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Estimating background concentrations of $PM_{2.5}$ for urban air quality modelling in a data poor environment

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HIGHLIGHTS

- Background data in local scale air quality models represent regional $PM_{2.5}$.
- Accounting for wind direction in models improves background estimates.
- Proximate sources close to monitoring sites influence model verification.
- Improved model outputs will aid assessment of acute and long-term health impacts.

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ABSTRACT

Atmospheric dispersion models are widely applied to simulate pollutant concentrations such as $PM_{2.5}$ for use in long- and short-term health studies. A significant proportion of $PM_{2.5}$ originates outside urban areas in which many people live. It is important to reflect this 'background' component in the modelling process in order to provide an accurate representation of the total pollution load experienced by human populations. To be credible, model outputs must be verified against available monitoring data, which, in the case of $PM_{2.5}$, may be limited to a small number of monitoring sites across a large urban area. Here we evaluate four different approaches to representing background $PM_{2.5}$ in an atmospheric dispersion model (ADMS-Urban) for Nottingham, UK. A directional approach, based on multiple urban background monitoring sites located outside the study area provides the most robust estimates. Our adopted approach allows us to model both short- and long-term air quality conditions, whilst accounting for local- and regional-scale variations in the pollution burden, and will ultimately enable us to assess short- and long-term effects of air pollution on health.

1. Introduction

Particulate matter with an aerodynamic diameter of ≤2.5 μm ($PM_{2.5}$) is associated with many adverse health impacts including cardiovascular, respiratory, and neurological diseases (Anderson et al., 2013; Samoli et al., 2016; Shi et al., 2020; Southerland et al., 2022). There is no evidence of a threshold below which no adverse health effects occur, nor evidence of a safe level of exposure to $PM_{2.5}$ (Wei et al., 2019; World Health Organization (WHO), 2013; 2021). The current health based UK annual Air Quality standard is 20 μg/m^3, with a target figure for England of 10 μg/m^3 by 2040 (The Environmental Targets Fine Particulate Matter England Regulations, 2023). The recently updated WHO annual air quality guideline for $PM_{2.5}$ is 5 μg/m^3 (WHO, 2021).

$PM_{2.5}$ can be a primary or secondary pollutant. Major sources of primary $PM_{2.5}$ in the UK include exhaust emissions, non-exhaust emissions such as road, tyre and brake wear, and emissions from industrial processes and industrial and domestic combustion (Department for Environment, Food and Rural Affairs (Defra), 2023a; McDuffie et al., 2021). However, a significant proportion (41%–72%) of $PM_{2.5}$ in the UK is believed to be secondary, caused by chemical reactions in the atmosphere (Harrison et al., 2012; Yin et al., 2010). $PM_{2.5}$ can travel long distances in air masses, meaning long-range transport, particularly from mainland Europe, can make a large contribution to the $PM_{2.5}$ load in the UK (Graham et al., 2020).

Ambient $PM_{2.5}$ concentrations should be regularly reviewed against legislative limits and to assess the effectiveness of interventions to

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reduce $PM_{2.5}$ (Forehead et al., 2020). However, $PM_{2.5}$ is only measured at 92 out of the 171 Automatic Urban and Rural Network (AURN) monitoring sites across the UK, far fewer than for other pollutants such as NO_2, which is measured at 145 AURN monitoring sites (Defra, 2022a). Monitoring and personnel costs, along with previous regulatory focus placed on other pollutants, e.g., NO_2 and SO_2, may contribute to the lack of $PM_{2.5}$ monitors in the AURN (Defra, 2022b; Giordano et al., 2021). Local authorities may also conduct limited $PM_{2.5}$ monitoring, but focus mainly on NO_2 via the use of passive diffusion tubes (PDT) (Broday et al., 2017; Kumar et al., 2015; Sun et al., 2019). PDTs offer a coarse temporal resolution of NO_2 concentrations; however, they are useful for assessing spatial variability in concentrations (Nash and Leith, 2010).

Previously there has been a focus on monitoring $PM_{2.5}$ in 'hot spots' e.g., close to busy roads, due to associations between short-term peaks in $PM_{2.5}$ and adverse health effects (Harrison et al., 2012). Research has also demonstrated the health risks of long-term exposure to fine particles, but monitoring has not increased based on this understanding (Southerland et al., 2022). In the absence of monitoring, air pollution models can be used to generate $PM_{2.5}$ estimates across a range of spatial scales. Local-scale models can generate high resolution estimates of $PM_{2.5}$ across a city, which can then be used in decision making for air quality interventions and health impact assessments (Ortiz and Friedrich, 2013).

Annual mean modelled concentrations are typically used to assess relationships between long-term health and air pollution across city scales (Huang et al., 2017). However, models can also be used to simulate air pollution episodes, which are known to link to acute adverse health impacts (Bell et al., 2013). In the UK, these episodes typically occur at regional scales, dominated by background $PM_{2.5}$ from mainland Europe and other conurbations upwind of the study area, with local emissions 'topping-up' concentrations, resulting in higher-than-average $PM_{2.5}$ concentrations and more extreme exceedances of air quality thresholds (Graham et al., 2020).

Background concentrations are used in local-scale dispersion models to characterise the contribution of pollution sources not considered explicitly in the model run (Tchepel et al., 2010). They are combined with concentrations originating from local sources to estimate total concentrations at the models chosen receptor sites. Background concentrations are important for both long- and short-term studies as they can be a major source of error (Tchepel et al., 2010), so estimates must be as reliable as possible.

There are many ways in which background concentrations can be estimated and incorporated within a local atmospheric dispersion model. For example, rural AURN sites can provide an indication of transboundary $PM_{2.5}$ contributions, while urban AURN sites also capture additional local contributions. To provide a more dynamic representation of background concentrations, local scale dispersion models can also be coupled with regional scale models (Kadaverugu et al., 2019; Zhong et al., 2022). It is essential to get the background and local proportions accurate in local-scale air quality modelling so that double counting of sources can be avoided and local sources can be targeted for management (Ortiz and Friedrich, 2013).

Previous UK studies have used gridded background annual mean concentrations of $PM_{2.5}$ on a 1-km x 1-km resolution generated using the Pollution Climate Mapping (PCM) model (Defra, 2018a) (e.g. Singh et al., 2013, 2019). To introduce temporal variability into the background dataset, Singh et al. (2019) used urban background monitoring data, representative of the model domain. Khreis et al. (2018) also used gridded background concentrations from the PCM to account for NO_x concentrations from non-road sources in Bradford, UK.

Background concentrations represented by data from a single monitoring site have been used in studies conducted at varying scales, for example, to assess intra-urban variability of $PM_{2.5}$ concentrations in Pittsburgh, USA (Michanowicz et al., 2016) and smaller scale studies determining agricultural emissions in Ohio, USA (Hadlocon et al., 2015). In Lithuania, Dėdelė and Miškinytė (2018) used a single monitoring site to derive an annual average background concentration for their atmospheric dispersion model.

Rittner et al. (2020) used a combination of urban and rural background monitoring sites as background concentrations in an atmospheric dispersion model to determine particle concentrations (PM_{10}, $PM_{2.5}$) in Sweden. Zhong et al. (2021) derived background $PM_{2.5}$ concentrations for their atmospheric dispersion model from hourly $PM_{2.5}$ recorded at rural background AURN sites, which they then scaled using a ratio of the annual average background concentration in rural areas bordering the model domain (West Midlands, UK) generated by the PCM (Defra, 2018a). In Beijing, China, Biggart et al. (2020) used three sites to the northwest, northeast and southeast of urban Beijing to represent background $PM_{2.5}$ and PM_{10} concentrations when modelling street-scale air quality.

Regional scale air quality models have also been coupled with urban scale atmospheric dispersion models to provide background concentrations. When estimating NO_x, NO_2 and O_3 in London, Beevers et al. (2012) found that coupled models yielded reasonable agreement with measurements, although there were some issues with double counting of emissions. In their study of Paris, France, Lugon et al. (2020) found that their regional models underestimated NO and NO_2 concentrations at a local level, but this improved when coupled with a local scale model.

In order to test model input parameters and assumptions about background concentrations, it is necessary to verify model outputs with monitored data (Defra, 2018b). This highlights the need for monitoring locations for $PM_{2.5}$ to be as representative of an area as possible. Previous studies have demonstrated the importance of using fine temporal resolution monitoring data to identify sources and PM components. Ferranti et al. (2008) recorded PM_{10} concentrations in a remote location in northwest England and were able to classify high concentration PM_{10} events by start time, duration, wind direction and particle size characteristics, resulting in the identification of an unregulated burn site. On a wider scale, Malley et al. (2016) used air mass trajectory data combined with measurements from monitoring sites, measuring PM in southeast Scotland and southeast England, to identify the contributions from different PM components. Conducting model verification can, however, be challenging when there are a small number of sites within the model domain that measure $PM_{2.5}$, and they have missing or poor quality data, for example, where monitored data are affected by sources that are not representative of the study area.

The aim of our larger study is to use an air pollution model (ADMS-Urban) to generate long- and short-term outputs for the city of Nottingham, UK, for use in long- and short-term health studies. Prior to doing this, however, we need to test whether the model is performing well for our chosen study area. This paper describes how we set up and validated our model by: a) exploring different approaches for creating background datasets to determine the most suitable regional background $PM_{2.5}$ to include in the model; b) scrutinising current monitoring data and ensuring it is suitable for model verification and c) evaluating model performance spatially and temporally across the city against data from automatic and non-automatic monitoring sites. Advantages and disadvantages of each modelling approach are also discussed.

2. Methods

2.1. Study area

Nottingham is situated in the East Midlands region of England, UK.

The Nottingham City Council (NCC) area is located in the centre of the Nottingham urban area, which extends into the surrounding boroughs of Ashfield, Gedling, Rushcliffe and Broxtowe. The study area for the purposes of this research covers areas within the NCC boundary (Fig. 1) and all model runs and validation were carried out for this area. The City of Nottingham is typical of larger urban areas in the UK, with a total population of 323,700 (2021 Census) (Nottingham City Council, 2023). This is important as 35.9% of the UK population live in major urban conurbations and there has been a 16% growth in population in cities since 2001 (to mid-2019) (Defra, 2021; Government Office for Science, 2021).

Evidence suggests regional background concentrations of $PM_{2.5}$ make up a considerable amount of $PM_{2.5}$ mass in urban areas of the UK (Air Quality Expert Group (AQEG), 2012). AQEG (2012) have suggested that approximately 60% of $PM_{2.5}$ mass recorded at urban background monitoring sites in central England are made up of secondary $PM_{2.5}$. NCC (2018) suggest Nottingham experiences regional background $PM_{2.5}$ pollution from other conurbations and sources, for example, the West Midlands conurbation and agricultural sources outside of the City's boundary.

In the City of Nottingham, there is one urban background AURN site that measures $PM_{2.5}$, PM_{10} and NO_2 (City Centre), and one roadside site that measures PM_{10} and NO_2 only (Western Boulevard) (Fig. 1). In line with the objectives of the AURN, these sites were established to assess compliance with the Ambient Air Quality Directives and associated air quality standards and measure reduction of pollutants over time. NCC also run a PDT network to monitor NO_2 concentrations at roadside locations (Fig. 1). One $PM_{2.5}$ monitoring site is unlikely to be representative for a population of >300,000 residents and therefore unlikely to be useful without additional data for air pollution – health assessments (Baca-López et al., 2021; Su et al., 2022). As $PM_{2.5}$ monitoring is sparse in this city, we must apply atmospheric dispersion models to provide high resolution estimates of $PM_{2.5}$ concentrations across Nottingham.

2.2. ADMS-urban atmospheric dispersion model

ADMS-Urban is a quasi-Gaussian plume dispersion model able to model pollution from sources with point, line, area, or volume geometry. It can model air quality across a range of spatial (street-scale, urban-scale, and even larger scales when coupled with a regional model (Zhong et al., 2022)) and temporal scales (short- and long-term average pollutant concentrations).

ADMS-Urban requires a number of input data files to produce air pollution estimates, including point sources, traffic count and composition data to model road sources explicitly, emissions data to input as a grid (for sources not explicitly modelled), meteorological data and background air pollution concentration data. Hourly meteorological data, including wind speed, wind direction, cloud cover and temperature data are used to drive dispersion calculations (Di Nicola et al., 2022; Zhong et al., 2021). Values representing background concentrations can be entered as either annual or hourly values (see section 2.4).

We adopted a reference year of 2019 for our modelling studies because this was the only year for which traffic data were available to us. This was also the last year before the COVID-19 pandemic, hence predates any changes in activity (e.g., commuting) caused as a consequence of national lockdowns. Annual $PM_{2.5}$ and NO_2 concentrations recorded at the City Centre AURN site between 2008 and 2019 are shown in Fig. S1. These data indicate that annual $PM_{2.5}$ concentrations at this urban background site are below the UK Air Quality Standard, but often exceed the more recent Environmental Target for England.

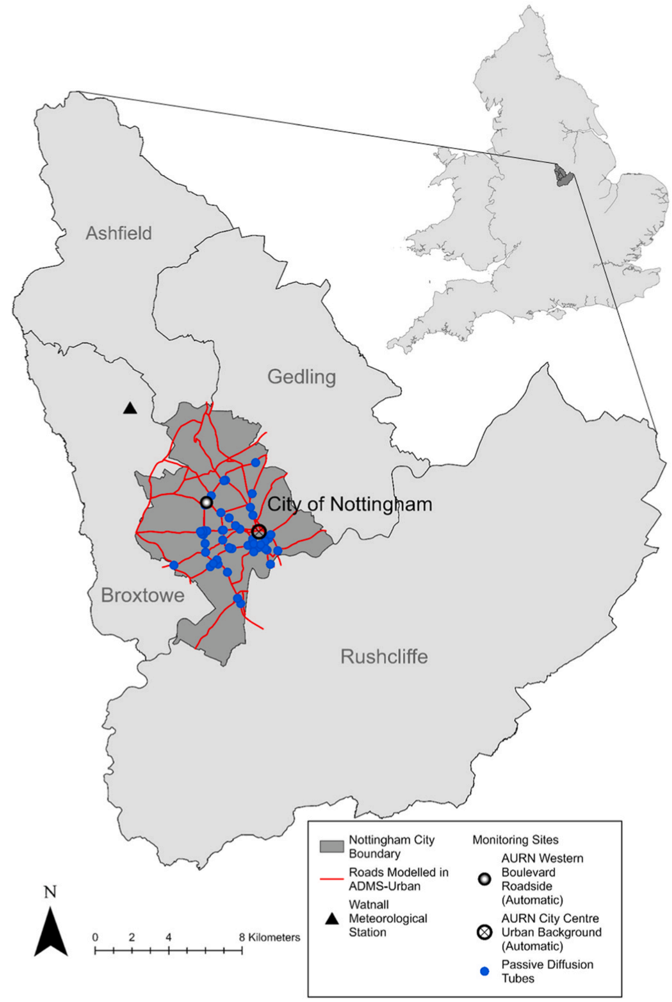

Fig. 1. Location of the Nottingham Urban Area within the UK, locations of administrative districts within the Nottingham Urban Area, the Nottingham City Boundary, AURN and PDT monitoring locations in Nottingham used for model verification, location of Watnall meteorological station, and the roads modelled in ADMS-Urban.

2.3. Model set-up

Hourly sequential meteorological data for 2019 were inputted into ADMS-Urban version 5.0. This included wind speed, wind direction, cloud cover and temperature data from Watnall weather station located 6 km northwest of the city (Fig. 1) (Centre for Environmental Data Analysis (CEDA), 2019). A single, fixed value was used to represent relative humidity.

Major roads within the Nottingham City boundary were modelled explicitly using daily average traffic count and vehicle composition data for individual road links supplied by NCC (Fig. 1). The UK Emissions Factor Toolkit (EFT) v10.1 was used to calculate road traffic emissions; this was the most current version at the time of modelling (Defra, 2020). Emissions were scaled depending on traffic flow by a local time-varying emissions factor, this drives diurnal variations on weekdays and lower concentrations on Saturdays and Sundays. This factor was calculated using traffic count data supplied by NCC.

All other emissions, including minor roads, were inputted into ADMS-Urban as 1-km x 1-km resolution emissions grids (Gulliver et al.,

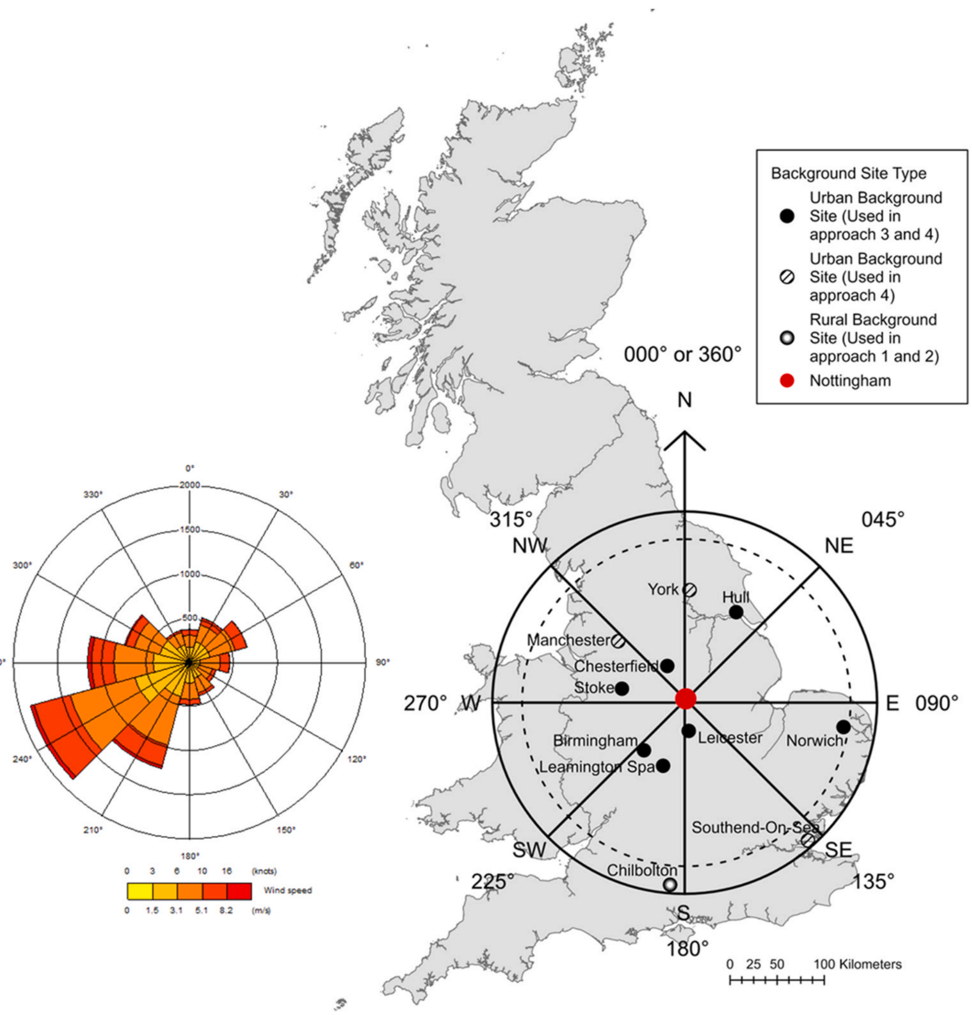

Fig. 2. Background AURN sites used to form background datasets for input into ADMS-Urban. Compass (right) shows 8 × 45° wind sectors used in Approach 3. The wind rose (left) shows the frequency of wind directions at Watnall and the 12 × 30° sectors used in Approach 4.

2018; NAEI, 2018). These are estimated annual averages which are compiled from a range of statistical datasets including energy, transport, pollution inventories and food and farming data (NAEI, 2022). Source disaggregation was turned on in ADMS-Urban to avoid duplication of sources and the Generic Reaction Set (GRS) Chemical Reaction Scheme was used to model photochemical reactions between NO, NO_x, O_3 and VOCs (O'Neill et al., 2021).

2.4. Determining a suitable background dataset for model input

ADMS-Urban provides options to incorporate background concentrations into the modelling process using single fixed values or hourly sequential values taken from an appropriate background monitoring site. This is important in the case of $PM_{2.5}$, because ADMS-Urban does not simulate any background particulate matter from either primary or secondary sources. Here we use hourly sequential background concentrations of pollutants recorded at monitoring sites outside the modelling domain. These background concentrations represent transboundary, national, and regional components of $PM_{2.5}$. It is important to get the best possible estimate of the background fraction of $PM_{2.5}$ concentrations to ensure the impact of local sources can be assessed realistically.

Table 1
Approaches to determining background concentrations.

Approach	Description
Approach 1	Hourly sequential background data from a rural AURN site, Chilbolton Observatory (southern England), 200 km from Nottingham City Centre (Fig. 2).
Approach 2	Hourly sequential background data from a rural AURN site, Chilbolton Observatory (Fig. 2), were scaled using fixed annual averages from the PCM Model (Defra, 2019). Four values were taken from the PCM Model at rural locations within Nottingham but outside the city (Fig. S2).
Approach 3	Hourly sequential background data from urban background AURN sites located within each of 8 × 45° sectors upwind of Nottingham (solid black dots Fig. 2) were used. These were all within a 170 km radius of the city. Monitoring sites as listed in Table S1. In calculating the background concentrations to be used in ADMS-Urban, contributions from each sector were weighted by the frequency of wind from that sector (wind rose, Fig. 2) representing air masses from each direction (Biggart et al., 2020; O'Neill et al., 2021; Zhong et al., 2021).
Approach 4	Hourly sequential background data from urban background AURN sites located in 12 × 30° sectors upwind of Nottingham (solid black dots and open dots with cross hatch Fig. 2) were used. These sites were within a 200 km radius of the city. Contributions from each sector were weighted by the frequency of wind from that sector (wind rose Fig. 2).

Four different approaches to deriving background concentrations of $PM_{2.5}$ for Nottingham were developed using data from different AURN sites in England. These included both rural and urban background sites, which were selected based on their relationship to Nottingham as determined by wind direction recorded at the nearby Watnall meteorological station (Fig. 2). Rural AURN sites, as defined by Defra, should be located more than 20 km away from an agglomeration and 5 km from other built up areas, whereas urban background AURN sites should be representative of a continuously built up area covering a few km^2 (Defra, 2023b).

Approach 1 used hourly sequential monitored background data from Chilbolton, a rural AURN site in southern England.

Approach 2 used the same hourly data from Chilbolton as Approach 1, however these data were scaled using fixed annual averages from the PCM (Fig. S2). This uplift should ensure that $PM_{2.5}$ values are more representative of background values on the outskirts of Nottingham (Zhong et al., 2021).

Approaches 3 and 4 used hourly sequential monitored background data from various urban background AURN sites in the UK, based on their locations within either 8 (Approach 3) or 12 (Approach 4) sectors from the Watnall wind rose (Fig. 2). If there was more than one site in a sector, then the site closer to Nottingham was generally chosen. Where there was no site in a particular sector, then the closest site in an adjacent sector was used (e.g. data from Hull for the 45–90° sector in Approach 3). Details of the monitoring sites used to represent each sector in Approach 3 are given in Table S1. Each approach is summarised in Table 1.

The AURN sites were chosen for a number of reasons. At the time of this study, there were only five rural AURN sites that measured hourly $PM_{2.5}$ concentrations. Chilbolton Observatory was selected as it was the

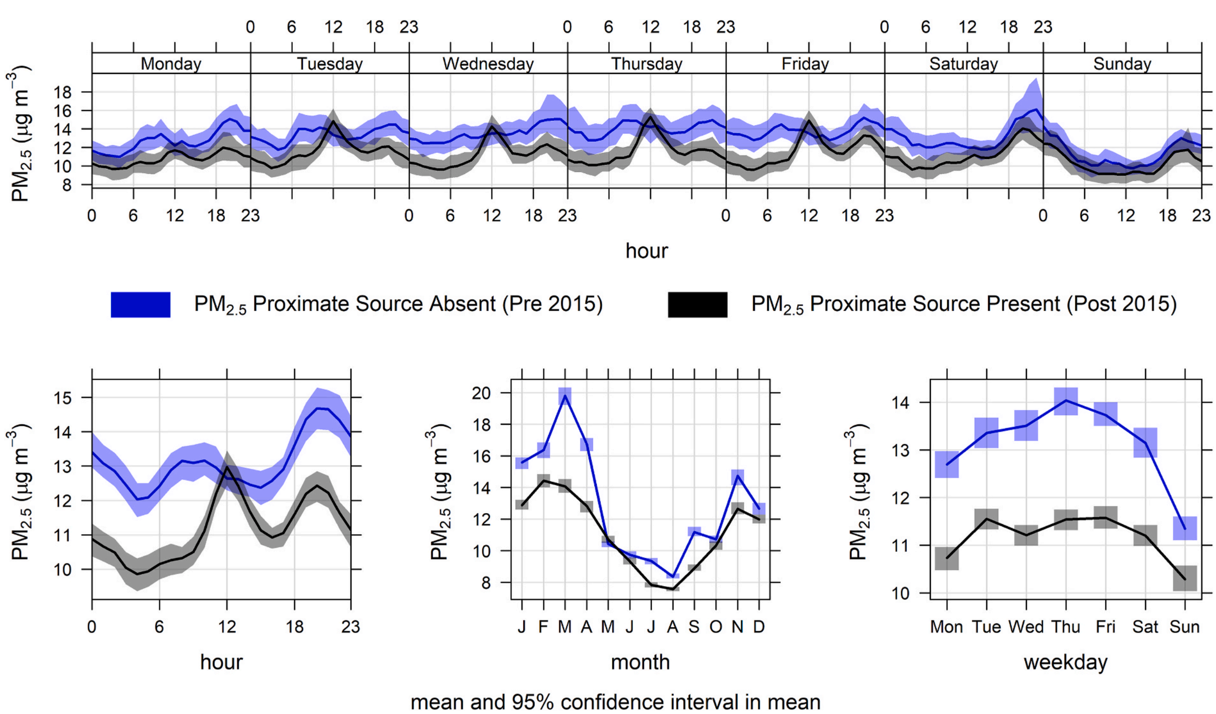

Fig. 3a. Time variation plot of $PM_{2.5}$ concentrations at the City Centre AURN site (2009–2019).

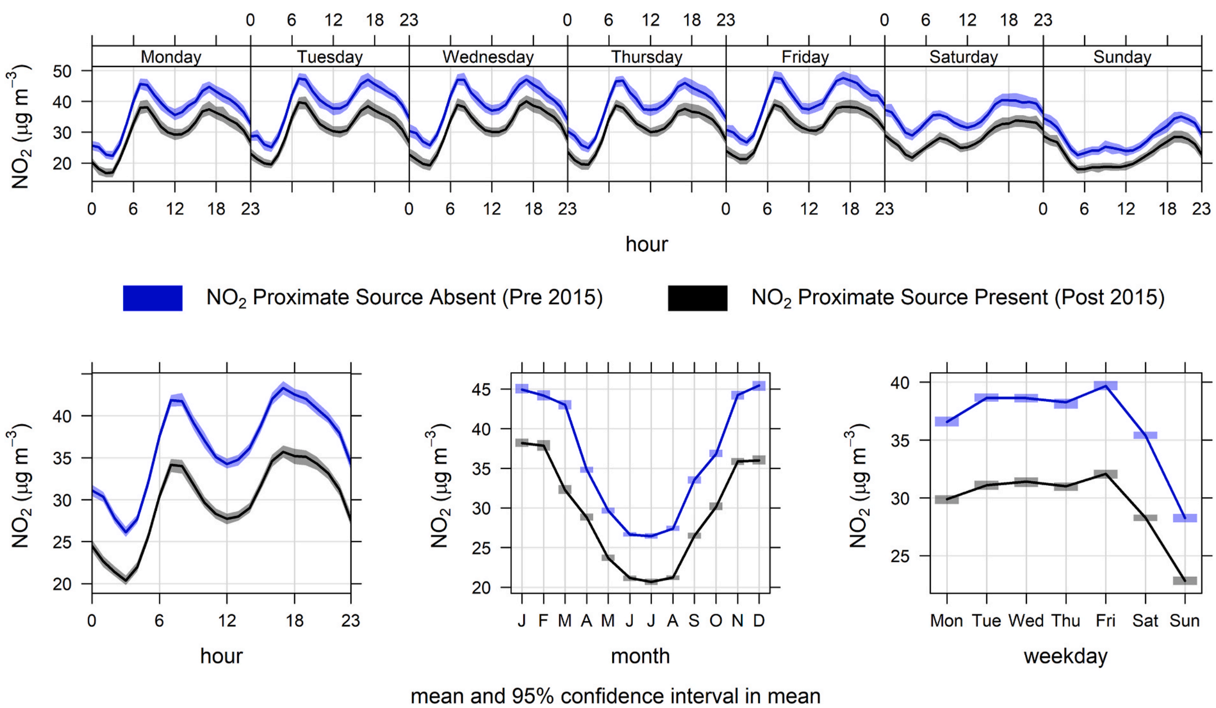

Fig. 3b. Time variation plot of NO_2 concentrations at the City Centre AURN site (2009–2019).

Fig. 4. Hot food stall located nearby to the Nottingham City Centre AURN site. Photograph taken: 21/02/2023.

closest of these rural AURN sites to Nottingham. Furthermore, the Chilbolton AURN site is often upwind of Nottingham as the prevailing wind comes from the south west (Fig. 2). The AURN sites located in Nottingham were not used to generate background values. It should be noted for the purpose of this study, straight-line trajectories have been assumed. Median annual concentrations of $PM_{2.5}$ for the AURN sites used in Approach 3 ranged from 4 to 10 μg/m^3. 5th percentile concentrations ranged from 1 to 3 μg/m^3. 95th percentile concentrations had a much larger range in comparison (20–40 μg/m^3) possibly due to above average concentrations recorded during $PM_{2.5}$ episodes. In Approach 3, 47% of hours in the year used values from two urban background AURN sites (Birmingham and Leamington Spa), reflecting the prevailing south-westerly wind (Fig. 2; Table S1).

As stated previously, $PM_{2.5}$ is only monitored at one urban AURN site in the City of Nottingham, whereas NO_2 is monitored more extensively, including at 55 sites using PDTs (see section 2.1). To get a broader understanding of model performance, specifically its ability to capture emissions from road traffic sources, we ran ADMS-Urban to these 55 sites. For consistency, we used the same rural and urban background AURN sites to generate background NO_2 concentrations as we did for $PM_{2.5}$, even though there were other AURN sites we could have used for NO_2 as it is more widely measured, and which might have provided a better representation of background NO_2.

2.5. Model verification

Both AURN sites in Nottingham (Fig. 1) were used to verify model performance for each approach to representing background pollutant concentrations (Table 1). The openair R package was used to visualise and analyse data prior to model verification (Carslaw and Ropkins, 2012; R Core Team, 2021). This package has been used widely to identify sources of air pollution using polar and time variation plots (Bodor et al., 2020; Grange et al., 2016; Munir and Mayfield, 2021). Here this approach allowed us to identify the influence of a proximate source of $PM_{2.5}$ at the City Centre AURN site. The lunchtime peaks of $PM_{2.5}$ in the time variation plots (Fig. 3a) present a very different temporal signature from that of expected local sources, such as nearby roads, that typically follow a diurnal pattern of the morning and evening rush-hours (Kendrick et al., 2015). As $PM_{2.5}$ was anomalous, NO_2 was tested to identify whether similar temporal patterns were seen for this pollutant. Fig. 3b shows that there were no differences in temporal patterns of NO_2 from the expected diurnal patterns. Further investigation into the $PM_{2.5}$ time series revealed that the cause was a mobile hot food outlet contributing much higher emissions to the area adjacent to the City Centre AURN site, not typical of the city as a whole (Fig. 4). Correspondence with the local authority revealed that the food outlet opened in 2015 and operated between the hours of 10:30 and 16:30. Due to the influence of the proximate source, data for busy periods (11:00–15:00 inclusive, Monday to Sunday), were removed from the verification dataset.

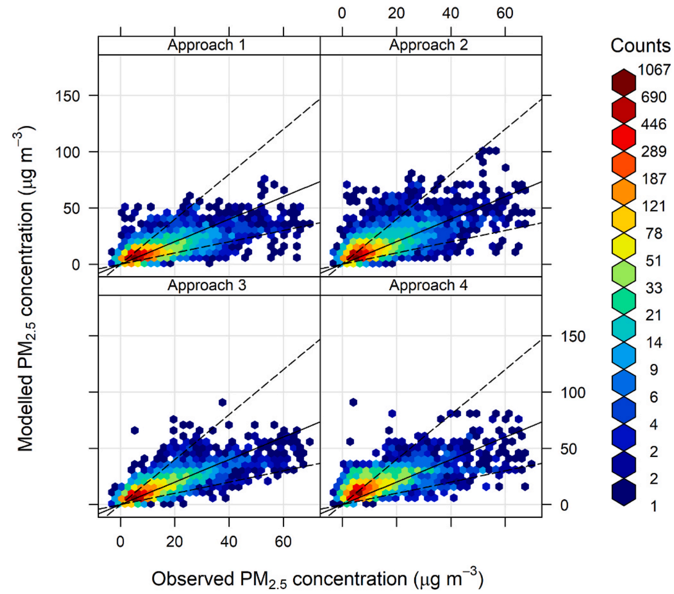

Fig. 5a. Observed vs. Modelled $PM_{2.5}$ scatter plots for all approaches for the City Centre AURN site. The solid line is the 1:1 line, the lower dashed line is the 1:0.5 line and the upper dashed line is the 1:2 line, these demonstrate how close the data points are to a 1:1 relationship and shows what data points are within a factor of two (FAC2) (Carslaw and Ropkins, 2012). Hexagonal binning shows the number of data points that lie within each shaded hexagon.

$PM_{2.5}$ is not monitored at the Western Boulevard AURN site (Fig. 1) hence it was estimated by applying a $PM_{2.5}$:PM_{10} ratio from the City Centre time series once the influence of the hot food stall was removed from the dataset. This ratio was calculated for hourly concentrations of PM_{10} and $PM_{2.5}$ and averaged for the year, giving a value of 0.58, which is similar to the ratios reported by Harrison et al. (2012), Munir (2017) and Spandana et al. (2021). The value was then applied to the measured PM_{10} values from Western Boulevard to yield estimated $PM_{2.5}$ values for that site.

Hourly concentrations of $PM_{2.5}$ modelled using ADMS were subsequently verified against both hourly monitored $PM_{2.5}$ concentrations at the City Centre AURN site, and hourly estimated $PM_{2.5}$ concentrations at the Western Boulevard AURN site.

Modelled NO_2 was verified using hourly concentrations at the City Centre and Western Boulevard AURN sites and average annual concentration data from the PDT network (Defra, 2022a; NCC, 2020). PDT data was taken from NCC annual air quality reports which include local bias adjustment (NCC, 2020).

Modelled and monitored values for Nottingham were compared using the ModStats model performance function in the Openair R Package (see also Borge et al., 2022; Carslaw, 2011; Frohn et al., 2022).

Table 2
Model statistics of model performance for $PM_{2.5}$ at City Centre and Western Boulevard AURN sites (hourly data).

Approach	n[a]	FAC2	MB	MGE	NMB	NMGE	RMSE	r	COE	IOA
City Centre										
Approach 1	6676	0.77	0.46	4.70	0.04	0.44	7.14	0.70	0.30	0.65
Approach 2	6676	0.70	3.43	5.97	0.32	0.56	8.96	0.70	0.11	0.56
Approach 3	6676	0.81	2.05	4.24	0.19	0.40	6.39	0.80	0.37	0.68
Approach 4	6676	0.60	5.62	7.28	0.53	0.69	9.48	0.69	−0.08	0.46
Western Boulevard										
Approach 1	8481	0.81	−0.54	4.60	−0.05	0.40	7.18	0.63	0.22	0.61
Approach 2	8481	0.78	2.35	5.72	0.20	0.50	9.12	0.62	0.03	0.51
Approach 3	8481	0.86	1.03	4.27	0.09	0.37	6.77	0.72	0.27	0.64
Approach 4	8481	0.72	4.74	6.60	0.41	0.58	9.10	0.64	−0.12	0.44

[a] n equals the number of hourly data points tested in the analysis.

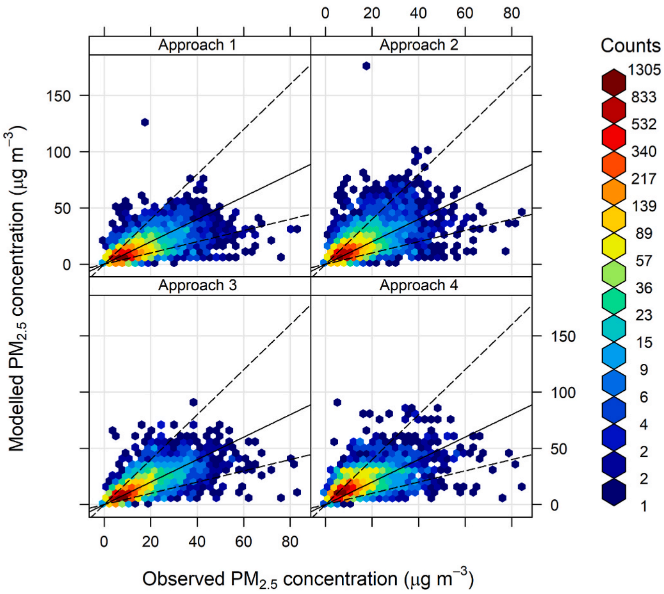

Fig. 5b. Observed (estimated) vs. Modelled $PM_{2.5}$ scatter plots for all approaches for the Western Boulevard AURN site. The solid line is the 1:1 line, the lower dashed line is the 1:0.5 line and the upper dashed line is the 1:2 line, these demonstrate how close the data points are to a 1:1 relationship and shows what data points are within a factor of two (FAC2) (Carslaw and Ropkins, 2012). Hexagonal binning shows the number of data points that lie within each shaded hexagon.

Model performance was assessed using metrics including fraction of predictions within a factor of two (FAC2), mean bias (MB), mean gross error (MGE), normalised mean bias (NMB), normalised mean gross error (NMGE), root mean square error (RMSE), correlation coefficient (r), coefficient of efficiency (COE) and index of agreement (IOA) (Carslaw and Ropkins, 2012; R Core Team, 2021).

3. Results

A range of issues affected the number of $PM_{2.5}$ data points available from observations and modelling. Fewer hourly observations ($n = 6676$) are included in the City Centre verification dataset, due to data loss (1825 data points, 21% data loss) associated with data cleaning to remove the influence of the proximate source (see above), compared to the PM_{10}:$PM_{2.5}$ scaled Western Boulevard verification dataset ($n = 8481$) (Table 2). Other differences in the number of observations (n) at the City Centre and Western Boulevard (Tables 2 and 3) are due to variable data capture at the monitoring sites, for example, missing data due to instrument failure ($PM_{2.5}$: City Centre – 191 missing data points (2% data loss), NO_2: City Centre – 148 missing data points (2% data loss); Western Boulevard – 42 missing data points (<1% data loss)). 195 data points were missing for PM_{10} at Western Boulevard, meaning we were unable to estimate $PM_{2.5}$ for these (see above), resulting in 2% data loss.

Missing data within the hourly sequential meteorological input file also impacted the number of modelled values. If meteorological data is not available, the calculation for that hour is skipped in ADMS-Urban. Across all model runs 97 h were skipped because of this, resulting in 1% data loss from missing meteorological data. Rows with missing air quality or meteorological data were deleted for the purposes of model verification.

Statistics of model performance based on the comparison of observation-based and modelled concentrations of $PM_{2.5}$ are given in Table 2. Approach 1 performed best in MB and NMB at both sites, with MB providing an indication of model over (+) or under (−) estimation. Approach 2 performed moderately well across all test metrics, except for r and RMSE where it performed the worst out of all approaches when verified against estimated $PM_{2.5}$ data at Western Boulevard.

Approach 3, using urban background concentrations from 8 × 45° wind sectors, shows good agreement with monitored concentrations at the City Centre AURN site (Fig. 5a) and with estimated concentrations at the Western Boulevard AURN site (Fig. 5b). This approach performed most strongly across a range of test metrics at both verification sites in the city (Table 2). High scores for FAC2 and IOA indicate good overall model performance (Chang and Hanna, 2004; Willmott et al., 2012).

Approach 4 performed worst out of all approaches across a range of statistics at both verification sites when compared to monitored and estimated $PM_{2.5}$ data.

Hourly modelled NO_2 concentrations were verified against hourly sequential monitored data at both the City Centre and Western Boulevard AURN sites and results varied across all four approaches. All approaches tended to underpredict NO_2 concentrations (MB), apart from Approach 3 at the City Centre AURN site (Table 3). Approach 3 showed generally good agreement with hourly monitored data at both the City Centre (Fig. 6a) and Western Boulevard sites (Fig. 6b). It performed best in FAC2 and r for both sites and best in IOA and RMSE at the Western Boulevard AURN site (Table 3). Approach 4 yielded a few large outliers where NO_2 was overestimated at both sites, however these were typically for a small number of simulations.

Modelled annual average NO_2 was also compared with monitored data from the PDT sites (Table 4). All approaches tended to underpredict (MB) concentrations at these sites. Approach 3 was able to predict

Table 3
Model statistics of model performance for NO_2 at City Centre and Western Boulevard AURN sites (hourly data).

Approach	n[a]	FAC2	MB	MGE	NMB	NMGE	RMSE	r	COE	IOA
City Centre										
Approach 1	8515	0.79	−2.96	10.83	−0.11	0.39	14.11	0.57	0.13	0.56
Approach 2	8515	0.80	−0.93	10.56	−0.03	0.38	13.72	0.60	0.15	0.58
Approach 3	8515	0.84	5.18	10.60	0.19	0.38	14.36	0.69	0.15	0.57
Approach 4	8515	0.72	−2.29	13.04	−0.08	0.47	22.92	0.63	−0.05	0.48
Western Boulevard										
Approach 1	8622	0.69	−10.94	14.20	−0.33	0.43	19.22	0.66	0.13	0.56
Approach 2	8622	0.73	−8.47	13.39	−0.26	0.41	18.10	0.65	0.18	0.59
Approach 3	8622	0.86	−1.76	10.75	−0.05	0.33	14.48	0.75	0.34	0.67
Approach 4	8622	0.67	−5.62	15.40	−0.17	0.47	24.56	0.61	0.06	0.53

[a] n equals the number of hourly data points tested in the analysis.

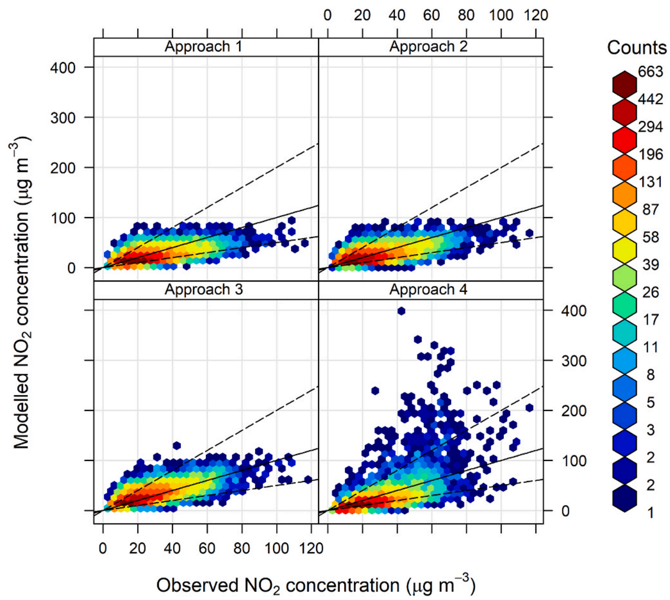

Fig. 6a. Observed vs. Modelled NO_2 scatter plots for all approaches for the City Centre AURN site. The solid line is the 1:1 line, the lower dashed line is the 1:0.5 line and the upper dashed line is the 1:2 line, these demonstrate how close the data points are to a 1:1 relationship and shows what data points are within a factor of two (FAC2) (Carslaw and Ropkins, 2012). Hexagonal binning shows the number of data points that lie within each shaded hexagon.

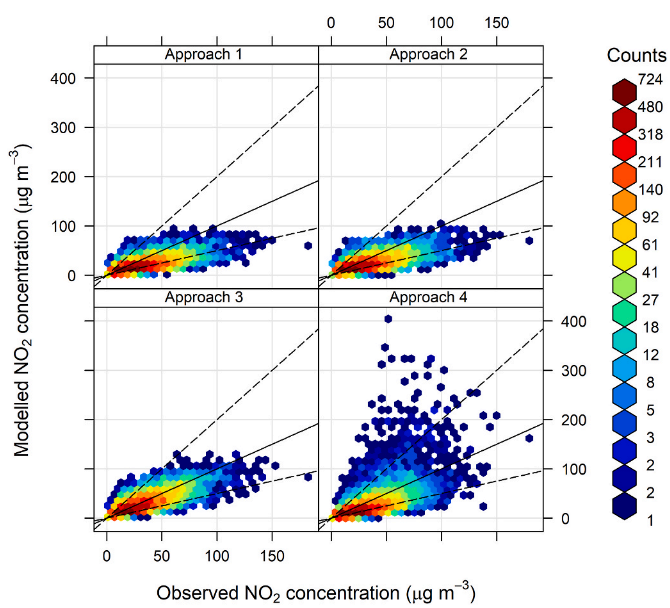

Fig. 6b. Observed vs. Modelled NO_2 scatter plots for all approaches for the Western Boulevard AURN site. The solid line is the 1:1 line, the lower dashed line is the 1:0.5 line and the upper dashed line is the 1:2 line, these demonstrate how close the data points are to a 1:1 relationship and shows what data points are within a factor of two (FAC2) (Carslaw and Ropkins, 2012). Hexagonal binning shows the number of data points that lie within each shaded hexagon.

annual mean concentrations across most sites in the city within 0–10 μg/m^3 of observed values (Fig. 7). This approach performed most strongly across all metrics apart from r, where Approach 4 performed better (Table 4).

Fig. 8 illustrates the difference between background concentrations derived from rural and urban background AURN sites used in Approaches 1 and 3 and observed concentrations recorded at the urban background AURN site located in Nottingham City Centre. The background concentrations generated by Approach 3 are typically within 5 μg/m^3 of observed total concentrations whereas the background concentrations generated by Approach 1 are much lower in comparison. Fig. 8 confirms that only a small proportion of $PM_{2.5}$ recorded in Nottingham City Centre usually originates from local sources, which is confirmed by running ADMS-Urban without the background datasets, giving a mean annual concentration of 2.5 μg/m^3. This emphasises why the input of the best possible estimate of background concentrations into the model is important to achieve accurate estimates of total concentrations.

Both approaches show seasonal trends in $PM_{2.5}$ consistent with those measured at the City Centre AURN site, including higher peaks in winter (December to February), spring (March to May) and occasionally summer (June to August). Peaks in November and December, which may relate to more local activities associated with celebrations such as Guy Fawkes Night (AQEG, 2012) are reflected better in Approach 3 but are not 'seen' by the distant rural AURN site at Chilbolton (Approach 1).

4. Discussion

4.1. *Interpretation of approaches to estimating background $PM_{2.5}$*

As noted in Section 2.1, regional and national background, and secondary contributions are known to dominate $PM_{2.5}$ concentrations across the UK (Kelly et al., 2023; Vieno et al., 2016; Wang et al., 2020), with local sources contributing very little (estimated to be < 2 μg/m^3 annual mean). Our study confirms that this is also true for the City of Nottingham (Fig. 8).

We have shown that background concentrations can be derived from available monitoring data to provide appropriate inputs into urban scale atmospheric dispersion models. As noted above, other researchers have coupled regional-scale pollution models with local-scale models to include background concentrations of $PM_{2.5}$, but this methodology was not open to us. Based on our case study, Nottingham, Approach 3, which used urban background $PM_{2.5}$ concentrations from $8 \times 45^\circ$ wind sectors, showed good agreement with monitoring data in the city and performed most strongly across a range of evaluation metrics (Table 2; Fig. 5). We have also highlighted the importance of screening and refining monitoring data, so that models can be verified to an acceptable standard, which can then be used to support air pollution – health studies.

Previous studies have demonstrated that $PM_{2.5}$ can travel long distances in air masses, and that $PM_{2.5}$ episodes in the UK can occur from stagnating air which has originated from mainland Europe or other parts of the UK (Fenech et al., 2019; Vieno et al., 2016). Back trajectory analysis has illustrated that these air masses can take a variety of paths across the UK. Approaches 1 and 2 used background data from a single, rural AURN site (Chilbolton), the closest rural AURN site upwind of Nottingham based on the prevailing wind direction. Comparison with

Table 4
Model statistics of model performance for NO_2 at diffusion tube sites (annual data).

Approach	n[a]	FAC2	MB	MGE	NMB	NMGE	RMSE	R	COE	IOA
Approach 1	55	0.93	−12.76	12.78	−0.33	0.33	14.71	0.33	−1.11	0.05
Approach 2	55	0.96	−10.69	10.80	−0.28	0.28	12.88	0.34	−0.78	0.11
Approach 3	55	1.00	−4.40	6.54	−0.12	0.17	8.33	0.34	−0.08	0.46
Approach 4	55	0.98	−10.92	10.92	−0.29	0.29	12.81	0.47	−0.81	0.10

[a] n equals the number of PDT sites used in the analysis.

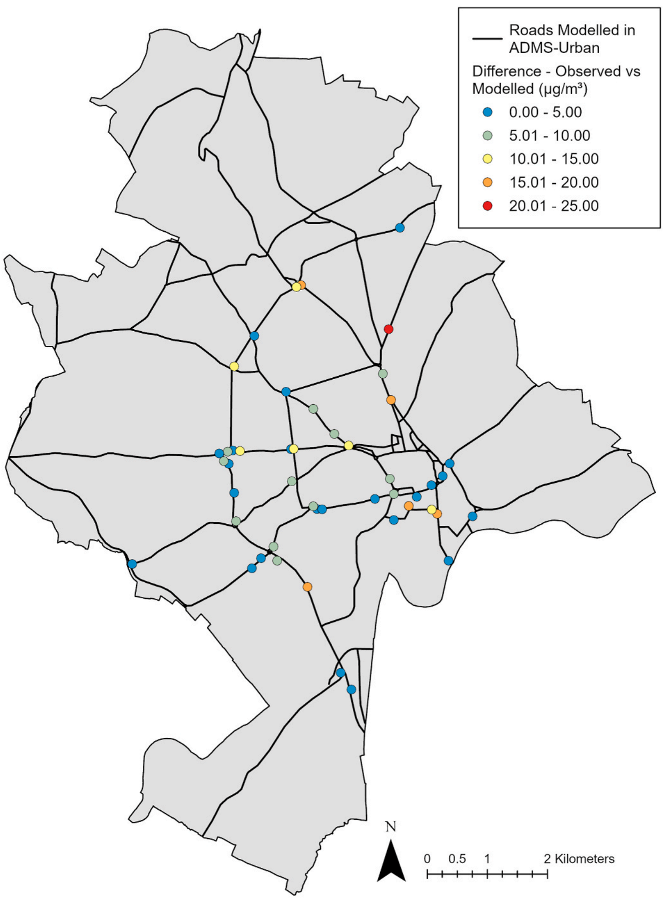

Fig. 7. Difference between observed and modelled annual average NO_2 concentrations (μg/m^3) at PDT sites across Nottingham.

measurements from the City Centre AURN site and modelling with ADMS using local sources only, indicates that these approaches are able to reproduce some episodes recorded in Nottingham and provide reasonable estimates of annual mean concentrations. Using this single monitoring site can, however, only capture certain air mass directions (Fig. 2) meaning that background $PM_{2.5}$ signals from other directions are at risk of not being represented.

In contrast, Approach 3, using data from multiple urban background AURN sites based on wind direction (Fig. 2) is more likely to reflect short-term variations in regional background $PM_{2.5}$ concentration because it considers a broader range of sectors and captures source contributions closer to the City of Nottingham. It is, therefore, more suitable to forecast both annual mean and episode-specific concentrations, for use in assessments of chronic and acute health impacts. Meteorology is a source of error in air quality models, and models will not predict observed pollution episodes if the modelled wind speed and direction does not replicate the conditions accurately enough to reflect the origins of an air pollution episode at a given location (Conti et al., 2017).

Our initial exploration of seasonality (Fig. 8) suggests possible different drivers for pollution episodes in Nottingham, which may also result in seasonal variations in PM composition (Kelly et al., 2023; Tang et al., 2018). Further work will be required to confirm this.

Nottingham is located in a central position in the UK meaning urban background AURN sites are positioned in most wind sectors. In some sectors, however, there were no AURN sites that measured $PM_{2.5}$ during the reference year, meaning some AURN sites had to cover more than one sector, or the closest AURN site to that wind sector was used (Table S1). However, capturing air mass characteristics from a range of directions, even if restricted by geographical location, will provide a better indication of $PM_{2.5}$ travelling in air masses than capturing air mass characteristics from a single background monitoring site alone. Further investigation is needed for other locations where there are fewer options for monitoring sites in certain wind sectors, for example, coastal cities.

4.2. Implications for long- and short-term health studies

The ultimate objective of long- and short-term studies that assess relationships between air pollution and health is to determine concentrations of a pollutant in space and time, so that exposure can be quantified and associations with health impacts can be determined (Kirwa et al., 2021). Although background contributions reduce the spatial heterogeneity at urban background locations (Beevers et al., 2013), other long-term health studies have found that finer spatial scale differences in concentrations of air pollution, driven by more local emissions, may have larger associations with health risks than differences in regional scale air pollution (Eeftens et al., 2012; Kirwa et al., 2021; Miller et al., 2007). For example, Gulliver et al. (2018) conducted long-term PM_{10} modelling in the Avon Longitudinal Study of Parents And Children (ALSPAC) study and found that background PM_{10} represented 83–85.6% of long-term PM_{10} concentrations. However, despite the large influence of background concentrations, local concentrations contributed to large differences in concentration experienced by different study participants.

$PM_{2.5}$ episodes are linked to acute health effects (Pope et al., 2011). Using background datasets that can accurately determine the background proportion of pollution in models during episodes is important,

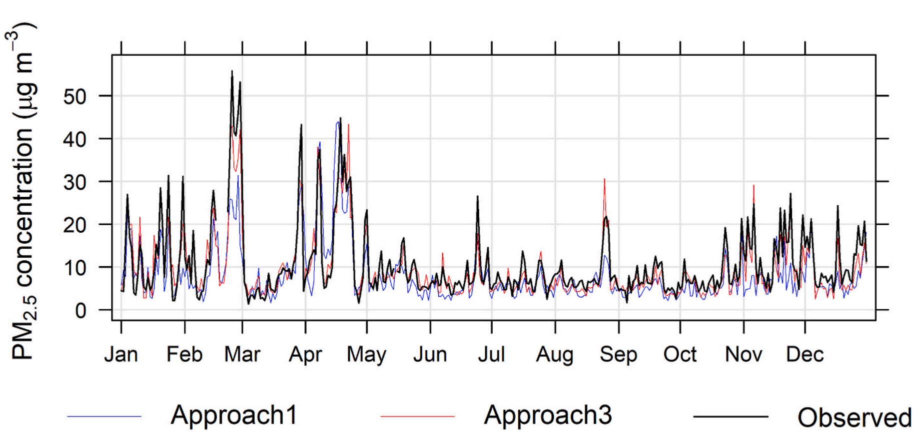

Fig. 8. Time series plot showing daily averages of the background $PM_{2.5}$ dataset for Approach 1 (Chilbolton only), Approach 3 (multiple sites) and observed $PM_{2.5}$ concentrations at the Nottingham City Centre urban background AURN site in 2019.

as episodes are often influenced by background air pollution (Graham et al., 2020). Therefore, being able to differentiate between background and local pollution contributions in models could help understanding on how local sources contribute to higher concentrations recorded during these periods. An et al. (2007) found that in Beijing, the contribution of $PM_{2.5}$ from background sources varied depending on location, ranging from 39% to 53% in the northwest to 15% in the southwest.

Modelling episodes is important to understand the relationship between above average concentrations of air pollution and acute morbidity and mortality recorded by hospital emergency departments, e.g., acute respiratory and cardiovascular events (Atkinson et al., 2014; Elliot et al., 2016; Sorek-Hamer et al., 2020). Effective models can identify locations with high concentrations of pollution which could be associated with a higher occurrence of acute health conditions.

The evidence above suggests it is important to ensure that both background and local components of $PM_{2.5}$ are accurately represented in the model, so that the total amount of $PM_{2.5}$ is estimated correctly. Background concentrations should be as representative as possible, so that the model can produce the 'right results for the right reasons.'

4.3. Model verification and impacts of proximate sources

This study found that the City Centre AURN site in Nottingham is influenced by a proximate source, not reflective of general conditions across the city which limits the number of observations at that site available for model verification. We decided to remove the influence of the proximate source from the verification dataset because of uncertainties in how best to parametrise this small local source in the model. In the absence of this discovery of a proximate source, erroneous conclusions could have been drawn when interpreting model verification results. This could have serious impacts if the model outputs were being used for decision making, e.g., conducting a study on air pollution interventions or an impact assessment on a new piece of policy (Holman et al., 2015).

The influence of proximate sources on monitoring sites may also affect the reporting required in accordance with air quality directives. Air quality in the wider vicinity may be much better than monitoring suggests. Therefore, proximate sources may obscure general reductions in $PM_{2.5}$ in an area subject to national and local emission reduction interventions, leading to incorrect conclusions on the effectiveness of such interventions. This also applies to epidemiological studies, where associations between air pollution and adverse health impacts may be inaccurately identified when using data that is not representative of actual human exposure (Bell et al., 2007; Fann et al., 2011).

In this study, we used the corrected time series from the City Centre AURN site to produce a PM_{10}:$PM_{2.5}$ ratio to apply to the Western Boulevard monitored PM_{10} data. This was useful to increase the number of verification sites (from one to two) to check model performance. However, data from one monitoring site monitoring $PM_{2.5}$ and scaled estimates of $PM_{2.5}$ from another monitoring site monitoring PM_{10} are unlikely to be spatially representative enough to inform decisions on air quality – health policy for medium to large sized cities (Piersanti et al., 2015). It is recommended that $PM_{2.5}$ monitoring should be added to Western Boulevard.

PDT monitoring at roadside locations, measuring annual mean NO_2 concentrations from traffic related air pollution, were used as a proxy to test how well models predicted concentrations from road sources. This helped to provide a broader spatial assessment of model performance, although model inputs were not optimised for NO_2 (see section 2.4). This gives us confidence that our model can estimate concentrations reasonably well from modelled road sources and enables us to apply the model widely across the city using the road source parameters. However, ultimately there is a need for more $PM_{2.5}$ monitors to record $PM_{2.5}$ concentrations in more locations and verify models. The rise of new technology for $PM_{2.5}$ monitoring, such as the use of low-cost air quality sensors, could provide more data for verifying air pollution models across a range of temporal scales even if of lower accuracy and precision compared to reference monitoring sites (Bi et al., 2021). Nevertheless, there is still a need for models to estimate pollution concentrations across scales that are not covered by monitors, run forecasts and back-casts and conduct source apportionment exercises.

5. Conclusion and recommendations

This study aimed to generate model output for use in short- and long-term health studies in a location where monitoring is compromised and insufficient. In this study we explored methods for determining a suitable background dataset for $PM_{2.5}$ models and identified ways to verify $PM_{2.5}$ models when monitoring data was limited.

We have shown that we can produce directionally-informed estimates of background $PM_{2.5}$ from urban background monitoring sites selected on the basis of wind direction. This enables us to include local and regional background contributions in our modelling studies and to evaluate their varying contributions to short-term pollution episodes and longer term air quality. Therefore, this approach can be applied in studies investigating the short- and long-term health impacts of $PM_{2.5}$.

This study also identified the influence of a proximate source near to a monitoring site used for model verification, which highlights the importance of ensuring that monitoring data for individual sites is scrutinised in detail prior to use in modelling and model verification. Furthermore, better controls are needed to prevent siting potential sources of pollution close to reference air quality monitoring sites. It is recommended that provision of robust statutory guidance on the siting of potential sources is needed to prevent this issue.

The approaches used in this study are relatively simple and accessible for modellers. They can be applied in different geographies, across a range of spatial and temporal scales, however, they cannot be easily used to assess future changes unlike coupled regional and local models used for forecasting (Baklanov and Zhang, 2020; Zhang et al., 2012).

Air pollution modelling is complex, it requires compromise to achieve the precision in concentrations, temporal and spatial resolution appropriate to the study. This study provides methods to overcome some of the compromises modellers need to consider when conducting an air pollution study, for example representing the complexity of the wider air quality climate within the model and overcoming limitations with monitored data.

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CRediT authorship contribution statement

Eve L. Draper: Conceptualization, Methodology, Investigation, Visualization, Writing – original draft. **J. Duncan Whyatt:** Conceptualization, Methodology, Writing – review & editing, Supervision. **Richard S. Taylor:** Resources, Supervision. **Sarah E. Metcalfe:** Funding acquisition, Conceptualization, Methodology, Writing – review & editing, Supervision.

Declaration of competing interest

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

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.atmosenv.2023.120107.

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