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Limited impact of diesel particle filters on road traffic emissions of ultrafine particles

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A R T I C L E I N F O

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A B S T R A C T

Diesel engines are a major contributor to emissions of both Black Carbon (BC) and ultrafine particles. Analysis of data from the only roadside monitoring site in Europe with a continuous dataset for size-segregated particle number count (Marylebone Road, London) from 2010 to 2021 reveals that the growing number of vehicles fitted with a Diesel Oxidation Catalyst (DOC) and Diesel Particle Filter (DPF) has been very effective in controlling the emissions of solid particles and hence BC, but that there has been little change in the liquid mode (<30 nm) particles, and that concentrations of ultrafine particles (<100 nm) still well exceed the threshold for "high" concentrations (>10^4 cm^{-3} /24-hour mean) defined by WHO. BC declined by 81% between 2014 and 2021, but the ultrafine particle (<100 nm) count declined by only 26%. Consequently, in locations worldwide with heavy diesel traffic, concentrations of ultrafine particles are likely to remain "high" for the foreseeable future unless more effective abatement technologies are implemented.

1. Introduction

Ultrafine particles (UFPs) are defined as those with a diameter<100 nm. There are concerns that they may have enhanced toxicity per unit mass and the World Health Organization has recommended enhanced surveillance and precautionary limits on concentration (WHO, 2021). UFPs make a negligible contribution to the PM mass in the ambient air, but they contribute dominantly to the particle number count (PNC) (Harrison et al., 2000; HEI, 2013). Thus, the concentration of UFPs is mainly expressed and quantified by number concentration (particles/cm^3) (Austin et al., 2019; Cassee et al., 2019).

Although there is no strict definition of classification based on size range, particle size distribution are generally classified into three groups which are the Nucleation mode (diameter particle or Dp < 30 nm), Aitken mode (Dp 30–100 nm), and Accumulation mode (Dp > 100 nm), which may reflect their formation process and origin (AQEG, 2018). Many studies have revealed that the PNC in urban traffic-related sites is relatively higher than in other urban environments (Giemsa et al., 2021; Oliveira et al., 2009) because of the emissions from the high volumes of road vehicles. Wahlin et al. (2001) found that petrol and diesel vehicles mainly emit particles with a diameter less than 300 nm. Another study in a street canyon found an association between accumulation mode particles and emissions from heavy duty traffic, while a stronger association was found between smaller size range particles (30–60 nm) with light duty traffic (Charron and Harrison, 2003).

Globally, policies to improve urban air quality have been implemented in recent decades. For the UK's road transport sector, these involve both requirements for strict emission standards (i.e., Euro standards), and local traffic regulations (e.g., Congestion Charge, Low emission Zone (LEZ), Ultra Low emission Zone (ULEZ)). A policy's effectiveness in improving air quality can be assessed through the trends of pollutant concentration (Gualtieri et al., 2014; Font and Fuller, 2016). Hence, long-term data sets from the air quality measurement site play a crucial role in evaluating and quantifying the impact of those regulation implementations. Studies in the UK to date (Font and Fuller, 2016; Holman et al., 2015; Ma et al., 2021; Harrison and Beddows, 2017) have generally focused on some of the regulated pollutants (e.g., NO_2, $PM_{2.5}$, PM_{10}), and there has been limited study focusing on the UFP, either number concentration (PNC) or number size distribution (PNSD).

In Europe, diesel vehicles are responsible for most of the exhaust particulate matter from road vehicles. Particles emitted in diesel exhaust fall into two main types: solid graphitic (Black Carbon) particles, often

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comprising a loose aggregate of primary spherules of around 40 nm diameter, and a second mode of generally smaller particles, comprising mainly liquid, derived substantially from condensed engine oil (hydrocarbon) vapour (Kittelson et al., 2006; Harrison et al., 2018). The latter particles form in the roadside atmosphere as the hot exhaust gases cool as they mix with cooler ambient air. Some of the first observations of the formation of the semi-volatile particles were made on the heavily trafficked Marylebone Road in London (Charron and Harrison, 2003). Formation of such particles depends upon the presence of condensation nuclei, typically sulphuric acid or tiny ash particles formed from metallic impurities in the fuel, upon which the semi-volatile hydrocarbons condense. A reduction in the sulphur content of motor fuel in 2007 led to a consequential major reduction in particle number concentrations at Marylebone Road, presumably because of a reduction in sulphuric acid nuclei, causing more vapour to condense on the larger, graphitic particles (Jones et al., 2012).

Our study has analysed 12-years (2010–2021) of particulate matter data derived from Scanning Mobility Particle Sizer (SMPS) and Condensation Particle Counter (CPC) instruments, and other related pollutant data derived from the Automatic Urban and Rural Network (AURN) measured at an urban traffic site in London. This is the only roadside measurement site in Europe with a continuous dataset over this period. Moreover, PNC and PNSD differentiated by wind direction were also further analysed. Some policy interventions implemented during the study period were identified to evaluate any associations between them and the particle number and size distribution.

2. Materials and methods

***Site Description*:** The London, Marylebone Road (LMR) site has been extensively studied (Harrison and Beddows, 2017; Hicks et al., 2021; Kamara and Harrison, 2021) for air quality research in the UK. Located at 51°31′21.1″N 0°09′16.6″W, the air monitoring equipment is in a large cabin on a sidewalk on the southern side of Marylebone Road. Marylebone Road is a six-lane highway which carries approximately 80,000 vehicles/day, and the nearest carriageway is only ∼ 1 m from the monitoring station. The road is about 32 m wide, and the buildings surrounding the site have a maximum height of around 25 m, forming a street canyon of typical height to width ratio.

***Data Collection*:** Multiple year (2010–2021) datasets of hourly concentration of PNC, PNSD and Black Carbon (BC) were downloaded from the UK-Air data repository. Total number concentrations were derived from a CPC (TSI 3022A to June 2017, then 3772-CEN, both measuring from a lower D_{50} cut point of 7 nm) (Tompkins et al., 2021). Particle size distributions were measured using a SMPS. This consists of a CPC (TSI model 3775) combined with an electrostatic classifier (TSI model 3080). The electrostatic classifier consists of a charge neutraliser (incorporating a Kr-85 radioactive source) and a Differential Mobility Analyser (DMA – TSI model 3081) (Tompkins et al., 2021). The range of particle size distribution data derived from the SMPS is 16.55–604.3 nm with 51 size channels. The sum of particles measured by the SMPS (TNCS) is typically substantially less than the total particles counted by the CPC (TNC), mainly due to the different lower measurement cut points. Black carbon is quantified based on light transmission through a filter-based sample using an Aethalometer instrument (model AE22 to November 2019, then AE33) that is operated at several wavelengths, from which the BC concentration is measured at 880 nm wavelength using the default mass absorption coefficient built into the instrument (Ciupek et al., 2021). Regulated-pollutants (CO, NO_x, NO_2, NO, $PM_{2.5}$, PM_{10}) concentrations and surface meteorological data were downloaded from the Openair package, which was already integrated with data from Automatic Urban and Rural Network (AURN) and London Heathrow (LHR) Airport. Although the distance between the measurement site and meteorological station at LHR is approximately 22.5 km, the study by Manning et al. (2000) shows a fair consistency of wind direction up to 40 km from the measurement station. In addition, data from LHR is representative of larger scale air flows and is not influenced by the nearby buildings in London. A wind rose from the entire period (2010–2021) shows a prevailing wind from the south-west direction, as seen in Fig. S1. The pollutant data capture is mostly higher than 90%, apart from TNC and SMPS data (Table S1) which has under 50% data capture in several years due to instrument maintenance.

***Data Analysis*:** Data analysis mainly used the Openair package in R software (Carslaw and Ropkins, 2012) and MS Excel. The Theil-Sen method (Theil, 1950; Sen, 1968) available in the R-Openair package was used to estimate the trend of pollutant concentrations which used the median slope among all lines through the paired sample points. It is very suitable for air quality data analysis because it tends to yield confidence intervals accurately, even for a non-normal dataset, and is also resistant to outliers (Carslaw and Ropkins, 2012). For the slope estimation, the de-seasonalised mode in which any seasonal fluctuation and cyclical variation was removed, was selected to yield a more unambiguous indication of the pollutant trend. This is done using Seasonal Decomposition of Time Series by Loess (STL) i.e a filtering procedure to decompose a time series into seasonal, trend and remainder. The Loess smoother is applied sequentially in STL, allowing for fast computation even for very long time series and significant quantities of trend and seasonal smoothing. This allows analysis of the procedure's properties. It can also decompose time series with missing values (Cleveland et al., 1990).

3. Results

***Long-term trend of Particle Number and Pollutant Concentrations*:** Data from the roadside Marylebone Road supersite in central London were analysed for trends in particle metrics and other traffic-related pollutants. Table 1 shows the overall Theil-Sen slope of all pollutants

Table 1
Overall trends for the period 2010–2021, and 2014–2021 at London Marylebone Road for de-seasonalised data. Trends are calculated for the various metrics using the Theil-Sen method. Negative values indicate a reduction with time. The brackets show 95% Confidence Intervals, while the statistical significance marked by "***", "**", "*", "+" represents $p < 0.001$, $p < 0.01$, $p < 0.05$, and $p < 0.1$, respectively.

Parameter	Trends 2010–2021		Trends 2014–2021	
	# cm^{-3} $year^{-1}$	% $year^{-1}$	# cm^{-3} $year^{-1}$	% $year^{-1}$
Nucleation	−242 [−290, −191] ***	−4.86 [−5.52, −4.14] ***	28.6 [−44.5, 99.8]	0.98 [−1.41, 3.69]
Aitken	−643 [−700, −597] ***	−6.18 [−6.53, −5.84] ***	−541 [−581, −494] ***	−7.18 [−7.54, −6.9] ***
Accumulation	−328 [−352, −309] ***	−7.28 [−7.64, −6.99] ***	−305 [−323, −283] ***	−10.18 [−10.14, −9.75] ***
TNCS	−1211 [−1333, −1101] ***	−6.11 [−6.5, −5.72] ***	−853 [−951, −749] ***	−6.26 [−6.72, −5.63] ***
TNC	−1815 [−2351, −1364] ***	−5.2 [−6.39, −4.15] ***	−74.5 [−742, 804]	−0.38 [−3.32, 4.85]
	μg m^{-3} $year^{-1}$	% $year^{-1}$	μg m^{-3} $year^{-1}$	% $year^{-1}$
BC	−0.83 [−0.9, −0.77] ***	−8.26 [−8.56, −7.98] ***	−0.76 [−0.82, −0.7] ***	−12.1 [−12.5, −11.8] ***
CO	−0.04 [−0.04, −0.03] ***	−5.2 [−5.51, −4.69] ***	−0.04 [−0.04, −0.03] ***	−7.15 [−8.22, −5.54] ***
NOx	−18.3 [−22.4, −14.0] ***	−4.73 [−5.29, −3.90] ***	−36.5 [−39.8, −33.4] ***	−9.31 [−9.61, −8.88] ***

in concentration units and per cent per year. It shows many statistically significant downward trends over the given period. The reduction in CO and NO_x is in line with a decline in national emissions from road transport for almost all pollutants in the UK from 2010 to 2019, based on National Atmospheric Emissions Inventory data (NAEI, 2022).

TNC can be derived from both CPC (TNC) data directly and SMPS (TNCS) data by summing the size bins, as well as for each size-range mode, and all show a downward trend.

Over 2010–2021, the Nucleation mode decreased more slowly (4.9%/year, with confidence interval (CI): 5.52, 4.14%/year) than other modes, while the accumulation mode showed the fastest rate of decline of 7.3%/year (CI: 7.64, 6.99%/year). The Aitken mode decreased at a similar rate to the TNCS. The inventory of total particle number emissions reported by the UK's Air Quality Expert Group (AQEG, 2018) also showed that the transportation sector was the dominant contributor to the TNC emissions in 2005 (as a base year). BC has the most significant decline among other pollutants over the study period, and decreased by 8.3%/year (CI: 8.56, 7.98%/year). BC is primarily released by combustion engines (particularly diesel), wood and coal burning from the residential sector, heavy oil or coal power stations, field burning of agricultural wastes, and forest and vegetation fires (WHO, 2012). In UK urban areas, diesel road traffic is the main source, as revealed by large roadside increments (Harrison and Beddows, 2017). The data for 2014–2021, corresponding to the growing penetration of Euro 6/VI into the vehicle fleet, show an almost level trend in Nucleation particles (+1% per year) and TNC (-0.4% per year). Over this period, Accumulation mode particles fell by 10.2% per year and BC by 12.1% per year.

The trends of TNC found in this study are comparable with another recent study by Mikkonen et al (2020) that analysed particle number concentration (Dp 6–1000 nm) over 2008–2018 in Budapest. They found a decreasing decennial statistical trend (4–5%) that was mainly associated with the decrease in road traffic emissions.

Due to a vortex in the Marylebone Road street canyon, the emissions from traffic on Marylebone Road are carried directly to the sampler by winds in a southerly sector, while air on northerly sectors is predominantly the background from north London (Harrison et al., 2019), and hence trend analysis differentiated by wind direction was conducted. The summary of the trends of all pollutants is presented in Fig. S2. It shows that the most significant reduction of pollutants mainly occurred when the air came from the southerly sectors, mainly south and south-westerly directions. The very significant reduction observed for BC and Accumulation mode particles over the period 2010–2021 was 8.7% (CI:-8.94,-8.34%) and 8% (CI: −8.45,-7.65%) per year, respectively. Smaller reductions of pollutant concentration mainly occurred when the air masses came from the north-easterly wind sector, reflecting a mix of particle sources in the north London area, and a regional background of largely secondary particles.

***Time Variation of Pollutants and Particle Size Modes Over 2010–2021*:** Fig. S3 shows the diurnal variation of the traffic-generated pollutants BC and NO_x for each year, while the yearly change is presented in Table S2. Overall, it is observed that concentrations reach a peak during the morning and afternoon rush hour and have a minimum at around 3 am coinciding with the minimum traffic volume, particularly on weekdays (Helfter et al., 2011). Pollutant concentrations, however, remain high during the middle of the day as the traffic volume passing on this road remains high.

The traffic-related pollutants have decreased substantially from 2016 onwards (Fig. S3), which is strongly associated with the rapid growth of the Euro 6/VI vehicle proportion as presented in Fig. S4. Pollutant concentrations were observed sometimes at a lower level in 2020 and 2021, attributable to movement restrictions during Covid-19 which at times led to a dramatic reduction in vehicle numbers in London. However, overall vehicle numbers on Marylebone Road varied little between the years. (Fig. S5). Black Carbon concentrations gradually decreased after 2012 and reduced substantially from 2016 onwards, likely associated with the growing numbers of Euro 6 and VI-compliant vehicles fitted with DPFs, which are reported to remove around 99.9% of all solid, carbonaceous emissions of UFP (Calderón-Garcidueñas and Ayala, 2022). A significant reduction in BC concentration following LEZ implementation and other clean air program policies was also observed at all measurement sites in Leipzig, Germany (Baldauf et al., 2016).

Diurnal variations in particle counts (Fig. S3) over 2010–2021 show a significant reduction of nucleation mode particles from 2013 to 2014, followed by a similar level from 2014 onward and slightly reduced in 2021. Unlike other pollutants, the concentration level of this mode in 2020 was observed not to be significantly different compared to that in 2014–2016. This trend was reflected in the annual average, particularly in the 225° wind sector with highest concentrations.

A gradual decrease of Aitken mode was observed from 2010 to 2020, which was very clearly observed during 2016–2020. The Accumulation mode, the solid particles mainly resulting from the combustion of diesel fuel as well as from the growth of Aitken mode (Vu et al., 2015), remains high starting from the morning rush hour and decreases after around 8 pm.

The annual average particle number concentration of each mode and regulated pollutants according wind direction were analysed (Fig. 1, and Fig. S6). As indicated by Fig. S3, a sharp decline of Nucleation mode particles was observed from 2013 to 2014, which then stabilises and changes little from 2014 to 2021, seen most clearly in the sectors from 135 to 270°in which the traffic emissions on Marylebone Road are sampled most effectively. An obvious continual decline was observed for the Accumulation mode, particularly in the 135 to 270° wind sectors which are most reflective of the emissions on Marylebone Road. The Aitken mode, also shows a strong decline, but not as substantial as the accumulation mode, presumably because it contains some liquid mode particles which are poorly removed by the DPF. The Nucleation mode proportion of the particles rose from 23% in 2015 to 39% in 2021, and the Accumulation mode shows the opposite trend, decreasing from 22% in 2015 to 12% in 2021. The trend in the proportion of nucleation mode particles out of the total number of particles is shown in Fig. S7.

***Continuous Number Size Distribution*:** Annual particle number size distributions are shown in Fig. 2, showing changes over the period 2010–2021. The distribution in 2010 is clearly bimodal, and as time progresses, the contribution of a mode at ca.70 nm gradually declines, until it is not visible in the data for 2021. For further investigation, an analysis of PNSD split by wind direction was conducted (Fig. S8). This shows that the change in size distribution affected all wind sectors indicating that the changes seen in the emissions on Marylebone Road also occurred in the air of north London, which dominates the air sampled on winds in the northerly sector.

***Emissions Regulation Implementation Over 2010–2021*:** In order to mitigate against the health impacts of fine particle emissions from road traffic, diesel particle filters (DPF) were made mandatory on new light duty diesel vehicles from Euro 5 which was enforced across Europe from January 2011, and continued with the subsequent Euro 6 from September 2015. Particle filters were also required for Gasoline Direct Injection engines by imposition of a similar standard in Euro 6, and for heavy duty vehicles by Euro VI from 2013. The Directive requiring the use of particle filters was framed in terms of a limit on the number of particles emitted per kilometre, and in order to make the test repeatable was limited to particles involatile at 300–400 °C and smaller than 23 nm. Due to the natural turnover of the vehicle fleet and the introduction of the London Ultra-Low Emissions Zone requiring diesels to conform to Euro 6 standards from April 2019, a progressively larger proportion of vehicles fitted with DPFs has been using Marylebone Road.

As it takes time for new vehicles which comply with the regulations to penetrate the market, the data on fleet composition, based on vehicle type and Euro standard, was downloaded from the National Atmospheric Emissions Inventory (NAEI, 2022). Data, available from 2013 onward, was filtered so as to use only data from within the Inner London area, as seen in Fig. S4. It shows a steady increase in the aggregate of Euro 5 and Euro 6 in all light duty categories from 2013 to > 80% in 3

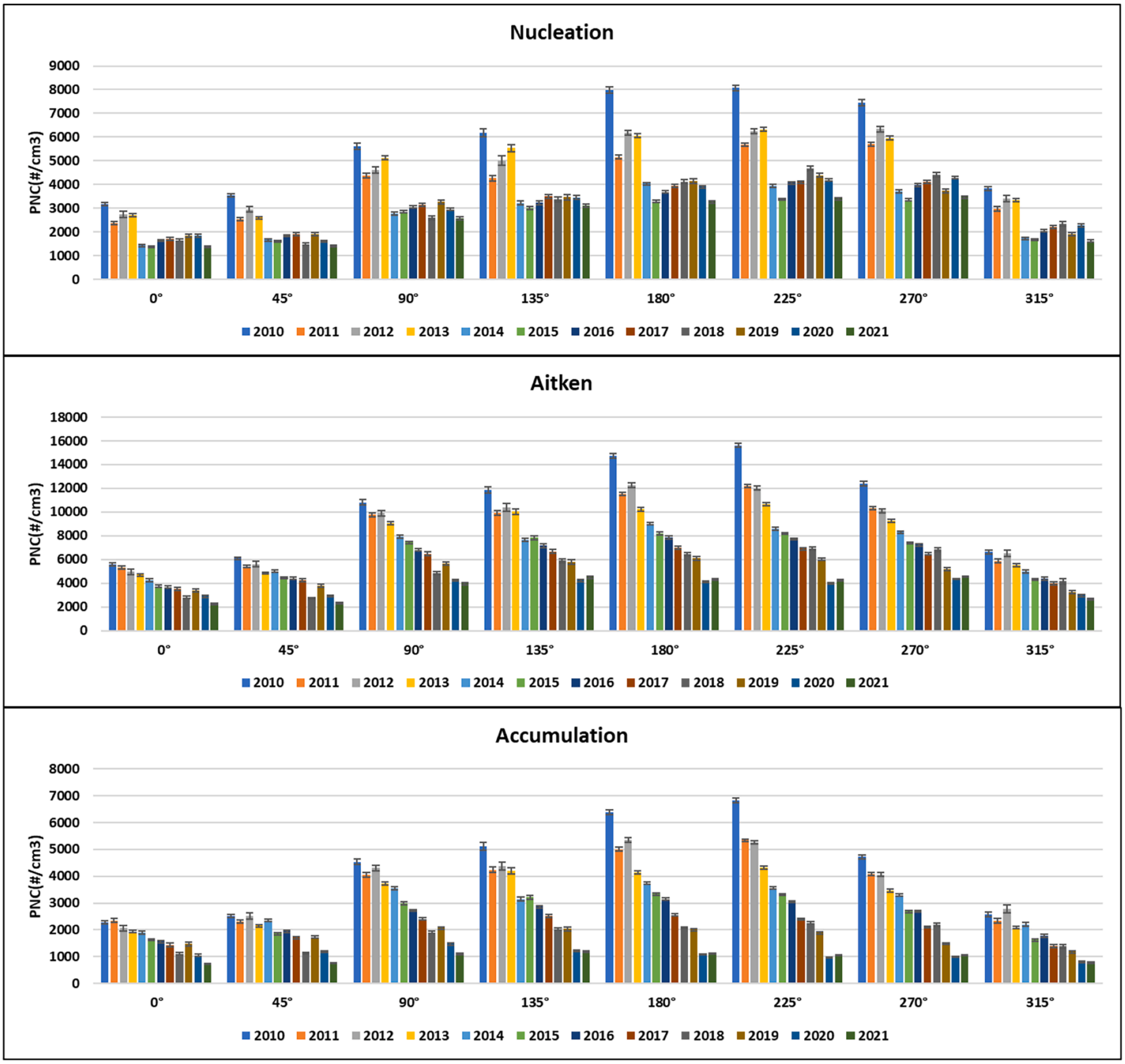

Fig. 1. Annual mean concentration of each particle size mode split by year (2010–2021) and wind direction. Data are filtered according to hourly average wind direction and averaged over one-year intervals. The error bars correspond to one standard error. The Nucleation mode data show a sharp drop from 2013 to 2014, after which there is no clear upward or downward trend. The Aitken and Accumulation mode data show a steady decline. The 180° and 225° data show the greatest contribution of Marylebone Road traffic.

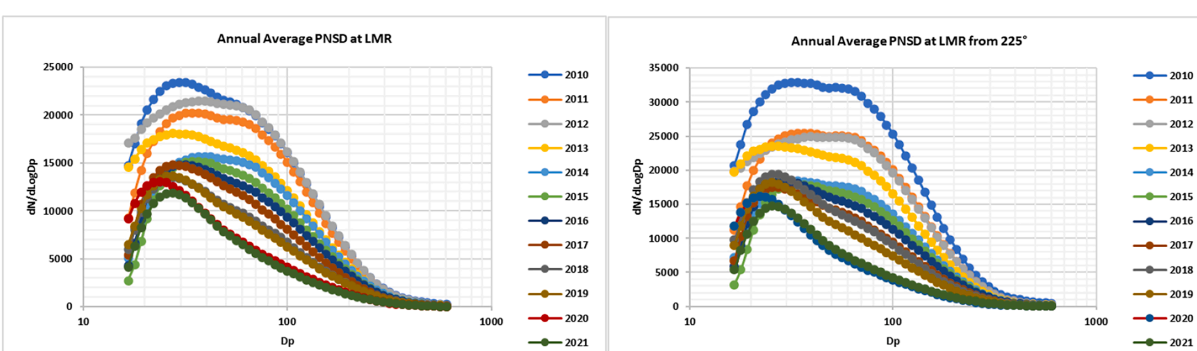

Fig. 2. Annual average particle number size distributions (PNSD) at London, Marylebone Road, 2010–2021. These show all data (left panel), and data only for the 225° wind direction (right panel) which is most affected by Marylebone Road traffic. A mode visible at around 70 nm in the earlier years is no longer visible at the end of the time series.

out of 4 vehicle categories in 2021. A more prominent increase of Euro VI HGVs can be seen from 2015 onwards, from 2% in 2014 and up to 87% in 2021. Buses and coaches were the slowest heavy duty category to adopt Euro VI but still reached > 70% by 2021.

A progressive reduction of vehicles non-compliant with DPFs was observed in all vehicle types during 2013–2021, within the range of 11–26% per year; larger decreases were seen in heavy-duty vehicles (Rigid and Artic, and TfL buses) (see Table S3). These rates of change closely follow the trends of BC and accumulation mode particle reductions during 2014–2021 as seen in Table 1.

The London Low Emission Zone (LEZ) was implemented in four phases from 2008 to 2012, covering initially only parts of central London (Fig. S9), and operates 24 h a day. This policy restricts vehicles within the zone based on PM emission standards. The London Ultra-Low Emissions Zone (ULEZ), was introduced in April 2019 and operates 24 h every day except Christmas Day. This requires Euro 6 or Euro VI for light and heavy duty vehicles and has accelerated the uptake of these technologies, and although initially Marylebone Road was on the northern border of the zone and not subject to its regulations, inclusion occurred when the ULEZ was extended in 2021.

The decline in Black Carbon correlated with the increased penetration of vehicles meeting the Euro 5, 6 and VI standards, as exemplified for heavy duty trucks in Fig. 3. The slower decline in Accumulation mode particles than Black carbon is attributable to the large background in secondary Accumulation mode particles which is only indirectly affected by vehicle emissions regulations.

The Nucleation mode particles comprise mainly semi-volatile compounds and partition between the vapour and particulate phases. At the high temperature of the exhaust gases, the vapour phase will predominate. The DPF is highly effective at removing particles of all sizes as indicated in the finding of present study that shows significant reduction of particle number, but is not designed to remove vapour. This should be removed by the DOC, which has been a standard fitment on diesels since Euro 3. However, a laboratory study on a Euro 5 light duty diesel engine (Alam et al., 2019) found a reduction of organic carbon in the sum of both phases of only 56% by a diesel oxidation catalyst (DOC) and diesel particulate filter (DPF) relative to engine-out emissions, hence explaining the small effect of emission controls upon the Nucleation particle fraction. A further consideration is that the formation of Nucleation mode particles is competitive with vapour condensation onto larger particles (Harrison et al., 2018), and if the condensation sink provided by those larger particles declines, as reflected in the reduction of Aitken and accumulation mode particles, the formation of Nucleation mode particles is favoured. Calderón-Garcidueñas and Ayala (2022) report the formation of Nucleation mode particles in the exhaust of catalysed, but not uncatalyzed DPFs.

Two further factors need to be considered. The number of vehicles burning neither petrol or diesel, i.e. hydrogen fuelled, electric vehicles (EVs) or hybrid vehicles in electric mode, and hence unlikely to emit nanoparticles, except in small quantities from the tyres and brakes, has been increasing as shown in Fig. S4. However, by 2021 only 0.5% of passenger cars in inner London were electric vehicles. Taxis comprise 6.4% of the vehicles in central London, and by 2021, 50% were either battery-electric, hybrid or hydrogen fuel cell. Hence, even allowing for the greater distance travelled by taxis, it seems likely that only a few percent of vehicles on Marylebone Road were neither petrol nor diesel, and hence would have little influence on the air quality trends observed. There has been a shift of petrol vehicles towards gasoline direct injection (GDI), which emit sub-23 nm solid particles (Pfau et al., 2022). However, the particle number emission limit set by Euro 6 requires the fitting of particulate filters which are highly efficient at removing solid particles of all sizes, and hence no effect upon the sub-23 nm particles in the atmosphere is anticipated.

4. Discussion

The long-term (2010–2021) trends of both particle number and size distribution and of some regulated pollutant concentrations have been analysed from an urban traffic site in the UK. The Aitken mode (Dp 30–100 nm), Accumulation mode particles (Dp > 100 nm), and Black Carbon decreased faster than other pollutants, and reduced by 6.2%/year (CI: 6.5–5.8%/year), 7.3%/year (CI: 7.64, 6.99%/year) and 8.3%/year (CI: 8.56, 7.98%/year), respectively. The most significant reduction occurred when the wind was blowing from the southerly sector, which at this site carries fresh emissions from traffic to the sampling station, confirming that the policy intervention in the road transport sector has effectively decreased those pollutant concentrations. The change in particle emissions is clearly seen in the changing size distribution. The Nucleation mode proportion of the entire particle population gradually rose from 23% in 2015 to 39% in 2021, and the Accumulation mode shows the opposite, decreasing from 22% in 2015 to 12% in 2021. Analysis of PNSD split by wind direction shows two different

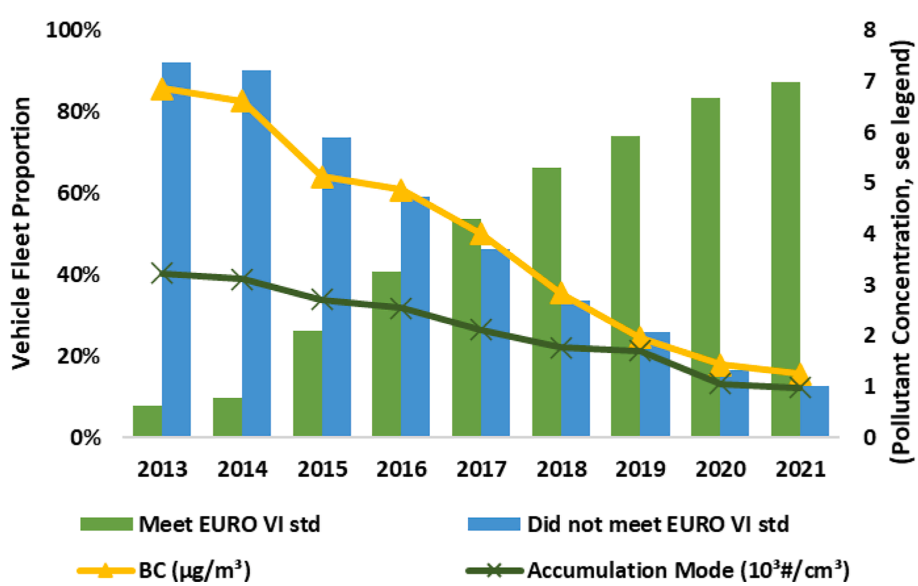

Fig. 3. Temporal trends in Black Carbon and Accumulation mode particle concentrations in relation to heavy duty truck traffic which did not meet the Euro VI emissions standard, 2013–2021. The continuous lines join annual averages for the particle metrics, and the bars show those heavy duty trucks which either did or did not meet the Euro VI emissions standard. A strong relationship is seen in the trends for Black Carbon and Euro VI non-compliant vehicles.

populations of data, in which the southerly winds (particularly 180°, 225°, and 270°) contribute higher concentrations than that from the northerly sector. Another study by Rönkkö et al (2017) that focused on nanocluster aerosol (Dp 1.3–3 nm) has also found a high concentration from wind sectors that were associated with emission from the road.

At LMR, a clear change was observed in the particle size distribution, most prominent in the 225° wind sector which is most influenced by the Marylebone Road traffic; a bimodal pattern is seen in the early years, in which a modal (~70 nm) peak gradually disappeared by the end of the period, indicating a significant change in the sources of these particle size ranges.

DPFs were introduced as a measure to reduce air pollution by diesel exhaust particles. As the mass of particles became difficult to measure in laboratory regulatory tests, the Euro 5, 6 and VI emission standards were defined in terms of a particle number concentration. As the formation of the nucleation mode particles occurs in the diluting exhaust gases, it is inherently variable, depending upon the dilution ratio and environmental conditions, and consequently the emission standards were set in terms of the number concentrations of involatile (solid) particles > 23 nm in the exhaust gases. The introduction of DPFs has clearly had a major beneficial impact upon concentrations of Black Carbon and Accumulation mode particle emissions, but there remains an issue of poor control of the Nucleation mode particles. As a consequence, the total particle number count declined by only 26% between 2014 and 2021, which compares poorly with the 81% decline in Black Carbon. The relative toxicity of different particle components and sizes is not well understood, but there remains enhanced concern over ultrafine particles due to their high surface area to volume ratio and ability for translocation within the human body (WHO, 2021).

In the Good Practice Statement issued by WHO (2021) as part of its updated Air Quality Guidelines a concentration of ultrafine particles exceeding 10,000 cm^{-3} is defined as “high”. It is noted that compliance with the WHO guideline is not only exceeded at Marylebone Road but is also observed at a suburban background site in London at Honor Oak Park (Ciupek et al., 2022). Current concentrations at Marylebone Road exceed the threshold for “high” values by around a factor of two, and at the suburban Honor Oak Park site in south London also exceeded that level in one of the past three years. Compliance with the recommendation is likely to require a much-increased uptake of battery-electric vehicles with a much reduced emission of ultrafine particles. High concentrations of ultrafine particles are a worldwide phenomenon (Kumar et al., 2014), and hence this is likely to be a widespread and persistent phenomenon.

Author contributions

The study was conceived by RH and FP. DB provided initial scrutiny of the data and SD carried out the data analysis and wrote the first draft of the paper. RH refined the paper, with inputs from FP and DC.

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Data availability

Data supporting this publication are openly available from the UBIRA eData repository at https://doi.org/10.25500/edata.bham.00000862.

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.2023.107888.

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