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Modelling the dispersion of particle number concentrations in the West Midlands, UK using the ADMS-Urban model

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ABSTRACT

Ultrafine particles (UFPs) are respirable particles with a diameter less than 100 nm, which some studies have associated with adverse effects upon health. UFPs are currently not regulated as the health evidence is insufficient and very few observational data are available in most cities. The 2021 WHO Global Air Quality Guidelines highlighted the pressing issue of UFPs and provided a good practice statement for UFPs, which recommends that more measurement and modelling studies are implemented in future. Particle number concentrations (PNC) are the most common metric for UFPs as this fraction normally dominates the total ambient PNC in urban environments. This study simulates the dispersion of particle number concentrations in the West Midlands (a metropolitan area), UK using the local scale ADMS-Urban model, which is an advanced quasi-Gaussian plume dispersion modelling system. ADMS-Urban implements a physics-based approach to represent the characteristics of the atmospheric boundary layer and has been widely used in the dispersion modelling of air pollutants. It can represent a variety of source types (such as road and grid emissions) occurring in urban environments and requires a range of input data. Particle number was used as a passive scalar, with no inclusion of aerosol microphysics within the model, as a first implementation in the ADMS-Urban model for the West Midlands, UK. Evaluation was conducted by comparing the modelled (from a receptor run) and measured data at the Birmingham Air Quality Supersite. Overall, the model performed well although there was a slight underestimation for PNC. Based on the modelling output from a contour run, PNC maps at a variety of spatial scales (i.e. street scale, ward level and local authority level) and temporal resolutions (i.e. annual, 24-hour, and 1-hour) were generated. PNC mapping could be linked to local population and health data for potential epidemiological studies.

1. Introduction

Ultrafine particles (UFPs with a diameter less than 100 nm) are tiny and respirable particles (Manigrasso et al., 2017). Because of their small sizes, UFPs can penetrate cells and tissue, accumulate in lungs, and cause health effects (Geiser et al., 2005, Panis et al., 2010). Although UFPs are currently not regulated in the same way as mass concentrations for larger particles such as $PM_{2.5}$ and PM_{10} (with diameters less than 2.5 µm and 10 µm, respectively), the 2021 WHO Global Air Quality guidelines (WHO, 2021) have highlighted the pressing issue of UFPs with a Good Practice Statement to guide stakeholders and researchers for future practices to reduce UFP concentrations. UFPs may possess a larger potential for adverse health effects than larger particles (e.g. $PM_{2.5}$ and PM_{10}). Particle number concentrations (PNC) are the most common measure for UFPs with tiny individual and almost negligible collective mass (Harrison et al., 2000). UFPs often dominate the total ambient PNC in urban environments, especially in the near source regions (Kumar et al., 2014). Road transport and other types of combustion are the predominant primary sources of UFPs in urban areas (Kumar et al., 2011, Zhong et al., 2020). Secondary particle formation mostly from nucleation and particle growth processes may be of importance at longer time scale (hours to days) (Reche et al., 2011).

UFP measurement can provide direct information of PNC in ambient air, which is useful in the evaluation of models. However, the current

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UFP measurements are sparse and local understanding of UFPs is very limited (WHO, 2021). It is necessary to expand UFP monitoring within the existing traditional air quality monitoring network. The largest automatic monitoring network in the UK is the Automatic Urban and Rural Network (AURN) (Defra, 2019). There are only 3 AURN sites (i.e. Chilbolton, London Honor Oak Park and London Marylebone Rd) measuring PNC in the UK. The lack of PNC monitoring may be due to the fact that PNC is not regulated at the moment and PNC measurements can be complex (Ketzel et al., 2021).

The variability of PNC of UFPs in the ambient urban air may be influenced by combined effects of UFP emissions, dispersion, deposition and aerosol dynamics (Kumar et al., 2014, Zhong et al., 2020). The dispersion processes depend on meteorological conditions (e.g. wind speed and direction, temperature, cloud cover etc) and the ventilation of cities, which is determined by the urban morphology which controls the exchange of air pollutants within the urban canyon layer and the overlying cleaner atmosphere (Hood et al., 2014, Zhong et al., 2017). UFPs may undergo non-linear aerosol dynamic processes, e.g. nucleation, coagulation, condensation and evaporation, which may change PNC on a range of timescales (Ketzel et al., 2021). In urban environments, emissions and dispersion are the most important processes (Wang et al., 2010, Ketzel and Berkowicz, 2005), compared with most aerosol dynamic processes.

A number of modelling studies have been conducted to predict PNC at different spatial and temporal scales. Zhong et al. (2018) conducted street canyon box modelling (considering condensation and evaporation processes of UFPs and canyon ventilation) to capture the significant contrast of PNC in street canyon compartments. This is limited to the a single street canyon environment. Nikolova et al. (2018) adopted a Lagrangian CiTTy-Street model with a multicomponent aerosol microphysics module to simulate PNC on the neighbourhood scale. Only a small time scale (in the order 100 s) was considered. Zhong et al. (2020) further coupled this multicomponent aerosol microphysics module with the WRF-LES model (Weather Research and Forecasting modelling system and large eddy simulation) to simulate Neighbourhood-scale dispersion of UFPs. This study is also limited to smaller region of 2.54 km × 2.54 km with only consideration of traffic-induced PNC. Kukkonen et al. (2016) conducted PNC modelling for five European cities at the regional background scale using the LOTOS-EUROS model and at the local scale using an urban-scale dispersion modelling of vehicular emissions. However, the effects of urban buildings were not explicitly modelled and particle dynamics were not considered in their urban-scale modelling. Ketzel et al. (2021) predicted long-term PNC at the local and street scale using the Danish air quality modelling system DEHM/UBM/AirGIS. However, particle dynamics were also not considered. Frohn et al. (2021) adopted the regional and urban models of the DEHM/UBM/AirGIS modelling system to simulate the regional background of PNC for the urban model. Although the model reproduced the patterns in the observations, the improvement of emission inventories is still needed.

Current PNC dispersion modelling has been limited to small areas (such as street canyon or neighbourhood scales) with a lack of comparison analysis at different spatial and temporal scales especially linking with health studies and potential regulation as highlighted by the 2021 WHO Global Air Quality guidelines. This current study will fill this gap. The paper aims to simulate the dispersion of particle number concentrations in the West Midlands (a metropolitan area), UK and provide street scale resolution maps of PNC for potential epidemiological studies. Section 2 provides a description of the modelling approach. Section 3 reports the results from the modelling at different spatial and temporal resolutions. Section 4 discusses the implication of the results. Section 5 presents the conclusions.

2. Methods

2.1. The ADMS-Urban modelling system

The model used in this study is an advanced quasi-Gaussian plume dispersion modelling system, called ADMS-Urban. The system was developed by Cambridge Environmental Research Consultants (CERC) (Owen et al., 2000, Hood et al., 2018) and has been widely used for air quality management and assessment of interventions and policies. ADMS-Urban implements a physics-based approach to represent the characteristics of the atmospheric boundary layer by using two governing parameters, i.e. Monin–Obukhov length and boundary layer height, rather than a single imprecise Pasquill-Gifford stability parameter (Arciszewska and McClatchey, 2001). It adopts normal quasi-Gaussian plume distributions in neutral and stable atmospheric conditions, while non-Gaussian vertical profiles are created in convective atmospheric conditions to represent the skewed nature of turbulence which causes high surface concentrations close to emission sources (Righi et al., 2009, Riddle et al., 2004).

ADMS-Urban can represent a variety of emission source types (such as explicit point, road, and grid emission sources) occurring in urban environments. The model can resolve concentration gradients in the vicinity of emission sources. It can model large industry sources as elevated point sources, which requires the inputs of stack parameters (e.g. exit velocity, efflux temperature, stack height and diameter). Road sources are represented by line sources with traffic-induced turbulence effects on local dispersion (CERC, 2021a). Regular grid sources (with uniform depth) are used to represent less detailed emissions sources, e.g. residential combustion, agriculture, minor industrial processes, waste treatment and disposal.

ADMS-Urban has several other modules, e.g. chemistry, urban canopy and advanced street canyon. The model accounts for fast photochemistry using Generic Reaction Set chemistry scheme for gas-phase air pollutants (Venkatram et al., 1994), which is not activated in this study. It has an urban canopy module (Hood et al., 2014), which estimates turbulence flow and wind speed profiles based on the spatially varying surface roughness length as derived from building data. It also has an advanced street canyon module, which can model the effects of street canyons for both asymmetric and symmetric, and the channelling and circulation of air flow for street canyon environments (Hood et al., 2021). The urban canopy and advanced street canyon modules are used in this study. A range of model input data are needed to run the model, e.g. emissions inventory, time varying emission profiles, background and meteorological data. These model inputs used in the case study for the UK West Midlands which will be briefly described below. The ADMS-Urban model has been applied to simulate the dispersion of traditional air pollutants (NO_2 and $PM_{2.5}$) for the West Midlands, UK (Zhong et al., 2021, Zhong et al., 2023). This current modelling study will extend the capability to explore the dispersion of PNC.

2.2. PNC dispersion modelling case study for West Midlands

2.2.1. Study area

The study area covers the West Midlands region (See Fig. 1), which includes 7 local authorities, i.e. Birmingham, Coventry, Dudley, Sandwell, Solihull, Walsall and Wolverhampton. West Midlands has a geographical area of about 902 km^2 with a population of 2.9 million. West Midlands is a metropolitan county (an administrative county with combined authorities, and heavily populated and urbanised). The component authorities have population densities ranging from 1,200/km^2 (Solihull) to 4,300/km^2 (Birmingham), typical of the range of UK cities outside of London (Wikipedia, 2023). It is unusual within the UK in having major motorways pass through, rather than around it. Birmingham is the second largest city in the UK. Birmingham has implemented a Clean Air Zone (BCC, 2021) for the city centre since June 2021 in order to tackle air pollution issues. Air pollution modelling can serve as a

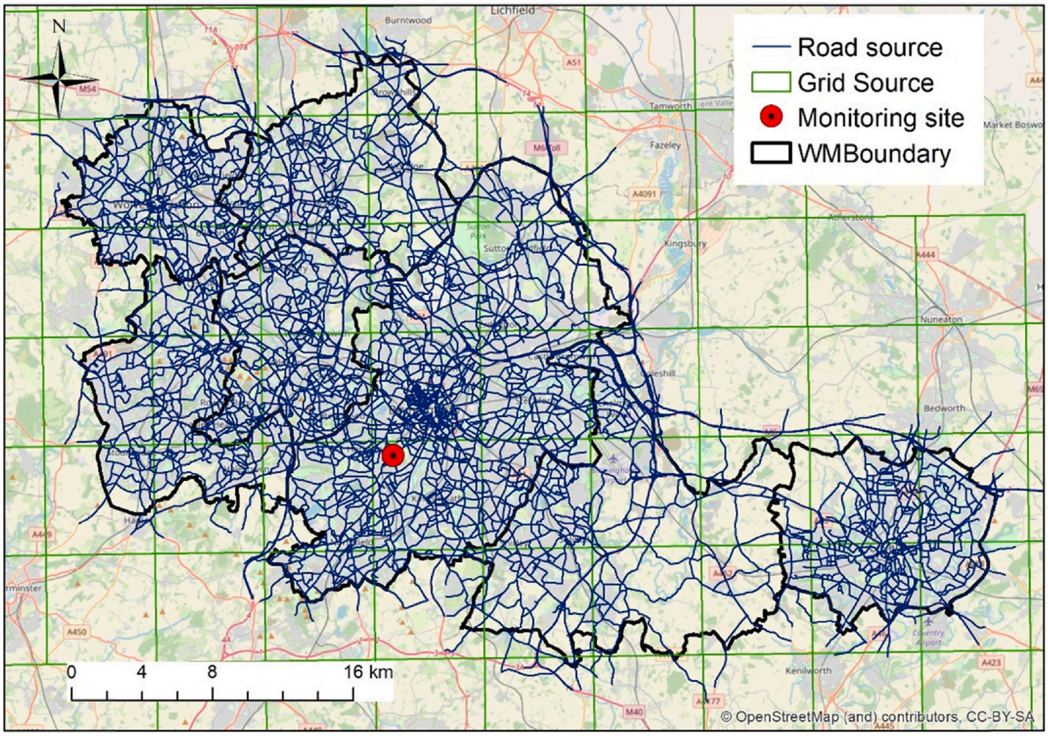

Fig. 1. Emission sources implemented in the ADMS-Urban model over the West Midlands, which covers 7 local authorities, i.e. Birmingham, Coventry, Dudley, Sandwell, Solihull, Walsall and Wolverhampton. The monitoring site at the Birmingham Air Quality Supersite (BAQS) is also indicated.

predictive tool for air pollution management for the region. The baseline modelling year in this study is 2019, which is the last normal year before Covid 19.

2.2.2. Birmingham air quality supersite

For the West Midlands, there was only 1 site with available PNC observation data, i.e. Birmingham Air Quality Supersite (BAQS indicated in Fig. 1). BAQS represents urban background site and is located at the University of Birmingham campus (lat: 52.45, lon:-1.93), with a distance of about 3 km southwest from the Birmingham City Centre. PNC data in the size range of 12–552 nm was measured by a scanning mobility particle sizer (SMPS, model TSI 3082) (Bousiotis et al., 2021).

2.2.3. Emission inventory

Unlike emission inventories for traditional air pollutants (e.g. NO_2 and $PM_{2.5}$, which can be available via the UK National Atmospheric Emissions Inventory with high spatial resolution of 1 km × 1 km), there are limited sources for the emission inventory of particle number for UFPs. Such an emission inventory has been developed by TNO in the RI-Urbans (Research Infrastructures Services Reinforcing Air Quality Monitoring Capacities in European Urban & Industrial AreaS) EU project. Emissions of particle number are calculated based on 250 different UFP source categories, aggregated to the TNO-GNFR classification, which is similar to the CAMS-REG inventory (Kuenen et al., 2022). For some source sectors with available necessary information (mostly transport-related sources), direct emission factors of particle numbers were used. For other source sectors (mostly industrial sources), particle number emissions were estimated based on $PM_{2.5}$ mass emissions and mass size distribution data, to first estimate the $PM_{0.3}$ mass fraction of $PM_{2.5}$, and thus eliminate the coarser part of $PM_{2.5}$ that is not relevant for particle numbers. Next, particle number below 300 nm was estimated based on representative particle number size distributions as found from the literature and particle geometry data (TNO, 2023). Almost all number size distributions found in literature peak at particle diameters well below 300 nm. The resulting emission inventory of particle number covers a total size range of 10 to 325 nm (aerodynamic) with 15 size bins at 6 km × 6 km resolution (See Fig. 1 for the illustration of the West Midlands region). The reference year of the inventory is 2018 and it covers both solid and semi-volatile particles. Traffic emissions for instance, especially those from diesels comprise both solid non-volatile particles and semi-volatile droplets formed during engine exhaust dilution (Harrison et al., 2018). We assume that there are nearly no changes for emissions between 2018 and 2019, so the 2018 emission inventory can be used for 2019 baseline year modelling. Fig. 2 shows the summary of emission sources of primary particle numbers for different SNAP (Selected Nomenclature for Air Pollution) sectors (derived based on the TNO GNFR sectors originally in the emission inventory; See Table S1. for the classification between SNAP and GNFR sectors) for the UK West Midlands. It reflects that road transport sector (SNAP07) is the largest source for particle number concentrations in this region, also with substantial contributions from Other Transport and Mobile Machinery (SNAP08: Birmingham Airport is located 13 km east-southeast of Birmingham city centre), Combustion in Commercial, Industrial, Residential and Agriculture (SNAP02), and Waste Treatment and Disposal (SNAP09: there is a very large waste incinerator at Tyseley, which is about 4 km to the east of Birmingham city centre). The exchange of the SNAP08 and SNAP09 shares in Fig. 2 between the two size fractions is due to aviation being part of SNAP08 and having a large emissions of particles <100nm, and the waste incinerator contributing particles mostly in the larger fraction in SNAP09.

For road transport sources, we simulated major roads as explicit sources for the West Midlands (shown as Fig. 1), with non-resolved minor roads remaining in grid emissions of the SNAP07sector to avoid double counting of road transport emissions. In other words, SNAP07 in Fig. 2 is split into explicit major road emissions and unresolved minor road grid emissions. The traffic network modelling data was obtained from Transport for West Midlands and Birmingham City Council, as previously used in Zhong et al. (2021) for ADMS-Urban modelling for traditional air pollutants. The traffic flows included activities for heavy vehicles, light vehicles and buses. The 2019 regional traffic fleet data

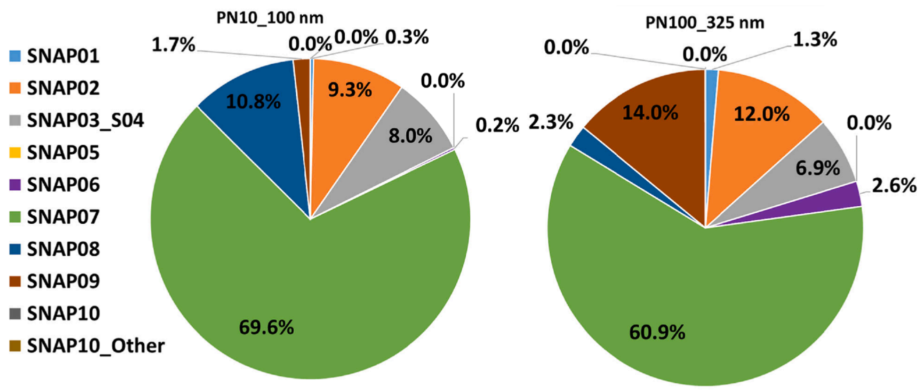

Fig. 2. Summary of emission sources of primary particle numbers for different SNAP sectors for West Midlands, UK. PN10_100nm: PN between 10 and 100 nm. PN100_325nm: PN between 100 and 325 nm. SNAP01: Combustion in Energy Production and Transformation, SNAP02: Combustion in Commercial, Industrial, Residential and Agriculture, SNAP03: Combustion in Industry,_SNAP04: Production Processes, SNAP05: Extraction and Distribution of Fossil Fuels, SNAP06: Solvent Use, SNAP07: Road Transport, SNAP08: Other Transport and Mobile Machinery, SNAP09: Waste Treatment and Disposal, SNAP10: Agriculture, Forestry and Landuse Change, SNAP10_Other: Agriculture_Other (Table S1).

was used to further split traffic flow into detailed Euro class-based vehicle categories. Emission factors of particle number for different vehicle categories adopted in Ketzel et al. (2021) were combined with traffic activity and fleet data to derive emission rates inputs for the ADMS-Urban model. Both explicit road transport emissions and grid emissions for particle numbers were pre-processed using EMIT (Atmospheric Emissions Inventory Toolkit) (CERC, 2021b) to produce ADMS-Urban format modelling inputs.

2.2.4. Time varying emission profiles

Time varying emission profiles (hour in day, day in the week and month in year) for grid emissions of different sectors were also available together with the TNO emissions inventory (Section 2.2.2), which can be used to scale annual emissions to hourly emissions as the model will run at hourly resolution. For explicit traffic emissions, major roads were categorised in to low, medium and high traffic flow based on measured hourly traffic activity and time varying profiles were applied for each road category to derive hourly traffic emissions, similar to those in Zhong et al. (2021).

2.2.5. Background input

The number of AURN sites for PNC in the UK is very limited. An appropriate background site with available PNC data for the year of 2019 is Chilbolton (lat: 51.149617, lon: −1.438228; It is roughly at ~ 149 km distance from the University of Birmingham). Chilbolton is the only rural background site for PNC available from the UK-Air website (Defra, 2019). For some periods during the year without available PNC, we used NO_x or $PM_{2.5}$ concentrations to estimate PNC based on available data for other periods during the year. A background input file for PNC was then generated and formatted for the ADMS-Urban model.

2.2.6. Meteorological input

The meteorological inputs required by the model include wind speed, wind direction, and cloud cover (alternatively Monin-Obukhov length or surface heat flux if known), air temperature, relative humidity and precipitation (Arciszewska and McClatchey, 2001), which are normally measured by weather stations. Birmingham Airport site is a representative synoptic site for this case study of the West Midlands. The meteorological data for Birmingham Airport site was extracted from the Met Office MIDAS dataset in the CEDA Archive (Met_Office, 2019).

2.2.7. Urban canopy and street canyon input

To generate inputs for the urban canopy and street canyon modules, two shapefiles for road centreline locations and building data (with a field for building height obtained from Digimap) were processed using ArcGIS tools developed by CERC (Jackson et al., 2016). Gridded building height and density parameters were used for the calculation of urban canopy flow field. Street canyon properties at each side were used for the calculation of street canyon effect on local dispersion for road emissions.

2.2.8. Computational approach

A novel task farming approach was implemented in ADMS-Urban v5.0.0.1 and was successfully tested in Zhong et al. (2021). Task farming enables the parallel running of the same or sequential code with different modelling parameters and inputs on multiple cores on supercomputer clusters or a PC network. Such an approach is specifically adopted in the ADMS-Urban modelling for a larger urban area, which can be spatially split into smaller sub-regions to optimise the overall computational time. The computational domain for the West Midlands was spatially split into 540 subdomains (same as that in Zhong et al. (2021)), with a size of 2 km × 2 km for most subdomains, and 1 km × 1 km or 500 m × 500 m for city centre areas with a denser road network. Each subdomain was also applied with a buffer zone of 750 m for major road sources.

3. Results

3.1. Model evaluation

The ADMS-Urban model was firstly configured as a "Receptor" mode for the receptor location where the air quality measurement site (i.e. BAQS indicated in Fig. 1) was located. The modelled hourly output for PNC was generated for BAQS, which was then imported into the Model Evaluation Toolkit (Stidworthy et al., 2018) together with the measured PNC for the evaluation of model performance.

Fig. 3 shows a comparison of annual averages of PNC_{10_100} and PNC_{100_325} at BAQS between the model and measurement for the year of 2019 using scatter plots. The model underestimated annual PNC by 14.3 % for PNC_{10_100} and by 11.9 % for PNC_{100_325}, respectively (See details in Table 1). Fig. 4 further shows a comparison of scatter plots for daily mean PNC_{10_100} and PNC_{100_325}. The model captured most of the features in daily means for both PNC_{10_100} and PNC_{100_325}.

Fig. 5 presents frequency scatter plots of hourly data between the model and observation at BAQS for PNC_{10_100} and PNC_{100_325}. For PNC_{10_100}, there is a large spread occasionally for high concentrations. A high density of counts lies within the factor of 2 lines, indicating that the model in general can capture these as observed. For PNC_{100_325}, there is generally good agreement for the most likely PNC concentrations, indicated by the red points lie within the gradient = 2/0.5 lines.

Fig. 6 illustrates a time variation comparison between the model and

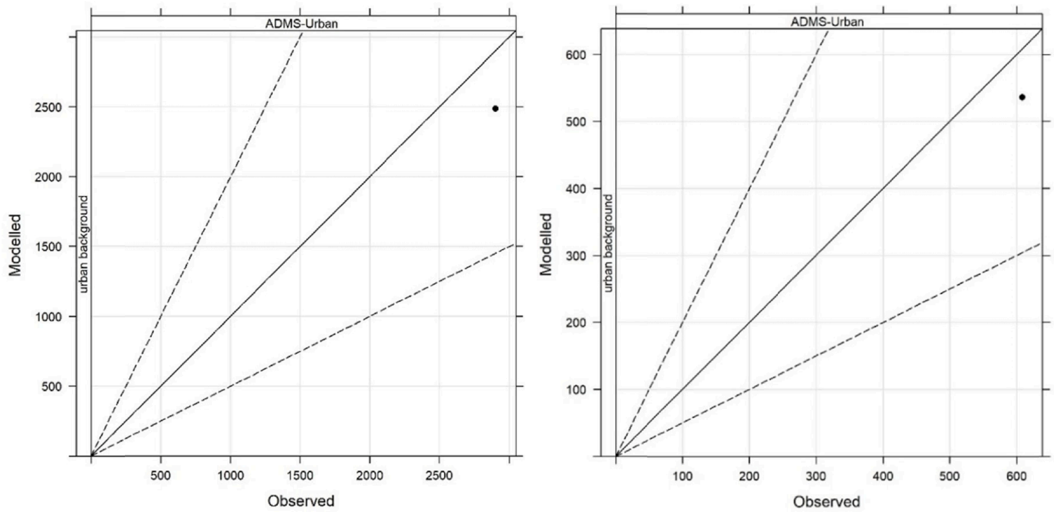

Fig. 3. Annual averages comparison between the model and observation (for the same period where both are available) at BAQS for (a) PNC between 10 and 100 nm (PNC_{10_100}, unit in # cm^{-3}) and (b) PNC between 100 and 325 nm (PNC_{100_325}, unit in # cm^{-3}) for the 2019 business-as-usual (BAU) case.

Table 1
Model evaluation statistics for the 2019 BAU case. Obs: measured PNC; Mod: modelled PNC; Fb: fraction bias; Fac2: fraction of modelling output within a factor of 2 of measurement; NMSE: normalised mean square error; R: correlation coefficient.

Pollutant	Site Type	Obs (# cm^{-3})	Mod (# cm^{-3})	Fb	Fac2	NMSE	R
PNC_{10_100}	urban background	2902	2487	−0.15	0.71	0.67	0.47
PNC_{100_325}	urban background	608	536	−0.13	0.73	0.89	0.49

(a) (b)

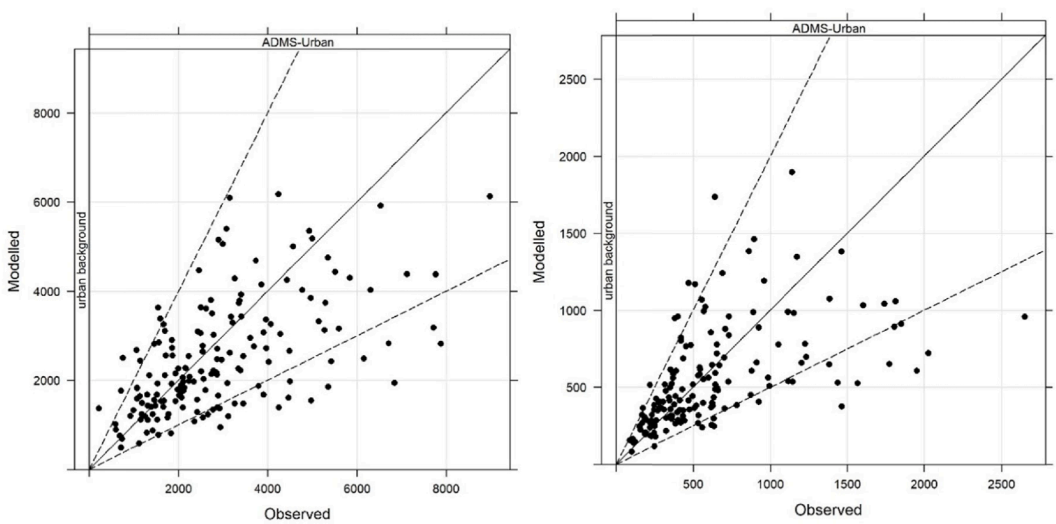

Fig. 4. Daily mean comparison between model and observation (for the same period where both are available) at BAQS for (a) PNC between 10 and 100 nm (PNC_{10_100}, unit in # cm^{-3}) and (b) PNC between 100 and 325 nm (PNC_{100_325}, unit in # cm^{-3}) for the 2019 business-as-usual (BAU) case.

observations at BAQS for PNC_{10_100} (See Figure S1 for PNC_{100_325}). The model typically captured the diurnal patterns for PNC_{10_100} and PNC_{100_325}, reflecting both morning and evening peaks related to traffic. The underestimation of PNC at night by the model may be due to the underestimation of residential combustion emissions (Font et al., 2022) and the missing representation of intensive cooking activities during the night. The mean daily profile over the week shows an underestimation of PNC_{10_100} by the model, and a variation of PNC_{100_325} between the model and observation. The monthly profile varies substantially between the model and observations, but there is good agreement of overall averages of PNC_{10_100} and PNC_{100_325} between the model and observation (indicated as Table 1). As BAQS is an urban background site, modelled concentrations at BAQS are heavily influenced by the incoming background input data (here from the Chilbolton site), as well as the contribution from local urban emissions (in which traffic emissions are dominant as shown in Fig. 2) (Figure S2). This can lead to discrepancies in the timing of peaks at BAQS due to the large distance of the Chilbolton site, which is predominantly on the upwind sector, but several hours away for an approaching airmass, and may also be downwind. With a more proximate and hence representative

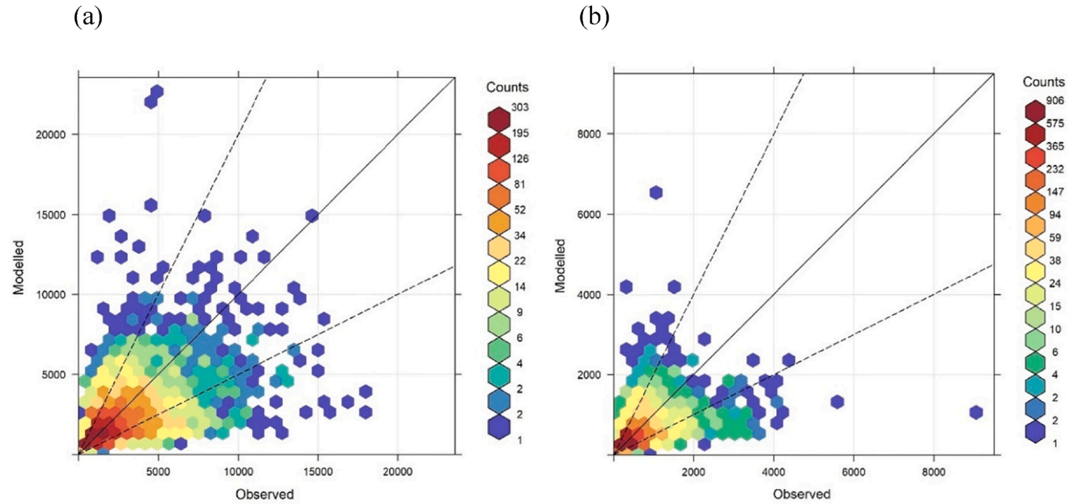

Fig. 5. Frequency scatter plots between the model and observation at BAQS for (a) PNC between 10 and 100 nm (PNC_{10_100}, unit in # cm^{-3}) and (b) PNC between 100 and 325 nm (PNC_{100_325}, unit in # cm^{-3}) for the 2019 business-as-usual (BAU) case.

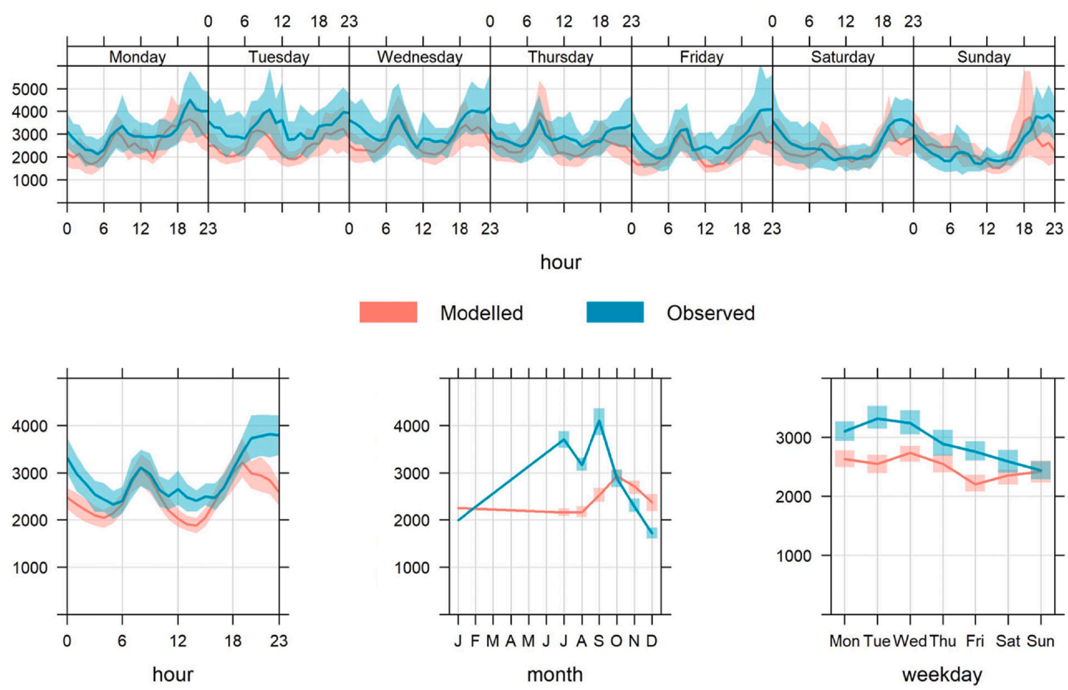

Fig. 6. Time variation comparison between the model and observation (for the same period where both are available from July to December 2019) at BAQS for PNC between 10 and 100 nm (PNC_{10_100}, unit in # cm^{-3}) for the 2019 business-as-usual (BAU) case.

background site as the model inputs, the model performance would be expected to improve.

Table 1 shows detailed model evaluation statistics. The model generally underestimates both PNC_{10_100} and PNC_{100_325}, compared with observation. Fb (Fractional bias) is a measure of how modelled mean concentration differs from these measured. Fb is calculated as −0.15 for PNC_{10_100} and −0.13 for PNC_{100_325}, respectively. Fac2 is a measure of how many modelled data is within a factor of 2 of the measured data. Fac2 has a value of 71 % for PNC_{10_100} and 73 % for PNC_{100_325}, respectively. NMSE (normalised mean square error) is a measure of normalised mean difference between the model and measurement. NMSE is 0.67 for PNC_{10_100} and 0.89 for PNC_{100_325}, respectively. R (Pearson's correlation coefficient) measures the extent of how the modelled data is correlated with measurement. R is calculated as 0.47 for PNC_{10_100} and 0.49 for PNC_{100_325}, respectively.

3.2. PNC concentration mapping

In order to generate PNC concentration maps, we need to run the ADMS-Urban model in the "Contour" mode. It covers intensive output points (~0.61 million covering regular grids and denser resolution near road sources) over the West Midlands region with a buffer zone of 1 km. For a big computational domain like the West Midlands, the splitting option using the task farming approach needs to be adopted and the domain was divided into 540 subdomains. The Linux version of the ADMS-Urban model was run on the supercomputer at the University of Birmingham using 540 cores each for a subdomain. The overall elapsed time for a typical annual simulation was about 22 h. The output netcdf files for all subdomains were then combined and interpolated to a big netcdf file (with a size of 83 GB) with all output points covering the whole West Midlands at every hour. This resulting netcdf file was then used to derive annual averages and percentiles of PNC for the whole West Midlands. The PNC concentration mapping over the West Midlands were generated using GIS tools (here Surfer for gridding and ArcMap for visualisation).

Fig. 7 shows annual concentration maps for PNC_{10_100} and PNC_{100_325} at 10 m × 10 m resolution for the 2019 BAU case. PNC_{10_100} ranged from 2304 to 4478 # cm^{-3}, while PNC_{100_325} ranged from 574 to 1062 # cm^{-3}. These are lower than those found in the case study of Copenhagen, Denmark, i.e. ranging from 6000 to 12,000 # cm^{-3} for PNC_{30_250} at the residential address locations (Ketzel et al., 2021). There were clear patterns of the influence of traffic emissions on the distributions of PNC with higher levels near major roads in the city centre and motorways. High levels of PNCs inside city areas were also caused by the high density of buildings, which reduced the ventilation between the street canyon and overlying atmosphere. Lower levels of PNCs were observed in rural areas and regions far away from major roads. These findings are consistent with Ketzel et al. (2021).

To link PNC information to health-related studies of air pollution, high resolution PNC mapping can be spatially aggregated into population related polygon layers, such as at the electoral ward level and local authority level. This does result in smoothing of the highest and lowest concentrations, in particular in proximity to line sources. Figs. 8 and 9 present aggregated annual maps for PNC_{10_100} and PNC_{100_325} for the ward level and local authority level, respectively. Ward level averaged PNC_{10_100} ranged from 2339 to 2894 # cm^{-3}, while PNC_{100_325} ranged from 583 to 718 # cm^{-3}. Local authority averaged PNC_{10_100} ranged from 2471 to 2680 # cm^{-3}, while PNC_{100_325} ranged from 611 to 666 # cm^{-3}. This spatial averaging was dependent on different resolution, and coarser spatial resolution would have narrower ranges of PNCs, as both lowest and highest concentrations were not fully represented any more.

Apart from annual PNC mapping, the short-term analysis of concentration distribution (percentiles) to assess exceedances is also useful. For 24-hour (daily) mean concentrations, the 99 percentile is normally adopted, which represents 4th highest levels of the annual daily series. For 1-hour mean concentrations, the 99.8 percentile is normally used to represent 18th highest levels of the annual hourly series. Figs. 10 and 11 present the 99 (for daily series) and 99.8 percentiles (for hourly series) PNC maps for PNC_{10_100} and PNC_{100_325}), respectively. The exceedances were observed mostly in the areas close to motorways and major roads directing to motorways. For the 99 percentile, PNC_{10_100} ranged from 5490 to 9365 # cm^{-3}, while PNC_{100_325} ranged from 2043 to 2901 # cm^{-3}. For the 99.8 percentile, PNC_{10_100} ranged from 12,257 to 17,840 # cm^{-3}, while PNC_{100_325} ranged from 3307 to 4703 # cm^{-3}. It is not surprising that the 99 percentile has narrower ranges than the 99.8 percentile for both PNC_{10_100} and PNC_{100_325}, as the diurnal peaks over the day cannot be fully represented by the 24-hour mean.

4. Discussion

A "Receptor" run in the ADMS-Urban model is normally required to evaluate the model configuration before it can be applied to a contour run for mapping. There is a very limited number of PNC measurement sites in the UK, with only 1 site in the West Midlands located at the University of Birmingham (i.e. BAQS). The 2021 WHO guideline (WHO, 2021) highlighted the need to expand UFP monitoring, which can be used to support future exposure estimation, modelling and epidemiological studies for UFP. The Model Evaluation Toolkit is a powerful tool to evaluate the performance of the model using the available measured and model datasets. Overall, the model performed well with slight underestimation for both PNC_{10_100} and PNC_{100_325}. This underestimation may be caused by the uncertainty of the background dataset, which was taken from the rural Chilbolton air quality site relatively far away from the West Midlands. An additional source of underestimation may be due to natural and semi-natural sources (such as resuspension). PNC_{10_100} and PNC_{100_325} have the similar patterns in terms of model evaluation statistics, indicating that they are highly correlated, which is consistent with Ketzel et al. (2021) for similar PNC metrics. Also, PNC was used as a passive scalar in this modelling study, with no inclusion of aerosol microphysics which may contribute to particle formation, coagulation or size shrinkage. This treatment was also used in Ketzel et al. (2021) for the implementation of PNC into their Danish air quality modelling DEHM/UBM/AirGIS. In an analysis of data from the southern UK, Bousiotis et al. (2019) show that atmospheric new particle formation does not impact heavily upon longer-term particle concentrations in the UK. Particle shrinkage, demonstrated in London (Dall'Osto et al., 2011, Harrison et al., 2016) may be affecting particle size distributions, but does not appear to be impacting significantly upon particle number concentrations.

Based on the modelling output from a contour run (normally required HPC clusters to optimise run time), annual PNC maps at a variety of spatial resolutions (i.e. street scale of 10 m × 10 m resolution, ward level and local authority level) can be generated. There are currently no regulations for PNCs due to the lack of consistent epidemiological evidence, but these annual concentration maps for PNCs can be useful in future. The street scale resolution can resolve the hotspots near road sources (a key feature of the ADMS-Urban model). Building geometry in urban areas would significantly influence the dispersion of PNC at the street locations in the city centre areas, as also highlighted by Ketzel et al. (2021) for their PNC dispersion modelling for Copenhagen, Denmark. The overall PNC maps look realistic, showing the similar pattern of spatial distribution of other traffic-related pollutants such as

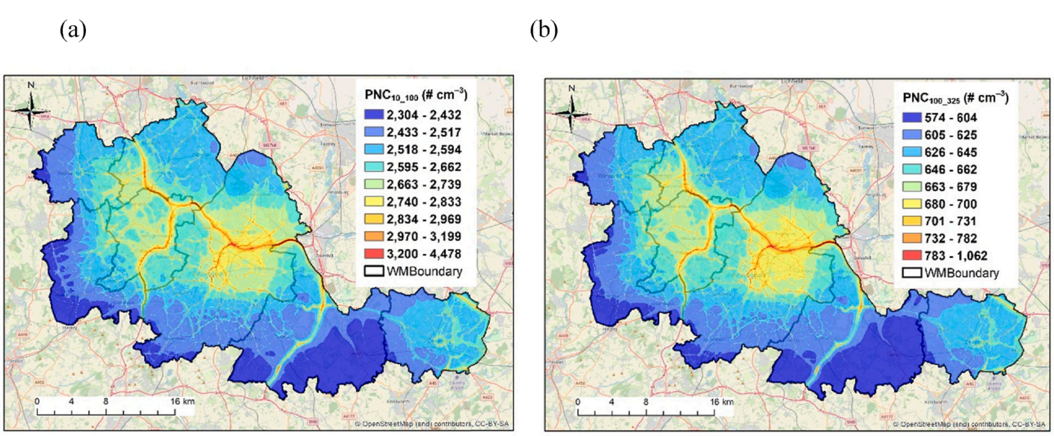

Fig. 7. Annual PNC maps for (a) PNC between 10 and 100 nm (PNC_{10_100}) and (b) PNC between 100 and 325 nm (PNC_{100_325}) at 10 m × 10 m resolution for the 2019 BAU case.

(a) (b)

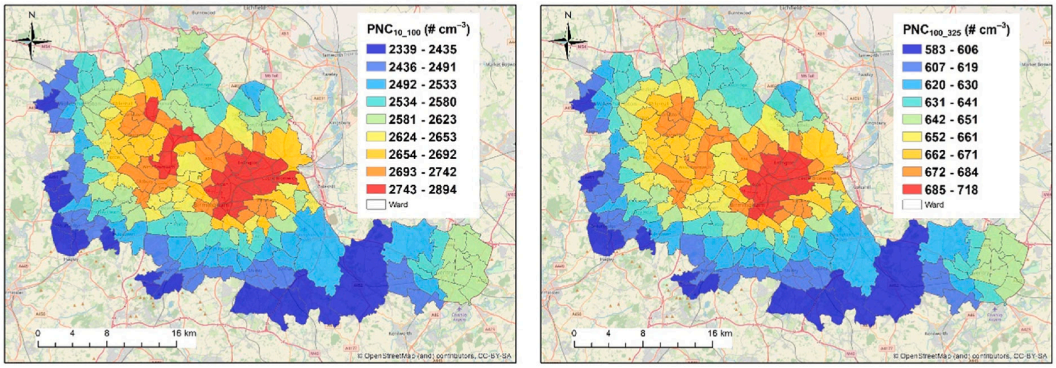

Fig. 8. Aggregated annual PNC maps for (a) PNC between 10 and 100 nm (PNC_{10_100}) and (b) PNC between 100 and 325 nm (PNC_{100_325}) at the electoral ward level for the 2019 BAU case.

(a) (b)

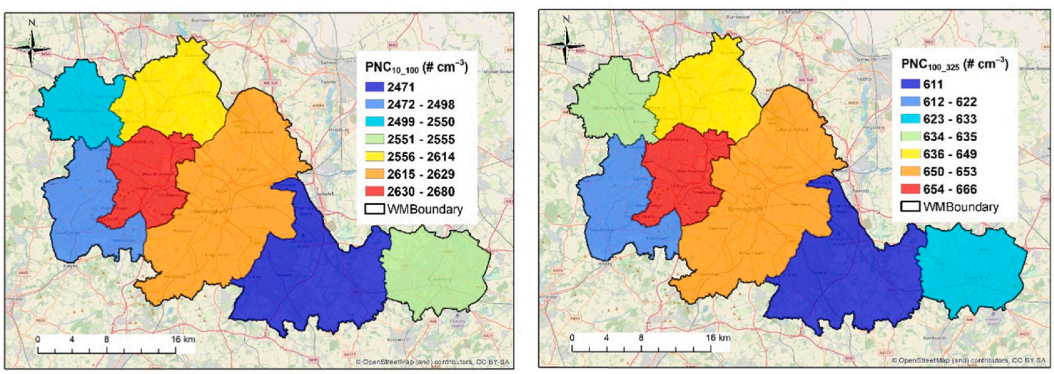

Fig. 9. Aggregated annual PNC maps for (a) PNC between 10 and 100 nm (PNC_{10_100}) and (b) PNC between 100 and 325 nm (PNC_{100_325}) at the local government authority level for the 2019 BAU case.

(a) (b)

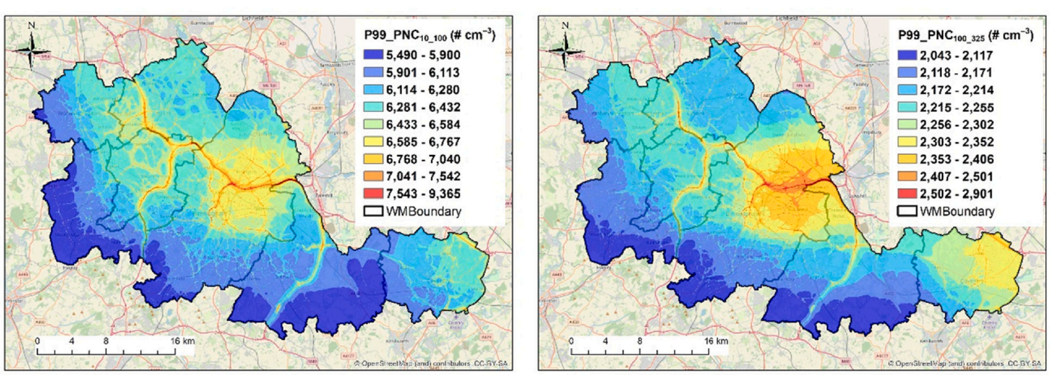

Fig. 10. The 99 percentile PNC maps for 1 day mean over the year for (a) PNC between 10 and 100 nm (PNC_{10_100}) and (b) PNC between 100 and 325 nm (PNC_{100_325}) at 10 m × 10 m resolution for the 2019 BAU case.

NO_2 as shown in Zhong et al. (2021). Coarser resolutions (at ward level and local authority level) tend to have a narrower range of PNC still capturing detailed gradients, as the local hotspots are averaged and therefore cannot be fully represented. However, this spatial aggregation can be useful to link local population and health data (normally available at the ward level) for potential exposure, health impact and epidemiological studies. The open source Air Quality Lifecourse Assessment Tool (AQ-LAT) (Hall et al., 2023) model developed by the WM-Air health strand is an electoral ward-level tool, although it is currently only applicable to the traditional air pollutants (i.e. NO_2 and $PM_{2.5}$). Local authority level PNC can be also useful to indicate the transboundary concentrations and local to regional combined efforts will be needed to reduce PNC.

Although there are no clear guidelines for PNC, the 2021 WHO

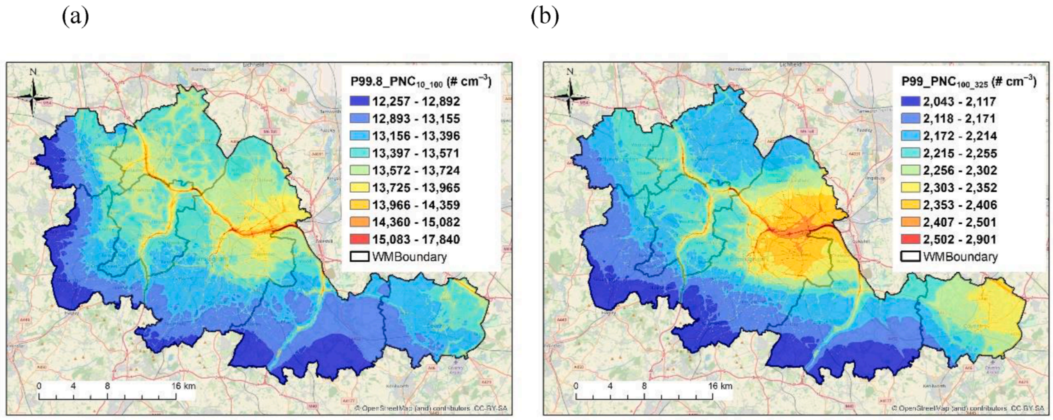

Fig. 11. The 99.8 percentile PNC maps for 1 h mean over the year for (a) PNC between 10 and 100 nm (PNC_{10_100}) and (b) PNC between 100 and 325 nm (PNC_{100_325}) at 10 m × 10 m resolution for the 2019 BAU case.

Guidelines incorporate guidance for PNC for UFP regarding short term concentrations based on expert opinion and available literature (WHO, 2021). It is suggested that a low PNC for UFP is considered below 1000 # cm^{-3} for a 24-hour mean (no statement for 1-hour mean), while high PNC is considered above 10,000 # cm^{-3} for the 24-hour mean or 20,000 # cm^{-3} for the 1-hour mean. The daily objective linking with the 99 percentile if proposed in future will mean the targeted value not to be exceeded more than 4 days in a whole calendar year, while the hourly objective linking with the 99.8 percentile would mean no exceedance beyond 18 times over an annual hourly time series. Here, we used PNC_{10_100} to compare with the suggested values of PNC for UFP, although the sub-10 nm nanoparticles were not represented by the model, which therefore underestimates the total particle number concentration. Our results for the 99 and 99.8 percentiles for PNC_{10_100} indicate that levels in the West Midlands lie between the suggested low and high PNC defined concentrations, although approaching the high PNC. Future policies targeting traffic emission reductions, e.g. Clean air strategy and Net Zero policies, would be expected to reduce PNC, towards low PNC as suggested by WHO. Recent work by Damayanti et al. (2023) shows the limited effectiveness of diesel particle filters in removal of UFP, and hence transition to a battery-electric vehicle fleet will be needed to achieve a substantial reduction in concentrations.

5. Conclusion

The ADMS-Urban model configuration for PNC over the West Midlands, UK has been set up to simulate the dispersion of PNC. Model evaluation has been conducted using PNC measurement data from BAQS. Overall, the model generally performed well although there was a slight underestimation for PNC. The ADMS-Urban model was then run on a contour mode to generate PNC maps at a variety of spatial scales (i.e. street scale, ward level and local authority level) and temporal resolutions (i.e. annual, 24-hour, and 1-hour). PNC mapping could be linked to local population and health data for potential epidemiological studies.

This modelling work is the first PNC dispersion case study for the West Midlands, UK and is limited to a baseline year of 2019. PNC was treated as a passive scalar, with no inclusion of aerosol microphysics and restricted to sizes above 10 nm. Future work can be 1) to extend model evaluation for other types of sites (e.g. roadside, airport and urban background sites) if future measurement data (including low-cost-sensor data) is available, 2) to conduct modelling scenarios to account for the effects of future interventions, 3) to link with epidemiological tool for health effects of PNC, 4) to provide scientific evidence to support potential future WHO guidelines for PNC, 5) to develop a reanalysis approach to account for aerosol microphysics, 6) to include aerosol microphysics towards an implementation of size-bin resolved PNC, and 7) to extend the current PNC modelling capability to other metropolitan areas (e.g. Greater Manchester, and Greater London).

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CRediT authorship contribution statement

Jian Zhong: Methodology, Software, Visualization, Formal analysis, Writing – original draft. **Roy M. Harrison:** Conceptualization, Supervision, Writing – review & editing, Funding acquisition. **William James Bloss:** Conceptualization, Supervision, Writing – review & editing, Funding acquisition. **Antoon Visschedijk:** Methodology, Writing – review & editing. **Hugo Denier van der Gon:** Methodology, Writing – review & editing.

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.

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Appendix A. Supplementary material

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envint.2023.108273.

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