Location of areas of emission of pollutants when poor urban air quality is detected

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Abstract: Air pollution constitutes an environmental risk, evidenced in large urban centres. This work applies a methodology capable of detecting the areas of emission of pollutants when episodes of poor urban air quality are observed. This is carried out coupling air quality indices proposed by United States

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Environmental Protection Agency with the receptor model known as Nonparametric Trajectory Analysis. As a control case, observed concentrations (2013–2015) in Bahia Blanca (Argentina) were analysed, highlighting particulate matter as a dominant pollutant for episodes of poor air quality. Likewise, the application of the methodology allowed to reduce the number of possible stationary emission sources by 75%, and to highlight the implication of nearby linear sources. The strength of the methodology lies in visualising in real time, or in diagnostic mode, the potential areas of emission and their significance.

Keywords: AQI; air quality index; criteria pollutants; receptor model; episodes of high concentrations; back-trajectories; SO_2 ; PM_{10} ; risk; decision-makers.

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1 Introduction

At present, the World Health Organization (WHO) concludes that air pollution is considered an environmental risk and states that one in eight deaths are a consequence of it. In addition, it is estimated that half of the urban populations are exposed to values higher than 2.5 times the guide levels established by WHO. This situation is magnified in low-to-middle income countries (WHO, 2014). Particularly, both Argentina and other Latin American countries have a decent regulatory framework and lack a robust monitoring network (PAHO, 2017).

Air pollution is explained by means of air quality indices, which are used by government agencies to communicate to the population about the state of air quality, at a certain site and time period. The United States Environmental Protection Agency (US EPA) defines an air quality index (AQI) based on the following pollutants: tropospheric ozone (O₃), particulate matter (PM_{10}), carbon monoxide (CO), sulphur dioxide (SO₂) and dioxide nitrogen (NO₂). This index is categorised into six levels (good, moderate, unhealthy for sensitive groups, unhealthy, very unhealthy, and hazardous) (US EPA, 2014).

Complementary to this, the receptor models are chosen by stakeholders in environmental air quality management as essential tools for the generation of mitigation policies and the development of public health plans. These models allow us to interpret which of the present sources are the most significant in the area and to design mitigation strategies. The choice of the model to implement will depend on the available information, such as source types and profiles and emissions inventory, among others (Behrentz et al., 2009; Belis et al., 2014; Hopke, 2016; WHO, 2000). For a correct performance, it is required that both the receptor site and the characterised species are representative of the region. Meteorological information and updated source profiles are considered relevant in this instance (Belis et al., 2014; Donnelly et al., 2011; Hopke, 2016).

The study of air pollution addressed in local scale with hybrid receptor models allows to georeferenciate the emission sources and its impact in greater urban centres. Through different statistical models that use the fundamental role of the wind (e.g., conditional probability function, conditional bivariate probability function, nonparametric wind regression, nonparametric trajectory analysis), authors were able to indicate that urban traffic, resuspended dust and other point-inventoried sources were the major emission sources. In general, these studies were carried out by recollecting air pollutants data in monitoring campaign (Argyropoulos et al., 2017; Cahill et al., 2016; Feinberg et al., 2019; Grange et al., 2016; Henry et al., 2019; Roig Rodelas et al., 2019; Squizzato et al., 2017; Wei et al., 2019). Furthermore, due to the proximity of important sources that exceed the territories with jurisprudence, the interest in identifying different sources of regional emissions, and with it, the predominant directions through the use of more complex hybrid models, is a common practice, normally, through HYSPLIT (Argyropoulos et al., 2017; Grange et al., 2016; Landis et al., 2019; Roig Rodelas et al., 2019; Squizzato et al., 2017; Urbančok et al., 2017; Yang et al., 2020).

The receptor model used in this work is the nonparametric trajectory analysis (NTA) (Henry, 2008), which seeks to identify the areas with the highest probability of contributing contaminants to the receptor site. This model consists of approximating the back-trajectories to the local scale through the kinematic model of uniform rectilinear motion, using local meteorological information. The advantage of the method consists in the association of each point of the trajectory with a spatial coordinate. The use of spatial coordinates is a modification of the original model by the authors of this work.

The back-trajectories are estimated for each concentration observed at the receptor site during the episode, according to the following equation:

$$z = (x, y) \begin{cases} x_{k}(t_{j}) = \sum_{i=0}^{k} v_{x}(t_{j-i}) \Delta t \\ y_{k}(t_{j}) = \sum_{i=0}^{k} v_{y}(t_{j-i}) \Delta t, \quad k = 1, ..., N. \end{cases}$$

where the coordinates z(x, y) correspond to the point on back-trajectory ending at time j, Δt is the time step (temporal resolution), N is the number of steps taken backwards in time, v_x and v_y are the decomposition of wind in Cartesian coordinates estimated from geometric expressions for the *k*th time. Each point of trajectory is associated to the concentration (C_k) observed at the receptor site. Then each point (x_i , y_i) generated in each back-trajectory, hereinafter referred to as coordinate z_i , is assembled to concentrations associated summarising the information in a new set (z_i , C_i). The resulting trajectory (z_i) expressed in the Cartesian system with longitudinal units (km), was approximated in its corresponding geographic coordinates. Then, for any point (z) defined in this grid, the concentration to the measurement matrix $\hat{C}(z)$ is estimated using a nonparametric regression of previously generated back-trajectories, using the Epanechnikov kernel (Härdle, 1990). The expected value of \hat{C} over grid point z is a result of a kernel regression by the following equation:

$$\hat{C}(z) = \frac{\sum_{k} K(z - z_{k} / h) c_{k}}{\sum_{k} K(z - z_{k} / h)}$$

where h is the smoothing parameter obtained by cross validation. It should be noted that this estimation is influenced by the proximity to the receptor site and the parameters used in the regression. Finally, the location of the major sources contributing to concentrations of the pollutant at the monitoring site is spatially visualised.

Many authors have used receptor models to study the areas of origin of criteria pollutants, with historical databases (Cahill et al., 2016; Donnelly et al., 2011; Feinberg et al., 2019; Grange et al., 2016; Han et al., 2017; Henry et al., 2019; Pérez et al., 2012; 2013; Wei et al., 2019). However, the coupling of these tools with an index that defines

air quality reinforces the relationship between pollution levels and the impact on population health. Currently, there is a vacancy in the offer of similar results by government agencies, which use indexes to communicate the current status of frequent exposure to local and regional emissions (Navebare et al., 2018; Sarigiannis et al., 2017; Xue et al., 2019). Such is its importance, that some authors work on advanced algorithms to be able to predict the state of the index (Wu and Lin, 2019) or evaluating chronic effects by combining the polluting criteria (Ruggieri and Plaia 2011, 2012). Another approach currently addressed is the geolocation of the criteria pollutant emissions associated with air quality in annual periods, to account for regional emissions (Yang et al., 2020). However, the evaluation of exposure events of short periods of time for analysis on an urban scale has not been strongly addressed. The study of air pollution addressed in local scale with hybrid receptor models allows to georeferenciate the emission. Based on the above, this work aims to apply a methodology capable of detecting in real time (or in diagnostic mode), on a local scale, the areas of origin of emission of pollutants against episodes of poor quality of air. This is done by coupling the air quality index (AQI – US EPA), that defines the episodes of interest in population health, and the hybrid receptor model (NTA).

2 Methods

The methodological scheme proposed here consists in the detection of episodes of high concentrations associated with alert levels for population health, and in the identification of the respective areas of contribution to the monitoring site.

2.1 Detection of high concentration episodes

The analysis of the databases seeks to define episodes, such as those concentrations that exceed levels of interest defined by an AQI. For example, if AQI - US EPA is used, episodes of poor air quality could be associated with those with a higher index than the 'unhealthy' category. In this step, the air quality is estimated on an hourly basis from the adaptation of the measurements observed in the monitoring site (receptor site) to the reference times of the chosen AQI. The 'dominant' pollutant, which characterises the air quality, is the one that presents the maximum value of the index individually estimated.

2.2 Application of the NTA receptor model

To estimate the contribution zones, the model makes use of the concentrations observed at the receptor site (*C*) that define the episode, together with the associated wind speed and direction data. The number of trajectories to be estimated per episode is a function of the average time of the dominant pollutant, those that take place during the presence of calm winds (less than 1 km h^{-1}) being rejected.

For the pollutant under study, the average distance (r), estimated from the maximum and minimum distances, of the most relevant emission sources with respect to the monitoring site is established. From this, the (t_r) back-trajectory time is calculated as the ratio between the radius (r) and the mode of the velocities included in the episode.

The NTA offers as an output a matrix of georeference average concentrations, which can be visualised with a geographic information system (GIS) on a chromatic scale on a map. Those polygons associated with the maximum concentrations represent the contribution areas of the contaminant to the receptor site.

3 Application to the study of criteria pollutants in the Bahia Blanca district

The district of Bahía Blanca, province of Buenos Aires (Argentina) consists of the homonymous city and seven other locations, among which Ingeniero White stands out due to its industrial-economic activity in the region. Here is located one of the largest petrochemical complexes of the country. Taking into account the population factor and the presence of many important industries, the region under study is a potentially exposed area to the risk of air pollution.

A criteria pollutants monitoring station, under the supervision of a Municipal Technical Committee, is located in the vicinity of the petrochemical area $(38^{\circ}45'32''S, 62^{\circ}17'08''W)$. This station reports average hourly values of tropospheric ozone (O_3) , particulate matter (PM₁₀), carbon monoxide (CO), sulphur dioxide (SO₂) and dioxide nitrogen (NO₂) on the air, since 1997, and they are published on a web platform (http://quepasabahiablanca.gov.ar/tiempo_real/calidad_de_aire/). In this work, the database corresponding to the 2013–2015 period, of pollutant observations and meteorological parameters, is analysed. The latter are provided by the national meteorological service station located at $(38^{\circ}43'13''S, 62^{\circ}09'27''W)$.

The technical survey of stationary emission sources was taken from the report of the Programa Integral de Monitoreo (PIM – Comprehensive Programme of Monitoring), updated for 2015, elaborated by the Executive Technical Committee of the Bahía Blanca municipality (BB ETC, 2016). It presents an update of the Inventory of Gaseous Emissions from point emission sources, provided by the municipality (Figure 1). It follows that the main contributors in sulphur oxides (SO_x) emissions are the thermoelectric power station 'Central Piedra Buena S.A.' (82.8% annual) and oil refinery 'Petrobras Argentina S.A.' (17.2% annual). The SO₂ emissions in the plant are directly related to the amount of sulphur in the fuel used. For PM₁₀ it is found that Petrobras Argentina S.A. emits 46% of the annual tons received, followed by Central Piedra Buena S.A. with 15%, out of a total of nine declared companies. According to the inventory of fixed emissions, of the eight companies that emit NOx, similar annual emissions are observed by Central Piedra Buena S.A. and TGS (29%), followed in third place by PBB Polisur (14%). With respect to CO, seven companies are declared, of which TGS (43.4%), Solvay Indupa (24.4%) and Cargill (22.7%) are highlighted.

Since the analysis is carried out in the territory of the Bahía Blanca district, province of Buenos Aires, Argentina, it is arranged to use the index proposed by the US EPA. This decision was made because there is no index that considers the exposure of population health to air quality for short periods of time. As a result, the district uses the US EPA index to report the daily status of air quality. The AQI -US EPA has values considered representative and reliable for the region. However, it is highlighted that the methodology is suitable to apply other criteria of air pollution.

Taking as an alert level the category of 'unhealthy' (AQI – US EPA), eight episodes of high concentrations were detected. The episodes have in common PM_{10} as the dominant pollutant; two of them were selected to deepen their study. The selection criteria applied were defined based on those with the highest AQI (from unhealthy to

hazardous) and greatest availability of contaminant data. On the other hand, a third case is analysed in which the air quality is 'moderate' (AQI – US EPA), dominated by SO_2 , in order to show the versatility of the model. The third case is selected because the contaminant has only two main sources declared in the source survey (PIM) and because it is a contaminant related to anthropogenic sources.

Figure 1 Location of the monitoring site (red star). The polygons represent the companies declared in the in the technical survey of stationary emission source for the year 2015 (see online version for colours)



Source: Executive Technical Committee of the Bahía Blanca Municipality

3.1 First episode: 18 November, 2014

This episode is governed by the PM_{10} ; the air quality remains 'unhealthy' from 2 am (red level) until 9 am on 18 November, 2014. From the time series that describes the episode (Figure 2), in which the concentrations observed at the monitoring site are analysed, it follows that the contaminant levels begin to increase from 10 am on 17 November until 2 am the next day. Previous to the episode, the air quality remained 'unhealthy for sensitive groups' (orange level) from 3 pm on 11/17/2014 to 2 am on 11/18/2014, during which the moving concentrations were between 155–254 µg m⁻³. From the time series it follows that the episode has a duration of 8 h (moving concentrations between 255–354 µg m⁻³).

3.2 Second episode: 24–26 April, 2015

On the other hand, the winds associated with the period in which the concentrations contribute to poor ("unhealthy") air quality, mainly comprise the south, southeast and east directions. The inventoried emission sources of the PM_{10} are located at an average distance of 2.5 km. Then, for a frequent speed of 46 km h⁻¹, the path time used in the NTA is 3.26 min. As a result, the polygons with the highest concentrations (365–572 µg m⁻³) are observed to the north-northwest of the receptor site, where oil refinery, National Route 3 and part of the perimeter roads of the city are located. It should be noted that the index is defined by a 24-hour moving average for this pollutant (Figure 3). This episode responds to the eruption of Calbuco volcano, located in southern

Chile, and to the west-southwest of the monitoring site (Ingeniero White), evidencing an air quality considered "unhealthy" (average concentrations higher than 354 μ g m⁻³), on the monitoring site, from 24 April, 2015 at 4:00 pm to 2:00 am on the 26th of the same month. In the present episode dominated by PM₁₀, the quality index exceeded the maximum proposed by the US EPA, whose category is "hazardous", from 10 pm on day 24 until 7 pm on day 25. In the time series (Figure 4) one can visualize how concentrations observed (measurements at the monitoring site) increase from 1 pm on 24 April, 2015 level of "unhealthy for sensitive groups" (155–254 μ g m⁻³), which quickly transits to the categories 'unhealthy' (255–354 μ g m⁻³), 'very unhealthy' (355–424 μ g m⁻³) and 'hazardous' (>425 μ g m⁻³) within 40 h.

Figure 2 The concentrations observed of PM_{10} (µg m⁻³) in the receptor site for the episode from 18 November, 2014 are observed in the upper box. Below this box, a time grid with the wind directions associated to each concentration observed and level of AQI (with colour scale) is exposed (see online version for colours)



Figure 3 Average concentrations of $PM_{10} (\mu g m^{-3})$ obtained by NTA during the event 18 November, 2014. The receptor site is represented with a light blue star and the polygons in blue chromatic scale represent the companies declared in the in the PIM for PM_{10} (see online version for colours)



Figure 4 The concentrations observed of PM_{10} (µg m⁻³) in the receptor site for the episode from 24–26 April, 2015 are observed in the upper box. Below this box, a time grid with the wind directions associated to each concentration observed and level of AQI (with colour scale) is exposed (see online version for colours)



As in case 1, the back-trajectory time of 8.8 min is associated with the most frequent winds of 17 km h⁻¹ for the episode. When applying the NTA for the period corresponding to the first 24 h that include the first value corresponding to the category of "unhealthy", it is observed that the concentrations of the PM₁₀ come from a wide range of directions, from west to south clockwise (Figure 5). In this regard, it is needed to record natural phenomena, outside the local scale, that can reach and impact the region under study.

Figure 5 Average concentrations of PM_{10} (µg m⁻³) obtained by NTA during the event 24–26 April, 2015. The receptor site is represented with a light blue star and the polygons in blue chromatic scale represent the companies declared in the in the PIM for PM_{10} (see online version for colours)



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3.3 Third episode: 2 August, 2015

In this case, an episode is studied where the air quality goes from being 'good' to "moderate". This case is used to observe the goodness of the tool, taking SO_2 as the representative pollutant. Developed at 10 pm on 2 August, 2015 (Figure 6), the moving average is equal to the concentration observed at the monitoring site, because the parameter integration time is equal to the measurement frequency.

Figure 6 The concentrations observed of SO_2 (ppb) in the receptor site for the episode from 2 August, 2015 are observed in the upper box. Below this box, a time grid with the wind directions associated to each concentration observed and level of AQI (with colour scale) is exposed (see online version for colours)



The application of the NTA (Figure 7), for a time of 11.5 min of back-trajectory, allows observing a polygon with higher concentrations located at north-northwest of the monitoring site, where the oil refinery is located, associated with 17% of emissions of the contaminant.

For the first two episodes defined by 24 h, Root Mean Square Error (RMSE) was calculated. For the first (18 November, 2014), the variability of the PM_{10} presents an average value of 23.85 µg m⁻³ in the range of 0 to 135.2 µg m⁻³ (median: 17.4 µg m⁻³, Q₁: 12.1 µg m⁻³, Q₃: 28.7 µg m⁻³), with 82% of the data with values less than 33.80 µg m⁻³. Similarly, in the second event developed on 24 April, 2015 by PM_{10} , the variability of the data presents an average of 13.27 µg m⁻³ in the range of 0 to 217.55 µg m⁻³ (median: 3.04 µg m⁻³, Q₁: 0.01 µg m⁻³, Q₃: 7.94 µg m⁻³), with 91% of values less than 54.39 µg m⁻³. On the other hand, in the episodes in which the air quality is defined by a pollutant with hourly impact, the variation of the observed data corresponding to the direction of the intervening wind (during the episode) was calculated for the period under study; without considering calm winds since they are not modelled. Then, the last episode that analyses SO₂ for 2 August, 2015 presents a variability of 2.94 ppb.

Figure 7 Average concentrations of SO₂ (ppb) obtained by NTA during the event of 2 August, 2015. The receptor site is represented with a light blue star and the polygons in blue chromatic scale represent the companies declared in the in the PIM for SO₂ (see online version for colours)



4 Discussion

The cases under study highlight the virtues and limitations of coupling the categorisation obtained through AQI with receptor model as an air quality analysis tool. By means of the AQI – US EPA it was observed that PM_{10} was the dominant pollutant of the episodes of poor urban air quality observed for the period under study. This situation is recurrent in an international context in developed countries (Argyropoulos et al., 2017; Grange et al., 2016; Guo and Lu, 2019; Karagulian et al., 2015). In addition, the application of the methodology allowed to limit the number of emission sources of the dominant pollutant by 75%, and to highlight the implication of nearby linear sources, especially in the resuspended dust of high traffic roads (National Route 3). These emissions are the most frequent in studies carried out in other large urban centres (Argyropoulos et al., 2017; Cahill et al., 2016; Feinberg et al., 2019; Grange et al., 2016; Squizzato et al., 2017). In the first case, the most probable area of contribution to poor air quality could be associated with a company that emits about 46% of the particulate matter (PM_{10}) in the region and a linear source (National Route 3). This last source concentrates an annual average daily traffic of approximately 13,852 vehicles between kilometres 685.7 and 689.3 (Seguridad Vial Nacional, 2018). On the other hand, the tool demonstrates its limitation by trying to describe an episode considered regional (ashes from the Capulco volcano), even if the directions of origin estimated with the model are consistent with those expected. The distributions of sources associated with regional emissions are usually estimated through hybrid models that analyse back-trajectories of more than 24 h, such as the HYSPLIT model (Argyropoulos et al., 2017; Nayebare et al., 2018; Roig Rodelas et al., 2019; Squizzato et al., 2017; Yang et al., 2020). Finally, the third case evidenced the good performance of the NTA tool in the definition of the probable contribution zone, where one of the two industries that contribute to sulphur dioxide (SO_2) emissions in the region is located.

The scheme limitations for diagnosing poor air quality events (proposed methodology) are mainly linked to the selected receptor model. The representativeness of the scenario has as a critical point the interpolation between the pollutant data and the meteorological parameters, which must have at least the same temporal resolution, the latter being representative of the monitoring site. Regarding the model, the estimated back-trajectories can be affected by the topography of the place, as well as by the presence of obstacles between the monitoring site and the area under study. When applying the methodology, a great knowledge of the emission sources in the region is required to see how close the model's output is to reality (Henry, 2008, 2019; Feinberg et al., 2019; Wei et al., 2019).

5 Conclusion

The benefits presented by the articulated use of an AQI and a receptor model are highlighted in this work. The results are descriptive, interoperable and simple to interpret, which facilitates their implementation as a management tool for analysing the results and/or communicating them to the population.

The coupling of the AQI-US EPA with the NTA allows the evaluation of the origin of criteria pollutants, in real time or in diagnostic mode, for episodes of high concentrations that cause poor air quality in a monitoring site. Besides, the coupling of these tools shows the ability to relate emission and local weather conditions, and to point out those potential sources of contribution, that are not necessarily those with the highest emissions emitted in the region. It is remarked that, during the period studied, episodes of poor urban air quality are associated with particulate matter.

Finally, the proposed methodology is intuitive, easy to interpret, and offers quick visual results to interpret the contamination levels of any site under study, at the local level.

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