Long-term trends in $PM_{2.5}$ mass and particle number concentrations in urban air: the impacts of mitigation measures and extreme events due to changing climates

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ABSTRACT

Urbanisation and industrialisation led to the increase of ambient particulate matter (PM) concentration. While subsequent regulations may have resulted in the decrease of some PM matrices, the simultaneous changes in climate affecting local meteorological conditions could also have played a role. To gain an insight into this complex matter, this study investigated the long-term trends of two important matrices, the particle mass ($PM_{2.5}$) and particle number concentrations (PNC), and the factors that influenced the trends. Mann-Kendall test, Sen's slope estimator, the generalised additive model, seasonal decomposition of time series by LOESS (locally estimated scatterplot smoothing) and the Buishand range test were applied. Both $PM_{2.5}$ and PNC showed significant negative monotonic trends (0.03–0.6 $\mu g.m^{-3}.yr^{-1}$ and 0.40–3.8 x 10^3 $particles.cm^{-3}.yr^{-1}$, respectively) except Brisbane (+0.1 $\mu g.m^{-3}.yr^{-1}$ and +53 $particles.cm^{-3}.yr^{-1}$, respectively). For the period covered in this study, temperature increased (0.03–0.07 $°C.yr^{-1}$) in all cities except London; precipitation decreased (0.02–1.4 $mm.yr^{-1}$) except in Helsinki; and wind speed was reduced in Brisbane and Rochester but increased in Helsinki, London and Augsburg. At the change-points, temperature increase in cold cities influenced PNC while shifts in precipitation and wind speed affected $PM_{2.5}$. Based on the LOESS trend, extreme events such as dust storms and wildfires resulting from changing climates caused a positive step-change in concentrations, particularly for $PM_{2.5}$. In contrast, among the mitigation measures, controlling sulphur in fuels caused a negative step-change, especially for PNC. Policies regarding traffic and fleet management (e.g. low emission zones) that were implemented only in certain areas or in a progressive uptake (e.g. Euro emission standards), resulted to gradual reductions in concentrations. Therefore, as this study has clearly shown that $PM_{2.5}$ and PNC were influenced differently by the impacts of the changing climate and by the mitigation measures, both metrics must be considered in urban air quality management.

Keywords: $PM_{2.5}$, particle number concentration, ultrafine particles, mitigation, climate variabilities

Introduction

Air quality has changed throughout history, but particularly over the past few decades. Elevated concentrations of air pollutants due to industrialisation and urbanisation, in particular, have become a global problem because of their impacts on human health and the environment. To address this problem, local and national authorities in an increasing number of countries have been introducing policies and strategies to mitigate anthropogenic emissions and improve air quality. As a result, improvements in air quality have been observed, for example, in the United States (USEPA, 2019), the European Union (EEA, 2018) and China (Fontes et al., 2017). Conversely, where policies and strategies are not implemented, air quality continues to worsen due to the emissions from an increasing number of local and regional sources, in particular the transportation sector and fossil fuels for energy generation (Al-Taani et al., 2019; Pant et al., 2019).

Airborne particulate matter (PM) is one of the most relevant pollutants to human health, with both short- and long-term exposure linked to increased morbidity and mortality (Atkinson et al., 2010; Tobías et al., 2018). To add to the complexity, the impacts of PM on health are related to the particle size: smaller particles, such as those emitted by combustion sources, have a lower deposition velocity and therefore stay suspended longer in the air (Rose et al., 2012; Schmale et al., 2011); and they also deposit deeper in the respiratory tract causing a range of local and systemic health effects (Fang et al., 2017; Fireman et al., 2017). With the growing understanding of the negative impacts of PM, standards for particle mass concentration have been introduced in many countries worldwide and compliance monitoring of $PM_{2.5}$ and PM_{10} has been conducted (mass concentration of particles with an aerodynamic diameter < 2.5 μm and < 10 μm, respectively). However, there are no standards, and therefore little monitoring is conducted, for ultrafine particles (UFPs, size <100 nm); although with traffic being a major pollution source in cities around the world, this size fraction of PM may be more significant in terms of health impacts than larger particles of higher mass in urban air (Kumar et al., 2014; Rönkkö et al., 2017). UFPs are measured in terms of particle number concentration (PNC), rather than mass.

An important factor that affects particles of different sizes somewhat differently is meteorology. A changing climate, which in turn affects local and global meteorological parameters, can also have an impact on particle characteristics, irrespective of the impact of changes in the sources. For example, stronger winds will, in general, result in higher resuspension of larger particles, but faster dispersal and thus dilution of smaller particles (Teinilä et al., 2019). On the other hand, colder ambient temperatures with high relative humidity (RH) can increase PNC by favouring nucleation, especially during winter (Jeong et al., 2006; Rönkkö et al., 2006), but higher temperatures with low RH (below 60%) enhance H_2SO_4 levels in the air, promoting new particle formation (An et al., 2015; Birmili & Wiedensohler, 2000; Hamed et al., 2011).

Smaller and larger particles in the air typically originate from different sources and, therefore, require different mitigation strategies. Conversely, mitigation strategies have different impacts on particles of different sizes; therefore, the concentration trends could differ between $PM_{2.5}$ and PNC as evidenced by the experience in Eastern Germany after the German reunification in 1990 (Kreyling et al., 2003; Pitz et al., 2001). A comprehensive review of the measurement metrics, source apportionment, health effects and legislations on PM by Heal et al. (2012) revealed that controlling $PM_{2.5}$ and PM_{10} resolves only a part of the problem, and does not necessarily address the problem of UFPs. Therefore, our study of the long-term trends of both $PM_{2.5}$ and PNC further illustrates that monitoring and characterising air quality in terms of PM mass concentrations only, without conducting any monitoring of PNC, might be insufficient given that the sources and drivers for $PM_{2.5}$ and PNC differ, as well as their impacts relating to human health.

Long-term studies of $PM_{2.5}$ and PNC have shown that the impacts of emission control strategies and policies can be either a steady decrease or a step change. For example, the consistent decrease of $PM_{2.5}$ in Seoul, South Korea, in the period from 2004 to 2013 can be partially explained by the implementation of several emission reduction strategies such as the use of natural gas as a bus fuel and the installation of emission control retrofits (Ahmed et al., 2015). However, an abrupt reduction in PNC was observed when London, England, introduced sulphur-free diesel fuel and a traffic pollution charge scheme for heavy goods vehicles in 2007 (Jones et al., 2012). Trend analysis for PM metrics is commonly done by using simple linear regression such as the Theil-Sen method to obtain the slope that quantifies gradual changes. However, this cannot capture significant patterns in time series data as effectively as curve fitting by applying smoothing functions. Moreover, doing a time series decomposition prior to analysis to separate trend, seasonality, and noise components are more precise when specific attribution is desired.

Considering the need to understand the effectiveness of mitigation measures on controlling particles in the air, but with the backdrop of other changes occurring, in particular climatic changes that will affect meteorological parameters, the aims of this work were to: (1) determine the long-term trends of $PM_{2.5}$ and PNC in cities using time series analysis; (2) evaluate the impact of changes in climate (based on key meteorological factors, after removing seasonality) on the observed trends of $PM_{2.5}$ and PNC; and (3) investigate whether the observed changes in $PM_{2.5}$ and PNC can be attributed to modifications in the operation of anthropogenic sources. Analysis of long-term trends in concentration changes using both $PM_{2.5}$ and PNC can provide an understanding of the magnitude of changes and of the factors that influenced their ambient concentrations; in particular, the efficiency of human interventions (e.g. changes in technology or fuels and the impact of new regulations). This information can provide a more complete picture for policy makers and state leaders to design a more effective and efficient regulatory approach.

Material and methods

The criteria for inclusion of data in this study were: (1) measurements of $PM_{2.5}$, PNC and the selected meteorological parameters (mean air temperature, total precipitation and mean wind speed) performed for at least 10 years; (2) $PM_{2.5}$, PNC and the selected meteorological parameters to be collected concurrently and from the same location or in proximity; and (3) measurements to be recorded at monthly resolution or higher. Data acquisition was done through convenience by connecting with colleagues on our collaborative network based on our knowledge of data availability. Five cities that fulfilled the above criteria for $PM_{2.5}$, PNC, and the meteorological parameters are listed in Table 1 together with the sources and mitigation measures. A brief description of the cities and stations is given in the supplementary material including station type and instruments used (Table S1). Urban background (UB) measuring stations for PM concentrations were preferred over roadside (RS) stations to represent the ambient condition of the whole city area, which has several other sources, rather than be biased to traffic as the source.

Table 1. Climate classification, identified PM sources and relevant legislations on emission control in the study areas

City [1]Climate	[2]Sources	[3]Regulations
Augsburg, Germany (Cfb, Marine West Coast)	Local traffic, biomass burning (for heating), secondary aerosol (Gu et al., 2011; Schäfer et al., 2016)	2005 – Limit values for air pollutants and the first air pollution control plan 2008 – Federal Emission Control Act 2009 – Low emission zone 2010 – Average Exposure Indicator
Brisbane, Australia (Cfa, Humid Subtropical)	Local traffic, secondary aerosol, biomass burning (controlled and forest fires), sea salt (Cheung et al., 2011; Friend & Ayoko, 2009; Friend et al., 2012)	1998 – AAQ NEPM (Ambient Air Quality National Environment Protection Measure) 2001 – Renewable Energy Target 2002 – Fuel Quality Standards Act 2003 – $PM_{2.5}$ standard included in the AAQ NEPM; Ultra-low sulphur diesel (ULSD) in bus 2010 – Congestion Reduction Unit 2014 – Emission Reduction Fund 2015 – $PM_{2.5}$ become part of the reporting standards
Helsinki, Finland (Dfb, Humid Continental)	Local traffic, secondary aerosol, biomass burning (for heating and regional), long-range transport (Carbone et al., 2014; Kupiainen et al., 2016; Pirjola et al., 2017; Saarikoski et al., 2008; Timonen et al., 2013)	2005 - "Sulphur-free" (max 10 $mg.kg^{-1}$ fuel) diesel and petrol 2008 – City of Helsinki Air Quality Action Plan 2008-2016 2010 – Low emission zone for city buses and garbage trucks ; Low-sulphur (max 1%) marine fuels in the Baltic Sea (IMO) 2015 – Very low-sulphur (max 0.1%) marine fuels in the Baltic Sea (IMO)
London, United Kingdom (Cfb, Marine West Coast)	Local traffic, secondary aerosol, crustal, sea salt, urban/regional background (Beddows et al., 2015; Charron et al., 2007; Crilley et al., 2017)	2003 – Congestion Charging Scheme 2007 – "Sulphur-free" diesel and petrol; Low-sulphur marine fuels in the North Sea and English Channel (IMO); UK Air Quality Strategy 2008 – Low Emissions Zone 2010 – London Air Quality Strategy 2011 – Progressive uptake of Euro 5 passenger cars
Rochester, United States of America (Dfb, Humid Continental)	Local traffic, secondary aerosol, crustal, biomass burning (for heating and wildfires), sea salt (Squizzato et al., 2018a, 2018b)	2003 – Regulation to reduce SO_2 and NO_x emissions from electricity generation 2004 – Increase renewable energy sources' contribution to electricity generation from 19% to 25% by 2013 2006 – Gasoline sulphur standard (30 ppm refinery average and 80 ppm per gallon cap) 2007 – *Heavy-Duty Highway Rule* – on-road diesel fuels with ultra-low sulphur (< 15ppm) 2010 – Non-road diesel fuel sulphur standard (< 15 ppm) 2012 – Locomotive and marine diesel fuel sulphur standard (< 15 ppm) from large refiners 2014 – Locomotive and marine diesel fuel sulphur standard (< 15 ppm) from small refiners

[1]Köppen climate classification – a system to classify climate based on the annual and monthly averages of temperature and precipitation
[2]Several terminologies are used by different authors to refer to traffic as a source, but for consistency in this paper, local traffic may mean only vehicle exhaust or may include resuspended road dust and other non-exhaust emissions; secondary aerosol as secondary ammonium sulphate and secondary ammonium nitrate from new particle formation; and long-range transport from airport, harbour and industrial emissions as well as regional sources.
[3]EU directives on emission standards and the introduction date, applied to all European cities – for light duty vehicles: Euro 2 (January 1996), Euro 3 (January 2000), Euro 4 (January 2005), Euro 5 (September 2009) and Euro 6 (September 2014); for heavy duty vehicles: Euro II (October 1996), Euro III (October 2000), Euro IV (October 2005), Euro V (October 2008) and Euro VI (January 2013); industrial emission (January 2011)

Concentration trends – time series analysis

Monthly mean concentrations of both $PM_{2.5}$ and PNC were used for the analysis for all cities. Meteorological parameters considered were monthly mean temperature (°C), monthly total precipitation (mm) and monthly mean wind speed ($m.s^{-1}$), all of which significantly affect PM concentrations (Barmpadimos et al., 2012) and contribute to the process of dilution, removal and recirculation of pollutants. If obtained data were of higher resolution (e.g. hourly or daily), to compute for the monthly mean, at least 50% of the daily concentrations must be available. Monthly time resolution was preferred over hourly or daily to account for the seasonality of the data. Time series analysis by fitting LOESS (locally estimated scatterplot smoothing) and GAM (generalised additive model) were applied to determine trends in $PM_{2.5}$ and PNC as well as the effects of the above-mentioned meteorological parameters on the concentration. Time series analysis techniques are used to determine correlation structure, understand the underlying cyclic content as to how the data evolve in time, and develop models that can forecast future trends (Woodward, 2012). This method has long been applied to air quality data, but usually in relation to epidemiology (Gouveia & Fletcher, 2000; Sagiv et al., 2005; Schwartz & Marcus, 1990). All data analysis and visualisation were done in R statistical software (RStudio Team, 2016).

General trends of PM concentration

The Mann-Kendall test for trend detection and the Sen's slope test for the magnitude of the trend were used to analyse the data. In particular, the correlated seasonal Mann-Kendall test, a non-parametric test that takes into account the seasonality of the data and is preferred when data are correlated, was applied using the *csmk.test* function of the *trend* package (Pohlert, 2018). The Mann-Kendall test measures the degree to which a trend is monotonic (a gradual change over time that is consistent in direction), thus, a p-value < 0.05 is strong statistical evidence that a monotonic trend exists. However, even if the trend was not monotonic, quantifying the rate of change is still important. Thus, the seasonal Sen's slope test using the *sea.sens.slope* function from the same package, which also considers the seasonality of the data, was applied. The Sen's slope is the median of a set of calculated linear slopes. Both *csmk.test* and *sea.sens.slope* functions in the R software are currently not capable of handling missing data; hence, the *tsclean* function (which identifies and replaces outliers and missing values in a time series) of the *forecast* package (Hyndman et al., 2018) was used to interpolate missing values in the data set.

Trends may occur in two ways: a monotonic trend or a step trend (an abrupt shift at a specific point in time). Thus, determining the linear rate of change (slope) may not completely capture the trend and is only appropriate if the change is gradual. In a time series, the underlying trend may not be apparent by simply plotting the data, because a particular repetitive pattern may emerge. Further, aside from the trend component, a time series may contain the seasonality component. Thus, in this study, the *stl* (seasonal decomposition of time series by LOESS) function from the *stats* package (R Core Team, 2018) that differentiates the time series data into seasonal, trend and irregular components was applied to obtain the long-term trend of the $PM_{2.5}$, PNC and meteorological parameters. LOESS, also known as local polynomial regression, is a non-parametric method for fitting a curve with more relaxed linearity assumptions compared with conventional regression. Weighted least squares is used to fit a smooth curve through points in a scatter plot, giving more weight to points near the point whose response is being estimated. The *tsclean* function was also applied prior to *stl*. The use of *tsclean* can potentially make the slope flatter in long periods of missing data, but precisely because of this, no inference can be made during this particular period.

To visualise the concentration changes in $PM_{2.5}$ and PNC over time as well as the evolution of the meteorology, the trend component was overlaid in the time series data for comparison. Then, to determine if the trend component after the LOESS decomposition had adequately captured the information in the $PM_{2.5}$ and PNC data, the mean absolute percentage error (MAPE) was computed using the *Measures of Accuracy* function of the *DescTools* package (Signorell et al., 2019). MAPE is a measure of error, thus high values suggest a bad fit, and subtracting this statistical measure from 100 gives the percentage accuracy of the model. MAPE is commonly used to evaluate the performance of obtained regression models, and is applied to air quality analysis (Cheng et al., 2014; Liu et al., 2015). Further, if the trend line has a good fit, the residuals (i.e. the difference between the observed and the predicted values): (1) are uncorrelated, (2) have constant variance, (3) are normally distributed, and (4) have zero mean. Thus, the *checkresiduals* function, also from the *forecast* package, was used to evaluate the residuals of the seasonally adjusted trend line.

Relationship between meteorology and PM concentration

Because seasonality was eliminated before establishing the trend, it was vital to determine how PM concentrations would have responded to changes in meteorological conditions. A GAM was fitted to the monthly $PM_{2.5}$ and PNC data with monthly mean temperature, total precipitation, and mean wind speed as additive predictors together with time. GAM is the combination of the additive model and the generalised linear model that uses non-parametric functions obtained from a scatterplot smoother, and then sums up these smooth functions instead of the linear combination of the effects of the individual predictors. The *gam* function from the *gam* package (Hastie, 2018) was applied using the LOESS smooth term in the GAM formula. To support the results of the correlation and significance between $PM_{2.5}$ and the meteorological parameters, and then PNC and the meteorological parameters, cross-correlation was done using the *ccf* function of the *tseries* package (Trapletti, 2005). Cross-correlation is used to determine whether one time series is affected by the other given a number of lags.

Impacts of changing climates

The long-term weather pattern in a particular area defines its climate, and climate categories are based on average temperature and precipitation then the existing climate system in the area dictates the prevailing winds (the heat from the sun creates the circulation). Climate variability, on the other hand, is defined by the short-term changes in climate patterns caused by factors such as the El Niño Southern Oscillation (ENSO). Thus, to analyse effects of the changing climate on $PM_{2.5}$ and PNC, the trend line of the meteorological parameters produced by the *stl* decomposition was analysed by applying the Buishand range test for change-point detection using the *br.test* function of the *trend* package. The Buishand range test is a non-parametric homogeneity test with a null hypothesis that there is no change-point and the *p*-value is estimated with a Monte Carlo simulation (20,000 replicates). The change-point detection analysis was done to assess where significant change had occurred in the time series, therefore identifying if the relevant meteorological parameters had changed over time (Jaiswal et al., 2015). If a significant change-point was detected, it was examined if such a meteorological change coincided with a change in PM concentrations to evaluate the impact of climate or the long-term trends in meteorology.

Effects of mitigation

To investigate the impact of the modifications in anthropogenic activities and PM sources on the $PM_{2.5}$ and PNC, which can either be a gradual decrease or an abrupt change, the Buishand range test was also used. Subgroups of the data were made to identify more change points. Initial analysis covered the whole period (P_T) of the time series per city, and then the data were broken up into shorter periods, namely P_1 (covering the time series up to 2006), P_2 (covering the time series from 2007 to 2011) and P_3 (covering the time series from 2012 to 2016). The year 2006 was selected as the base year because the PM concentration guidelines by the World Health Organisation (WHO) were released in 2006, followed by a five-year interval. The detected change-points were then studied if change corresponded to a particular implemented regulatory control measure and variation in contributing sources.

Results and discussion

The amount of missing data in each city for the period covered in this study varied from 0–7.5% for $PM_{2.5}$, 7–11.0% for PNC except for the 30% in London-RS, 0–6.6% for temperature, 0–7.9% for precipitation except for the 45.5% in Augsburg, and 0–6.6% for wind speed. The PNC data for Brisbane, on the other hand, were only from 1998 to 2000 and 2011 to 2015. Despite this limitation in available data, we could still use them to derive important information about long-term variations in PM concentration and meteorological parameters that could signify the impact of changes.

Concentration trends

General trends in PM concentration

The Mann-Kendall test detected that at a level of significance of 0.05, the $PM_{2.5}$ in all cities except London had a monotonic trend, and all had a negative slope except Brisbane (+0.1 $\mu g.m^{-3}.yr^{-1}$). The magnitude of reduction was greatest in Augsburg (-0.6 $\mu g.m^{-3}.yr^{-1}$), while Helsinki (-0.2 $\mu g.m^{-3}.yr^{-1}$) had the lowest of those that are significant ($p < 0.05$). London (-0.03 $\mu g.m^{-3}.yr^{-1}$) had the smallest magnitude of reduction, but no monotonic trend existed. The PNC had a significant negative monotonic trend for all cities except in Brisbane (+53 $particles.cm^{-3}.yr^{-1}$), which was positive and not monotonic. The greatest concentration reduction was in London-RS (-3.8 x 10^3 $particles.cm^{-3}.yr^{-1}$) and the lowest was in Rochester (-4.0 x 10^2 $particles.cm^{-3}.yr^{-1}$). Results are presented in Table S2.

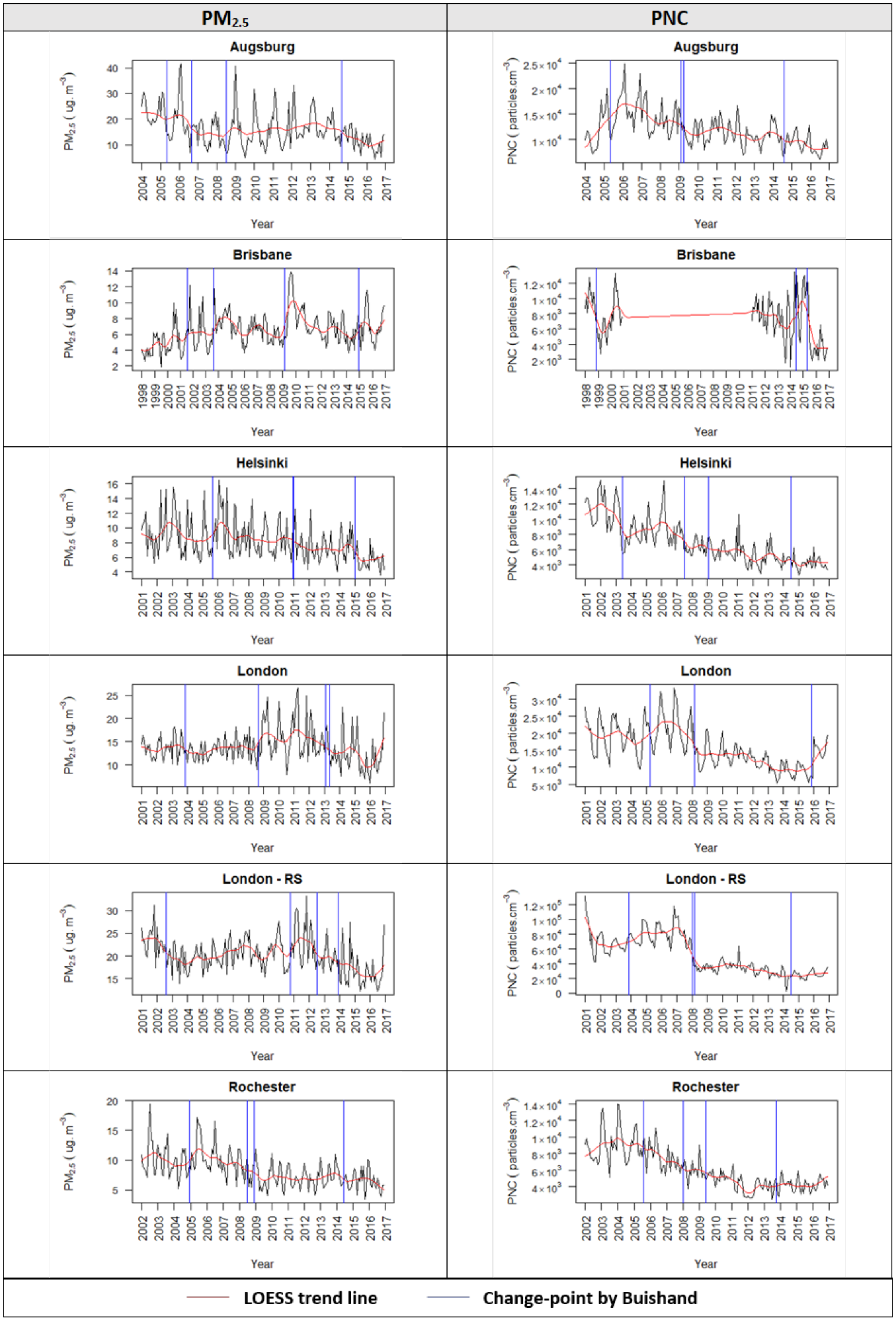

Figure 1. Fitted trend of the monthly $PM_{2.5}$ ($\mu g.m^{-3}$) and PNC ($particles.cm^{-3}$) using LOESS (red line) with the change-points detected by the Buishand range test (blue line).

The increasing trend observed in Brisbane for $PM_{2.5}$ may be attributed to industrialisation as the location of the station is zoned for industrial land use. Increases in vehicle numbers over time and the changing conditions being more favourable to particle formation have contributed to PNC, discussed in the subsequent section. In the case of London, Font and Fuller (2016) has reported the effectiveness of exhaust emission abatement policies therefore reducing PNC especially at the roadside, while $PM_{2.5}$ is influenced more by regional transport, hence improvement is limited. The small decline in PNC for Rochester is a combination effect of the increased traffic while simultaneously implementing emission reduction strategies (Masiol et al., 2018) since the station is in proximity to major roads with ~230,000 vehicles.d^{-1}. Despite population increases and economic growth, a decreasing trend in PM concentrations due to pollution control has been observed in most cities around the world (Cusack et al., 2012; Lurmann et al., 2015).

Further analysis showed that the two metrics behaved differently over time (i.e. significant changes in concentrations occurred at different times and at varying magnitudes) and that the reduction in concentration was more evident in PNC than in $PM_{2.5}$. Figure 1 presents the generated trend line from the seasonally-adjusted data set. More discussion about LOESS and the *stl* function is given in the supplementary material. Visual observation showed that $PM_{2.5}$ decreased to some extent in some cities, but this was not as evident as in the PNC for all cities. It can also be observed that the years in which the reduction in concentration occurred do not coincide for $PM_{2.5}$ and PNC. The difference in trend was further confirmed by the Buishand range test that detected several change-points, but none for $PM_{2.5}$ and PNC occurred at the same time. These change-points will be discussed subsequently in the sections on change in climate conditions and effects of mitigation. Factors responsible for the variation are discussed below.

Relationship between meteorology and PM concentration

Figure 2 shows how the ambient temperature, amount of precipitation and prevailing wind speed differed among the investigated cities. Helsinki is the coldest among the cities under investigation, while Rochester has the most varied monthly mean temperature. Brisbane is the warmest and has the narrowest temperature range among the cities; however, the amount of precipitation is very varied. Rochester has the highest monthly mean total precipitation, while Augsburg has the lowest monthly mean total precipitation and the weakest wind.

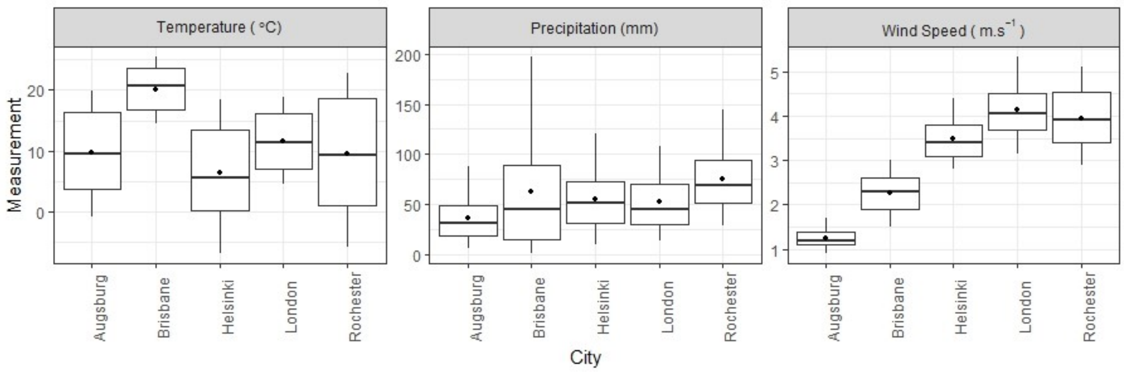

Figure 2. Boxplot with the mean (black dot), median, interquartile range (IQR), 5th percentile, and 95th percentile of the monthly mean temperature (°C), total precipitation (mm) and mean wind speed ($m.s^{-1}$) in the investigated cities.

Meteorology is one major factor in concentration variation at shorter time scales, with the diurnal and seasonal cycle most often pre-determining the occurrence of peak concentrations. The main effect of temperature, precipitation and wind speed as a predictor of $PM_{2.5}$ and PNC is indicated by the fitted GAMs (Figures S3 and S4). An autoregressive integrated moving average (ARIMA) model was also utilised to determine the effects of the meteorological parameters to the PM concentrations and results are, further discussion given in the supplementary material. $PM_{2.5}$ and PNC are mostly negatively correlated with the meteorological factors tested, except for the temperature and PNC of Brisbane; when the predictors increase, the function (LOESS) of the predictors decreases, hence decreasing the response variables ($PM_{2.5}$ and PNC). The correlation of temperature with $PM_{2.5}$ and PNC in Helsinki and Rochester are similar; for both metrics at temperatures below zero, the curve has a steeper downward slope, becomes more stable at higher temperature, and then at about 15 °C, the correlation becomes positive for $PM_{2.5}$ but not for PNC. The precipitation in Augsburg and London has a similar effect; the observed decrease in $PM_{2.5}$ and PNC (i.e. more evident for $PM_{2.5}$) is more pronounced when total precipitation is below 40 mm, and then becomes more stable. On the contrary, the PNC at London-RS, develops a positive correlation with increasing precipitation. Increasing wind speed has more effect in reducing $PM_{2.5}$ than PNC in cities except in the case of Brisbane, which showed the opposite influence of wind speed for $PM_{2.5}$ and PNC.

A negative correlation between particle concentrations ($PM_{2.5}$ and PNC) and the meteorological parameters (ambient temperature, amount of precipitation, and prevailing wind speed) suggests that reduced temperature promotes particle formation, high precipitation causes a washout effect, and weak wind speed concentrates particles and precursors due to stagnation (Zhang et al., 2016). However, statistically based on the GAMs (Figures S3 and

S4), precipitation and wind speed are significant to $PM_{2.5}$ while PNC is more affected by temperature. The dilution effect of increasing wind speed that affects particles >30 nm in size (Charron & Harrison, 2003) provides further support for the significance of wind speed to $PM_{2.5}$ rather than PNC, since larger particles have more significant mass. Thus, the impact of regional and long-range transported pollutants is important to $PM_{2.5}$ concentrations rather than to PNC, which is always dominated by smaller particles (and originate mainly from local combustion sources, particularly traffic), especially in urban areas. Similarly, the cleansing effectiveness of precipitation is greater to particles with size > 1 µm (Nicolás et al., 2009). Low temperature can result in the cooling of air masses causing stagnation, thus increasing pollution concentration (Hussein et al., 2006) and then enhancing nucleation due to the presence of high levels of precursors (Ripamonti et al., 2013); this increases PNC with less impact on $PM_{2.5}$ due to the lower mass of the formed particles. These results further demonstrate that the factors affecting the two metrics are different as those reported by De Hartog et al. (2005) that particle mass and number "are two separate indicators of airborne particulate matter" and by de Jesus et al. (2019) that they are not representative of each other.

The conditions in Brisbane, where wind speed and temperature were not statistically significant to $PM_{2.5}$ and PNC, respectively, but were positively correlated, can be attributed to its humid subtropical climate, with mean temperatures of above 20 °C and lower wind speed. The positive correlation of $PM_{2.5}$ with wind speed happens when the incoming air mass contains more air pollutants and the increased rate of nucleation and particle growth during relatively windy and warm days increases PNC. Long-range transport from distant sources (e.g. Brisbane airport and Port of Brisbane) has been observed in Brisbane (Rahman et al., 2017); aged particles contribute to $PM_{2.5}$ while additional precursors during a high insolation day favour particle formation elevating PNC (Shi et al., 2001). Thus, the prevailing winds must be carrying clean air masses for the dilution effect of winds to occur. Further, the positive correlation between PNC and precipitation at London-RS can be associated with the prevailing south-westerly winds during heavy rains. The station is located on the southern side of the road and. due to the vortex in the canyon, PNC concentrations are enhanced when winds have a southerly component (Harrison et al., 2019). The effect of precipitation on $PM_{2.5}$ and PNC as reported by Ikeuchi et al. (2015) and Zhang et al. (2016), respectively, is more on the influence of precipitation pattern (i.e. precipitation duration and precipitation occurrence) rather than the precipitation intensity. This could explain the observation in Rochester that despite the high monthly total precipitation, it had no significant effect on any of the two metrics.

Impacts of changing climates

As observed from the fitted LOESS (Figure 3), the monthly mean temperature (±2 °C) was more stable compared with total precipitation (±50 mm except for Augsburg with only ±5 mm and the extreme rainfall in Brisbane at the end of 2010) and mean wind speed (±1 $m.s^{-1}$), which were more varied over time. The results of the Mann-Kendall test confirmed that the meteorological parameters had no monotonic trend except for the wind speed of Augsburg, Brisbane and Rochester (Table S3). Additionally, the magnitude of change from the Sen's slope ranged from 0–0.07 $°C.yr^{-1}$ for temperature (positive for all cities), 0.02–1.43 $mm.yr^{-1}$ for precipitation (Helsinki is the only positive) and 0.01–0.03 $m.s^{-1}.yr^{-1}$ for wind speed (negative for both Brisbane and Rochester). Further, the Buishand range test detected change-points in the ambient temperature, precipitation and wind speed for the duration considered in this study (dates of shift shown in Table S8) and the trends agreed with the obtained Sen's slopes. Based on the fitted LOESS (Figure 3), except for London (~ -1 °C), there was a slight increase in the mean monthly temperature for the cities after the change-point. The trend for precipitation was in contrast with ambient temperature; the amount of rainfall decreased in all cities after the change-point except for Helsinki (~ +10 mm). For wind speed, the shift was varied; a minute increase for Augsburg, Helsinki and London but a slight decrease in Brisbane and Rochester.

The change-points in the meteorological parameters (Tables S8), did not match any of the change-points for $PM_{2.5}$ (Table S9, versus precipitation and wind speed) and PNC (Table S10, versus temperature) except for the $PM_{2.5}$ and wind speed of Rochester. However, trends of $PM_{2.5}$ and PNC during the change-points were visually inspected using Figures 1 and 3 then summarised in Table S11. For Rochester, wind speed and $PM_{2.5}$ declined in July 2008. Although wind speed is significant to $PM_{2.5}$ based on GAM and ARIMA, they are negatively correlated. Thus, this observed decline in concentration is not due to a weakening wind speed in Rochester. Similarly, the observations in $PM_{2.5}$ trends and wind speed in London (increase) and Brisbane (decrease) were as expected based on GAM. On the contrary, the change in wind speeds for Augsburg (increase) and Helsinki (increase) may have had effects on PM concentrations. Additionally, since precipitation is also significant to $PM_{2.5}$, its increase in Helsinki and decrease in Brisbane in 2003 and 2012, respectively, may have influenced the reduction in $PM_{2.5}$, while having no effect in other cities.

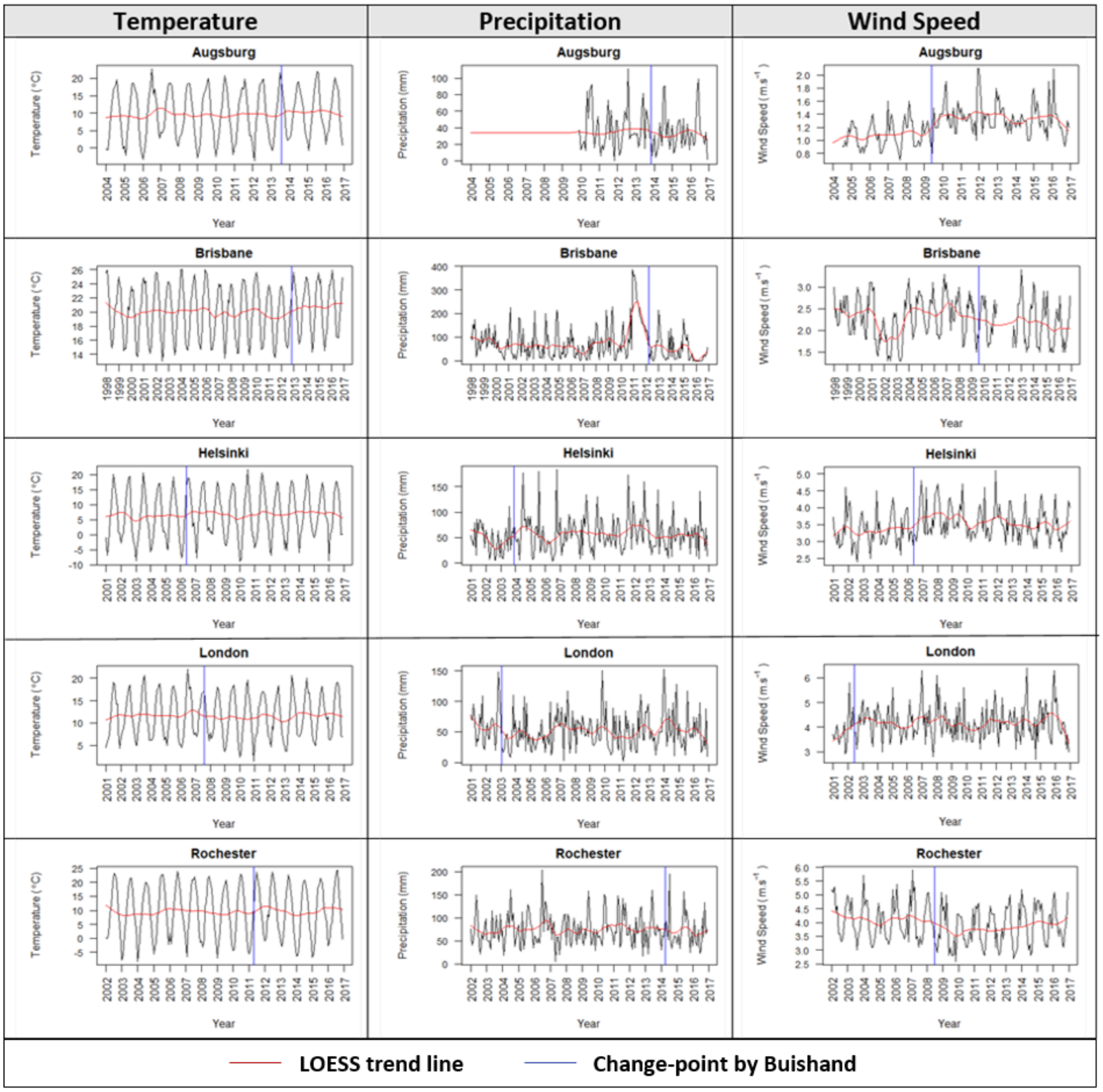

Figure 3. Fitted trend of the monthly mean temperature (°C), total precipitation (mm) and mean wind speed (m.s^{-1}) in the investigated cities using LOESS (red line) with the change-point detected by the Buishand range test (blue line).

For the effect of temperature as a significant factor on PNC in Helsinki and Rochester, no change-points were detected in PNC in 2006 and 2011, respectively (Figure 3), but there was an observable decrease in PNC at the same time the temperature increased. Hence, since temperature is significantly negatively correlated with PNC, this change in concentration may be partly attributed to the change in temperature in Helsinki and Rochester. Further, for London, although a change occurred in 2007 for both temperature and PNC, the observed trends are of the same direction, contrary to the results of GAM and ARIMA. Therefore, long term changes in meteorological parameters could impact PM concentrations; an increase in temperature in cold cities (Helsinki and Rochester) may slow down particle formation, a decrease in precipitation in a relatively wet city (Brisbane) or an increase in a relatively dry city (Helsinki) may minimise or promote wet deposition, respectively, and an increase in wind speed in less windy cities (Augsburg and Helsinki) when incoming air masses are cleaner supports particle dilution.

One important driver of the year-to-year variability in climate particularly in the Pacific region is the ENSO, which impacts wind circulation, precipitation, and temperature. The shifting from the warm phase (El Niño) to the cool phase (La Niña) happens every two to seven years and triggers a very predictable disruption affecting global climate. In this study's timeframe, two very strong El Niño events (1997–1998 and 2015–2016) and three strong La Niña events (1999–2000, 2007–2008, and 2010–2011) occurred. However, these particular events did not have observable effects in the meteorological parameters (Figure 3) of the cities under study except for the 2011 extreme precipitation in Brisbane, being in proximity to Pacific Ocean; a corresponding decreasing concentration trend in $PM_{2.5}$ can be observed. In contrast, another phenomenon called the Indian Ocean Dipole (IOD) played an important role in the Australian drought in 2009 (i.e. a positive IOD dominated the weak El Niño) causing bushfires and dust storms (Cai et al., 2009), and resulting in an abnormally high $PM_{2.5}$ in Brisbane.

The North Atlantic Oscillation (NAO) is another climate fluctuation but with effects that are more local than global like the ENSO. NAO strongly affects the winter weather in Europe and North America particularly precipitation (Dai et al., 1997). When a positive NAO is accompanied by an El Niño, European winters tend to be wetter and less severe; during a La Niña, formation of Atlantic hurricanes is favoured but the position of the Azores high influences the direction (Huang et al., 1998; Nakamura et al., 2015; Oshika et al., 2015). The winter of 2009–2010 was linked with the presence of negative NAO during an El Niño affecting the eastern North America and northern Europe (Seager et al., 2010). Rochester and Helsinki received less precipitation during this time based on the precipitation data (Figure 3), and a corresponding increase in $PM_{2.5}$ with no such effect in PNC can be observed in Figure 1. London, in contrast, received an unusually high amount of precipitation, and a decrease in $PM_{2.5}$ in the urban background can be seen.

Effects of mitigation

Table 2 lists the years detected by the Buishand range test (the change-point dates are in Tables S8 and S9) and the changes in concentration that occurred in the $PM_{2.5}$ and PNC of every city. When the change-points overlaid in the fitted LOESS (Figure 1) were examined, some points were positioned along a step-change (i.e. a rapid decrease within a year), but some were contained within periods of gradual change. The probable cause was identified by studying the existing conditions prior to, during, and after each change-point, ensuring that the lag effects of policy were considered. Step-changes occurred for $PM_{2.5}$ in 2006 – Augsburg (decrease), in 2009 – Brisbane (increase), in 2005 – Helsinki (increase), 2008 – London (increase) and 2002 – London-RS (decrease). For PNC, they were in 2014 and 2015 for Brisbane (increase and decrease, respectively), in 2003 – Helsinki (decrease), and in 2007 – London and London-RS (both decrease). The rest of the change-points were part of a period with gradual changes. It can also be observed that at some points, the changes in $PM_{2.5}$ and PNC were not the same.

Mitigation is another important factor that affects ambient PM concentration, and causes either a step-change or a monotonic change. Control techniques that involve traffic management usually result in localised and gradual reductions, and those that modify the quality of emissions using cleaner technologies have varying results; control retrofits reduce $PM_{2.5}$ but not always PNC (Järvinen et al., 2019), natural gas as fuel may increase PNC with little or no clear reducing effect on $PM_{2.5}$ depending on operating conditions (Pirjola et al., 2016), and fuel with reduced sulphur content causes an abrupt reduction in PNC as observed in Helsinki and London. The application of traffic schemes has been reviewed in the following cities: in London, UK (Atkinson et al., 2009; Beevers et al., 2016), Delhi, India (Kumar et al., 2017), Dublin, Ireland (Tang et al., 2017), and Lanzhou, China (Zhao et al., 2014). Shifting to low emission vehicles and fuels was applied in India (Gurjar et al., 2016), Japan (Hasunuma et al., 2014) and California, USA (Kuwayama et al., 2013). Integrated emission control strategies and regulatory policies have been proven effective in reducing ambient PM concentrations despite population growth and the increased vehicle count in urban centres (Lurmann et al., 2015; Wu et al., 2017).

Table 2. Trends of $PM_{2.5}$ and PNC at the detected change-points and the probable cause of change in concentration in relation to modifications in emission sources.

Change-point	Trend at change-point		Probable cause
	$PM_{2.5}$	PNC	
Augsburg			
2005	Decrease	Increase	No particular attribution
2006	Decrease	Decrease	Reduced sulphur content in diesel and petrol
2008	Increase	None	Warm and calm year
2009	None	Decrease	No particular attribution
2014	Decrease	Decrease	Continued improvement from Euro 5 passenger cars uptake
Brisbane			
1998	Increase	Decrease	No particular attribution
2001	Increase	N/A	Start of extreme drought in Australia causing bushfires and dust storms, with very low wind speed
2003	Increase	N/A	Drought continued, with low wind speed
2009	Increase	N/A	Dust storm (September 2009) due to extreme drought
2014	Increase	Increase	Relatively warm and dry year and increase in vehicle count
2015	Decrease	Decrease	Increased in precipitation; monitoring of $PM_{2.5}$ for compliance started
Helsinki			
2003	Decrease	Decrease	Reduced sulphur content in diesel and petrol
2005	Increase	None	Long-range transported particles during regional wildfires and agricultural burns causing high PM in 2006
2007	None	Decrease	No particular attribution
2009	None	Decrease	No particular attribution
2010	Decrease	None	Long-range transported particles during regional wildfires causing high PM in summer 2010
2014	Decrease	Decrease	Very low sulphur (max 0.1%) marine fuels in the Baltic Sea (IMO) and warm winter in 2015 causing low PM
London			
2003	Decrease	Decrease	No particular attribution
2005	Increase	Increase	Calm weather and Smog episodes
2007	None	Decrease	Reduced sulphur content in diesel and petrol
2008	Increase	None	Calm and dry 2009
2013	Increase	None	No particular attribution
2016	Increase	Increase	Warm and dry year
London - RS			
2002	Decrease	None	Reduction of traffic volume due to implementation of bus lanes in 2001
2003	Decrease	Increase	No particular attribution
2007	Decrease	Decrease	Reduced sulphur content in diesel and petrol
2010	Increase	None	No particular attribution
2012	Decrease	Decrease	Low emissions zone with Euro IV heavy duty trucks and Euro 5 passenger cars
2013	Decrease	Decrease	Continued improvement from Euro 5 passenger cars uptake
2014	Decrease	Increase	No particular attribution
Rochester			
2004	Increase	Decrease	No particular attribution
2005	Decrease	Decrease	Car fleet change; implementation of emission standards for on-road vehicles, use of after treatment technologies.
2008	Decrease	Decrease	Reduced economic activity due to recession (2007 – 2009); new heavy-duty diesel vehicles equipped with catalytic regenerative traps (CRT) and reduced sulphur content in diesel (2007)
2009	Decrease	Decrease	Reduced use of coal for power generation
2013	Increase	Increase	Increased vehicular traffic (recovery from 2008 recession)
2014	Decrease	Decrease	Reduced sulphur content in diesel for non-road, locomotive and marine

The extent of changes in PM concentration is very much dependent on the type of fleet and volume of vehicles if traffic is a main contributor, and on other existing contributors such as energy generation sources. The Euro standards, for instance, apply only to new vehicles, which then take time to penetrate the vehicle fleet, and hence any improvements take place over a number of years. Squizzato et al. (2018b) and Masiol et al. (2018), on the contrary, reported in separate studies that the decrease in $PM_{2.5}$ and PNC trends in Rochester that accounted for a significant change in concentration could be attributed to a shift in fuel use for power generation due to price changes and lowered activity in 2007–2009 because of the economic recession on top of policy initiatives. In New York City when emissions from burning heating oil were being regulated, Kheirbek et al. (2014) calculated that a complete phase-out scenario of high sulphur heating fuel could reduce $PM_{2.5}$ by about 0.71 $\mu g.m^{-3}$. Additionally, precursors originating from other sources besides local traffic (e.g. harbours and airports, industrial, agricultural, forest and marine) via long-range transport are also substantial in increasing PM concentration (including both $PM_{2.5}$ and PNC) in urban air (Aranda et al., 2015; Donateo et al., 2014; Hasheminassab et al., 2014; Sarkar et al., 2019; Stettler et al., 2011). In a study done by Venkataraman et al. (2018) about different emission pathways involving the source sectors in India, significant reduction in $PM_{2.5}$ can be achieve through aggressive regulation of biomass-fuelled technologies, industrial coal-burning and agricultural burning.

Conclusions

The long-term trends in $PM_{2.5}$ and PNC in five cities in Australia, Europe, and the United States were assessed with regard to the changing climates and regulatory policies. However, determining which of the factors affecting PM concentration took effect in a particular event is complicated because of the complex dynamics of pollution formation and transport. Both $PM_{2.5}$ and PNC declined in all cities except Brisbane for the course of the study, with a greater magnitude of reduction for PNC. In general, $PM_{2.5}$ and PNC were negatively correlated with temperature, precipitation and wind speed. Temperature is significant to PNC, while $PM_{2.5}$ is greatly affected by precipitation and wind speed. The long-term changes observed in the meteorological conditions caused changes in $PM_{2.5}$ and PNC that were similar to the changes caused by seasonality, such as low particle formation at higher temperatures, higher rate of wet deposition during increased precipitation and dilution enhanced by strong winds (Fontes et al., 2017). Both $PM_{2.5}$ and PNC had a monotonic downward trend while long-term measurements of temperature, precipitation, and wind speed had no particular trend. Additionally, the increasing intensity and frequency of climate variabilities causing extreme events due to changing climates also influenced PM concentrations (Jeong et al., 2018; Markakis et al., 2016; Messori et al., 2018).

Given the complex interplay of available emission sources and mitigation strategies, then the influence of climate, controlling anthropogenic emissions through improved technology still has significant impacts in the concentrations of PM in urban ambient air. The same findings have been reported about emission management in terms of $PM_{2.5}$ in China (Vu et al., 2019) and PNC in Germany (Sun et al., 2019). The planning and implementing of urban air quality management will be more effective and efficient if both $PM_{2.5}$ and PNC are considered. As $PM_{2.5}$ and PNC were affected differently by the above-mentioned factors, a separate regulatory standard for particulate mass and number would be a positive step in abating negative health and environment effects. Additionally, a separate emission targets should be applied since the sources of $PM_{2.5}$ and PNC are different.

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Declaration of interests

 The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

☐The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

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Alma Lorelei de Jesus: Conceptualisation, Methodology, Software, Formal Analysis, Writing – Original Drafts, Visualisation
Helen Thompson: Methodology, Software, Writing – Review & Editing
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Supplementary Material

Long-term trend in $PM_{2.5}$ mass and number concentrations in the urban air: the impacts of mitigation and extreme events due to changing climates

Site description

Augsburg, Germany

An urban district in southeast Germany, Augsburg is the third largest city in the state of Bavaria with Munich as the largest and the capital city. In 2013, the population density is 1,896 persons.km^{-2} but the number of inhabitants in the built-up area is much higher since the city has a large forest area (2,420 km^{-2} if excluding Districts XV and XVI). Augsburg is situated beside the Lech Valley and the Wertach River on the west side. Augsburg has warm summers and no dry season, the wetter season is from May – August while the driest month is October. Though the summer (June- Aug) mean temperature is still comfortable at about 20°C, winter (November – February) is chilly and windy with mean temperature of 6°C but can go as low as -3°C in February. In spring up to autumn, the wind becomes calmer but prevailing wind is from the west throughout the year. The Fachhochschule station is located in Innenstadt and the nearest main road from the station is 152 m with 19,503 vehicles.d^{-1} (2008). Though there is a nearby road (110 m away) but the estimated daily vehicles is only 500.

Brisbane, Australia

Brisbane is located in the South East Queensland region, which is mountainous with urban centres mostly at the coast. Brisbane City is the state capital and its CBD lies along the Brisbane River that extends in all direction within the floodplain. Brisbane's climate is described as warm humid summer and mild winter without extreme seasonal variability; the four seasons are not distinct but changes in temperature can be experienced. The hottest month is January, the wettest is February and the coldest is July. The Wet/Dry pattern is more appropriate in describing Brisbane's climate, where the wet season is between November and March while the dry season is from April to October. The PNC measurement was done at the Gardens Point Campus, Queensland University of Technology. This area is where the air quality monitoring for Brisbane CBD is done and is <100 m away from the South East Freeway (aka M3 Pacific Motorway). The Rocklea station, on the other hand, is surrounded by light industry and residential areas.

Helsinki, Finland

A city along the coast of the Baltic Sea with a fairly flat terrain, Helsinki is the centre of Finland's cultural, educational, financial and political activities. Like any other city, vehicular traffic is the major source of air pollutants in the metropolitan area, the inner city with busy roads and streets lined with tall buildings are the most challenging places and springtime is the worst for fine particles when road surfaces start to dry out. During winter (December – March), the days only last for almost 6 hours, with the average temperature at around -4°C in January and February but in summer (June- August), Helsinki enjoys the longest daylight for almost 19 hours. Thunderstorms occur during summer. The SMEAR III station, which monitors the urban environment, is located in the Kumpula Campus, University of Helsinki, beside the Finnish Meteorological Institute about 5 km northeast of the city centre. The Kallio station is located in the Kallio Sports Field to measure air quality of the residential areas in the inner city of Helsinki. The Kaisaniemi station (FMISID100971) is located 3.5 km distance from SMEAR III station.

London, United Kingdom

London, located in the south east of the island of Great Britain, is the capital city of England, a part of the United Kingdom. London is considered as one of the most important cities in the global economic system; with the busiest airspace and ports, has one of the longest metro system, and one of the largest bus network. The city is situated along the River Thames then surrounded by gently rolling hills. London has a temperate oceanic climate, which features cool summer but not so cold winter; an average temperature of 24°C in the warmest month and above 0°C in the coldest month. Winter (January – February) is relatively damp and cloudy, with less chances of snowfall while summers (June- August) are mild but occasional heat waves are experienced. In general, the city of London is warmer compared to the suburbs and outskirts due to urban heat island effect. North Kensington station is located in the Sion Manning School surrounded mainly by a residential area with the nearest road in 5 m while Marylebone station is located in a street canyon and with A501, a 6-laned frequently congested road (~72,000 vehicles.d^{-1}) just 1 m away. North Kensington is 4km west of Marylebone. The Heathrow station (25 masl, ID 708) measures weather data for Greater London.

Rochester, United States of America

Rochester is in the north-eastern United States, situated along the southern shore of Lake Ontario and is the third largest city in the State of New York. Several hills and mounds as well as numerous streams and ponds can be found in Rochester, which were formed when the continental glacier reached standstill. The Genesee River, a tributary of Lake Ontario, traverses the city and giving it a fertile valley that lead to the development of a manufacturing hub that founded Rochester as one of America's boomtowns. Downtown Rochester, on the other hand, is lined with skyscrapers for both residential and office space. Rochester has cold, snowy winters and moderately humid summers with generally comfortable temperatures but with significant precipitation year round. The month with most sunshine is July while January has the most number of rainy days. Before April 2004, the data for PM were collected from a site situated within 1 km of downtown Rochester on the roof of the central fire headquarters, 100 m south of an inner loop road (~86,000 vehicles.d^{-1}). It was transferred to its present location (130 masl, USEPA Site code 36-055-1004) in a residential area ~300 m from the intersection of two major highways (I-490 and I-590) with an average traffic count of ~230,000 vehicles.d^{-1}. The Rochester Greater International Station (164 masl) is about 5 km southwest of the metropolitan area and 18 km to the north of Lake Ontario.

Table S1. Location and characteristics of the monitoring sites and the data used for the different cities

City Monitoring station/s	Coordinates	[1]Station type	Parameters measured	Period (years)	Instrument	
					$PM_{2.5}$	**PNC (size range)**
Augsburg, Germany						
Fachhochschule Station by the *Helmholtz Zentrum München*	48.36°N, 10.91°E	UB	$PM_{2.5}$, PNC, Temp., [3]Prec., WS	2004–2016	TEOM 1400A / FDMS 8500	CPC TSI 3025A (3 – 3000 nm)
Brisbane, Australia						
Rocklea Station by the *Department of Science, Information Technology and Innovation*	27.54°S, 152.99°E	UB	$PM_{2.5}$, Temp., Prec., WS	1998–2016	TEOM 1405-DF	---
Queensland University of Technology (QUT) by the *International Laboratory for Air Quality and Health*	27.48°S, 153.03°E	UB	PNC	1998–2000 2011–2015	---	CPC TSI 3787 (5 – 1000 nm)
Helsinki, Finland						
Kallio Station by the *Helsinki Region Environmental Services Authority*	60.19°N, 24.95°E	UB	$PM_{2.5}$,	2001–2016	[4]TEOM 1400AB and Eberline FH 62 I-R	---
SMEAR III Station by the *Department of Physical Sciences, Chemistry and Forest Ecology, University of Helsinki*	60.20°N, 24.96°E	UB	PNC	2001–2016	---	CPC TSI 3022 7nm size
Kaisaniemi Station by the *Finnish Meteorological Institute*	60.18°N, 24.94°E	NA	Temp., Prec., WS	2001–2016	---	---
London, United Kingdom						
(1) North Kensington Station by the *Automatic Urban and Rural Network* (AURN)	51.52°N, 0.21°W	UB	$PM_{2.5}$, PNC	2001–2016	TEOM Thermo 1400AB /FDMS 8500	CPC TSI 3022A (7 – 1000 nm)
(2) Marylebone Road Station by the *Automatic Urban and Rural Network* (AURN)	51.52°N, 0.15°W	[2]RS	$PM_{2.5}$, PNC	2001–2016	TEOM Thermo 1400AB /FDMS 8500	CPC TSI 3022A (7 – 1000 nm)
Heathrow Station by the *Met Office*	51.48°N, 0.45°W	NA	Temp., Prec., WS	2001–2016	---	---
Rochester, United States of America						
Rochester Station by the *New York State Department of Environmental Conservation*	46.16°N, 77.6°W	UB	$PM_{2.5}$, PNC	2002–2003	TEOM 1400A	CPC TSI 3010 (11 – 500 nm)
Rochester Station by the *New York State Department of Environmental Conservation*	43.17°N, 77.55°W	UB	$PM_{2.5}$, PNC	2004–2016	TEOM 1400A	CPC TSI 3010 (11 – 500 nm)
Rochester Greater International Station by the *National Centres for Environmental Information*	43.12°N, 77.68°W	NA	Temp., Prec., WS	2002–2016	---	---

[1]UB: urban background; RS: roadside station (classification for PM monitoring); NA: not applicable (for stations collecting only meteorological data)
[2]Since data were available for a roadside station; we opted to present the difference in trend between the two site categories
[3]Precipitation data for Augsburg is only from 2009
[4]Eberline was used from 2001–2006 and TEOM from 2007 – 2016. Methods are reliably comparable after using calibration functions (Waldén et al., 2010)

Trend Analysis

1. Trend Analysis using Mann-Kendall test and Sen's slope

Table S2. The obtained *p*-value from the Mann-Kendall test and the Sen's slope for $PM_{2.5}$ ($\mu g.m^{-3}.yr^{-1}$) and PNC ($particles.cm^{-3}.yr^{-1}$) for each city.

City	$PM_{2.5}$ ($\mu g.m^{-3}$)		PNC (particles.cm^{-3})	
	p-value	slope	*p*-value	slope
Augsburg	0.020	-0.6	0.004	-497
Brisbane	0.037	0.1	0.194	53
Helsinki	0.001	-0.2	<0.001	-436
London	0.536	-0.03	0.003	-752
London - RS	0.022	-0.3	<0.001	-3769
Rochester	<0.001	-0.3	<0.001	-397

Table S3. The obtained *p*-value from the Mann-Kendall test and the Sen's slope for temperature (°C.yr^{-1}), precipitation (mm.yr^{-1}), and wind speed ($m.s^{-1}.yr^{-1}$) for each city.

City	**Temperature** (°C)		**Precipitation** (mm)		**Wind Speed** ($m.s^{-1}$)	
	p-value	slope	*p*-value	slope	*p*-value	slope
Augsburg	0.096	0.07	0.540	-0.017	0.008	0.03
Brisbane	0.246	0.03	0.111	-1.429	0.049	-0.02
Helsinki	0.093	0.05	0.437	0.417	0.079	0.02
London	0.704	0	0.776	-0.134	0.274	0.01
Rochester	0.305	0.05	0.306	-0.408	0.040	-0.03

2. Trend Analysis using Locally Estimated Scatterplot Smoothing (LOESS)

Smoothing by LOESS is a non-parametric fitting that uses local regression wherein a line is fitted to the points that fall within a specified window. The points nearest to the centre of the window are given more weight (i.e. they have the greatest effect on the calculation of the regression line). As the points move further away from the regression line, the weighting is reduced. The regression process is repeated several times within the window then moving the window across the data to obtain the LOESS curve. Each point on the resulting LOESS curve is the intersection of the regression line and the centre of each window. In order to obtain the $PM_{2.5}$ and PNC trends, an additive time series decomposition by applying the LOESS smoothing was done. In an additive model, the seasonal variation remains constant and does not change with increasing time unlike the multiplicative model. Further discussion about decomposition by LOESS is given by Cleveland et al. (1990).

In the *stl* function, the *s.window* was set to 'periodic' where the seasonal component was computed by getting the mean values for each month. Seasonality in a time series is the regular and predictable pattern that recurs at a fixed interval of time, while trend is the overall direction of the data. The seasonally adjusted data is then LOESS-smoothed to determine the trend. After decomposition, the seasonal component corresponds to the variations in the data associated to calendar cycles; the trend component gives the overall pattern of the time series that are not seasonal and the remaining component of the time series that cannot be attributed to either seasonal or trend, often referred as the residual or error. The mean absolute percentage error (MAPE) of the LOESS trend line is presented in Table S4. The values ranged from 12.1 to 33.5% for $PM_{2.5}$ and from 15.3 to 23.9% for PNC, these values are comparable to the calculated MAPE values by other regression techniques applied to ambient particulate matter (Sajjadi et al., 2017). The residuals were also tested by getting the correlation and distribution. Checking the residuals is necessary to demonstrate the validity of the fitting of the LOESS. Figure S1 showed that the residuals were uncorrelated and were normally distributed.

Table S4. Mean absolute percentage error of the LOESS trend line for the monthly $PM_{2.5}$ and PNC.

City	**MAPE (%)**	
	$PM_{2.5}$	**PNC**
Augsburg	33.5	16.3
Brisbane	20.7	23.9
Helsinki	22.9	16.6
London	18.0	20.8
London - RS	12.1	19.0
Rochester	21.0	15.3

Four plots were generated for each PM2.5, PNC, temperature, precipitation and wind speed per city (Figure S2 a to f). The plot on top is the original data, next is the extracted periodic seasonal pattern, then the trend and the remaining components. The y-axis scale is placed alternatingly on each side and a scale bar on the right hand side of each graph is for relative comparison of the magnitude of each component. The decomposition showed different patterns for the seasonal component, not only among parameters, but even between the same parameters of different cities (e.g. precipitation data of Helsinki against London against Rochester). Only the seasonal component of temperature is comparable for all cities.

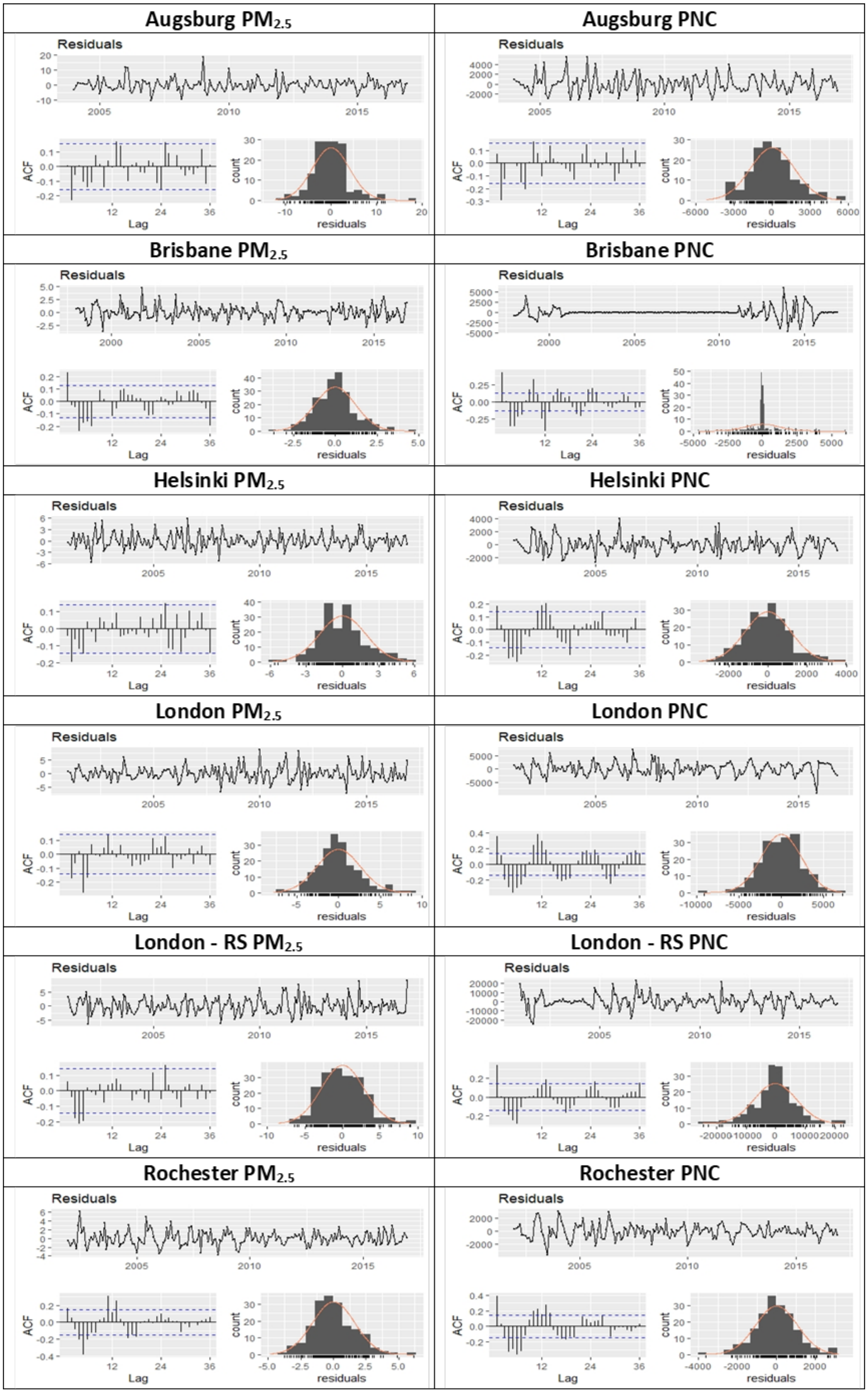

Figure S1. Time plot, ACF plot, and histogram of the residuals of the LOESS trend line for the monthly $PM_{2.5}$ and PNC.

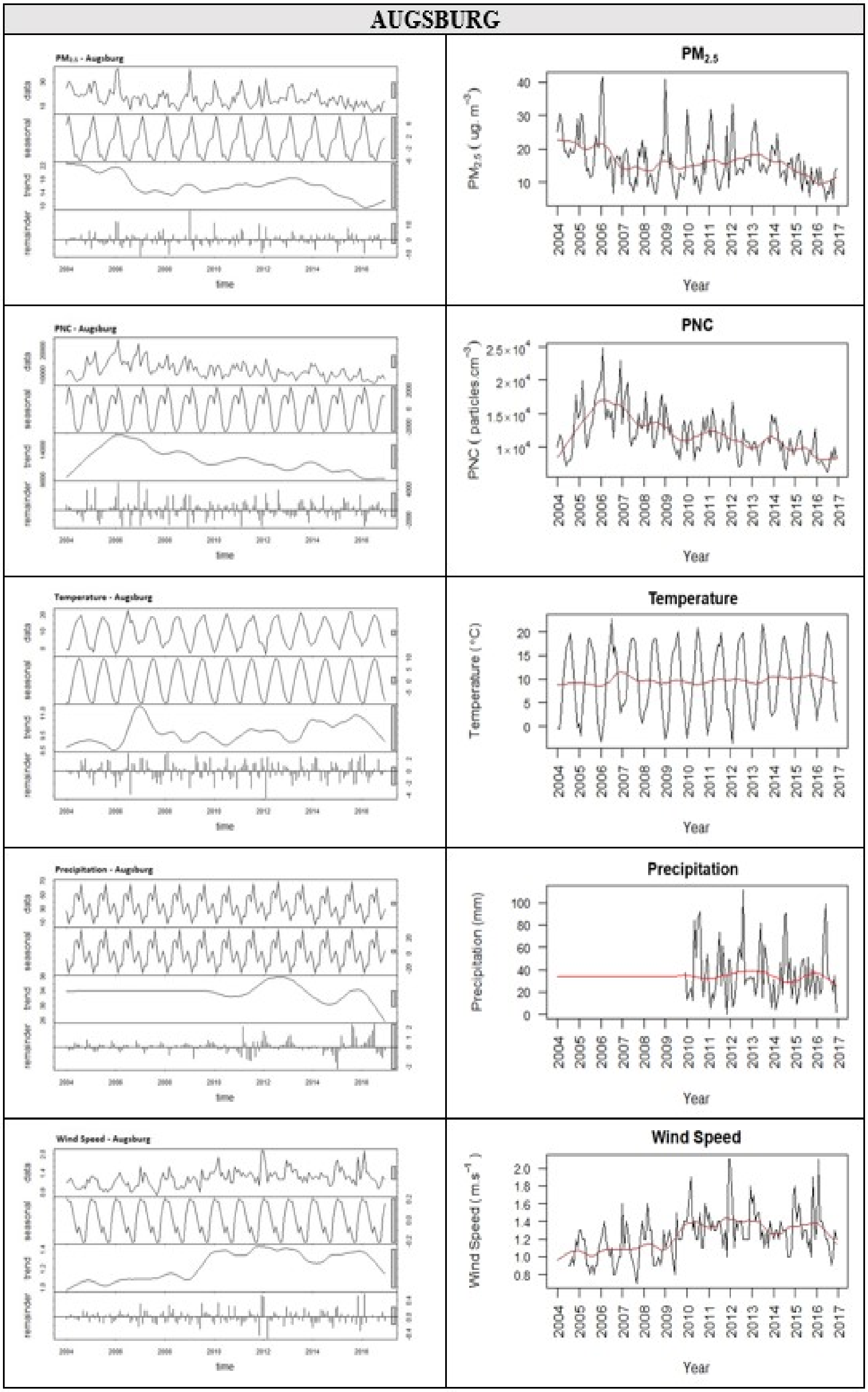

Figure S2a. Decomposition of the monthly $PM_{2.5}$ (μg.cm^{-3}), PNC (particles.cm^{-3}), mean temperature (°C), total precipitation (mm) and mean wind speed (m.s^{-1}) time series into seasonal, trend and stochastic (remainder) components using *stl* then the fitted trend for Augsburg.

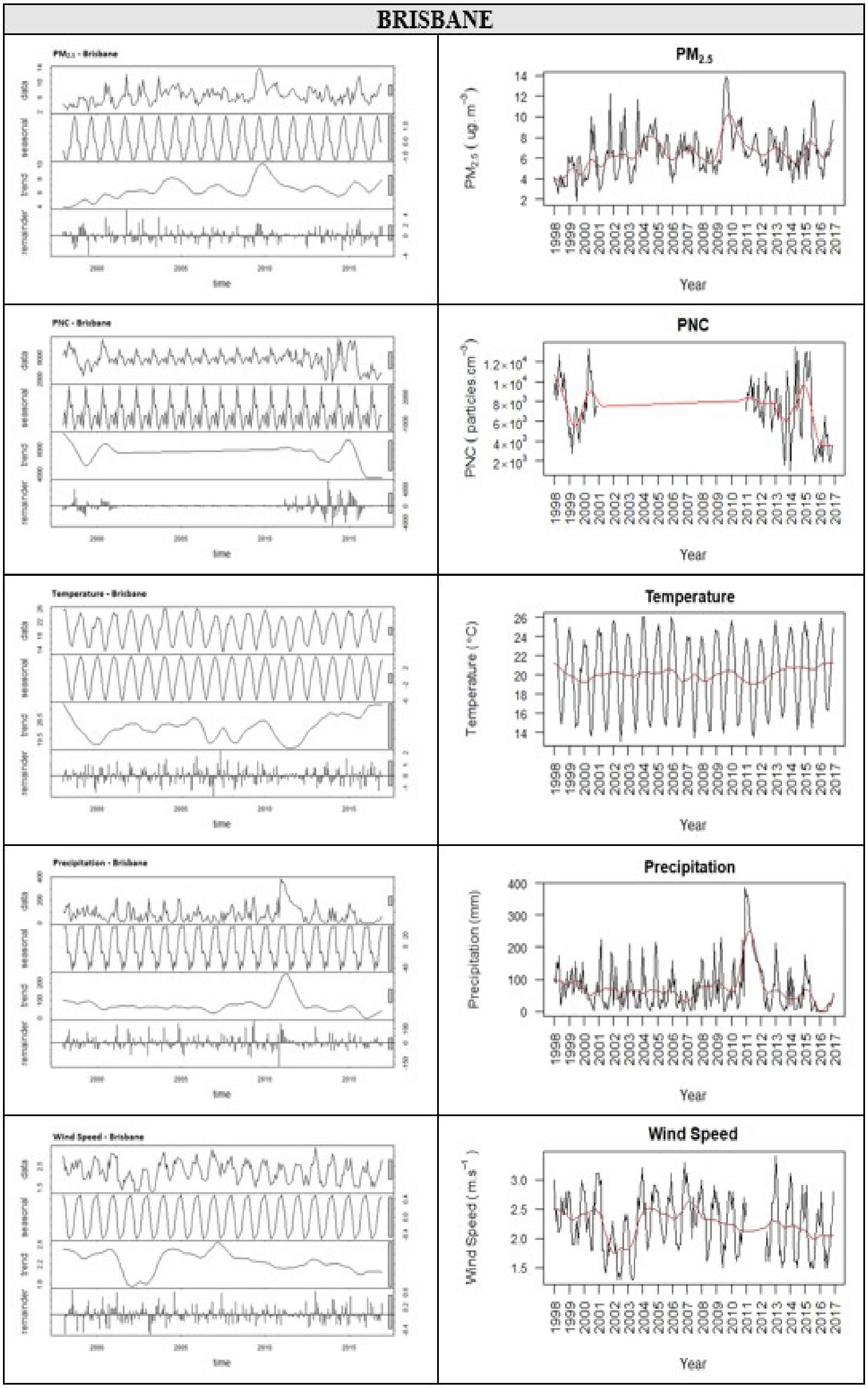

Figure S2b. Decomposition of the monthly $PM_{2.5}$ (µg.cm^{-3}), PNC (particles.cm^{-3}), mean temperature (°C), total precipitation (mm) and mean wind speed (m.s^{-1}) time series into seasonal, trend and stochastic (remainder) components using *stl* then the fitted trend for Brisbane.

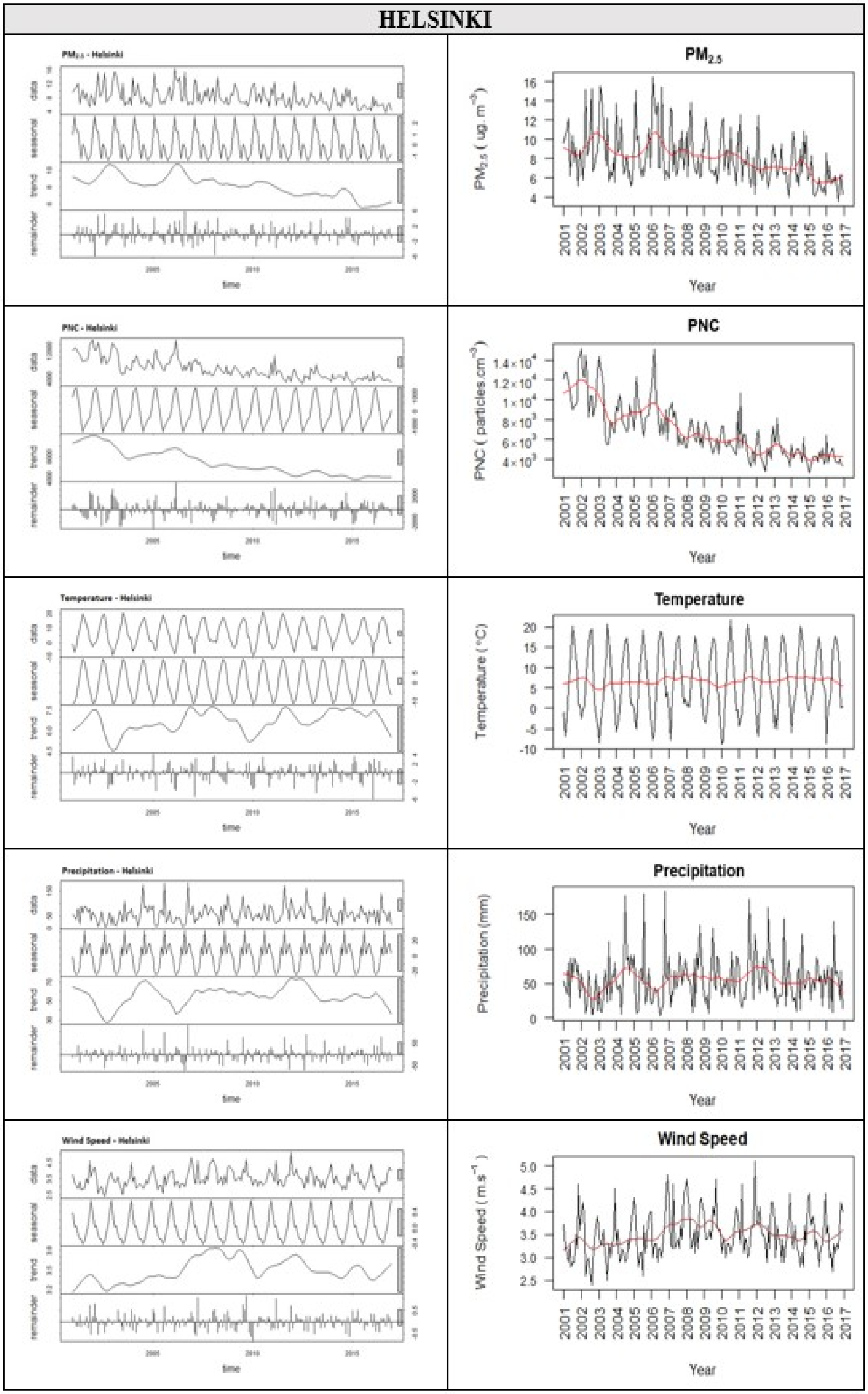

Figure S2c. Decomposition of the monthly $PM_{2.5}$ (μg.cm^{-3}), PNC (particles.cm^{-3}), mean temperature (°C), total precipitation (mm) and mean wind speed (m.s^{-1}) time series into seasonal, trend and stochastic (remainder) components using *stl* then the fitted trend for Helsinki.

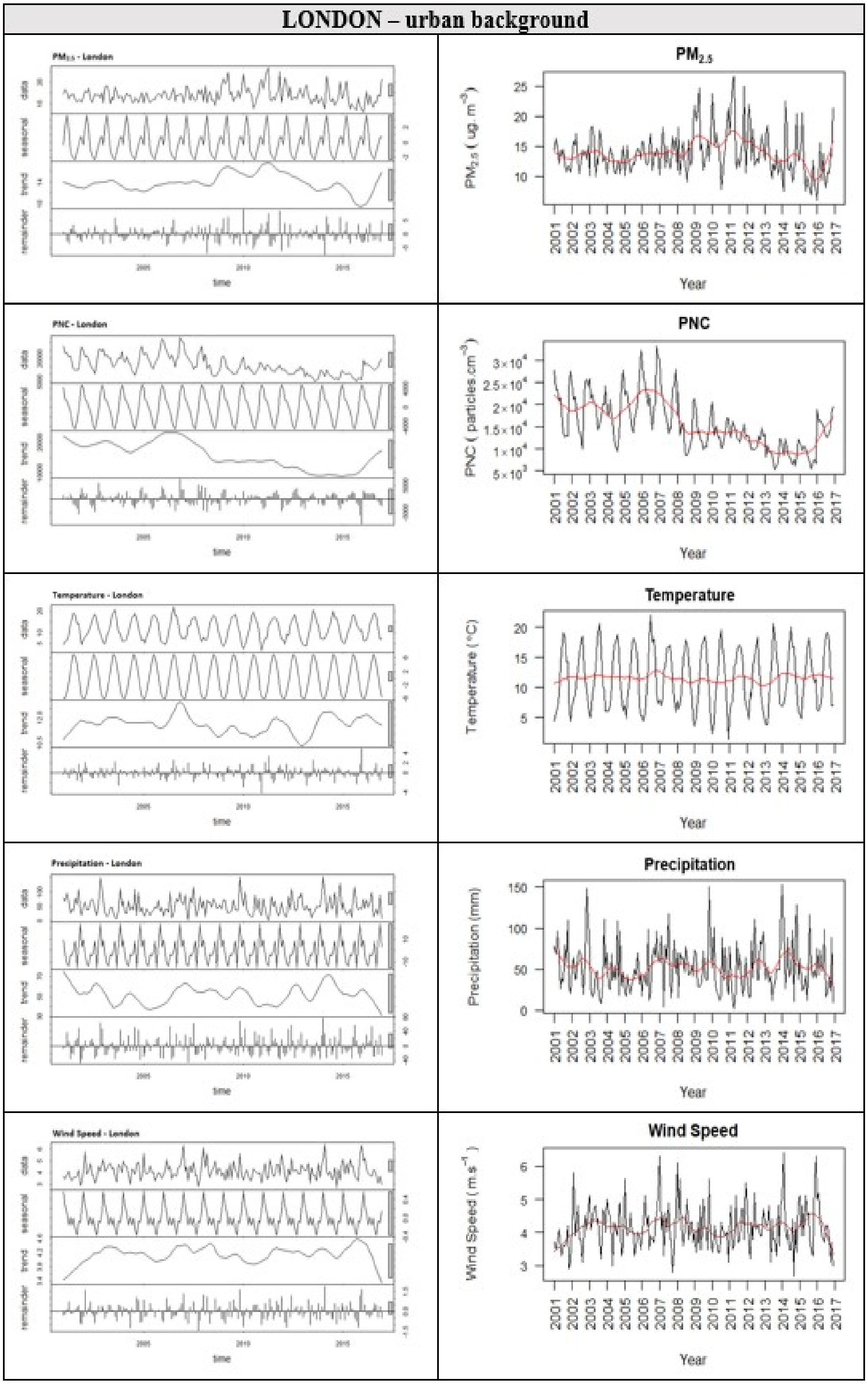

Figure S2d. Decomposition of the monthly $PM_{2.5}$ (μg.cm^{-3}), PNC (particles.cm^{-3}), mean temperature (°C), total precipitation (mm) and mean wind speed (m.s^{-1}) time series into seasonal, trend and stochastic (remainder) components using *stl* then the fitted trend for London – UB

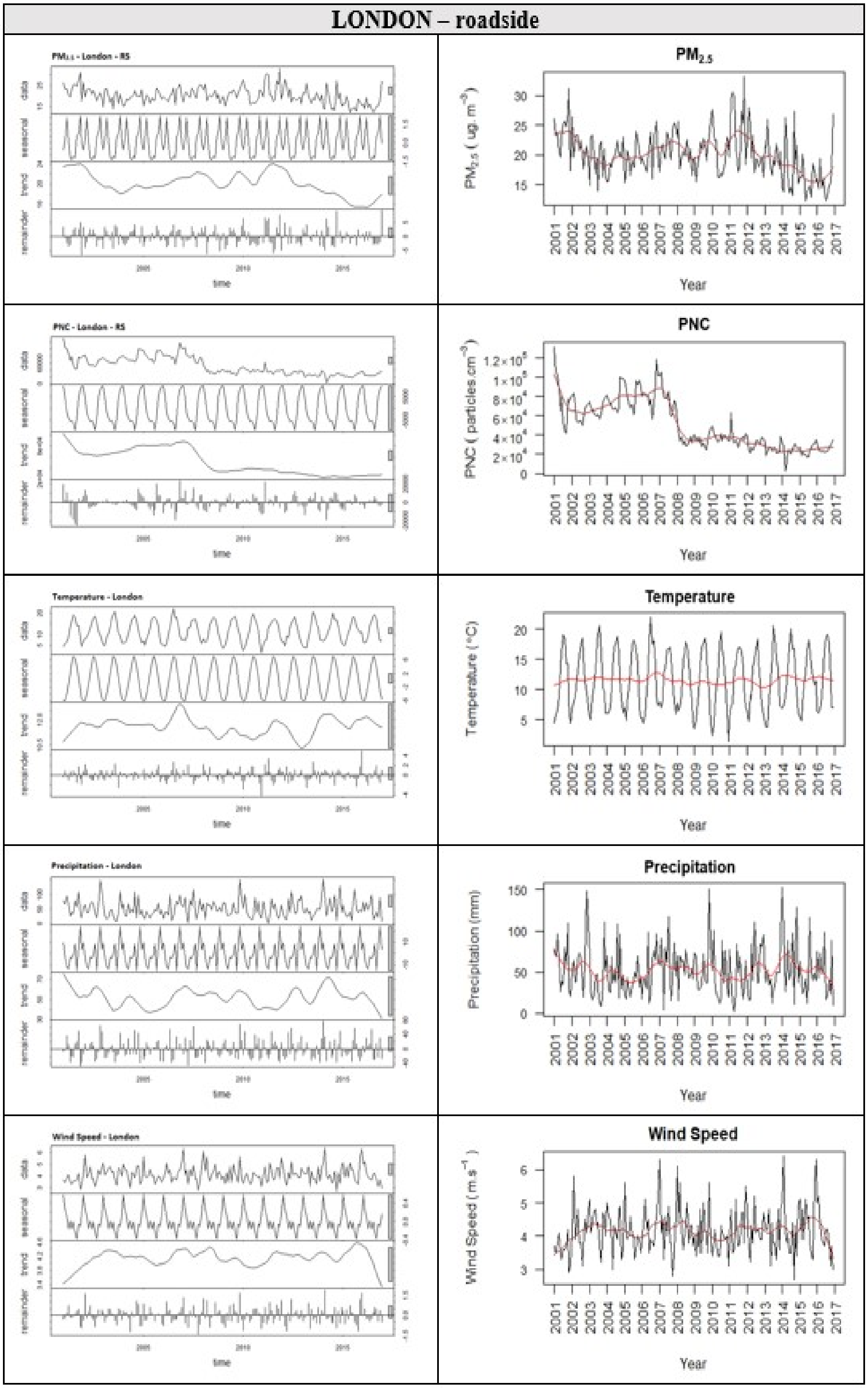

Figure S2e. Decomposition of the monthly $PM_{2.5}$ (µg.cm^{-3}), PNC (particles.cm^{-3}), mean temperature (°C), total precipitation (mm) and mean wind speed (m.s^{-1}) time series into seasonal, trend and stochastic (remainder) components using *stl* then the fitted trend for London – RS.

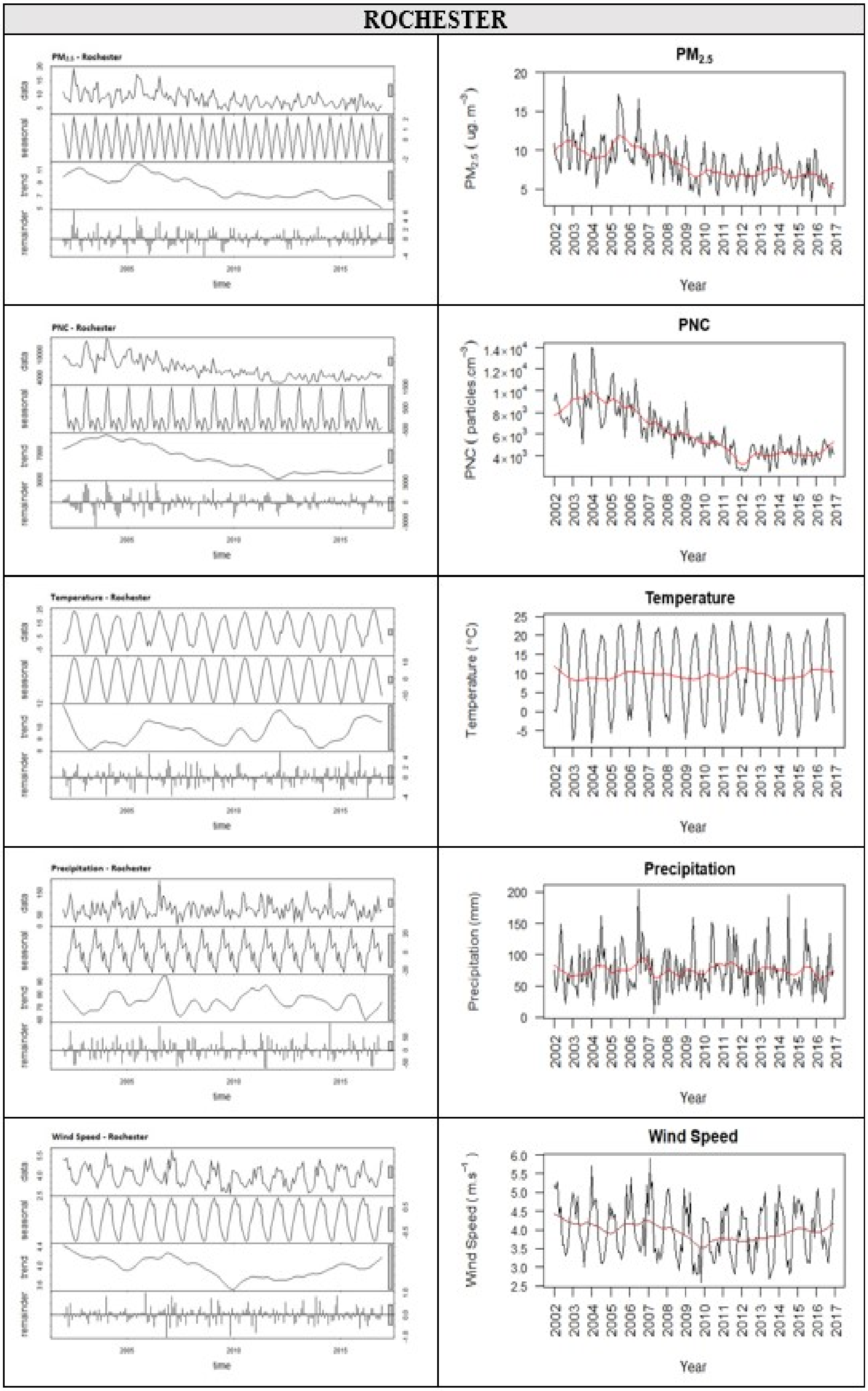

Figure S2f. Decomposition of the monthly $PM_{2.5}$ ($\mu g.cm^{-3}$), PNC ($particles.cm^{-3}$), mean temperature (°C), total precipitation (mm) and mean wind speed ($m.s^{-1}$) time series into seasonal, trend and stochastic (remainder) components using *stl* then the fitted trend for Rochester.

Predictor Analysis

1. Predictor Analysis using Generalised Additive Model (GAM)

The Generalised Additive Model (GAM) is an enhanced generalised linear model that uses a smoothing function to get the contribution of the linear predictors (a.k.a. explanatory variables) and with the properties of an additive model, which is a non-parametric regression method. GAM is preferred for analysis that does not favour a fitted straight line but rather a 'squiggly' line that best describes the data. The obtained smoothed curve (Figure S3 and S4) is the sum of the smooth functions based on the smoothing term applied (e.g. cubic regression splines, LOESS). A GAM can also be used for time series analysis since a time series can be viewed as a summation of individual trends. The effects of each predictors on the response variable are not based on the explanatory variables themselves but on the functions, therefore, are not restricted by the linearity assumption of regression.

GAM was used to estimate the relationships between the response variables ($PM_{2.5}$ and PNC) and the explanatory variables (time, temperature, precipitation, and wind speed). LOESS was used as the smoother and the formula used in the *gam* function were:

$$PM_{2.5} \sim lo(time) + lo(temp) + lo(prec) + lo(wind) \qquad (1)$$
$$PNC \sim lo(time) + lo(temp) + lo(prec) + lo(wind) \qquad (2)$$

In order to evaluate the GAMs, the *deviance explained* (Table S4) were computed as follows:

$$\textit{Deviance Explained} = [(\text{null deviance} - \text{residual deviance}) / \text{null deviance}]*100 \qquad (3)$$

Deviance is a measure of goodness-of-fit of a model, similar to the R^2 of Gaussian data. There are two forms, the null deviance and the residual deviance. The null deviance reflects how well the response variable is predicted by the model with just a constant term (an intercept) while the residual deviance is just the same as the basic residuals of a fitted model, therefore *deviance explained* is the percentage of the null deviance explained by the model.

The obtained *deviance explained* ranged from 23.3 to 75.6% for $PM_{2.5}$ and 38.1 to 79.0% for PNC (Table S5). The performance of the GAMs was better (i.e. higher values indicate a good fit) for PNC than $PM_{2.5}$; therefore, the effects of meteorology on $PM_{2.5}$ are more difficult to model. Among the cities, Brisbane has the lowest *deviance explained,* indicating that the GAMs have poorly captured the effects of meteorology for both $PM_{2.5}$ and PNC, in contrast with Rochester, which has high *deviance explained* for both metrics. Helsinki and London have similar GAM performance for both metrics; the models can explain the variability in PNC due to meteorology more than the variability in $PM_{2.5}$. The GAMs for Augsburg have the reverse performance; the effects of meteorology are captured more for $PM_{2.5}$ than for PNC. London-RS has an interesting response wherein the GAM satisfactorily explained the variability of PNC due to meteorology (high *deviance explained* at 79.0%) but was ineffective for $PM_{2.5}$ (low *deviance explained* at 37.0%).

Table S5. *Deviance explained* for the GAMs of the monthly $PM_{2.5}$ and PNC with the meteorological parameters as additive explanatory variables.

City	***Deviance Explained (%)***	
	$PM_{2.5}$	**PNC**
Augsburg	75.6	57.6
Brisbane	23.3	38.1
Helsinki	58.2	77.1
London	53.0	70.1
London – RS	37.0	79.0
Rochester	64.1	79.0

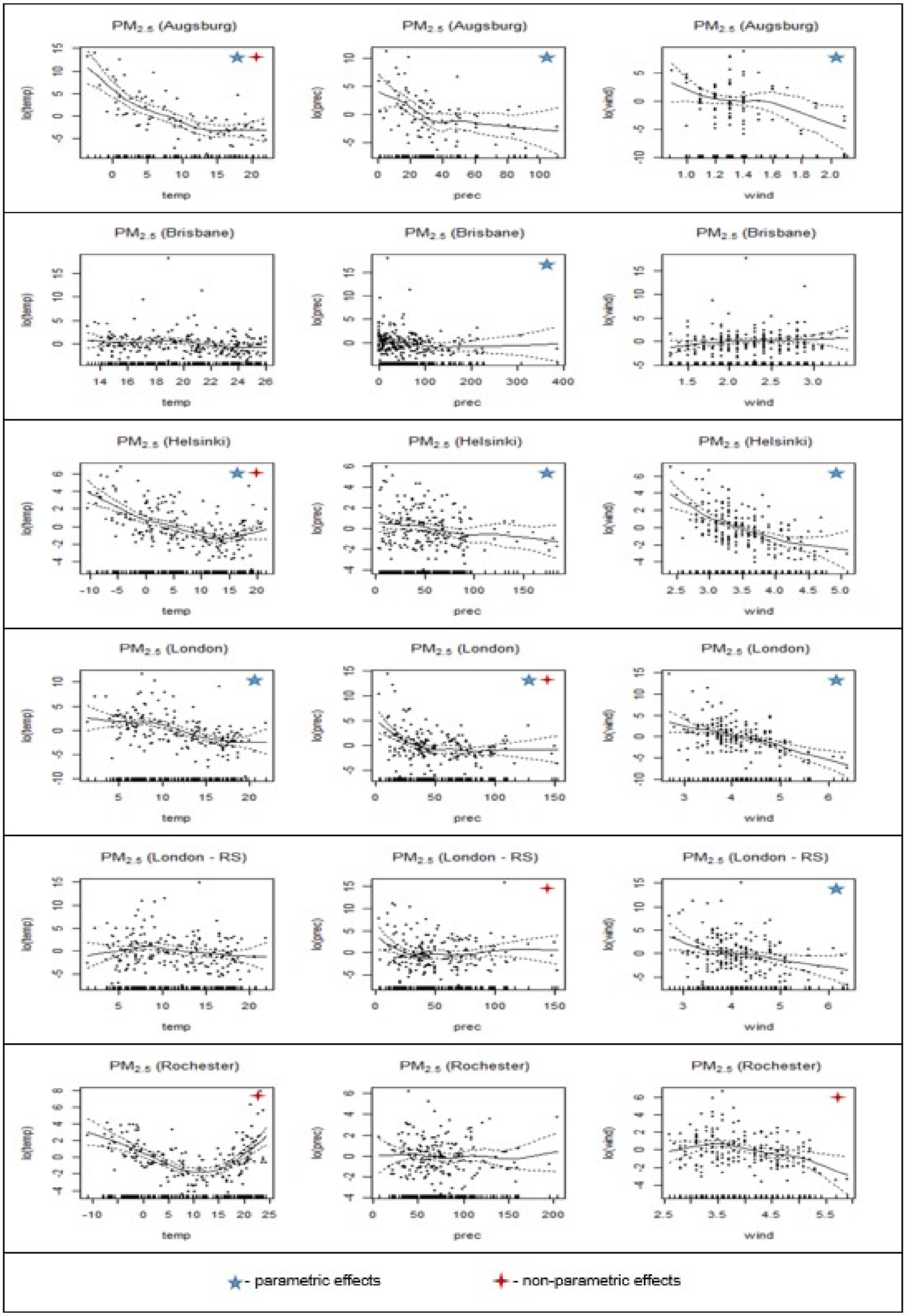

Figure S3. Fitted GAM for $PM_{2.5}$ (solid line) including standard error (dashed line) with monthly mean temperature (°C), total precipitation (mm) and mean wind speed ($m.s^{-1}$) as predictors. Significant ($p < 0.05$) parametric and non-parametric effects are denoted by a blue star and red cross, respectively. (Note: graphs have a different scale for the y-axis).

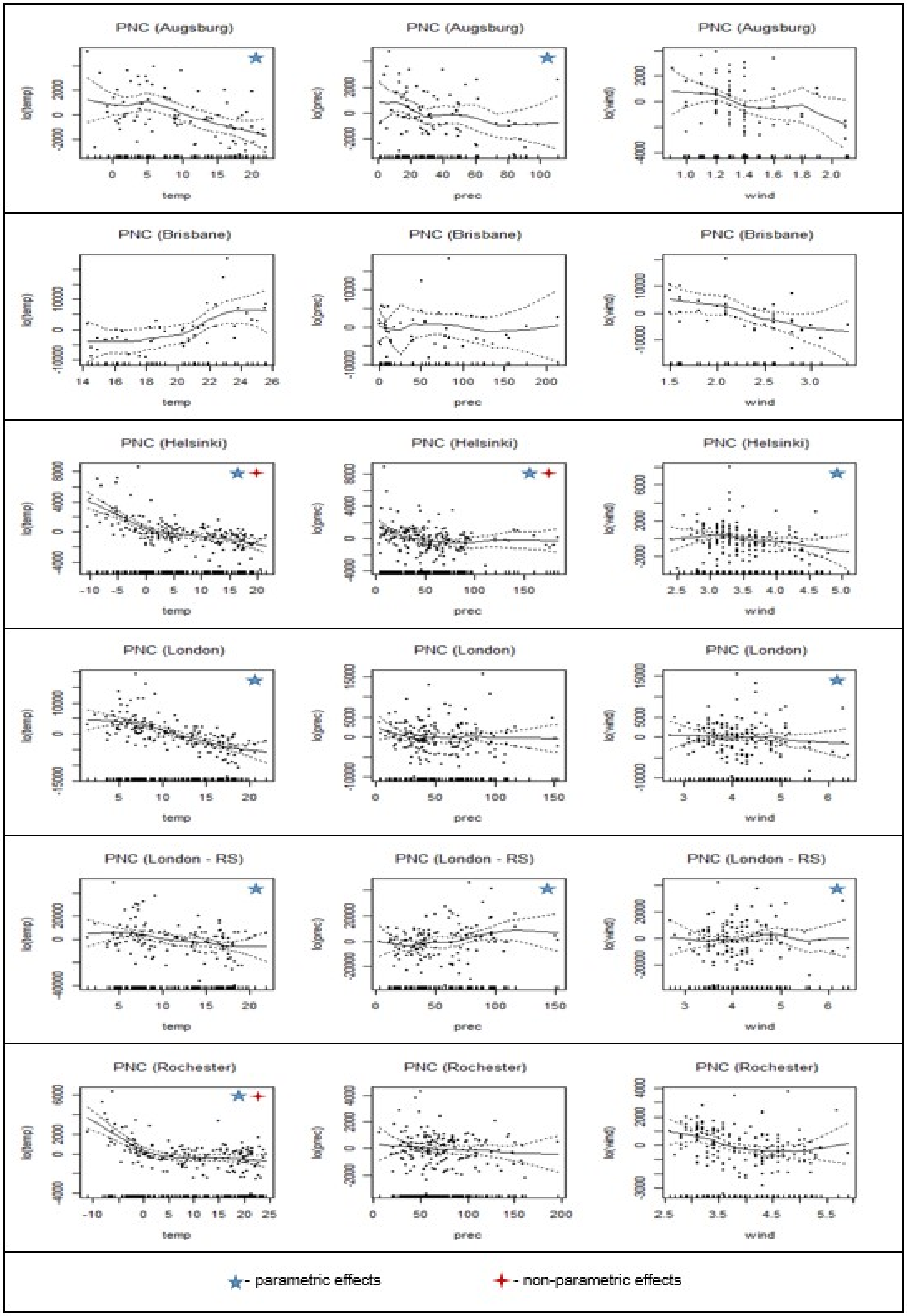

Figure S4. Fitted GAM for PNC (solid line) including standard error (dashed line) with monthly mean temperature (°C), total precipitation (mm) and mean wind speed ($m.s^{-1}$) as predictors. Significant ($p < 0.05$) parametric and non-parametric effects are denoted by a blue star and red cross, respectively. (Note: graphs have a different scale for the y-axis).

An analysis of variance (ANOVA) as a post-test was also part of the *gam* function, which is similar to a classical F test for the mean square if a linear model with multiple covariates (or functions of covariates) is fitted. The parametric and non-parametric effects of all the terms in the model – time, temperature, precipitation and wind speed – were estimated and significance tested. The parametric effects correspond to the linear parts of the fitted smoother, while the non-parametric effects correspond to the non-linear portion; their significance per predictor to $PM_{2.5}$ and PNC per city is indicated in Figures S3 and S4. The parametric effects of wind speed and precipitation dominate $PM_{2.5}$, while temperature is more linearly significant for PNC. The time factor has a more significant non-parametric effect for $PM_{2.5}$, but a more significant parametric effect for PNC.

2. Predictor Analysis using SARIMA Model

An autoregressive integrated moving average (ARIMA) model was fitted in the time series to determine how meteorology played a role in the variation of the PM concentrations in the cities investigated. Temperature, precipitation, and wind speed were considered as the explanatory variables or regressors. An ARIMA model is a stochastic time series model and a special type of regression model mostly used for forecasting that can account for shocks and not only seasonality. Understanding the time series is vital prior to any model fitting and the preliminary step is examining the autocorrelation (ACF) and partial autocorrelation (PACF) plots to determine whether the time series is stationary or not. These plots present a summary of the relationship strength of an observation in a time series with the observations of the previous time period. ACF shows the correlation of the series with itself at different lags while PACF shows the amount of autocorrelation at a particular lag that is not yet accounted for. The *Acf* and *Pacf* functions of the *forecast* package (Hyndman et al., 2018) were used to obtain the ACF and PACF plots, respectively. Both ACF and PACF were used to identify initially the number of terms the time series needs to address the lags for a stationarised series.

ARIMA is classified as a non-stationary time series model wherein the process of interest have (a) a non-constant mean, (b) an infinitive variance, or (c) an autocorrelation function that depends on time. In an ARIMA process, being autoregressive (AR) means that the value of Y at time t depends on its value in the previous time period and a random term. Being integrated (I) implies that the time series needs to be converted into stationary. The moving average (MA) process, which is a type of linear filter, indicates that Y_t is equal to a constant and a moving average of the current and past error terms. Hence, in developing the ARIMA model, three terms were needed to be identified: ***p*** is the AR order that addresses the autocorrelation and the lags of the ‘stationarised’ series, ***d*** is the difference order or the number of times the series needs to be ‘differenced’ to become stationary, and ***q*** is the MA order that deals with the lags of the forecast errors. However, the model was extended since the data was known to possess a seasonal pattern, therefore applying the seasonal ARIMA (SARIMA), which depends on seasonal lags and differences. Four more additional terms were then required: **P** is the seasonal AR order, **D** is the seasonal difference order, **Q** is the seasonal MA order and **S** is the seasonal period (e.g. 4 for quarterly data and 12 for monthly data).

In identifying the best fit model for $PM_{2.5}$ and PNC data, the *ndiffs*, *nsdiffs* and *auto.arima* functions of the *forecast* package were utilised in conjunction with the Box-Jenkins Approach. The *ndiffs* function estimates the number of differences required to make the time series stationary (i.e. gives the value for *d*) applying the KPSS test (Kwiatkowski et al., 1992). The *nsdiffs*, on the other hand, determines the number of seasonal differences required (i.e. gives the value for D) using the measure of seasonal strength (Wang et al., 2006). Given the *d* and D, the *auto.arima* was employed with the Bayesian information criterion (BIC)(Schwarz, 1978) for model selection and the Augmented Dickey-Fuller (ADF)(Dickey & Fuller, 1979) for the stationarity test. The BIC is a residual analysis partly based on the likelihood function that provides an estimate of how much information would be lost if a given model is chosen;

the model with the least BIC is the most preferred. ADF test has a null of a unit root and a non-stationarity assumption. After obtaining the best fit model, the *sarima* function of the *astsa* package (Stoffer, 2017) was used with temperature, precipitation and wind speed as external regressors to determine the relationship between these meteorological parameters and the two PM metrics.

The identified *d* was 1 while *D* was 0 using the *ndiffs* and *nsdiffs* functions with the KPSS test. Then running the *auto.arima* with the ADF test for the stationarity assumption and the BIC as the information criterion, the model that best fit all data is ARIMA(1,1,1)(1,0,0)[12]. Thus the equation for ARIMA(1,1,1)(1,0,0)[12] model is

$$\hat{Y}_t = Y_{t-1} + \alpha_1 Y_{t-1} - \alpha_1 Y_{t-2} - \alpha_1\beta_1 Y_{t-13} + \alpha_1\beta_1 Y_{t-14} + \beta_1 Y_{t-12} - \beta_1 Y_{t-13} + e_t + \theta_1 e_{t-1} \qquad (4)$$

where α_1 is the non-seasonal autoregressive (AR) coefficient, β_1 is the seasonal AR (SAR) coefficient and θ_1 is the non-seasonal moving average (MA) coefficient. Table S6 gives the estimates for the AR(1), MA(1) and the SAR(1) term in the model then an asterisk to indicate significance at 0.05. The lower the α_1 in an AR(1), the quicker is the rate of convergence to the mean. It can be observed that PNC returns to its mean slower than $PM_{2.5}$ and that AR(1) term is significant for all PNC. Since the sum of AR(1) and SAR(1) is less than 1, the fitted model is therefore stationary. The MA(1) coefficient is the fraction of the “shock” from the last period that is still felt in the current period. The relatively high value of the θ_1 in the MA(1), on the other hand, suggests that substantial smoothing was done to estimate the local level and trend. The negative sign attached to θ_1 is merely a convention used by Box and Jenkins. The *p*-value for the term determines if the association between the response and each term in the model is statistically significant. Significant terms can also be interpreted as having significant effects.

Both $PM_{2.5}$ and PNC apparently undergo random “shocks” in a similar way for all urban background stations; the PNC pattern at the London roadside station seemed to be more stable. The values for the BIC ranged from 0.9 – 3.2 for $PM_{2.5}$ and 14.3 – 18.2 for PNC. The computed MAPE ranged from 13.1 – 27.4 for $PM_{2.5}$ and 13.5 – 21.5 for PNC (Table S6). These values are comparable to other SARIMA models for air quality (Rahman et al., 2015). Based on the ACF plot of the residuals and the Ljung-Box statistics (Figure S5 a-c), no significant correlations for the autocorrelation function of the residuals can be observed from for all cities (i.e. residuals are randomly distributed with no regular pattern and are very small that are generally within the significance bounds) and most *p*-values for the Ljung-Box chi-square statistics are >0.05, hence the residuals are independent and that the model meets the assumption (Figure S3 a to c). Further, the standardized residuals indicated no trend, generally no outliers and no changing variance across time. In the Q-Q plot, showed a normally distributed residuals.

Initially, our premise was that different SARIMA would fit the $PM_{2.5}$ and PNC data, knowing that these two metrics behave differently. ARIMA(1,1,1)(2,0,0)[12] was used for $PM_{2.5}$ while ARIMA(0,1,1) for PNC. But it turned out that ARIMA(1,1,1)(1,0,0)[12] had lower BIC and lower MAPE for both $PM_{2.5}$ and PNC compared to the first two. Therefore, only one model was used to analyse the effects of meteorology. The coefficients of the regressors (Table S7) can be interpreted similarly to a linear model, that temperature, precipitation and wind speed were negatively related to both $PM_{2.5}$ and PNC in all cities except for Brisbane’s $PM_{2.5}$ and the temperature, Brisbane’s PNC with temperature and precipitation and for the London – RS station with precipitation and wind speed. The negative correlation means PM concentration decreases as temperature, precipitation, and wind speed increases.

Table S6. Coefficients and significance of each term in the SARIMA(1,1,1)(1,0,0)[12] model fitted to the monthly $PM_{2.5}$ and PNC data of each city with temperature (°C), precipitation (mm) and wind speed ($m.s^{-1}$) as external regressors and the mean absolute percent error (MAPE).

City	$PM_{2.5}$				PNC			
	AR1	MA1	SAR1	MAPE	AR1	MA1	SAR1	MAPE
Augsburg	0.12	-0.88*	0.38*	27.4	0.28*	-0.89*	0.05	15.3
Brisbane	0.63*	-0.98*	0.10	18.2	0.39*	-1.00*	-0.84*	21.5
Helsinki	0.13	-0.93*	0.21*	21.6	0.64*	-0.94*	0.24*	15.1
London	0.30*	-0.93*	0.10	18.3	0.68*	-0.92*	0.05	13.5
London-RS	0.34*	-0.92*	0.17*	13.1	0.55*	-0.81*	0.26*	17.3
Rochester	0.33*	-0.94*	0.36*	19.3	0.42*	-0.91*	0.17*	15.7

*term is statistically significant (p-value ≤ 0.05)

Table S7. Coefficients and significance of the external regressors, temperature (°C), precipitation (mm) and wind speed ($m.s^{-1}$) to the monthly $PM_{2.5}$ and PNC data of the fitted SARIMA(1,1,1)(1,0,0)[12] model for each city.

City	$PM_{2.5}$			PNC		
	Temperature	Precipitation	Wind Speed	Temperature	Precipitation	Wind Speed
Augsburg	-0.58*	-0.04*	-8.21*	-114*	-13	-1834*
Brisbane	0.04	-0.01*	-0.17	394	4.6	-3473
Helsinki	-0.13*	-0.01*	-2.60*	-212*	-3.7	-1355*
London	-0.27*	-0.03*	-2.76*	-700*	-8.6	-1240*
London-RS	-0.01	-0.00	-1.55*	-748*	53	2695*
Rochester	-0.01	-0.00	-1.20*	-81*	-1.6	-552*

*term is statistically significant (p-value ≤ 0.05)

Both models GAM and ARIMA, determined the effects of the meteorological parameters on $PM_{2.5}$ and PNC conjointly. The effect of the meteorological parameters on the particulate matter concentrations can only be determined by fitting a model (He et al., 2017; Kumar & Goyal, 2011; Pearce et al., 2011). A temporal dependence between particle concentration and the predictors was also captured in the ARIMA and alternatively including time in the GAM. The advantage of an ARIMA model is that when dealing with the existence of time correlations, adjacent observations may not be independent and identically distributed (temporal dependence) and the model can account for both seasonal variability and shocks. However, GAM is better for exploratory analysis and predicting correlations if no high-order autocorrelation errors exist (Chen et al., 2001). ARIMA has more rigid assumptions, such as that trends should have regular periods (i.e. hence the differentiation) with constant mean and variance; therefore, GAM is more flexible. Nevertheless, the results on the effects of meteorology are the same – mostly negatively correlated and with precipitation and wind speed as the most important factors for $PM_{2.5}$ and temperature to PNC. In the fitted SARIMA model, the precipitation effect is not significant to PNC in all cities (Table S7). The cross-correlation plots further demonstrate this (Figure S6 a–c). A strong autocorrelation mostly occurs at lag 0 but there are also other autocorrelations that occur throughout the series at different lags. The $PM_{2.5}$ and PNC correlation plots with temperature evidently illustrate a six-month pattern of dependence, which did not exist with precipitation and wind speed.

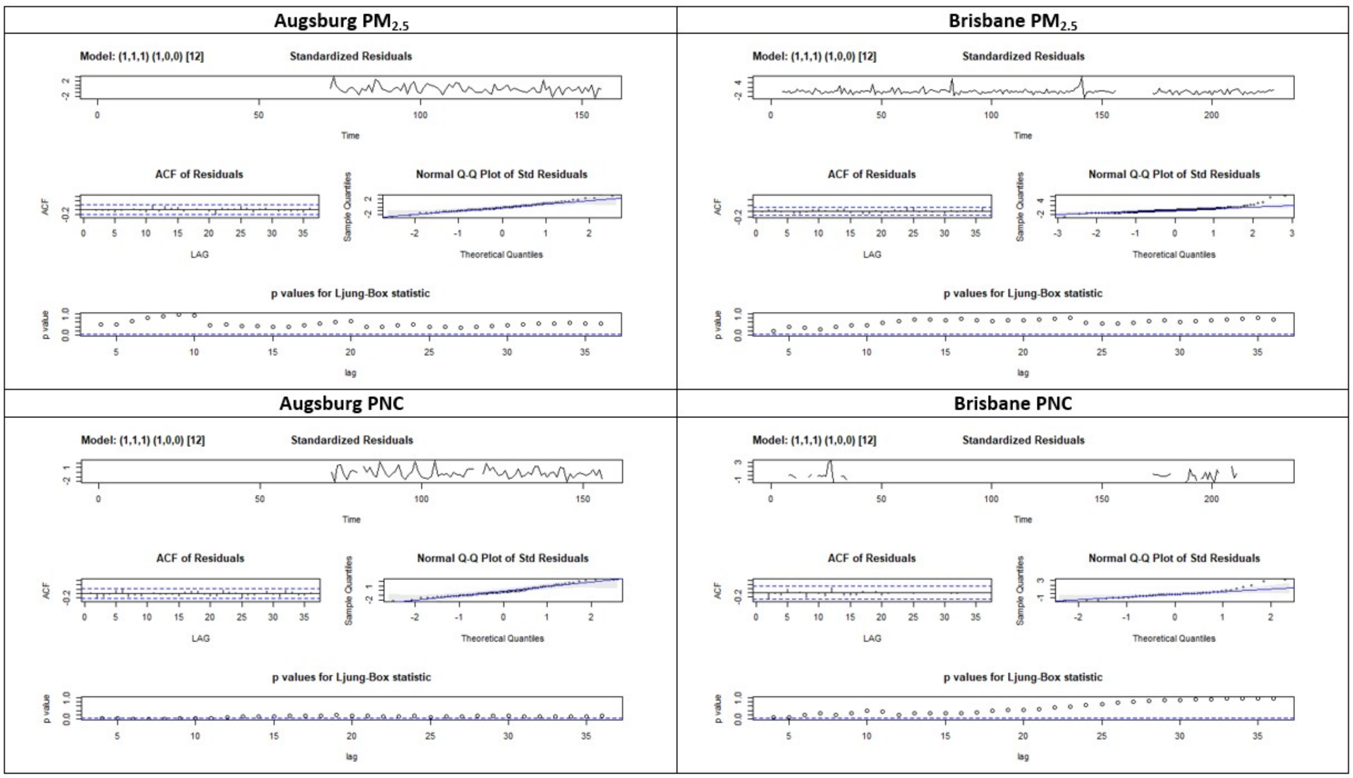

Figure S5a. ACF of Residuals and the Ljung-Box statistics for SARIMA(1,1,1)(1,0,0)[12] model fitted in the $PM_{2.5}$ and PNC data of each city with temperature (°C), precipitation (mm) and wind speed ($m.s^{-1}$) as external regressors.

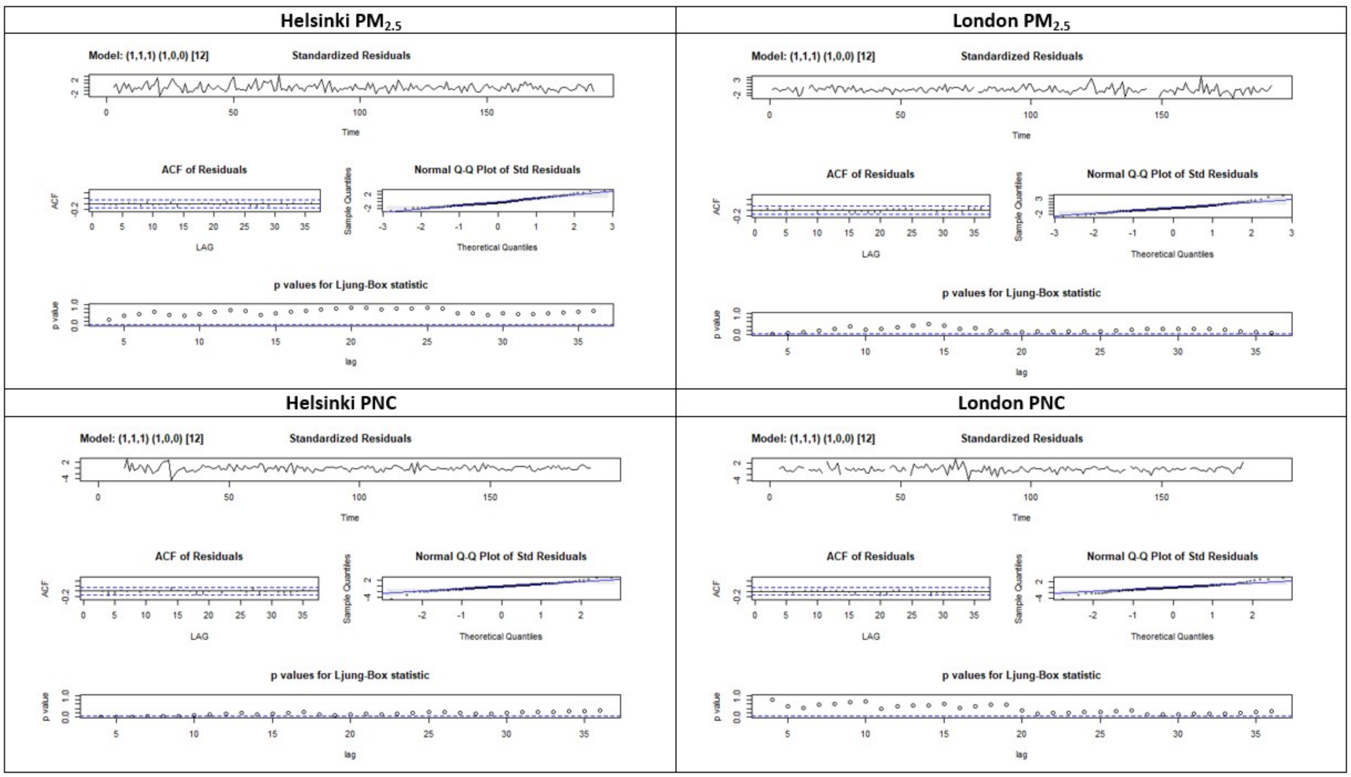

Figure S5b. ACF of Residuals and the Ljung-Box statistics for SARIMA(1,1,1)(1,0,0)[12] model fitted in the $PM_{2.5}$ and PNC data of each city with temperature (°C), precipitation (mm) and wind speed ($m.s^{-1}$) as external regressors.

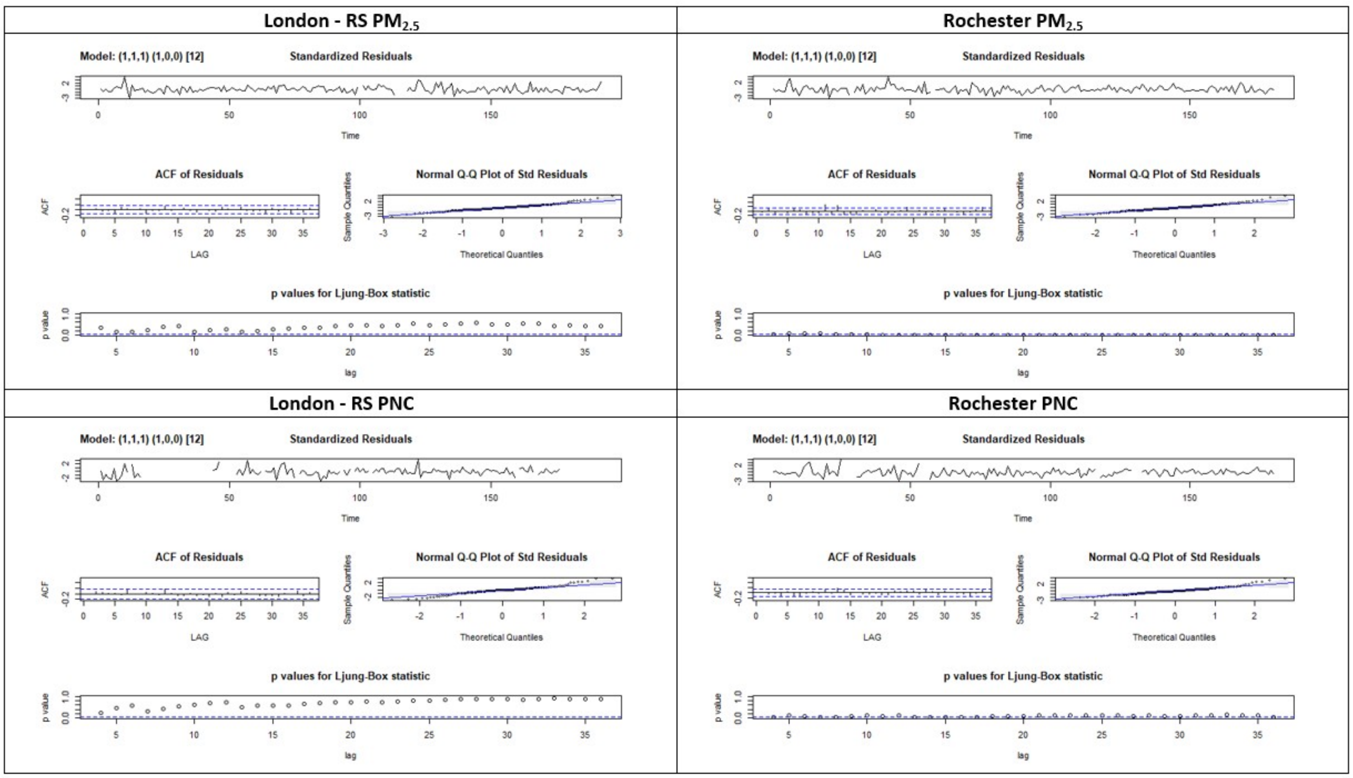

Figure S5c. ACF of Residuals and the Ljung-Box statistics for SARIMA(1,1,1)(1,0,0)[12] model fitted in the monthly $PM_{2.5}$ and PNC data of each city with mean temperature (°C), total precipitation (mm) and mean wind speed ($m.s^{-1}$) as external regressors.

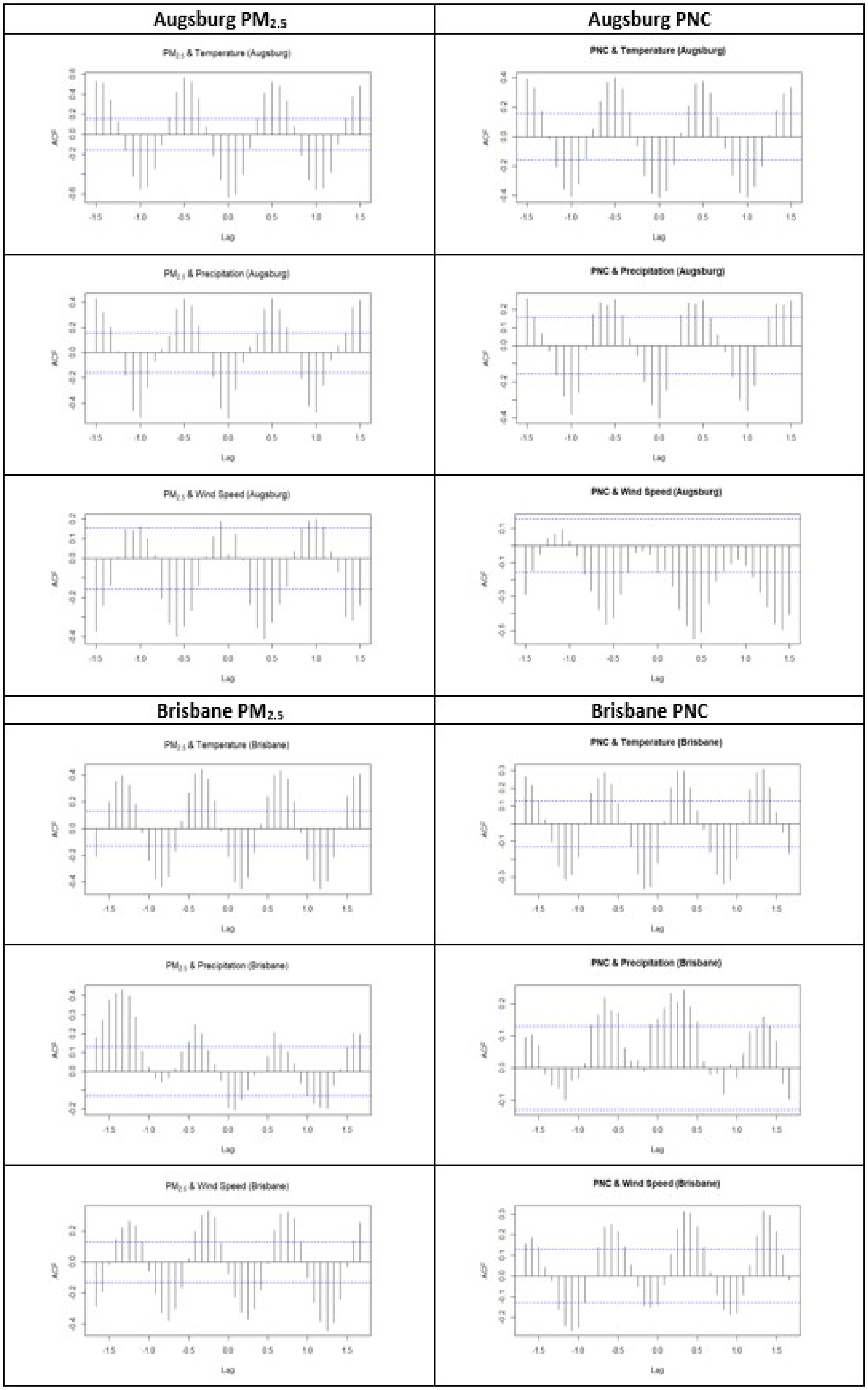

Figure S6a. Cross-correlation of monthly $PM_{2.5}$ and PNC data of each city with mean temperature (°C), total precipitation (mm) and mean wind speed ($m.s^{-1}$).

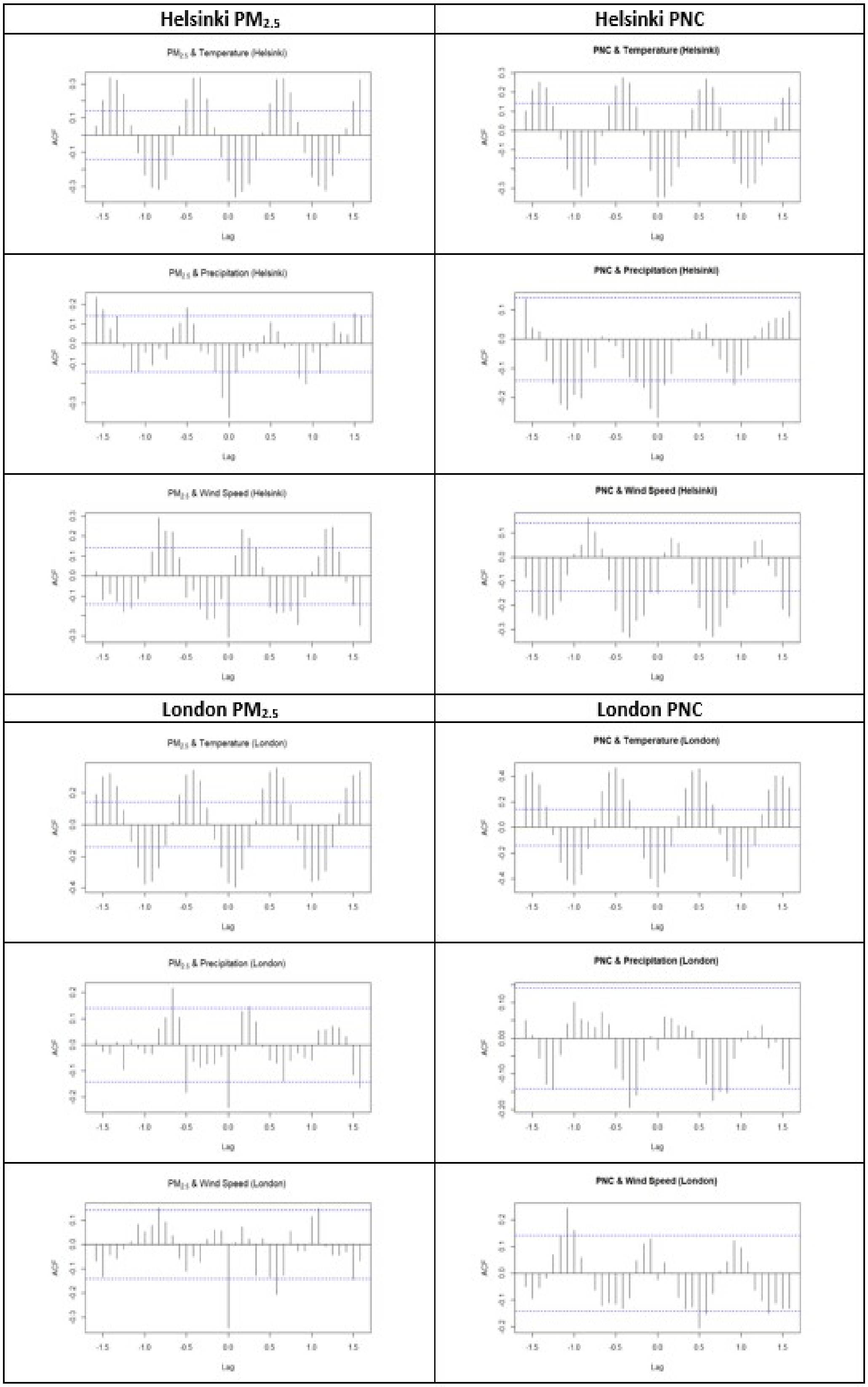

Figure S6b. Cross-correlation of monthly $PM_{2.5}$ and PNC data of each city with mean temperature (°C), total precipitation (mm) and mean wind speed ($m.s^{-1}$).

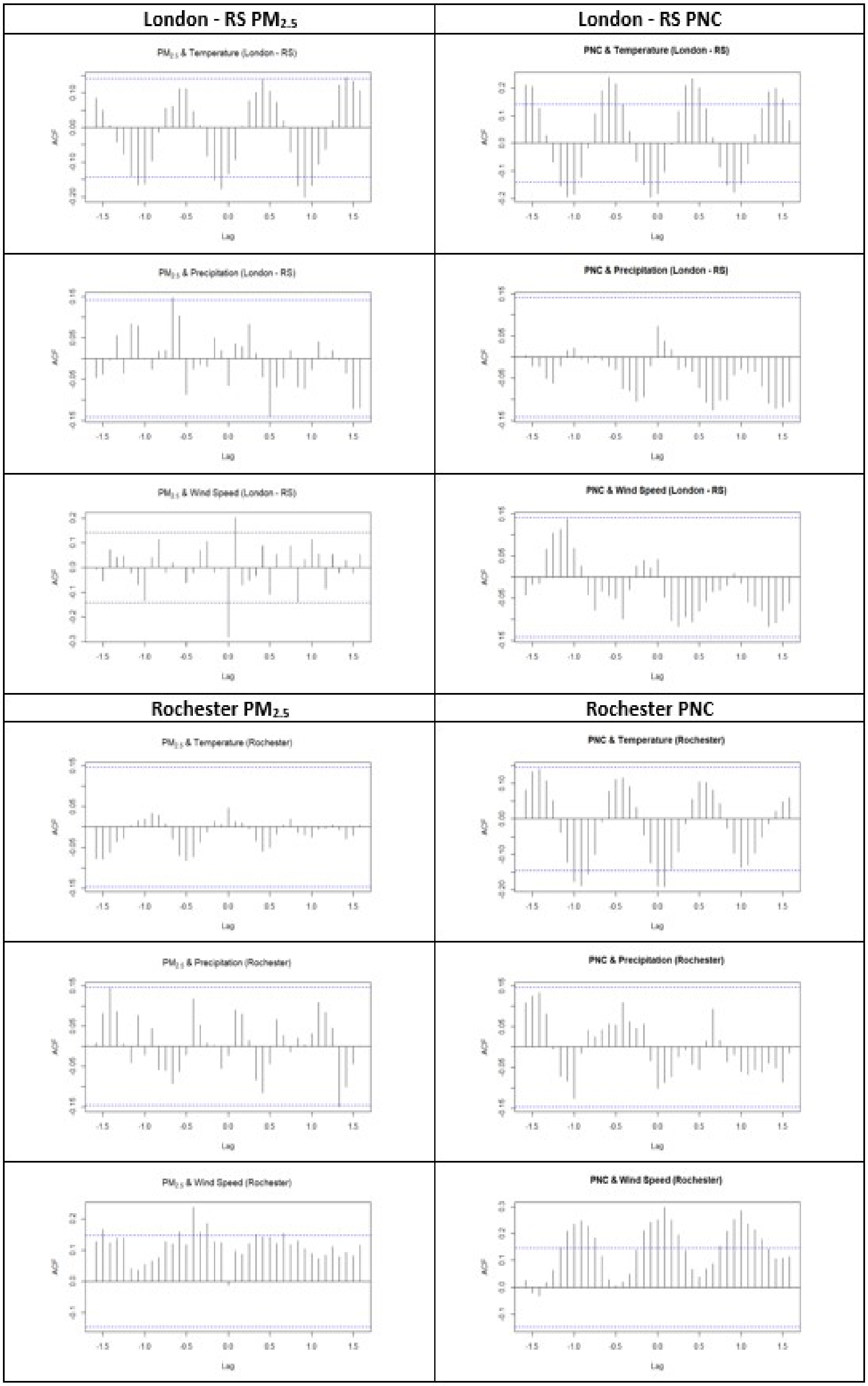

Figure S6c. Cross-correlation of monthly $PM_{2.5}$ and PNC data of each city with mean temperature (°C), total precipitation (mm) and mean wind speed ($m.s^{-1}$).

Change-point detection

Table S8. Detected change-points for the imputed monthly meteorological parameters using Buishand Range Test.

Parameter	Augsburg	Brisbane	Helsinki	London	Rochester
n	156	228	192	192	180
Temperature					
Date of shift	Aug 2013	Nov 2012	Jun 2006	Aug 2007	May 2011
Precipitation					
Date of shift	Nov 2013	Apr 2012	Dec 2003	Feb 2003	Apr 2014
Wind Speed					
Date of shift	Jun 2009	Aug 2009	Jun 2006	Jun 2002	Jul 2008

Table S9. Detected change-points for the imputed monthly $PM_{2.5}$ using Buishand Range Test for different periods.

Period	Augsburg	Brisbane	Helsinki	London	London-RS	Rochester
P_T						
n	156	228	192	192	192	180
Date of shift	Sep 2006	Aug 2003	Jan 2011	Feb 2013	Jul 2012	Jul 2008
P_1 (≤2006)						
n	36	108	72	72	72	60
Date of shift	May 2005	Aug 2001	Sep 2005	Nov 2003	Aug 2002	Dec 2004
P_2 (2007 – 2011)						
n	60	60	60	60	60	60
Date of shift	Jul 2008	Mar 2009	Dec 2010	Sep 2008	Oct 2010	Dec 2008
P_3 (2012 – 2016)						
n	60	60	60	60	60	60
Date of shift	Apr 2014	Dec 2014	Jan 2015	May 2013	Dec 2013	Jun 2014

Table S10. Detected change-points for the imputed monthly PNC using Buishand Range Test for different periods.

Period	Augsburg	Brisbane[1]	Helsinki	London	London-RS	Rochester
P_T						
n	156	228	192	192	192	180
Date of shift	Apr 2009	May 2015	Jun 2007	Mar 2008	Jan 2008	Jan 2008
P_1 (≤2006)						
n	36	36	72	72	72	60
Date of shift	May 2005	Nov 1998	Jun 2003	Apr 2005	Nov 2003	Aug 2005
P_2 (2007 – 2011)						
n	60	---	60	60	60	60
Date of shift	Feb 2009	---	Feb 2009	Mar 2008	Mar 2008	Jun 2009
P_3 (2012 – 2016)						
n	60	60	60	60	60	60
Date of shift	Aug 2014	Jun 2014	Jul 2014	Nov 2015	Jul 2014	Oct 2013

[1]*Note: Brisbane data is only from 1998 – 2000 and P_3 is 2011 – 2015*

Table S11. Summary of trends at the detected change-points for the selected meteorological parameters and for $PM_{2.5}$ and PNC.

City	**Change-point**	**Trend**		**Change-point**	**Trend**		**Change-point**	**Trend**	
		Prec.	**$PM_{2.5}$**		**Wind**	**$PM_{2.5}$**		**Temp.**	**PNC**
Augsburg	2013	dec	dec	2009	inc	dec	2013	inc	inc
Brisbane	2012	dec	inc	2009	dec	inc	2012	inc	dec
Helsinki	2003	inc	dec	2006	inc	dec	2006	inc	dec
London	2003	dec	dec	2002	inc	inc	2007	dec	dec
London-RS	2003	dec	dec	2002	inc	dec	2007	dec	dec
Rochester	2014	dec	dec	2008	dec	dec	2011	inc	dec

Note: Prec. Is precipitation, Wind is wind speed, and Temp. is temperature, while dec means decrease and in is increase.

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