Assessing air pollution in European cities to support a citizen centered approach to air quality management

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Abstract

European cities have made significant progress over the last decades towards clean air. Despite this progress, several cities are still facing acute air pollution episodes, with various urban areas frequently exceeding air quality levels allowed by the European legal standards and WHO guidelines. In this paper, six European cities/ regions (Bristol, UK; Amsterdam, NL; Sosnowiec, PL; Ljubljana, SI; Aveiro, PT; Liguria, IT) are studied in terms of air quality, namely particulate matter, nitrogen dioxide and ozone. The concentrations trends from 2008 – 2017 in the different typology of monitoring stations are addressed, together with the knowledge of daily, weekly and seasonal pollution patterns to better understand the city specific profiles and to characterise pollutant dynamics and variations in multiple locations. Additionally, an analysis of the duration and severity of air pollution episodes is also discussed, followed by an analysis of the fulfillment of the legislated limit values.

Each of our 6 case study locations face different air pollution problems, but all these case studies have made some progress in reducing ambient concentrations. In Bristol, there have been strong downward trends in many air pollutants, but the levels of NO_2 remain persistently high and of concern. In recent years, decreasing concentration levels point to some success of Amsterdam air quality policies. PM_{10} exceedances are a seasonal pollution problem in Ljubljana, Sosnowiec and Aveiro region (even if with different lev-

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els of severity). While, exceedances of NO_2 and O_3 concentrations are still problematic in Liguria region.

The main findings of this paper are particular relevant to define and compare future citizen-led strategies and policy initiatives that may be implemented to improve and fulfill the EU legislation and the WHO guidelines.

Keywords: European cities, EU legislation, ClairCity project, air quality, monitoring data, temporal patterns and trends

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1. Introduction

Many European cities are affected by poor air quality levels and regularly exceed both the European standards prescribed by the Ambient Air Quality Directive (AQD) (2008/50/CE) and guidelines recommended by the World Health Organization (WHO) (EEA, 2019, 2020). This is particularly the case for particulate matter with diameters of 10 μm and smaller (PM_{10}), for which both the daily and the yearly average limit values are often exceeded in many European cities (EEA, 2019, 2020). For fine inhalable particles ($PM_{2.5}$), the EU limit value is generally met (EEA, 2019, 2020), but only a few cities manage to keep concentrations below the levels recommended by the WHO (EEA, 2020; Thunis et al., 2018). According to the latest report released by the European Environment Agency (EEA) on air quality in Europe, the WHO guideline for $PM_{2.5}$ annual mean was exceeded at 70% of the monitoring stations across Europe (EEA, 2020). Additionally, the EEA estimates that in 2018 nitrogen dioxide (NO_2) was linked to 54,000 premature deaths, and ground-level ozone was linked to 19,400 premature deaths across the European Union countries (EEA, 2020). Consequently, poor air quality is recognized as one of the most pressing environmental issues in urban areas and remains the largest environmental risk in Europe. More than an environmental issue, air pollution has become the world's largest environmental health threat (Lelieveld et al., 2020).

To reduce these air pollution effects, particularly in cities where most of the European population lives, it is important to define effective planning strategies for air quality improvement (Miranda et al., 2015; Monteiro et al., 2018b; Pisoni et al., 2019a; Viana et al., 2020). The 2008 European AQD requires Member States to design appropriate air quality plans for zones where the air quality does not comply with the AQD limit values, to plan and implement possible emission reduction measures to improve air quality (Coelho et al., 2020; Pisoni et al., 2019b; Thunis et al., 2016).

ClairCity, an innovative European project funded by the EU Horizon 2020 program (Ref: 689289), engaged thousands of citizens across Europe to define policy measures that consider the optimal local interventions that are more citizen centered in their design to achieve a low carbon, clean air, health future. The project focussed on six distinct European urban areas, all of them with over 50,000 inhabitants: Bristol in the United Kingdom, Amsterdam in the Netherlands, Ljubljana in Slovenia, Sosnowiec in Poland, the Aveiro region in Portugal and the Liguria region around Genoa in Italy. The focus of the project was to take a more citizen-centered approach to air quality management by primarily focusing on the relationships between citizens day to day behaviors, practices and activities and the links to air pollution and carbon emissions (Hayes, 2017). The current air quality management practices need to go beyond the traditional approach to provide a new perspective based instead on citizens daily activities behaviour and practices which will clearly allow the connection to be made between pollution and behaviour, and link these to the various practices that constitute everyday life within our cities. Therefore, the research question addressed in this paper is: would it be possible to support air quality management practices with a citizen-centered approach through a historical air quality assessment study? To understand the local context, a complete diagnosis of the air quality and its main emission sources for each case study was implemented. The main objective of this paper is to present a comprehensive air quality assessment, based on existing air quality monitoring data, to support a citizen centered approach. The main findings of this paper will allow the identification of the main problems and causes of air pollution, to then support the development of more effective local policies for emission abatement in European cities by initiating new modes of engaging citizens, stakeholders and policy makers.

In this paper, air quality data recorded in these different European cities was analysed for a 10–year period (where available), focusing on the main critical pollutants in urban areas, combining different approaches (Flemming et al., 2005; Gama et al., 2018; Henschel et al., 2013; Jo and Park, 2005; Liu et al., 2015; Lonati et al., 2006; Zhao et al., 2009). The paper is organised as follows: in Section 2, the air quality data collection methodology along the six cities is described in detail, followed by a description of the six urban case studies main characteristics. Section 3 focuses on the analysis and interpretation of the monitoring data, considering the daily, weekly and seasonal pollution patterns (sub-section 3.1), the concentrations trends in the different typology of monitoring stations (sub-section 3.2), and the duration and severity of air pollution episodes (sub-section 3.3). In addition, an analysis of the fulfilment of the legislated limit values is presented as Supplementary Material (SM 5.2). Finally, in Section 4, the main conclusions are summarized.

2. Air quality assessment framework

An assessment of measured ambient air quality data was performed for the period from 2008 to 2017, focusing on PM_{10}, $PM_{2.5}$, NO_2 and O_3 concentrations (where available). The selection of ozone for this analysis, even if it is not directly related with citizens behaviour, is justified by its health-related effects, and assuming that a citizen-centered approach is not just about the generation of pollution but also about the protection of health through exposure minimization. The main findings of this assessment support the baseline characterization of the air quality status of the six cities and regions and will be the basis for the validation of the air quality modelling tools applied in the ClairCity project. The air quality assessment was performed for the study areas included in the computational domains, shown in Figure 1, and cover the urban areas of each case study and were selected based on a preliminary discussion with local stakeholders.

2.1. Air quality assessment methodology

The air quality monitoring data was retrieved from the European Air Quality Database (EEA, b) for the years 2008 to 2012, and from the Air Quality e-Reporting database (EEA, a) for the years 2013 to 2017, for all the case studies. Additionally, for Bristol data was obtained from the UK Automatic Urban and Rural Network (AURN), which is part of the national monitoring network and five additional monitoring stations maintained by Bristol City Council. These monitoring stations follow the same QA/QC procedures as the national AURN network.

The monitoring stations were selected based on their data capture for each year. A station was considered eligible when half of the years (at least 5 out of 10) had more than 75% data capture. An exception was made for $PM_{2.5}$ in the Liguria Region station IT0858A, where only 4 years out of 10 were available, otherwise there would be no data for this pollutant. Preference was given of stations that have more recent data, meaning if a station fulfils the criteria, but does not have data for any of the five more recent years it was not selected. A list of all selected stations is presented in Table 1. In Ljubljana, Sosnowiec and Liguria region some stations do not have hourly PM data. In addition, all the selected monitoring stations are automatic and use the chemiluminescence method to measure NO_2 concentrations, and the ultraviolet (UV) photometry method to measure O_3 concentrations. While different methods are used to measure PM_{10} and $PM_{2.5}$ concentrations, the tapered element oscillating microbalance (TEOM), the gravimetric analysis and the beta ray attenuation, depending on the country and site. Data measured using the TEOM or the beta ray attenuation method cannot always be considered equivalent to the manual gravimetric reference method, which is required in Europe for compliance measurements. Correction procedures are employed by each Member State to obtain reference equivalent PM_{10} and $PM_{2.5}$ data series from automatic TEOM and beta-attenuation monitors.

Although all cities meet the monitoring requirements established by the AQD, Amsterdam is clearly the city with the highest density air quality monitoring network, with 17 air quality monitoring stations (AQS), distributed over an area of 500 km^2, and encompassing a population of 834,713 inhabitants. The assessment of the spatial representativeness (SR) of air quality monitoring stations is an important subject linked with several research and management areas, including risk assessment and population exposure, the design of monitoring networks, model development, model evaluation and data assimilation. The European Commission is working on the implementation of a harmonised programme for the monitoring of air pollutants and to ensure that the information collected on air pollution is sufficiently representative and comparable across the Community. However, there is not yet detailed provisions on the methods for assessing the SR (Kracht et al., 2017). Also in the scientific literature, there is no unified agreement to address this complex problem, and no well-established procedure for assessing SR has been identified so far. All the monitoring stations included in this study follow the EU directive classification scheme based on two indicators on different scales (Decision 2011/850/EU): “type of area” (rural, suburban, urban), and “type of station” (in relation to predominant emission sources relevant for the measurement: background, traffic, industrial). Concentrations measured at background stations are assumed to be representative of a wider area (EU, 2008), referring to “exposure of the general population”. While the selected traffic and industrial stations in this study are not representative of the “exposure of the general population”. Nevertheless, and having in mind the main goal of the paper, which focuses on the relationship between citizens behavior and air quality management, it is crucial to consider all the stations within the boundaries of the city/ region.

This study employed classic statistical methods for time series analysis by using the R package OpenAir (Carslaw and Ropkins, 2012; Ropkins and Carslaw, 2012), developed for the purpose of analyzing air quality data. Concentrations of PM_{10}, $PM_{2.5}$, NO_2, and O_3 registered at the six cities were used to characterize the variability of mean pollutant concentrations on the timescales from diurnal to annual, addressing the processes driving this variability. In addition, long-term temporal trends of mean pollutant concentrations have been estimated. To characterize extreme values in air pollutant concentrations, the duration and severity of air pollution episodes were also assessed in this study.

Furthermore, this analysis also integrated some field-knowledge from the ClairCity engagement activities, namely local interviews with citizens, stakeholders, decision– and policy–makers, which were crucial to identify the most critical air pollution problems of each pilot city/ region, and the public perception of those problems. All the collected data are compiled in Artola and Bolscher (2018); Slingerland et al. (2018a, 2017); Slingerland and Smith (2018); Slingerland et al. (2018b); Smith et al. (2017).

These approaches contribute to the historical air quality assessment study, providing essential data to inform and engage citizens.

2.2. Summary of the six EU case studies

No two cities are the same, so the six EU case studies were chosen to represent diversity such as different air pollution sources, geographies, meteorology, economies, demographics, and local air quality capacity and capabilities. Table 2 provides a summary of each case study city and region. Further information can be found as Supplementary Material (SM5.1). Figure 1 shows the location of the AQS considered for each case study and the corresponding classification.

Table 1: Summary of the air quality monitoring network of the case studies.

		Number of stations measuring			
Case study	Type of stations	PM_{10}	$PM_{2.5}$	NO_2	O_3
Bristol	2 urban background	1	1	2	1
	4 urban traffic	–	–	4	–
Amsterdam	6 urban background	2	2	4	2
	6 urban traffic	4	3	5	1
	2 urban industrial	2	1	1	–
	3 rural background	3	2	1	–
Ljubljana	2 urban background	2	1	1	1
Sosnowiec	1 urban background	1	–	1	–
	1 urban traffic	1	1	1	–
Aveiro region	1 urban traffic	1	–	1	–
	1 suburban background	1	–	1	1
	1 suburban industrial	1	1	1	1
Liguria region	3 urban background	1	1	2	3
	5 urban traffic	–	–	5	–
	1 urban industrial	1	–	1	–

Table 2: Summary of the main characteristics of each case study (information was gathered from Artola and Bolscher (2018); Slingerland et al. (2018a, 2017); Slingerland and Smith (2018); Slingerland et al. (2018b); Smith et al. (2017)).

	Bristol	Amsterdam	Ljuljana	Sosnowiec	Aveiro region	Liguria region
Population[1]	450,000	834,713	288,919	206,000	363,752	855,834
Population density (hab/km^2)	4,000	4,700	1,075	2,376	215	466
Climate classification[2]	Temperate Oceanic (Cbf)	Temperate Oceanic (Cbf)	Warm–summer humid continental (Dfb)	Warm–summer humid continental (Dfb)	Warm–summer mediterranean (Csb)	Hot-summer mediterranean (Csa)
Domain area	20 km × 20 km	25 km × 20 km	20 km × 20 km	20 km × 20 km	40 km × 55 km	25 km × 15 km
Main economic activities	Services Industry Higher-education	Port Airport Tourism Industry Services	Limited industry Services Tourism	Heavy industry Services	Heavy industry Port Services Agriculture (inland)	Services (mainly tourism) Industry Port
Population distribution within the area	Mainly central	Mainly central	Mainly central	More equally distributed	Mainly coastal	Mainly coastal
Number of stations per 100,000 inhabitants	1.3	2.0	0.7	1.0	0.8	1.1

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[1] data from 2016

[2] classified following the Köppen-Geiger Climate Classification System

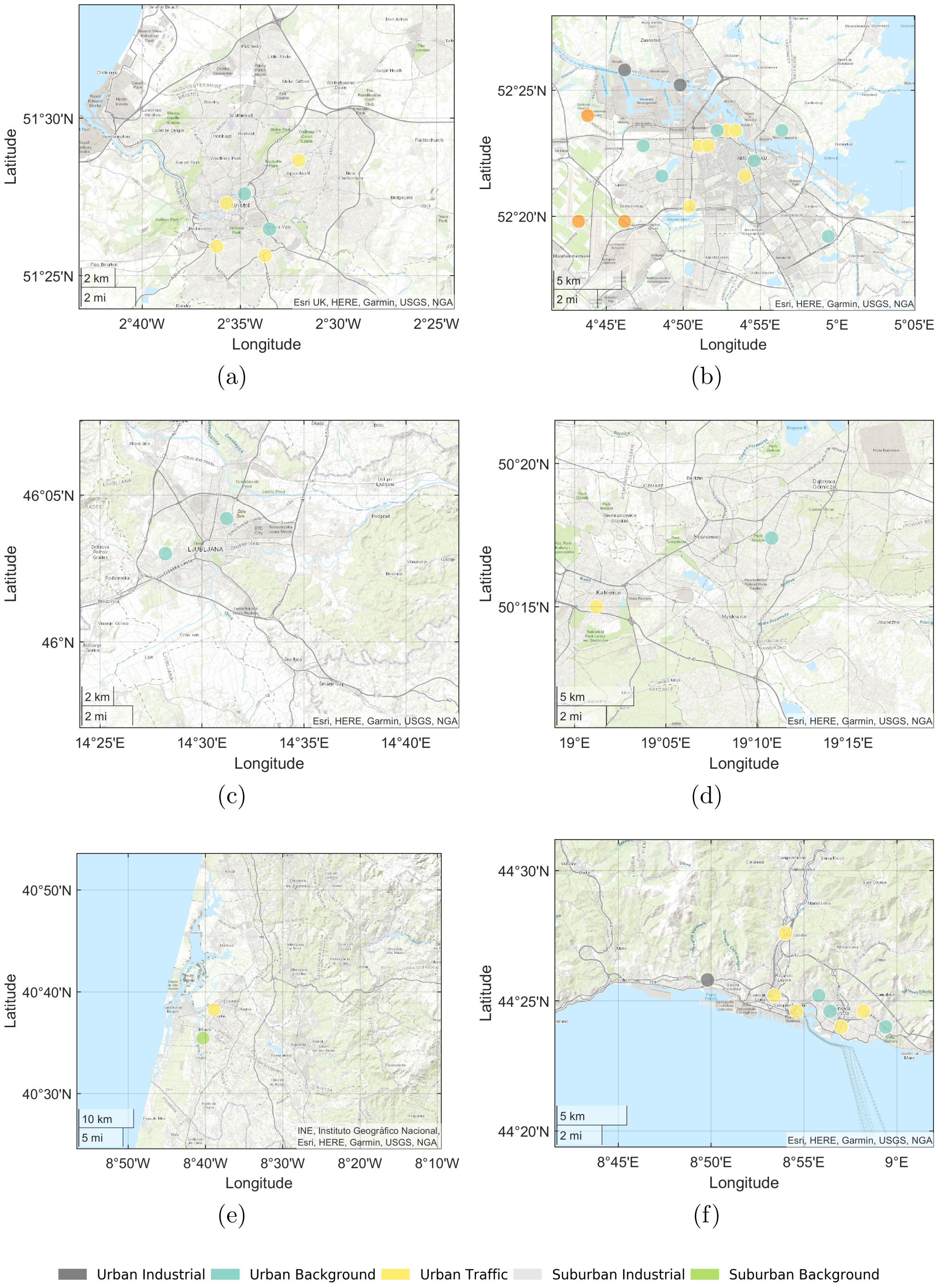

Figure 1: Location of the air quality monitoring stations within the selected study areas of each case study: Bristol (a), Amsterdam (b), Ljubljana (c), Sosnowiec (d), Aveiro region (e), and Liguria region (f)

3. Air quality assessment

The results of the air quality assessment are presented in this section considering the variability of pollutant concentrations on the timescales from diurnal to annual (sub-section 3.1), the trend describing the mean concentrations evolution during the 10 years period (sub-section 3.2), and the duration and severity of air pollution episodes (sub-section 3.3). In addition, an analysis of the fulfillment of the legislated limit values is presented as Supplementary Material (SM 5.2).

3.1. Time profiles

To characterize the air quality temporal patterns at the six case studies, air quality observations have been grouped considering different time scales. For each pollutant, PM_{10}, $PM_{2.5}$, NO_2, and O_3, hourly (mean hour of day variation), daily (day of the week variation), and monthly (monthly plot) cycle plots have been done, using the OpenAir package for R (Carslaw and Ropkins, 2012; Ropkins and Carslaw, 2012). In these plots the mean and the 95% confidence interval are depicted and the color of each line/shadow represents the type of station: blue for the urban background; green for the suburban background; yellow for the urban traffic; grey for the urban industrial and orange for the rural background.

3.1.1. PM_{10} and $PM_{2.5}$ concentrations

Figures 2 and 3 show the variation of PM_{10} and $PM_{2.5}$ concentrations, respectively, by hour of the day, by day of the week and by month of the year, considering all data observed between 2008 and 2017, for each case study.

Bristol, Amsterdam, Ljubljana, and Liguria hourly profiles show a peak of PM10 and PM2.5 concentrations in the morning (between 7 and 10h), which may be linked with road traffic emissions. For Bristol, Ljubljana and Sosnowiec, a similar peak is also observed in the evening. High PM10 and PM2.5 concentrations are also observed during night-time in Sosnowiec and Aveiro region, that may be related with both the daily evolution of the urban atmospheric boundary layer, which gets thinner during the night (Oke et al., 2017), and with a contribution of semi-volatile material condensing on ambient particles with the lower night-time temperatures (Harrison et al., 2012). In turn, Liguria shows a strong decrease of PM concentrations in the evening, an opposite pattern to the observed in the other cities. This may be explained by the penetration of sea breezes in the evening, bringing cleaner air into the city (e.g. Viana et al. (2005) found minimum PM levels during night-time due to reductions on the average mixing height and night-time catabatic winds, for a regional background site in the Barcelona city area).

Regarding the daily profiles, there is a negligible variability in Bristol for both PM_{10} and $PM_{2.5}$ concentrations, while in Amsterdam the profile indicates a decrease of PM concentrations during weekends, more notably at the traffic stations. During Sundays, PM_{10} and $PM_{2.5}$ concentrations are about 3.1 and 1.8 μg m^{-3}, respectively, lower than average weekdays concentration, which reflects the importance of coarse particles of anthropogenic origin in Amsterdam. For Sosnowiec the decrease of concentrations in weekends it is not so evident. In Aveiro region, the daily profiles indicate slightly lower concentrations on Sundays. Additionally, the traffic station monitored higher concentrations (about 6.8 μg m^{-3}) than the suburban background station, which may be used as an estimation for the traffic contribution to PM_{10} and $PM_{2.5}$ concentrations (Pant and Harrison, 2013). In addition, for Liguria, the daily profile shows a decrease of PM_{10} concentrations during the weekend at the industrial station (about 3.6 μg m^{-3} lower on Sundays than during average weekdays), while the background station profiles kept constant.

On contrary to the other cities/ regions, Liguria region monthly profile shows peaks of PM concentrations during summer months, particularly for $PM_{2.5}$. This may be linked with the enhancement of photochemically driven secondary formation of aerosols, from anthropogenic precursors transported from populated and industrialized areas such as the Po Valley.

In turn, Bristol and Amsterdam indicates a slight decrease of PM_{10} and $PM_{2.5}$ concentrations in spring and summer months, with slightly higher levels in winter months potentially linked with residential heating practices. For Ljubljana, Aveiro, and particularly in Sosnowiec, monitored PM_{10} and $PM_{2.5}$ concentrations are much higher during winter months than during summer months, and are also higher than in the other cities. For example, in Sosnowiec, mean winter concentrations are about 2.6 times higher than those in the summer period. These results are in accordance with the great seasonal variability of the $PM_{2.5}$ concentrations in Poland described by Rogula-Kozlowska et al. (2014). In this study, this variability is attributed to the seasonal fluctuations of the emissions of PM and its precursors from hard and brown coal combustion for energy production, growing in a heating season, reaching maximum in winter, and decreasing in a non-heating period.

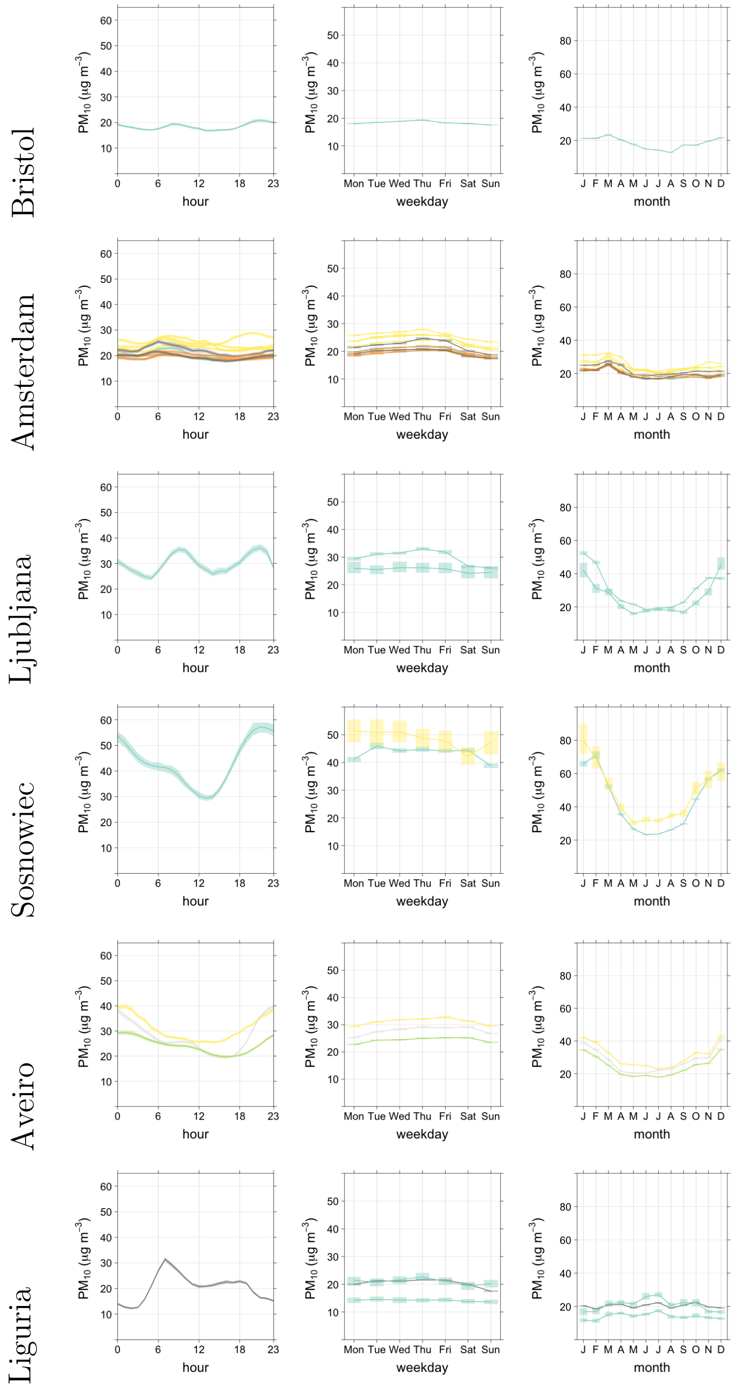

Figure 2: Hourly, daily, and monthly variability of the PM_{10} concentrations observed between 2008 and 2017, for the 6 case studies. The solid line shows mean concentrations, while the bars in the daily and monthly plots, as well as the band in the hourly plots represent the 95% confidence intervals in the mean. Data from urban background stations is depicted in blue, suburban background in green, urban traffic in yellow, urban industrial in grey and rural background in orange.

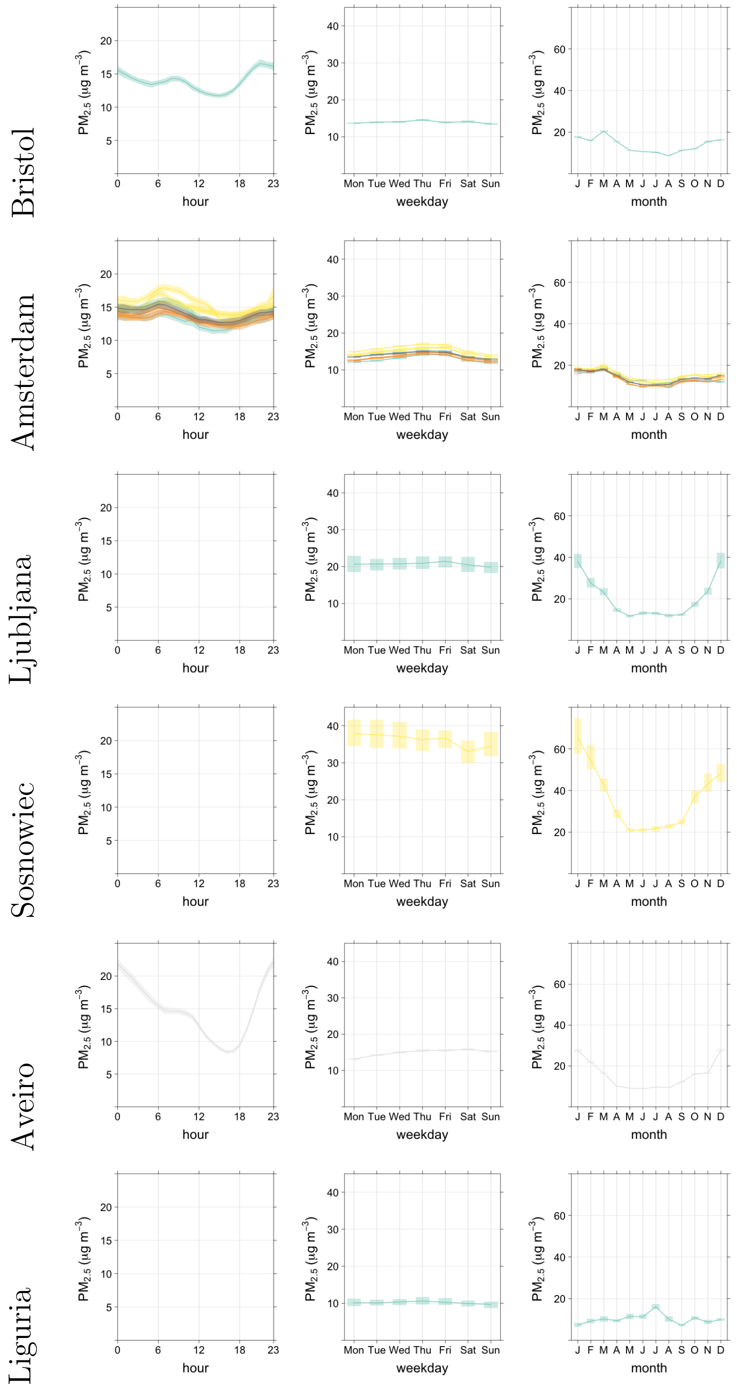

Figure 3: Hourly, daily, and monthly variability of the PM$_{2.5}$ concentrations observed between 2008 and 2017, for the 6 case studies. The solid line shows mean concentrations, while the bars in the daily and monthly plots, as well as the band in the hourly plots represent the 95% confidence intervals in the mean. Data from urban background stations is depicted in blue, suburban background in green, urban traffic in yellow, urban industrial in grey and rural background in orange.

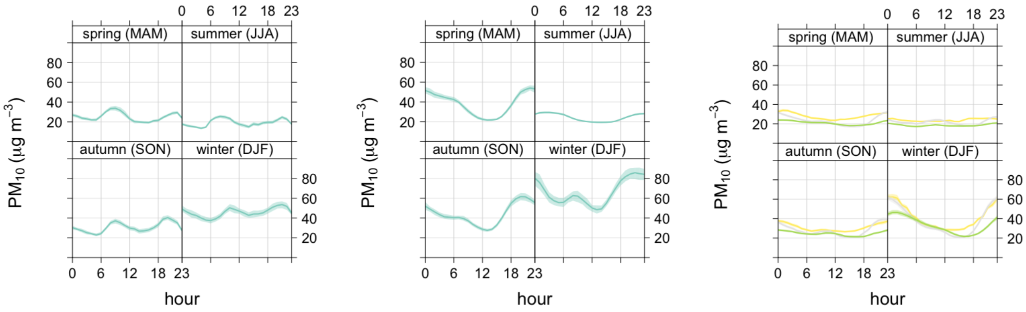

Figure 4: Hourly variability by season of the year of the PM_{10} concentrations observed between 2008 and 2017, for Ljubljana (a), Sosnowiec (b) and Aveiro (c). The solid line shows mean concentrations while the shading shows the 95% confidence interval in the mean. Data from urban background stations is depicted in blue, suburban background in green, urban traffic in yellow and urban industrial in grey.

Moreover, during winter meteorological conditions which favour the accumulation of pollutants at surface levels are common, which may contribute to episodic increases in PM concentrations (Chen et al., 2020). To complement this analysis, Figure 4 presents the hourly variation of PM_{10} concentrations by season of the year, for the three cities/ regions. At Ljubljana, the PM_{10} daily profile has a similar progression between seasons. Conversely, Sosnowiec and Aveiro exhibit a huge peak in mean hourly concentrations during late evening and night, during winter, but without significant peaks during summer. PM_{10} mean concentrations at 00h are 52.4 and 32.8 μg m^{-3} higher than during summer, at Sosnowiec and Aveiro, respectively. As previously stated, evening peaks may be related with daily evolution of the atmospheric boundary layer, evening contribution of domestic sources such as heating (Gonçalves et al., 2012; Vicente et al., 2015) and cooking, and contribution of semi-volatile material condensing on ambient particles (Harrison et al., 2012). All these causes are more important during winter (due to thinner and more stable boundary layers, more emissions from heating, and colder night-time temperatures), which may explain the results shown on Figure 4.

Sosnowiec also has large smog problems in wintertime. According to the literature (Adamczyk et al., 2017; Lubecki et al., 2019; Woźniak et al., 2020), the main sources of particulate matter are low stack emissions from household stoves burning coal and waste. Episodes of high concentrations of PM are most often associated with increased dust emissions from communal-living sources, which is accompanied by unfavorable conditions of air pollution spread (anticyclones situations with a large territorial range, weak wind, strong thermal inversion, negative average daily air temperatures).

Previous studies for the northern and central part of Portugal, Aveiro Region included, indicate that, overall, residential and commercial combustion units for heating, followed by industrial combustion processes, are the main source of PM_{10} (Borrego et al., 2010; Figueiredo et al., 2013; Gonçalves et al., 2012; Lopes, 2018; Monteiro et al., 2018a).

3.1.2. NO_2 and O_3 concentrations

The temporal variability of NO_2 and O_3 concentrations in the troposphere is connected, since these two pollutants are both involved in several specific chemical reactions which play a key role in their concentrations. Typically, the diurnal cycle of O_3 and NO_2 exhibit an inverse relationship where O_3 shows a peak during the afternoon (due to photochemical production) and lower night-time concentrations. Close to emission sources, freshly emitted NO locally scavenges O_3, yielding NO_2, which contributes to the night-time drop in O_3 concentrations. In addition, dry deposition of O_3 plays also an important role in the decrease of the concentrations of this pollutant during the night and early morning. In terms of monthly profiles, as sunlight triggers OH production, causing NO_2 to be removed from the atmosphere (Melkonyan and Kuttler, 2012), lower NO_2 concentrations are expected during summer. On the other hand, higher values of NO_2 are expected in winter, when the solar activity and OH concentrations are lower (Melkonyan and Kuttler, 2012). Moreover, winter is the season with the strongest anthropogenic emissions in Europe because of heating (Cincinelli et al., 2019; Vicente and Alves, 2018).

Figures 5 and 6 show the time variation of NO_2 and O_3 concentrations, respectively, considering all data observed between 2008 and 2017. Note that for O_3 only 5 case studies were considered, due to the lack of data for Sosnowiec.

The hourly profiles of NO_2 concentrations in the six cities/ regions show two peaks of concentrations, one in the morning, and the other in the evening, associated with the peak road traffic in the cities. In general, traffic stations (data plotted in yellow) show the largest NO_2 concentrations. This behavior was expected, since NO_2 in ambient air is in large part derived from the oxidation of NO, a pollutant which is emitted from combustion processes.

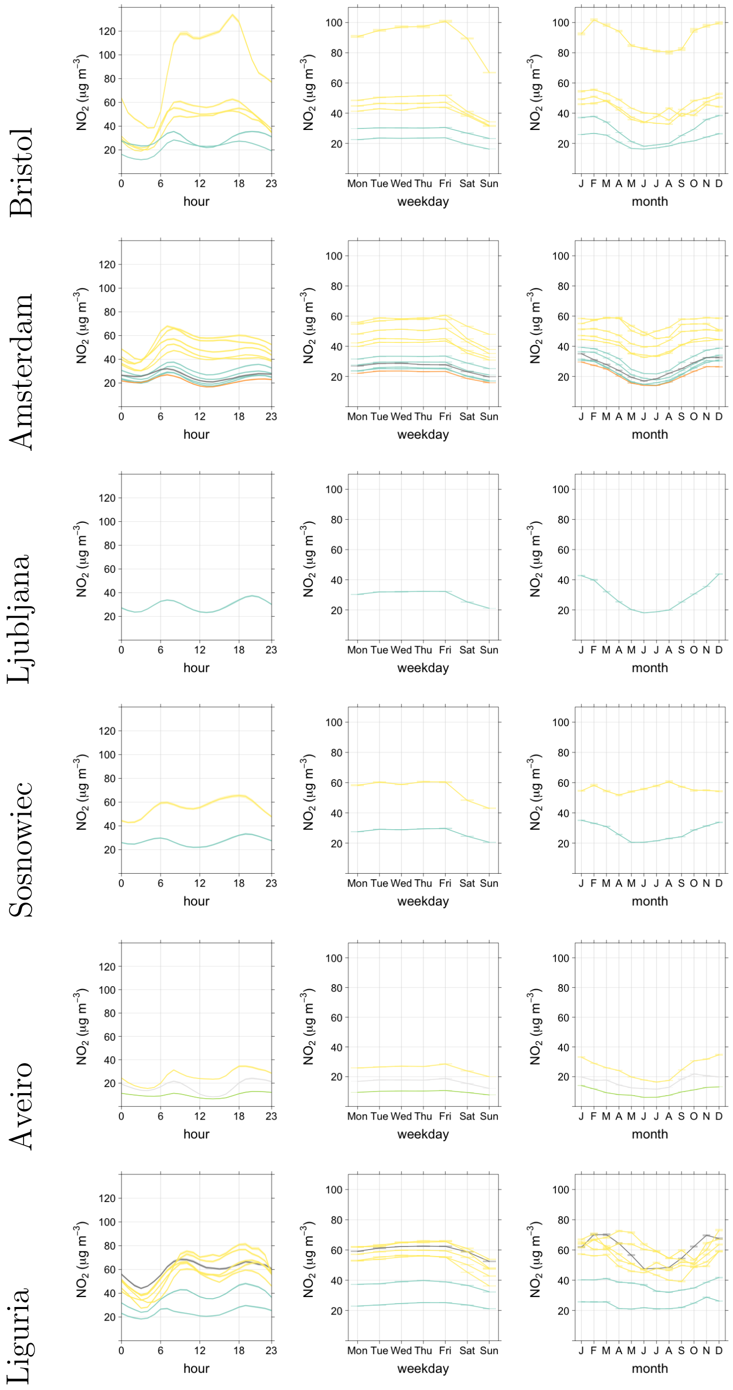

Figure 5: Hourly, daily, and monthly variability of the NO$_2$ concentrations observed between 2008 and 2017, for the 6 case studies. The solid line shows mean concentrations, while the bars in the daily and monthly plots, as well as the band in the hourly plots represent the 95% confidence intervals in the mean. Data from urban background stations is depicted in blue, suburban background in green, urban traffic in yellow, urban industrial in grey and rural background in orange.

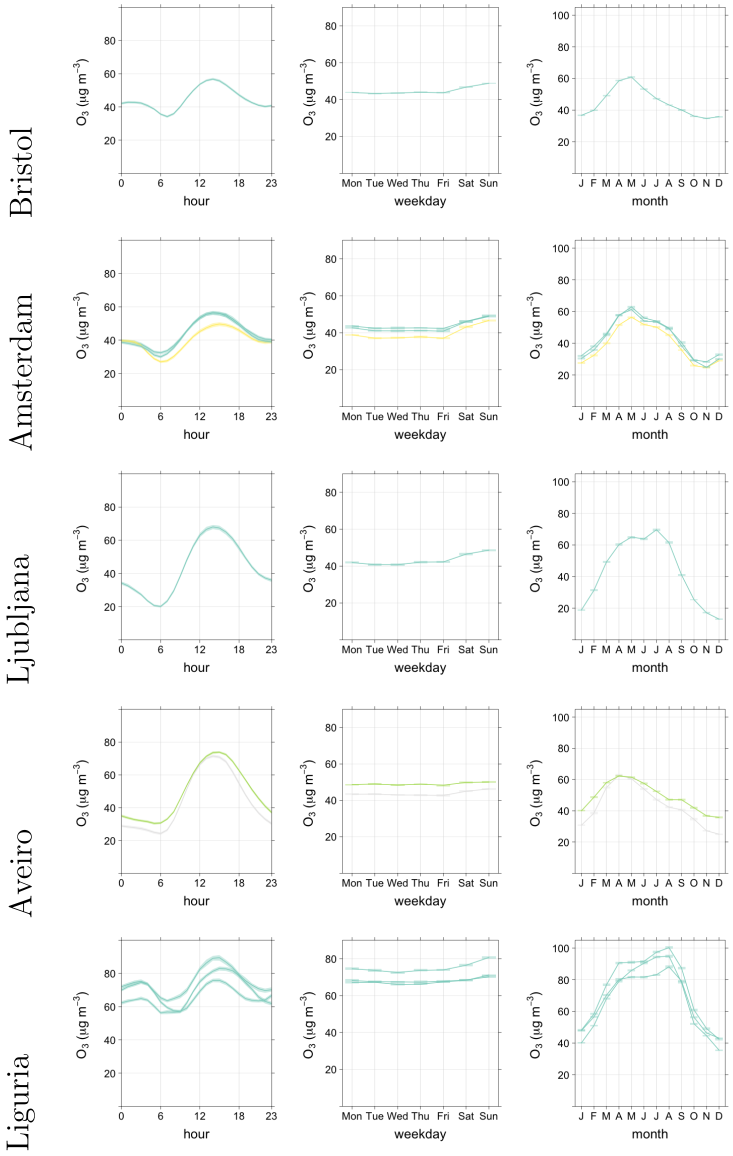

Figure 6: Hourly, daily, and monthly variability of the O_3 concentrations observed between 2008 and 2017, for the 6 case studies. The solid line shows mean concentrations, while the bars in the daily and monthly plots, as well as the band in the hourly plots represent the 95% confidence intervals in the mean. Data from urban background stations is depicted in blue, suburban background in green, urban traffic in yellow and urban industrial in grey.

Since NO sources, at urban areas, are mainly from vehicular exhaust, NO_2 is therefore a clear indicator for road traffic (Rafael et al., 2020). One traffic station is highlighted in Bristol, as having very high mean NO_2 concentrations between 9h and 18h (higher than 110 μg m^{-3}). Although the NO_2 concentrations observed in this station present lower values during the weekend, due to lower traffic levels and less pronounced peaks (mean difference between week days and Sundays is about 23.4 μg m^{-3}, very high mean levels are registered even on Sundays (higher than 50 μg m^{-3}).

In Ljubljana, cars are the most used means of transportation, which causes problems of traffic congestion (Slingerland et al., 2018b). Although the city's air quality monitoring network does not include traffic stations (see Table 1), the observed NO_2 cycles at background stations still denote traffic influence, with peaks at rush hours.

Sosnowiec faces typical urban congestion problems, which explain the higher NO_2 concentrations at the traffic station than at the background station (with a mean delta of about 28.5 μg m^{-3}).

In the Aveiro region, a contribution of 64% from traffic emissions for the NO_2 concentrations was found (Gama et al., 2021).

Liguria also shows significantly higher concentrations at the traffic stations, when compared to the background stations (mean difference between traffic and background stations is about 18.1 μg m^{-3}). The industrial station in this city also shows NO_2 peaks in the morning and evening and mean concentrations during daytime of the same levels as the monitored range at the traffic stations. However, during nighttime, the NO_2 concentrations are higher at the industrial station than at traffic stations, denoting that the industrial facilities operate during the entire day.

As expected, since NO_2 is converted to O_3 in a reaction catalyzed by sunlight (UV radiation), NO_2 (Figure 5) and O_3 (Figure 6) hourly profiles show an inverse relationship. O_3 concentrations peak is found in the early afternoon (between 1 and 4 pm), typically associated with local production of O_3 which reaches maximum levels with the highest solar radiation. O_3 concentrations start to decrease in the evening with the absence of sunlight, when the ozone production ceases, and the loss processes dominate. Minimum mean concentrations are reached around 6 - 9 am (due to O_3 scavenging by NO, during morning rush hours), and then start to increase, reaching their maximum in the afternoon, around 2 pm. The hourly peak in O_3 mean concentrations is the highest in Liguria (75.8 - 89.5 μg m^{-3}), followed by Aveiro (71.5 - 73.8 μg m^{-3}) and Ljubljana (69.6 μg m^{-3}). Liguria region shows indeed mean O_3 concentrations higher than the other case studies. This finding is particularly relevant during the nighttime period, when mean observed concentrations in Liguria are in the range 60 - 80 μg m^{-3} while in other regions are lower than 40 μg m^{-3}.

In general, the NO_2 weekly profiles of all the monitoring stations of all case studies, show lower concentrations during the weekend. These finding highlight that, as expected over urban areas, the air quality is marked by anthropogenic cycles. On contrary, O_3 concentrations are higher during weekends. This corresponds to the so-called weekend effect (Sicard et al., 2020). High concentrations of freshly emitted NO locally scavenge O_3, a process leading to formation of NO_2. Close to the sources this titration process can be considered as an ozone sink. In addition, high NO_2 concentrations deflect the initial oxidation step of VOCs by forming other products (e.g. nitric acid), which prevents the net formation of O_3. Because of these reactions, a decrease in NO_x can lead to an increase in O_3 at low VOC/NOx ratios, as is the case in cities. In this often-called VOC-limited regime, emission control of organic compounds is more efficient to reduce peak values of ozone pollution locally (Sicard et al., 2020). Due to the titration effect (reaction of O_3 with NO), lower O_3 are usually recorded by stations monitoring busy traffic and this pollutant is commonly not measured at traffic stations. Amsterdam network is an exception, with O_3 data at an air quality traffic station. As expected, O_3 concentrations are lower (with a delta of 3.9 μg m^{-3}, which corresponds to 9%) at this site than at the urban background ones.

In all the six cities/ regions, seasonal profiles indicate higher NO_2 concentrations in winter months. As previously mentioned, this behaviour is related, on the one hand, with the chemical reactions where NO_2 is involved (as sunlight triggers OH production) and, on the other hand, with extra NO_x emissions, during winter, from combustion processes for heating purposes.

Regarding the ozone seasonal profiles, higher mean concentrations are recorded during spring, specifically in May in Bristol (60.9 μg m^{-3}) and Amsterdam (62.5 μg m^{-3}), and in April and May in Aveiro region (61.7 μg m^{-3}). The ozone spring maximum is a common characteristic of many mid-latitudes regions in the northern hemisphere (Monks, 2000; Parrish et al., 2021; Zara et al., 2021). The physical and chemical mechanisms behind the spring maximum have been revised by Monks (2000) and Vingarzan (2004) and include both enhanced photochemistry in the free troposphere and stratospheric input. Indeed, O_3 concentrations in Europe are very much influenced not only by local and regional production but also by northern mid-latitudes background concentrations. From all the case studies, Aveiro, Bristol and Amsterdam are the ones located closer to the Atlantic. Their location and the dominant synoptic conditions, and the similarity of their spring mean concentrations (about 60 μg m^{-3}) point out the relevance of the high background O_3 concentrations received from the Atlantic to the high mean concentrations observed in these regions during spring (Auvray and Bey, 2005).

In Ljubljana and Liguria, the highest O_3 mean concentrations are registered from April to August/ September. During this period, mean O_3 concentrations are higher than 60 μg m^{-3} in Ljubljana and higher than 80 μg m^{-3} in Liguria. Although high mean concentrations are recorded during spring, the maximum mean concentrations occur in July and August in Ljubljana and Liguria, respectively. This behaviour indicates that in these regions, the observed O_3 concentrations have a strong contribution from local and/or regional ozone production, which is favoured by the summer higher atmospheric temperature leading to enhanced photochemical reactions and O_3 formation.

To complement our analysis on O_3 variability, Figure 7 presents the hourly variation of O_3 concentrations by season of the year, for the three cities/regions with the highest concentrations: Liguria, Aveiro and Ljubljana.

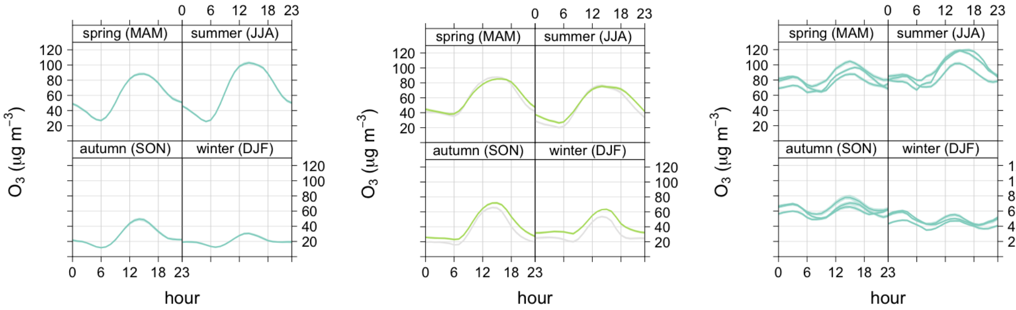

Figure 7: Hourly variability by season of the year of the O_3 concentrations observed between 2008 and 2017, for Ljubljana (a), Aveiro (b) and Liguria (c). The solid line shows mean concentrations while the shading shows the 95% confidence interval in the mean. Data from urban background stations is depicted in blue, suburban background in green and urban industrial in grey.

Ljubljana has a typical seasonal variation of the O_3 hourly profiles. During winter, concentrations are more constant during the day (low diurnal amplitude), and O_3 mean peaks achieve higher magnitude during autumn, spring and summer. The mean difference between daily minimum and maximum O_3 concentrations is higher during summer (about 70 μg m^{-3}) and lower during winter (about 15 μg m^{-3}). In Aveiro, this delta is similar during spring and summer (about 45-50 μg m^{-3}). However, as nighttime O_3 concentrations are lower during summer than during spring, the mean profile of the summer season shows lower concentrations than during spring.

The seasonal variation of the O_3 hourly profiles in Liguria is quite interesting. Observed concentrations during the nighttime period are in average much higher than in the other case studies, for all the seasons. During winter, the mean highest daily concentrations are recorded during the nighttime period. Liguria is located on the north-west of Italy, in the Mediterranean coast. Several studies have been published regarding the ozone concentrations in the Mediterranean Basin, which are relatively high when compared to other European areas (Cristofanelli and Bonasoni, 2009). In addition, high O_3 values are typical not only for ground level measurements in the Mediterranean, but in the entire boundary layer (Kalabokas et al., 2017). The transport of polluted air masses from Europe and other continents to southern Europe/ Mediterranean Basin, favours photochemical O_3 production in a region frequently characterised by high solar radiation intensity (Cristofanelli and Bonasoni, 2009). Within the Western Mediterranean area and based on a cruise ship measurements between April and October for two years, the Liguria region/ Gulf of Genoa was identified as one of the two main ozone "hot spots" (Velchev et al., 2011). The main cause of high O_3 levels in the Gulf of Genoa during this period (between April and October) was found to be outflow of polluted air from the Po Valley (with contributions also from the Genoa area) and, to a minor extent, from Marseille area as well. During specific meteorological events, the vertical motion of stratospheric air into the lower troposphere may represent a non-negligible source of background O_3. This stratosphere–troposphere exchange process exhibits a strong seasonality with a maximum in winter and spring and a minimum in summer (Sharma et al., 2017), and may partially explain the winter O_3 hourly profile plotted for Liguria (Figure 7).

3.2. Trend analysis

A trend analysis was performed to investigate the evolution registered and expected to the future. Long-term temporal trends of pollutant concentrations have been estimated with the TheilSen function of the OpenAir package for R (Carslaw and Ropkins, 2012), which quantifies monotonic trends in unit/year, and calculates the associated p value through bootstrap simulations. Trend is estimated for mean monthly values, and the 95% confidence interval of the slope is presented (see Table 3). In this analysis, data has been deseasonalized using the seasonal-trend decomposition procedure based on locally weighted scatterplot smoothing LOESS (Cleveland et al., 1990). The symbols shown next to each trend estimate relate to how statistically significant the trend estimate is: $p < 0.001 = ***$, $p < 0.01 = **$, $p < 0.05 = *$ and $p < 0.1 = +$.

Overall all the cities and regions have made significant progress over the last decade towards a clean air. This progress was mainly achieved due to the implementation of effective air quality management policies nationally and locally (e.g. the European legislation, such as air quality directive (EU, 2008)).

PM_{10} concentrations are decreasing in all the cities and regions where statistically significant trends were computed (Bristol, Amsterdam, Ljubljana, Sosnowiec and Aveiro). Similarly, $PM_{2.5}$ concentrations are also decreasing, despite the limited data available for the 10-years period, thus only Amsterdam, Sosnowiec and Aveiro show statistically significant evolution trends for this pollutant. Overall decreasing trends of PM concentrations may be associated with emissions reductions from the residential sector, as well as from industries. NO_2 concentrations are typically decreasing in all the case studies (exception for the background station in Aveiro). On contrary, O_3 mean concentrations are increasing in Bristol, Amsterdam, Ljubljana and Liguria region, but decreasing in Aveiro. NO_2 concentration trends are mostly associated with reductions on NO_x emissions from on-road transport. The increasing O_3 trends reflect the trends in NO_2 concentrations.

Table 3: Trend analysis for the 6 case studies. Trend estimates represent the change of concentrations per year, as an average over the entire period (from 2008 to 2017) and are shown in μg m^{-3} yr^{-1}. The symbols shown next to each trend estimate relate to how statistically significant the trend estimate is: $p < 0.001 = ***$, $p < 0.01 = **$, $p < 0.05 = *$ and $p < 0.1 = +$. n.s.s. stands for not statistically significant.

Case study	Monitoring station (type)	PM_{10}	$PM_{2.5}$	NO_2	O_3
Bristol	GB00203 (UT)			-0.51 [-0.69, -0.37] ***	
Bristol	GB00215 (UT)			-1.27 [-1.79, -0.71] ***	
Bristol	GB00270 (UT)			-1.04 [-1.44, -0.72] ***	
Bristol	GB00318 (UT)			-2.89 [-3.76, -1.89] ***	
Bristol	GB00463 (UT)			-0.73 [-1.13, -0.32] ***	
Bristol	GB0884A (UB)	-0.57 [-0.81, -0.36] ***	n.s.s.	-0.81 [-1.11, -0.47] ***	0.52 [0.12, 0.89] **
Amsterdam	NL00002 (UT)			-1.61 [-1.94, -1.29] ***	
Amsterdam	NL00003 (UB)				0.90 [-0.06, 1.68] +
Amsterdam	NL00007 (UT)	-1.23 [-1.62, -0.79] ***	-0.82[-1.35, -0.31] *	-2.02 [-2.48, -1.55] ***	
Amsterdam	NL00012 (UT)	-1.17 [-1.55, -0.78] ***	-0.67[-1.09, -0.14] *	-1.64 [-1.96, -1.33] ***	0.61 [-0.04, 1.33] +
Amsterdam	NL00014 (UB)	-0.82 [-1.23, -0.50] ***	-1.05 [-1.72, -0.45] ***	-0.76 [-1.02, -0.56] ***	1.24 [0.47, 1.98] **
Amsterdam	NL00016 (UB)	n.s.s.	n.s.s.		
Amsterdam	NL00017 (UT)	-1.43 [-1.81, -1.00] ***	-1.11 [-1.75, -0.56] ***	-1.13 [-1.37, -0.90] ***	
Amsterdam	NL00019 (UB)			-0.56 [-0.80, -0.38] ***	
Amsterdam	NL00020 (UT)			-1.44 [-1.75, -1.12] ***	
Amsterdam	NL00021 (UB)			-0.54 [-0.74, -0.39] ***	
Amsterdam	NL00022 (UB)			-0.39 [-0.61, -0.19] ***	
Amsterdam	NL00545 (UT)	-0.84 [-1.17, -0.52] ***			
Amsterdam	NL00546 (UI)	-1.29 [-1.90, -0.66] ***			
Amsterdam	NL00561 (RB)	-0.79 [-1.24, -0.35] ***	-0.72 [-1.35, 0.06] +		
Amsterdam	NL00565 (RB)	-0.60 [-1.04, -0.23] ***			
Amsterdam	NL00703 (RB)	-0.91 [-1.27, -0.59] ***	-0.96 [-1.66, -0.25] *	-0.57 [-0.87, -0.30] ***	
Amsterdam	NL00704 (UI)	n.s.s.	-0.88 [-1.44, -0.24] *	n.s.s.	

Table 4: Trend analysis for the 6 case studies. Trend estimates represent the change of concentrations per year, as an average over the entire period (from 2008 to 2017) and are shown in μg m^{-3} yr^{-1}. The symbols shown next to each trend estimate relate to how statistically significant the trend estimate is: $p < 0.001 = ***$, $p < 0.01 = **$, $p < 0.05 = *$ and $p < 0.1 = +$. n.s.s. stands for not statistically significant.

Case study	Monitoring station (type)	PM_{10}	$PM_{2.5}$	NO_2	O_3
Ljubljana	SI0003A (UB)	-0.76 [-1.08, -0.47] ***		-0.24 [-0.51, 0.05] +	0.50 [0.09, 0.81] *
Ljubljana	SI0058A (UB)	n.s.s.	n.s.s.		
Sosnowiec	PL0529A (UB)	-2.00 [-2.67, -1.37] ***		-2.20 [-3.29, -1.00] ***	
Sosnowiec	PL0567A (UT)	n.s.s.	-1.12 [-1.83, -0.17] **	n.s.s.	
Aveiro	PT02004 (SI)	-1.01 [-1.37, -0.65] ***	-0.70 [-0.98, -0.46] ***	-0.99 [-1.21, -0.77] ***	-0.91 [-1.47, -0.48] ***
Aveiro	PT02017 (UT)	-1.95 [-2.34, -1.56] ***		-0.59 [-0.87, -0.35] ***	
Aveiro	PT02018 (SB)	-0.57 [-0.86, -0.30] ***		0.30 [0.08, 0.57] *	-0.88 [-1.27, -0.48] ***
Liguria	IT0852A (UI)	n.s.s.		-1.59 [-2.36, -0.82] ***	
Liguria	IT0853A (UT)			n.s.s.	
Liguria	IT0854A (UB)	n.s.s.		-0.91 [-1.24, -0.50] ***	2.57 [1.51, 3.68] ***
Liguria	IT0856A (UB)				2.65 [1.78, 3.52] ***
Liguria	IT0858A (UB)		n.s.s.	-2.24 [-2.91, -1.51] ***	0.89 [0.28, 1.45] ***
Liguria	IT1698A (UT)			-1.63 [-2.37, -0.76] ***	
Liguria	IT1850A (UT)			-1.85 [-2.77, -0.81] ***	
Liguria	IT1884A (UT)			-2.15 [-2.91, -1.57] ***	
Liguria	IT1887A (UT)			-2.51 [-3.40, -1.56] ***	

These results are different from the trends estimated by Guerreiro et al. (2014) for the 93.15 percentile of maximum daily 8-h mean concentrations (as indicator for the EU target value for the protection of health), for the period 2002 – 2011. In that study, although 80% of the European monitoring stations did not reveal a clear trend, 18% registered a statistically significant decreasing trend, and 2% registered a significant increasing trend, most of them in the Iberian Peninsula (where Aveiro region is located). The difference in those results is probably related with the choice of the O_3 parameter (93.15 percentile of maximum daily 8-h mean concentrations against mean monthly concentrations) for the trend analysis.

The highest decreasing trends for the evolution of mean PM_{10} concentrations are estimated for Sosnowiec (-2.00 μg m^{-3}/yr at the urban background PL0529A station) and Aveiro (-1.95 μg m^{-3}/yr at the urban traffic PT02017 station). Those two cities were highlighted in the previous section, due to their high PM concentrations, in particular during winter late evening and night-time period. In Sosnowiec, pollutant concentrations decreased over the last decades, and according to Slingerland and Smith (2018) this was due to closure and modernisation of industries after the political and economic change of the 1990s. The main drivers of the observed reductions in concentrations in Sosnowiec, most of which have been largely driven by EU regulation, include cleaner power generation, lower increases in energy demand per household due to more efficient housing and appliances, improved road transport technologies and fuels, and reductions in industrial emissions measures, particularly regarding transport. To address the problem of local low-stack residential heating, subsidies for replacing the commonly used low-efficiency household stoves and boilers have been introduced (Slingerland and Smith, 2018). Decreasing trends in particulate matter concentrations over Portugal had already been shown by Gama et al. (2018), using observations from background air quality monitoring stations recorded from 2007 to 2016. According to this study, the main factor contributing to the PM10 decrease in urban areas is the decrease in the coarse PM (2.5-10 μm) concentrations.

The highest decreasing trends for the evolution of mean NO_2 concentrations are estimated for Bristol (-2.89 μg m^{-3}/yr at the BCC urban traffic GB00318 station) and Liguria (-2.51 μg m^{-3}/yr at the urban traffic IT1887A station).

In Bristol, the potential reasons behind those decreasing trends are the local policies included in the air quality action plans because of the designation of parts of Bristol as an Air Quality Management Area. The measures in the plan were almost entirely transport focused (Smith et al., 2017).

While, in Liguria region the downward trends have been the result of implemented measures to reduce industry emissions (EU legislation and the decommissioning of plants), harbour emissions (standards for fuels), and transport emissions (standards for diesel cars and traffic and mobility measures related to improving the railway, the metro, the bus fleet, and fostering electric mobility). These measures have helped bring down NO_2 concentrations, albeit not enough to comply with the EU limit values at all the traffic stations. The closing of different industrial plants, due to a lack of compliance with the regulation on air pollutant emissions, is likely an influential factor (Artola and Bolscher, 2018).

3.3. Pollution episodes

In the previous sections, air quality at the six case studies was characterized using averaged quantities. However, when assessing air quality, we are often interested in the extremes of these quantities, e.g., the concentrations of a given pollutant which may be harmful to the ecosystems and the human health. Thus, in this section, we will look at these extremes, using the short-term thresholds established in the European Air Quality Directive (Directive 2008/50/EC; (EU, 2008)) for the protection the human health. In addition, a detailed analysis of the fulfillment of the legislated limit values was performed for the six case studies and is available as Supplementary Material (SM 5.2).

For each case study, the observed concentrations above the short-term thresholds for the protection of the human health, defined for PM_{10}, NO_2, and O_3 (Table 5), were used to assess the occurrence of pollution episodes, and to characterize those episodes based on their magnitudes and duration. In this study, an episode is defined as a period of consecutive days (for PM_{10} and O_3) or hours (for NO_2) where a concentration above the threshold was observed in at least one station of the case study air quality monitoring network. This approach has however some limitations: for example, in a week with concentrations above the threshold for a given pollutant, if there is a day where no station recorded data, this period will be divided into two separate episodes.

Between 2008 and 2017, all the six case studies recorded PM_{10} concentrations above the daily limit value for the protection of the human health (50 $\mu g\ m^{-3}$). Those exceedances occurred mainly from October to March (Figure S3). However, in Bristol and Liguria region, although there are exceedances,

Table 5: Short-term limit values or target values established in the Directive 2008/50/EC for the protection of the human health for PM_{10}, NO_2, and O_3.

Pollutant	Time aggregation	Threshold (μg m^{-3})
PM_{10}	mean daily concentrations	50
NO_2	hourly concentrations	200
O_3	maximum daily eight-hour mean concentrations	120

the PM_{10} daily mean limit value was not exceeded more than 35 days per year in any station and thus there is compliance with the EU legislation (see Supplementary Material for details).

The frequency distribution graphs of the duration of PM_{10} episodes in days, for each case study, are presented in Figure 8. In this analysis, Amsterdam, Ljubljana, Sosnowiec and Aveiro are highlighted as the case studies with a higher number of PM_{10} episodes.

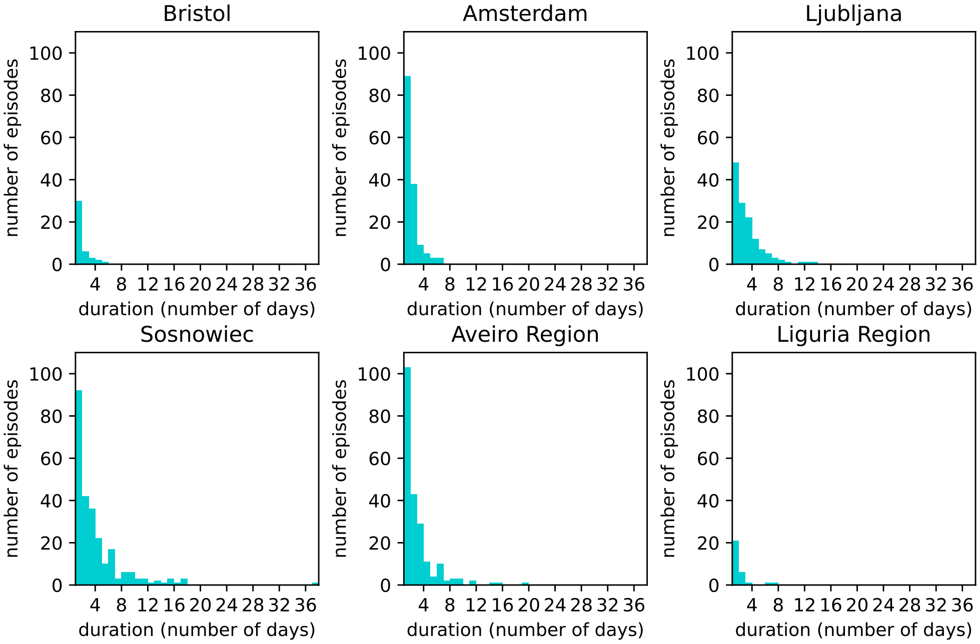

Figure 8: Characterization of the PM_{10} episodes which took place from 2008 till 2017, for the 6 case studies.

Three case studies recorded pollution episodes with mean PM_{10} concentrations above the limit value for the protection of the human health during 10 or more consecutive days: Sosnowiec (18 episodes), Ljubljana (3 episodes) and Aveiro region (5 episodes). The complete list of episodes with a duration of 10 or more days among the 10-year period is given as Supplementary Material (Table S1). Those episodes affected all type of monitoring stations existent in those three case studies (e.g., urban traffic and background in Sosnowiec, urban background in Ljubljana, and urban traffic, background and industrial in Aveiro region).

Contrary to what happens, for example, in Aveiro region, where the most persistent episodes were not recorded in the most recent years, in Sosnowiec the most persistent episode (37 consecutive days with PM_{10} exceedances), which is also the one where one of the monitoring stations recorded its highest value (306.2 μg m^{-3} at PL0567A station), took place in the latest study year (between 14 Jan and 19 Feb 2017). This evidence indicates that, despite the observed reduction in particulate matter mean concentrations through the study period (-2.00 μg m$^{-3} yr^{-1}$ for PM_{10} at PL0529A, as presented in Table 3), PM_{10} continues to be a pollutant of great concern at Sosnowiec. Another great PM_{10} episode that affected this city occurred between 31 Jan and 14 Feb 2012 (15 consecutive days with PM_{10} exceedances), when the values recorded in the PL0529A station reached 541 μg m^{-3}.

For NO_2, between 2008 and 2017, all the case studies but Ljubljana recorded concentrations above the hourly limit value for the protection of the human health (200 μg m^{-3}). Although Amsterdam and Sosnowiec did not record more than the 18 NO_2 exceedances per year permited in the AQD, the annual limit value for this pollutant (40 μg m^{-3}) was exceeded at specific traffic stations during several years of the study period in these two case studies (see the Supplementary Material for details). Both Ljubljana and Aveiro region are compliant with the two (annual and hourly) EU limit values for the protection of the human health. Contrary to PM_{10}, NO_2 exceedances do not show a marked seasonality (Figure S7).

The frequency distribution graphs of the duration of NO_2 episodes in hours, for each case study, are presented in Figure 9. Bristol is highlighted is this analysis due to the high number of NO_2 episodes, 2 of them which persisted for 12 hours, and another one for 10 hours.

Two case studies recorded pollution episodes with mean NO_2 concentrations above the limit value for the protection of the human health during 5 or more consecutive hours: Bristol (19 episodes) and Liguria region (2

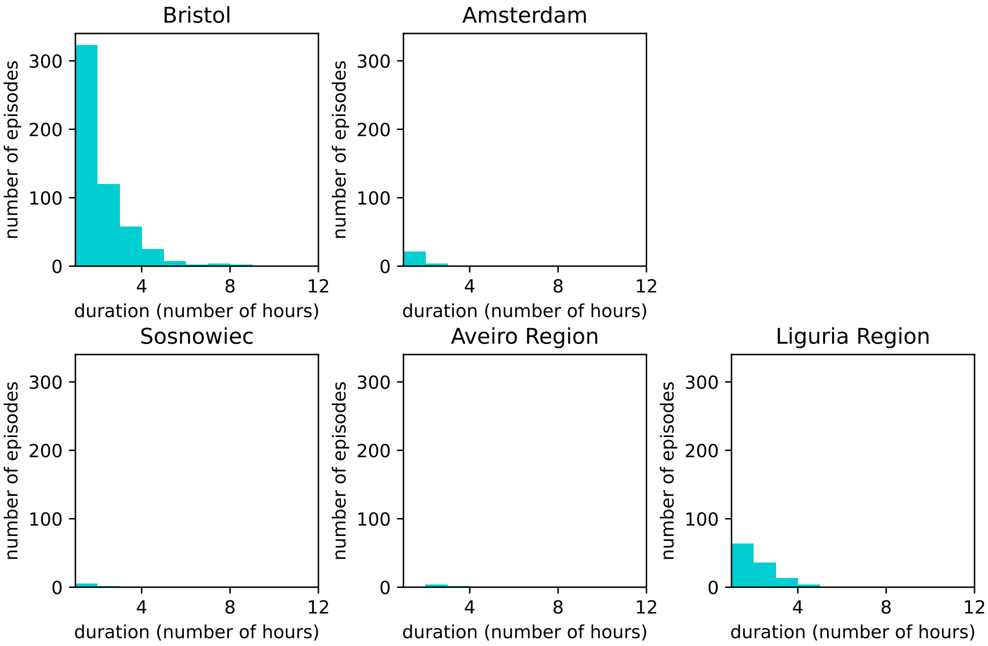

Figure 9: Characterization of the NO_2 episodes which took place from 2008 till 2017, for the 6 case studies.

episodes). The complete list of episodes with a duration of 5 or more hours among the 10-year period is given as Supplementary Material (Table S2).

During the two episodes highlighted in the Liguria region, which occurred in 20 Dec 2009 and 3 Dec 2012, exceedances have been recorded in one traffic station only. At Bristol, from the 19 episodes with 5 or more consecutive hours of exceedances, only one (2 June 2008) is associated with exceedances in more than one station. From those 19 episodes, which were registered in traffic stations, the ones which occur between 17 and 19 Mar 2009 and between 26 and 29 Aug 2015 can be considered exceptionally persistent, not only because of the number of hours with concentrations above 40 μg m^{-3}, but also because they occur during consecutive days.

All the five case studies with O_3 data recorded days with eight-hour mean concentrations higher than 120 μg m^{-3} during the study period. However, Bristol and Amsterdam present a low number of exceedances per year, showing compliance with the EU legislation. As expected, O_3 exceedances to the target value for the protection of the human health show a marked seasonality, with a higher number of exceedances from April to September (Figure S9). Although some case studies presented mean maximum concentrations during spring (e.g., Aveiro, Amsterdam and Bristol, see Figure 6), the maximum number of exceedances is observed during summer in all case studies.

For O_3, frequency distribution graphs of the duration of pollution episodes in days, are presented in Figure 10. Only the Liguria region recorded pollution episodes with maximum daily eight-hour mean concentrations above the target value for the protection of the human health during 10 or more consecutive days. In this region, 26 of these long-lasting episodes were recorded among the 10-year period, distributed within the study period (Table S3). An exceptional episode occurred from 20 Jun to 17 Sep 2016, when O_3 remained higher than 120 μg m^{-3} during 90 consecutive days. This episode was exceptional not only because of its persistence, but also regarding the observed concentrations: the three urban background monitoring stations that measure O_3 recorded the maximum concentrations (206.0, 245.0 and 216.0 μg m^{-3}) over the study period within this episode.

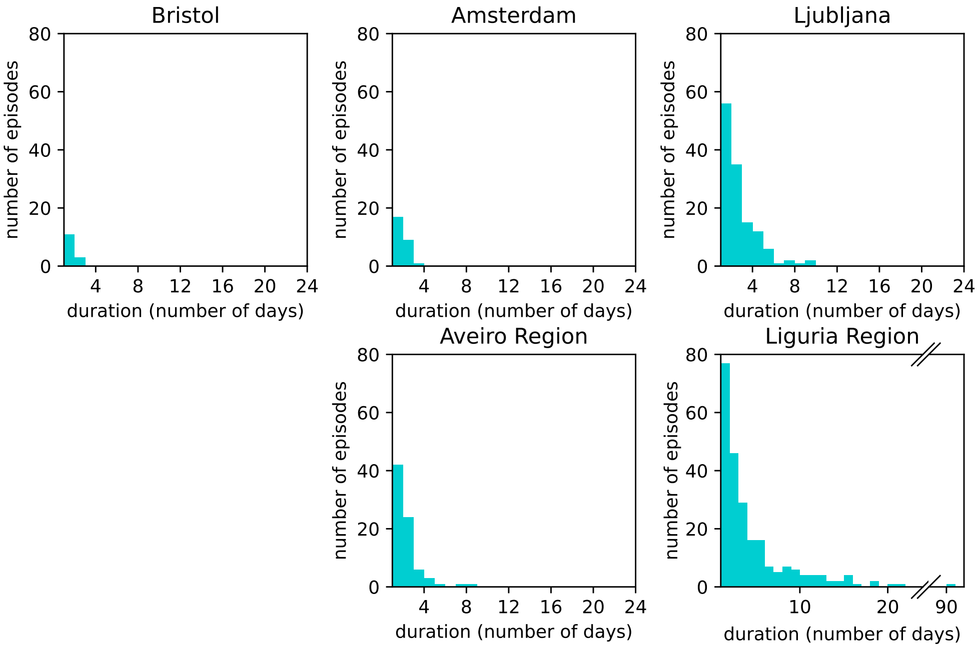

Figure 10: Characterization of the O_3 episodes which took place from 2008 till 2017, for the 5 case studies.

3.4. Summary analysis

To evaluate the air quality status of each case study, a set of criteria were established aiming to classify an air pollutant as indicator of historical air pollution issues. Table 6 summarizes the cities/ regions per pollutant and per type of station. The following criteria were established for PM_{10}, $PM_{2.5}$ and NO_2 concentrations: i) the annual mean concentrations above the yearly EU limit value in any station, and in at least two years (YR); ii) and/ or a number of exceedances registered above the allowed per year, in any station, and in at least two years (HD). The criteria established for O_3 concentrations is the number of exceedances registered for O_3 target value over the allowed per year (HD). Additionally, the occurrence of exceptionally persistent pollution episodes of PM_{10}, NO_2, and O_3 concentrations were also considered as an indicator (PE), using the criteria mentioned in sub-section 3.3 (e.g., episodes are classified as persistent when lasting for 10 or more consecutive days in the case of PM_{10} and O_3, and for 5 or more consecutive hours in the case of NO_2).

Table 6: Summary of the main air pollution problems identified for each case study and split by type of station, for the 10-years period. HD points out a problem of hourly or daily exceedances, YR represents a problem with exceedances to the annual mean limit value, and PE indicates the existence of persistent episodes.

	Type of station	PM_{10}	$PM_{2.5}$	NO_2	O_3
Bristol	traffic			YR HD PE	
	background				
Amsterdam	traffic			YR	
	background				
	industrial				
	background				
Ljubljana	background	HD PE			HD
Sosnowiec	traffic	YR HD PE	YR	YR	
	background	YR HD PE			
Aveiro region	traffic	HD PE			
	background	HD PE			
	industrial	HD PE			
Liguria region	traffic			YR HD PE	
	background			YR	HD PE
	industrial			YR	

PM_{10} is classified as a critical pollutant, according to the established criteria, in Sosnowiec, followed by Ljubljana and Aveiro. These three cities are facing different levels of severity regarding this pollutant, and thus, the future policies and measures to control this problem require distinct levels of ambition.

$PM_{2.5}$ is highlighted as a serious issue only in Sosnowiec. Nevertheless, besides the spatial limitations of the stations monitoring $PM_{2.5}$, together with the limited available measurements, there is also time-drawbacks due to the fact that several air quality stations have only started to monitor $PM_{2.5}$ concentrations in the recent years. Additionally, the AQD only sets annual thresholds for $PM_{2.5}$, and there is no criteria to assess short-term pollution for this pollutant. However, PM_{10} is only highlighted as a problem in Aveiro region and Ljubljana, when considering the HD and PE indicators, and thus if similar indicators were available for $PM_{2.5}$ probably a similar pattern would be found. All these factors may indicate that $PM_{2.5}$ concentrations may be of concern on other cities, together with the fact that all the cities do not met the WHO recommended limit values.

Monitored NO_2 concentrations along the 10-years period denote a pollution problem, mainly associated with road-traffic emissions in Bristol, Amsterdam, Sosnowiec and Liguria. Among those cities, Bristol and Liguria region had the highest number of exceedances to the hourly limit value.

Liguria presents also a pollution problem for O_3 concentration, as well as Ljubljana. In addition, both cities present an increasing trend for the 10-years period.

It is of note the limitations of this analysis associated with the spatial representativeness of the available measurements, since each city/region has a very different number of air quality stations with valid measurements, and therefore a city with a greater number of stations, may potentially have more air pollution issues highlighted by the available measurements. While no measurements available may hide some important air pollution problems.

The pollution problems identified for each case study are independent on the type of station, except in Liguria region where the traffic stations indicate issues related with exceedances to the annual mean of NO_2 concentrations, together with exceedances to the hourly limit, as well as the existence of persistent episodes, while the background and industrial stations highlight a problem of exceedances only to the annual limit.

4. Conclusions

This work aimed to assess the air quality status of six European cities for the period from 2008 to 2017, identifying the main and common air quality challenges between these different cities/ regions, and its main priorities in terms of pollutants and mitigation strategies. The specific context of the different regions and cities of Europe and their complex systems dynamics are considered in this analysis. The results are discussed considering the hourly, daily and seasonal variation of concentrations; a trend analysis providing the evolution during the 10-years period; and the number of persistent air pollution episodes, followed by the fulfillment of the EU legislated limit values, together with the stricter, but still voluntary, WHO guideline values.

Each city/ region faces different issues and causes of air pollution, but all these case studies have been working on to improve their air quality. In Bristol there have been strong downward trends in many air pollutants, but the levels of NO_2 remain persistently high, with transport as the key contributor. PM on the other hand is not widely monitored in Bristol, but background levels are under limit values. Similarly, the main sources of air pollution in Amsterdam are traffic, for NO_2. Decreasing concentration levels point to some success of Amsterdam air quality policies in recent years. PM_{10} exceedances are a seasonal pollution problem in Ljubljana, with the main particulate matter sources attributed to residential heating, which is still significantly outdated in some parts of the city, where households still heat with burning wood and biomass during winter (Slingerland et al., 2018b). The most pressing issue for air quality within Sosnowiec is particulate matter (PM_{10} and $PM_{2.5}$), linked with the use of inefficient heating systems, together with poor quality fuels, in winter (Slingerland and Smith, 2018). On the other hand, NO_2 limit values are also exceeded in Sosnowiec. Air quality in the Aveiro region is relatively good, due to an overall relatively low population density in the region, and an open landscape in a maritime climate. PM_{10} (particularly exceedances to the EU daily limit value) and O_3 exceedances do occur occasionally. Wood burning for residential heating and industrial activities are important contributors to air polluting emissions (Slingerland et al., 2018a). Exceedances of NO_2 and O_3 concentrations are still problematic in Liguria region, with road transport, industrial plants and port activities being the main contributors to these problems.

Sosnowiec is the only city presenting no compliance with the EU AQ objectives for $PM_{2.5}$ concentrations, considering the reduced measurements available in each case study and their potential lack of spatial representativeness of the entire study areas. However, assuming a transition towards the establishment of the WHO stricter, but still voluntary, guideline values, as the formal EU legal limits, the cities and regions will move to an overall situation of no compliance with the EU legislation, exception made for Liguria Region. Therefore, monitoring networks, particularly the stations measuring $PM_{2.5}$, should be designed to consider the optimum data that it can generate for public health purpose, and not only for compliance with the AQD.

Nowadays, in European urban areas, the current levels of atmospheric emissions, the growing of epidemiological evidence on the health effects of air pollution, the threat of fines by the European Commission towards Member States and the high-profile court cases taken forward by distinct organizations against Member States Governments has raise the media and political profile of air pollution. Together with a recent growth of citizens' sciences activities, where citizens are measuring air quality by themselves. Recently, low-cost sensors were made available to everyone, which implies that every citizen in any city will be able to monitor air quality levels in the surroundings of their home, or their work place, or any other place. This democratic access to monitoring devices could contribute to strengthen air quality management practices, also considering data from citizen science. Nevertheless, this will require from local stakeholders, decision- and policy-makers a strong investment on training to provide citizens with the required knowledge to understand what they are measuring. In summary, the methodology we propose in this paper represents an useful approach, which could support any local stakeholder, decision- and policy-maker to start processes of citizens' engagement in their city or region. The fact that we use data available on the European database allows everyone to have access to data to reproduce a similar analysis to any European city, and which could be adapted and adopted by any global city.

The main findings of this paper highlight the overall decreasing trends of most of the analyzed pollutants during the past decade. These achievements were possible due to a set of air quality policies technological-centered, which have been implemented in Europe, and in each case study, during the last decades. On the other hand, a considerable number of implemented policies were not followed by stronger improvements on air quality and there is still severe air pollution problems within the European regions and cities, as highlighted by the main findings of this study. In addition, most of the identified air pollution problems over the case studies have a strong link with citizens' daily behaviour, practices and activities. Therefore, air quality policies aiming to reduce air pollutant emissions further should focus on changing individual and societal behaviour in parallel with technological changes. People and their behaviour need to be included in the way air quality is managed and communicated. This assessment provides a basis to better understand the role of citizens behaviour in the generation of pollution allowing for a realignment of policy process to go beyond the traditional techno-centric approaches to manage air quality.

The air quality assessment provided in this paper should be the first step of a citizens engagement process. With this data analysis, citizens will start to understand their recent historical air quality, from the past decade. Therefore, this paper presents a comprehensive methodology suited for any air quality assessment that may be performed for any city, taking advantage of the data available from the official air quality monitoring networks, and using this data to inform citizens. Therefore, the answer to the research question which motivated this paper is that it is possible to support air quality management with a citizen-centered approach through a historical air quality assessment study, and vice-versa, it is essential to support citizen-centered approaches with historical air quality information. The knowledge on the recent historical air quality levels, through a systematic approach, will be a key contribution to improved air quality city policies in the future, as policies, not only local, but also national and European policies, to date have failed to successfully engage citizens because, unlike technological solutions, people and their behaviour are not obviously present in the way that air quality is managed and communicated.

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5. Supplementary Material

5.1. Description of the six EU case studies

5.1.1. Bristol, UK

Bristol is located in the South West of England with a population around 450,000 and an area of 110 km^2, resulting in a population density around 4000 hab/km^2. The population was found to be mainly concentrated in the central area of the considered domain. Bristol's economy is highly service based, although the industrial sector is still strong. The education sector plays a major role in the economy of the city (Smith et al., 2017). The climate is Temperate Oceanic (Cfb), classified by the Köppen-Geiger Climate Classification System (Peel et al., 2007). The Department for Environment Food & Rural Affairs is the government department responsible for safeguarding the natural environment over the UK. The AURN is the UK's largest automatic monitoring network and is the main network used for compliance reporting against the AQD. In this paper, the dimensions of the study area for Bristol are 20 km by 20 km, and so there is only one urban background station located in the city. In addition, as part of the Local Air Quality Management duties, Bristol City Council measures some air quality concentrations in the city, at five continuous analyser sites: an urban background site, and the remaining mainly influenced by road traffic. The local authority stations monitor only Nitric Oxide (NO), Nitrogen Oxides (NOx) and NO_2 (Smith et al., 2017). For the considered stations in Bristol the number of stations per 100,000 inhabitants is 1.3. The location of AQS is presented in Figure 1a). Furthermore, an extensive passive diffusion tube network is used for indicative monitoring of NO_2.

5.1.2. Amsterdam, Netherlands

Amsterdam is the capital of the Netherlands and is in the western part of the country, in the province of North Holland. Amsterdam is the most populous city of the Netherlands with a total population of 834,713 inhabitants and a high population density around 4700 hab/km^2. Amsterdam's economy is strongly influenced by the harbour, the airport, tourism, the creative industries, and financial and business services (Slingerland et al., 2017). The population was found to be mainly concentrated in the central area of the considered domain. Amsterdam has a Temperate Oceanic climate (Cfb), following the Köppen-Geiger Climate Classification System (Peel et al., 2007).

The Dutch National Institute for Public Health and the Environment is responsible for the air quality monitoring and assessment. In this paper, the dimensions of the study area are 25 km by 20km, where it exists a vast monitoring network with 17 monitoring stations, 6 of which are classified as urban traffic, 2 urban industrial, 6 urban background and 3 rural background. For the considered stations in Amsterdam the number of stations per 100,000 inhabitants is 2, the highest of the six cases. The location of these AQS is presented in Figure 1b.

5.1.3. Ljubljana, Slovenia

Ljubljana is the capital and it is located in the central part of Slovenia. The city, with an area of 275 km^2, has a total population of 288,919 inhabitants, with a population density around 1075 hab/km^2. In Ljubljana industry, services and tourism are the main further dynamic sectors generating employment and economic activity (Slingerland et al., 2018b). The population was found to be mainly concentrated in the central area of the domain. Ljubljana's climate is classified, by Köppen-Geiger Climate Classification System, as a warm-summer humid continental climate (Dfb) (Peel et al., 2007). In Slovenia, the Environmental Agency of the Republic of Slovenia is responsible for the air quality protection, in terms of monitoring of the outdoor air quality, collecting emission data and performing administration procedures for air quality protection. In this paper, the dimensions of the study area are 20 km by 20km, which covers the urban area of Ljubljana where two urban background stations exist, belonging to the national automatic air pollution monitoring network. For the considered stations in Ljubljana the number of stations per 100,000 inhabitants is 0.7, the lowest of the six cases. The location of the AQS is presented in Figure 1c.

5.1.4. Sosnowiec, Poland

Sosnowiec is in the southern part of Poland. The city has a total population around 206,000 and an area of 91 km^2, resulting in a population density around 2376 hab/km^2. In Sosnowiec, similar to the whole Silesian region, industry plays the major role in the economic sector. Although, a transition to a more service-oriented economy can be noticed in recent years (Slingerland and Smith, 2018). For the considered domain the population was found to be more equally distributed. The climate is classified, by Köppen-Geiger Climate Classification System, as a warm-summer humid continental climate (Dfb) (Peel et al., 2007). In Poland, the air quality monitoring system is set under the State Environmental Monitoring established by the Inspection of Environmental Protection since 1991. In this paper, the dimensions of the study area in Sosnowiec are 20 km by 20km, which covers 2 monitoring stations, of which one is influenced by traffic and the other is classified as urban background. For the considered stations in Sosnowiec the number of stations per 100,000 inhabitants is 1. The location of the AQS within the study area is presented in Figure 1d.

5.1.5. Aveiro region, Portugal

The Aveiro region is in the central part of Portugal, over the coastal line, with a total population of 370,394. The Region consists of 11 municipalities, namely Águeda, Albergaria-a-Velha, Anadia, Aveiro, Estarreja, Ílhavo, Murtosa, Oliveira do Bairro, Ovar, Sever do Vouga and Vagos. The Aveiro Region has a total area of 1,693 km^2 resulting in a population density of 218.1 hab/km^2, being Aveiro, Ovar and Águeda the most populated municipalities. In the region, industry represents a major part of the economy of the region, while the port also has an important role, followed by tourism and services. Agriculture plays a prominent role mainly in the inland parts of the region (Slingerland et al., 2018a). The region's climate is classified, by Köppen-Geiger Climate Classification System, as a warm-summer mediterranean climate (Csb) (Peel et al., 2007). In this paper, the dimensions of the study area for the Aveiro region are 40 km by 55km. Within the region there are three AQS managed by the Regional Coordination and Development Commission of the Center, namely: (i) Aveiro (urban traffic); (ii) Ílhavo, (suburban background); and (iii) Estarreja, (suburban industrial). For the considered stations in Aveiro Region the number of stations per 100,000 inhabitants is 0.8. The location of the AQS is presented in Figure 1e.

5.1.6. Liguria region, Italy

Genoa is the capital of the Ligura region located on the north-west of Italy. The metropolitan area of Genoa is the biggest province of the Liguria region, with an area of 1,838 km^2 and a total of 855,834 inhabitants in 2011 (population density of 465.5 hab/km^2). The strongest economic sectors in Liguria are services (mainly tourism), and then industry. The Port of Genoa is also a key local and regional source of income (Artola and Bolscher, 2018). The population is mainly concentrated in the coast. The region has a hot-summer mediterranean climate (Csa) in the Köppen-Geiger Climate Classification System (Peel et al., 2007). The Regional Agency for the

Protection of the Ligurian Environment is responsible for the environmental matters in the Liguria Region. Being responsible for the planning, protection and management of the environment and nature. In this paper, the dimensions of the study area for this region are 25 km by 15km, which includes a vast air quality monitoring network distributed over the study area, with 5 urban traffic, 1 urban industrial and 3 urban background stations. For the considered stations in Liguria Region the number of stations per 100,000 inhabitants is 1.1. The location of the AQS is presented in Figure 1f.

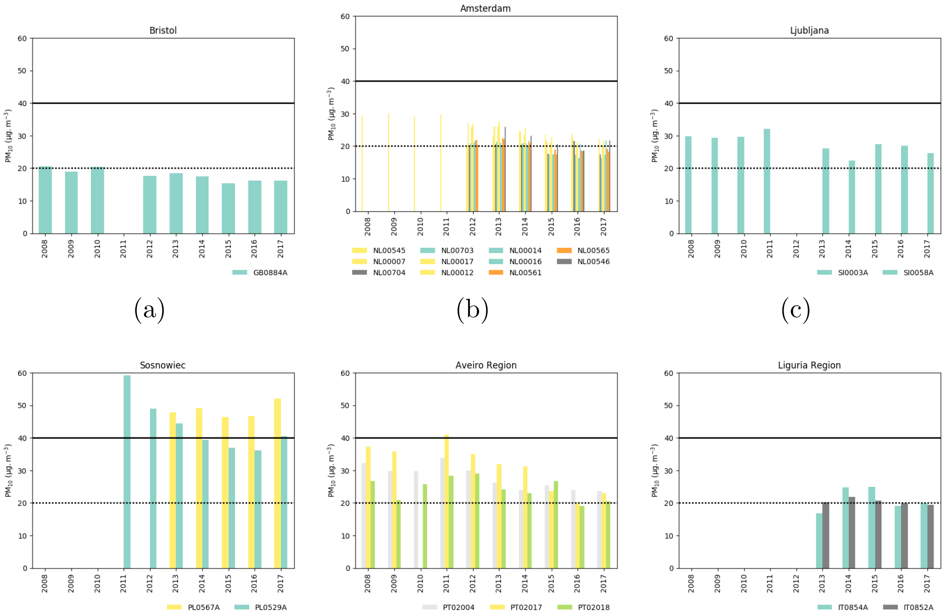
5.2. Legislation fulfilment

The following figures show the annual mean values for PM10, PM2.5, NO_2, and O_3, together with the exceedances observed for the daily limit value established for PM10 concentrations, the hourly limit value established for NO_2 concentrations, and the O_3 target value for the protection of human health, for all the 6 case studies. The comparison between the measurements in the monitoring stations and the EU objectives allows the characterization of the air quality status in each ClairCity Pilot. The WHO guideline values were also considered, since the authors believe that the current legislation could be updated to be aligned with these more strict guidelines. In the figures below, the color of each bar represent the classification type of the stations (e.g. blue for the urban background, green for the suburban background, yellow for the urban traffic, grey for the urban industrial and orange for the rural background).

5.2.1. PM10 concentrations

Figures S1, S2 and S3 show the annual mean concentrations, the exceedances observed for the EU daily limit value and the distribution per month of these exceedances, respectively, for PM10, for all the 6 case studies.

Bristol has only one urban background station measuring PM10 concentrations. This background station has recorded no exceedances to the yearly EU limit value, and two exceedances to the yearly guideline value from the WHO (in 2008 and 2010; the records have only slightly exceeded the guideline value). The minimum value (of 15.4 μg.m^{-3}) of PM10 annual mean concentrations was measured in 2015, while the highest was recorded in 2008 (Figure S1a). The station also recorded exceedances for the PM10 EU daily mean limit value, but never more than 35 days per year. 2008 was the year with the highest number of days with exceedances (Figure S2a).

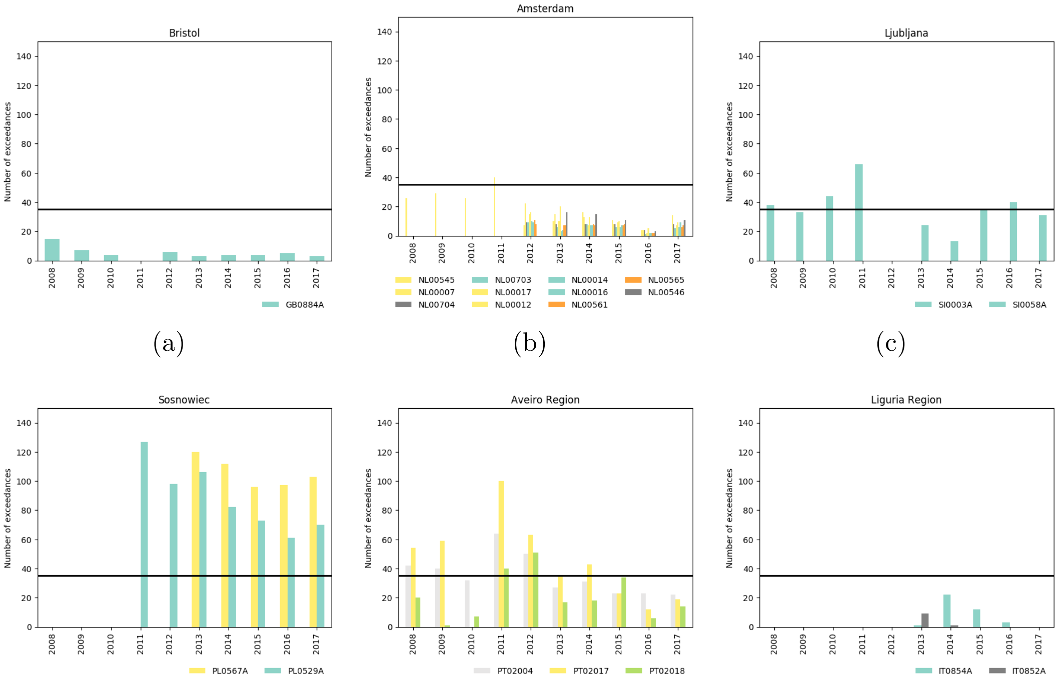


(a) (b) (c)

(d) (e) (f)

Figure S1: PM10 annual mean concentrations, from 2008 till 2017, for the 6 case studies. The dashed line represents the WHO air quality guideline value, while the solid line represents the PM10 yearly EU limit value for the protection of human health, as defined by the Directive 2008/50/EC.

Amsterdam is the ClairCity pilot city with the greatest number of stations monitoring PM10 concentrations, with a set of 9 air quality stations with valid records, since 2012 until 2017 (3 traffic, 2 urban background, 3 rural background, and an urban industrial stations). In addition, there is an urban industrial station with valid records that has started to measure in 2013 till 2017, and an urban traffic station with valid records from 2008 until 2016. All the monitoring sites within the study area of Amsterdam have monitored PM10 annual mean concentrations in compliance with the PM10 yearly EU limit value. While when considering the stricter, but still not mandatory, WHO guidelines, the urban traffic stations tend to be above the yearly guideline value, exception for the station NL00017 in 2016. Particularly, the urban traffic stations are the ones registering the highest PM10 annual mean concentrations (Figure S1b). Regarding the exceedances for the PM10 EU daily mean limit value, the 35 allowed days were only surpassed



(a) (b) (c)

(d) (e) (f)

Figure S2: Number of exceedances registered for PM10 daily mean limit value for the protection of human health, registered per calendar year from 2008 till 2017, for the 6 case studies. The solid line represents the number of exceedances of 35 allowed per year, as defined by the Directive 2008/50/EC.

in a urban traffic station in 2011. Although, there is days with exceedances in multiple other stations (Figure S2b).

Ljubljana has two air quality stations classified as urban background stations with valid data (station SI0003A recorded valid measurements from 2008 to 2011, while the station SI0058A started to have valid measurements in 2013 until 2017). The measurements from both stations point out compliance with the yearly EU limit value during the entire analysis period. However, annual mean concentrations are always above the WHO yearly guideline value. PM10 annual mean concentrations are lower in the more recent five years (Figure S1c). Exceedances for the EU daily mean limit value were recorded on more than 35 days per year in 2008, 2010, 2011, and 2016 (Figure S2c).

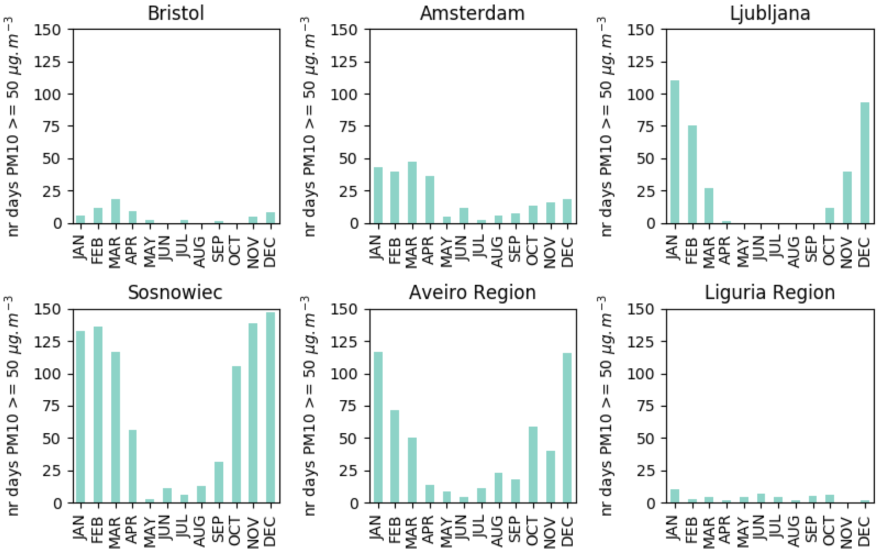


Figure S3: Monthly distribution of the days where PM10 concentrations above the threshold for the protection of human health were observed in at least one monitoring station in each case study, from 2008 till 2017.

Despite the reduced number of air quality stations available within the study area, together with the lack of data before 2010, Sosnowiec is the city with the most critical PM10 pollution problem. The urban traffic station, which had valid measurements from 2013 till 2017, had registered annual mean concentrations always above the yearly EU limit value. While the urban background station (valid measurements from 2011 till 2017), had recorded PM10 annual mean concentrations above the yearly EU limit value between 2011 and 2013, and again in 2017 (Figure S1d). Similarly, both stations have exceeded the EU daily mean limit value on more than 35 days per year, in all the years with valid measurements. The highest number of 120 days with exceedances was recorded in the traffic station in 2013, while 127 days with exceedances were recorded in the background station in 2011 (Figure S2d).

In Aveiro Region there are only three stations monitoring PM10 concentrations, despite being the larger study area. In this Region, the PM10 annual mean concentrations are compliant with the yearly EU limit value, except the exceedance registered in 2011 on the traffic station. However, the annual mean concentrations are always above the WHO guideline value, except for one of the stations in 2016 (Figure S1e). The industrial station exceeded the EU daily mean limit value on more than 35 days in 4 years, while the traffic station exceeded this limit on more than 35 days in 5 years. The suburban background station exceeded the daily limit value on more than 35 days in 2011, and 2012 (Figure S2e). Despite the compliance with the yearly EU limit value, all the three stations registered exceedances for the PM10 EU daily mean limit value, during the entire period, except the urban traffic station in 2010. This indicates a problem of PM10 pollution, mainly associated with episodes, detailed in subsection 3.2.

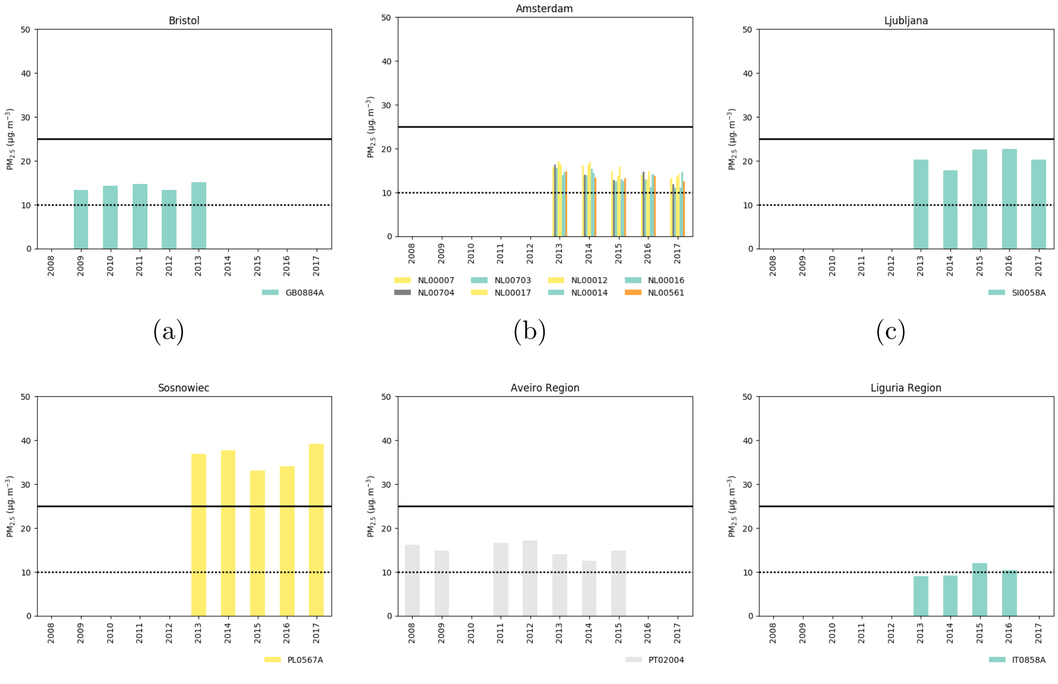
In Liguria Region, only two out of the nine air quality stations available have valid measurements for PM10 concentrations, an urban background station and an urban industrial station. Both stations comply with the yearly EU limit value, for the entire period. However, the PM10 annual mean concentrations are above the WHO yearly guideline value in 2014 and 2015 on the background station, and between 2013 and 2015 on the industrial station (Figure S1f). The EU daily mean limit value was not exceeded on more than 35 days per year in either station (Figure S2f).

5.2.2. PM2.5 concentrations

Figure S4 shows the annual mean PM2.5 concentrations, for all the 6 case studies.

The urban background in Bristol is the only station measuring PM2.5 concentrations, within the study area, with valid measurements only during half of the analysis period. In Amsterdam, there are eight air quality stations monitoring PM2.5 concentrations, all of them with valid records also only during half of the analysis period. In Ljubljana, one of the urban background stations is measuring PM2.5 concentrations, while within the Aveiro Region, the suburban industrial station was monitoring PM2.5 concentrations. In all these four cities/ regions (Figures S4a, S4b, S4c and S4e), PM2.5 annual mean concentrations were compliant with the yearly EU limit value, but exceeding the yearly guideline value from the WHO in all the years and stations with valid measurements.

The urban traffic station in Sosnowiec is measuring PM2.5 annual mean concentrations exceeding the yearly EU limit value, and thus exceeding also the yearly guideline value from the WHO (Figure S4d). On contrary, in Liguria Region the urban background station monitoring PM2.5 concentrations, from 2013 till 2016, indicates compliance with the yearly EU limit value in all the years. The measurements were also compliant with the yearly guideline



(a) (b) (c)

(d) (e) (f)

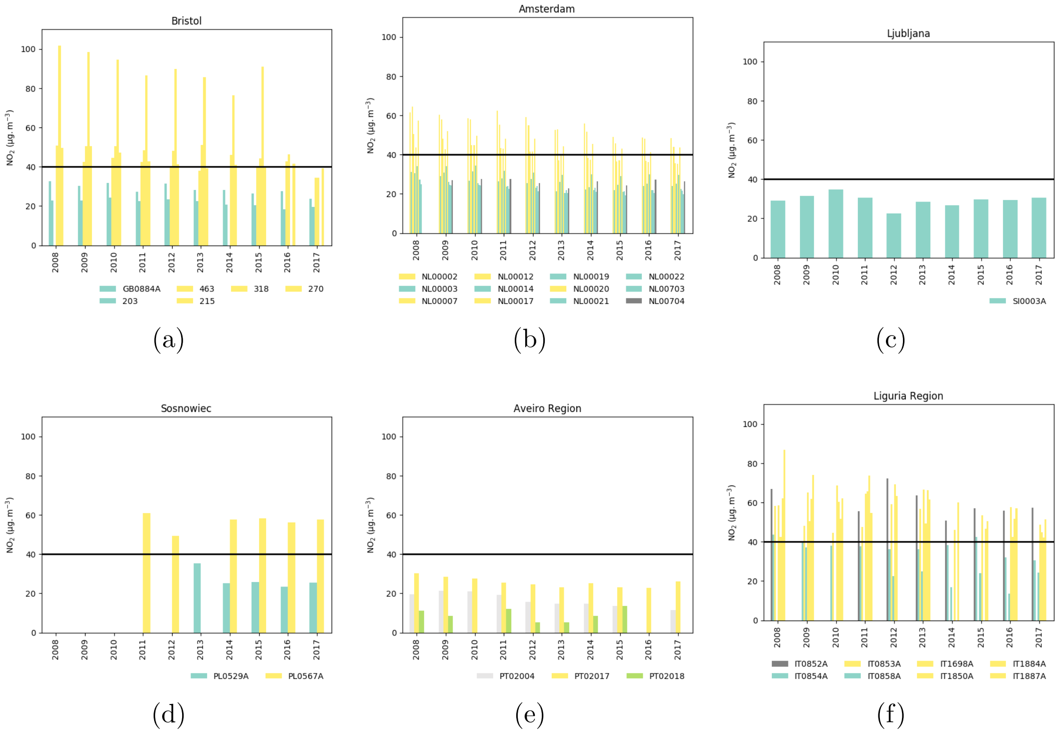
Figure S4: PM2.5 annual mean concentrations, from 2008 till 2017, for the 6 case studies. The dashed line represents the WHO air quality guideline value, while the solid line represents the PM2.5 yearly EU limit value for the protection of human health, as defined by the Directive 2008/50/EC.

value from the WHO in 2013 and 2014, and only slightly above the guideline value for 2015 and 2016 (Figure S4f).

5.2.3. NO_2 concentrations

Figures S5, S6 and S7 show the annual mean concentrations, number of exceedances registered for EU hourly limit value for the protection of human health and the distribution per month of these exceedances, respectively, for NO_2, for all the 6 case studies.

The urban background station from the national monitoring network in Bristol, together with five additional automatic stations (an urban background and four traffic stations from the local authority), are measuring NO_2 concentrations, within the study area. The NO_2 annual mean concentrations monitored by the two background stations are compliant with the



(a) (b) (c)

(d) (e) (f)

Figure S5: NO$_2$ annual mean concentrations, from 2008 till 2017, for the 6 case studies. The solid line represents the NO$_2$ yearly EU limit value for the protection of human health, as defined by the Directive 2008/50/EC.

yearly EU limit value during the entire period (Figure S5a). These background stations have only recorded each an exceedance of the EU hourly limit value for the NO$_2$ concentrations (Figure S6a). On contrary, the traffic stations often indicate situations of no compliance with the legislated limit values. Particularly, the Rupert Street station, which is located on a busy traffic thoroughfare in the city centre, is exceeding the yearly EU limit value in all the years with valid measurements. This station is also exceeding the EU hourly limit value always on more than eighteen times a calendar year, with the highest number of 284 days in 2008. The three remaining traffic stations are often no compliant with the yearly limit value [3], while they tend

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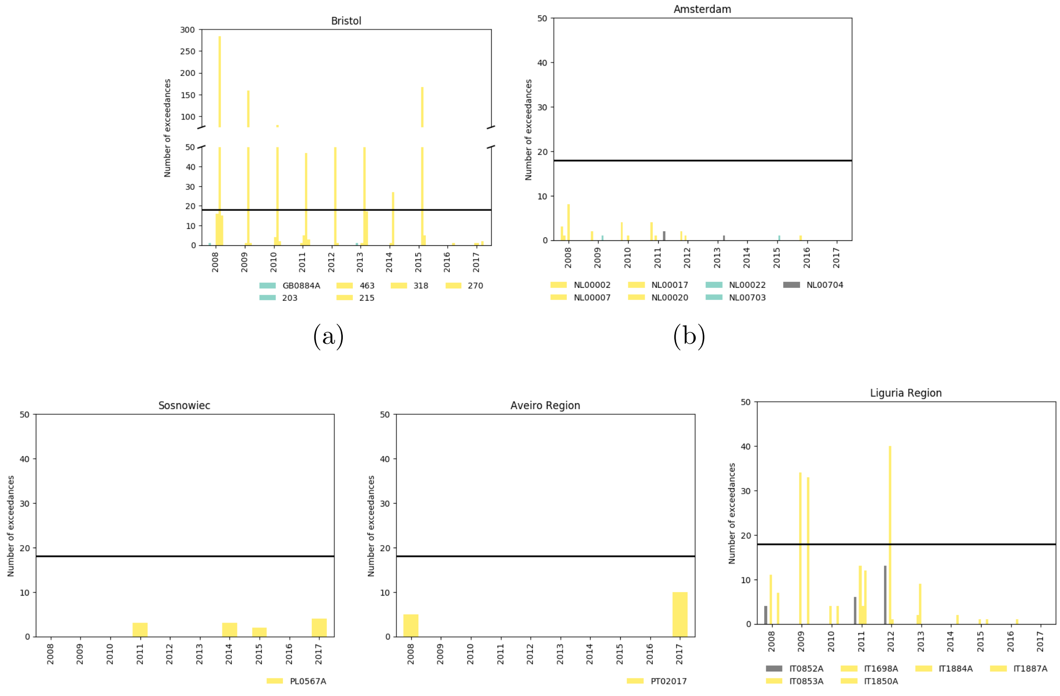
[3] Fishponds Road station records 4 out of 8, Parson Street School station records 9 out of 10, and Wells Road station records 7 out of 10 years exceeding the yearly limit value.



(a) (b)

(c) (d) (e)

Figure S6: Number of exceedances registered for NO_2 hourly limit value for the protection of human health, registered per calendar year from 2008 till 2017, for the 6 case studies. The solid line represents the number of exceedances allowed per year, as defined by the Directive 2008/50/EC.

to be compliant with the hourly limit value, with only a few acute situations (e.g. in 20018 the Parson Street School station exceeded on 16 days the limit value; while the Wells Road station recorded 15 and 17 days exceeding this limit in 2008 and 2013, respectively). In addition, an existing network of NO_2 diffusive samplers distributed over the urban area is used for indicative monitoring of ambient nitrogen dioxide in the city. An analysis of these measurements indicates NO_2 exceedances to the yearly EU limit value, denoting traffic-related NO_2 pollution.

Amsterdam has valid measurements of NO_2 concentrations in five urban traffic stations, and five urban background stations for the 10-years period. In addition, one rural background and an urban industrial stations have valid measurements from 2009 till 2017. All the five urban background stations,

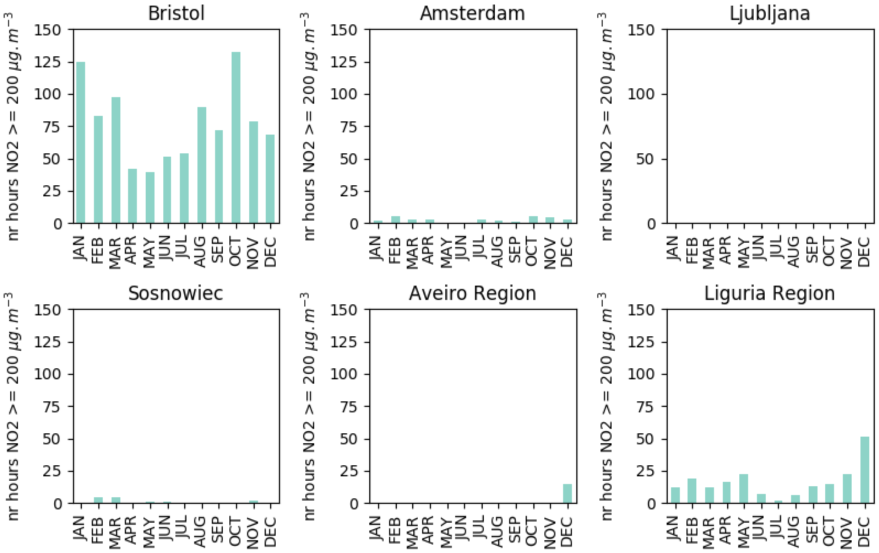


Figure S7: Monthly distribution of the hours where NO_2 concentrations above the threshold for the protection of human health were observed in at least one monitoring station in each case study, from 2008 till 2017.

together with the rural background and the urban industrial stations, are compliant with the yearly EU limit value during the entire period. On contrary, three of the traffic stations are not compliant with the yearly EU limit value for all the period, while the remaining two traffic stations are not compliant with the yearly EU limit value only from 2008 till 2012 (Figure S5b). Despite few exceedances, all the stations in Amsterdam are compliant with the EU air quality objectives, which establishes the limit value of 200 μg.m^{-3} for NO_2 concentrations not to be exceeded on more than eighteen times a calendar year (Figure S6b).

In Ljubljana, there are only valid measurements in one of the urban background stations, where NO_2 annual mean concentrations are fully compliant with the yearly EU limit value during the entire period (Figure S5c). In addition, this station has recorded no exceedances of the EU hourly limit value.

In Sosnowiec, the NO_2 annual mean concentrations measured in the urban background station are compliant with the yearly EU limit value for all the 5 years with valid data. While, in the traffic station, the NO_2 annual mean concentrations are exceeding the EU limit value in all the 6 years with valid data (Figure S5d). In addition, the urban traffic station has recorded exceedances of the EU hourly limit value, always on more than eighteen times a calendar year (Figure S6c).

In Aveiro, the NO_2 annual mean concentrations measured in all the three stations are compliant with the yearly EU limit value for all the years with valid data (Figure S5e). Furthermore, the background and the industrial stations are also compliant with the hourly EU limit value. However, the urban traffic station has recorded few exceedances of the EU hourly limit value (Figure S6d).

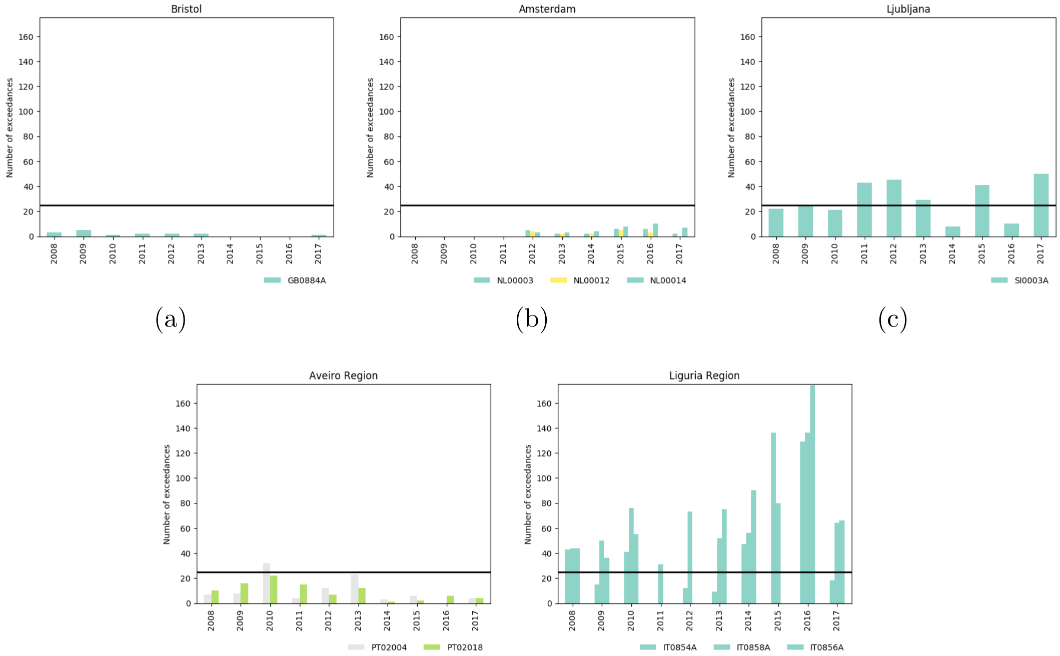
The NO_2 annual mean concentrations measured in all the stations within the Liguria region are always exceeding the yearly EU limit value for all the years with valid data (Figure S5f). Two of the traffic stations with valid data present the highest number of exceedances, greater than the eighteen exceedances legally allowed (Figure S6e). There are also valid measurements from an urban industrial station, which are exceeding the EU limit value in all the 8 years with valid data. This industrial station measure also few exceedances of the EU hourly limit value, but still compliant. The two urban background stations were compliant with the yearly EU limit value, except for the station IT0854A, which has no compliance in 2008 and 2015. These urban background stations have recorded no exceedances of the EU hourly limit value for the NO_2 concentrations.

5.2.4. O_3 concentrations

Figure S8 shows the number of exceedances registered for O_3 target value for the protection of human health, registered per calendar year from 2008 till 2017. The WHO guideline value for O_3 ($100\ \mu g.m^{-3}$) is not displayed in this figure since it is calculated from eight-hour mean concentrations, a different metric from the one used by EU legislation. Figure S9 indicates the distribution per month of those days of exceedance for O_3.

The background station in Bristol registered few exceedances to the O_3 target value almost every year of the 10-years period, with the highest number of five exceedances registered in 2009. Despite those exceedances, the background station is always compliant with the 25 exceedance days allowed per year, averaged over three years (Figure S8a).

Amsterdam recorded lower ozone concentrations in the traffic station, as expected. Few exceedances to the O_3 target value were registered from 2012 till 2017. Amsterdam was compliant with the 25 days of exceedances allowed per year (Figure S8b).



(a) (b) (c)

(d) (e)

Figure S8: Number of exceedances registered for O_3 target value for the protection of human health, registered per calendar year from 2008 till 2017, for five case studies. The solid line represents the number of exceedances allowed per year (averaged over three years), as defined by the Directive 2008/50/EC.

The background station in Ljubljana registered exceedances to the O_3 target value in all the years, being no compliant with the 25 days of exceedances allowed per year from 2011 till 2013, in 2015, and in 2017 (Figure S8c).

In Aveiro Region, there are valid measurements of ozone concentrations in two air quality stations, a suburban background station, and a suburban industrial station. Both stations registered exceedances to the target value in all the years with valid data. With the industrial station being no compliant with the 25 days of exceedances (Figure S8d).

The stations in Liguria Region recorded exceedances to the O_3 target value in all the years with valid data, always greater than the allowed 25 days of exceedances. These results indicate a problem of no compliance with the legislated target value for O_3 concentrations, with more critical numbers

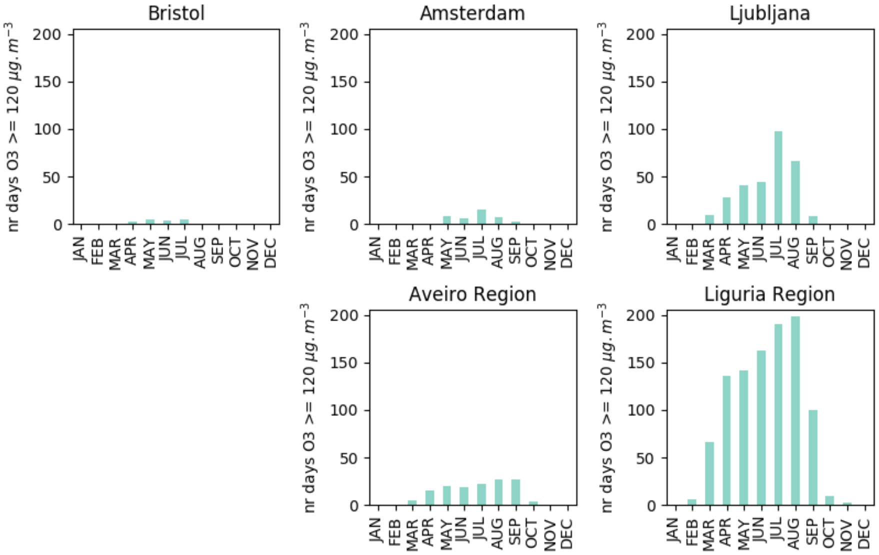


Figure S9: Monthly distribution of the days where O_3 concentrations above the target value for the protection of human health were observed in at least one monitoring station in each case study, from 2008 till 2017.

in 2015 and 2016 (Figure S8e).

5.3. Persistant episodes during 2008-2017

The following tables show the complete list of PM_{10} (Table S1), NO_2 (Table S2), and O_3 (Table S3) persistant episodes, which affected the ClairCity case studies between 2008 and 2017. As explained in Section 3.3, in this study, an episode is defined as a period of consecutive days (for PM_{10} and O_3) or hours (for NO_2) where a concentration above the threshold was observed in at least one station of the case study air quality monitoring network. An episode is classified as persistant when it lasts for 10 or more consecutive days in the case of PM_{10} and O_3, and for 5 or more consecutive hours in the case of NO_2.

Table S1: PM_{10} episodes that last for 10 or more consecutive days, between 2008 and 2017.

ClairCity Pilot	first hour of exceedance	last hour of exceedance	duration (days)	max concentration (μg m^{-3})
Sosnowiec	03/01/2011	13/01/2011	11	150
Sosnowiec	16/02/2011	27/02/2011	12	121
Sosnowiec	27/10/2011	09/11/2011	14	147
Sosnowiec	12/11/2011	26/11/2011	15	192
Sosnowiec	31/01/2012	14/02/2012	15	541
Sosnowiec	12/01/2013	28/01/2013	17	219
Sosnowiec	09/02/2013	21/02/2013	13	98
Sosnowiec	02/10/2013	18/10/2013	17	100
Sosnowiec	12/11/2013	21/11/2013	10	149
Sosnowiec	26/12/2013	04/01/2014	10	114
Sosnowiec	29/01/2014	14/02/2014	17	187
Sosnowiec	24/10/2014	03/11/2014	11	132
Sosnowiec	01/12/2014	10/12/2014	10	155
Sosnowiec	10/02/2015	24/02/2015	15	180
Sosnowiec	23/10/2015	07/11/2015	16	248
Sosnowiec	14/01/2016	24/01/2016	11	143
Sosnowiec	14/01/2017	19/02/2017	37	306
Sosnowiec	21/11/2017	03/12/2017	13	136
Ljubljana	30/01/2011	11/02/2011	13	93
Ljubljana	12/11/2011	22/11/2011	11	102
Ljubljana	18/01/2016	29/01/2016	12	115
Aveiro region	19/01/2008	01/02/2008	14	154
Aveiro region	16/12/2008	26/12/2008	11	163
Aveiro region	08/10/2011	18/10/2011	11	118
Aveiro region	17/12/2011	04/01/2012	19	113
Aveiro region	30/12/2014	13/01/2015	15	121

Table S2: NO_2 episodes that last for 5 or more consecutive hours, between 2008 and 2017.

ClairCity Pilot	first hour of exceedance	last hour of exceedance	duration (days)	max concentration (μg m^{-3})
Bristol	16/01/2008 09:00	16/01/2008 15:00	7	236
Bristol	13/02/2008 08:00	13/02/2008 14:00	7	274
Bristol	26/03/2008 14:00	26/03/2008 18:00	5	237
Bristol	02/06/2008 15:00	02/06/2008 19:00	5	243
Bristol	12/10/2008 08:00	12/10/2008 14:00	7	290
Bristol	17/03/2009 15:00	17/03/2009 20:00	6	276
Bristol	18/03/2009 14:00	18/03/2009 21:00	8	287
Bristol	19/03/2009 08:00	19/03/2009 19:00	12	274
Bristol	12/10/2009 16:00	12/10/2009 20:00	5	247
Bristol	06/04/2010 15:00	06/04/2010 19:00	5	247
Bristol	02/06/2012 08:00	02/06/2012 12:00	5	285
Bristol	26/08/2015 14:00	26/08/2015 18:00	5	577
Bristol	27/08/2015 08:00	27/08/2015 19:00	12	310
Bristol	28/08/2015 07:00	28/08/2015 12:00	6	268
Bristol	29/08/2015 10:00	29/08/2015 19:00	10	277
Bristol	18/09/2015 10:00	18/09/2015 16:00	7	268
Bristol	14/10/2015 11:00	14/10/2015 18:00	8	285
Bristol	16/10/2015 15:00	16/10/2015 19:00	5	238
Bristol	20/10/2015 08:00	20/10/2015 16:00	9	280
Liguria region	20/12/2009 17:00	20/12/2009 22:00	6	234
Liguria region	03/12/2012 18:00	03/12/2012 22:00	5	288

Table S3: O_3 episodes that last for 10 or more consecutive days, between 2008 and 2017.

ClairCity Pilot	first hour of exceedance	last hour of exceedance	duration (days)	max concentration (μg m^{-3})
Liguria region	23/04/2008	13/05/2008	21	167
Liguria region	28/06/2009	08/07/2009	11	177
Liguria region	19/08/2009	28/08/2009	10	174
Liguria region	08/04/2010	17/04/2010	10	157
Liguria region	01/07/2010	13/07/2010	13	154
Liguria region	17/08/2011	27/08/2011	11	160
Liguria region	18/06/2012	02/07/2012	15	166
Liguria region	04/07/2012	15/07/2012	12	149
Liguria region	25/07/2012	06/08/2012	13	174
Liguria region	14/07/2013	25/07/2013	12	174
Liguria region	10/08/2013	19/08/2013	10	168
Liguria region	15/07/2014	29/07/2014	15	174
Liguria region	13/04/2015	26/04/2015	14	162
Liguria region	08/05/2015	22/05/2015	15	164
Liguria region	24/05/2015	03/06/2015	11	158
Liguria region	06/06/2015	16/06/2015	11	190
Liguria region	18/06/2015	05/07/2015	18	197
Liguria region	31/07/2015	14/08/2015	15	193
Liguria region	02/09/2015	13/09/2015	12	150
Liguria region	18/03/2016	29/03/2016	12	149
Liguria region	09/04/2016	26/04/2016	18	156
Liguria region	14/05/2016	29/05/2016	16	169
Liguria region	20/06/2016	17/09/2016	90	245
Liguria region	19/09/2016	02/10/2016	14	169
Liguria region	18/04/2017	27/04/2017	10	164
Liguria region	25/05/2017	13/06/2017	20	210