

1 MULTIPLEX VISIBILITY GRAPHS AS A COMPLEMENTARY TOOL  
2 FOR DESCRIBING THE RELATION BETWEEN GROUND LEVEL  
3 O<sub>3</sub> AND NO<sub>2</sub>

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12 DECLARATION OF INTERESTS

13 Declarations of interest: none

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## 14 ABSTRACT

15 The usage of multilayer complex networks for the analysis of correlations  
16 among environmental variables (such as  $O_3$  and  $NO_2$  concentrations from the  
17 photochemical smog) is investigated in this work. The mentioned technique is  
18 called *Multiplex Visibility Graphs (MVG)*. By performing the joint analysis of those  
19 layers, the parameters named Average Edge Overlap and Interlayer Mutual  
20 Information are extracted, which accounts for the microscopical time coherence  
21 and the correlation between the time series behavior, respectively.

22 These parameters point to the possibility of using them independently to  
23 describe the correlation between atmospheric pollutants (which could be  
24 extended to environmental time series). More precisely the first one of them is  
25 considered to be a potential new approach to determine the time required for the  
26 correlation of  $NO_2$  and  $O_3$  to be observed, since it is obtained from the correlation  
27 of the pollutants at the smallest time scale. As for the second one, it has been  
28 checked that the proposed technique can be used to describe the variation of the  
29 correlation between the two gases along the seasons. In short, MVGs parameters  
30 are introduced and results show that they could be potentially used in a future for  
31 correlation studies, supplementing already existing techniques.

## 32 KEYWORDS

- 33 - Photochemical smog
- 34 - Complex networks
- 35 - Multiplex Visibility Graphs
- 36 - Nonlinear time series
- 37 - Multivariate analysis

## 38 1. INTRODUCTION

39 In the last years, many studies have been conducted to give some light on  
40 formation and dynamics of ground-level ozone. The important of these analyses  
41 lie on the fact that it is one of the main photochemical oxidants (due to its  
42 abundance) and it can lead to serious damage for human health and harvest for  
43 high concentrations (Doherty et al., 2009). According to Miao (Miao et al., 2017),  
44 its impact can be quantified in losses of billions of dollars from the economical  
45 point of view.

46 The formation and destruction of this secondary pollutant is known to be  
47 governed by photochemical and nonlinear processes (Graedel and Crutzen,  
48 1993; Trainer et al., 2000) that depend highly on meteorological conditions such  
49 as temperature, wind direction and mainly solar radiation (Trainer et al., 2000). In  
50 addition to all that, O<sub>3</sub> concentration also depends on the behavior of its chemical  
51 precursors. These are mainly nitrogen oxides (amongst them, NO<sub>2</sub> is studied  
52 here) and volatile organic compounds produced from the urban and industrial  
53 activity (Sillman, 1999). It is because of all those factors that the analysis of the  
54 temporal evolution of ozone is a very complex task. Hence, some questions  
55 remain open such as the influence of the working time scale on these studies and  
56 the relevance of NO<sub>2</sub> as precursor depending on the season of the year.

57 A very recent method (called *Multiplex Visibility Graph*, MVG) to analyze  
58 nonlinear multivariate timeseries (Lacasa et al., 2015) is used in this work to see  
59 the relation between O<sub>3</sub> and its precursor NO<sub>2</sub> to answer the questions posed  
60 above. This method consists basically on turning the time series into complex  
61 networks and then forming multilayered structures that can be analyzed  
62 afterward, thanks to the last advances in this field. MVGs have shown to be useful

63 for several applications already, from economics to neurology (Bianchi et al.,  
64 2017; Lacasa et al., 2015; Sannino et al., 2017). Those works have used last  
65 developments in multilayer networks (Bianconi, 2013; Boccaletti et al., 2014; De  
66 Domenico et al., 2013; Kivela et al., 2014) to obtain information to describe and  
67 compare the signals.

68 The reason to use this approach instead of other ways to construct functional  
69 networks is because those usually require performing a pre-processing or  
70 symbolization, associated with loss of information (Kantz and Schreiber, 2004).  
71 Also, one of the advantages of using complex networks for analyzing time series  
72 is that they are becoming powerful tools when one seeks to construct feature  
73 vectors that can be used to automatically feed classifiers with low computational  
74 cost (Lacasa et al., 2015).

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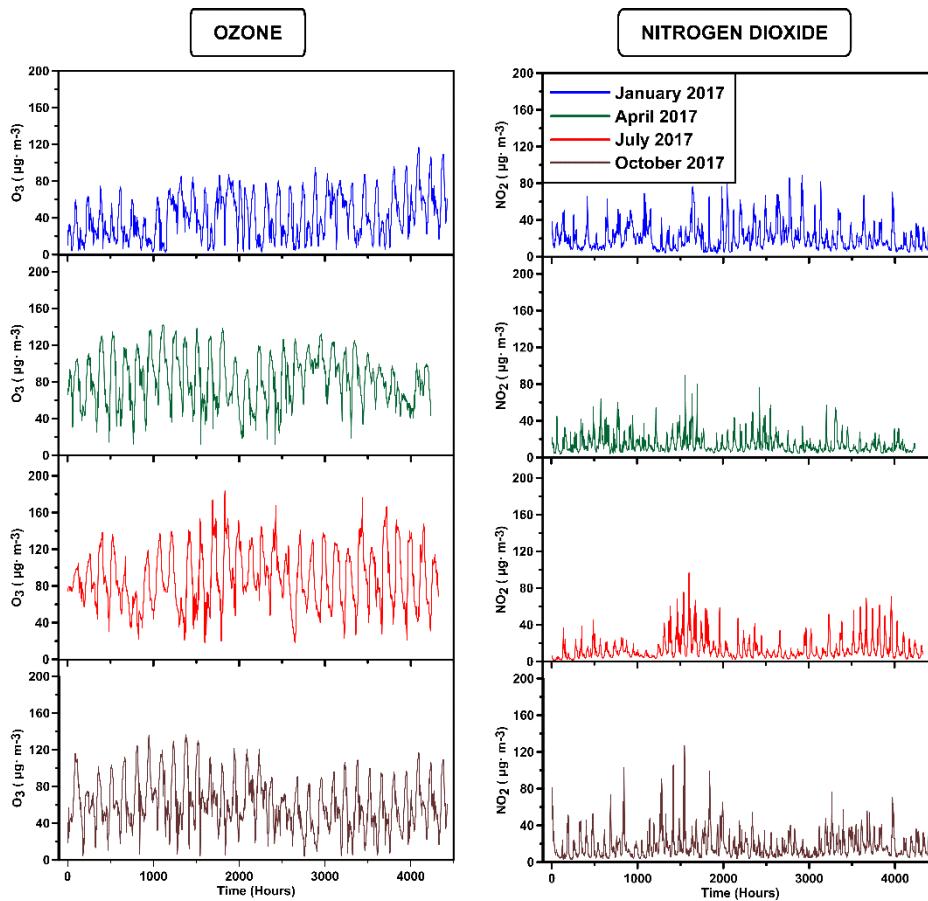
## 76 2. MATERIALS AND METHODS

### 77 2.1. Experimental data

78 The data of pollutant used for this analysis correspond to ozone and nitrogen  
79 oxide concentration values collected from 2010 to 2017, with a frequency of 10  
80 minutes between each measure. Then, these data were separated into the  
81 different months in order to perform the analysis. The measurements were  
82 performed at the urban station located in Lepanto, Córdoba (37.53° N, 4.47° W).  
83 The cited station belongs to the regional network in charge of controlling the air  
84 quality in Andalusia, co-financed by the Consejería de Medioambiente (Regional  
85 Environmental Department) and the European Union. This station is located at  
86 117 m of altitude and the average temperature and solar radiation is maximum

87 on July and minimum on January every year. The region where it is placed is the  
88 western part of Andalusia (Spain). Since as exposed previously (Domínguez-  
89 López et al., 2014), this area meets the weather conditions (high temperatures  
90 and solar radiation), orographic (the valley of the Guadalquivir river) and  
91 anthropic ones to be potentially vulnerable to pollution by ground-level ozone and  
92 nitrogen oxide. The climate of the zone of study, according to the Köppen-Geiger  
93 classification, is defined as Csa, with warm average temperatures and hot and  
94 dry summer. Furthermore, the city of Córdoba is surrounded by two main  
95 industrial parks. One of them is located at southwest, and the other is at east.  
96 Moreover, there is a highway at southeast with frequently high traffic volume from  
97 both directions.

98 Authors have also employed the temperature, wind direction and average  
99 solar radiation in this work. They are shown further in the text (see Figure 7), were  
100 they are plotted along with the results in question, in order to illustrate their  
101 apparent relationship. These meteorological quantities have been provided by  
102 the Andalusian Agency of Energy.



103

104 Figure 1: Sample time series of  $O_3$  and  $NO_2$  for four different months (year 2017).

105

106

107 2.2. Visibility graph

108 A graph can be defined as a set of vertices, points or nodes connected to

109 each other by lines that are usually called *edges*. A tool to transform time series

110 into a graph was presented in the last decade (Lacasa et al., 2008). This new

111 complex network receives the name of *Visibility Graph* (VG) and has been proven

112 to inherit many of the properties of the original signal (Lacasa and Toral, 2010).

113 This means that, for instance, a periodic time series would lead to a regular graph

114 and a fractal series to a scale-free one.

115 In order to construct the visibility matrix which contains the information of all  
 116 the nodes in the system, it is necessary to establish a criterion to discern whether  
 117 two points would be connected or not. This criterion reads as follows: two arbitrary  
 118 data from the time series  $(t_a, y_a)$  and  $(t_b, y_b)$  have visibility (and would become  
 119 two connected nodes in the graph) if any other data point  $(t_c, y_c)$  between them  
 120  $(t_a < t_c < t_b)$  fulfills the following condition:

$$y_c < y_a + (y_b - y_a) \frac{t_c - t_a}{t_b - t_a} \quad (1)$$

121 The result of applying this visibility method is a  $N \times N$  adjacency binary matrix,  
 122 being  $N$  the number of points in the set. Each row of the matrix contains the  
 123 information of a different node. For example, an element as  $a_{ij} = 1$  means that  
 124 the node  $i$  and  $j$  have visibility; whereas  $a_{ij} = 0$  means that there is no edge  
 125 between them. The resulting matrix has several properties that can be used to  
 126 simplify the algorithm and thus reduce the computational required time: it is a  
 127 hollow matrix ( $a_{ii} = 0$ ), symmetric ( $a_{ij} = a_{ji}$ ) and all the nearest neighbors have  
 128 visibility between each other ( $a_{ij} = 1$  for  $j = i \pm 1$ ). Its general form is shown  
 129 below:

$$A = \begin{pmatrix} 0 & 1 & \dots & a_{1,N} \\ 1 & 0 & 1 & \vdots \\ \vdots & 1 & \ddots & 1 \\ a_{N,1} & \dots & 1 & 0 \end{pmatrix} \quad (2)$$

130 In Figure 2, the application of the VG to two arbitrary time series is shown,  
 131 highlighting the connections of two given time points (nodes in the graph) for the  
 132 sake of clarity.

### 133 2.3. Degree centrality

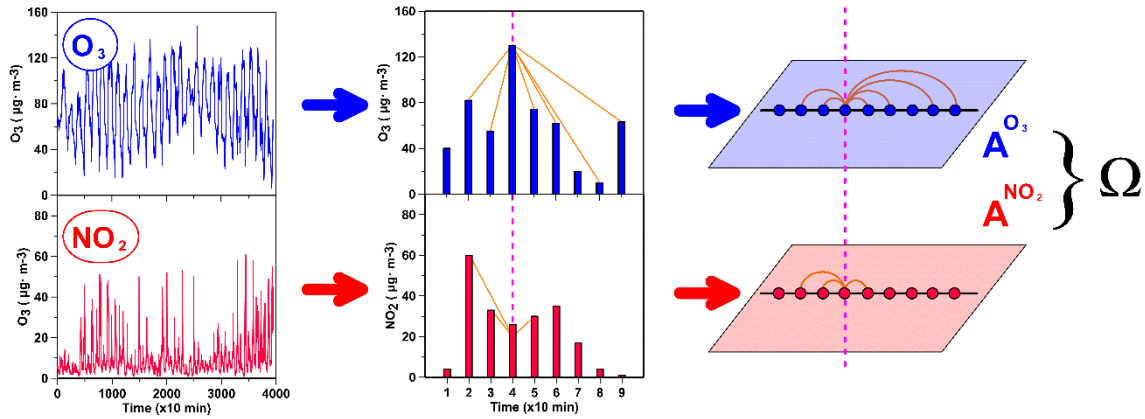
134 To study the main properties of a complex network, centrality parameters  
135 become convenient mathematical tools to take into account. This kind of  
136 parameters measure the node importance within the graph in relation to the  
137 others by different approaches (Latora et al., 2017).

138 A very frequently used centrality parameter and an important feature of graphs  
139 in general is the degree. The degree of a node ( $k_i$ ) can be defined as the number  
140 of nodes that have reciprocal visibility (in an undirected graph) with the first one  
141 ( $k_i = \sum_j a_{ij}$ ). In Figure 2, the degree of the node that is highlighted is  $k = 6$  for  $O_3$   
142 and  $k = 3$  for  $NO_2$ .

143 From the degree of each one of the nodes present in the VG, it is possible to  
144 obtain the degree distribution of the sample  $P(k)$ , which is nothing but the  
145 probability that every degree has within the graph. This distribution can be  
146 analyzed to get a deep insight of the intrinsic nature of the time series, as  
147 previously demonstrated by (Lacasa et al., 2008; Mali et al., 2018; Pierini et al.,  
148 2012). The degree distribution of VGs whose right tails can be fitted to a power  
149 law in the way  $P(k) \propto k^{-\gamma}$ , are associated to fractal time series (Lacasa et al.,  
150 2008). The right tails are related to hubs, which are unlikely highly-connected  
151 nodes in the graph and therefore points with large values of degree. In a log-log  
152 plot, one can fit  $P(k)$  to a simple linear regression, obtaining the so-called  $\gamma$   
153 coefficient, which has been directly related to the Hurst exponent of the time  
154 series in the Brownian motion (Lacasa et al., 2009).

## 155 2.4. Multiplex visibility graph





156

157 Figure 2: Time series of ozone and nitrogen dioxide concentrations (left) are  
 158 transformed into complex networks through the VG algorithm (center), which is defined  
 159 by an adjacency matrix ( $A^{O_3}$  and  $A^{NO_2}$ ). Finally, the two of them are combined to create  
 160 a two-layered MVG, called  $\Omega$  (right image).

161 In the case of a multivariate time series of  $M$  variables, it is possible to  
 162 construct a  $M$ -dimensional network from the VG of each one for the description  
 163 of the signals (Lacasa et al., 2015). This multilayer network is called *Multiplex*  
 164 *Visibility Graph* and each one of its  $M$  layers corresponds to the VG of one of the  
 165 variables from the underlying time series (see Figure 2). The MVG is represented  
 166 by a vector of adjacency matrices  $\Omega = \{A^{[1]}, A^{[2]}, \dots, A^{[M]}\}$ , being  $A^{[\alpha]}$  the matrix  
 167 corresponding to the VG of the  $\alpha$ -dimension (or layer in the multiplex) from the  
 168 multivariate time series.

169 When it comes to analyzing the information that lies within these complex  
 170 multilayer networks, there are several measures that can be used (Nicosia and  
 171 Latora, 2015). Here, two quantities have been chosen. The first one is the  
 172 *Average Edge Overlap* ( $\omega$ ), that measures the number of layers on which a given  
 173 edge between two nodes is found, on average. The other one, that captures the  
 174 presence of inter-layer correlations of the degree distributions between two layers  
 175  $\alpha$  and  $\beta$ , is the so-called *Interlayer Mutual Information* ( $I_{\alpha,\beta}$ ). These layers in the

176 presented study correspond to the VGs of O<sub>3</sub> and its precursor NO<sub>2</sub> concentration  
 177 time series, hence the notation  $I_{O_3,NO_2}$  will be used in this work.

178 The formula for the calculation of  $\omega$  is presented in Equation 3, where  
 179  $\delta_{0,\sum_{\alpha} a_{ij}^{[\alpha]}}$  corresponds to a Kronecker Delta and the other quantities are already  
 180 defined.

$$\omega = \frac{\sum_i \sum_{j>i} \sum_{\alpha} a_{ij}^{[\alpha]}}{M \sum_i \sum_{j>i} (1 - \delta_{0,\sum_{\alpha} a_{ij}^{[\alpha]}})} \quad (3)$$

181 The maximum possible value of this quantity is  $\omega = 1$ , and corresponds to the  
 182 case where all the layers are identical. On the other hand, the minimum value is  
 183  $\omega = 1/M$  (being  $M$  the number of layers), meaning that a case with each edge in  
 184 the multiplex existing just in one layer.

185 In Equation 4,  $I_{\alpha,\beta}$  is defined:

$$I_{\alpha,\beta} = \sum_{k^{[\alpha]}} \sum_{k^{[\beta]}} P(k^{[\alpha]}, k^{[\beta]}) \log \frac{P(k^{[\alpha]}, k^{[\beta]})}{P(k^{[\alpha]})P(k^{[\beta]})} \quad (4)$$

186 Where  $P(k^{[\alpha]}, k^{[\beta]})$  is the joint probability of finding a node having a degree of  
 187  $k^{[\alpha]}$  in the layer  $\alpha$  and  $k^{[\beta]}$  in the layer  $\beta$ . This joint probability is computed as  
 188 follows:

$$P(k^{[\alpha]}, k^{[\beta]}) = \frac{N_{k^{[\alpha]},k^{[\beta]}}}{N} \quad (5)$$

189 With  $N_{k^{[\alpha]},k^{[\beta]}}$  being the number of nodes that have the corresponding degree  
 190 of  $k^{[\alpha]}$  and  $k^{[\beta]}$  in layers  $\alpha$  and  $\beta$ , respectively. Since  $N$  is the total amount of  
 191 nodes, it must be fulfilled that it is equal to the sum over all the possible  $N_{k^{[\alpha]},k^{[\beta]}}$   
 192 values.

193

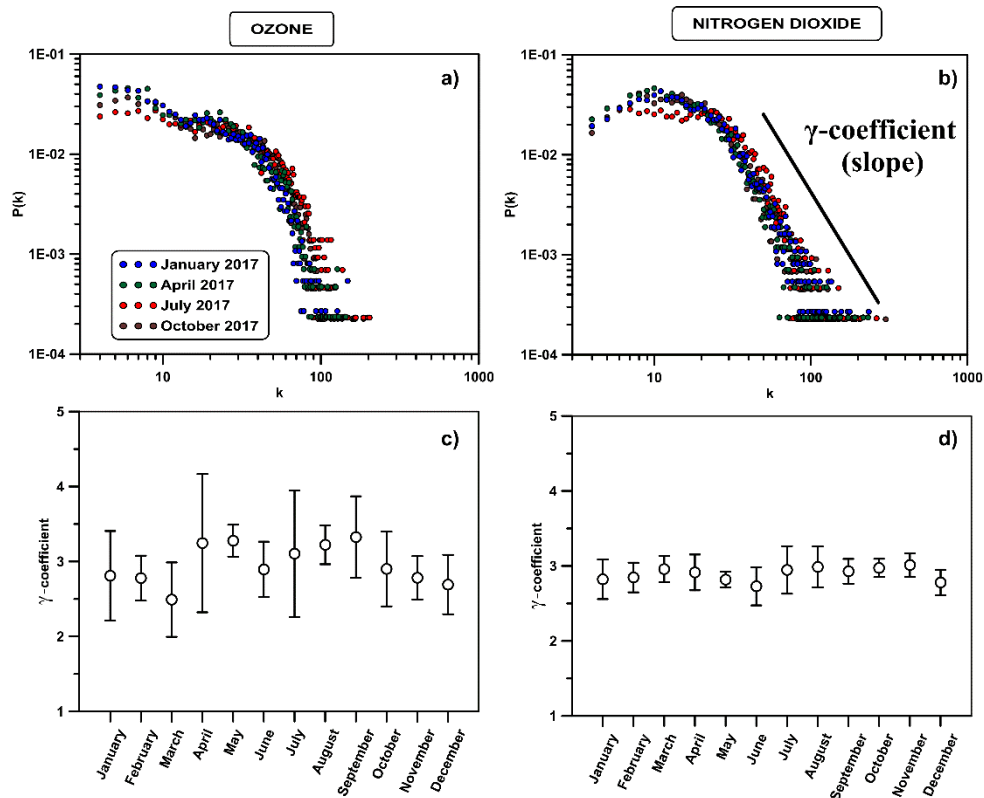
194 3. RESULTS AND DISCUSSION

195 3.1. Exploratory analysis

196 Before applying the MVG methodology, authors have performed a  
197 preliminary analysis of the data employed in this work. To do so, the first  
198 feature to consider has been the distribution of the degree of the independent  
199 VGs obtained from each time series. Some examples of these distributions  
200 can be regarded at Figure 3 (a and b). In these plots, only four months (the  
201 same ones for both) are depicted for illustrative purposes: January, April, July  
202 and October. The reason for choosing these months is that they are equally  
203 spaced through the year, each one represents a different season and they  
204 have been used in previous works in the same location (Carmona-Cabezas  
205 et al., 2019a; Jiménez-Hornero et al., 2010a). The year shown in this case is  
206 2017, the most recent one that has been used here. All data used has not  
207 undergone any deseasonalizing because, as it has been previously  
208 discussed, VGs are not suitable for this kind of preprocessing approaches  
209 (Lange et al., 2018).

210 Looking at the mentioned figures, it can be noticed that both pollutants  
211 present a power-law behavior in the tail of the degree distribution obtained  
212 from their respective VGs, which points to fractal behavior of the time series.  
213 As commented in Section 0, from the linear regression in the log-log plot of  
214 this tail, one is able to obtain the  $\gamma$  coefficient. At plain sight, it can be already  
215 seen Figure 3a and b how the distribution of the degree for ozone varies more  
216 along the year than those of the nitrogen dioxide in the given examples.  
217 Authors have computed the  $\gamma$  coefficient of each month from 2010 to 2017  
218 and shown their average monthly value in Figure 3c and d; where the previous

219 statement is checked. The error bars correspond to the standard deviation of  
220 that quantity along the studied years. Therefore, the ozone concentration has  
221 a different behavior along the seasons, being the mean values of the  
222 coefficients between 2 and 4.25. Moreover, it can be appreciated that those  
223 are also irregular from one year to another, as the standard deviation is higher  
224 in comparison to the second gas by looking at Figure 3c and d. This was  
225 already seen in a previous work by the authors (Carmona-Cabezas et al.,  
226 2019a). On the other hand, NO<sub>2</sub> coefficients do not vary as much as the  
227 previous one, being always its value between 2.5 and 3.25. These results  
228 seem to indicate a different trend in the likeliness of hubs coming from VGs of  
229 O<sub>3</sub> and NO<sub>2</sub> and so distinct unlikely large values variation. Although in some  
230 cases the physical meaning of those coefficients has been related to fractal  
231 parameters such as the Hurst exponent (see Section 0 for more details),  
232 authors have employed these as a preliminary study before going deeper into  
233 the analysis with MVGs.



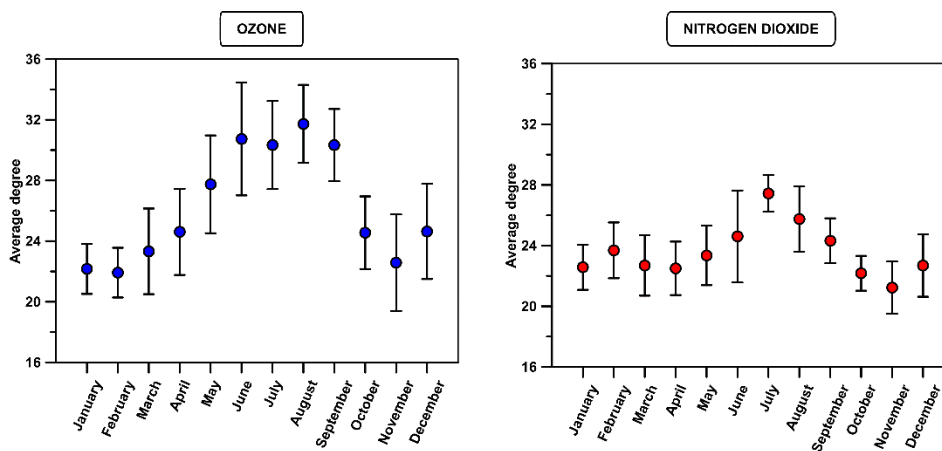
234

235 Figure 3: Degree distributions of four example months in 2017 for O<sub>3</sub> (a) and NO<sub>2</sub> (b)  
 236 and the actual values of the  $\gamma$  coefficient obtained for every studied month and  
 237 averaged from 2010 to 2017 (c and d). The error bars come from the standard  
 238 deviation of the values obtained for all the years.

239 From the degree computed for each node of a given VG, it is possible to obtain  
 240 also its total average value, which has been used in previous papers to describe  
 241 the behavior of the maxima in the series (Carmona-Cabezas et al., 2019b;  
 242 Donner and Donges, 2012). As in previous studies, a clear seasonal pattern is  
 243 observed for both pollutants in Figure 4. The values of this figure correspond to  
 244 the average obtained over the whole period from 2010 to 2017, while the error  
 245 bars come from their standard deviation. This seasonality is less intense in the  
 246 case of NO<sub>2</sub>, which is in accordance to what was discussed in Figure 3.

247 In both cases, the maxima of the average degrees correspond to summer  
 248 months. It was expected in ozone, since those months have the most favorable  
 249 conditions for its creation and therefore there will be a higher number of maximal

250 values (hubs) that increase the average degree. Nevertheless, the behavior of  
 251 nitrogen dioxide is not as acute during summer. This difference might be due to  
 252 the different factors that influence on both pollutants. Results point to the  
 253 possibility that this quantity could be used to identify the known correlations  
 254 between the two pollutants, and hence, authors have tested this hypothesis by  
 255 using the MVG parameters. More precisely,  $I_{\alpha,\beta}$  that is directly based on the  
 256 degree of the two time series.



257  
 258 Figure 4: Monthly average degree values for each pollutant from 2010 to 2017. Again,  
 259 the standard deviation along the different years is reflected through the error bars.

260

### 261 3.2. MVG analysis

262 After applying the VG algorithm to transform the O<sub>3</sub> and NO<sub>2</sub> concentration  
 263 time series into complex networks, the MVGs for each month were built. With  
 264 these multilayer networks, it was possible to compute  $\omega$  and  $I_{O_3,NO_2}$  for all the  
 265 months considered. Figure 5a shows that  $\omega$  values obtained are very similar for  
 266 all the months and during the different studied years. This is clearly seen, as it  
 267 has been averaged over the different studied years (2010-2017) and the standard  
 268 deviation is as well remarkably low.

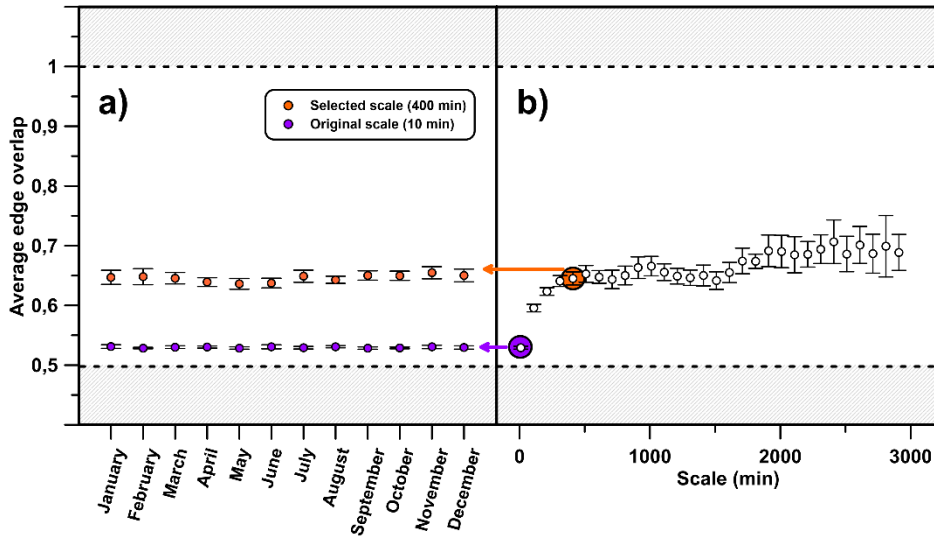


Figure 5: a) Parameter  $\omega$  is computed for each month and then averaged over all the analyzed years (2010-2017), first with the original time scale (purple) and finally with a bigger one (orange) derived from the analysis shown in b). In this one,  $\omega$  is recalculated with several scales for constructing the MVGs. The selected scale is taken from the point where the saturation starts. The white area between the grey zones corresponds to all the possible values that  $\omega$  can have for MVGs with two layers.

270

271 As  $\omega$  accounts for the overall coherence of the original time series, high  
 272 values of it indicates a strong correlation in the microscopic structure and vice  
 273 versa (Lacasa et al., 2015). In the context of VGs applied to time series, the  
 274 microscopic structure can be understood as the most likely scale of the edges of  
 275 the graph. This scale can be considered as the time resolution of the recorded  
 276 signal. For that reason, a study was performed to see how the  $\omega$  parameter  
 277 behaves when the minimal size of the edges of the concentration of  $O_3$  and  $NO_2$   
 278 is changed (see Figure 5b).

279 The result is that the edge overlap increases rapidly as the number of points  
 280 in the sample is decreased (the VGs become smaller and their edges bigger) and  
 281 then it starts to saturate around  $\omega \approx 0.65$  for scales greater than 400 minutes per  
 282 measured point (about 6 hours). On the other hand, for the original scale of the

283 measure  $\omega \approx 0.53$ , which is very close to the minimum value that quantity can  
284 hold ( $\omega = 0.5$ ). From that, this result can be understood as follows: the correlation  
285 between the  $O_3$  and the  $NO_2$  concentrations is very weak at a scale of 10 min  
286 (the order of magnitude of the smallest edges in the graphs). Nevertheless, this  
287 correlation becomes greater as the minimal size of these edges (the microscopic  
288 scale) is increased, until this rising saturates. Authors attribute this effect to the  
289 time needed for the two systems (gases) to reach coupling, so that it affects the  
290 correlation between their concentrations. The posterior saturation would  
291 correspond to the fact that they are already coupled for higher scales, so there is  
292 no increase on  $\omega$  due to physical processes. The slight increase observed would  
293 be due to the reduction of the number of nodes of the VGs (mathematical artifact).  
294 Hence, this approach to determine the time scale at which the saturation starts  
295 could be used to determine the effective time of a given reaction. This could be  
296 useful for later works describing the relationship between some other pollutants.

297 As a previous step before introducing the computed  $I_{O_3,NO_2}$ , authors have  
298 depicted in Figure 6 the quantity  $P(k^{O_3}, k^{NO_2})$ . Again, the same months as before  
299 are used in these plots as an example, for the sake of clarity. Also, only one year  
300 is taken (2017, the most recent), since the results have been found to be  
301 equivalent and the same conclusions could be drawn for different years. The  
302 meaning of these figures can be interpreted as combined degree distributions,  
303 where the colors indicate the probability of the two VGs having degree  $k^{O_3}$  and  
304  $k^{NO_2}$ , simultaneously. It is regarded how the most likely combinations of values  
305 of  $k$  are those of the lowest values of the degree ( $k \in [0, 50]$ ). By contrast, as the  
306 degree increases, the joint probability becomes less and less significative.



307 It must be pointed out that the probability approaches asymptotically to both  
308 X and Y axis. It means that as the degrees increase, the probability of  
309 encountering relatively similar both  $k^{O_3}$  and  $k^{NO_2}$ , decreases exponentially. This  
310 translates into the alternation between the hubs of the two time series. The  
311 reason behind this is the time shift that exists between both  $NO_2$  and  $O_3$  maximal  
312 concentrations throughout the day (previously mentioned). One of the reactions  
313 that governs the ozone creation and destruction is  $NO_2 + O_2 \leftrightarrow O_3 + NO$  (Graedel  
314 and Crutzen, 1993). According to this one, when the ozone reaches a maximum,  
315 the concentration of nitrogen dioxide decreases in general, leading to what has  
316 been argued here.

317 Another characteristic of the plot is that as the year advances, the distribution  
318 of the joint probability changes, being more concentrated around the  $k^{NO_2}$  axis  
319 for January, while in July it is more equally distributed (April and October present  
320 an intermediate behavior). Since the value of the degree and concentration are  
321 related (Carmona-Cabezas et al., 2019b; Pierini et al., 2012), the interpretation  
322 can be seen as follow:

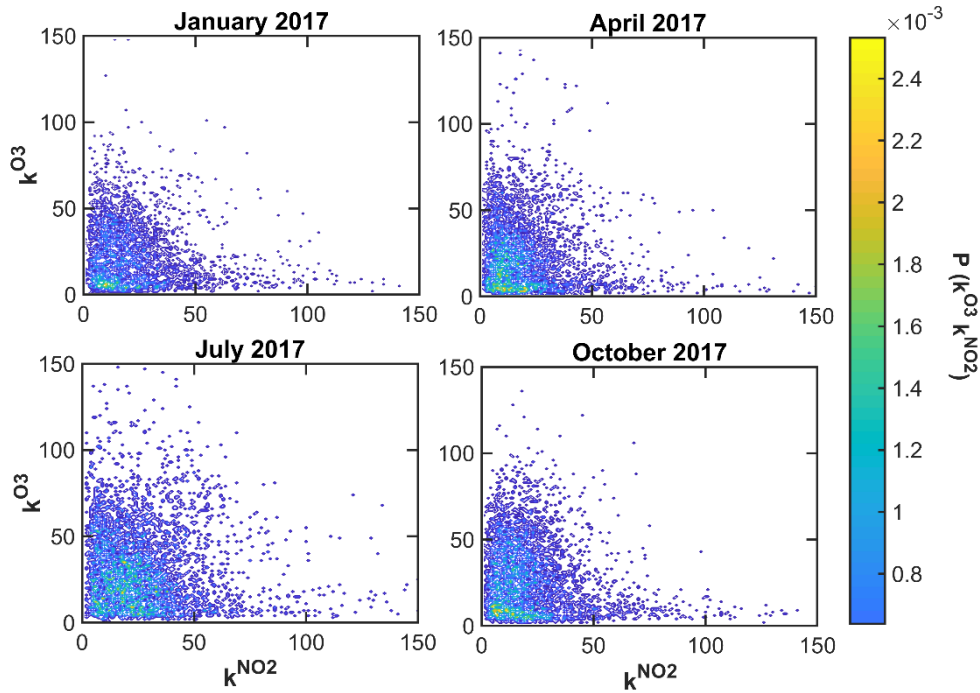
- 323 • In January,  $P(k^{O_3}, k^{NO_2})$  is more concentrated to specific combinations  
324 of degrees of the gases, specially, low  $k^{O_3}$  and higher  $k^{NO_2}$ . This  
325 translates into fewer values of the temporal series that are  
326 considerably correlated and thus, the overall correlation will decrease  
327 (Figure 7). As a result, high concentrations of  $NO_2$  will not necessarily  
328 lead to greater production of ozone, as can be regarded in Figure 1;  
329 where there are many days with extreme values of nitrogen dioxide  
330 concentration, whereas the ozone levels remain at minima with  
331 respect to the rest of the year. The reason is that although high

332 concentrations of NO<sub>2</sub> are available, there is not enough solar  
333 radiation to make the optimal interaction possible.

334 • In July, the concentration of ozone rises, as it widely known. In Figure  
335 6,  $P(k^{O_3}, k^{NO_2})$  is in this case more homogeneous and non-null for the  
336 values of  $k$  where the vast majority of points are located:  $k \in [0, 50]$ .

337 The result of this will be an increase in the  $I_{O_3, NO_2}$  that is shown on  
338 average in Figure 7. In contrast to January, now the extreme  
339 concentrations of NO<sub>2</sub> coincide with those of O<sub>3</sub>, for instance in Figure  
340 1 for July, from  $1.5 \cdot 10^4$  to  $2 \cdot 10^4$  minutes both reach their highest  
341 concentrations (taking into account that there exist a time delay  
342 between both quantities).

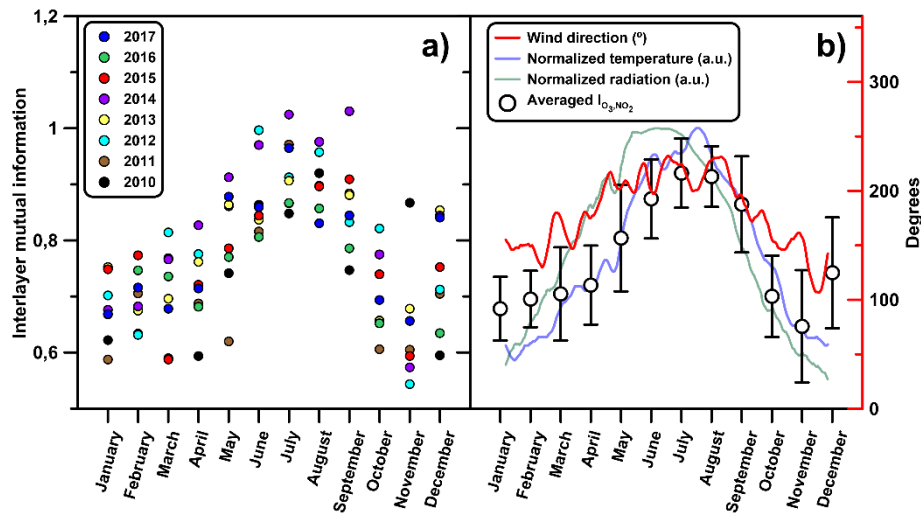
343 • Lastly, April and October are intermediate cases, where the  
344 distribution of  $P(k^{O_3}, k^{NO_2})$  is neither as acute as in January, nor as  
345 regular as July. In both cases, the probability is higher for finding low  
346  $k^{O_3}$  when  $k^{NO_2}$  is greater.



347

348 Figure 6: Graphical representation of the joint probability distribution of the degrees of  
 349 both layers for four sample months in 2017, the most recent year. Each point  
 350 represents the probability that degree is exactly  $k^{O_3}$  in the ozone VG, while is  $k^{NO_2}$  in  
 351 the nitrogen dioxide case, at the same time node.

352 The monthly computed values of  $I_{O_3,NO_2}$  are presented in Figure 7a, where the  
 353 different years are indicated by several colors. Figure 7b shows the monthly  
 354 average value (over all the mentioned years) and the standard deviation along  
 355 with the temperature, the average global radiation (normalized for the sake of  
 356 clarity) and wind direction.



357

358 Figure 7: a) Seasonal pattern obtained for  $I_{O_3,NO_2}$  for each one of the studied months  
 359 and years. b) Averaged  $I_{O_3,NO_2}$  over the whole period (2010-2017) for every month and  
 360 normalized temperature, solar radiation and wind direction for comparison. 0, 90, 180  
 361 and 240 degrees correspond to winds coming from the East, North, West and South  
 362 directions, respectively.

363

364 It can be appreciated that there is a sensible increase of the mutual  
 365 information between O<sub>3</sub> and NO<sub>2</sub> from May to September, coinciding with the  
 366 period of highest temperature and radiation. By definition,  $I_{\alpha,\beta}$  describes the  
 367 average correlation of the degree distributions and these can be used to describe  
 368 the nature of the time series (Lacasa et al., 2008; Mali et al., 2018). From that, it  
 369 could be inferred that the greater this parameter is, the more similarly are  
 370 expected to behave the signals underlying the VGs studied. Therefore, in this  
 371 case the degree distributions of the two pollutants are more correlated during the  
 372 summer months and the opposite during winter. It might indicate that they will  
 373 have a more similar behavior in the first case. For instance, O<sub>3</sub> is known to have  
 374 multifractal nature (Carmona-Cabezas et al., 2019a; He et al., 2017; Jiménez-  
 375 Hornero et al., 2010a; Pavon-Dominguez et al., 2013), hence NO<sub>2</sub> could be  
 376 expected to have higher multifractality in the summer months, being in  
 377 accordance with the findings reported in a prior work for the same area (Jiménez-

378 Hornero et al., 2010b). A higher multifractality is understood as a greater degree  
379 of multifractal behavior, which is usually related to complex systems with  
380 fluctuations appearing at different scales (Kantelhardt, 2011).

381 The reason for this clear seasonal pattern seems obvious looking at the  
382 distribution of solar radiation and temperature, which change greatly along the  
383 seasons in the study area (both maximal in summer and vice versa in winter).  
384 The first one of them (solar radiation) is the main source of energy for the  
385 photochemical reaction needed for the formation of  $O_3$  in the troposphere  
386 (Graedel and Crutzen, 1993), since nitrogen dioxide ( $NO_2$ ) and oxygen ( $O_2$ ) are  
387 recombined and produce ozone gas in the presence of ultraviolet light. The  
388 second one has been found to have relation as well with the production of ozone  
389 from nitrogen oxides in previous works in the same area (Pavón-Domínguez et  
390 al., 2015). Many of the previous studies are based on multifractal approaches to  
391 analyze time series, which have some disadvantages such as the need for  
392 choosing a scale interval where the searched behavior holds, leading to errors in  
393 the computation and some undesired ambiguities of the results. On the other  
394 hand, VGs are univocal for a given time series and their computation do not have  
395 any error associated, since their results are based on basic graph theory  
396 arithmetic.

397 Furthermore, regarding the average wind direction that can be seen in Figure  
398 7b, it is clearly seen how the direction is roughly  $\sim 220^\circ$  when the correlation  
399 between  $NO_2$  and  $O_3$  reaches its maximal value for summer, as commented  
400 before. This corresponds to wind coming as an average from the South-West.  
401 This fact has been previously described for the Guadalquivir Valley (Guardans  
402 and Palomino, 1995), where the pressure and temperature differences make

403 more predominant the wind coming from South-West direction when the  
404 temperature is maximum (summer). The reason behind this phenomenon is that  
405 the air mass moves from the plain areas towards the upper parts of the valley  
406 (North-East). The opposite case occurs in winter following an equivalent  
407 reasoning.

408 It must be pointed out the fact that in that predominant direction, the main  
409 sources of  $NO_2$  in the vicinities of Cordoba are located, since there are two of the  
410 main industrial parks of the city. Also, the most populated capital of the region  
411 (Sevilla) is situated in that direction, as well as the main highway that connect the  
412 two cities, which is one of the most transited roadways in Spain. This fact further  
413 corroborates the adequation of the correlation results, as was seen with the  
414 temperature and solar radiation previously.

#### 415 4. CONCLUSION

416 All the stated results confirm the capability of the two parameters provided by  
417 the MVGs for describing the interaction between ozone and nitrogen dioxide in  
418 the troposphere. On the one hand, authors consider that the first studied  
419 parameter ( $\omega$ ) may be used to infer the time shift in the coupling of the two  
420 systems, represented by the two layers. Given that the value of this parameter  
421 does not vary significantly neither in the different seasons, nor along the years,  
422 it means that  $\omega$  does not depend on any external factors, such as meteorological  
423 features. Authors believe that this quantity might be used to check theoretical  
424 models of  $O_3$  -  $NO_2$  interaction, although further investigation will be needed.

425 On the other hand,  $I_{O_3,NO_2}$  and  $P(k^{O_3}, k^{NO_2})$  could be used to have an insight  
426 of the correlation in the behavior of the series (from the degree, which is

427 associated to the concentration itself). Regarding the first one, one can obtain an  
428 overall look of how the  $\text{NO}_2$  and  $\text{O}_3$  are correlated depending on the  
429 concentration. For instance, it is possible to see how great values of  $\text{NO}_2$  correlate  
430 with large or low ones of  $\text{O}_3$ , and vice versa. Furthermore, the second quantity  
431 corresponds to a numerical value that measures the correlation of the whole set  
432 of series that are transformed into the different layers of the MVG.

433 To authors' mind, the outcomes of this research support the capability of  
434 multilayer complex networks for looking at the relation between several variables  
435 such as atmospheric pollutants. By using this approach, one can take advantage  
436 of some of its assets, as the computation efficiency or the univocity of the results.  
437 All this situates MVG as a suitable complementary technique for tackling analyses  
438 within the environmental problem. It is well compatible with others that have  
439 demonstrated to give satisfactory results, such as multifractal algorithms, as it has  
440 been demonstrated (Carmona-Cabezas et al., 2019a; Mali et al., 2018; P. Pavón-  
441 Domínguez et al., 2017). Hence one possible application of MVGs in this context  
442 could be the enlargement of databases used for predictive techniques that rely  
443 on data mining and machine learning. For this aim, it remains open the study of  
444 more pollutants and other variables through the methodology analyzed here.  
445 Since it is possible to focus MVGs on only two variables or to see the correlation  
446 of many of them at the same time, it allows a flexible analysis. All these  
447 considerations could come in handy for describing the many mechanisms  
448 involved in the dynamics of the photochemical smog for future works.

449

450

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455

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576

## **Declaration of interest**

There is no conflict of interests to declare.

1 **HIGHLIGHTS**

- 2 - Multiplex visibility graphs are used to check correlations between NO<sub>2</sub> and O<sub>3</sub>.
- 3 - NO<sub>2</sub> and O<sub>3</sub> have different behavior of their degree distributions along the year.
- 4 - Average edge overlap between the two pollutants remains constant in every
- 5 case.
- 6 - Interlayer mutual information evolves with a seasonal behavior every year.

