



Said Munir ^{1,*}, Haibo Chen ² and Richard Crowther ³

- ¹ National Center for Meteorology, Jeddah 21431, Saudi Arabia
- ² Institute for Transport Studies, Faculty of Environment, University of Leeds, Leeds LS2 9JT, UK; h.chen@leeds.ac.uk
- ³ Environmental Advisory, Transport Strategy, Leeds City Council, Leeds LS1 1UR, UK
- * Correspondence: s.munir@ncm.gov.sa

Abstract: Atmospheric nanoparticles, due to their tiny size up to 100 nanometres in diameter, have negligible mass and are better characterised by their particle number concentration. Atmospheric nanoparticle numbers are not regulated due to insufficient data availability, which emphasises the importance of this research. In this paper, nanoparticle number emissions are estimated using nanoparticle number emission factors (NPNEF) and road traffic characteristics. Traffic flow and fleet composition were estimated using the Leeds Transport Model, which showed that the road traffic in Leeds consisted of 41% petrol cars, 43% diesel cars, 9% LGV, 2% HGV, and 4.5% buses and coaches. Two approaches were used for emission estimation: (a) a detailed model, which required detailed information on traffic flow and fleet composition and NPNEFs of various vehicle types; and (b) a simple model, which used total traffic flow and a single NPNEF of mixed fleet. The estimations of both models demonstrated a strong correlation with each other using the values of R, RMSE, FAC2, and MB, which were 1, 2.77×10^{17} , 0.95, and -1.92×10^{17} , respectively. Eastern and southern parts of the city experienced higher levels of emissions. Future work will include fine-tuning the road traffic emission inventory and quantifying other emission sources.

Keywords: nanoparticles; ultrafine particles; traffic emissions; nanoparticle number emission factors; emission inventory; emission modelling

1. Introduction

Atmospheric nanoparticles (NPs), also known as ultrafine particles (UFPs), are tiny particles up to 100 nm in diameter (<100 nm). Due to smaller size, their mass is negligible, and therefore, NPs are characterised by their number concentrations or number counts, measured in units of particles per cubic centimetre (p/cm³). Nanoparticle number emissions are expressed as the nanoparticle number emitted per km (p/km) or per second (p/sec) or per unit fuel consumed (p/litre). It is important to mention that atmospheric NPs are more dangerous to human health than the fine and course particles due to several reasons [1–3]: (a) NPs are suspended in the atmosphere for longer time and can travel larger distance. (b) NPs are smaller in size and, therefore, can enter the circulatory and lymphatic system and can pass through the blood–brain barriers. (c) NPs act as a precursor to coarser particles through their aggregation during the atmospheric ageing process, and (d) NPs have higher surface area, which is more related to the health effects than the particle mass. The human body's inflammatory response to particles has a strong correlation with particle surface area, rather than with particle mass [4,5]. The large surface area of NPs is linked with the high concentrations of reactive chemicals at deposition sites, causing oxidative



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stress to body cells [6]. Additionally, an increased surface area could also act as a carrier for other pollutants, which could negatively affect cells [7]. Therefore, it has been suggested that a particle surface area could be an appropriate metric for the health effect of UFPs [8].

Health effects are also related to NP composition. NPs are mainly composed of organic compounds, elemental carbon, trace metal oxides, sulphate ions, and nitrate ions [9,10]. Most of the mass of NPs is typically composed of carbonaceous materials with small contributions of inorganic ions, thus confirming combustion as their dominant source [11]. The chemical composition of NPs demonstrates spatiotemporal variability due to variations in local sources and their proportional contribution. Kuhn et al. [12] reported that in Los Angeles, CA, USA, NPs were predominantly composed of organic compounds and ammonium and sulphate salts, contributing 45–55% and 35–40%, respectively. Sardar et al. [13] analysed the composition of NPs and found that the contribution of organic compounds ranged from 32 to 69%, elemental carbon from 1 to 34%, sulphate from 0 to 24%, and nitrate from 0 to 4% in Los Angeles. According to Pakkanen et al. [14], trace elements (Ca, Na, Fe, K, and Zn) in NPs were in higher proportions than heavy metals (Ni, V, Cu, and Pb) in Helsinki. Ermolin et al. [15] analysed the concentration of several toxic elements (Ni, Zn, Cd, Ag, Sn, Se, Te, Hg, Tl, Pb, and Bi) in NPs and found that their concentration was higher in volcanic ash than in any other sample. They analysed samples collected in Kamchatka, Far East Russia, and Andes and Chile. Exposure to NPs induces several end health points (diseases), including asthma, lung cancer, COPD (chronic obstructive pulmonary disease), diabetes, colon cancer, Alzheimer's disease, Parkinson's disease, ischemic stroke, heart arrhythmia, myocardial ischemia, coronary heart diseases, cerebral epilepsy, DNA damage, infertility, carcinogenicity, oxidative stress, and inflammation [1,2,16,17].

Road transport is a major source of particle pollution in the urban environment [18]. The NP emission contribution of road transport varied spatially and ranged from 32% in Greece to 97% in Luxemburg [19]. In some other European countries (Spain, France, Germany, the UK, Italy, and Poland), road transport contributed about 72% of the total PM emissions [19]. Road traffic contributes significantly to atmospheric nanoparticle number concentrations and, hence, alters the particle number size distribution in the urban atmosphere [20]. Road transport, in addition to particles, emits large quantities of several gaseous pollutants, e.g., CO, NOx, SO₂, and VOCs. Pollutant emissions from vehicles are related to the vehicle type (such as light and heavy-duty vehicles), vehicle age, operating and maintenance conditions, exhaust treatment, type and quality of fuel, quality of tyre and brake materials, type of engine lubricants used, and driving behaviour [21]. Pollutant emissions have significantly decreased from 1990 to 2017, demonstrating a reduction of 87% in CO, 66% in SO₂, 40% in NOx, 44% in PM_{2.5}, and 35% in PM₁₀ across Europe [22]. Nanoparticle number emissions from road transport can be divided into exhaust emissions and non-exhaust emissions. Exhaust emissions are emitted through the exhaust when the vehicle engine is running, whereas non-exhaust emissions are not emitted by the vehicle exhaust and are rather released into the air by brake wear, tyre wear, and road wear. The proportion of particle emissions from non-exhaust emissions is predicted to increase in the future due to the significant reduction in exhaust emissions, and by 2030, non-exhaust emissions will constitute about 90% of all particle emissions from road traffic in the UK [21]; however, the proportion of exhaust emissions to non-exhaust emissions varies spatially among various cities and countries. Although uncertainty remains with respect to the amount of particles emitted from non-exhaust emissions under real-world driving conditions, non-exhaust emissions are likely to increase in the future due to the growing number of electric vehicles.

Monitoring techniques, source apportionment, and emission inventory of different emission sources of fine particles (PM_{2.5}) and coarse particles (PM₁₀) are more mature and

well defined, whereas the use of these tools for nanoparticle number is generally a continuation from PM_{2.5} and PM₁₀ [23]. Such techniques consist of emission inventories [24], receptor modelling [25], and dispersion modelling [26]. Dispersion modelling is generally carried out after the emission sources are quantified using the first two techniques. The main purpose of these three approaches is related to different aspects of air quality research, such as air pollutant characteristics, sources, processes, effects, and control [27]. Various techniques have been developed and used to collect high-resolution temporal data of nanoparticle number concentrations in urban areas; however, high-resolution spatial data are still lacking due to the limited number of monitoring stations in the UK and elsewhere [28]. Here, several emission inventories are provided as references, which are developed in different countries around the world, covering a range of air pollutants [27]: (1) Multi-resolution Emission Inventory for China: Region—China; pollutants covered—SO₂, NO_x, CO, NMVOC, NH₃, PM₁₀, PM_{2.5}, BC, OC, and CO₂. (2) Intercontinental Chemical Transport Experiment Phase B (INTEX-B): Region—Mexico City and Asian countries; pollutants covered— O_3 and precursors, aerosols and precursors, and greenhouse gases (GHGs). (3) Regional Emission Inventory in Asia (REAS): Region—Asia; pollutants covered—SO₂, NO_x, CO, NMVOC, PM₁₀, PM_{2.5}, BC, OC, NH₃, and CO₂. (4) Clean Air Policy Support System (CAPSS): Region—Korea; pollutants covered—CO, NOx, SOx, total suspended particle (TSP), PM₁₀, PM_{2.5}, VOC, NH₃, and black carbon (BC). (5) Representative Concentration Pathway (RCP): Region-Global; pollutant covered-black carbon (BC), organic carbon (OC), CH₄, sulfur, NOx, VOC, CO, and NH₃. (6) Greenhouse Gas and Air Pollution Interactions and Synergies (GAINS): Region-Europe, China, and India; pollutant covered—SO₂, NO_x, PM_{2.5}, PM₁₀, NH₃, VOC, and CO₂. Furthermore, there are some examples of the nanoparticle number emission inventory, e.g., Kukkonen et al. [29], who developed an emission inventory for five major cities across Europe, namely, Helsinki (Finland), Oslo (Norway), London (UK), Rotterdam (the Netherlands), and Athens (Greece). Kuenen et al. [30] developed a regional emission inventory, which estimated emissions over the whole of Europe. In their emissions inventory, they not only included the main air pollutants (CH₄, CO, NH₃, NMVOC, NOx, SO₂, PM₁₀, and PM_{2.5}) but also NPs. It is important to mention that particle concentrations in the atmosphere are affected not only by emissions sources but also by meteorology and interaction with other pollutants (e.g., ozone) [31].

Even though NPs are more dangerous to public health, atmospheric NPs are still not regulated due to insufficient data availability. Compared to PM_{2.5} and PM₁₀, much fewer studies have considered NPs. This is probably due to two main reasons: (a) The data availability problem—hourly concentrations of gaseous pollutants (e.g., NO₂, NO, CO, and O_3) and mass concentrations of PM_{10} and $PM_{2.5}$ are available abundantly in the UK and elsewhere; in contrast, the NP data are very limited. (b) According to the UK Air Quality Standards Regulations 2010, NO₂, O₃, PM₁₀, and PM_{2.5} are among the regulated outdoor air pollutants, and their monitoring and routine reporting are mandatory, whereas NPs are not regulated and their monitoring is not mandatory. Due to these constraints, it has been challenging to develop dispersion models for nanoparticle number concentrations and assess the impact of various policies for reducing nanoparticle number concentrations in urban areas. The purpose of the paper is to quantify the emissions of nanoparticle numbers from road transport in Leeds and analyse their spatial variability. To achieve the purpose, two approaches have been used: a simple model and a detailed model and their performance has been compared. The main strong points, which highlight the importance of this study, are:

- 1. This study uses detailed traffic flow and composition data. Data on road traffic flow and composition were estimated for both urban roads and motorways employing Leeds Transport Model.
- 2. Emissions of NPs from both exhaust and non-exhaust emissions are estimated in this study. For this purpose, a detailed literature review was conducted to collect nanoparticle number emission factors (NPNEF) for exhaust emissions. NPNEF for non-exhaust emissions were calculated from mass-based EFs using previously established models.
- 3. Two approaches were employed for the nanoparticle number emission estimation, which were referred to as 'a detailed model' and 'a simple model' (described in Section 2).
- 4. Estimated emissions of the two models were compared using several statistical metrics, such as R, R², RMSE, FAC2, and NMB.
- 5. The road transport emission inventory developed for the estimation of nanoparticle number emissions and emission maps produced in this paper can be used for air quality management, assessing the impact of policy interventions, and for developing a dispersion model for the estimation of nanoparticle number concentrations in urban areas. However, no previous nanoparticle number emission inventory existed and there were no nanoparticle number monitoring stations in Leeds, which was a challenge for validating the models' outputs.

2. Materials and Methods

In this paper, nanoparticle number emissions from road transport are characterised in the City of Leeds, UK. In the Section 2, we first analysed road traffic flow and fleet composition, followed by the description of NPNEF for both exhaust and non-exhaust emissions. Finally, the methodology for nanoparticle number emission calculations on different road links in Leeds is described.

2.1. Traffic Data

Traffic data used in this study were estimated by employing a computer transport model, referred to as the Leeds Transport Model, which is described in detail in the Leeds Clean Air Zone Report 2020 [32]. In the past, the Leeds Transport Model has been used to help support business cases for major transport schemes and to assess the impact of the site allocation plan. The highway network element of the Leeds Transport Model used the Saturn modelling suite. The model covered the whole of Leeds District, together with neighbouring local authorities and national roads and rail links. Figure 1 shows the coverage of the Saturn simulation network, which extends to the Leeds District boundary or beyond. The 'fully modelled area' encircled by a red line in the figure relates to the area where the bulk of model calibration has been carried out, whereas the full extent of the road network is encircled by a black line.

The road traffic flow level is reported as annual average daily traffic (AADT), which is the average amount of traffic on an average day (including weekends and holidays). The daily flows were further broken down into four time periods: morning peak (a.m.) 07:00–10:00 h; interpeak (IP) 10:00–16:00 h; evening peak (p.m.) 16:00–19:00 h; and off-peak (OP) 19:00—24:00 and 00:00–07:00 h. An aggregation of several central urban automatic number plate recognition (ANPR) datasets was used to generate an average Leeds fleet composition. Road traffic in Leeds, on average, consisted of 41% petrol cars, 43% diesel cars, 9% LGV, 2% HGV (1.5% rigid and 0.5% artic), and 4.5% coaches [32]. According to the UK National Atmospheric Emissions Inventory in 2020 [33], the UK average fleet composition was different from the one observed in Leeds, as shown in Table 1, demonstrating the



variability of fleet composition spatially from city to city and showing the importance of local data collection.



| Traffic Category | Motorway | Rural | Urban |
|------------------|----------|-------|-------|
| Electric car | 0.4% | 0.5% | 0.5% |
| Petrol car | 33.7% | 42.2% | 47.5% |
| Diesel car | 38.9% | 35.3% | 33.1% |
| Electric LGV | 0.0% | 0.0% | 0.0% |
| Petrol LGV | 0.2% | 0.2% | 0.2% |
| Diesel LGV | 14.9% | 15.9% | 15.4% |
| Rigid HGV | 3.1% | 2.3% | 0.9% |
| Artic HGV | 8.0% | 2.4% | 0.4% |
| | | | |

Table 1. The UK national fleet composition for road traffic in 2020 [33].

2.2. Nanoparticle Number Emission Factors

Emission factors relate the amount of pollutants emitted to traffic flow and composition. Emission factors are generally expressed as the amount of emitted pollutants divided by a unit activity (weight, volume, distance, or duration of the activity emitting the pollutants). NPNEF are expressed as the number of nanoparticles emitted per unit activity. For road transport, here, NPNEFs are expressed as the number of nanoparticles emitted per km travelled by a vehicle. Nanoparticle number emissions are the product of NPNEF, number of vehicles, and distance travelled, as shown in Equation (1).

$$NPNE = NPNEF \times Ni \times Di$$
(1)

where NPNE is nanoparticle number emission, NPNEF is nanoparticle number emission factors, N*i* is the number of vehicles in each vehicle category, and D*i* is the distance travelled by each vehicle in each category.

Vehicle's EFs depend on many parameters, such as vehicle characteristics, emission control technology, fuel specifications, and ambient and operating conditions (e.g., cold-start, cruising, acceleration, etc.). Vehicle-specific nanoparticle number emissions are typically measured using (a) NP measurements in the laboratory, such as chassis and engine dynamometer; (b) roadside remote-sensing; (c) vehicle chasing experiments; (d) road tunnel studies; and (e) onboard measurements with portable emission measurement systems (PEMS) [34,35]. Franco et al. [35] have provided a detailed review of various emission measurement techniques both in the laboratory and in real driving conditions.

NPNEFs for the exhaust emissions of different traffic categories, as well as for mixed fleets, were collected from the literature (provided in Section 3.1). In this paper, NPNEF for the mixed fleet was used from [36]. The following steps were taken by Wang et al. [36] to estimate NPNEF for mixed fleet road traffic under real-world conditions:

- Measure nanoparticle number concentrations (NPNC) at roadsides using differential mobility particle sizers (DMPS). The average NPNC was 27,100 particles/cm³.
- Measure NPNC at the background site (average NPNC at the background site was 5311 particles/cm³).
- Subtract background NPNC (5311) from the roadside NPNC (27,100) to calculate the contribution of road traffic, referred to as ΔC .
- Monitor traffic flow (AADT).
- Estimate dilution rate.
- Convert nanoparticle number concentrations to NPNEF using measured traffic volume and dilution rate [36]:

$$PNEFi = [\Delta Ci(t) \times D(t)]/N \text{ (total)}$$
(2)

where, in Equation (2), delta C (particles/cm³) is the concentration increment of species i, D(t) (m²/s) is the dilution rate, and N is the number of vehicles passing by per unit time. The dilution rate was estimated using the Danish Operational Street Pollution Model (WinOSPM) [36] where wind speed, wind direction, and traffic-generated turbulence were considered. However, Wang et al. [36] did not explicitly report the dilution rate values used in their calculations. Instead, their focus was on deriving emission factors for NOx, particle number, and particle mass based on modelled dilution.

NPNEFs for non-exhaust emissions were calculated from mass-based EFs of NPs. The following equation was used to calculate NPNEF (particles/km) from mass-based EF in units of (g/km) for non-exhaust emissions [37]:

$$Nij = mij/\rho_i v_i$$
(3)

where, in Equation (3), N is NPNEF for non-exhaust emissions, m is EF in terms of mass (generally expressed as $PM_{0.1}$) for non-exhaust emissions of NP, and rho (ρ) is the density of non-exhaust emissions of NP with a value of 0.0016 kg/cm³ for tyre wear and road wear and 0.001 kg/cm³ for brake wear [38–43]. V is the volume of a single NP assuming that each particle is spherical. The volume was calculated using the formula 4/3 π r³ where r is the average radius of the particle and was assumed to be 0.025 µm for NPs. Mass-based EF for NPs was collected from the national atmospheric emission inventory (NAEI), UK [44], given in Table 2. NPNEFs for exhaust emissions were added to non-exhaust emissions to estimate the final NPNEF for different vehicle categories and mixed fleets, which are provided in Section 3.1.

| Vehicle Type | EF for PM _{0.1} (g/km) |
|---------------------------------|---------------------------------|
| Car—petrol | $3.26 	imes 10^{-7}$ |
| Car—diesel | $2.04	imes10^{-6}$ |
| Car—brake wear | $9.37	imes10^{-4}$ |
| Car—tyre wear | $9.37	imes10^{-4}$ |
| Car—road abrasion | $6.12 	imes 10^{-4}$ |
| LGVs—petrol | $1.60 	imes 10^{-7}$ |
| LGVs—diesel | 1.61×10^{-6} |
| LGVs—brake wear | $1.46 	imes 10^{-3}$ |
| LGVs—tyre wear | 1.09×10^{-3} |
| LGVs—road abrasion | $6.12	imes10^{-4}$ |
| Buses and coaches—diesel | 1.22×10^{-6} |
| Buses and coaches—brake wear | $4.29	imes10^{-3}$ |
| Buses and coaches—tyre wear | $1.69 	imes 10^{-3}$ |
| Buses and coaches—road abrasion | $3.10 	imes 10^{-3}$ |
| HGV articulated—diesel | 5.06×10^{-7} |
| HGV articulated—brake wear | $4.08 	imes 10^{-3}$ |
| HGV articulated tyre wear | 3.71×10^{-3} |
| HGV articulated—road abrasion | $3.10 	imes 10^{-3}$ |
| HGV—rigid diesel | $9.67 	imes 10^{-7}$ |
| HGV—rigid brake wear | $4.08	imes10^{-3}$ |
| HGV—rigid tyre wear | $1.64 	imes 10^{-3}$ |
| HGV—rigid road abrasion | $3.10 	imes 10^{-3}$ |

Table 2. Nanoparticle emission factors (NPEF) expressed in the unit of mass (g/km) for different vehicle types [44].

2.3. Nanoparticle Number Emission Estimation and Mapping

In this paper, nanoparticle number emissions were estimated using two approaches: (1) detailed model, which used the flow of various traffic categories and NPNEFs for each category of vehicle types, e.g., cars, buses, LDVs, and HDVs; (2) simple model, which used combined traffic flow of all vehicle categories and NPNEF for the mixed fleet. The simple model is particularly useful for situations when detailed traffic flow, fleet composition, and EF for each vehicle type are not available. In the detailed model, the total nanoparticle number emissions per link were calculated by multiplying the traffic flow of each category type by their NPNEF and distance and then summing them up (Equation (4)).

$$[NPNE]_{ij} = \sum_{i=1}^{i=n} (Xij \times NPNEFij \times Di)$$
(4)

where NPNE is the number of nanoparticles emitted by a traffic category *i* on road link *j* with distance D. *Xij* s the road traffic flow of traffic category *i* on road link *j*, and *NPNEFij* is the emission factor for transport category *i* on road *j* (urban or motorway). Note that the

model is run separately for each link. To present the above formula in a simple way for a single link, it would look like (Equation (5)):

$$[NPNE] = \{((Petrol_Car \times NPNEF) + (Diesel_Car \times NPNEF) + (HGV \times NPNEF) + (LGV \times NPNEF) + (Bus \times NPNEF)) \times D\}$$
(5)

In the simple model, nanoparticle number emissions (NPNE) per link were calculated by multiplying the total traffic flow (the sum of all traffic categories for each link) by the NPNEF of the mixed fleet for both motorways (1.78×10^{14}) and urban roads (2.15×10^{14}) [36], as in Equation (6).

$$NPNE = Traffic_flow \times EF_mixed_fleet \times D$$
(6)

After the nanoparticle number emission calculation using the above equations, emission maps were developed by importing the emissions to a Geographical Information System. Emissions of both methods were compared to quantify the difference.

Although there was a strong correlation between the outputs of the two models, the simple model underestimated the nanoparticle number emissions compared to the detailed model. The outputs of the two models are compared using several statistical metrics: correlation coefficient (r), coefficient of determination (R^2), root-mean-squared error (RMSE), factor of tow (FAC2), and normalised mean bias (NMB). RMSE is a good measure of the error between two datasets, which calculates how close or far the compared values are from each other. NMB estimates the average over or underprediction of the simple model, compared to the detailed model. NMB value between +0.02 and -0.02 shows acceptable performance of the simple model. The correlation coefficient (r) shows the strength of the linear relationship between the two variables (here, the estimation of the simple and detailed models). 'r' should have a value as close to one (± 1) as possible; however, generally, a value ranging from ± 0.5 to ± 0.99 indicates reasonably good performance of the simple model. R-squared is simply the squared value of the correlation coefficient and its value ranges from 0 to 1. FAC2 is the fraction of the simple model estimation within a factor of 2 of the detailed model [45]. In addition to these metrics, previously, several other metrics have been used for model evaluation, including the mean absolute error (MAE) and mean absolute percentage error (MAPE) (e.g., [31,46]).

3. Results and Discussion

Section 3 is made of two subsections: Section 3.1 describes and compares the results of the simple and detailed models. In this section, the NPNEFs for different vehicle categories are also presented and briefly discussed; however, a detailed discussion of the NPNEF is provided in Section 3.2. Section 3.2 reviews the relevant literature on NPNEFs, which provides further insights into understanding this work and other relevant work carried out previously.

3.1. Calculation and Mapping of Nanoparticle Number Emissions

In this paper, NPNEFs are used for both exhaust and non-exhaust emissions of road traffic. NPNEFs for exhaust emissions were collected from already published literature (references are provided in Table 3), whereas for non-exhaust emissions, NPNEFs were calculated from mass-based NPEFs [44]). The final NPNEFs (particles/km/vehicle), which are the sum of the exhaust and non-exhaust EFs for different traffic categories for both urban roads and motorways, provided in Table 3, were used to calculate nanoparticle number emissions for each road link. Table 3 presents NPNEFs for both urban roads and motorways. It is interesting to see that NPNEFs are higher for urban roads than for

motorways. Generally, urban roads experience higher congestion and face more stopand-start traffic situations due to more traffic lights and junctions, whereas traffic flow on motorways is generally smoother, experiencing comparatively less congestion. It is reported that particle emissions were higher on urban roads than on rural roads and motorways, a result of frequent braking in urban areas, which increased the amount of brake wear emissions [47–49]. Probably due to these reasons, EFs are higher for urban roads. Furthermore, NPNEFs are higher for diesel vehicles than for petrol vehicles, which is expected. According to the Office of National Statistics [50], although vehicle miles have increased on the roads by 29%, the total fuel use for road transport has remained relatively stable between 1990 and 2017 in the UK because of the improvement in the fuel use efficiency of new vehicles. Furthermore, petrol use in the UK has decreased by 52% from 27 Mtoe (million tonnes of oil equivalent) in 1990 to 13 Mtoe in 2017, whereas the use of diesel has increased by 145% from 11 Mtoe in 1990 to 27 Mtoe in 2017 [50]. The temporal trend showed that petrol use was higher than diesel use; however, in 2005, diesel use exceeded petrol use [50]. The switch between petrol and diesel use has made atmospheric particles the pollutants of concern in urban areas.

Table 3. Nanoparticle number emission factors (NPNEFs) (particle/km) for different vehicle types on urban roads and motorways (EE = exhaust emissions, NEE = non-exhaust emissions).

| Veh. Category | Road Type | Exhaust | Brake Wear | Tyre Wear | Road Wear | EE and NEE |
|------------------|-----------|-------------------------------|--------------------|--------------------|-------------------|-----------------------|
| Petrol car | Urban | $8.00	imes10^{12}\mathrm{a}$ | $8.95 	imes 10^3$ | $8.95 	imes 10^3$ | $5.84	imes10^3$ | $8.00 	imes 10^{12}$ |
| Diesel car | Urban | 6.08×10^{14b} | $8.95 	imes 10^3$ | $8.95 	imes 10^3$ | $5.84	imes10^3$ | $6.08 	imes 10^{14}$ |
| LGV petrol | Urban | $5.00	imes10^{12}$ a,c | $1.39 	imes 10^4$ | 1.0410^4 | $5.84 	imes 10^3$ | $5.00 	imes 10^{12}$ |
| LGV diesel | Urban | $4.86	imes10^{13}$ a,c | $1.39 	imes 10^4$ | $1.04 	imes 10^4$ | $5.84	imes10^3$ | $4.86 	imes 10^{13}$ |
| Coach | Urban | $7.06\times10^{14\text{b}}$ | $4.09	imes10^4$ | $1.61 	imes 10^4$ | $2.96	imes10^4$ | $7.06	imes10^{14}$ |
| HGV artic | Urban | 3.45×10^{14d} | $3.89 	imes 10^4$ | $3.54 	imes 10^4$ | $2.96	imes10^4$ | $3.45 	imes 10^{14}$ |
| HGV rigid | Urban | 3.45×10^{14d} | $3.89	imes10^4$ | $1.56 	imes 10^4$ | $2.96	imes10^4$ | $3.45 	imes 10^{14}$ |
| Mixed fleet | Urban | $2.15\times10^{14\mathrm{e}}$ | $2.35 	imes 10^4$ | 1.51×10^4 | $1.60 	imes 10^4$ | $2.15 	imes 10^{14}$ |
| Petrol car | Motorway | $1.64 	imes 10^{12}$ | $1.86 	imes 10^3$ | $4.39 	imes 10^3$ | $5.84	imes10^3$ | $1.64 	imes 10^{12}$ |
| Diesel car | Motorway | 4.380×10^{14} | $1.86 	imes 10^3$ | $4.39 	imes 10^3$ | $5.84	imes10^3$ | $4.380 	imes 10^{14}$ |
| LGV petrol | Motorway | $3.60\times10^{13}{}^{\rm g}$ | $4.17	imes10^3$ | 7.33×10^3 | $5.84	imes10^3$ | $3.60 	imes 10^{13}$ |
| LGV diesel | Motorway | $2.20 	imes 10^{14}$ | $4.17	imes10^3$ | 7.33×10^3 | $5.84	imes10^3$ | $2.20 	imes 10^{14}$ |
| Coach | Motorway | $3.60\times10^{13}{}^{\rm g}$ | $1.26 	imes 10^4$ | $1.86 	imes 10^4$ | $2.96	imes10^4$ | $3.60 	imes 10^{13}$ |
| HGV artic | Motorway | $7.02 	imes 10^{13}$ g | $1.85 	imes 10^4$ | $2.79 	imes 10^4$ | $2.96	imes10^4$ | $7.02 	imes 10^{13}$ |
| HGV rigid | Motorway | $3.60 	imes 10^{13}$ g | 1.85×10^4 | 1.24×10^4 | $2.96 	imes 10^4$ | 3.60×10^{13} |
| Mixed fleet | Motorway | $1.78	imes10^{14}$ e,f | 8.81×10^3 | $1.18 	imes 10^4$ | $1.60 	imes 10^4$ | $1.78 	imes 10^{14}$ |

^a [34]; ^b [51]; ^c [52]; ^d [53]; ^e [36]; ^f [54], ^g [55].

According to the outputs of the LTM, diesel cars were 43% of the total flow in Leeds, whereas petrol cars were 41%, LGVs were 9%, HGVs were 2%, and buses and coaches were 4.5%. Nanoparticle number emissions were calculated for each road linked in Leeds and mapped using ArcGIS 10.6. Emissions were calculated for annual average daily traffic (AADT), which was the total volume of vehicle traffic on a road for a year divided by 365 days. Figure 2 shows nanoparticle number emissions (particle numbers per km per AADT) in Leeds for road transport calculated from different traffic categories using

the detailed model. Diesel cars not only had relatively higher NPNEFs but also had the highest percentage of traffic flow in Leeds, which resulted in the highest amount of nanoparticle number emissions. The highest number of NPs per AADT was emitted by diesel vehicles (2.26×10^{21} particles), whereas the lowest was emitted by petrol LGVs (1.99×10^{19} particles). The total number of particles for the various vehicle categories and both simple and detailed models are provided in Table 4. These values are the sum of nanoparticles emitted on all links in Leeds. Among the vehicle categories, diesel emitted the highest number of nanoparticles, followed by HGVs and petrol cars.

Figure 3 shows the outcome of the simple model, which used NPNEF of the mixed fleet $(2.15 \times 10^{14} \text{ for urban roads and } 1.78 \times 10^{14} \text{ for motorways})$ (Table 3) and AADT on each road link. Both models presented a similar pattern of nanoparticle number emissions in Leeds. The highest concentrations shown by the red colour are in the southern and eastern parts of the city, which are linked to the motorway traffic, such as M1, M62 and M621, and other busy roads. The detailed model shows slightly higher levels of nanoparticle number emissions (maximum 4.73×10^{18} particles/link/AADT) than the simple model (maximum 3.51×10^{18} particles/link/AADT). The sum of nanoparticles emitted for all links was also higher for the detailed model than for the simple model (Table 4). The outputs of the simple and detailed models are compared in Figure 4, which presents a similar pattern in the outputs of both models. However, the resultant nanoparticle number emissions are higher for the detailed model than for the simple model. In addition to the line plot, a scatter plot was produced for the simple model vs. the detailed model. The scatter plot shows that the line is not 45 degrees. All the points fall below the line of 45 degrees, which shows that the number of particles is less for the simple model. However, all points make a straight line indicating a strong correlation between the two models. For further analysis, several statistical metrics were calculated to quantify the association between the simple and the detailed model: (i) FAC2 (Factor of 2) is a measure of how well the simple model outputs compare to the detailed model outputs. A value closer to 1 is ideal, as it indicates that the outputs of both models are close to each other within a factor of 2. A value of 0.95 suggests that, on average, the simple model outputs are within about 95% of the detailed model outputs. (ii) MB (mean bias) measures the average difference between the two model outputs. A negative value indicates that the simple model is underpredicting on average. Here, -1.92×10^{17} suggests a slight negative bias in the simple model's predictions, meaning the simple model tends to slightly underestimate the emissions in comparison to the detailed model. (iii) MAE (mean absolute error) is the average of the absolute differences between the simple and detailed model estimations. It provides a measure of the magnitude of errors without considering their direction (ove- or under prediction). A value of 1.92×10^{17} suggests that, on average, the errors (whether positive or negative) are about 1.92×10^{17} in magnitude. (iv) RMSE (root-mean-square error) is one of the most commonly used measures of the difference between the two datasets. It provides a sense of how much error exists in the simple model predictions compared to the detailed model prediction. A value of 2.77×10^{17} is relatively low, suggesting that the two models' predictions are fairly close to each other. (v) r is the correlation coefficient between the simple and detailed model estimations. A value of 0.998 indicates a very strong positive correlation between the two models' estimation, indicating that they are closely related and move in the same direction. A *p*-value of 0 indicates very strong evidence that the correlation between the two models' outputs is significant.



Figure 2. Nanoparticle number emissions (number particles/km) from road transport in Leeds using the detailed model for the year 2020.



Figure 3. Nanoparticle number emissions (number of particles/km) from road transport in Leeds using the simple model for the year 2020.



Figure 4. Comparing the outcomes of the simple vs. detailed model, showing a strong association between the estimations of the two models: the upper panel shows a line plot of both simple and detailed models, and the lower panel shows a scatter plot between the simple and detailed models' outputs. In the upper panel *x*-axis, "Index" is simply a sequence of numbers (showing the counts of data points).

| Vehicle Category | NPN Emissions (AADT) |
|------------------|-----------------------|
| Petrol cars | $1.09	imes 10^{20}$ |
| Diesel cars | $8.81	imes 10^{21}$ |
| LGV | $8.63	imes10^{19}$ |
| HGV | $1.76	imes 10^{20}$ |
| Artic HGV | $6.86	imes 10^{19}$ |
| Coaches | $3.37	imes 10^{20}$ |
| Detailed model | $9.58	imes 10^{21}$ |
| Simple model | 5.79×10^{21} |

Table 4. NPN emissions for all links in Leeds per AADT for different vehicle categories and both detailed and simple models.

3.2. Discussion on Nanoparticle Number Emissions

This was the first research project of its kind in Leeds, which described road transportrelated emission sources and NPNEFs; therefore, due to the lack of previous data, it was not possible to make a comparison with previous studies. Previously, several studies had been carried out in other cities, which analysed nanoparticle emissions from different sources, including road traffic [29,56]. Kukkonen et al. [29] modelled particle number concentrations in five major cities across Europe, namely, Helsinki (Finland), Oslo (Norway), London (UK), Rotterdam (Netherlands), and Athens (Greece). They compiled emission inventories and ran dispersion models for the five cities. They found that the concentrations of particle numbers in the selected cities were dominated by the emissions originating from local vehicular traffic. They reported that megacities such as London and Athens experienced higher particle number concentrations than Helsinki and Oslo. In the current study, we estimated the amount of nanoparticle number emissions in Leeds using previously established NPNEF for road transport and found that nanoparticle number emissions were dominated by diesel vehicles. The findings of the current study were in agreement with Harrison et al. [56] who reported that diesel engine emissions were by far the largest source of NPs and presented a threat to public health in urban areas.

Recently, Kuenen et al. [30] developed a regional emission inventory, which estimated emissions over the whole of Europe. In their emissions inventory, they included not only the main air pollutants (CH₄, CO, NH₃, NMVOC, NOx, SO₂, PM₁₀, and PM_{2.5}), but also NPs. The results were, however, not comparable to the current study due to the different nature of the two studies. Kuenen et al. [30] developed a regional emission inventory, whereas the current study quantified nanoparticle number emissions only for an urban area. Vouitsis et al. [55] carried out a detailed review of NPNEF for road vehicles and came up with gap-filling methods for those vehicles or fuel types for which EFs were not available. Vouitsis et al. [55] presented exhaust particulate emission factors for light and heavy-duty vehicles for different driving conditions (urban, rural and highway). The authors of [55] estimated EF for both the total particle number (TPN) and solid particle number (SPN). The TPN included both solid and semi-volatile particles. Semi-volatile particles may be removed by gradual oxidation to lighter species and/or evaporation [57]. TPN emissions may be several times higher than SPN, especially at high vehicle load operation because of the sulphur-driven nucleation. The NPNEFs proposed by Vouitsis et al. [55] after the gap-filling estimation are provided in Table 5. Given the limited number of measurements of Euro 6 cars as well as the identical emission limits with Euro 5 cars and the compliance of the latter with limits, the final Euro 6 EFs proposed were assumed equal to the Euro 5 ones. For details on the gap-filling methodology and the original source

of the NPNEF, readers are referred to Vouitsis et al. [55] and the references therein. Vouitsis et al. [55] only estimated EF for nanoparticles but did not quantify emissions for a given area (e.g., a city or a country); therefore, although their results are useful for this study, they were not comparable directly.

Table 5. The total particle number (TPN) and solid particle number (SPN) emission factors expressed as 10^{11} p/km, as proposed by Vouitsis et al. [55]. CNG = compressed natural gas, LPG = liquefied petroleum gas, DPF = diesel particulate filters, E10 and E50 = 10% and 50% ethanol, and B10 and B50 = 10% and 50% biodiesel.

| TPN | Cars 1.4–2.0 L | | | |
|------|----------------------|----------|----------|----------|
| 1110 | Vehicle Type | Urban Rd | Rural Rd | Highways |
| | Gasol. PFI Euro 4 | 15.3 | 12.2 | 16.4 |
| | Gasol. DI Euro 4 | 163 | 148 | 1183 |
| | Diesel Euro 4 | 1280 | 1080 | 1750 |
| | B10 Euro 4 | 610 | 524 | 848 |
| | B20 Euro 4 | 487 | 418 | 703 |
| | B100 Euro 4 | 468 | 402 | 651 |
| | E10 Euro 4 | 5.8 | 5.1 | 5.4 |
| | E75 Euro 4 | 3.2 | 2.8 | 3.0 |
| | CNG Euro 4 | 1.8 | 3.9 | 410 |
| | LPG Euro 4 | 3.8 | 3.2 | 3.5 |
| | Gasol. PFI Euro 5 | 0.2 | 7.2 | 0.9 |
| | and 6 | 9.2 | 7.3 | 9.8 |
| | Gasol. DI Euro 5 and | 10.1 | 10.0 | 07 E |
| | 6 | 12.1 | 10.9 | 87.5 |
| | Diesel Euro 5 and 6 | 4.1 | 1.6 | 16.4 |
| | B10 Euro 5 and 6 | 2.0 | 0.8 | 7.9 |
| | B20 Euro 5 and 6 | 1.6 | 0.6 | 6.6 |
| | B100 Euro 5 and 6 | 1.5 | 0.6 | 6.1 |
| | E10 Euro 5 and 6 | 3.5 | 3.1 | 3.2 |
| | E75 Euro 5 and 6 | 1.9 | 1.7 | 1.8 |
| | CNG Euro 5 and 6 | 1.1 | 2.3 | 245 |
| | LPG Euro 5 and 6 | 2.3 | 1.9 | 2.1 |
| SPN | Cars 1.4 0 2.0 L | | | |
| 51 N | Gasol. PFI Euro 4 | 9.0 | 7.9 | 8.4 |
| | Gasol. DI Euro 4 | 95 | 76 | 606 |
| | Diesel Euro 4 | 748 | 552 | 900 |
| | B10 Euro 4 | 357 | 269 | 469 |
| | B20 Euro 4 | 282 | 213 | 387 |
| | B100 Euro 4 | 271 | 213 | 358 |
| | E10 Euro 4 | 1.0 | 1.0 | 3.0 |
| | E75 Euro 4 | 0.5 | 1.0 | 1.0 |
| | CNG Euro 4 | 1.7 | 3.9 | 13.6 |
| | LPG Euro 4 | 3.0 | 2.6 | 2.8 |
| | Gasol. PFI Euro | 2 (| 0.1 | 1.0 |
| | 5and6 | 3.6 | 3.1 | 1.3 |
| | Gasol. DI Euro 5and6 | 20 | 11 | 7.5 |
| | Diesel Euro 5 and 6 | 2.2 | 0.9 | 2.3 |
| | B10 Euro 5 and 6 | 1.2 | 0.4 | 4.3 |
| | B20 Euro 5 and 6 | 0.9 | 0.3 | 3.6 |
| | B100 Euro 5 and 6 | 0.9 | 0.3 | 3.6 |
| | E10 Euro 5 and 6 | 0.4 | 0.4 | 0.5 |
| | E75 Euro 5 and 6 | 0.2 | 0.4 | 0.4 |
| | CNG Euro 5 and 6 | 0.7 | 1.5 | 2.1 |
| | LPG Euro 5 and 6 | 0.5 | 0.4 | 1.0 |

| TPN | | Heavy-Du | ty Vehicles | | | | |
|------|---------------------|------------------------------|-----------------|--------|--|--|--|
| | | Rigid | < 7.5 t | | | | |
| | Euro I | 4594 | 3917 | 10,066 | | | |
| | Euro II | 3190 | 2720 | 6990 | | | |
| | Euro III | 3190 | 2720 | 6990 | | | |
| | Euro IV | 673 | 682 | 1880 | | | |
| | Euro V | 673 | 682 | 1880 | | | |
| | Euro VI | 0.7 | 0.7 | 1.9 | | | |
| | | Rigid 7 | 7.5–14 t | | | | |
| | Euro I | 9749 | 7186 | 14,832 | | | |
| | Euro II | 6770 | 4990 | 10,300 | | | |
| | Euro III | 6770 | 4990 | 10,300 | | | |
| | Euro IV | 1430 | 1250 | 2770 | | | |
| | Euro V | 1430 | 1250 | 2770 | | | |
| | Euro VI | 1.4 | 1.2 | 2.8 | | | |
| | | Rigid and Articulated > 14 t | | | | | |
| | Euro I | 15,264 | 11,102 | 19,584 | | | |
| | Euro II | 10,600 | 7710 | 13,600 | | | |
| | Euro III | 10,600 | 7710 | 13,600 | | | |
| | Euro IV | 2240 | 1930 | 3670 | | | |
| | Euro V | 2240 | 1930 | 3670 | | | |
| | Euro VI | 2.2 | 1.9 | 3.7 | | | |
| CDN | Heavy-Duty Vehicles | | | | | | |
| SEIN | | Rigid | < 7.5 t | | | | |
| | Euro I | 3170 | 1528 | 1913 | | | |
| | Euro II | 2210 | 1060 | 1340 | | | |
| | Euro III | 2210 | 1060 | 1340 | | | |
| | Euro IV | 467 | 266 | 360 | | | |
| | Euro V | 467 | 266 | 360 | | | |
| | Euro VI | 0.5 | 0.3 | 0.4 | | | |
| | | Rigid 7 | 7.5–14 t | | | | |
| | Euro I | 6727 | 2803 | 2818 | | | |
| | Euro II | 4700 | 1950 | 1970 | | | |
| | Euro III | 4700 | 1950 | 1970 | | | |
| | Euro IV | 992 | 489 | 530 | | | |
| | Euro V | 992 | 489 | 530 | | | |
| | Euro VI | 1.0 | 0.5 | 0.5 | | | |
| | | Rigid and art | iculated > 14 t | | | | |
| | Euro I | 10,532 | 4330 | 3720 | | | |
| | Euro II | 7350 | 3010 | 2610 | | | |
| | Euro III | 7350 | 3010 | 2610 | | | |
| | Euro IV | 1550 | 755 | 702 | | | |
| | Euro V | 1550 | 755 | 702 | | | |
| | Euro VI | 16 | 0.8 | 07 | | | |

Table 5. Cont.

Giechaskiel et al. [34] analysed SPN emission factors both on the chassis dynamometer in the laboratory and on the road using PEMS. According to their analysis, cold-start and strong accelerations substantially increased SPN emissions. Two heavy-duty vehicles were tested for their emissions. Among them Euro V truck on the road showed emissions of around 2×10^{13} p/km and the Euro VI truck showed emissions of around 6×10^{10} p/km. The light-duty vehicles equipped with the diesel particle filter showed emissions of 8×10^{11} p/km. The port-fuelled injection (PFI) car had SPN emissions a little higher than 1×10^{12} p/km. The on-road gasoline direct injection (GDI) emissions for cars ranged from 8×10^{11} to 8×10^{12} p/km (Table 6). This shows how exhaust emissions change with vehicle engine size and type. Newer engines (e.g., Euro VI) have significantly fewer emissions compared to older engines. Therefore, replacing older vehicles or retrofitting them can cause a reduction in particle emissions. Huang et al. [51] carried out on-road emission measurements of both gasoline- and diesel-fuelled vehicles using a PEMS in Shanghai, China. The driving-based EFs of gaseous pollutants and particle mass and number were obtained on various road types. The particle number emission factors for diesel buses, diesel cars, and gasoline cars were 7.06×10^{14} , 6.08×10^{14} , and 1.57×10^{14} p/km, respectively. The size distribution of the particles emitted from the diesel vehicles were mainly concentrated in the accumulation mode, while those emitted from the gasoline car were mainly distributed in the nucleation mode. Huang et al. [51] reported that the particle number emission rates of petrol cars increased with increasing VSP (vehicle-specific power) and speed. From idling to the highest speed and VSP of a petrol car, the particle number emission increased by 3 orders of magnitude. They concluded that aggressive driving and heavy loads were the main reasons for high emissions. Therefore, it is important to note that driving behaviour and road types can affect particle emissions from both exhaust and non-exhaust sources, which are not accounted for in this study. Table 6 provides a summary of the NPNEFs for various vehicle categories. In addition, Jones and Harrison [58], Kumar et al. [59], and Gidhagen et al. [60] have provided a useful collection of EFs for particle numbers. Wang et al. [61] provided NPNEF in different units, including particles/mile, particles/kg, and particles/bhp-hr (brake horsepower-hour). Lahde and Giechaskiel [62] analysed how solid NPN emissions of size > 4 nm, >10 nm, and >23 nm from bi-fuel vehicles (CNG + Gasoline or LPG + gasoline) and mono-fuel vehicles (CNG) varied with ambient temperature in laboratory at 23 °C and sub-zero temperature (-7 °C). In this study, emissions from CNG and LPG vehicles were not considered, assuming their numbers were not significant in Leeds. Furthermore, how emissions vary with ambient temperature was not accounted for, which, probably, introduces a degree of uncertainty in the outcomes of the models.

Table 6. NPNEF for various vehicle engine technologies using different fuel types collected from the literature review. CNG = compressed natural gas, LPG = liquefied petroleum gas, DPF = diesel particulate filters, E10 = 10% ethanol, B10 = 10% biodiesel, HDV = heavy-duty vehicle, LDV light-duty vehicle, PFI = port fuel injection, GDI = gasoline direct injection, TWC = three-way catalyst.

| Vehicle Type | Fuel Type | Emission Factors (p/km) | Reference |
|--------------|-------------------|---------------------------------------|-----------|
| HDV, Euro V | Diesel with DPF | $2	imes 10^{13}$ | |
| HDV, Euro VI | Diesel with DPF | $6	imes 10^{10}$ | |
| LDV | Diesel with DPF | $8	imes 10^{11}$ | [34] |
| Car PFI | Gasoline with TWC | 1×10^{12} | |
| Car GDI | Gasoline with TWC | $8 	imes 10^{11}$ - $8 	imes 10^{12}$ | |
| Buses | Diesel | $7.06 	imes 10^{14}$ | |
| Car | Diesel | $6.08 	imes 10^{14}$ | [51] |
| Car | Petrol | $1.57 	imes 10^{14}$ | |
| HDV | Diesel | $1.1 4.9 	imes 10^{14}$ | |
| HDV | Diesel | $0.5 7.4 	imes 10^{14}$ | [50] |
| HDV | Diesel | $1.00 	imes 10^{16}$ | [33] |
| HDV | Diesel with DPF | $1.4	imes10^{13}$ | |

Table 6. Cont.

| Vehicle Type | Fuel Type | Emission Factors (p/km) | Reference |
|---------------------|-----------|---|-----------|
| Mixed vehicle fleet | 1.5%HDV | $5.9 	imes 10^{14}$ – $3.3 	imes 10^{14}$ | |
| LDV | Gasoline | $1.93	imes10^{14}$ | |
| LDV | Gasoline | 4.29×10^{13} | |
| LDV | Gasoline | $3.52 	imes 10^{13}$ | |
| LDV | Gasoline | $1.00 	imes 10^{13}$ | [52] |
| LDV | Diesel | $5.91	imes10^{14}$ | |
| LDV | Diesel | $1.53 	imes 10^{14}$ | |
| LDV | Diesel | $3.33	imes10^{14}$ | |
| LDV | Diesel | $9.64 	imes 10^{13}$ | |
| Car Euro 2 | Diesel | $4.380	imes10^{14}$ | [63] |
| Car Euro 3 | Diesel | $2.747	imes10^{14}$ | [64] |
| Car Euro 4 | Diesel | $2.327	imes10^{14}$ | [65] |
| Euro3 + DPF | Diesel | $3.5 	imes 10^{11}$ | [63] |
| Euro4 + DPF | Diesel | $2.2 	imes 10^{12}$ | [65] |
| Euro 5 | Diesel | $1.7	imes10^{11}$ | [66] |
| Euro3, DI | Gasoline | $2.97	imes10^{13}$ | [63] |
| PFI Euro 1 | Gasoline | $1.88	imes10^{13}$ | [67] |
| PFI Euro 3 | Gasoline | $2.5 	imes 10^{11}$ | [68] |
| PFI Euro 4 | Gasoline | $1.64	imes10^{12}$ | [65] |
| PFI Euro 5 | Gasoline | $5.7	imes10^{11}$ | [69] |
| DI Euro 3 | Gasoline | 7.8×10^{12} | |
| DI Euro 4 | Gasoline | $5.9 	imes 10^{12}$ | [70] |
| DI Euro 5 | Gasoline | $3.7 	imes 10^{11}$ | |
| Car Euro 2 | Biodiesel | $1.3 	imes 10^{14}$ | |
| Car Euro 2 B50 | Biodiesel | $1.95 	imes 10^{14}$ | [71] |
| Car Euro 2 B100 | Biodiesel | $4.45	imes10^{14}$ | |
| Car Euro 3 | Biodiesel | $1.20 	imes 10^{14}$ | [70] |
| Car Euro 3 B10 | Biodiesel | $9.58	imes10^{13}$ | [72] |
| Car Euro 3 | Biodiesel | 3.32×10^{13} | [73] |
| Car Euro 3 | Biodiesel | 2.75×10^{13} | [75] |
| Car Euro 2 | Biodiesel | 1.64×10^{14} | |
| Car Euro 2 B50 | Biodiesel | $1.82 	imes 10^{14}$ | [71] |
| Car Euro 2 B100 | Biodiesel | 1.76×10^{14} | |
| Car Euro 3 | Biodiesel | 1.60×10^{14} | [70] |
| Car Euro 3 B10 | Biodiesel | $1.32 	imes 10^{14}$ | |

١

Car EU4 + E10

Car EU4 + E10

Car EU4 + E > 10

Car EU4 + E > 10

| 10 | | | | |
|--|---------------------------------------|----------------------------|------------------------------|--|
| /ehicle Type | Fuel Type | Emission Factors (p/km) | Reference | |
| Car Euro 3 | Biodiesel | $2.52 	imes 10^{13}$ | [72] | |
| Car Euro 3 | Biodiesel | 2.21×10^{13} | [73] | |
| Car Euro 4 | CNG | $1.8	imes10^{11}$ | [7]4] | |
| Car Euro 4 | CNG | $4.10 	imes 10^{13}$ | [/4] | |
| Car EU5 | LPG | $2.00 	imes 10^{10}$ | [75] | |
| Car EU4 | LPG | $7.00 	imes 10^{10}$ | [76] | |
| Car Euro 3 Car Euro 4 Car Euro 4 Car EU5 Car EU4 | Biodiesel CNG CNG LPG LPG | | [73] [74] [75] [76] | |

Table 6. Cont

Bioethanol

Bioethanol

Bioethanol

Bioethanol

There are three main types of potential uncertainties in this study: (a) uncertainties in the estimation of traffic flow and fleet composition using the Leeds Transport Model (LTM), (b) uncertainties in the emission factors of nanoparticles, and (c) other uncertainties. The uncertainty of nanoparticle emission factors is considered a major part of the overall uncertainties in all transport emission models. These uncertainties originate from the variability of the underlying sample data, i.e., the variability in the emission level of each individual vehicle that has been included in the sample of vehicles used to derive the emission factors. The sample vehicle may not be a true representative of the population and may introduce a degree of uncertainty. In addition, the nanoparticle EFs used in this study were not calculated in Leeds, they were calculated in different urban areas of the UK, EU, or other countries around the world. Also, these EFs were calculated during different environmental conditions and different seasons of the year, which might have introduced a degree of uncertainty. The transport model used for traffic flow estimation, such as LTM, has its own built-in uncertainties, which are based on the assumptions and correction factors of the model. Furthermore, such models use different types of external data (such as population, jobs, and land use) to predict the number of trips. These parameters have their inherited uncertainties, which would add to the uncertainties of the model. Furthermore, various intervention policies on road networks, vehicle speed, and those related to clean air and net zero may affect traffic flow and, hence, may introduce a degree of uncertainty in the model. In this study, we adopted a simple traffic classification framework and did not consider alternative fuel-type vehicles, including electric, CNG, and LPG vehicles, assuming their number at present was not significant, which is a potential weakness of the study. Such simplification of the vehicle categories might have a considerable effect on the amount of nanoparticle emissions estimated in this study. Therefore, the results reported here need to be viewed considering these uncertainties.

 6.00×10^{10}

 $1.8 imes 10^{11}$

 5.10×10^{10}

 1.00×10^{11}

4. Conclusions

Atmospheric NPs, though more dangerous to public health, are relatively less studied pollutants compared to $PM_{2.5}$ and PM_{10} . The reason is that PM_{10} and $PM_{2.5}$ along with some gaseous pollutants are regulated and local authorities are required by the law to monitor and model their concentrations. In contrast, NPs are not regulated and are not monitored and modelled by the local authorities. More recently, researchers have started focusing on atmospheric NPs by measuring their levels, developing emission inventories in

[77]

different EU cities, and investigating their health impacts. In this paper, the main aim was to compile a nanoparticle number emission inventory for road transport in Leeds, UK, using a methodology, which would be transferrable to other urban areas. A literature review was conducted to collect previously published NPNEFs for various traffic categories. Gaps filling was carried out for those emission sources for which NPNEF did not exist, especially for non-exhaust emission sources. Existing NPNEFs varied among different studies due to spatial and temporal variability and variations in the characteristics of the emission sources and measuring techniques. Traffic flow and fleet composition were estimated using the Leeds Transport Model. The amount of nanoparticle number emissions on each road link was calculated using traffic flow and NPNEF. Diesel and petrol vehicles were the dominant sources of nanoparticle numbers in Leeds. Spatially, the eastern and southern parts of the city, which experienced high traffic volumes and were linked with M1, M62, and M621 motorways, showed higher levels of nanoparticle number emissions. It was not possible to validate the results as previous data did not exist for comparison in Leeds. Two models were developed, referred to as the 'detail model' and the 'simple model'. The former used detailed traffic composition and EF data, whereas the latter used total traffic flow and a single EF for the mixed traffic flow. The simple model slightly underestimated nanoparticle number emissions compared to the detailed model. However, the emissions from both models demonstrated a strong correlation with each other, demonstrated by the values of various statistical metrics, i.e., R, R-squared, RMSE, FAC2, and NMB. The simple model provides a simple and easy-to-run alternative for circumstances where detailed data are not available. This paper highlights the importance of atmospheric NPs and presents a framework for nanoparticle number emission modelling. Future work will include developing a detailed dispersion model for modelling NP concentrations in urban areas; however, dispersion models could be data-hungry and time-consuming; therefore, an alternative approach will be explored for such analysis based on machine learning approaches. The road traffic emission inventory will be further fine-tuned and other emission sources included in the future.

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