important consequence of this is that a slight increase of emissions in the central and elevated part of the city will likely have a much smaller impact on concentrations, than what the same increase would have in the topographical depressions. In other words, *the pollutant concentration in the depressions, where the dispersion conditions are not favorable, is much more sensitive to changes in the emissions, than in the central and elevated part of the city.*

Marsden et al. (2020) pointed out that significant reductions in carbon emission can be obtained by introducing innovations in people's behavior. The disruptive changes introduced in our lives by the tragic COVID-19 pandemic contributed to create the technological infrastructures and break cultural resistance, that allows new and more flexibles working arrangements. This new situation can form the basis for the installation, after the pandemic, of *behavioral changes that not only reduce the total amount of emissions, but also shift part of them to time windows, and city areas, most favorable for pollutant dispersion.* This is something that could be achieved in several ways, like modifying the working schedule, allowing people to work from home during all or part of the day, limiting the circulation of highly pollutant heavyweight vehicles to selected hours of the day, or modifying the opening and closing hours of the shops, restaurants, or schools. Another option would be to introduce flexible fees that people have to pay to drive on busy roads, as is done in Singapore (Ratti, 2020), and adjust them to the meteorological conditions. For example, the meteorological model could be part of a digital platform, that would increase the fees to drive on the parts of the city and at the time of the days when the dispersion conditions are less favorable.

Finally, we would like to stress that, even if the results are restricted to the region of Madrid and cannot be generalized, the methodology applied is indeed universal and can be applied to any city in similar conditions.

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Figure 1. Upper left: Topography and air quality stations (red dots are traffic stations, blue are background stations, and green are rural stations). The diagonal solid line represents the vertical section of Fig. 7 and 9; Upper right: time evolution of the spatially averaged emissions of NOx for a week-day (solid line) and for week-end (dashed); Lower left spatial distribution of the emissions at 0800 UTC; Lower right spatial distribution of emissions at 1900 UTC.

Figure 2. Air quality zones of Madrid's city council. Color of stations is the same of Fig.1.

NO_x

Figure 3. Time evolution of NO_x concentration averaged over all the stations that belong to zone 1, 2, 3, and 5 (from top to bottom). Diamonds represent measurements, and solid lines the three simulations (see legend). The vertical dashed lines indicate the three days used to characterize the different phases of the episode.

Figure 4. Log-log plot of hourly measured NO_2 vs NO_x for November and December 2016 in the 24 air quality stations of Madrid. The data are binned based on NO_x values, with a bin size of 10 ppb. The solid line is the linear fit, obtained with data between the two vertical dashed lines. The vertical segments centered on each point represents one standard deviation.

Figure 5. Same as Figure 3, but for NO₂. Values here have been transformed to micrograms/m³.

Figure 6. NO_x distribution (left) and wind field (right) produced by 25%SMOIS at 0800 UTC the 24th (top), 27th (middle) and 30th (bottom). The small boxes represent the stations, and the color the concentration. Note the logarithmic scale. Wind field is at the lowest model level.

Figure 7. Vertical section of the NO_x concentration (left) and Turbulent Kinetic Energy (right) through a SW-NE transect indicated by the solid line in Fig. 1. Top is for the 24th, middle for the 27th and bottom for the 30th. The black bar indicates the city. The boxes are the stations that are on the transect, and the colors indicate the concentration. Note the log scale of the NO_x plots.

Figure 8. Same as fig. 6, but for the 1900 UTC

Figure 9. Same as fig. 7 but for the 1900 UTC.

Figure 10. Percentage of tracer concentration due to same day emissions (green) and previous day emissions (orange) during the first 12 hours of the day, for zones 1, 2, 3, and 5 (from top to bottom), for the 24th (left), 27th (center), and 30th (right).

Tracer day before 0900 UTC 30/12

MADRID

Tracer day before 0900 UTC 27/12

Figure 11. Concentration of the pollutants emitted the day before in the morning of the following day. Above, emissions of the 26th at 0900 UTC of the 27th. Below emissions of the 29th at 0900 UTC of the 30th.

Figure 12. Percentage of the tracer concentration in zone 1, 2, 3, and 5 (from top to bottom), for the 24th (left), 27th (center), and 30th (right). Due to the morning rush hour emissions (red), evening rush hour emissions (yellow), and emissions outside the rush hours (blue), for the 24 hours of the day.

Figure 13. Time evolution of the pollutant concentration for zone 1, the 30th, for the five emission scenarios: Triangles represent the base case, and solid line the scenario. See text for the definition of the scenarios.

Figure 14. Percentage reduction of NO_x daily mean (left column), morning peak (center), and evening peak (right) due to the different scenarios for the period before the episode (22-25, left bars), and during the episode (26-30, right bars), for zones 1,2,3, and 5 from top to bottom.

Figure 15. Spatial distribution of the ratio between the maximum pollutant concentration and the maximum amount of pollutant emitted in the lowest model level in one hour, at the time of the morning peak (0800 UTC, left), and of the evening peak (1900 UTC, right), for the 27th (top) and the 30th (bottom).

Supplemental Information

0800 UTC 27/12

Point on the hill (683m a.s.l.)

Figure S1. Vertical profile of NOx (upper left), TKE (upper right), Potential Temperature (lower left), and the components of the TKE budget, for a point on the hill, at 0800 UTC 27/12 for simulation 25%SMOIS

Point on the hill (683m a.s.l.)

Figure S2. Same as Fig. S1, but at 1900 UTC of the 27/12. Note that the horizontal axis of the TKE budget figure is different than Fig. S1.

0800 UTC 30/12

Point on the hill (683m a.s.l.)

Figure S3. Same as Fig. S1, but at 0800 UTC of the 30/12. Note that the horizontal axis of the TKE budget figure is different than Fig. S1.

Point on the hill (683m a.s.l.)

Figure S4. Same as Fig. S1, but at 1900 UTC of the 30/12. Note that the horizontal axis of the TKE budget figure is different than Fig. S1.

0800 UTC 27/12

Point in the topographic depression (586m a.s.l.)

Figure S5. Same as Fig. S1, but at 0800 UTC of the 27/12, and for a point in the depression west of the city center. Note that the horizontal axis of the TKE budget figure is different than Fig. S1.

Point in the topographic depression (586m a.s.l.)

Figure S6. Same as Fig. S5, but at 1900 UTC of the 27/12. Note that the horizontal axis of the TKE budget figure is different than Fig. S5.

0800 UTC 30/12

Point in the topographic depression (586m a.s.l.)

Figure S7. Same as Fig. S5, but at 0800 UTC of the 30/12. Note that the horizontal axis of the TKE budget figure is different than Fig. S5.

Point in the topographic depression (586m a.s.l.)

Figure S8. Same as Fig. S5, but at 1900 UTC of the 30/12. Note that the horizontal axis of the TKE budget figure is different than Fig. S5.

Fig. S9. Same as Fig. 14, but for NO₂