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ESTIMATING REGIONAL BACKGROUND CONCENTRATIONS OF $PM_{2.5}$ AND VERIFYING LOCAL SOURCE CONTRIBUTIONS IN A DATA POOR ENVIRONMENT

Eve L. Draper[1], J. Duncan Whyatt[2], Richard Taylor[3] and Sarah E. Metcalfe[1]

[1]School of Geography, University of Nottingham, University Park, Nottingham, NG7 2RD, UK
[2]Lancaster Environment Centre, Lancaster University, Bailrigg, Lancaster, LA1 4YQ, UK
[3]Nottingham City Council, Loxley House, Station St., Nottingham, NG2 3NG, UK

Abstract: Long- and short-term exposure to $PM_{2.5}$ is linked to adverse health impacts. Compared to other pollutants, $PM_{2.5}$ is monitored at far fewer stations across the UK's Automatic Urban Rural Network (AURN) hence models are widely used to simulate concentrations. A significant proportion of local $PM_{2.5}$ may be attributed to regional sources, so an accurate estimation of that component is necessary to ensure that the model gets the 'right results for the right reasons'. This study explores alternative methods of deriving background $PM_{2.5}$ concentrations in Nottingham, UK, to represent the regional component in an atmospheric dispersion model (ADMS-Urban). A model variation employing hourly background concentrations from AURN stations located up to 200km upwind of the city performed best when verified against $PM_{2.5}$ concentrations for two monitoring locations in the city. This study also uses alternative verification datasets to assess model performance in a data poor environment. This gives us confidence to apply ADMS-Urban more widely across the city to assess associations between $PM_{2.5}$ and human health.

Key words: *$PM_{2.5}$, ADMS, background sources, hyperlocal sources.*

INTRODUCTION

Particulate matter with an aerodynamic diameter of ≤2.5µm ($PM_{2.5}$) is associated with many health conditions including cardiovascular diseases, respiratory diseases and reduced cognitive function (Southerland *et al*., 2022). In spite of this, in the UK, $PM_{2.5}$ is only measured at 79 of the 171 Automatic Urban and Rural Network (AURN) monitoring sites, which is far fewer than for other pollutants e.g. NO_2, measured at 145 (Defra, 2022). The association between short-term peaks in $PM_{2.5}$ and adverse health effects has been recognised, hence in the past there has been a focus on monitoring in 'hot-spot' areas, e.g. busy roads (Harrison *et al*., 2012). Research has also demonstrated the risks of long-term exposure to fine particles on human health, but monitoring has not increased based on this understanding (Southerland *et al.*, 2022). This means there is heavy reliance on air quality models to predict $PM_{2.5}$ concentrations over a city-wide scale for the purposes of decision making and human health assessments (Ortiz and Freidrich, 2013).

$PM_{2.5}$ can be a primary pollutant, from exhaust emissions, tyre and brake wear, or emissions from industrial and household combustion. However, some studies have identified that a significant amount (41% - 72%) of $PM_{2.5}$ in the UK is secondary, which is 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, also contributes to the $PM_{2.5}$ load in the UK (Graham *et al.*, 2020). Rural AURNs provide an indication of transboundary $PM_{2.5}$ contributions whilst urban AURNs reflect additional local contributions. It is essential to get the background and local proportions accurate in local-scale air quality modelling so that local sources can be targeted for management accordingly (Ortiz and Friedrich, 2013).

This study explores methods for determining background concentrations of $PM_{2.5}$ for use in an atmospheric dispersion model (ADMS-Urban 5.0) for local-scale $PM_{2.5}$ modelling in Nottingham, using wind direction as a predictor for background concentration. Methods for overcoming hyperlocal source contributions for the purposes of model verification are also presented.

Nottingham, UK

In Nottingham, there is one urban background AURN station that measures both $PM_{2.5}$ and PM_{10} (city centre), and one roadside station that measures PM_{10} only (Western Boulevard) (Figure 1a). In line with the objectives of the AURN, these were established to assess compliance with the Ambient Air Quality Directives and measure reduction of pollutants over time. Nottingham has an annual mean $PM_{2.5}$ of 12 μg/m^3 of which 8 μg/m^3 (67%) may be attributed to background sources (Nottingham City Council, 2018).

METHODOLOGY

Determining a suitable background dataset for model input

Four different approaches to estimating background concentrations of $PM_{2.5}$ were developed, based on data from one rural and several urban background AURN sites (Figure 1b). The year 2019 was used as this is the most recent year that was not affected by emission reductions due to the COVID-19 pandemic national lockdowns:

1. Model 1: Used hourly background data from a rural AURN, Chilbolton Observatory (southern England). This site was chosen as it was the closest rural AURN site to Nottingham that measures hourly $PM_{2.5}$ concentrations.
2. Model 2: The Chilbolton AURN values were scaled using average annual background values in rural areas outside of the Nottingham conurbation generated by the Pollution Climate Mapping (PCM) model (Defra, 2019). A single annual value was applied to uplift all hourly values from the Chilbolton AURN so they were more reflective of the $PM_{2.5}$ background climate in Nottingham (Zhong *et al.*, 2021).
3. Model 3: Hourly background data were taken from other urban background AURNs within 200km of Nottingham using wind direction as a predictor for background concentrations. Model 3 selected stations based on 8 x 45° sectors upwind of Nottingham.
4. Model 4: Takes the same approach as Model 3 but selects stations based on 12 x 30° sectors upwind of Nottingham.

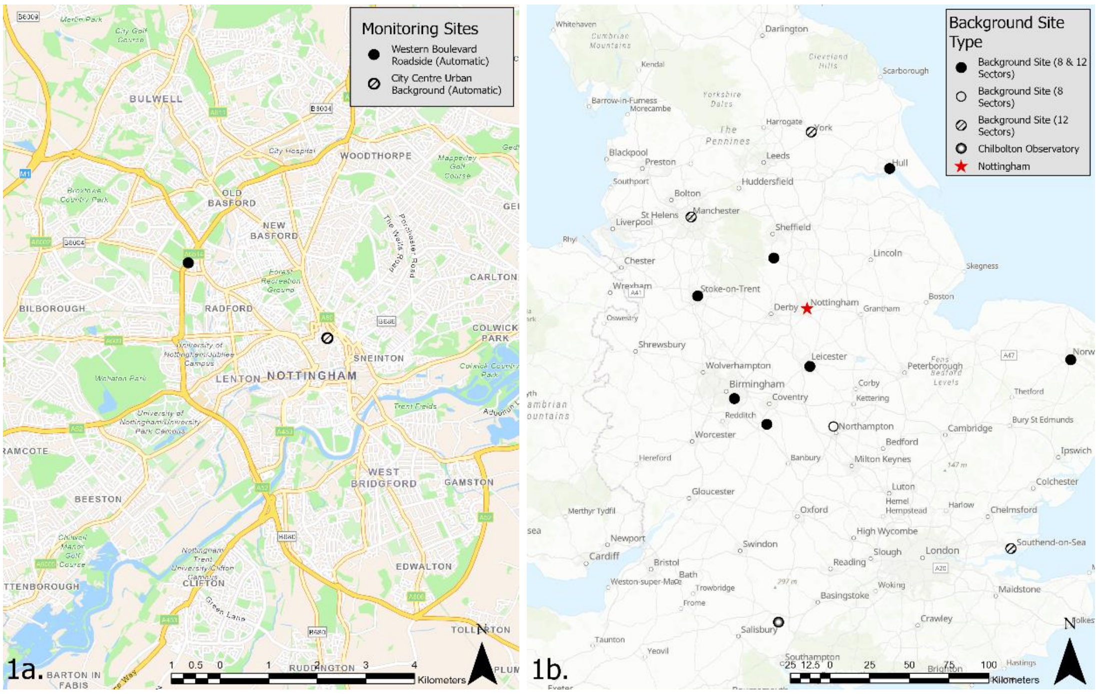

Figure 1a: Nottingham AURN sites. **Figure 1b:** Background AURN locations used in models.

Model verification

Both AURN sites in Nottingham were used to verify $PM_{2.5}$ models (Figure 1a). Analysis of the city centre site prior to verification revealed a hyperlocal source of $PM_{2.5}$, which was a mobile hot food stall not representative of the wider $PM_{2.5}$ climate in Nottingham. Correspondence with the Local Authority revealed that the food outlet started operating in 2015. The lunchtime peaks present a very different temporal signature from expected local sources, such as nearby roads, that typically follow a diurnal pattern of the morning and evening rush-hours (Kendrick *et al.*, 2015) (Figure 2). For the purposes of model verification, data during periods whilst the hot food stall was operating were removed from the verification dataset.

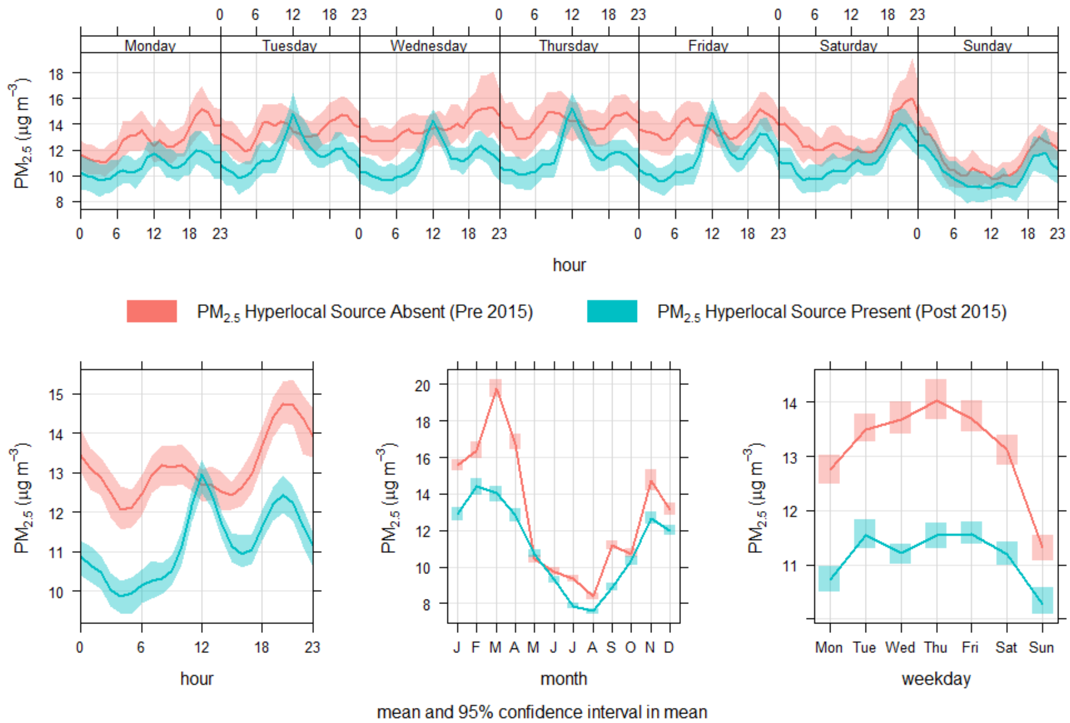

Figure 2: Time variation of $PM_{2.5}$ at the city centre AURN, pre (2008-2014) and post operation (2015-2019) of the hot food stall.

$PM_{2.5}$ was estimated at Western Boulevard (Figure 1a) by applying a $PM_{2.5}$:PM_{10} ratio from the city centre site 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. Hourly concentrations of the model outputs were verified against recorded hourly $PM_{2.5}$ concentrations at city centre, and hourly estimated $PM_{2.5}$ concentrations at Western Boulevard.

Model performance for $PM_{2.5}$ was tested using fraction of predictions within a factor of two (FAC2), mean bias (MB), mean gross error (MGE), normalised mean bias (NMB), correlation coefficient (r), and index of agreement (IOA).

RESULTS

Model 3 performed best for FAC2, MGE, r and IOA at both the city centre and Western Boulevard sites (Table 1). The city centre site has fewer data points for $PM_{2.5}$ than Western Boulevard due to the removal of hours influenced by the hot food stall.

Table 1: Model statistics of model performance for $PM_{2.5}$ at city centre and Western Boulevard AURN sites (hourly data). n is the number of hourly data points tested in the analysis.

Model	n	FAC2	MB	MGE	NMB	r	IOA
City Centre							
Model 1	6676	0.77	0.46	4.70	0.04	0.70	0.65
Model 2	6676	0.70	3.43	5.97	0.32	0.70	0.56
Model 3	6676	0.81	2.05	4.24	0.19	0.80	0.68
Model 4	6676	0.60	5.62	7.28	0.53	0.69	0.46
Western Boulevard							
Model 1	8481	0.81	-0.54	4.60	-0.05	0.63	0.61
Model 2	8481	0.78	2.35	5.72	0.20	0.62	0.51
Model 3	8481	0.86	1.03	4.27	0.09	0.72	0.64
Model 4	8481	0.72	4.74	6.60	0.41	0.64	0.44

DISCUSSION AND CONCLUSIONS

This study used four different datasets to represent background $PM_{2.5}$ concentrations in ADMS-Urban. Model 3, applying urban background AURNs in eight 45° wind sectors using wind direction as a predictor for $PM_{2.5}$ concentration performed most strongly across a range of test statistics at the two verification sites in the city, including FAC2 - a model is considered as acceptable when more than half of the model predictions lie within a factor of two (Derwent *et al.*, 2010). MB and MGE provide useful measures of model over- and under-estimation. Model 1 performed best for MB. Over-estimation in Model 3 may be attributed to local contributions from other urban AURN sites included within the background data input for Nottingham (Derwent *et al.*, 2010). Model 3 performed best for MGE at both sites. NMB is a measure of relative difference between modelled and observed concentrations. Model 1 performs best in this metric, however Model 3 is within the accepted range of -0.2 and +0.2 for both sites, whilst Models 2 and 4 are not (Derwent *et al.*, 2010). There is good overall agreement for modelled and observed concentrations (r = 0.80, city centre and r = 0.72, Western Boulevard) for Model 3, which performed the best in this metric out of the four models. Model performance is slightly poorer at the Western Boulevard site than the city centre. There could be two explanations for this: First, modelled concentrations at Western Boulevard are compared to concentrations based on a predicted $PM_{2.5}$: PM_{10} ratio; second, there are fewer observations at the city centre site, meaning that r may appear stronger due to a smaller number of pairings (Derwent *et al.*, 2010). Lastly, IOA is a good indicator for overall model performance, Model 3 performed the best out of the four models in this metric for both sites, giving confidence to apply this model across the city to assess relationships with health outcomes (Willmott *et al.*, 2012).

Annual mean modelled concentrations are typically used to assess relationships between 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). These typically occur at regional scales, dominated by background $PM_{2.5}$ from mainland Europe and other conurbations in the UK, 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). The method used to generate background datasets for Models 1 and 2 supports the prediction of annual mean concentrations, but not episodes. In contrast, the method used to generate background datasets for Model 3 can be used to predict both annual mean and episode-specific concentrations so can be used in assessments of chronic and acute health impacts.

Verifying model performance is challenging in Nottingham. There is one site in the city centre that measures $PM_{2.5}$ which is unlikely to be representative for a population of >300,000 residents. The city centre site is also influenced by a hyperlocal source, not reflective of general conditions across Nottingham, limiting the observations available for model verification. From a wider policy perspective, the influence of hyperlocal sources on monitoring stations may affect the reporting required in accordance with Air Quality Directives. Hyperlocal sources may obscure general reductions in $PM_{2.5}$ due to national and local emission reduction interventions (Figure 2). This confirms the need to carry out rigorous assessment of monitoring data prior to use in model verification. Provision of robust statutory guidance for requirements of siting potential sources, for example, mobile hot food outlets, near to air quality monitoring stations is needed to prevent this issue.

It is difficult to assess the impacts of air pollution from monitoring alone, hence the importance of using a modelling approach to achieve better spatial representation (Conti *et al.*, 2017). Through careful scrutiny of pollutant measurements and assessment of background concentrations it is possible to generate modelled $PM_{2.5}$ concentrations at spatial and temporal scales consistent with available health outcome data to make a more rigorous assessment of associations between air pollution and human health.

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