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Detecting high emitting vehicle subsets using emission remote sensing systems

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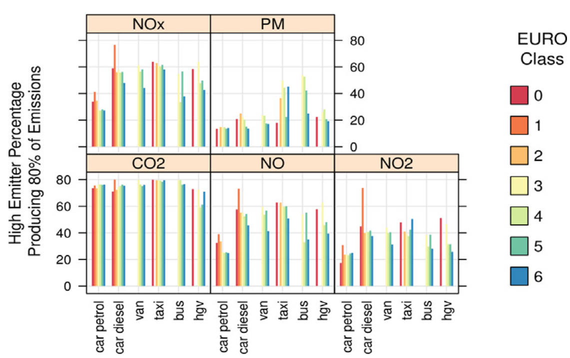
HIGHLIGHTS

- A large vehicle emission remote sensing system (VERSS) dataset is used.
- A new approach, using enrichment factor in cumulative Pareto analysis is used to assess high emitters.
- Different high emitter trends are observed for both different pollutants, and different vehicle classes.
- High emitter trends are observed to be very distinct between diesel and car fleets.
- Heavy goods vehicles show a distinct enrichment in PM emissions.

GRAPHICAL ABSTRACT



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ABSTRACT

It is often assumed that a small proportion of a given vehicle fleet produces a disproportionate amount of air pollution emissions. If true, policy actions to target the highly polluting section of the fleet could lead to significant improvements in air quality. In this paper, high-emitter vehicle subsets are defined and their contributions to the total fleet emission are assessed. A new approach, using enrichment factor in cumulative Pareto analysis is proposed for detecting high emitter vehicle subsets within the vehicle fleet. A large dataset (over 94,000 remote-sensing measurements) from five UK-based EDAR (emission detecting and reporting system) field campaigns for the years 2016–17 is used as the test data. In addition to discussions about the high emitter screening criteria, the data analysis procedure and future issues of implementation are discussed. The results show different high emitter trends dependent on the pollutant investigated, and the vehicle type investigated. For example, the analysis indicates that 23 % and 51 % of petrol and diesel cars were responsible for 80 % of NO emissions within that subset of the fleet, respectively. Overall, the contributions of vehicles that account for 80 % of total fleet emissions usually reduce with EURO class improvement, with the subset fleet emissions becoming more homogenous. The high emitter constituent was more noticeable for pollutant PM compared with the other gaseous pollutants, and it was also more prominent for petrol cars when compared to diesel ones.

1. Introduction

The transport sector has had a key and constructive role in modern civilization (Araghi et al., 2017), while the associated combustion-based emissions are one of the leading drivers of urban air pollution and climate change (Franco et al., 2013; Liu et al., 2017; Ropkins et al., 2009). Correspondingly, abatement strategies and regulations have been established to mitigate the environmental drawbacks of transportation (Fontaras and Dilara, 2012; Singh and Kennedy, 2015). Several emission standards are used across the world to set quantitative limits on the permissible vehicular emissions of different air pollutants. For EU countries, the EURO class

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standard was first introduced in 1992 (EURO 1), and updated in subsequent years with EURO6d most recently being introduced in 2021.

In addition to vehicle emission standards, placed-based strategies have also been developed to reduce vehicle-related air pollution. Clean air zones (CAZs) or low emission zones (LEZs) are one strategy applied to either prohibit or discourage, via financial charges, certain classes of vehicles that are expected to be high emitters from entering populated urban environments (Bigazzi and Rouleau, 2017; Burns et al., 2020). Over 200 LEZs have been implemented in EU countries (please see www.lowemissionzones.eu), while there are many debates on the long and short-term effectiveness of LEZs (Panteliadis et al., 2014; Tartakovsky et al., 2020). Recent vehicle emission remote sensing system (VERSS) campaigns across the world have demonstrated that real-world emission performance and emission improvements due to fleet renovation or EURO upgrading can often show differences in emissions from what is calculated from emission inventories, see (Carslaw et al., 2011; Carslaw and Rhys-Tyler, 2013; Chen and Borken-Kleefeld, 2014; Ghaffarpasand et al., 2020b; Osei et al., 2021).

High emitters are defined as vehicles that are responsible for a disproportionate of the total vehicle fleet emissions, or within vehicle class emissions. High-emitter vehicle subsets are often considered a major reason that the total fleet emissions do not meet the standards expected by emission inventories (Cames and Helmers, 2013; Grange et al., 2017; Hassler et al., 2016). The reasons for high-emitter vehicles can be due to various reasons including vehicle malfunction, tampering and poor maintenance. (Bishop et al., 2012) found that the 1 % high emitter vehicles were responsible for 10 % of the total fleet emissions in the late 1980s and the ratio was raised to near 30 % in the 2000s.

Remote sensing measurements can be used to analyse high-emitter vehicle subsets (Bishop, 2019). VERSSs are usually placed beside the roadside to monitor vehicular emissions by the attenuation of either infrared (IR) or ultraviolet (UV) beams that are intersected by the exhaust plume of passing vehicles (Huang et al., 2019; Bishop et al., 1989). VERSS can measure the real-world exhaustive emissions of passing vehicles. They do this without stopping the vehicles and hence introduce little interruption to the traffic flow. Hence, they provide great potential for the assessment of fleet emissions and the screening of high-emitter vehicle subsets. If VERSS measurements can be successfully used to detect high emitters, then they have great potential to be used within vehicle emissions management policies that specifically target the dirtiest vehicles on the road. However, there are still several technical challenges that need to be addressed before remote sensing can be used to reliably target individual high-emitter vehicles.

The main advantage of VERSSs is the capability of fast real-world assessment of a significant number of vehicles at a relatively low-cost per-vehicle. However, this rapid measurement of the fleet is also considered a limitation of these systems. Since VERSSs measure vehicle emissions for only a short while (less than a second) as the vehicle passes the light beam. Each measurement only contains a single snapshot of a vehicle's emissions for one engine condition. (Qiu and Borken-Kleefeld, 2022) argue that the identification of high emitters is quite uncertain when only supported by single measurements. Furthermore, high emitter screening criteria can be significantly influenced by measurement accuracy and the number of vehicles detected, see for example (Borken-Kleefeld, 2013; Park and Rakha, 2010). Presumably because of the above-mentioned challenges, to date, only a few countries and regions such as mainland China, Hong Kong, Scotland, and certain states in the US have identified high emitters by this type of approach (Qiu and Borken-Kleefeld, 2022).

Another strategy to study high emitters is to identify subgroups of the total fleet which have disproportionately high emissions. This typically involves defining emissions cut points at which a vehicle is considered a high emitter. Different research groups have suggested different cut point criteria, including fixed value cut points as well as several statistical metrics such as n × mean, mean + (n × standard deviations), n^{th} percentile of the population, etc., have been proposed by previous investigators, see the review paper of (Huang et al., 2018b).

(Pujadas et al., 2017) employed screening criteria in which they use the 95 percentile of the respective emission ratios as the screening criteria for light-duty vehicles (LDVs). Their results show approximately 46 % of petrol LDVs were targeted as high emitter vehicles and Pre-Euro vehicles dominantly contributed to the high emitter fleet. (Huang et al., 2018a) used a two-year (2014–2016) remote-sensing measurement dataset to study and analyse the emission rates of the high emitter population of the Hong Kong fleet. They employed fixed cut points for the vehicular emissions of CO, HC, and NO, i.e. 65, 49, 9.95, and 29.11 g/kg fuel, respectively, and targeted a vehicle as a high emitter when it exceeds any of those triple cut points. Results show approximately 12 % of the fleet might be identified as high emitter vehicles and contributed to 70 %, 45 %, and 23 % of the total fleet emission of CO, HC, and NO, respectively. (Huang et al., 2019) used a three year long remote sensing dataset, from April 2014 to April 2017, of Hong Kong as well as transient chassis dynamometer experiments to study the high emitter population in diesel vehicles. Their results show that 36 % and 47 % of the population of EURO4 and EURO5 studied vehicles were high emitters. (Hassani et al., 2021) identify high emitters using fixed cut points which are determined for different remote volumetric concentration ratios ($CO/CO_2 = 0.24$, $HC/CO_2 = 122E\text{-}04$, and $NO/CO_2 = 18E\text{-}04$) measured during VERSS campaigns. Results show that almost one-fifth of the LDV fleet in Tehran, Iran, were high emitters and responsible for over half of the total CO, HC, and NO_x emissions. (Yang et al., 2022) used Gumbel distribution to identify candidate high NO_x emitting diesel light commercial vehicles (vans) based on remote sensing measurements. Results show that 4 % of EURO 6 diesel vans are high emitters and responsible for around 45 % of total NO_x fleet emissions. The application of Gumbel distributions for identifying high emitters was initially proposed by (Rushton et al., 2021).

Most of the fixed cut points or screening criteria in the studies above are based on the absolute emission percentage, while vehicle emission performance is affected by a wide variety of factors. Hence, transforming VERSS measurements, taken under various operating conditions and ambient environments, to target high-emitters is difficult (Kang et al., 2021). Due to such a level of complexity, advanced data analysis technologies and/or methods are used by a few investigators to somehow resolve the problem. For example, (Xie et al., 2019) proposed an algorithm which identifies high-emitters using cut points that are updated automatically. In their method, the VERSS measurements were clustered and labelled by the k-mean clustering algorithm and then the k-nearest neighbour algorithm was used to identify high emitters. (Kang et al., 2021) developed a machine-learning algorithm to combine VERSS measurements and periodic emission inspection records to identify the high emitters. (Qiu and Borken-Kleefeld, 2022) designed a scheme which combines VERSS measurements and second-by-second emission measurements from individual vehicles. The approaches discussed above do not allow for the investigation of the trends of high emitters within different vehicle subsets.

In this paper, we describe and assess a new statistical population-based metric with which to assess and identify trends in high-emitting subgroups of the total fleet using data from VERRS. This approach can provide vehicle fleet managers and city planners with a robust indicator of the relative effectiveness of different fleet management strategies. For example, in regions where vehicle emissions are dominated by a small number of high emitters, then the region is likely to have effective emission-management using stop-and-inspect style activities. Within area with little or high emitter contribution, they would be better addressed using fleet-level initiatives.

2. Materials & methods

2.1. Emissions detection and reporting (EDAR) remote sensing device

The EDAR device applies on-road remote sensing to measure vehicle emissions under real-world conditions. The device is deployed over a section of road at a height of 5 m alongside other instruments, including an automatic number plate recognition (ANPR) camera, and a weather station. EDAR uses open-path laser-based differential absorption LIDAR (DiAL) to provide a measure of the exhaust emissions of a vehicle as that vehicle passes under it. A schematic diagram of EDAR deployment is presented in

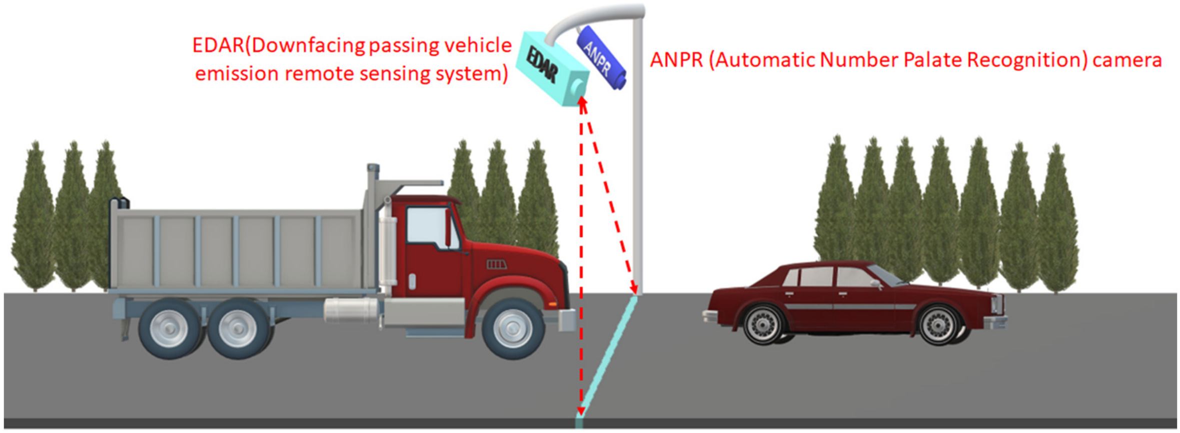


Fig. 1. A schematic picture of EDAR deployment.

Fig. 1. DiAL is widely reported to be more sensitive, selective, and less susceptible to drift than the other techniques applied in the different VERSSs approaches (Abshire et al., 2010; Hager, 2015; Menzies and Tratt, 2003). The EDAR device directly measures NO and NO_2 together to give true NO_x values. EDAR also provides a better measure of PM compared to other available VERSSs (Ropkins et al., 2017), and when compared to emission measurements from Portable Emissions Measurement Systems (PEMS), an acceptable agreement was observed (Ropkins et al., 2017).

2.2. EDAR data

Data from the EDAR device is used from campaigns in five UK urban environments: Tyburn Road in Birmingham, Marylebone Road in Central London, and Blackheath in Greenwich, Edinburgh and Broxburn in West Lothian in 2016–2017. These campaigns generated multiple linked data sets including passing vehicle emission measurements of CO_2, NO_2, NO, and PM (the methodology used for estimating emission rates is provided in the next section) and passing vehicle information from automatic number plate recognition (ANPR), vehicle information data are included vehicle type, model year, model style, manufacturer company, EURO classification, and fuel type. During those campaigns over 94,000 measurements were collected. Table 1 summarizes the overall average distributions of measurement campaigns, testing conditions and distribution of vehicle specific power (VSP) for the major vehicle subsets studied here. The distributions are plotted in different colours to distinguish between vehicles with different EURO classes. It should be reminded that VSP which is developed by (Jimenez-Palacios, 1998) is a function of speed and acceleration and defined as the required power per vehicle mass to have a constant speed, see for example (Ghaffarpasand et al., 2020a).

2.3. EDAR data analysis

Most VERSSs, including EDAR, report the concentration ratios of pollutants over CO_2 (a proxy of fuel consumption) in the tailpipe exhaust plume. A method which has been widely used by previous investigators such as (Bernard et al., 2022; Ghaffarpasand et al., 2020a; Ropkins et al., 2017) is used here to convert measured emission ratios into distance-specific metric, i.e. grams of pollutants per kilometre. It is worthwhile to note that (Ropkins et al., 2017) assessed the EDAR-measured real-world emission factors which were produced by the current method and the results of portable emission measurement systems (PEMS) and found very encouraging agreements. Similarly (Bernard et al., 2022) assesed the distance-specific emission factors achieved from the several RS campaigns across the EU against the PEMS measurements, with an already satisfactory agreement found.

The gaseous species CO_2 emission ratios were converted to g/km using the following equation:

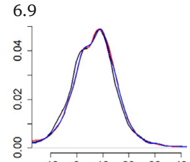
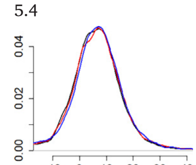
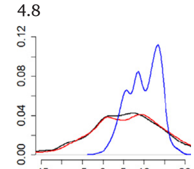
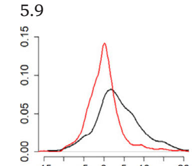
$$[n]_{g/km} = ratio_{[n]/CO_2} \times [CO_2]_{g/km} \times mwt_{[n]/CO_2} \tag{1}$$

where n is the species (CO, NO, NO_2, and PM); $ratio_{[n]/CO_2}$ is the EDAR measurement; and $mwt_{[n]/CO_2}$ is the molecular weight ratio of n to CO_2. As can be observed, the CO_2 emission rate of the studied vehicles ($[CO_2]_{g/km}$) is required to convert EDAR measured pollutant data into emission rates in units of g/km. Where available, CO_2 emission rate (g/km) values were taken from the archives of Driver Vehicle Licensing Authority (DVLA), the Society of Motor Manufacturers Traders (SMMT), and Motor Vehicle Registration Information System (MVRIS). These were predominately cars, taxis, and vans for which DVLA/MVRIS/SMMT have more complete records and in these cases accounted for about 97 %, 96 %, and 62 % of observations for the Blackheath, Marylebone, and Tyburn sites, respectively. Where these were used, the real-world corrections were applied in the form:

$$[CO_2]_{g/km,rw} = [CO_2]_{g/km,archive} \times \left(1 + \left(\frac{RWC}{100\%}\right)\right) \tag{2}$$

where $[CO_2]_{g/km,\ archive}$ is the DVLA/MVRIS/SMMT CO_2 g/km value; and RWC is the percentage real-world correction (see (Stewart et al., 2015)).

Table 1
Summary of the monitored fleet characteristics for EDAR campaigns across the five UK cities for 2016–2017. The blue, red, and black solid lines are corresponded to EURO 4, EURO 5, and EURO 6 vehicles in the studied vehicle classes, respectively. The y-axis is the density of the distribution functions.

	Car petrol	Car diesel	Vans	HGV & buses
# of measurement	36,668	29,146	10,736	2567
Average age (year)	6.9	5.4	4.8	5.9
VSP (kW/ton)				

Where these values were not available, CO_2 g/km values were taken from COPERT5 if available (EMISIA/EEA, 2016). Here, following the guidance of Emisia, hot urban emission factors were used rather than speed-curve outputs based on individual vehicle instantaneous speed measurements (EMISIA/EEA, 2016). For this average speed (and time contributions) of 26.68 km/h (75 %) and 13.90 km/h (25 %) were assumed for urban off-peak, and urban peak conditions (TFL, 2016), and COPERT5 values were assigned to EDAR data on basis of COPERT5 model inputs (vehicle type, segment, engine size, and weight) where information was available. This generated a further 1 % of car values, 4 % of taxis values, and 20 % of van values, as well as 49 % of heavy-duty vehicle (HGV) values and 11 % of bus values, mostly Blackheath, Marylebone, and Tyburn values. Although COPERT5 motorcycle values were available, these could not be unambiguously aligned with DVLA/MVRIS/SMMT records in early work, so a purpose written engine-size motorcycle emission factor model was used to assign motorcycle emission factors where engine size was available. The remaining missing cases were hole-filled using same-vehicle or nearest-vehicle matching mean values. Vehicles with no license plate information or license plate records that could not be matched with EDAR records (about 3 % of cases) were analysed separately. However, the absolute values (g/km) calculated in this manner should not be compared with g/km values derived from, for example, type approval test cycles without careful consideration. Ratios observed using the EDAR or any other VERSSs in similar remote sensing surveys are single-point measurements of a passing vehicle and do not necessarily represent emissions across an urban test cycle or urban journey. It should be noticed that NO_x emissions were calculated as NO_2 equivalents (NO_2 + NO as NO_x), and that PM emissions were reported in a nominal unit that approximates g/km.

Particulate emissions were reported by EDAR in units of nanomole/mol. As an alternative to reporting emissions in these molar units, the PM comparison plot generated from data collected as part of the PEMS drive-through EDAR evaluation exercise of the Birmingham and London EDAR study (Ropkins et al., 2017) is used as a field calibration to convert the EDAR nanomole/mol outputs to gram of particulate per kilogram CO_2 equivalents, before applying the above method to derive an estimate of PM emissions in g/km units.

2.4. Data aggregating

Where data is aggregated by vehicle subset, for example, vehicle type or EURO class, a bootstrapping approach was adopted based on that previously used by Carslaw & Rhys-Tyler (Carslaw and Rhys-Tyler, 2013). In bootstrapping methods, the selected sample is repeatedly randomly subsampled and descriptive statistics such as the arithmetic mean calculated for each subsample. The mean of these means is then taken as the mean and additional statistics such as confidence intervals are calculated based on distributions of these values. For this work, the process was undertaken using the R package boot (Canty and Ripley, 2015, Davison and Hinkley, 1997).

2.5. Pareto analysis and high-emitter vehicle subsets

A cumulative Pareto approach is applied here for the characterisation and analysis of potential high-emitter vehicle subsets in the fleet. The Pareto analysis orders data according to the magnitude of the numerical value i.e. from highest to lowest emitter. This provides an indication of variance within the sample set and the significance/number of outliers (Backhaus, 1980). When converted to a cumulative sum plot it becomes possible to characterize high emitter subsets. The first vehicle counted is the highest emitter, the second added is the second-highest emitter, and so on. In other words, emission measurements are reordered, highest to lowest, and converted to a cumulative sum. The Pareto ordering method provides a robust basis for the characterisation of emission data, for example, high emission values significantly

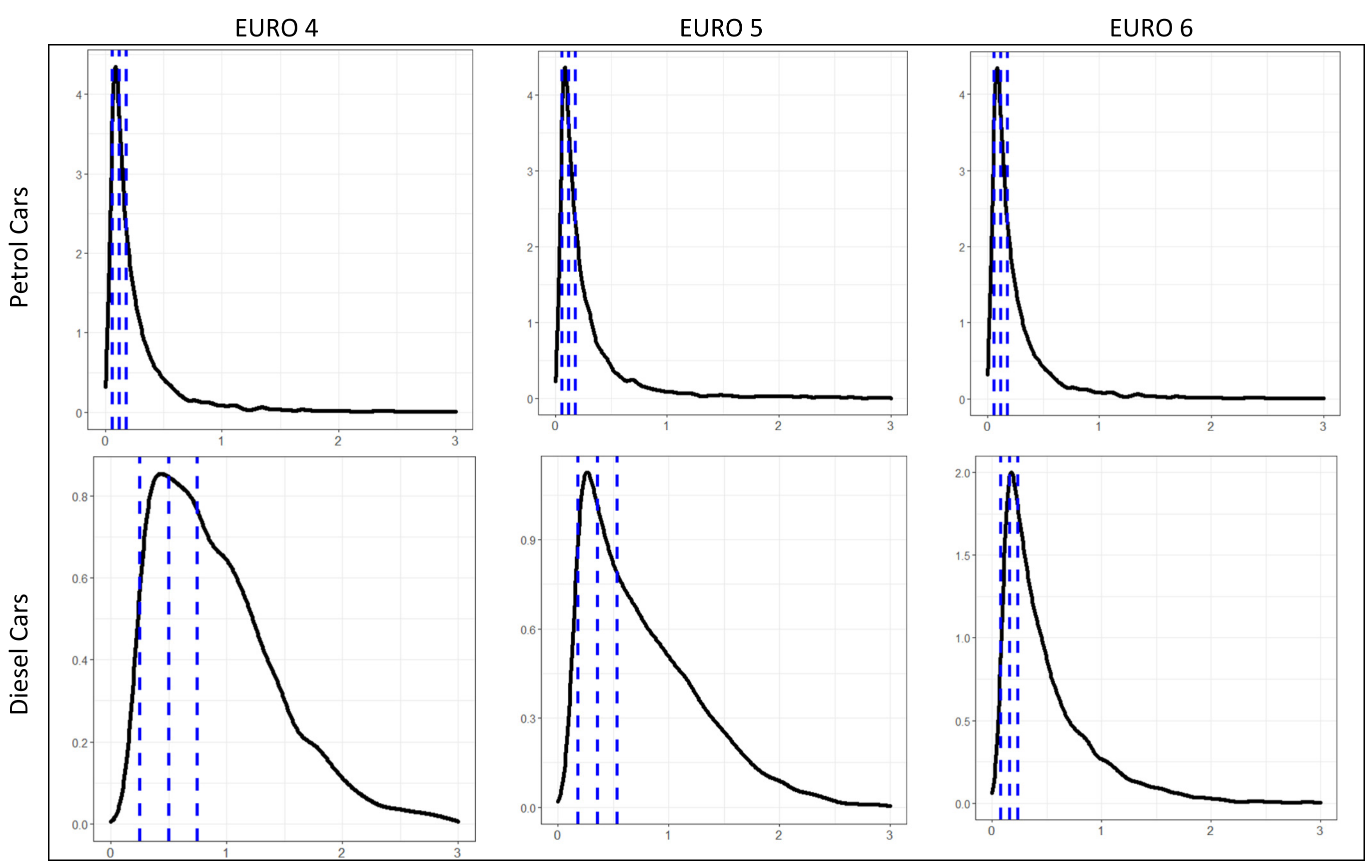


Fig. 2. The probability distribution function of the real-world NO_x emission factors measured by EDAR for petrol and diesel passenger cars. The blue dashed lines represent 1×, 2× and 3× the type-approved emission limits for the given EURO class and fuel type. The y and x-axis labels are density and emission factor, (g/km), respectively.

outside the ranges estimated based on the distribution of the larger sample set of emission measurements are indicative of high emitter behaviour. The closer the cumulative Pareto curve is to a second-order parabola the less likely you are to have high emitters. This is achieved in the ideal case for normally distributed data with a mean of zero. As high emitter vehicle subsets are added to the distribution the cumulative Pareto curve starts to plateau sooner, forming a 'knee' in the curve. Similarly, dividing a larger sample by cumulative Pareto value and comparing data distributions of vehicle information for vehicles associated with higher and lower cumulative Pareto values, respectively, provides an indication of the nature of high emitter behaviour. For example, for a cumulative Pareto sequence of NO emissions of diesel cars, the 80 % contributor point could be selected as a cut-point and the distribution of vehicles by the manufacturer could be compared above and below the 80 % cut-point. If high emitters were randomly distributed across the fleet, the two distributions would look similar. However, if the proportion of one manufacturer's vehicles is higher amongst the 80 % subset, then this is indicative of a make or model that had a greater tendency to be a high-emitter vehicle subset, which is named HEVS for the sake of brevity. This approach is referred Pareto 'enrichment' analysis because the methods are analogous to those used in enrichment studies, used, for example, by environmental scientists and geochemists. Pareto enrichment factor (PEF) is defined by the following equation:

$$\mathrm{PEF} = \frac{F_{n,veh\_subset}}{F_{n,veh\_set}} \tag{3}$$

where $F_{n,\ veh\_subset}$ and $F_{n,\ veh\_set}$ are the fractions of vehicles producing n% of the emissions in the investigated subset, and the larger sample that the subset was taken from, respectively. Here, enrichment factors >1 indicate a higher than average high emitter contribution. As discussed earlier in the paper, the screening criterion is a controversial challenge for identifying high emitters. In this study, a statistical population-based metric is applied which is arguably more accessible to study the underlying trends of high-emitters over different vehicle subsets.

3. Results & discussions

3.1. High emitter identifications by official limits

The EDAR-measured NO_x emission factors (EFs) are assessed against the published official EU limits (https://dieselnet.com/standards/eu/ld.php) to achieve a preliminary view of the high-emitter vehicles across the studied fleet. It should be noted that the approved limits for the heavier class of vehicles such as buses or HGVs are introduced in g/kWh, and vehicle weight plays a vital role in estimating the real-world emissions in distance-specific metric (g/km). For heavy-weight vehicles such as HGVs or buses, however, there is no reliable and certain weight data in almost all transport archives across the globe such as the ones used in this study. Hence, the comparisons are just provided for passenger cars.

Fig. 2 shows the distributions of EDAR-measured NO_x EFs for different passenger car subsets as well as corresponding type approval limits. Two- and three-times official limits are also plotted here as vertical dashed lines to better analyse the distribution of real-world NO_x EF of passenger cars. The contributions of different subsets due to the approved limits are reported in Table 2. A small contribution of passenger cars (<10 %) met the approved official limits. A similarly small contribution was observed in the study of (Qiu and Borken-Kleefeld, 2022), in which a remote sensing (RS) dataset for nine European cities was analysed.

However, along with the rationale aforementioned above, a very important technical fact should be taken into consideration. As was mentioned by (Ghaffarpasand et al., 2020a), there is an obvious difference between the VSP distribution of the test cycles such as the new European driving cycle (NEDC), which the official limits were introduced based on that, and the EDAR sample. Whilst EDAR measurements were on average collected under higher power conditions (frequency maxima about 10 kW/ton), NEDC tends to be conquered by lower power events with a VSP frequency that is highest very near to 0 kW/ton, please see (Ghaffarpasand et al., 2020a). This is not unique to the EDAR sample, whereby (Borken-Kleefeld et al., 2018) also reported quite similar conclusions for the much larger pan-European dataset of OPUS remote sensing devices. Hence, it is expected that real-world emission factors measured by VERSSs are meaningly higher than those measured through the test procedures. This fact re-emphasises the importance of achieving a certain methodology for the identification of high-emitter vehicles through the data collected by VERSSs. However, (Ghaffarpasand et al., 2020a) reweighted the EDAR data to the NEDC VSP distribution and then assessed it against the official EU limits though.

Table 2
The percentage contribution of the measured NO_x emission factors for passenger cars within different multiples of the type-approved emission limits (TAEL) for the given EURO class and fuel type 1 ×.

Vehicle type		<1 × TAEL	1–2 × TAEL	2–3 × TAEL	> 3 × TAEL
Petrol Cars	EURO 4	15	25	15	45
	EURO 5	7	29	17	48
	EURO 6	8	29	17	47
Diesel Cars	EURO 4	6	21	20	53
	EURO 5	3	13	15	69
	EURO 6	8	12	11	69

3.2. High emitter vehicle subsets (HEVSs)

Vehicle counts and percentages at the different sites are illustrated by general vehicle classification (car, taxi, bus, HGV, etc.) in Fig. 3. Cars

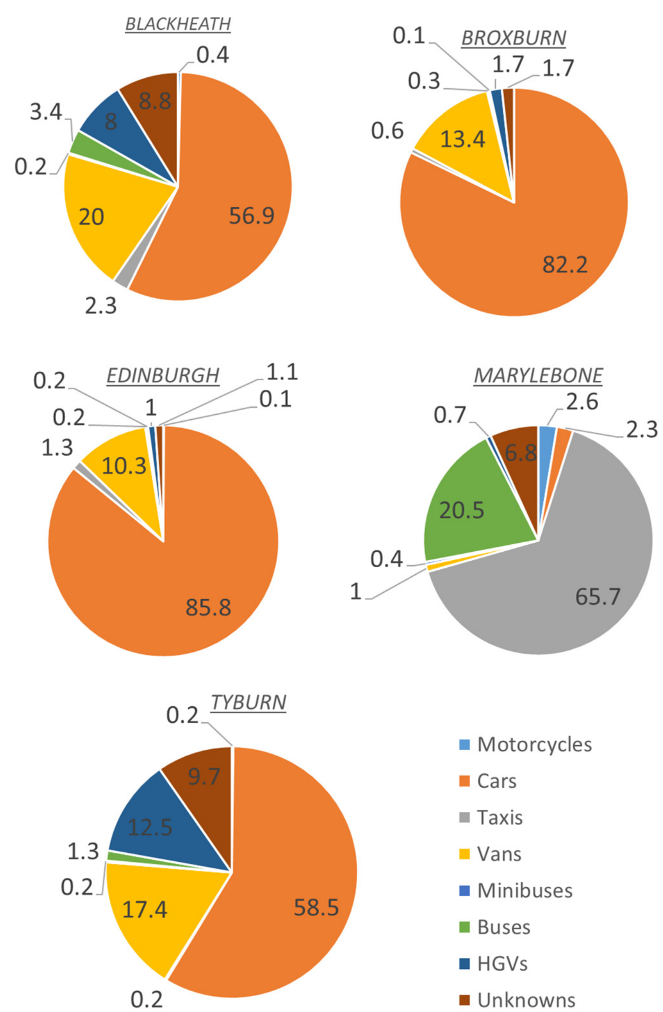


Fig. 3. The percentage of EDAR measurements at the different monitoring sites.

Table 3
80 % NO_x emission (g/km) contributions of the high emitting petrol and diesel cars at different sites studied in the recent EDAR campaigns. AT is the average temperature here.

site	AT (°C)	Petrol cars				Diesel cars			
		Cumulative Emissions (g/km)	Producing 80 % of Emissions	Total	Percentage of Vehicles (%)	Cumulative Emissions (g/km)	Producing 80 % of Emissions	Total	Percentage of Vehicles (%)
Blackheath	7 ± 3	770	664	2626	25	1754	1120	2096	53
Broxburn	12 ± 2	2668	2314	13,390	18	6487	5464	10,391	53
Edinburgh	8 ± 4	4542	5979	19,461	30	9118	8239	15,365	54
Marylebone	13 ± 4	92	37	88	42	138	77	149	52
Tyburn	7 ± 3	249	348	990	35	547	534	1000	53

were the most frequently observed vehicle class at all sites except Marylebone where the EDAR was measuring emissions from vehicles in a bus lane, where buses, taxis, and motorcycles are allowed.

As defined earlier, high-emitter vehicle subsets (HEVSs) are a small population of the fleet, which significantly contributes to the total fleet emission. As an example, the contribution of HEVSs which produce 80 % of the total NO_x emission of the total fleet at different sites is registered in Table 3. The most pronounced petrol car NO_x high-emitter contribution was seen at Broxburn where just 18 % of vehicles were responsible for 80 % of emissions. The observation most likely reflects a larger proportion of older petrol cars with EURO 3 or lower emission standards at this site (over 30 % at this site, compared with an average of 22 % at other sites). The least pronounced high emitter component was observed at Marylebone where 42 % of petrol cars were responsible for 80 % of petrol car NO_x emissions, however, here it should be reminded that the EDAR sampled vehicles in a bus lane at Marylebone and that, as a result, petrol car counts at this site were low. The contribution of high-emitting vehicle subsets which produce 80 % of the total NO_x emission of diesel cars was around 53 % for all sites. Table 3 also shows the average ambient temperature of measurements for different sites.

The impacts of ambient temperature on the real-world NO_x emission of passenger cars have been studied by previous investigators. Higher NO_x emissions have been observed for EURO 3 to EURO 5 diesel cars when ambient temperatures were below 15 °C (Borken-Kleefeld and Dallmann, 2018; Lakshminarayanan, 2022; Suarez-Bertoa and Astorga, 2018; Suarez-Bertoa et al., 2019). (Grange et al., 2019) observed the lowest NO_x emissions in diesel cars at the highest ambient temperatures encountered, up to 25 °C. For petrol cars, (Grange et al., 2019) found very little evidence of dependence NO_x emissions upon ambient temperature.

In this study, measurements at all five EDAR locations were conducted in the narrow temperature range of approximately 7-12 °C. For both petrol and diesel cars and no relationship between the average ambient temperature and the estimated high emitter percentage was found, see Table 3.

Table 4 summarizes the high-emitter trends NO and NO_x emissions for passenger cars. It can be seen that 23 % and 51 % of petrol and diesel cars contributed 80 % of the NO emissions produced by the petrol and diesel car segment of the EDAR sampled vehicle fleet, respectively. Meanwhile, near a quarter and a half of petrol and diesel cars are ranked as high emitter vehicles in terms of NO_x emissions, respectively. So, the diesel car fleet can be seen to exhibit a pronounced high emitter component.

The Pareto curve for NO and NO_x emissions of passenger cars is shown in Fig. 4 to better understand the contribution of high-emitter vehicle subsets to the NO and NO_x emission of the studied fleet. It shows that although older petrol cars are higher NO and NO_x emitters and their relatively small numbers create the petrol car fleet-level trends, within EURO classes it is the newer cleaner vehicles that exhibit the most pronounced high emitter trends. It can also be seen by the position of the steep gradient in the initial vehicles in the Pareto analysis that the trend within the all-petrol-car subset is driven by a relatively small number of older (pre-EURO and EURO1) petrol cars. The most rapid plateauing is for pre-EURO (EURO0) vehicles indicating that while there are not many of these vehicles on the roads, they are typically some of the dirtiest. The contributions of HEVSs within individual EURO classes are reported in Table 5. This suggests that with respect to NO and NO_x emissions, the petrol car fleet is getting cleaner but that the high emitter component is becoming a more important contributor to emissions. A different trend is observed in Fig. 4 and Table 5 for diesel

Table 4
NO and NO_x emissions (g/km) contributions of the highest emitting petrol and diesel cars.

Vehicle type	Cut point (%)	Cumulative emissions (g/km)	Vehicles	Percentage of vehicles (%)
Petrol Cars (NO)	20	1207	495	1
	40	2414	1511	4
	60	3621	3641	10
	80	4828	8385	23
	100	6035	36,555	100
Petrol Cars (NO_x)	20	2031	551	2
	40	4061	1704	5
	60	6092	4092	11
	80	8123	9285	25
	100	10,154	36,555	100
Diesel Cars (NO)	20	2414	881	7
	40	4828	3440	20
	60	7442	9006	31
	80	9656	14,684	51
	100	12,070	29,001	100
Diesel Cars (NO_x)	20	4511	2211	8
	40	9023	5393	19
	60	13,533	9554	33
	80	18,045	15,370	53
	100	22,556	29,001	100

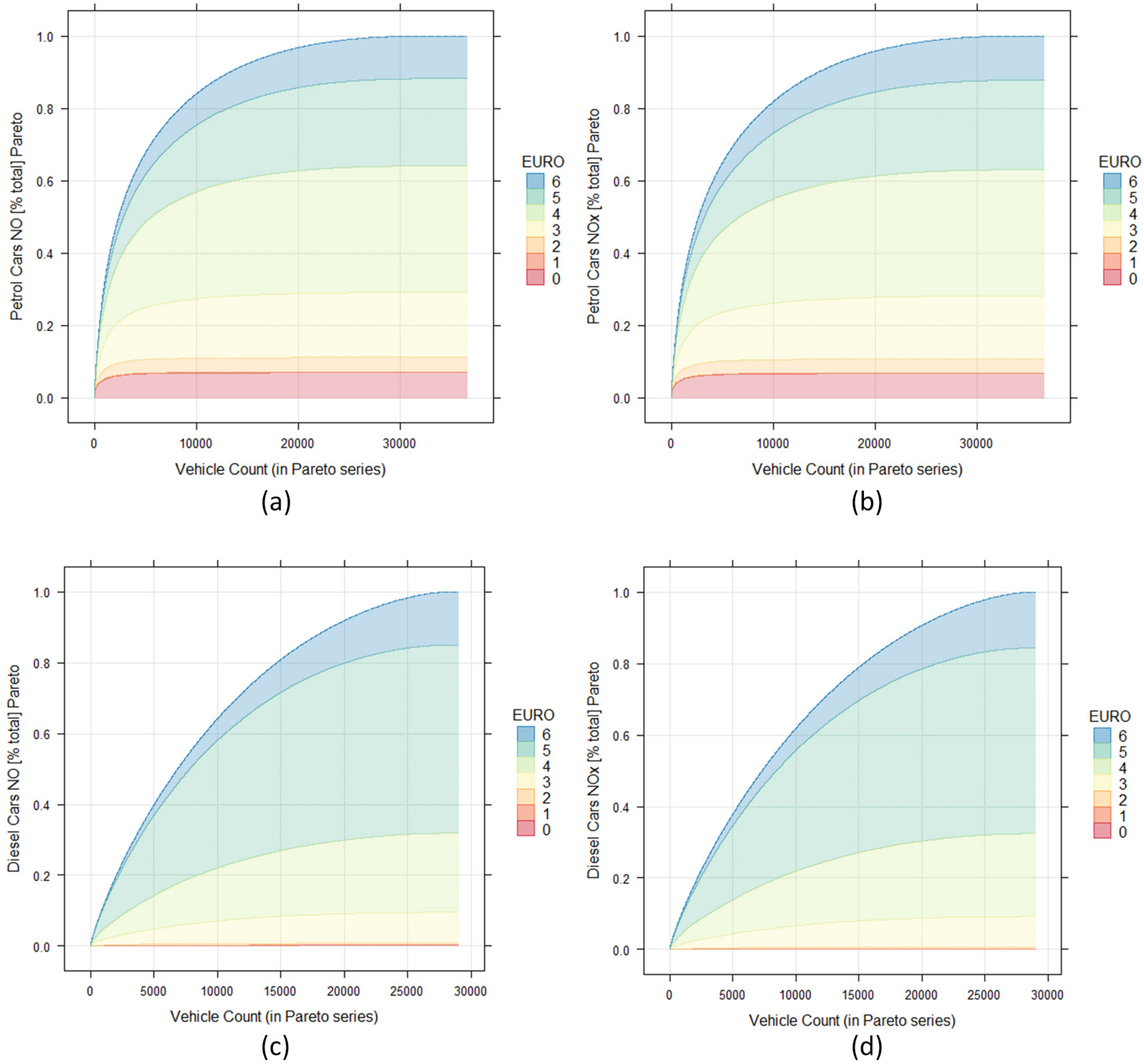


Fig. 4. The Pareto curve for (a) petrol cars NO emissions, (b) petrol cars NO_x emissions, (c) Diesel cars NO emissions and (d) diesel cars NO_x emissions, colour-coded to show relative contributions of different EURO classification vehicles.

cars. In particular, the EURO 5 diesel cars provide a much greater overall percentage of NO_x emissions within the diesel car subset compared to the petrol car subset. A similar analysis was undertaken for different vehicle types and pollutants, and the findings are summarised in Fig. 5.

The high emitter trends for different vehicle types were mostly similar. It seems that with perhaps the exception of taxis (PM and NO_2), the percentage of vehicles responsible for 80 % of emissions generally decreased with increasing EURO class indicating more pronounced high emitter contributions for newer subsections of the vehicle. The HEVS component was more pronounced for petrol cars by comparison to other vehicle types, and strongest for PM by comparison to other measured pollutant species. The less pronounced HEVS contribution and small variation in CO_2 reflect the fact that emissions were estimated here rather than measured.

It is important to note that although the assumption that 'a high measurement equals a high emitter' was widely accepted in the earliest remote sensing work, see for example (Stephens et al., 1997), this is not necessarily true. A single VERRS measurement captures the instantaneous emission at a certain point in the driving cycle and does not represent the average emission value of the vehicle across a journey. For example, a low emission from an otherwise dirty vehicle could generate a false-negative or a high emission from an otherwise clean vehicle would generate a false-positive. If regulators were looking to move to a policy where vehicle owners are penalised if their vehicles fail remote sensing tests, false-positives are likely to be the biggest legal challenge. The relative contributions from such false-negatives, false-positives, and the true-positives (the actual high-emitters) are currently unknown. Various strategies have been proposed to minimise the likelihood of such misassignment influencing follow-on action, e.g. only taking action on the basis of several consistent measurements to increase sampling confidence and excluding very high load, acceleration or VSP, etc., measurements to reduce false-positives, see e.g. (Pujadas et al., 2017). In recent years there has been a significant increase in efforts to better understand this issue, demonstrate the capability of VERSSs to detect faulty vehicles and so identify the emission signatures that would provide a credible basis for legal activities, see e.g. (Huang et al., 2018b). Such work is ongoing. So, for this study, we are studying the trends over a group of vehicles identified as high-emitter vehicle subsets (HEVS), see Section 2.5.

Table 5
80 % NO and NO_x emission (g/km) contributions of the highest emitting (petrol and diesel) cars in each EURO classification observed in the studied EDAR campaigns.

Vehicle type	EURO class	Cumulative emissions (g/km)	Vehicle		Percentage of Vehicles (%)
			Producing 80 % of Emissions	Total	
Petrol Cars (NO)	preEURO	334	156	466	33
	1	7	4	9	39
	2	196	122	354	34
	3	866	971	3602	26
	4	1689	3035	11,482	25
	5	1167	3237	11,830	25
	6	568	1729	6440	25
Petrol Cars (NO_x)	preEURO	542	161	466	35
	1	12	3	9	33
	2	316	125	354	35
	3	1408	1047	3602	29
	4	2836	3324	11,482	29
	5	2015	3562	11,830	30
	6	992	1903	6440	30
Diesel Cars (NO)	preEURO	23	25	44	57
	1	1	1	2	50
	2	53	54	97	55
	3	838	1094	1982	55
	4	2162	3152	6021	52
	5	5127	7443	13,714	54
	6	1450	3266	7141	46
Diesel Cars (NO_x)	preEURO	39	25	44	57
	1	2	1	2	50
	2	90	54	97	56
	3	1527	1140	1982	58
	4	4186	3358	6021	56
	5	9375	7709	13,714	56
	6	2825	3420	7141	48

Vehicle information extracted from the DVLA and SMMT archives was used here to extend the analysis from HEVS to vehicle make and model. The correct identification of vehicle makes and models that are more prone to being 'high emitter' would be a significant asset for policymakers. Therefore, it is perhaps worth first making a cautionary observation. All efforts to identify high-emitter groups require the ranking of high-emitter potential for different subsamples of a vehicle sample. Given the inherent variation in emission measurements, either atypically high or low outliers can bias a subsample especially if the subsample size is smaller. Counts of the order of about 100 (typically $n >$ 50–200) have previously been recommended as a lower threshold for such analysis. However, a higher threshold should be considered when accessing subsets within larger datasets in this fashion. Many of the

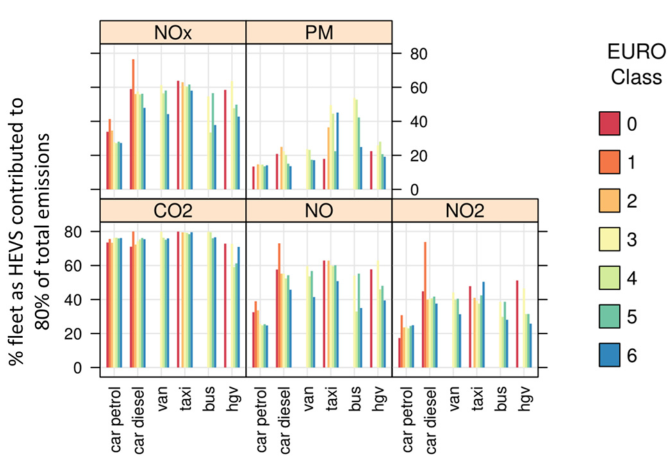


Fig. 5. High-emitter vehicle subset (HEVS) contributions of different vehicle types with various EURO classifications as observed during the EDAR campaigns (assuming 'the percentage of vehicles responsible for 80% of emissions' indicator).

less common vehicle makes or make and model combinations will have sample sizes at or below an $n = 100$ threshold and could be excluded from the analysis.

Fig. 6 illustrates this by comparing the Pareto enrichment factors (Eq. (3)) for the high emitter potential of different petrol car makes using sample size thresholds of $n = 20$ and $n = 200$. It should be noted that the enrichment factor was determined using the percentage of the fleet responsible for 80 % of the emissions benchmark. A value >1 indicates that a vehicle make was more likely to be seen in the high emitter population rather than the low emitter population. When the analysis is undertaken assuming a sample size threshold of n = 20 (Fig. 6(a)), four petrol car makes are seen that have high-emitter enrichment scores significantly larger than 1 (enrichment >1.5), namely MAKE0104, MAKE0068, MAKE0115 and MAKE0083. However, when the sample analysis is repeated with a larger sample size ($n = 200$; Fig. 6(b)) these makes are absent from the analysis. There are several makes (e.g. Land Rover and Mercedes) that often contribute highly to high emitter trends at the car/fuel/EURO group level but that are typically only present at about the $n = 50$ sample size and are therefore excluded for make or make/model level analysis. Likewise, there are several makes that are relatively common, e.g. Ford and Vauxhall, which are not always as pronounced high-emitters in larger samples but tend to score more highly in the more detailed analysis because they typically have large enough sample sizes to not be excluded, see Fig. 7 which presents the distribution of vehicle count by make attained during all EDAR campaigns in the UK available at the time of this analysis. Please note, however, this does not mean that Ford and Vauxhalls are always clean. Some of the less common Ford models appear to be moderately high emitters and the Vauxhalls at Tyburn looked particularly dirty. Rather than it is easy to overestimate the high emitter contributions of the most highly populated data subsets whenever you introduce subset size thresholds. Although often only moderately high emitters in parent samples, more commonplace makes or make/model/Euro class, etc., subsampled from them can easily be assigned as the high emitters in such analyses if their emission contributions are not carefully tracked and confirmed as significant as part of the larger fleet.

This tendency for the very dirtiest vehicle subgroups to be those with relatively small sample sizes could reflect measurement uncertainty, which was mentioned by many previous investigators such as (Chen et al., 2019, Martin Jerksjö et al., 2022, Qiu and Borken-Kleefeld, 2022). There is also a similar but arguably less pronounced tendency for the very cleanest subgroups to have relatively small sample sizes which would suggest that non-representative sampling could be a contributing factor. However, there is also the possibility that these observations reflect actual trends within the vehicle fleet. For example, the less common vehicle makes and models are likely to include special builds, the high-specification (sports, etc.) vehicle variations, and these may have been designed with higher performance to emissions trade-off. Similarly, the fact that the vehicles are less common make/model combinations may also mean that they may not be as comprehensively optimised during development or tested as extensively during regulatory approvals.

Finally, despite the relatively small sample size of the HGV subgroup of the EDAR dataset, it is worth highlighting one notable feature of the HGV PM emission distribution. Again, strictly we lack the sample sizes for unambiguous individual vehicle and make/model subgroup analysis, but Pareto enrichment trends were highly pronounced for such a small sample. The plots of Pareto and Pareto enrichment for by-make heavy-duty vehicles PM emissions are pictured in Fig. 7. It seems that the HGV PM emissions exhibited perhaps the most pronounced individual vehicle behaviour by make. Future work needs to get greater HGV sample sizes to further test this observation (Fig. 8).

4. Conclusions

This study is conducted to investigate and analyse the high emitter population and emission trends in the UK vehicles fleet. Conventional

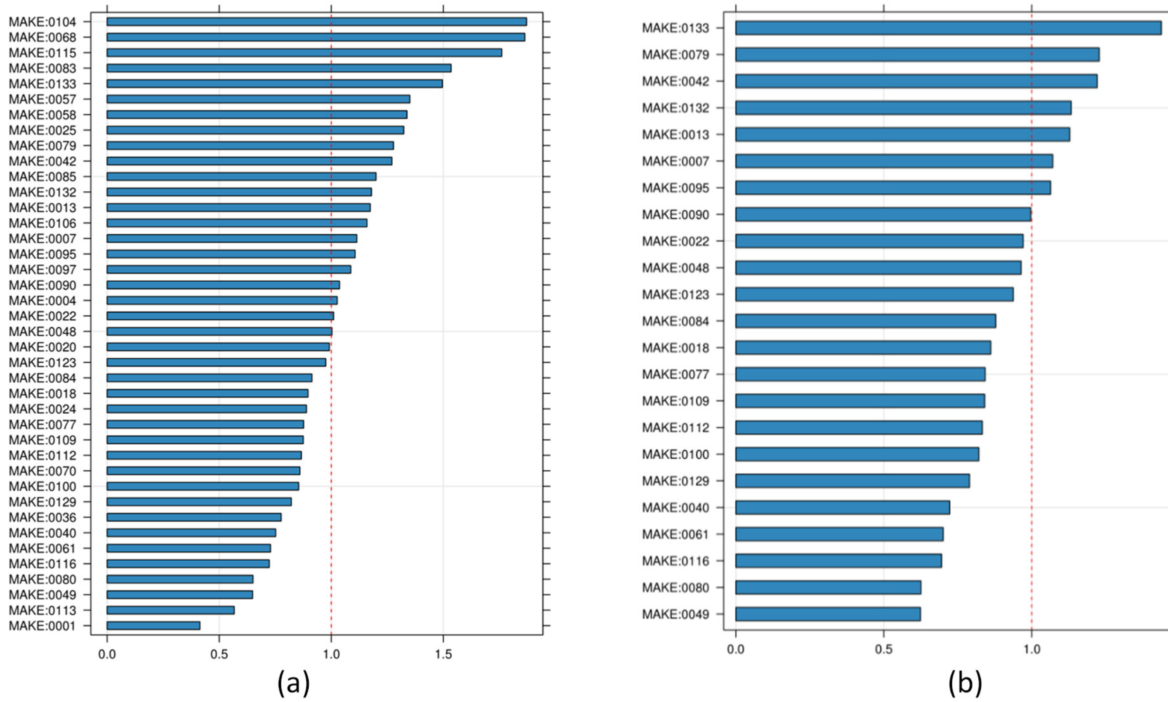


Fig. 6. Pareto enrichment plots of the NO high emitter potential of petrol cars of different (anonymised) vehicle makes, applying sample size thresholds of (a) $n = 20$ and (b) $n = 200$. The Pareto enrichment factor is defined in Eq. (3).

cumulative Pareto analysis was used in combination with enrichment factor methods, more commonly applied in geochemical studies, to target high emitter vehicle subsets (HEVS) from the large dataset including >94,000 real-world emission measurements of different vehicles. The remote sensing field campaigns had been established by deploying the EDAR system within the five UK urban environments for the years 2016–2017. The Pareto analysis was applied to characterize and analyse the proportion of vehicles responsible for 80 % of emissions originated by the fleet, i.e. HEVS. Pareto analysis indicates that there are vehicles that could be high emitter vehicles within the vehicle fleet, but there is still potential uncertainty regarding the overlap between the false-positive of a good vehicle's occasional high emissions and the true-positive of an unambiguously bad vehicle's emissions given the current size of EDAR dataset. Results show a similar trend for different vehicle types, whereby the contribution of HEVS reduced with increasing EURO class. Moreover, the strongest evidence for high-emitter-like behaviour is arguably seen for HGVs and PM. However, there are high uncertainties and multiple confounders associated with the measurement

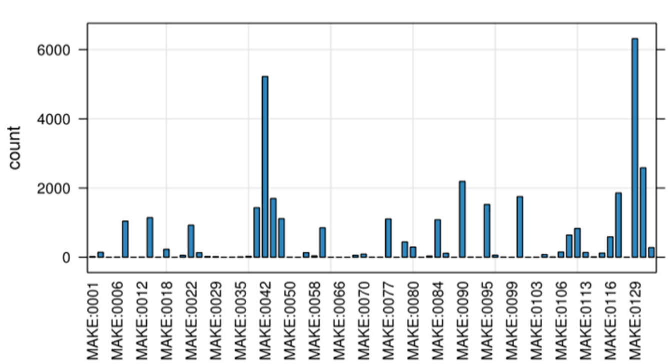


Fig. 7. Vehicle count by make for EDAR petrol cars observed during recent EDAR campaigns. MAKE0042 and MAKE0129 are Ford and Vauxhall.

of vehicle types seen in smaller numbers (e.g. HDVs and motorcycles) and species like PM which are more challenging using open path techniques. Therefore, there is a need for further work to generate more data to reduce uncertainties.

The HEVS identification approach proposed here might be useful in the short-term to help identify the best LEZ management strategies, and in the longer term to build the case for a threshold-based approach for individual vehicle surveillance and inspection and maintenance schemes for ageing vehicle fleets.

CRediT authorship contribution statement

Omid Ghaffarpasand: Formal analysis, Methodology, Writing – original draft. **Karl Ropkins:** Formal analysis, Conceptualization, Methodology, Visualization, Investigation, Writing – review & editing. **David C.S. Beddows:** Formal analysis, Software, Data curation, Writing – review & editing. **Francis D. Pope:** Formal analysis, Conceptualization, Methodology, Supervision, Writing – review & editing.

Data availability

Data will be made available on request.

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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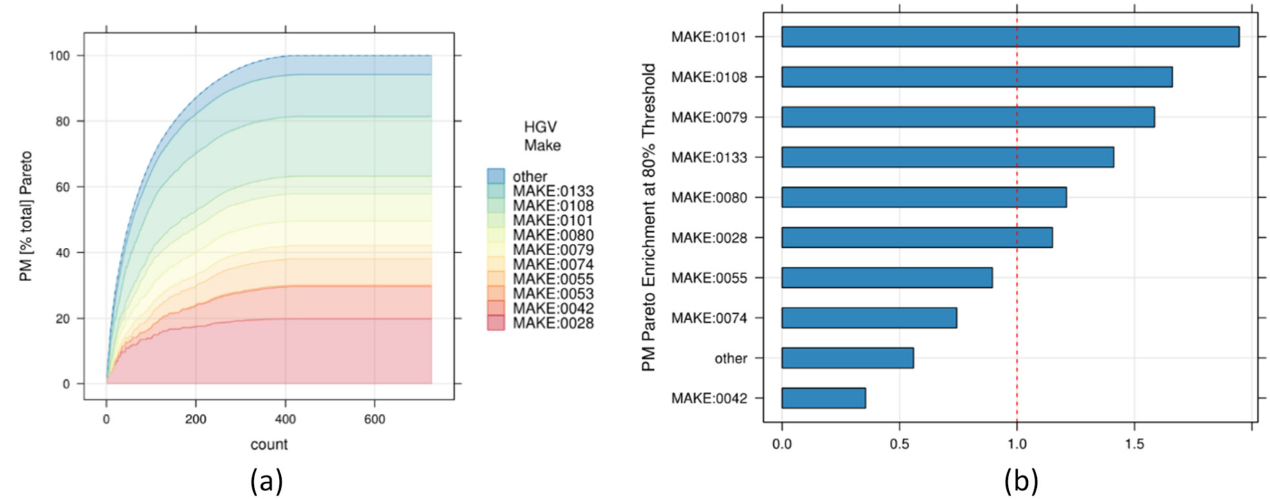

Fig. 8. (a) Pareto plot, and (b) Pareto enrichment plot for different makes of HGV PM emissions.

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