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Gasoline and diesel passenger car emissions deterioration using on-road emission measurements and measured mileage

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ABSTRACT

Modern gasoline and diesel vehicles are equipped with highly effective emission control systems that result in low emissions of pollutants such as nitrogen oxides (NO_x) when new. However, with increasing age or mileage, the emissions performance of vehicles can deteriorate over time, leading to increased emissions. In this work we use comprehensive vehicle emission remote sensing measurements collected over a wide range of conditions, together with individual vehicle measured mileage to quantify vehicle emissions deterioration. A quantile regression modelling approach is used to provide a more complete understanding of the distribution of deterioration effects that is not captured by considering mean changes over time. The approach accounts for factors such as driving conditions and ambient temperature, as well as determining whether deterioration affects whole populations of vehicles or a smaller subset of them. Accounting for these factors, we find that for most pollutants the rate of deterioration of emissions from pre-Euro 5 gasoline passenger cars is highly skewed. Between 5% and 10% of pre-Euro 5 gasoline passenger cars have emissions similar to a Euro 5 diesel car, suggesting that policies should be developed to accelerate their removal from the fleet. Furthermore, we find evidence that there are differences between vehicle manufacturers in the way emissions of NO_x deteriorate.

1. Introduction

Worldwide, progressively more stringent vehicle emissions legislation has been developed to reduce the emissions of many important air pollutants from road vehicles. These developments have resulted in increasingly more sophisticated technologies being used to reduce emissions. The introduction and refinement of technologies such as the three way catalyst, particle filters and selective catalytic reduction (SCR) systems have led to considerable reductions in emission species such as nitrogen oxides (NO_x) and particulate matter (Wang et al., 2014; Praveena and Martin, 2018; Miller and Jin, 2019). While much of the focus of vehicle emission measurements is on newer vehicles with improved emission control technology, it is imperative to quantify the change in emissions from vehicles over their full lifetime, which can exceed 20 years. As a vehicle is driven, changes in emission behaviour can occur due to wear of engines, deterioration of emissions control systems and after-treatment technologies such as catalysts and particle filters. Moreover, with increasingly complex and sophisticated after-treatment technologies being adopted, it is important to ensure their effective performance throughout the lifetime of the vehicle.

In Europe, the legislation for the most recently regulated vehicles (Euro 6) specifies “Manufacturers’ obligations”, among which include an obligation that any technologies which limit tailpipe and evaporative emissions are effective “throughout the normal life of the vehicles under normal conditions of use” (Council of European Union, 2014; Williams and Minjares, 2016). It is stipulated that, *a)*, in-service conformity testing eligibility continues until a vehicle is either 5 years old or has driven 100,000 km, and *b)*, manufacturers must conduct pollution control system durability tests to over 160,000 km of driving. Recently, the Consortium for ultra Low Vehicle Emissions (CLOVE) has suggested bringing future Euro 7 legislation in-line with US Tier 3, which defines a “normal life” for a vehicle as either 15 years or 150,000 miles (roughly 240,000 km) (United States Environmental Protection Agency, 2014; ICCT5, 2021).

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Emission factors and inventories recognise that emissions deterioration occurs and attempt to provide pragmatic approaches to account for such deterioration. In Europe, the joint European Monitoring and Evaluation Programme (EMEP)/European Environment Agency (EEA) air pollutant emission inventory guidebook 2019 (Ntziachristos and Samaras, 2019) details the use of correction factors to account for emission deterioration due to vehicle age. Importantly, deterioration factors are only applied to gasoline vehicles and it is assumed that the carbon monoxide (CO), nitrogen oxides (NO_x) and hydrocarbon emissions of Euro 3 and later gasoline cars and light commercial vehicles stop deteriorating at 160,000 km. The Handbook Emission Factors for Road Transport (HBEFA) version 4.1 updated its deterioration factors for CO, NO_x and hydrocarbon emissions (Matzer et al., 2019) based on vehicle emission remote sensing data from the large European CONOX database (Sjödin et al., 2018). The updated deterioration functions cover Euro 1 through to projected Euro 7 diesel and gasoline light duty vehicles within a mileage range of 0–200,000 km. They report that NO_x emissions are less than 1.5 times higher for all Euro standards of diesel vehicles at 200,000 km compared to 50,000 km. Gasoline vehicles, however, are found to be over 3 times as high for Euro 3 and 2.5 times as high for Euro 5, though the ratio for Euro 6 vehicles is only around 1.25.

Emission deterioration functions are typically based on a limited number of chassis dynamometer tests in a limited range of mileages (Keller et al., 2017; Ntziachristos and Samaras, 2019). An unavoidable aspect of in-lab and even on-board methods (e.g., Portable Emission Measurement Systems (PEMS)) is a small sample size owing to the time and cost requirements to measure a vehicle. Furthermore, these methods are typically used to measure newer vehicles, so there are sparse data for older or higher mileage vehicle emissions. Using these methods to get a broad sample of vehicles of different model years, meeting different Euro standards, and from different manufacturers, would be prohibitively expensive and time consuming.

Vehicle emission remote sensing has the potential to overcome some of these issues. The non-selective, real-world nature of remote sensing ensures that, with a sufficiently large sample size, the full spectrum of age, mileage and emission deterioration of a fleet will be captured. Furthermore, with the large data-sets obtained using remote sensing, multivariate statistical analysis can be conducted to isolate the effect of deterioration from other influences such as driving characteristics (e.g., instantaneous engine power) or ambient conditions. Indeed, much of the literature on emission deterioration focuses on the use of vehicle emission remote sensing (Borken-Kleefeld and Chen, 2015; Bishop et al., 2016; Chen and Borken-Kleefeld, 2016; Bishop and Stedman, 2008; Zhan et al., 2020; Sjödin and Andréasson, 2000), although a smaller number of studies using other methods such as PEMS (Huo et al., 2012) and chassis dynamometers (Chiang et al., 2008; Zhang et al., 2017, 2018) do feature. An important limitation of many of these remote sensing studies is that individual vehicle mileage is not available, leading to vehicle age being a frequent proxy. For example, in Borken-Kleefeld and Chen (2015) and Chen and Borken-Kleefeld (2016) the difference between the year of measurement and the year of first registration is taken to be a vehicle's age, which is then used to estimate mileage using statistics from the Swiss government.

Deterioration factors as used in emission factor development generally provide fleet-average linear relationships to correct an emission from a vehicle when assumed to be new. However, these factors do not capture potentially important information on the nature of deterioration, such as whether all vehicles tend to deteriorate similarly over time or whether the changes are dominated by significant deterioration from relatively few vehicles. These considerations are important from a policy perspective because different responses might be required depending on the nature of emissions deterioration. For example, it is arguably more efficient and cost effective to identify and fix (or remove) a small population of high emitters than it is to deal with a large population of vehicles that deteriorate by a more modest amount. To understand these issues, there is a need to consider large populations of vehicles and to establish the full distribution of effects rather than a mean response.

In this study, comprehensive vehicle emission remote sensing data is paired with measured vehicle mileage from individual vehicles using data from annual passenger car technical inspections. These paired data are used to study the deterioration of emissions from passenger cars, as well as consider the appropriateness of vehicle age as a mileage proxy. This study uses measured mileage data from 197,000 gasoline and diesel passenger cars. The nature of any deterioration effects on emissions is complex and is not fully described by simple relationships that relate mileage and emissions. Therefore, this study adopts a quantile regression approach to consider the entire conditional distribution of effects. This approach allows for the control of other factors such as vehicle driving conditions and ambient temperature which also affect measured emissions. Finally, with large sample sizes available, we consider manufacturer effects on how emissions deteriorate with mileage.

2. Materials and methods

2.1. Vehicle emission remote sensing

The principles of vehicle emission remote sensing have been described in extensive detail elsewhere (Bishop and Stedman, 1996; Burgard et al., 2006), so only a short summary is provided here. Remote sensing is a non-obtrusive, curbside method for measuring real-world vehicle emissions. A remote sensing device is typically deployed to be as unobstructive as possible, ensuring vehicles can drive through the set-up unimpeded. As a vehicle drives through, each individual module of the remote sensing device simultaneously activates. These are an ultraviolet/infrared (UV/IR) source and detector to measure exhaust emissions, optical speed-acceleration bars to capture instantaneous driving conditions, a camera to photograph number plates, and sensors to record ambient conditions such as temperature, pressure and relative humidity. As the triggering of all these modules is achieved in just a fraction of a second, remote sensing observations are often referred to as 'snapshots' of a vehicle's journey.

Spectrometry is achieved with a collinear beam of IR and UV light. Carbon monoxide (CO), carbon dioxide (CO_2), hydrocarbons and a background reference are measured using the IR component, and ammonia (NH_3), nitrogen oxide (NO) and nitrogen dioxide (NO_2) by the UV component. 100 measurements are taken of each plume in just half a second. Pollutant concentrations are typically given as a ratio to CO_2, which is assumed to remain constant as the plume disperses. These ratios can be used to calculate fuel-specific (g kg^{-1}) emissions (Burgard et al., 2006).

The photographed vehicle number plates can be cross-referenced with vehicle technical databases to obtain key information about the measured vehicles, such as fuel type, emissions standards and manufacturers. In this study, technical information was sourced from the Driver and Vehicle Licensing Agency and the Society of Motor Manufacturers and Traders Motor Vehicle Registration Information System. These data were obtained from the commercial supplier CDL Vehicle Information Services Ltd. (Cheshire Datasystems Limited, 2018).

An important aspect of the current work is the use of measured mileage information. While vehicle age is readily available, it is not an ideal metric for emissions deterioration. Data relating to the total mileage of each vehicle at its last annual "MOT" test was obtained through CDL for vehicles greater than three years old. Vehicles younger than three years do not require an annual technical inspection in the UK. As a result, the proportion of mileage information available for Euro 6 vehicles (introduced in 2016) is lower than that for older Euro standards (24% of Euro 6 observations have associated mileage information, compared to 63–76% for Euro 3–5). The date at which the mileage information is available and the emissions measurement date could be up to 12 months different, i.e. the measured mileages available would tend to underestimate the actual mileage at the time the emissions

Table 1
A statistical summary of the vehicle emission remote sensing data, split into diesel and gasoline passenger cars. Statistics provided are only for measurements with an associated mileage value. Statistics presented: [1]Mean (Standard deviation); [2]Number of measurements (Percentage of the column total). Generated using the *gtsummary* R package (Iannone et al., 2021; Sjoberg et al., 2021).

Characteristic	Euro 3	Euro 4	Euro 5	Euro 6
Gasoline Passenger Cars				
# Measurements	22,952	36,327	38,855	10,776
# Manufacturer Groups	23	23	20	20
(with ≥100 Measurements)	18	18	15	13
Vehicle & Ambient Characteristics [1]				
VSP (kW t^{-1})	7.61 ± 7.65	7.80 ± 7.07	8.12 ± 6.88	8.96 ± 5.08
Speed (km h^{-1})	36.4 ± 9.2	36.6 ± 9.3	37.0 ± 9.5	38.4 ± 9.4
Acceleration (km h^{-1} s^{-1})	0.96 ± 2.33	0.99 ± 2.19	1.02 ± 2.15	1.07 ± 1.66
Ambient Temp. (K)	287.9 ± 5.4	288.1 ± 5.2	288.2 ± 5.2	288.2 ± 4.8
Cumulative Mileage (10^4 km)	15.7 ± 6.9	12.2 ± 5.6	7.1 ± 3.9	4.6 ± 2.5
Vehicle Age (years)	14.4 ± 1.6	10.4 ± 1.8	6.0 ± 1.9	4.1 ± 1.0
Remote Sensing Device [2]				
OPUS RSD 5000	20,387 (89%)	33,236 (91%)	36,181 (93%)	10,501 (97%)
Denver FEAT	2565 (11%)	3091 (8.5%)	2674 (6.9%)	275 (2.6%)
Diesel Passenger Cars				
# Measurements	9143	24,030	44,442	10,475
# Manufacturer Groups	22	22	21	18
(with ≥100 Measurements)	14	17	16	12
Vehicle & Ambient Characteristics [1]				
VSP (kW t^{-1})	8.26 ± 8.38	8.09 ± 7.70	8.23 ± 7.56	8.72 ± 6.14
Speed (km h^{-1})	36.3 ± 9.4	36.5 ± 9.8	36.6 ± 9.8	37.1 ± 9.8
Acceleration (km h^{-1} s^{-1})	1.06 ± 2.38	1.07 ± 2.26	1.14 ± 2.27	1.19 ± 1.79
Ambient Temp. (K)	288.2 ± 5.5	288.1 ± 5.4	287.9 ± 5.2	287.8 ± 4.9
Cumulative Mileage (10^4 km)	24 ± 13	19 ± 10	11 ± 7	7 ± 5
Vehicle Age (years)	14.0 ± 1.5	10.2 ± 1.8	5.7 ± 1.8	3.9 ± 1.2
Remote Sensing Device [2]				
OPUS RSD 5000	8223 (90%)	21,894 (91%)	40,692 (92%)	10,146 (97%)
Denver FEAT	920 (10%)	2136 (8.9%)	3750 (8.4%)	329 (3.1%)

measurements were made.

Vehicle emission measurements were conducted between 2017 and 2020 at 39 sites across 14 regions in the United Kingdom mainly using the Opus AccuScan RSD 5000 (OPUS5, 2018), augmented with a relatively small number of measurements using the Denver FEAT instrument (University of Denver, 2011). Previous literature has shown good agreement between the two remote sensing devices, so the combination of these data sets is appropriate (Rushton et al., 2018). Of interest to this study are measurements of diesel passenger cars or gasoline passenger cars. 197,000 of these measurements include mileage data from annual technical inspection "MOT" tests (Carslaw et al., 2019) and are therefore relevant to this study. Note that these are not 197,000 unique vehicles; available number plate data shows that around 13% of vehicles in the data set were measured twice, 4% three times, and 3% four or more times.

A statistical summary of these measurements is provided in Table 1. The speed ranges reflect that remote sensing is typically conducted in urban conditions, but this is not likely to be of detriment to the objectives of this study; it is unlikely that deterioration patterns would meaningfully differ under rural, motorway or any other driving conditions. Measurements being taken in urban areas may mean that a small proportion include cold start emissions, though this is unlikely as exhaust after-treatment technologies tend to reach effective operating temperatures in a few minutes (Han et al., 2021). The majority of emission measurements can therefore be assumed to be of hot, stabilised emissions.

2.2. Statistical methods

While ordinary least squares (OLS) linear regression may provide some insight into emission deterioration, it is intuitively likely that different vehicles deteriorate at different rates. An important question to address is whether it is the case that there is a general deterioration in the emissions performance of all vehicles or whether there is a smaller population of much higher emitters that have a disproportionate effect. For this reason, a quantile regression-based approach was used, which can account for the full distribution of responses and not just the mean response that is considered by OLS.

Linear quantile regression can be understood in analogue to linear OLS regression; while an OLS regression line minimizes the sum of the squared differences between it and the data, a quantile regression line ensures that some proportion of the data is below and above it. For example, the quantile regression line for the median ($\tau = 0.50$) ensures that half of the data is above it and half below. For the 75th percentile ($\tau = 0.75$), 75% of the data would be found below the line and 25% would be above. A more thorough description of quantile regression can be found in the Supplementary Information (Section S1).

In this study, quantile regression — using the *quantreg* R package (Koenker, 2021) – is used to explore the relationship, *a)*, between vehicle age, *AGE*, and cumulative vehicle mileage, *MIL* and, *b)*, between cumulative vehicle mileage and fuel-specific (g kg^{-1}) emissions. In the former case, a second-order polynomial was used. This is given in Equation (1), where $\hat{\mu}(\tau|AGE)$ represents the predicted quantile of vehicle mileage.

$$\hat{\mu}(\tau|AGE) = \hat{\beta}_0 + \hat{\beta}_1(\tau)\cdot AGE + \hat{\beta}_2(\tau)\cdot AGE^2 \quad (1)$$

When examining emissions, four air pollutants are initially explored: nitrogen oxides (NO_x), carbon monoxide (CO), ammonia (NH_3) and particulate matter (PM, measured by the OPUS remote sensing device using percentage UV opacity). Note that NH_3 is only pertinent to the gasoline vehicles and Euro 6 diesel vehicles, the latter of which have after-treatment systems that use SCR systems which can result in emissions of NH_3. When fitting models, multivariate analysis is used to predict fuel-specific emissions using vehicle mileage and additional covariates with known influences on vehicle emissions. The key covariate is vehicle specific power, *VSP* (Equation (2)), though for diesel NO_x emissions ambient temperature, *AT*, is also included (Equation (3), Grange et al. (2019)). B-splines, denoted using *f*, are used to smooth these covariates, both set to 3 degrees of freedom.

An interaction effect with some vehicle category, *VC*, is also included. The first category considered is Euro standard, to gain an understanding for the potential differences in deterioration as emission control technology has changed with legislation. The second category considered is vehicle manufacturer group, used to examine the differences in deterioration between the varying technologies employed by different vehicle manufacturers. In this second case, models are fit separately for each of Euro 3, 4 and 5; Euro 6 is excluded due to limited data and range of mileage.

$$\widehat{\mu}(\tau|MIL, VSP, VC) = \widehat{\beta}_{0,VC}(\tau) + \widehat{\beta}_{MIL,VC}(\tau)\cdot MIL + f_{VSP,VC}(\tau, VSP) \quad (2)$$

$$\widehat{\mu}(\tau|MIL, VSP, AT, VC) = \widehat{\beta}_{0,VC}(\tau) + \widehat{\beta}_{MIL,VC}(\tau)\cdot MIL + f_{VSP,VC}(\tau, VSP) + f_{AT,VC}(\tau, AT) \quad (3)$$

Data processing was carried out using the R programming language (R Core Team, 2021), with uncertainties calculated and term significance estimated using bootstrap resampling. The *tidymodels* (Kuhn and Wickham, 2020; Silge et al., 2021) collection of R packages was used for the bootstrapping simulations. The *bootstrap* function was used with 100 resamples with the sampling stratified (using the *strata* argument) by the relevant interaction term (Euro standard or manufacturer group) and otherwise default parameters. The stratification by the vehicle category ensures proportionally smaller categories (i.e. Euro 3 or 6 vehicles, or more niche manufacturers) are well represented in the bootstrap samples.

The "percentile" method was used to calculate 95% confidence intervals around the coefficients. If the 95% confidence interval of the bootstrapped mileage coefficients includes 0, the p-value is taken to be less than 0.05 and the term (and therefore the vehicle category's rate of deterioration) is judged to be insignificant. Furthermore, if the 95% confidence intervals of the *difference* between any given terms includes 0, it is taken that they are not significantly different from one another.

3. Results

3.1. Mileage and age characteristics for light duty vehicles

In this section the potential drawbacks of using vehicle age as a measure of emissions deterioration are considered, highlighting the benefits of this study's use of measured mileage. Second-order polynomial quantile regression fits for measured cumulative vehicle mileage as a function of vehicle age from the remote sensing data are shown in Fig. 1. Diesel London taxis ("black cabs", n = 556, 0.5% of the measured diesel passenger car fleet) have unique technical data relating to their body type so can be treated separately to the rest of the diesel passenger cars.

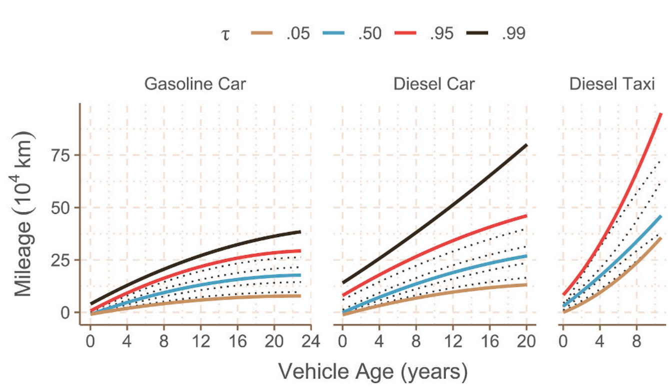

Fig. 1. Second-order polynomial quantile regression fits for cumulative vehicle mileage as a function of vehicle age at the date of their MOT (Equation (1)), where $\tau \in \{05, .10, .30, .50, .70, .90, .95, .99\}$. $\tau \in \{05, .50, .95, .99\}$ are solid and labelled; the dotted lines represent the unlabelled quantiles. $\tau = 0.99$ is not shown for the taxis due to the limited number of observations; the 99th percentile would represent just 5 vehicles.

There is a clear distinction between the three vehicle types. Gasoline passenger cars are among the oldest of the three categories, but have the lowest maximum mileage. The maximum age decreases and maximum mileage increases progressing through the gasoline cars, the diesel vehicles, and finally the taxis.

The quantile regression fits shown in Fig. 1 highlight a weakness of using simple linear regression to derive mileage from vehicle age, in that it would not fully represent the underlying and significant distribution of cumulative vehicle mileages of vehicles of the same age. For example, the average age of a vehicle in the vehicle emission remote sensing data set is around 8 years. Using a second-order polynomial ordinary least squares (OLS) model, 8 year-old gasoline passenger cars have driven 98,000 km. This is similar to the median vehicle according to quantile regression, which has driven 94,300 km (−3.8% of the OLS). However, the lowest mileage 5% of these vehicles have only driven 41,700 km (−57.4% of the OLS), whereas the 5% highest mileage have driven 163,000 km (+66.3% of the OLS). The top 1% have driven 206,000 km (+110%). Similarly, an ordinary least squares approach indicates an 8 year-old diesel non-taxi passenger car has driven 148,000 km. The *median* vehicle from quantile regression has driven 136,000 km (−8.1% of the OLS), the bottom 5% have driven 68,100 km (−54.0%), the top 5% 266,000 km (+79.7%), and the top 1% 383,000 km (+157%).

While it could be argued that the "average" vehicle modelled using ordinary least squares is similar to the median vehicle modelled using quantile regression, relying on OLS regression ignores a significant distribution of vehicle mileage at any given vehicle age. A straightforward assumption that mileage increases linearly with age is useful in the absence of measured mileage data, but is not optimal in representing the inherent distributions of mileages that exist. This is particularly relevant if considering taxi fleets or other commercial vehicle fleets that may likely have a distinct mileage-age relationship, as clearly demonstrated in Fig. 1. Moreover, if high mileage vehicles have emissions that are significantly different from average-age vehicles, then the estimated emissions response will also be erroneous.

3.2. Exploratory analysis of emission deterioration

We first explore the relationship between vehicle mileage and emissions without taking account of the influence of other factors such as Euro standard, ambient temperature and engine power demand. The distribution of vehicle NO_x and PM emissions at different cumulative mileages is given in Fig. 2. The equivalent figure for CO and NH_3 is given in Fig. S2.

These distributions highlight the benefit of a quantile regression-based approach. The median intensity of emissions from the species typically increase at higher mileage deciles to various extents, which suggests the presence of a mileage-based deterioration effect. More relevant for quantile regression, it also appears that for some species-fuel type combinations the spread of emission values (seen in both ranges between the 25th and 75th, and the 5th and 95th percentiles) appears to increase at higher deciles of mileage. This suggests that the *rate* of increase in emissions is itself greater at higher emission quantiles, indicating that an ordinary least squares linear regression may not fully represent the true range of responses different emitters have to increasing cumulative mileage.

From this exploratory analysis, it is clear that the species behave distinctively from one another with respect to mileage deterioration. The four species, NO_x, PM, CO and NH_3, will now be discussed in turn.

NO_x emissions show clear differences between the two vehicle types. The five visualised quantiles of fuel-specific NO_x in diesel vehicles all appear to show a gentle increase from the lowest to highest mileage deciles across all Euro standards, with the exception of the 95th percentile of Euro 6. The large interquartile ranges seen in the diesel

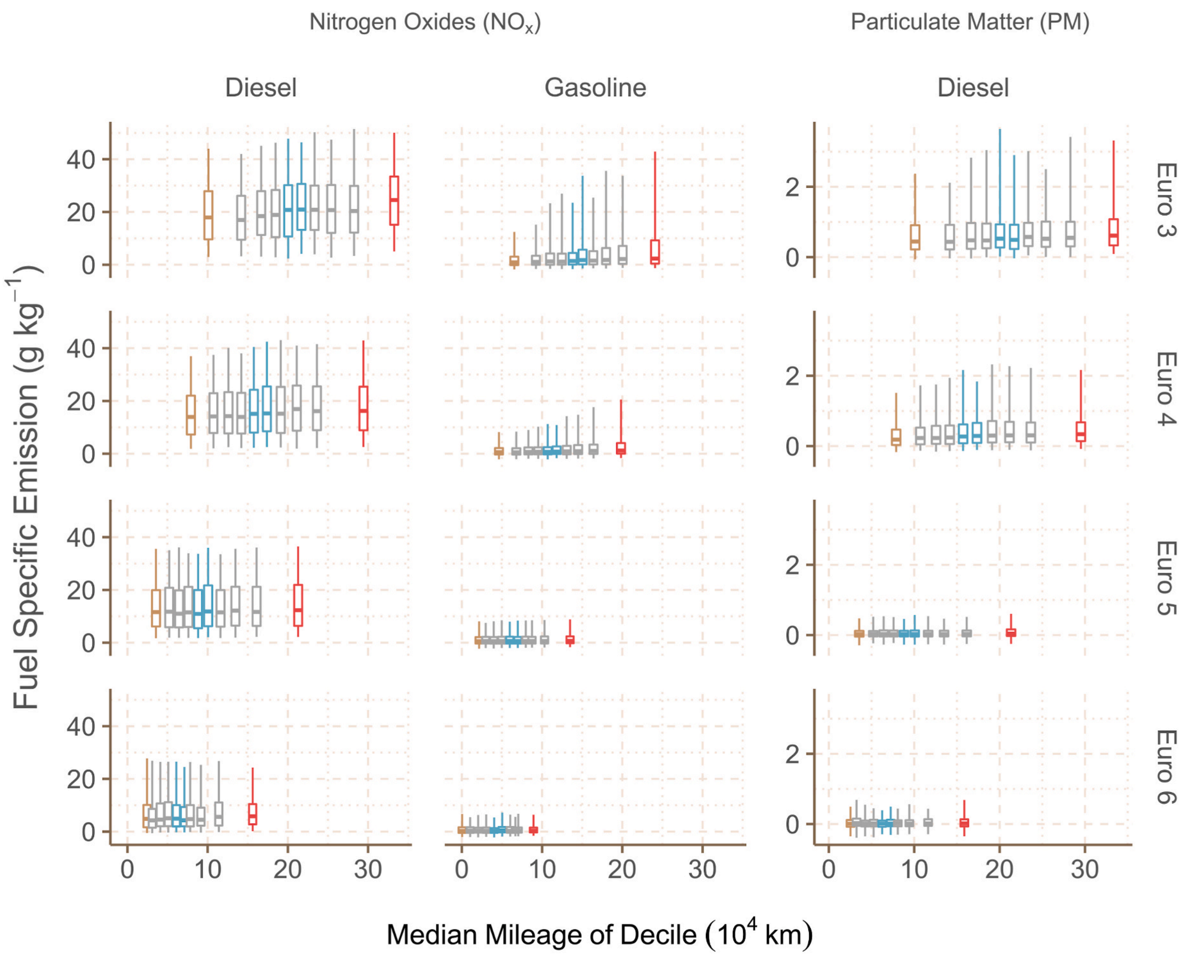

Fig. 2. Fuel-specific emissions (g kg^{-1}) of NO_x and PM as a function of vehicle mileage. The box plots show the distribution of emissions per decile of mileage, with each decile plotted at its median mileage value. The lowest, highest and middle mileage deciles are coloured to aid in comparison between panels. The hinges of the boxplots represent the 25th and 75th emission percentiles, and the whiskers the 5th and 95th percentiles.

passenger cars (6.9–19.5 g kg^{-1} across all Euro standards) are consistent with the wide range of NO_x performance in diesel vehicles widely reported in the literature (Davison et al., 2020; Bernard et al., 2018; Dallmann et al., 2018a,b).

The distributions of the gasoline passenger car NO_x emissions differ considerably from the diesel passenger cars and between the Euro standards. Euro 5 and 6 gasoline vehicles show a flat trend across the mileage deciles, and very small interquartlile ranges (2.4–2.7 and 1.8–2.2 g kg^{-1} respectively). Conversely, the Euro 3 and 4 gasoline vehicles show relatively larger interquartile ranges (3.2–8.8 and 2.4–4.0 g kg^{-1}), flatter trends for the lower NO_x quantiles and much steeper trends in the higher NO_x quantiles. The differences in the 95th percentile of fuel-specific NO_x between the first and tenth mileage deciles are +30.5 g kg^{-1} in Euro 3, +12.3 g kg^{-1} for Euro 4, +0.7 g kg^{-1} for Euro 5 and -0.3 g kg^{-1} for Euro 6. For comparison, the highest equivalent value for the diesel vehicles is seen in Euro 3 at +6.11 g kg^{-1}. All of this suggests a small proportion of high NO_x emitters among the Euro 3 and 4 gasoline fleets that are particularly sensitive to deterioration.

An unavoidable aspect of the data when making comparisons between Euro standards is that Euro 5 and 6 data is for a much lower range of cumulative mileage than the Euro 3 and 4 data, owing to the former vehicles being much younger. It is therefore possible that at higher cumulative mileages that are present in the remote sensing data set, Euro 5 and 6 gasoline passenger cars may show similar patterns of deterioration to the Euro 3 and 4 vehicles. However, it should be noted that there is overlap between the cumulative mileages of the four gasoline Euro standards. Even up to 135,000 km of cumulative mileage (representing the tenth mileage decile of Euro 5 and fifth decile of Euro 3), the Euro 3 and 4 distributions are widening, suggesting a deterioration effect, whereas the Euro 5 and 6 distributions remain flat, suggesting well controlled emissions.

A limited number of recent remote sensing studies focus on PM emissions (Chen et al., 2020; Smit et al., 2021), with Chen et al. (2020) noting that black smoke emissions have been following PM legislation limits, unlike many gaseous tailpipe emissions which have continued to exceed their respective limits. Gautam et al. (2010) sets a threshold of 1.5 g kg^{-1} of PM as a flag for a potentially compromised diesel particulate filter (DPF), which is well outside of the distributions of the DPF-equipped Euro 5 and 6 vehicles in Fig. 2. The fuel-specific PM emissions of Euro 5 and 6 diesel passenger cars appear to be well controlled overall, with a flat trend across mileage deciles at most PM quantiles. However there does appear to be evidence of deterioration at the higher quantiles of Euro 3 and 4 vehicles.

Carbon monoxide is generally understood to be well controlled and CO emissions are typically comfortably below emission limits (Chen and Borken-Kleefeld, 2014). While not visualised in Fig. 2, both gasoline and diesel vehicles appear to show similar trends — the five fuel-specific CO quantiles all increase at higher mileage deciles, with the difference between the lowest and highest deciles being greatest at $\tau = 0.95$, not unlike the trends seen in Euro 3 and 4 gasoline NO_x emissions. The key difference between the gasoline and diesel passenger cars is the lower absolute emissions of CO for diesel vehicles, particularly in the older Euro standards.

Recently, more attention has been paid toward ammonia emissions using remote sensing (Zhang et al., 2021; Farren et al., 2020), including limited analysis on their deterioration (Farren et al., 2021). There is some evidence of the deterioration of ammonia emissions for gasoline vehicles which is most pronounced in Euro 4 and 5. The gasoline vehicles tend to show a relatively low median but high 95th percentile fuel-specific NH_3 emission, reflecting the skewed nature of NH_3 emissions seen in Zhang et al. (2021). There appears to be no significant deterioration effect for the Euro 6 diesel passenger cars, suggesting that SCR-equipped vehicles are robust as far as NH_3 emissions are concerned.

Statistical modelling has some clear advantages over this sort of exploratory and visual analysis as it can be difficult to make real-world inferences from simple statistical tools like box plots. Firstly, there is arbitrariness that often comes with binning continuous data like vehicle mileage. More importantly, a key limitation is the inability to easily isolate the influences of other variables known to be important — in this case the instantaneous driving condition of the vehicle and ambient conditions. A modelling framework removes some of this arbitrariness and allows for the control of other covariates, as well as providing what could be described as a “rate of deterioration” as a function of vehicle mileage.

3.3. Multivariate statistical modelling

The introduction of new Euro standards tends to be associated with improvements in emissions control technologies and it is therefore important to consider how emissions deteriorate within a single Euro standard. Two pollutants were chosen to be further examined in a multivariate framework; NO_x due to being of significant interest owing to its air pollution impacts, and PM due to the considerable health effects associated with fine particulate matter.

We first consider deterioration effects over the normal lifetime of a vehicle, i.e., after 160,000 km of driving under Euro 6 legislation (Council of European Union, 2014). The models given in Equation (2) for gasoline NO_x and diesel PM and Equation (3) for diesel NO_x were fit with $\tau \in \{05, .10, .30, .50, .70, .90, .95\}$ and used to predict emissions at 0 and 160,000 km of cumulative mileage. VSP and ambient temperature were taken to be 7 kW t^{-1} and 288 K respectively, equal to the mean value of the variables in the remote sensing data set. Fig. 3 presents the predicted absolute deterioration of the fuel-specific NO_x and PM emissions for different quantiles for gasoline and diesel passenger cars. These values are also tabulated in Table S1.

The highest quantiles of Euro 3 and 4 gasoline vehicles increase by 17.1 and 9.1 g kg^{-1}, respectively. To put these values in context, the mean emissions of NO_x from Euro 3, 4, 5 and 6 passenger cars are 5.3, 2.9, 1.9 and 1.5 g kg^{-1} for gasoline and 21.0, 17.1, 15.9 and 8.2 g kg^{-1} for diesel. This analysis of absolute deterioration reveals some important characteristics for NO_x emissions. First, that the rate of deterioration of NO_x emissions is greater for a population of high-emitting gasoline passenger cars compared to diesel passenger cars. Second, Euro 5 and 6 gasoline passenger cars appear to have much better NO_x control than Euro 3 or 4 vehicles over the same range of cumulative mileage.

The trends in particulate matter for diesel vehicles reveal some interesting characteristics. For context, Euro 3 legislation did not require the use of a diesel particulate filters (DPF) whereas Euro 5 onward did.

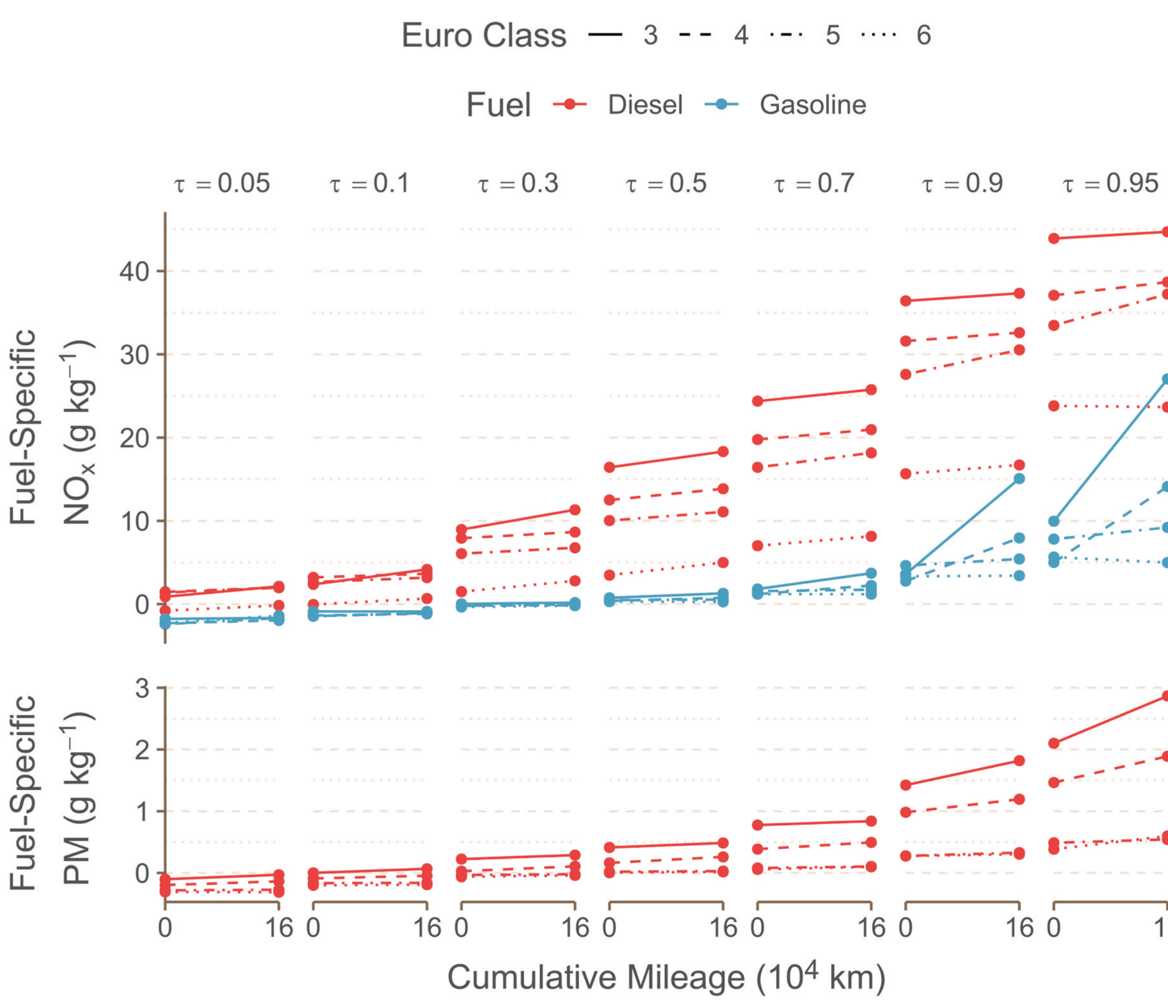

Fig. 3. Plot showing the modelled linear deterioration of passenger cars from 0 to 160,000 km of cumulative mileage (a vehicle’s “normal life” under Euro 6 legislation (Council of European Union, 2014)).

DPF technology became more widespread for Euro 4 vehicles even though they were not necessary, so the Euro 4 observations in the remote sensing data will contain a mixture of DPF and non-DPF-equipped vehicles. The results show that there are small populations of Euro 3 and Euro 4 diesel passenger cars where there is evidence of increased emissions of PM due to deterioration. However, Euro 5 and 6 vehicles appear to be well-controlled, suggesting that DPF technology provides a robust way of controlling particulate emissions over the lifetime of a vehicle.

One benefit of a statistical modelling approach is being able to comment directly on the magnitude and statistical significance of the rates of deterioration (represented by the models' mileage coefficients), which can provide a more comprehensive assessment of deterioration than is shown in Fig. 3. The magnitudes and bootstrapped confidence intervals of these rates of deterioration are tabulated in Table S2 and Table S3, and are briefly discussed below. When tabulated and presented in-text, deterioration rates are provided with their 95% confidence interval in parentheses and are expressed in the units of g kg^{-1} per 10^4 km driven for NO_x and mg kg^{-1} per 10^4 km driven for PM.

Gasoline passenger cars can be considered in two groups. First, Euro 3 and 4 gasoline vehicles show an exponential increase in their rates of deterioration as a function of τ. All terms are significant with the exception of $\tau \in \{05, .10\}$ for Euro 3 vehicles. These Euro standards both reach maxima at $\tau = 0.95$, at 1.06 (0.87–1.3) and 0.57 (0.49–0.67), respectively. Second, the Euro 5 and 6 gasoline vehicles show a relatively flat trend in rates of deterioration that are almost all insignificant. At higher quantiles ($\tau \geq 0.50$), Euro 5 and 6 are the only Euro standards where the differences are insignificant from one another, confirming well-controlled emissions for these vehicles.

For NO_x emissions from diesel passenger cars there is no consistent pattern of rates of deterioration changing with τ. The rates of Euro 3, 4 and 6 diesel cars reach maxima of 0.15 (0.12–0.18) at $\tau = 0.30$, 0.087 (0.064–0.11) at $\tau = 0.50$ and 0.094 (0.054–0.12) at $\tau = 0.50$ respectively, suggesting that the highest emitting vehicles are not necessarily any more sensitive to mileage-based deterioration than average emitters. Conversely, the deterioration rates of Euro 5 diesel cars reach a maxima of 0.24 (0.18–0.34) at $\tau = 0.95$, with rates increasing roughly linearly with $\tau (R^2 = 0.91)$. This pattern of behaviour is consistent with diesel passenger cars having a wide range of NO_x emissions, but emissions that do not show evidence of increases with mileage.

Across all four studied Euro standards, the rates of deterioration of diesel PM emissions possess a slight positive gradient with respect to τ up to $\tau = .70$. After this point the Euro standards deviate; the deterioration rates of the high emitting ($\tau > 0.70$) Euro 3 and 4 vehicles increase rapidly, reaching maxima of 53 (27–86) and 30 (16–46) mg kg^{-1} respectively, whereas the rates for Euro 5 and 6 remain low. Importantly, all but one of the deterioration rates for Euro 3 and 4 are seen to be significant, whereas the majority of the rates for Euro 5 and 6 are insignificant. This reinforces the insight from Fig. 3 that the PM emissions of DPF-equipped vehicles are well controlled.

3.4. Vehicle manufacturer effects

The differences between the real driving emissions of vehicles from different manufacturers is well reported (Davison et al., 2020, 2021; Grange et al., 2019, 2020; Borken-Kleefeld and Dallmann, 2018; Bernard et al., 2018; Dallmann et al., 2018a), but little has been reported on the differences in emission *deterioration* between manufacturers. Gasoline vehicles are of particular interest in this study due to the evidence of deterioration effects for some Euro standards. The exponential increase in the rate of deterioration seen in Euro 3 and 4 vehicles may be driven by only certain manufacturers, for example. Conversely, some manufacturers of gasoline vehicles may have a significant deterioration effect for Euro 5 and 6 cars, despite no strong effect being present when considered on a bulk level.

The density functions of the bootstrapped deterioration rates from Equation (2) using vehicle manufacturer group as the interaction effect are visualised in Fig. 4. The 8 most common manufacturer groups are shown for each Euro standard, each with at least 1000 observations. The distribution of cumulative mileage for each of these manufacturers are similar, although not identical (visualised in Fig. S3).

There are clear differences between gasoline car manufacturers within each Euro standard. The influence of mileage on NO_x emissions from almost all Euro 5 vehicles is insignificant; the confidence intervals includes 0 for most manufacturers and values of τ. The only Euro 5 manufacturers with a significant mileage effect are Ford with 0.0294

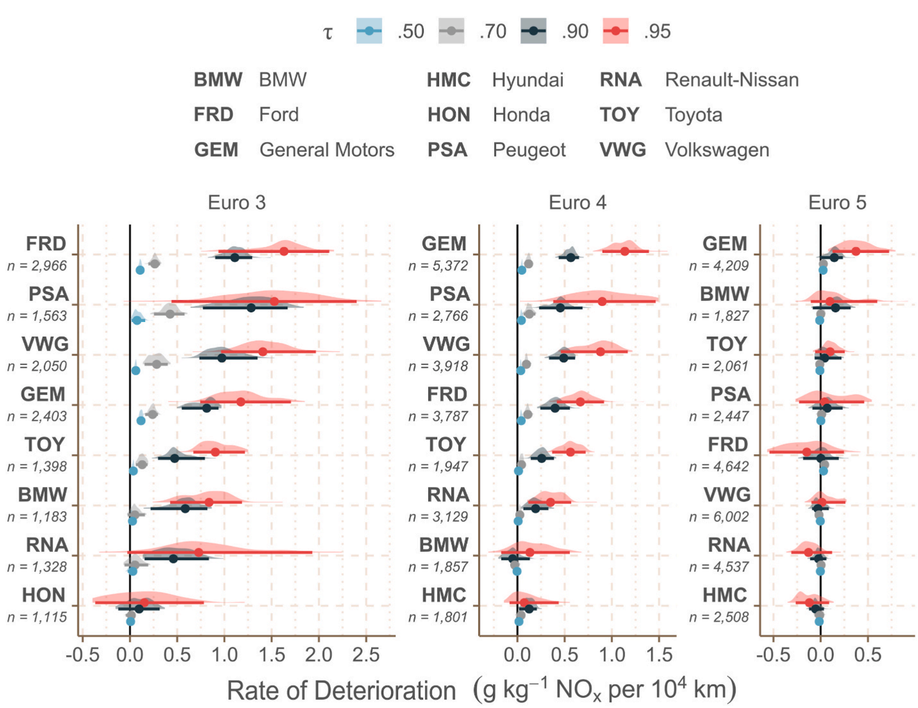

Fig. 4. Density functions of bootstrapped rates of deterioration from multivariate linear quantile regression fits ($\tau \in \{50, .70, .90, .95\}$) predicting NO_x as a function of vehicle mileage and vehicle specific power for gasoline passenger car manufacturers (Equation (2)). Density functions are normalised per individual function, so their peak heights should not be directly compared. Manufacturers are ordered by the magnitude of their 95th percentile deterioration effect. The number of observations for each manufacturer, *n*, is shown. $x = 0$ is indicated with a solid black vertical line. The median value and the 95% confidence intervals are shown beneath each density function as circles and horizontal lines, respectively. Distribution functions and confidence intervals were calculated and visualised using the *ggdist* R package (Kay, 2021).

and General Motors with 0.0255 g kg^{-1} NO_x per 10^4 at $\tau = 0.50$, and General Motors with 0.373 g kg^{-1} NO_x per 10^4 at $\tau = 0.95$. For Euro 3 and 4 manufacturers there is more evidence of diverging behaviours with some manufacturers demonstrating good NO_x control across a range of quantiles and others showing much stronger deterioration effects, especially at higher quantiles.

The analysis reveals that the progressive improvement of vehicle technology through Euro 3 to Euro 6 vehicles demonstrably led to improvements in vehicle emissions control. This finding is supported by the availability of vehicle-specific mileage information which shows that at 160,000 km, Euro 5 and Euro 6 gasoline passenger cars have much improved emissions control than Euro 3 and 4 vehicles for the *same* mileage.

The analysis of emissions deterioration of gasoline passenger cars shows that there exist small fractions of older vehicles in the fleet (Euro 4 and older) that have emissions that are similar to Euro 5 diesel cars for NO_x. From a policy perspective, it would be beneficial to target those vehicles for replacement or scrappage, given that there are relatively few of them. The current work broadly supports recent Low Emission Zone developments such as the ULEZ (Ultra Low Emission Zone) in London (Transport for London, 2021), which prohibits gasoline passenger cars older than Euro 4 and pre-Euro 6 diesel cars. However, the analysis suggests that restricting pre-Euro 5 gasoline cars would be advantageous given the consistently low emissions of Euro 5 and 6 gasoline cars, even with high mileage.

4. Conclusion

The large data sets that can be acquired by vehicle emission remote sensing measurements provide many opportunities to develop a good understanding of vehicle emission characteristics. Such data also offers the potential to adopt more sophisticated analysis approaches that extend beyond simple aggregations such as mean emissions by Euro standard. By adopting statistical modelling approaches, inferences drawn from the data will be stronger, with valuable information provided on uncertainties. An important advantage of this approach is that in determining vehicle mileage effects on emissions, other influences such as ambient temperature and vehicle power demand can be controlled for.

In the current work, the adoption of quantile regression as a technique fits well with the characteristics of the data being studied. The main benefit is the determination of whether all vehicles deteriorate similarly with increased mileage, or whether deterioration is controlled differently by different strata of a vehicle fleet. For gasoline passenger cars, where deterioration effects are most apparent, our results show that NO_x emission deterioration is significantly greater in a small population of vehicles.

In contrast to most other studies on vehicle emissions deterioration, the availability of measured mileage for individual vehicles in the current study is a considerable benefit, which avoids the use of proxy vehicle age-based data. For particulate matter, only pre-DPF vehicles show evidence of increasing emissions at higher mileages, and that DPF-equipped vehicles retain effective PM control even at high mileages. The results also show that while there is evidence of different deterioration behaviour depending on vehicle manufacturer for pre-Euro 5 vehicles, post-Euro 4 vehicles show no such evidence.

CRediT authorship contribution statement

Jack Davison: Formal analysis, Writing – original draft, Methodology, Visualization. **Rebecca A. Rose:** Investigation, Data curation, Writing – review & editing. **Naomi J. Farren:** Investigation, Writing – review & editing. **Rebecca L. Wagner:** Investigation, Data curation, Writing – review & editing. **Shona E. Wilde:** Investigation, Writing – review & editing. **Jasmine V. Wareham:** Investigation, Writing – review & editing. **David C. Carslaw:** Conceptualization, Methodology, Supervision, Writing – review & editing, Funding acquisition.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.aeaoa.2022.100162.

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