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Combining Modelling with Clustering Analysis to Characterise Exceedances of NO_2 Concentrations in the Metropolitan Area of Buenos Aires

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Abstract: Ground-level air pollutant concentrations are known to present marked spatial variations in the urban atmosphere. In large urban areas, air quality models can contribute to assess potential exposure concentration levels. The Metropolitan Area of Buenos Aires (MABA), the third mega-city of Latin America, concentrates around 13 million inhabitants in approximately 3830 km^2. However, despite of its extension and the potential impact of air quality on human health, only a few monitoring campaigns have measured ground-level nitrogen dioxide (NO_2). In this work, an urban scale atmospheric dispersion model (DAUMOD-GRS) is applied in the MABA, considering three years of hourly meteorological information and a high resolution (1 h, 1 km^2) emission inventory to analyse potential exceedances of urban background NO_2 hourly concentrations of its corresponding air quality standard (200 ppb) and the one suggested by the World Health Organization (110 ppb). When the WHO criterion is considered, despite of the low (<2%) maximum annual frequency of events, the surface area where at least four exceedances per year occur increases 34 fold, from 58 km^2 to 1979 km^2. In order to classify leading conditions and their spatio-temporal distributions, a clustering analysis including all relevant variables associated with exceedances is performed. Results indicate that exceedances can be grouped into six families with different characteristics and marked spatio-temporal distributions.

***Keywords**:* air quality standard, Buenos Aires, clustering analysis, nitrogen dioxide, urban scale model.

1 INTRODUCTION

The presence of nitrogen dioxide (NO_2) at relatively high concentrations can cause adverse effects on human health and the environment. Epidemiological studies have shown that bronchitis symptoms in asthmatic children increase with prolonged exposure to NO_2 (Hoek et al., 2013). In addition, nitrogen dioxide is a major source of nitrate aerosols, which constitute a considerable fraction of fine particulate matter that may be carcinogenic to humans (WHO, 2016). NO_2 can be formed from chemical reactions involving nitrogen oxides (NO_x) and volatile organic compounds (VOCs), while a part of it is emitted directly to the atmosphere, mainly from fossil fuel combustion sources. Therefore, large urban areas not only have the largest emission sources but also are susceptible to receive the greatest negative impact, due to the population size that may be exposed to the air pollution they cause. To guarantee that the air quality is adequate to protect citizens health, local environmental agencies determine the air quality standards (AQS) that must not be exceeded with a given frequency (e.g., the 98th percentile) during a period (e.g., three years), and design a monitoring network for controlling its attainment. However, air quality monitoring stations are expensive and therefore urban areas often have only a few measurement sites for this purpose. In addition, although they provide precise information on the pollutant concentration at the sites, these may not be representative of the whole urban area where marked spatial variations of emissions in combination with chemical transformations and dispersion, lead to large spatial variations in the pollutant concentrations. Air quality models allow to complement this information in any place and period whenever there is adequate input data to feed them. High resolution (1 km^2, 1 h) long-term (i.e., several years) simulations produce large sets of model results containing valuable information on the type of model solutions that is often under analysed.

Clustering is one way to extract qualitative information about the conditions leading to pollutant concentration exceedances and their spatio-temporal distributions. It reduces the dimensionality of the analysis by identifying groups of systematically co-occurring conditions. In air quality studies, it has been widely applied to analyse observed pollutant concentrations (e.g., Flemming et al., 2005; Beaver and Palazoglu, 2006; Rimetz-Planchon et al., 2008; Unal et al., 2011; Khedairia and Khadir, 2012; Wang et al., 2016). However, despite of its potential to produce low dimensional descriptions of large datasets, only a few works have used it in combination with modelled gridded pollutant concentrations (e.g., Jin et al., 2011).

The Metropolitan Area of Buenos Aires (MABA) presents a population of around 13 million inhabitants over a surface of approximately 3830 km^2. Despite of the emission sources of air pollutants that have the potential to cause damage in human health and the environment, there are only three air quality monitoring stations within the city which measure NO_2 concentrations regularly, one since 2006 and two others since 2009 (http://www.buenosaires.gob.ar/agenciaambiental). On the other hand, there have only been a few air quality measuring campaigns in the city of Buenos Aires (e.g., Bogo et al., 1999; Mazzeo et al., 2005). Hence, our knowledge of the spatio-temporal distribution of the NO_2 concentration in the MABA is scarce. In this work, we apply the urban scale atmospheric dispersion model DAUMOD-GRS (Pineda Rojas and Venegas, 2013a) to obtain high resolution (1 km^2) maps of potential exceedances of hourly NO_2 concentrations resulting from NO_x and VOCs emissions in the MABA, considering the local AQS and the Air Quality Guideline suggested by the World Health Organisation (WHO, 2005). The results obtained are studied applying clustering analysis in order to characterise the emission and atmospheric conditions that lead to such exceedances and to unveil patterns of spatio-temporal distributions. The objective is to explore the potential of this tool to qualitatively describe leading co-occurring conditions of pollutant concentration exceedances.

2 METHODOLOGY

2.1 The DAUMOD-GRS Model

The DAUMOD-GRS is an atmospheric dispersion model that results from the coupling of the DAUMOD (Dispersión Atmosférica Urbana - MODelo) model (Mazzeo and Venegas, 1991) and the GRS (Generic Reaction Set) simplified photochemical scheme developed by Azzi et al. (1992). DAUMOD is based on the bidimensional equation of diffusion (Arya, 1999) and assumes stationary conditions and that there is no transport of pollutant through the upper boundary of the plume. It was originally developed to estimate urban background concentrations of primary pollutants emitted to the atmosphere from area sources. This model has been used extensively to assess diverse aspects of atmospheric pollution in the MABA (e.g., Venegas and Mazzeo, 2006; Pineda Rojas and Venegas, 2009; 2013b). The GRS allows to estimate the concentrations of nitrogen dioxide (NO_2) and ozone (O_3) resulting from NO_x and VOCs emission sources with only seven reactions. Due to its simplicity and reasonably good performance, it has been widely adopted by the urban air quality community as an acceptable option to assess the photochemical transformations of NO_2 and O_3 at urban scale (e.g., Wallace et al., 2008; Pournazeri et al., 2014; Chaney et al., 2011; Miranda et al., 2016; Kimbrough et al., 2017).

A detailed description of the DAUMOD-GRS model can be found in Pineda Rojas and Venegas (2013a). The statistical evaluation of the model (i.e., the comparison of modelled concentrations with observed values) has shown a good performance of the DAUMOD-GRS to estimate hourly NO_2 and O_3 concentrations in the MABA (Pineda Rojas and Venegas, 2013a; Pineda Rojas, 2014).

2.2 Simulation Conditions and Input / Output Data

Simulations are performed for a domain of 85 km x 75 km including the MABA, with a horizontal resolution of 1 km x 1 km and a temporal resolution of 1 hour. Model input data consist of: i) three years (2006-2008) of hourly surface and sounding meteorological data from the stations located in the Domestic and International Airports, respectively, ii) high resolution emission rates of NO_x and VOCs from area sources in the MABA (Venegas et al., 2011), and iii) pollutant concentration boundary conditions. Since the MABA is surrounded by non-urban areas and due to the lack of data, "clean air" concentrations are considered for the regional background levels, except for O_3 where a concentration value of 20 ppb is considered based on a previous study (Mazzeo et al., 2005). At each hour, horizontally homogeneous meteorological conditions are assumed. In this way, only the emissions vary spatially.

At each hour and each receptor, the ground-level NO_2 hourly concentration value is compared with its corresponding air quality standard for the MABA (200 ppb) and with the Guideline suggested by the World Health Organisation (110 ppb) (WHO, 2005). In this first assessment only diurnal hours are considered. If an exceedance of any of these levels occurs, this concentration is stored together with the receptor (X,Y), date (YYYYMMDD) and hour (H) of occurrence, and seven relevant model input variables at that time: wind speed (WS), wind direction (DIR), air temperature (T), atmospheric stability class (KST), sky cover (SC), local NO_x emission rate (QNO_x) and local VOCs emission rate (QVOC). Therefore, output data consist of twelve variables for each receptor and event. For simulations of many years, this may lead to a significant amount of data. Considering all receptors, a total of 7,294 exceedances of the local AQS (200 ppb) and 79,440 of the WHO guideline values (110 ppb) are obtained from this 3-year simulation. The latter are considered as the objects in the clustering analysis.

2.3 Implementation of Clustering Analysis

Clustering analysis is a widely used statistical method that aims to group high (M) dimensional objects by their similarity in an unbiased way. The k-means algorithm (e.g., Lu et al, 2006) is applied in order to classify N = 79,440 objects, defined as the set of conditions leading to an event. A cluster is represented by its centroid: the mean over each of the M variables of all objects belonging to it. The algorithm starts with a random distribution of *k* centroids in a M-dimensional space and iteratively repeats two steps until a stable situation is reached: i) assign each object to the cluster with the closest centroid and ii) re-calculate the centroid of each cluster as the geometrical mean of all objects belonging to it.

In this work, the algorithm is implemented using the Matlab (MathWorks, Natick, MA) function *kmeans*. In order to be able to use Euclidean distance between objects, all variables are normalized by their standard deviation and their mean subtracted (with their mean and standard deviation applied over the whole dataset). Wind coordinates are transformed into their x and y components (Wx and Wy, respectively) that span a Euclidean space. Thus, each object is represented by a point in a M=8 dimensional space with normalized dimensions of the variables: H, Wx, Wy, T, KST, SC, QNO_x and QVOC. To avoid local minima, n=100 repetitions of the algorithm are performed with different random initial conditions, and the one with lowest sum of object-to-centroid distances is kept. Note, however, that this commonly used improvement does not change the heuristic nature of the solution. The crucial user-defined parameter in k-means is the number of clusters *k* and there is no generally accepted way to set it. For the data used in this study, the focus is put into avoiding an overfitting situation, as represented by too many clusters, each specialized in a few events. To find the optimal *k*, the number of clusters is progressively incremented starting from the minimum *k = 2* until the first cluster grouping less than N/(2*k*) objects appears. This criterion aims to find the largest possible number of different clusters of regular size. The highest *k* that only includes highly populated clusters according to this criterion is *k* = 6, which is thus used throughout this work.

3 RESULTS

Figure 1 shows the annual mean number of situations (hours) when the NO_2 concentration is above the local air quality standard (AQS) and the WHO level at each receptor. Exceedances of the local

AQS occur in the most urbanised area of the MABA, with a very low maximum frequency of occurrence (13 events/yr = 0.3%). When a more restrictive criterion is considered, the surface area where at least four exceedances per year occur increases 34 fold, from 58 km^2 to 1979 km^2 (see Figure 1.b), and the maximum frequency of occurrence of these events reaches 1.8%.

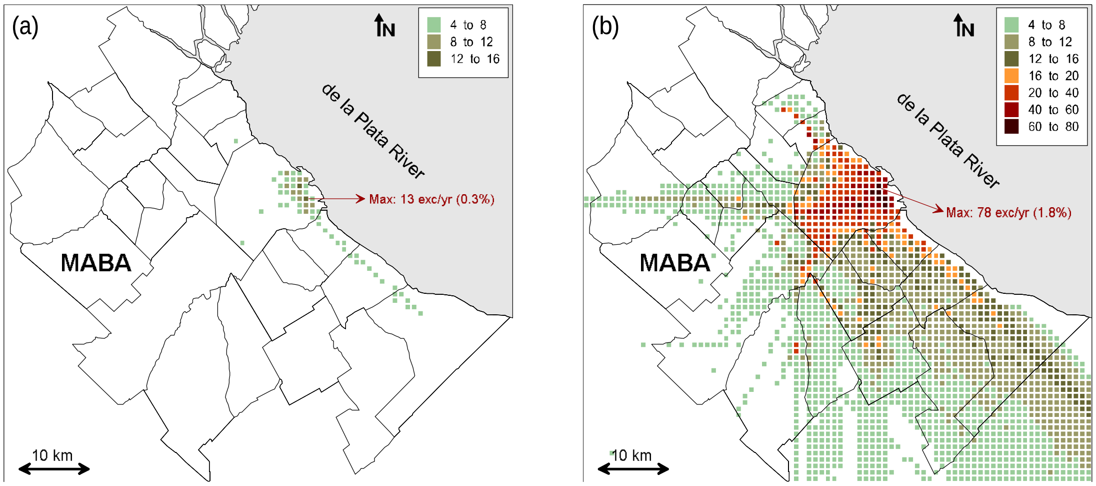


Figure 1. Annual mean (2006-2008) number of exceedances of hourly NO_2 concentrations of: (a) the local air quality standard (200 ppb), and (b) the guideline suggested by the WHO (110 ppb).

3.1 Description of Clusters

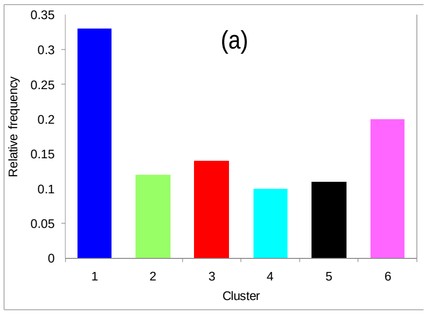
Figure 2 shows the spatial distribution of the dominant cluster, and Figure 3 presents the mean (a) and standard deviation (b) of the normalised variables [H, WS, DIR, T, KST, SC, QNO_x and QVOC] for each cluster. From Figure 2, the dominant cluster distribution clearly presents a spatial variation that is associated to that of the NO_x and VOCs emission rates, which are dominated by the traffic. Cluster 1 presents the largest number of objects (Figures 2.a) that are dominant in a large area southeast of the city of Buenos Aires (Figures 2.b), where the NO_2 exceedances occur with the lowest mean T and WS, and the highest mean KST (see Figure 3.a) and DIR from the NW sector (not shown). Cluster 2 dominates at the city of Buenos Aires with the greatest emission rates, where an exceedance occurs, on average, during comparatively later morning hours (Figure 3.a) and with winds from SW and NW sectors. Cluster 3 occurs in a narrow area close to the highway (relatively high emission rates) (Figure 2.b) and presents the greatest mean value of WS of all clusters (Figure 3.a). Cluster 4 dominates in an area west of the city (Figure 2.b) with the highest mean SC (Figure 3.a) and variable wind direction. Cluster 5 dominates at an area south of the city of Buenos Aires (Figure 2.b) with winds from the N, and this is the only cluster in which NO_2 exceedances occur at late evening hours (Figure 3.a). Finally, cluster 6 is mostly present to the southwest and further south of the city (Figure 2.b) with the largest mean value of T and the lowest KST (Figure 3.a) and variable DIR (not shown).

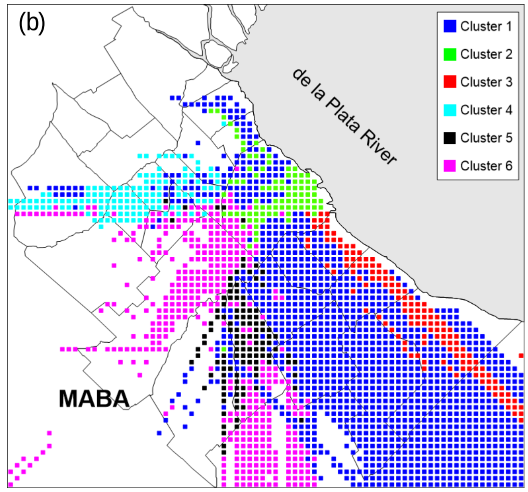
In summary, these results allow the association of spatial patterns of occurrence with qualitative footprints, evidenced by 2D projections in Figure 4. Cluster 2 is associated with high emissions, cluster 3 with the highest levels of WS, cluster 4 with the highest SC, cluster 5 with the latest hours and cluster 6 with the highest temperatures. Cluster 1, the most prevalent spatially, represents instead a baseline condition from which all others appear to deviate.

3.2 Spatio-temporal Distribution of Clusters

N = 79,440 events (situations of hourly NO_2 concentrations exceeding the WHO guideline value) take place during roughly one third of the days (i.e., 380 out of 1,095 days in the 3-year simulation). Cluster 2 (the one which dominates in the city of Buenos Aires with the highest emission rates) is present in at least one receptor most of the days in which exceedances occur, implying that it is present on the same days as other clusters. In contrast, the other clusters are distributed along a comparatively smaller fraction of days (see Figure 5.a) and tend not to co-occur (Figure 5.b).

Figure 2. (a) Frequency of occurrence of each cluster and (b) spatial distribution of the dominant cluster (i.e., group of similar conditions leading to exceedances of NO_2 concentrations of the WHO guideline).





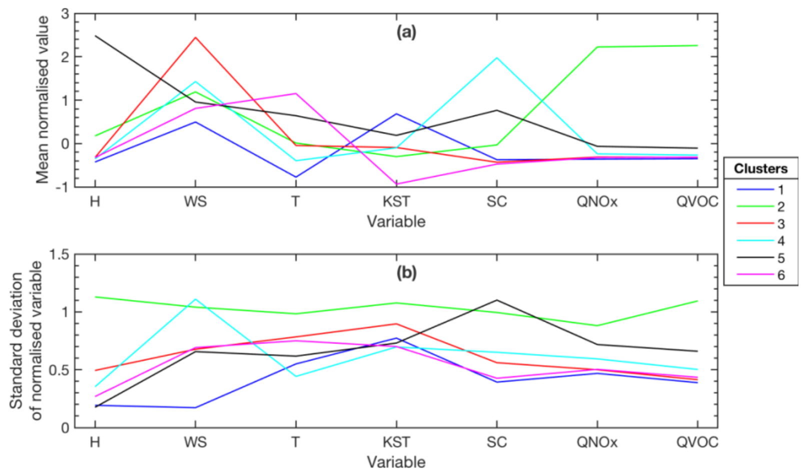


Figure 3. (a) Mean and (b) standard deviation of normalised variables of each cluster. [Note that the whole dataset has mean=0 and standard deviation=1]

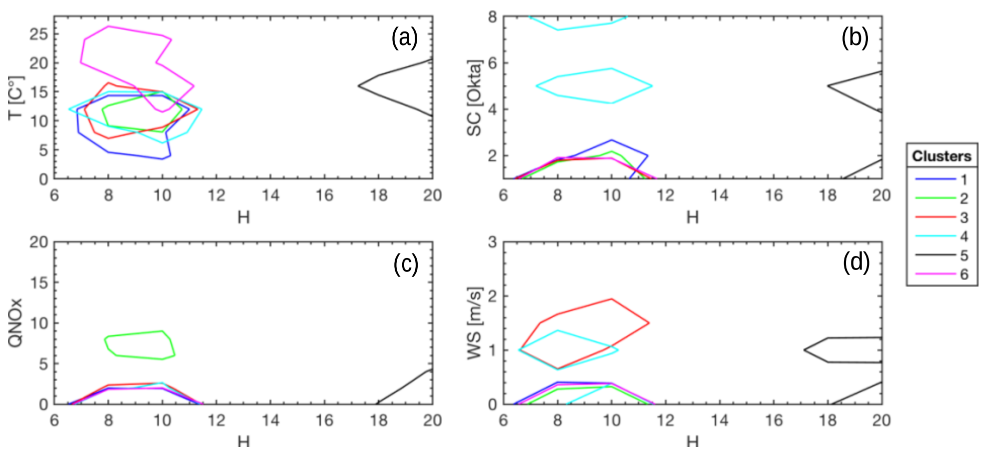
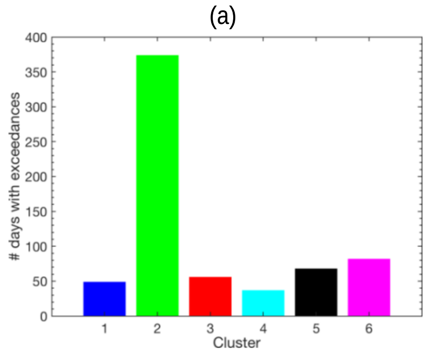


Figure 4. Curves containing 90% of objects of each cluster in the planes: a) T-H, b) SC-H, c) QNO_x-H and d) WS-H.



(b)

	1	2	3	4	5	6
1	1.00	1.00	0.00	0.00	0.00	0.02
2	1.00	1.00	1.00	0.95	1.00	0.95
3	0.00	1.00	1.00	0.00	0.02	0.00
4	0.00	0.95	0.00	1.00	0.03	0.00
5	0.00	1.00	0.02	0.03	1.00	0.03
6	0.02	0.95	0.00	0.00	0.03	1.00

Figure 5. a) Number of days in which clusters occur and b) fraction of days in which a given pair of clusters co-occur (red: > 0.95, black: < 0.03).

Figure 6 shows the spatial distribution of the dominant cluster for each month. Clusters 1 [the most frequent one in the whole dataset (Figure 2.a)] and cluster 4 dominate mostly during colder months (May to July). Cluster 2 dominates in the city of Buenos Aires throughout the year, but less frequently in November and December, when NO_2 exceedances occur only in a small area of the MABA due to enhanced dispersion conditions during warm months. Cluster 3 is mainly present in April, May and August to the southeast of the city. Cluster 5 (the only cluster with events occurring at late evening hours) dominates over large areas in April and September, and in a few receptors close to the urban area during warm months. Finally, cluster 6 dominates in warm months, mostly at suburban areas southeast of the city, alternating with cluster 1.

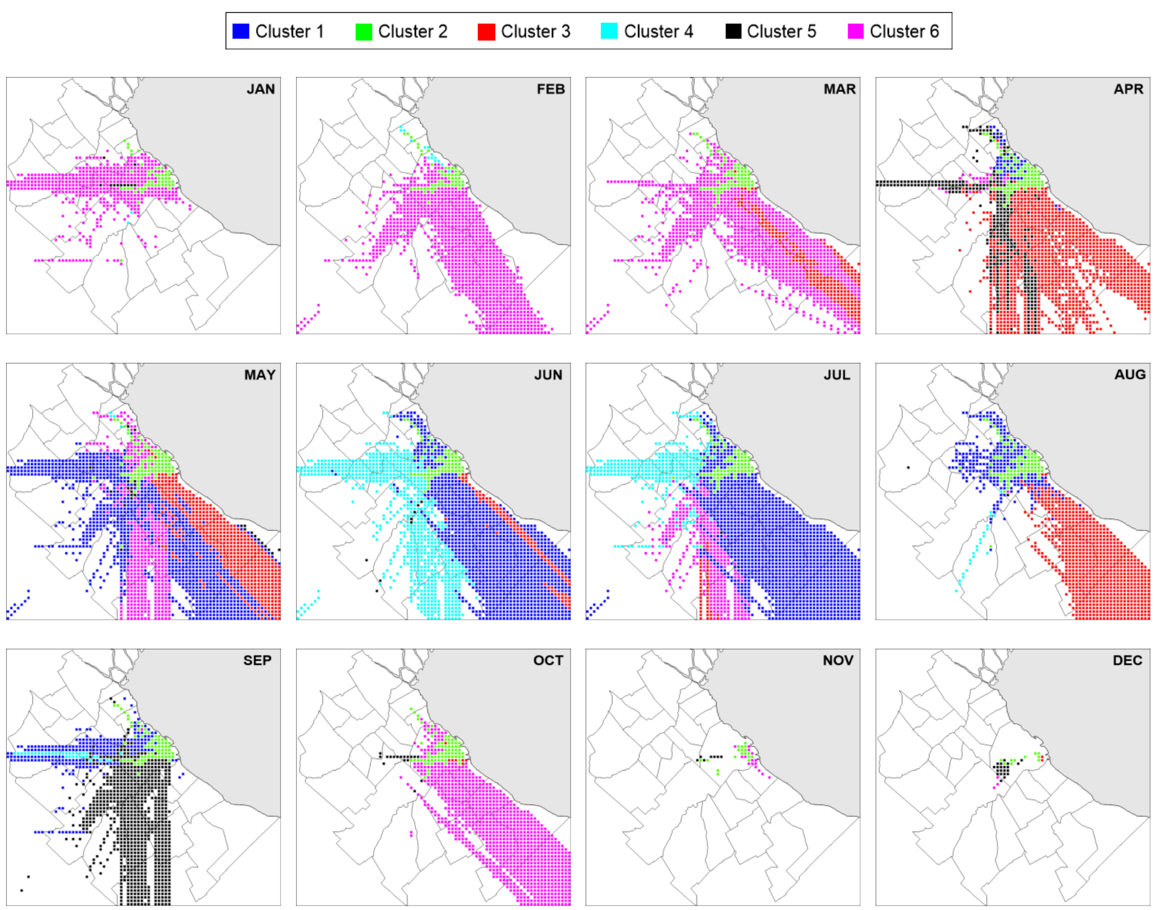


Figure 6. Monthly variation of the dominant cluster spatial distribution.

4 SUMMARY

In this work, an urban scale atmospheric dispersion model (DAUMOD-GRS) is applied in the Metropolitan Area of Buenos Aires (MABA) to estimate exceedances of urban background concentrations of nitrogen dioxide (NO_2) with respect to the local air quality standard (200 ppb) and the guideline suggested by the WHO (110 ppb). High resolution (1 h, 1 km^2) long-term (3-years) simulations are performed in a domain including the MABA area (3830 km^2) and its surroundings. Only a few exceedances of the local AQS are obtained, while a considerably number of events is obtained when the WHO level is considered. In order to characterise the conditions leading to these exceedances, clustering analysis is applied, greatly reducing the dimension and complexity of the data. Results show that model outcomes can be grouped into 6 clusters representing different sets of conditions that, when combined, lead to exceedances in large but compartmentalized regions of the MABA. In general, they occur mostly during early morning hours (i.e., maximum emission rates from traffic sources) and in winter months (due to reduced dispersion conditions of the atmosphere), as expected. However, other characteristics are less trivial to explain. For example, exceedances occurring at receptors with no emission and far from sources during calm conditions suggest that chemistry and/or the memory effect of the model could be playing a more important role than previously thought.

Overall, it is worth noting that even with a simple model such as the DAUMOD-GRS, different exceedance conditions are obtained at different parts of the MABA. This highlights the importance of having a good estimation of the NO_x and VOCs emissions and their spatial distributions throughout the area of study. With three years of simulations we have identified distinct conditions that consistently lead to exceedances in specific regions of MABA and seasons of the year. We plan to expand our work by increasing the number of simulated years and focus on conditions that lead to exceedances recurrently every year. This may help us to identify regions of MABA where efforts in monitoring NO_2 concentrations could be particularly valuable.

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