Air Quality in London: Evidence of Persistence, Seasonality and Trends

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ABSTRACT

The poor air quality in the London metropolis has sparked our interest in studying the time series dynamics of air pollutants in the city. The dataset consists of roadside and background air quality for seven standard pollutants: nitric oxide (NO), nitrogen dioxide (NO_2), oxides of nitrogen (NO_x), ozone (O_3), particulate matter (PM_{10} and $PM_{2.5}$) and sulphur dioxide (SO_2), using fractional integration to investigate issues such as persistence, seasonality and time trends in the data. Though we notice a large degree of heterogeneity across pollutants and a persistent behaviour based on a long memory pattern is observed practically in all cases. Seasonality and decreasing linear trends are also found in some cases. The findings in the paper may serve as a guide to air pollution management and European Union (EU) policymakers.

Keywords: Air quality; time trends; long memory; fractional integration; seasonality; London

JEL Classification: C22, Q53, Q58

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1. INTRODUCTION

The quality of air in London has improved significantly over the past decades (Browne et al. 2007; Colette et al. 2011; EEA 2017; Andrade et al. 2018; Lang et al. 2019), but exceedances of the legislative limit values still persist in some pollutants such as ozone (O_3), nitrogen dioxide (NO_2) and particulate matters (PM_{10} and $PM_{2.5}$), in both background and roadside datasets. London failed to meet the air quality European Union (EU) standard since 2010, largely due to diesel vehicles on its roads which resulted in high emissions of NO_2, and congestion has further compounded the effect of diesel fumes in the city. Diesel engines release NO_x, and large concentrations of NO_2 are experienced along roadsides in urban areas (Gardner and Dorling 1999; Font and Fuller 2016).

In 2016, 43 breaches of annual pollution were recorded in London. In 2017, the first breach of annual pollution limits was experienced in London, less than 10 days into the New Year, and this continued for about a month into 2018. However, for the first three months in 2010, no breach occurred (King´s College, 2019). Currently, people in London still live under poor air quality, however, NO_2 levels are falling and could reach the normal level for living in the next six years. The NO_2 and particulate matters exceed the EU standard, particularly during winter and early spring, not only in London but also in many other European cities (Bessagnet et al. 2005; Petit et al. 2017; etc.). Based on the rate of reduction in NO_2 between 2010 and 2016, Font et al. (2019) estimated that it would take about 193 years for NO_2 to reach legal levels.

The pollution has both health and social care costs, estimated to reach about £5.3 billion by 2035 (O'Hare 2018). This author further reported that the costs of air pollution in 2017 to the National Health Service (NHS) and social care were estimated at about £157 million. The research further predicted about 2.5 million new cases of coronary heart diseases, strokes, childhood cancer, lung cancer, pulmonary disease, diabetes, low birth weight and dementia by 2035. Fine particulate matter ($PM_{2.5}$) and NO_2 are particularly responsible for these costs. $PM_{2.5}$ is made up of particles with a diameter of less than 2.5 microns. These are emitted during the combustion of vehicle engine fuels, braking and tyre wearing, while NO_2 is released during the burning of fossil fuels, particularly diesel fuel. Both pollutants have been targeted by the UK government, seeking a strategy to reduce their exceedances. The Brexit deal could further challenge London's fight to improve the quality of air since the EU has stricter standards and rules than the UK is obliged to meet. Hence, concern is rising in the minds of dwellers regarding the future of London's air. Air pollution has proved to be stubborn in its behaviour, even if vehicle numbers are curbed, aircraft and agricultural pollution could prove more of a challenge to inhabitants, and further damage human health. There is a need to investigate the dynamics of the evolvement of the chemistry of air pollution since this will provide recommendations to modellers and predictors on the appropriate models to employ in the analysis of air pollution in London. Also, the findings in this paper will complement the management of air quality by the EU since there is a long-standing tradition of using modelling techniques to support the design of air quality policies by government authorities with regards to regulations on the emissions of pollutants.

In the present paper, we investigate the statistical properties of the data by looking at issues such as persistence, seasonality and time trends in the dynamic evolution of air quality chemistry in London. For this purpose, we use methods based on fractional integration, which extend the classical analysis of stationarity/non-stationarity that only use integer degrees of differentiation (i.e., 0 for stationarity and 1 for nonstationarity) to fractional values. Thus, by allowing the differencing parameter to be a fractional number, we allow for a much richer degree of flexibility in the dynamic specification of the data.

Studying air quality persistence, seasonality and trends inform us regarding the possible change in the current trend of the time series; the level of persistence gives us an idea of the impact that shocks would have on the series. In other words, it will tell us if the series would soon revert to its mean level or would be further pushed away from its mean path. In a highly persistent series, a shock to the series tends to persist for long periods of time and the series drifts away from its historical mean path. Then, a time trend indicates the general direction in which the series is moving, and the level of persistence will tell us if shocks will have a transitory or a permanent effect.

It is noticeable that most empirical research on air pollution has focused on a type of air pollutants such as NO_2, particulate matter 2.5 ($PM_{2.5}$) or sulphur dioxide (SO_2). The air pollution is determined by the chemical compositions of the mixture of pollutants such as SO_2, NO_2, carbon monoxide (CO), ozone (O_3), $PM_{2.5}$ and PM_{10}, as detailed by the World Health Organization (WHO, 2018) standards. Our research is also different from other existing studies in this field as it involves datasets of standard air pollutants in London.

The remainder of the paper is structured as follows: Section 2 presents a brief review of air quality modelling, while Section 3 is devoted to the methodology used in the paper. Section 4 describes the data. The empirical results are displayed in Section 5 and Section 6 contains some concluding comments.

2. LITERATURE REVIEW

Being able to assess and forecast the level of chemical composition of pollutants in the air can help in preventing harmful effects on public health and facilitate the efficiency of government policies aimed at improving air quality.

Most studies focus on two issues, on the one hand, seeking the connection between pollution and harmful health effects; and, on the other hand, quantifying, modelling and making predictions regarding air pollution by assessing the most suitable models for air pollution. The present paper is more related to this second line of research.

Dealing with the first line of research, Schwartz and Marcus (1990), using AutoRegressive Moving Average (ARMA) models, examined the connection between air pollution and mortality in London in the period 1958-1972. The results of the study demonstrated a high degree of correlation, in particular with SO_2. On the other hand, using predictive methods, Gardner and Dorling (1999) analyzed NO_x (a combination of NO and NO_2) using hourly meteorological data obtained from Central London using a multilayer perceptron (MLP) neural network model. The findings in the study showed that this model is more effective in modelling these pollutants compared to regression-based models.

Atkinson et al. (1999) examined the relationship between emergency admissions for respiratory problems and air pollution in London for the period from 1992 to 1994. They used Poisson regressions to determine the correlation between hospital admissions and the concentration of PM_{10} and SO_2. Salini and Pérez (2006) used the same methodology as Gardner and Dorling (1999), reaching similar results for the city of Santiago de Chile. They found that the simple perceptron with a linear function is more reliable for prediction than the persistence method; however, the function did not outperform a multi-layered network in its predictability. "It can be said that when nonlinear effects are not too important in modelling, multilayer networks are not significantly better than perceptron. However, as in this case, when these non-linear effects become important, multilayer networks are better in terms of their predictability, compared to linear models" (Salini and Pérez 2006; page 290). Pan and Chen (2008), using air pollution data in Taiwan, concluded that long memory AutoRegressive Fractional Integrated Moving Average (ARFIMA) models are more accurate than AutoRegressive Integrated Moving Average (ARIMA)-type models. Zamri et al. (2009), in assessing Malaysian air pollution, applied the Box-Jenkins ARIMA approach by modelling the maximum monthly chemical compositions of CO and NO_2 and showing an increasing trend since the 1996 limits set by the US National Ambient Air Quality Standards (NAAQS).

Beevers et al. (2013) combined two models for the study of pollution in London during 2008. The first is the KCL urban model that provides annual predictions of the air chemical compositions of NO, NO_2, O_3, PM_{10} and $PM_{2.5}$. The second one is the urban air quality model on a multiscale (CMAQ), which predicts air quality per hour. CMAQ is an acronym for the Community Multi-Scale Air Quality Model, a sophisticated atmospheric dispersion model developed by the US Environmental Protection Agency (EPA) to address regional air pollution problems. An example of a regional air pollution problem is a multi-state area where O_3 or $PM_{2.5}$ levels exceed the US health standards. The use of both models allows the quality of air, temporal and by source category to be predicted. However, the authors addressed the difficulty of measuring factors such as exhaust emissions, car braking, types of used tyres and, on the other hand, the need for a sociological study that includes data on public and private transport use, education, etc., to correct the uncertainty of the models.

Li et al. (2017) analysed air quality in Beijing from 2014 to 2016 to validate the effectiveness of the Long Short-Term Memory Neural Network Extended (LSTME) model. The models used are the Spatio-temporal Deep Learning (StDL), the Time Delay Neural Network (TDNN) model, the ARMA model, the Support Vector Regression (SVR) model, and the traditional LSTME model and concluded that the LSTME model is more effective than the other models as it is able to model time series with long-term dependencies with optimal time delays (Li et al. 2017; page 1002) and to capture more accurately effective space-time correlations and improving predictions. Naveen and Anu (2017) studied air quality in India using ARIMA and seasonal ARIMA (SARIMA) models, with the former being more effective than the latter. They also proposed the use of alternative models to improve prediction.

Regarding the studies focusing on air pollution and health effects in London, we should mention the studies of King's College London and the work of Anderson et al. (1996), in which, through a Poisson's regression model, they found a causal link between outdoor air pollution levels and mortality in London.

Our methodological approach is invariant to those in the literature, since it is based on long memory processes and use fractional integration in the analysis of air quality chemistry in London metropolis. Specifically, we dwelled on three properties of air quality in London, that is, the persistence, seasonality and time trend. The outcome of the findings in the paper would henceforth serve as eye opener to the choice of a forecasting model for air quality chemistry level.

3. METHODOLOGY

Long memory is a feature in time series that indicates that observations are highly dependent across time even if they are far distant apart. Many models can describe this behaviour and one very popular in Econometrics is the one based on fractional integration. Given a process $\{x_t, t = 0, \pm 1, \ldots\}$, we say that it is fractionally integrated or integrated of order d (i.e., $x_t \approx I(d)$) if after taking d-differences, the new process becomes stationary I(0). In other words, x_t is I(d) if its d-differences are short memory including here the white noise model but also the stationary ARMA-type of models.

Using L as the lag operator, i.e., $L^k x_t = x_{t-k}$, x_t is I(d) if:

$$(1 - L)^d x_t = u_t, \quad t = 1, 2, ... \tag{1}$$

where u_t is I(0) or short memory, and long memory takes places as long as the parameter d is positive, which may be a fractional value. This is clearly an advantage with respect the classical methods that exclusively consider integer degrees of differentiation, (usually, 1) and that based its statistical analysis on the unit root methods, simply distinguishing between stationarity (if d = 0) and nonstationarity (if d = 1). Thus, allowing d to be fractional we permit a much richer degree of flexibility in the dynamic specification of the data.

Our selected model is the following one,

$$y_t = \alpha + \beta t + x_t, \quad (1 - L)^d x_t = u_t, \quad t = 1, 2, ..., \tag{2}$$

where y_t is the observed time series, and α and β are unknown coefficients referring, respectively, to an intercept and a time trend; x_t is the regression error series, supposed to be I(d) and thus, u_t is an I(0) process, described first as a white noise process and then allowing for weak autocorrelation, in the latter case using the non-parametric spectral approach of Bloomfield (1973). The latter is a model for the I(0) term that is specified exclusively in terms of its spectral density function and that fits extremely well in the context of fractional integration. Moreover, its autocorrelation function decays exponentially fast as in the AR case, but unlike the AR models, it is stationary across all its values.

We estimate the fractional differencing parameter d along with the rest of parameters in equation (2) by using the Whittle function in the frequency domain, and, along with the estimates of the parameters in the model, we also presented the 95% confidence intervals of the non-rejection values of d using a simple version of the tests of

Robinson (1994) widely employed in the empirical literature on I(d) processes. (see, e.g., Gil-Alana and Robinson 1997; Gil-Alana 2005; Gil-Alana and Trani 2019 and others).

4. THE DATA

The London monthly average air quality levels dataset was obtained from the London Data website at https://datahub.io/core/london-air-quality, with both roadside and background datasets of standard air quality chemistry such as nitric oxide (NO), nitrogen dioxide (NO_2), oxides of nitrogen (NO_x), ozone (O_3), particulate matter (PM_{10} and $PM_{2.5}$) and sulphur dioxide (SO_2). These datasets are site averages, obtained from London Air Quality Network (LAQN) at:

https://www.londonair.org.uk/london/asp/datadownload.asp, at a daily frequency, and representing London daily air quality level. At LAQN, there were 30 background and 14 roadside sites; with 130 monitoring sites in Greater London with 51 background and 79 roadside sites (see, Font et al., 2019). Note that not all locations measured all pollutants, therefore it was more convenient to use datasets produced by the London Data website. The data are measured in micrograms per cubic meter of air (ug/m^3).

INSERT TABLE 1 ABOUT HERE

Time series ranges, as well as the corresponding number of observations of these air quality chemistries, are tabulated in Table 1. We observe the commencement of pollutants chemistry in 2008 for some variables, while others start in 2010 and all of them end in December 2018. Plots of each air quality chemical composition are given in Figures 1 and 2 for roadside and background readings, respectively. As noted in the plots for NO, NO_2, NO_x and O_3, seasonality is clearly noticeable, as the highest values of ozone level are found during summer periods in London (from June to August), while NO_2 and PM_{10} are at their lowest level during this period. For PM_{10}, $PM_{2.5}$ and SO_2, in both data reading sources (roadside, Figure 1 and background, Figure 2), we observe irregular time dynamics, albeit with occasional long spikes mimicking seasonality in the datasets. Summary statistics for the mean, minimum and maximum values of air quality chemistry levels for both roadside and background readings are given in Table 2. For NO_2, the mean value for the series is above the EU standard of 40μg/m^3 for the roadside readings of the pollutant, while this is below the standard for the background readings (34μg/m^3), whereas the maximum background value is above the standard (60.237μg/m^3). For PM_{10} and $PM_{2.5}$ in the roadside readings, the maximum values are above the exceedances limit, while only the $PM_{2.5}$ is above the EU standard, though the $PM_{2.5}$ is known to be more hazardous. Generally, there are wide disparities between maximum and minimum air pollution chemistry levels of all the seven pollutants for roadside and background readings.

INSERT FIGURES 1 AND 2 ABOUT HERE

INSERT TABLE 2 ABOUT HERE

5. EMPIRICAL RESULTS

The results in Table 3 are obtained under the assumption that the error term u_t in (2) is a white noise process. Thus, no autocorrelation is permitted apart from the one produced by the fractional differencing structure. We display the estimates of d (and the 95% confidence bands) for the three classic cases of i) no regressors, ii) with an intercept, and iii) with an intercept and a linear time trend, marking in bold in the table the selected case for each series depending on the significance of the coefficients based on their corresponding t-values.

INSERT TABLES 3 AND 4 ABOUT HERE

We observe in Table 3 that the time trend coefficient is significant in a number of cases, in particular for PM_{10} and $PM_{2.5}$ in the two cases of roadside and background series. Looking at the estimates of the differencing parameter, we observe that the unit root null hypothesis, i.e., d = 1 cannot be rejected in the cases of NO and NO_x for the London mean roadside, and in the cases of NO_x and O_3 for the background series. In all the other cases, the estimates of d are found to be statistically significantly smaller than 1 implying mean-reverting behaviour, and for two of the series ($PM_{2.5}$ in the roadside and PM_{10} in background), the I(0) hypothesis of short memory behaviour cannot be rejected, implying a very short degree of dependence between the observations in these two cases.

In Table 4, we allow for autocorrelation in the error term. As earlier mentioned, we use here a non-parametric approach due to Bloomfield (1973) which is quite convenient in the context of fractional integration and that approximates very well highly parameterized ARMA models with few parameters.[1] Here we observe that the time series is significant in a large number of cases and the estimated values of d are now smaller than 1 in all cases. Moreover, the I(0) hypothesis cannot be rejected now in the majority of the cases, and evidence of long memory (i.e., d > 0) only takes place in the cases of NO for roadside and SO_2 for background. Thus, it seems that the competition between the two structural approaches (No autocorrelation and Bloomfield autocorrelation cases) is the cause of the reduction in the values of the differencing parameter. These results, however, though allowing for autocorrelation, do not take into account the potential seasonal (monthly) nature of the data. Thus, in the following table, we take this feature into consideration by allowing for a seasonal monthly AR(1) structure on u_t, i.e.,

$$u_t = \phi u_{t-12} + \varepsilon_t , \qquad t = 1, 2, \dots , \qquad (3)$$

where ε_t is now a white noise process.

[1] See Gil-Alana (2004) for the modelization of Bloomfield (1973) in the context of fractional integration.

INSERT TABLES 5 AND 6 ABOUT HERE

The results using this model are presented in Table 5 with further results for selected cases in Table 5 displayed in Table 6. In Table 5, the long memory feature is found in the majority of the cases with the values of d belonging to the interval (0, 1). Evidence of short memory (d = 0) is only found in the cases of PM_{10} (roadside and background) and $PM_{2.5}$ (roadside) particulates. Table 6 displays the estimated model coefficients under the selected specification in Table 5. We observe significant seasonal AR coefficients and, in those cases where the time trend is statistically significant, it is found to be negative in all cases, implying a decreasing deterministic pattern in the data. This is consistent with other works that also find a decreasing trend in London air pollutants (e.g., Lang et al., 2019).

INSERT TABLE 7 ABOUT HERE

As a second approach in the analysis of the seasonality issue, we conducted seasonality tests using the method detailed in Beaulieu and Miron (1993). This is Hylleberg et al.'s (HEGY, 1990) test version for monthly frequency series. Nonstationarity and seasonality can both be tested in this approach simultaneously, following Box, Jenkins and Reinsel (2008) who proposed carrying out the lag operation $\left(1-L^{s}\right)x_{t}$ for seasonal differencing in order to obtain a transformed stationary weakly dependent series. For monthly data, as in our case, s = 12. The Seasonal Monthly Integration (SMI) process representing such a time series is,

$$\left(1-L^{12}\right)x_{t}=u_{t}, \quad t=1,2,... \tag{4}$$

where all the 12 roots lie within the unit circle. Using then the decomposition $(1 - L^{12})$ = $(1 - L)\,\phi(L)$, where $\phi(L) = (1 + L + L^{2} + \dots + L^{11})$, the first of these components, $(1 - L)$ is related to the regular (zero frequency) unit root test using the t_1 statistic, which is equivalent to the classical ADF unit root test. The remaining factors are then used in the seasonal unit root testing, while at annual frequency, the t statistic t_2 is used in testing annual seasonal unit roots. For all other frequencies, the joint test for seasonal unit root at any data frequency is carried out using the F test statistic $F_{2\text{-}12}$. (Details on HEGY testing procedure for monthly frequency dataset is found in Beaulieu and Miron 1993).

We conducted the HEGY test for the cases of i) an intercept only, ii) an intercept with a linear time trend and iii) with an intercept, a trend and seasonal dummies. The results are presented in Table 7 with the unit root test results for roadside readings presented in the upper panel (i) and those of background readings presented in the lower panel (ii). The results indicate non-rejection of the null hypotheses for non-seasonal unit root and seasonal unit root tests, for intercept only and an intercept with trend, based on t statistics t_1 and t_2, implying evidence of regular unit root and annual seasonal unit roots in the series. For all other frequencies other than the zero frequency (non-seasonal frequency), we conducted F tests F_{2-12} and these are significant throughout, implying that seasonality is only detected at the annual frequency but it is not found at any other frequency.

As a final step, we also examined the possibility of non-linear trends with smooth breaks, still within the context of fractional integration, and for this purpose, we used the method proposed in Cuestas and Gil-Alana (2016) which is based on the Chebyshev's polynomials in time.[2] In particular, we consider here the following model,

$$y_t \;=\; \sum_{i=0}^{m} \theta_i \, P_{iT}(t) \;+\; x_t\,; \qquad (1-L)^d x_t \;=\; u_t, \qquad t = 1, 2, \ldots, \qquad (5)$$

with m indicating the order of the Chebyshev polynomial $P_{i,T}(t)$ defined as:

$$P_{0,T}(t) \;=\; 1,$$

[2] This nonlinear deterministic function allows for modelling smooth breaks in the time series. It is similar in application to flexible Fourier function of Enders and Lee (2012a,b), recently applied in fractional integration framework in Gil-Alana and Yaya (2020).

$$P_{i,T}(t) = \sqrt{2}\cos\left(i\pi(t-0.5)/T\right), \qquad t = 1, 2, \ldots, T; \quad i = 1, 2, \ldots \qquad (6)$$

Hamming (1973) and Smyth (1998) showed detailed descriptions of these polynomials in time, and Bierens (1997) and Tomasevic and Stanivuk (2009) argued that it is possible to approximate highly non-linear trends with low degree polynomials. Thus, if $m = 0$ in (5), the model contains an intercept; if $m = 1$ it also includes a linear trend; and if $m > 1$ it becomes non-linear - the higher m is, the less linear the approximated deterministic component becomes. For our dataset, we set m = 3 and thus, significant values of the estimates of θ_2 and/or θ_3 will provide us with evidence of non-linearity.

INSERT TABLE 8 ABOUT HERE

Table 8 reproduces the results under the assumption that u_t in (5) is a white noise process, though almost identical results were obtained under weak (seasonal and non-seasonal) autocorrelation. The first thing we observe in this table is that there are very few cases of non-linearities. In fact, only for PM_{10} under the roadside series, and for SO_2 in the background reading do we find significant parameter estimates θ_2 and/or θ_3, implying nonlinearity of series dynamics. Focussing on the estimates of d, the results once more indicate some shreds of evidence of long memory patterns. Starting with the background case, there is a single case of a short memory pattern, i.e., d = 0 (PM_{10}); four series with values of d constrained between 0 and 1: SO_2, (d = 0.15), $PM_{2.5}$ (0.24), NO, (0.67) and NO_2, (0.68); and finally, there are two cases where the unit root null, i.e., d = 1 cannot be rejected: NO_x, (0.74) and O_3, (0.83). For the roadside series, evidence of I(0) behaviour is found in the two PM particulates (PM_{10} and $PM_{2.5}$). The unit root hypothesis cannot be rejected for NO, (with d = 0.73), and evidence of long memory with d ranging between 0 and 1 is obtained for the remaining cases.

6. CONCLUSIONS

The analyses carried out in this article and its results are transcendent as they examined the air quality in London by providing evidence of persistence, seasonality and time trends in various air quality pollutants. In particular, we have examined two datasets, these being roadside and background standard air quality chemistry readings such as nitric oxide, nitrogen dioxide, oxides of nitrogen, ozone, particulate matter (PM_{10} and $PM_{2.5}$) and sulphur dioxide. Our results indicate that long memory is present in the majority of the cases, implying high degrees of persistence measured in terms of a fractional differencing parameter that is constrained between 0 and 1. Mean reversion is also found practically in all cases since the values are found to be significantly smaller than 1. This means that shocks will tend to disappear by themselves in the long run. Seasonality is another relevant factor and the time trend if significant, is found to be negative in all cases indicating a reduction in pollutant levels.

The evidence of long memory is consistent with numerous other works that found this feature in many other disciplines including hydrology (Hurst, 1951), climatology (Gil-Alana, 2005; Rea et al., 2011; Gil-Alana and Sauci, 2019), environmental sciences (Barassi et al., 2011; Belbute and Pereira, 2017; Gil-Alana and Trani, 2019), etc. On the other hand, seasonality is something to be expected due to the monthly frequency of the data and the mixing layer dynamics, meteorological conditions and emission patterns.

The possibility of structural breaks has not been considered in this work. This is an important issue, noting that several authors have shown that fractional integration and structural breaks are very much related (Diebold and Inoue, 2001; Granger and Hyung, 2004; Ohanissian et al., 2008; etc.). Instead of that and based on the small number of observations used in this application, we have considered a nonlinear approach based on Chebyshev polynomials in time, and that approximates breaks in a rather smooth way.

The fractional integration modelling framework can easily be generalized to the Seasonal Autoregressive Fractionally Integrated Moving Average (SARFIMA) model or other variants which allow for forecasting values of the chemical composition of air quality, but the outcome of these findings would have served as eye opener to the choice of forecasting model, other than the SARFIMA model.[3] The findings could further help in the designing of environmental policies aimed at challenging and further reducing atmospheric pollution by 2050 in line with the UN 2030 Agenda, which focuses on the Sustainable Development Goals. It is essential to redesign the management of air pollution in London by focusing on improving emissions mainly from transportation. Also, appropriate measures should be taken in each seasonal quarter of London's weather since air quality is at its poorest level during the summer. This is because the tropospheric ozone, unlike other pollutants, is not emitted directly into the atmosphere, but it is a secondary pollutant produced by the reaction between nitrogen dioxide, hydrocarbons and sunlight; therefore, high levels of ozone pollution take place in the central hours of the day during the summer.

The data that support the findings of this study are openly available in the London Data website at https://datahub.io/core/london-air-quality

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Table 1: Time series of London Air Quality

London mean roadside			
Series	Starting period	Ending period	N. of Obs.
Nitric oxide, NO	January 2010	December 2018	108
Nitrogen dioxide, NO_2	January 2008	December 2018	132
Oxides of nitrogen, NO_x	January 2010	December 2018	108
Ozone, O_3	January 2008	December 2018	132
PM_{10} particulate	January 2008	December 2018	132
$PM_{2.5}$ particulate	January 2008	December 2018	132
Sulphur dioxide, SO_2	January 2008	December 2018	132
London mean background			
Nitric oxide, NO	January 2010	December 2018	108
Nitrogen dioxide, NO_2	January 2008	December 2018	132
Oxides of nitrogen, NO_x	January 2010	December 2018	108
Ozone, O_3	January 2008	December 2018	132
PM_{10} particulate	January 2008	December 2018	132
$PM_{2.5}$ particulate	May 2008	December 2018	128
Sulphur dioxide, SO_2	January 2008	December 2018	132

Table 2: Data Summary

Series	London mean roadside			London mean background		
	Mean	Min. value	Max. Value	Mean	Min. value	Max. value
Nitric oxide, NO	78.339	27.211	180.933	22.123	4.172	79.245
Nitrogen dioxide, NO_2	55.757	38.950	75.922	34.865	20.050	60.237
Oxides of nitrogen, NO_x	139.490	82.235	250.743	56.383	25.642	129.152
Ozone, O_3	27.174	10.658	46.266	36.895	13.869	62.562
PM_{10} particulate	25.122	16.285	43.315	19.272	11.927	36.933
$PM_{2.5}$ particulate	15.715	7.898	32.581	13.293	6.395	29.912
Sulphur dioxide, SO_2	3.263	-1.687	8.541	3.362	1.079	6.734

The Air Quality Standards Regulations 2010 is found at http://www.legislation.gov.uk/uksi/2010/1001/schedule/2/made. This documents the safe limits for NO_2 as 40 $\mu g/m^3$. For PM_{10} and $PM_{2.5}$, they are 40 $\mu g/m^3$ and 25 $\mu g/m^3$, respectively.

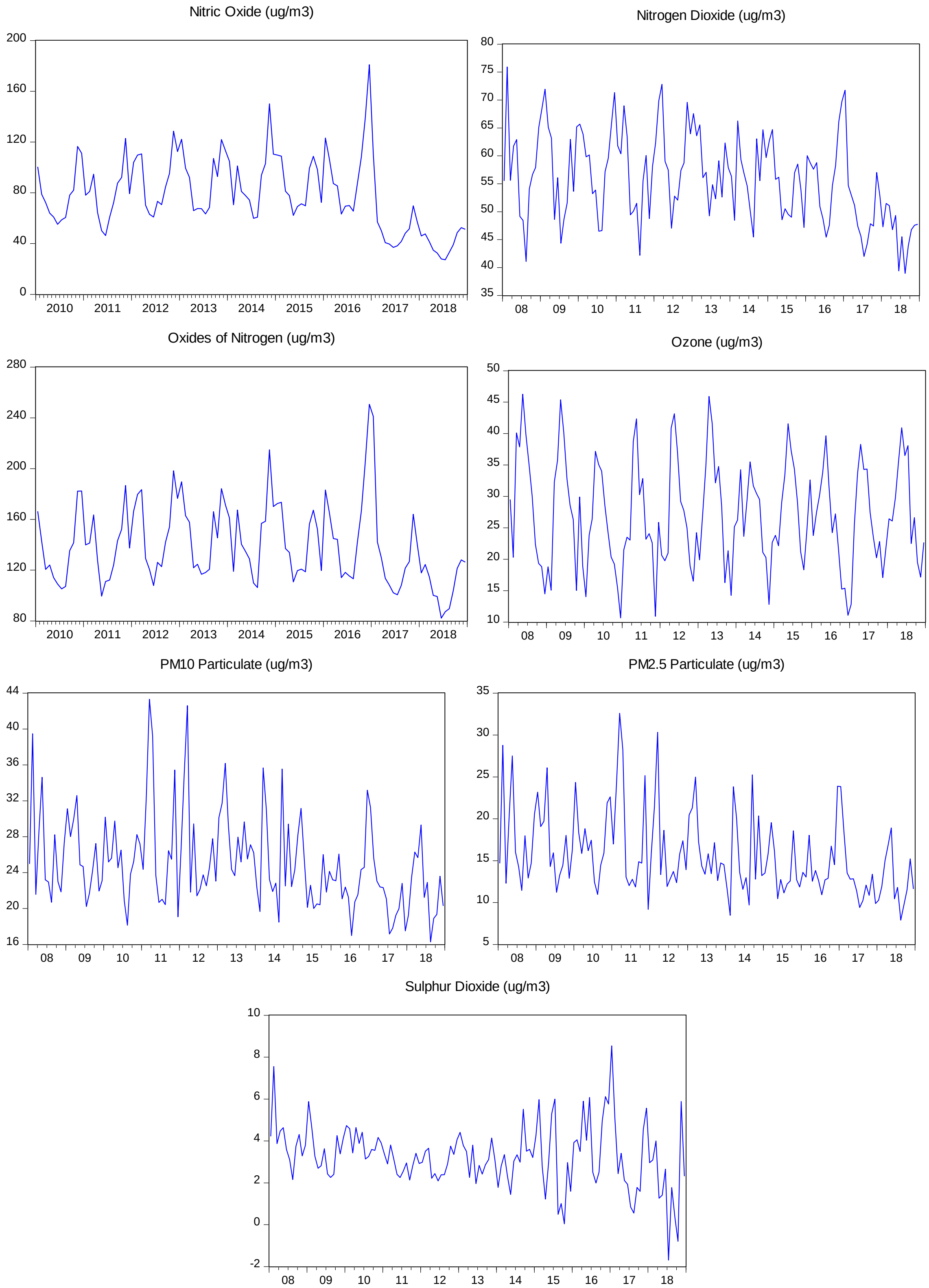

Figure 1: London Mean Roadside Air Quality readings on y-axis, measured in ug/m^3 and plotted year, the x-axis of each plot.

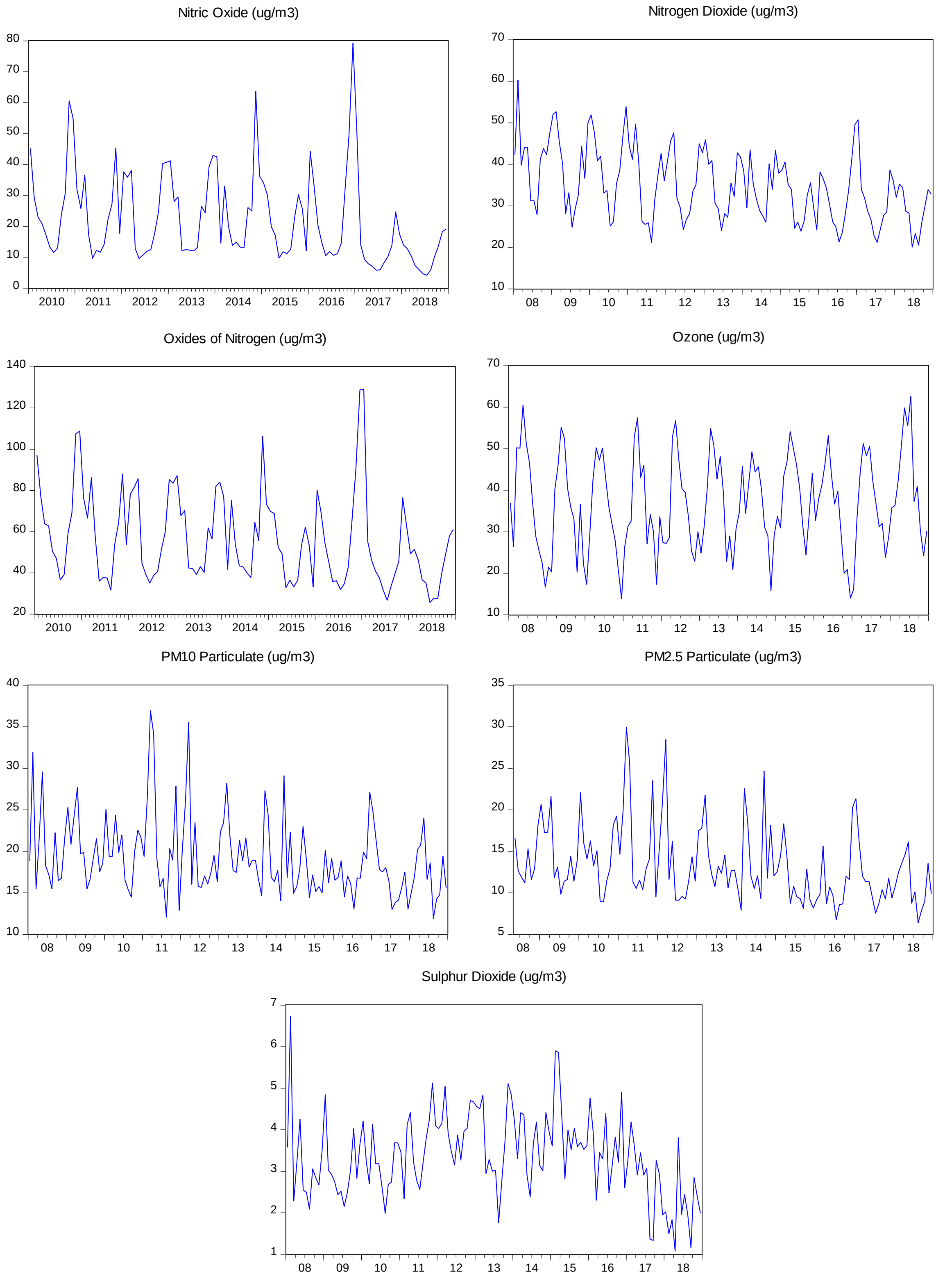

Figure 2: London Mean Background Air Quality readings on y-axis, measured in ug/m^3 and plotted year, the x-axis of each plot.

Table 3: Estimates of d under the assumption of no autocorrelation

London mean roadside			
Series	No terms	With intercept	With a time trend
Nitric oxide, NO	0.72 (0.62, 1.00)	**0.76 (0.56, 1.03)**	0.76 (0.55, 1.03)
Nitrogen dioxide, NO_2	0.82 (0.70, 0.99)	**0.50 (0.34, 0.70)**	0.48 (0.31, 0.70)
Oxides of nitrogen, NO_x	0.79 (0.63, 1.00)	**0.73 (0.50, 1.00)**	0.73 (0.50, 1.00)
Ozone, O_3	0.80 (0.63, 1.01)	**0.74 (0.51, 0.98)**	0.74 (0.51, 0.98)
PM_{10} particulate	0.58 (0.46, 0.72)	0.24 (0.12, 0.41)	**0.18 (0.04, 0.39)**
$PM_{2.5}$ particulate	0.48 (0.36, 0.63)	0.23 (0.12, 0.39)	**0.12 (-0.06, 0.35)**
Sulphur dioxide, SO_2	0.46 (0.31, 0.64)	**0.31 (0.17, 0.50)**	0.31 (0.16, 0.50)
London mean background			
Series	No terms	With intercept	With a time trend
Nitric oxide, NO	0.69 (0.48, 0.95)	**0.67 (0.42, 0.98)**	0.68 (0.42, 0.98)
Nitrogen dioxide, NO_2	0.87 (0.72, 1.07)	**0.68 (0.47, 0.93)**	0.68 (0.45, 0.93)
Oxides of nitrogen, NO_x	0.77 (0.58, 1.01)	**0.73 (0.46, 1.02)**	0.74 (0.47, 1.02)
Ozone, O_3	0.85 (0.67, 1.07)	**0.83 (0.60, 1.07)**	0.83 (0.60, 1.07)
PM_{10} particulate	0.52 (0.40, 0.66)	0.20 (0.09, 0.38)	**0.12 (-0.06, 0.35)**
$PM_{2.5}$ particulate	0.49 (0.37, 0.64)	0.31 (0.18, 0.51)	**0.27 (0.08, 0.50)**
Sulphur dioxide, SO_2	0.47 (0.34, 0.62)	**0.33 (0.24, 0.46)**	0.33 (0.24, 0.46)

In bold, the significant cases according to the deterministic terms. In parenthesis, the 95% confidence intervals of the values of d.

Table 4: Estimates of d under the assumption of autocorrelation

London mean roadside			
Series	No terms	With intercept	With a time trend
Nitric oxide, NO	0.51 (0.22, 0.91)	0.35 (0.15, 0.76)	**0.32 (0.11, 0.76)**
Nitrogen dioxide, NO_2	0.71 (0.51, 0.99)	0.24 (0.03, 0.73)	**0.15 (-0.11, 0.73)**
Oxides of nitrogen, NO_x	0.54 (-0.03, 0.98)	**0.18 (-0.10, 0.74)**	0.14 (-0.13, 0.74)
Ozone, O_3	0.32 (-0.12, 1.04)	**-0.26 (-0.57, 0.73)**	-0.23 (-0.58, 0.73)
PM_{10} particulate	0.51 (-0.06, 0.76)	0.07 (-0.06, 0.27)	**-0.10 (-0.26, 0.27)**
$PM_{2.5}$ particulate	0.45 (-0.13, 0.70)	0.09 (-0.05, 0.30)	**-0.37 (-0.59, 0.30)**
Sulphur dioxide, SO_2	0.28 (-0.14, 0.73)	0.02 (-0.24, 0.39)	**0.03 (-0.23, 0.39)**
London mean background			
Series	No terms	With intercept	With a time trend
Nitric oxide, NO	-0.01 (-0.19, 0.67)	0.00 (-0.23, 0.76)	**-0.18 (-0.47, 0.45)**
Nitrogen dioxide, NO_2	0.59 (0.35, 0.96)	0.17 (-0.01, 0.73)	**-0.43 (-0.81, 0.83)**
Oxides of nitrogen, NO_x	-0.12 (-0.19, 0.89)	-0.08 (-0.34, 0.74)	**-0.46 (-1.04, 0.58)**
Ozone, O_3	-0.05 (-0.11, 1.11)	-0.17 (-0.54, 0.73)	**-0.28 (-0.66, 0.90)**
PM_{10} particulate	0.49 (0.27, 0.73)	0.05 (-0.08, 0.27)	**-0.30 (-0.49, 0.07)**
$PM_{2.5}$ particulate	0.45 (-0.11, 0.71)	0.12 (-0.03, 0.30)	**-0.17 (-0.38, 0.22)**
Sulphur dioxide, SO_2	0.32 (0.09, 0.59)	**0.29 (0.13, 0.39)**	0.30 (0.17, 0.51)

In bold, the significant cases according to the deterministic terms. In parenthesis, the 95% confidence intervals of the values of d.

658 **Table 5: Estimates of d under the assumption of seasonal autocorrelation**

London mean roadside			
Series	No terms	With intercept	With a time trend
Nitric oxide, NO	0.72 (0.54, 0.95)	**0.60 (0.41, 0.91)**	0.60 (0.39, 0.91)
Nitrogen dioxide, NO_2	0.79 (0.66, 0.96)	0.36 (0.23, 0.56)	**0.32 (0.16, 0.54)**
Oxides of nitrogen, NO_x	0.74 (0.57, 0.96)	**0.54 (0.33, 0.86)**	0.53 (0.32, 0.86)
Ozone, O_3	0.55 (0.40, 0.75)	**0.24 (0.09, 0.50)**	0.25 (0.10, 0.50)
PM_{10} particulate	0.52 (0.39, 0.68)	0.14 (0.03, 0.29)	**0.08 (-0.06, 0.27)**
$PM_{2.5}$ particulate	0.42 (0.28, 0.58)	0.16 (0.05, 0.32)	**0.06 (-0.10, 0.27)**
Sulphur dioxide, SO_2	0.44 (0.30, 0.60)	**0.30 (0.17, 0.46)**	0.28 (0.14, 0.46)
London mean background			
Series	No terms	With intercept	With a time trend
Nitric oxide, NO	0.59 (0.36, 0.88)	**0.47 (0.25, 0.85)**	0.46 (0.19, 0.85)
Nitrogen dioxide, NO_2	0.77 (0.62, 0.96)	0.34 (0.22, 0.53)	**0.20 (0.00, 0.50)**
Oxides of nitrogen, NO_x	0.68 (0.48, 0.94)	**0.47 (0.25, 0.85)**	0.46 (0.19, 0.85)
Ozone, O_3	0.52 (0.34, 0.74)	**0.27 (0.12, 0.52)**	0.26 (0.09, 0.53)
PM_{10} particulate	0.45 (0.31, 0.61)	0.12 (-0.03, 0.28)	**0.04 (-0.11, 0.25)**
$PM_{2.5}$ particulate	0.44 (0.31, 0.60)	0.26 (0.13, 0.44)	**0.21 (0.05, 0.42)**
Sulphur dioxide, SO_2	0.45 (0.31, 0.60)	**0.28 (0.18, 0.42)**	0.28 (0.18, 0.42)

659 In bold, the significant cases according to the deterministic terms. In parenthesis, the 95% confidence
660 intervals of the values of d.
661
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Table 6: Estimates of d under the assumption of seasonal autocorrelation

London mean roadside				
Series	d (95% interval)	Intercept	Time trend	AR
Nitric oxide, NO	0.60 (0.41, 0.91)	84.921 (5.54)	---	0.354
Nitrogen dioxide, NO_2	0.32 (0.16, 0.54)	60.442 (19.15)	-0.080 (-1.99)	0.428
Oxides of nitrogen, NO_x	0.54 (0.33, 0.86)	143.642 (8.51)	---	0.420
Ozone, O_3	0.24 (0.09, 0.50)	27.478 (14.20)	---	0.669
PM_{10} particulate	0.08 (-0.06, 0.27)	28.009 (25.18)	-0.044 (-3.10)	0.382
$PM_{2.5}$ particulate	0.06 (-0.10, 0.27)	18.819 (19.38)	-0.047 (-3.76)	0.286
Sulphur dioxide, SO_2	0.30 (0.17, 0.46)	3.410 (8.52)	---	0.203
London mean background				
Series	d (95% interval)	Intercept	Time trend	AR
Nitric oxide, NO	0.47 (0.25, 0.85)	26.871 (4.02)	---	0.359
Nitrogen dioxide, NO_2	0.20 (0.00, 0.50)	41.813 (17.55)	-0.101 (-3.40)	0.646
Oxides of nitrogen, NO_x	0.47 (0.25, 0.85)	66.240 (6.46)	---	0.429
Ozone, O_3	0.27 (0.12, 0.52)	37.105 (13.76)	---	0.692
PM_{10} particulate	0.04 (-0.11, 0.25)	21.795 (24.58)	-0.038 (-3.32)	0.646
$PM_{2.5}$ particulate	0.21 (0.05, 0.42)	15.642 (10.77)	-0.038 (-2.04)	0.646
Sulphur dioxide, SO_2	0.28 (0.18, 0.42)	3.332 (13.20)	---	0.230

In parenthesis in the second column, the 95% confidence intervals of the estimated values of d. in the third and fourth column, t statistics values are in parenthesis. The "AR" is the estimated seasonal AR(1) values.

Table 7: Results of the HEGY Quarterly Seasonal unit root

i) London mean roadside

Series	Regression	t_1	t_2	F_{2-12}
Nitric oxide, NO	Intercept only	0.2429	-0.8343	**7.9221**
	Intercept and trend	-0.7950	-0.8413	**7.7816**
	Intercept, trend and seasonal dummy	0.8748	0.5871	**13.8999**
Nitrogen dioxide, NO_2	Intercept only	-0.7176	-0.8088	**6.1134**
	Intercept and trend	-2.3521	-0.8124	**6.1146**
	Intercept, trend and seasonal dummy	2.8932	**2.9362**	**9.9875**
Oxides of nitrogen, NO_x	Intercept only	-0.8530	-0.6957	**7.5510**
	Intercept and trend	-1.8307	-0.6930	**7.4601**
	Intercept, trend and seasonal dummy	2.6703	2.6227	**14.6369**
Ozone, O_3	Intercept only	-1.2149	-1.8058	**4.5338**
	Intercept and trend	-1.6516	-1.8080	**4.4319**
	Intercept, trend and seasonal dummy	**3.6920**	**3.5954**	**10.5347**
PM_{10} particulate	Intercept only	-0.2150	-0.8383	**5.0926**
	Intercept and trend	-2.8297	-0.6235	**5.0243**
	Intercept, trend and seasonal dummy	3.2142	3.2061	**6.9362**
$PM_{2.5}$ particulate	Intercept only	-0.0164	-0.7408	**4.9104**
	Intercept and trend	-1.9584	-0.6437	**4.8029**
	Intercept, trend and seasonal dummy	2.4029	2.4659	**7.7611**
Sulphur dioxide, SO_2	Intercept only	1.3854	-0.8968	**5.3395**
	Intercept and trend	0.3269	-0.8863	**4.9650**
	Intercept, trend and seasonal dummy	1.3805	1.6499	**5.8573**

ii) London mean background

Series	Regression	t_1	t_2	F_{2-12}
Nitric oxide, NO	Intercept only	1.4599	-0.7625	**6.8367**
	Intercept and trend	0.5360	-0.7741	**6.8876**
	Intercept, trend and seasonal dummy	-0.1487	-0.2389	**12.0501**
Nitrogen dioxide, NO_2	Intercept only	0.9932	-0.5471	**5.5080**
	Intercept and trend	-0.8502	-0.5334	**5.4032**
	Intercept, trend and seasonal dummy	1.5246	1.6500	**8.1662**
Oxides of nitrogen, NO_x	Intercept only	0.4292	-0.5903	**6.6705**
	Intercept and trend	-0.1071	-0.6044	**6.6629**
	Intercept, trend and seasonal dummy	0.9679	0.8257	**12.3667**
Ozone, O_3	Intercept only	-1.9739	-1.3880	**3.6973**
	Intercept and trend	-1.9417	-1.3786	**3.6504**
	Intercept, trend and seasonal dummy	**3.9302**	**3.7862**	**10.5785**
PM_{10} particulate	Intercept only	0.3369	-0.8042	**4.7729**
	Intercept and trend	-3.0612	-0.6396	**4.6386**
	Intercept, trend and seasonal dummy	**3.4327**	**3.4149**	**6.6551**
$PM_{2.5}$ particulate	Intercept only	0.1899	-0.6368	**5.5896**
	Intercept and trend	-2.4430	-0.4708	**5.4172**
	Intercept, trend and seasonal dummy	3.1601	**3.2390**	**8.3624**
Sulphur dioxide, SO_2	Intercept only	-0.9885	**-2.3177**	**5.6763**
	Intercept and trend	-0.9644	-2.2991	**5.5756**
	Intercept, trend and seasonal dummy	2.4293	2.0294	**7.4713**

In bold in the column for t statistic t_1 indicates evidence of no regular unit root at 5% level, while in bold for a column for t statistic t_2 indicates evidence of no seasonal unit roots at annual frequency, vice-versa. In the last column, the joint F tests F_{2-12} are reported with Estimates in bold implying no evidence of seasonal unit roots at all other frequencies other than annual frequency. Critical values of the test are given in Franses and Hobijn (1997).

Table 8: Estimates based on a non-linear I(d) model with white noise errors

i) London Mean Roadside					
Series	d	θ_0	θ_1	θ_2	θ_3
Nitric oxide, NO	0.73 (0.47, 1.01)	**86.585** **(2.31)**	9.415 (0.44)	-11.400 (-0.80)	5.712 (0.53)
Nitrogen dioxide, NO_2	0.46 (0.25, 0.68)	**55.833** **(13.20)**	2.918 (1.12)	-1.845 (-0.87)	1.015 (0.56)
Oxides of nitrogen, NO_x	0.72 (0.47, 0.99)	**150.178** **(3.30)**	6.606 (0.25)	-7.393 (-0.42)	3.911 (0.29)
Ozone, O_3	0.74 (0.51, 0.98)	**27.362** **(1.91)**	0.668 (0.08)	0.045 (0.01)	0.828 (0.20)
PM_{10} particulate	0.11 (-0.09, 0.37)	**25.152** **37.06)**	**1.692** **(2.85)**	**-0.968** **(-1.71)**	-1.816 (-0.03)
$PM_{2.5}$ particulate	0.11 (-0.08, 0.35)	**15.717** **(24.53)**	**1.797** **(3.21)**	-0.294 (-0.55)	-0.004 (-0.09)
Sulphur dioxide, SO_2	0.28 (0.12, 0.48)	**86.585** **(2.31)**	9.415 (0.44)	-11.400 (-0.80)	5.712 (0.53)
ii) London Mean Background					
Series	d	θ_0	θ_1	θ_2	θ_3
Nitric oxide, NO	0.67 (0.40, 0.97)	**27.937** **(1.78)**	4.822 (0.48)	-0.990 (-0.13)	2.534 (0.46)
Nitrogen dioxide, NO_2	0.68 (0.45, 0.93)	**36.407** **(3.45)**	3.852 (0.64)	0.564 (0.13)	0.784 (0.24)
Oxides of nitrogen, NO_x	0.74 (0.47, 1.02)	**71.022** **(2.00)**	7.532 (0.37)	1.332 (0.10)	2.748 (0.27)
Ozone, O_3	0.83 (0.60, 1.07)	**36.431** **(1.85)**	-0.300 (-0.02)	0.219 (0.02)	0.527 (0.07)
PM_{10} particulate	0.09 (-0.11, 0.34)	**19.281** **33.77)**	**1.486** **(2.91)**	-0.324 (-0.66)	-0.203 (-0.42)
$PM_{2.5}$ particulate	0.24 (0.04, 0.49)	**13.277** **(12.71)**	**1.557** **(1.98)**	-0.418 (-0.59)	-0.309 (-0.47)
Sulphur dioxide, SO_2	0.15 (0.00, 0.35)	**3.373** **(23.85)**	0.146 (1.24)	**-0.438** **(-3.96)**	**0.224** **(2.12)**

In parenthesis in the second column, the 95% confidence interval for d. In the third to the sixth column are t-values for parameters θ_0, θ_1, θ_2 and θ_3, respectively. Figures in bold indicate the significance of estimates at 5% level.