Environment International 182 (2023) 108330

Contents lists available at ScienceDirect

Environment International

journal homepage: www.elsevier.com/locate/envint

Full length article

The impact on passenger car emissions associated with the promotion and demise of diesel fuel

Samuel Wilson [a], Naomi J. Farren [a], Rebecca A. Rose [b], Shona E. Wilde [a], Jack Davison [b], Jasmine V. Wareham [b], James D. Lee [a], David C. Carslaw [a,b,*]

[a] Wolfson Atmospheric Chemistry Laboratories, University of York, Innovation Way, Heslington, York YO10 5DD, United Kingdom
[b] Ricardo Energy & Environment, The Gemini Building, Fermi Avenue, Harwell OX11 0QR, United Kingdom

ARTICLE INFO

Handling Editor: Xavier Querol

Keywords:
Vehicle emissions
Nitrogen oxides
Remote Sensing
Diesel

ABSTRACT

The promotion and growth in the use of diesel fuel in passenger cars in the UK and Europe over the past two decades led to considerable adverse air quality impacts in urban areas and more widely. In this work, we construct a multi-decade analysis of passenger car emissions in the UK based on real driving emissions data. An important part of the study is the use of extensive vehicle emission remote sensing data covering multiple measurement locations, time periods, environmental conditions and consisting of over 600,000 measurements. These data are used to consider two scenarios: first, that diesel fuel use was not promoted in the early 2000s for climate mitigation reasons, and second, that there was not a dramatic decline in diesel fuel use following the Dieselgate scandal. The strong growth of diesel fuel use coincided with a time when diesel NO_x emissions were high and, conversely, the strong decrease of diesel fuel use coincided with a time when diesel vehicle aftertreatment systems for NO_x control were effective. We estimate that the growth in diesel car use in the UK results in excess NO_x emissions of 721 kt over a three decade period; equivalent to over 7 times total annual passenger car NO_x emissions and greater than total UK NO_x emissions of 681.8 kt in 2021 and with an associated damage cost of £5.875 billion. However, the sharp move away from diesel fuel post-Dieselgate only reduced NO_x emissions by 41 kt owing to the effectiveness of modern diesel aftertreatment systems.

1. Introduction

1.1. Policy context

Exhaust emissions from road vehicles are a dominant source of ambient air pollution, particularly in densely populated urban environments. The proximity of road vehicles to the general population, particularly in urban areas, is a continued cause for concern given the myriad of health and wider environmental impacts associated with traffic-related air pollution (Hoek et al., 2013; Bell et al., 2011; Anenberg et al., 2019). Over the past two decades the contribution of diesel–fuelled vehicles to adverse air pollution has been of particular concern. For light duty vehicles in the early 2000's, diesel fuel offered fuel-efficiency and carbon (25 – 29% lower CO_2 emissions) advantages over gasoline (Sullivan et al., 2004). Whilst technology advancements and trends in passenger car characteristics (engine size, vehicle mass etc.) have since reduced this advantage, it's presence still resulted in the active promotion of diesel fuel in many countries (Dornoff and Rodriguez, 2019). Furthermore, diesel vehicles are significant contributors to the emission of important air pollutants such as fine particulate matter ($PM_{2.5}$) and nitrogen oxides (NO_x).

With respect to emissions of NO_x, the principal concern from a human health perspective is related to concentrations of nitrogen dioxide (NO_2), which resulted in the development of ambient limits in Europe and elsewhere (European Council, 2008). The current limit for annual mean NO_2 concentrations in Europe is 40 $\mu g\ m^{-3}$, which has been widely exceeded across many European countries for the past two decades. However, the evidence surrounding the adverse health effects associated with NO_2 exposure has grown stronger over recent years, prompting the World Health Organisation (WHO) to recommend an annual mean guideline of 10 $\mu g\ m^{-3}$ (WHO, 2021). Elevated concentrations of NO_2 are of particular concern to local air quality and especially in urban environments where there is close proximity of vehicular emissions to large populations. For this reason, cities across the UK and

* Corresponding author at: Wolfson Atmospheric Chemistry Laboratories, University of York, Innovation Way, Heslington, York YO10 5DD, United Kingdom.
E-mail address: david.carslaw@york.ac.uk (D.C. Carslaw).

https://doi.org/10.1016/j.envint.2023.108330
Received 7 August 2023; Received in revised form 23 October 2023; Accepted 14 November 2023
Available online 18 November 2023
0160-4120/

Europe have adopted mitigation measures such as low emission zones that seek to reduce emissions and concentrations of NO_2 and other traffic-related air pollutants, e.g., the Ultra Low Emission Zones in London, Paris, and Milan (Transport for London, 2023; République Français, 2022; Comune di Milano, 2023).

Emissions of NO_x also contribute to significant wider environmental damage. For example, NO_x is a key precursor in the formation of secondary particulate matter (PM) and tropospheric ozone, both of which are associated with a range of adverse health and environmental impacts (Ngatia et al., 2019; Selman and Greenhalgh, 2010; Kroll et al., 2005; Ebi and McGregor, 2008). It is for these reasons that the past few decades has seen an increase in policy designed to mitigate the multiple impacts of NO_x and other pollutants. At a national level in Europe, total emission limits have been set as part of The National Emissions Ceiling Directive (NECD), which sets annual pollutant emission ceilings for member states for a range of pollutants such as $PM_{2.5}$, Volatile Organic Compounds (VOCs) and NO_x (European Environment Agency, 2015; European Environment Agency, 2022).

The promotion of diesel passenger cars in the UK corresponds to the time period between 2001 to 2015 during which company car tax (CCT), a benefit in kind tax applied to vehicle sold commercially, and vehicle excise duty (VED), a tax levied on vehicles for use on public roads, were restructured based on a vehicle's reported carbon dioxide (CO_2) emissions (UK Government, 2023; HM Treasury, 2000). These policies, which were initially introduced for CO_2 reduction and climate change mitigation reasons, provided a financial incentive to purchase a diesel car over one powered by gasoline. A combination of the restructured CCT and VED policies resulted in new diesel passenger car registrations increasing by 38% in one year from 2001 to 2002, and the total number of diesel passenger cars licensed in the UK increasing by a factor of 3 from 2001 to 2015 (UK Goverment Asset Publishing Service, 2018). Whilst the air quality drawbacks associated with diesel fuel emissions were generally well understood at the inception of the restructured CCT and VED policies, CO_2 reduction and climate mitigation reasoning took precedence, and full consideration was not given to the air quality implications of the adoption of diesel technology on a mass scale (HM Treasury, 1998). The significant increase in diesel car use was more generally observed across Europe (Cames and Eckard, 2013).

Following the 2015 'Dieselgate' scandal, diesel passenger car sales in the UK plummeted (the 'demise' of diesel fuel). The scandal was directly associated with emissions of NO_x and the discovery of deliberate tampering that led to excess emissions. In 2017 VED was further restructured such that only the first year of the tax was calculated based on reported CO_2 emissions, before a fixed annual rate is applied for all internal combustion engines (ICE) powered passenger cars. This change removed much of the financial incentive to own a diesel vehicle, and when combined with the growing negative reputation of the fuel, resulted in the share of new diesel passenger car registrations falling from 48.3% in 2015 to 7.7% in 2022 (Department for Transport, 2022c).

1.2. Passenger car vehicle emissions

Passenger car tailpipe emissions have been regulated in the UK according to the European Type Approval emission standards (Euro standards) since 1992 (European Union Law, 1991; European Commission, 2022). Vehicle Type Approval is a Conformity of Production (CoP) process that all passenger cars must undergo before being eligible for sale in the UK (Vehicle Certification Agency, 2022). Early Euro standards were enforced during Type Approval through predefined testing methods (drive cycles) under standardised laboratory conditions, with emissions limits set for each pollutant. Over the following three decades Euro standard emission limits and testing methods have become increasingly stringent, and now extend from 'Euro 1' (1992) to 'Euro 6d' (2021) for passenger cars. The current Euro 6d standard regulates the exhaust emission of carbon monoxide (CO), hydrocarbons (HC), NO_x, and PM, measured as the amount of pollutant emitted per unit distance travelled ($g\,km^{-1}$); a CO_2 emission factor is also reported in $g\,km^{-1}$ and used to determine the VED rate (European Union Law, 2016). Different pollutant limits are set for diesel and gasoline fuels, Table 2 in the supporting information shows the pollutant emissions limits for each fuel type and Euro standard. In order to address the progressively demanding emissions limits, vehicle manufacturers have developed a variety of exhaust aftertreatment system strategies.

Gasoline passenger cars have used the three-way catalyst for emissions control in Europe since the early 1990s due to the introduction of the Euro 1 standard (Westerholm et al., 1996; European Commission, 2022). The three-way catalyst has remained the aftertreatment system used by gasoline vehicles since this time but has been refined alongside developing Euro standards. By contrast, the technology used in diesel passenger cars has greatly evolved since the early 1990s as shown in Fig. 1. The principal technologies used to control diesel passenger NO_x emissions include exhaust gas recirculation (EGR), diesel oxidation catalysts (DOCs), and diesel particle filters (DPFs). EGR routes exhaust gases back into the combustion chamber, lowering combustion temperatures and reducing emissions of NO_x (Zheng et al., 2004). DOCs facilitate the high temperature conversion of CO and HC into CO_2 and water, as well as oxidising NO to NO_2, the latter of which is necessary for the use in other forms of aftertreatment (Twigg, 2005). DPFs physically capture exhaust particulate matter during engine operation before oxidising stored material into CO_2. A major milestone in the reduction of NO_x emissions from diesel passenger cars was the introduction of selective catalytic reduction (SCR) and lean NO_x traps (LNTs) that were designed specifically to control NO_x emissions. These technologies were introduced on a mass scale following the increasingly stringent NO_x emissions limits that accompanied the Euro 6 standards (2014) for diesel passenger cars.

As a result of technology improvements, diesel passenger car exhaust systems have become increasingly complicated, often comprising a combination of different aftertreatment components. Successful vehicle emission control therefore presents a significant engineering challenge for vehicle manufacturers. To ensure that passenger cars continue to meet emissions limits across a range of operating parameters the Euro 6 standards introduced real driving emissions (RDE) testing, during which vehicle emissions are measured using portable emissions measurement systems (PEMS) under real driving conditions. The introduction of RDE testing greatly expedited the integration of SCR and LNT systems, which were required to meet the lowered NO_x emission limits outside of traditional laboratory testing. Whilst RDE legislation was under development prior to Dieselgate, the occurrence of the scandal increased awareness of passenger car NO_x emission issues and likely accelerated implementation of the policy (ICCT, 2017; Commission, 2016).

Vehicle emission remote sensing is an unobtrusive way of measuring emissions from vehicles under real driving conditions, which has been extensively described elsewhere (Bishop and Stedman, 1996; Burgard et al., 2006; Carslaw and Rhys-Tyler, 2013). The instrumentation is deployed on the roadside. Individual measurements are less than 1 s in duration and the measured emissions represent a single driving condition. However, a very large number of vehicles can be measured in a short period of time, allowing for measurements to be aggregated (e.g., by fuel type, vehicle category, Euro class) and used to provide detailed emissions behaviour insight over a range of driving and ambient conditions.

1.3. Objectives

The aim of this paper is to quantify the impact of the promotion of diesel fuel usage in passenger cars on emissions of important air pollutants at a UK level, with a specific focus on the emission of NO_x. We also aim to quantify the impact of the rapid move away from diesel fuel on passenger car emissions in the wake of the Dieselgate scandal. An important aspect of our analysis is the use of real driving emissions of NO_x and other pollutants based on comprehensive vehicle emission

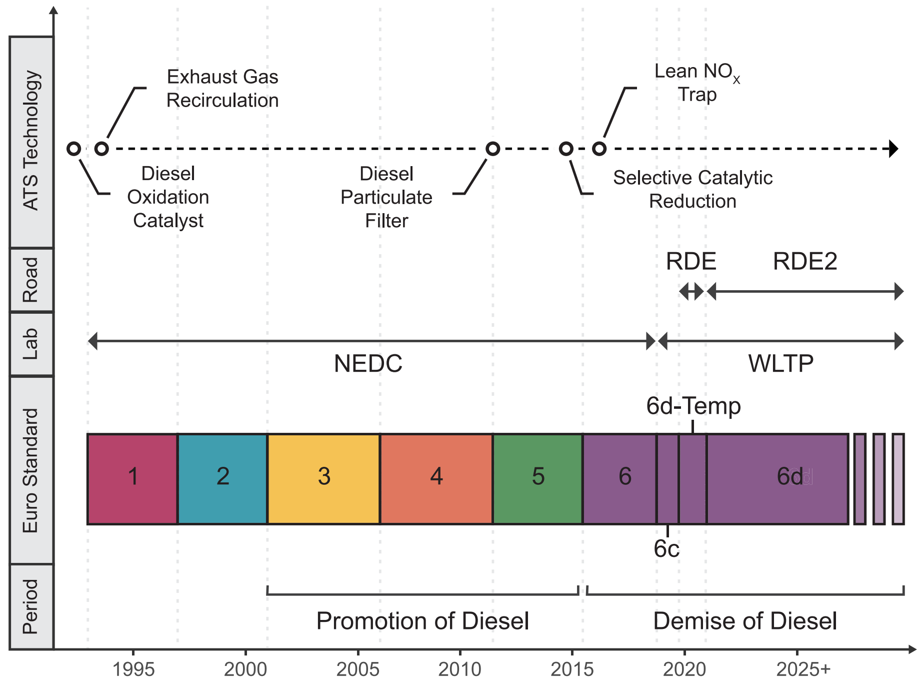

Fig. 1. Timeline of diesel passenger car Euro standards, laboratory and road testing methods, and aftertreatment technology adoption. ATS Technology = Aftertreatment Technology System technology. NEDC = New European Drive Cycle; WLTP = Worldwide Harmonised Light Vehicle Test Procedure; RDE = Real Driving Emissions.

remote sensing measurements. The vehicle emission remote sensing data are used to construct a timeline of passenger car emissions over three decades, which is then used to evaluate the air quality impacts associated with the promotion and demise of diesel fuel. The results are compared with the UK National Atmospheric Emissions Inventory (NAEI) emissions to put their impacts into context and the wider implications for urban and regional air quality discussed.

2. Data and methods

2.1. Instrumentation

The measurements in this work were made with two remote sensing systems: the Fuel Efficiency Automobile Test (FEAT) research instrument supplied by the University of Denver and the commercially available Opus AccuScan RSD 4600/5000 (University of Denver, 2023; Opus Inspection, 2023). Vehicle emission remote sensing instrumentation consists of three fundamental components: a spectroscopic unit for the measurement of exhaust gases, apparatus for measuring vehicle speed and acceleration, and a camera for photographing vehicle registration plates.

Both systems operate perpendicular to the flow direction of a single lane of traffic and utilise collinear open-path infrared (IR) and ultraviolet (UV) spectroscopy. A remote sensing observation, which includes the emission measurement, speed and acceleration measurement, and registration plate photograph, is made whenever the collinear light beam is obstructed by a passing vehicle. 100 spectroscopic measurements are taken of each exhaust plume in half a second and emission values are reported as molar volume ratios to CO_2, eliminating the variation associated with vehicle position and exhaust plume dispersion. All observations are preserved regardless of measurement validity, which is important for deriving information about the vehicle fleet composition.

The FEAT instrument measures carbon monoxide (CO), carbon dioxide (CO_2), hydrocarbon (HC), and a background reference using four non-dispersive IR detectors, whereas ammonia (NH_3), nitrogen oxide (NO) and nitrogen dioxide (NO_2) measurements are obtained using two separate dispersive UV spectrometers. An extensive description of the development and operation of the FEAT instrument, as well as a detailed comparison of the two remote sensing systems is available in the literature (Burgard et al., 2006; Bishop and Stedman, 1996; Rushton et al., 2018).

2.2. Data description

The remote sensing data set used in this work combines information from measurement campaigns conducted at 77 sites across the UK between 2012 and 2022. To ensure safe instrument operation and maximise measurement validity, data was collected during only dry, daylight hours (0800 to 1800 h). The ambient temperature throughout the measurement campaigns ranged from −1 to 29 °C with a mean temperature of 14 °C.

The specific vehicle information for each remote sensing observation was obtained by cross-referencing the registration plate photographs with vehicle technical databases. These data were supplied by CDL Vehicle Information Services Limited, who retrieved information from the Driver and Vehicle Licensing Agency and the Society of Motor Manufacturers and Traders Motor Vehicle Registration Information System. All measurements were assigned technical information relating to the fuel type, emission standard and registration date of the vehicle. Additional vehicle information was matched to approximately 70% of vehicles and included details on the manufacturer, vehicle dimensions and vehicle mass.

A total of 604,435 measurements of Euro 2 to 6 passenger cars are contained within the data in five different fuel classes: diesel vehicles, gasoline vehicles, gasoline full-hybrid electric vehicles (FHEV), gasoline plug-in–hybrid electric vehicles (PHEV), and electric vehicles — other fuel types were not measured in sufficient numbers to include in this analysis. For the evaluation of passenger car fleet composition trends all measurements were used, regardless of the validity of the emissions or speed/acceleration data, or the presence of additional technical information; this was essential for ensuring that the composition information derived from the remote sensing data was an accurate representation of the on road vehicle fleet. The calculation of distance based emission factors requires valid emissions and speed/acceleration data, as well as additional technical information Davison et al., 2020). Therefore only observations with a complete measurement profile were used. 56 % of measurements were used to derive emission factors for CO and HC, 49 % of measurements were used to derive emission factors for NO_x, and 34 % of measurements were used to derive emission factors for NH_3. The lower proportion of valid NH_3 emissions measurements resulted from issues with instrument calibration. A statistical summary of the data set and the subsets used for emission factor calculation is provided in Table 1.

2.3. Emission factor calculation

The conversion of remote sensing pollutant molar volume ratios to CO_2 into fuel-specific emission factors (mass of pollutant emitted per mass of fuel consumed in $g\,kg^{-1}$) is straightforward, and relies on stoichiometric assumptions about the composition and combustion of hydrocarbon fuels. This methodology has been described in detail elsewhere (Bishop and Stedman, 1996; Burgard et al., 2006). Whilst fuel-specific emission factors are often reported directly, their distance-based equivalents (mass of pollutant emitted per unit distance travelled in $g\,km^{-1}$) are more versatile, and can be compared to the emission factors generated during vehicle type approval, as well as those used for emission inventory development. Distance-based emission factors were calculated from the remote sensing data using a method developed by Davison et al. (2020), a brief overview of which is provided here.

For each remote sensing observation, a physics-based approach was applied to calculate vehicle specific power (VSP) in kW t^{-1} from a combination of the speed and acceleration measurements as well as the vehicle technical information. Road load and aerodynamic drag coefficients provided by Davison et al. (2020) were also used in this calculation. Next, fuel consumption in $kg\,s^{-1}$ was determined based on the linear relationship with VSP derived from the passenger car and heavy-duty emissions model (Hausberger, 2003). The parameters used in this model were based on Euro 5 and 6 vehicles, therefore the fuel consumption for Euro 2 to 4 vehicles was increased by 5 % to approximate poorer fuel efficiency. Fuel consumption values for gasoline hybrid vehicles, which comprised 2 % of the remote sensing measurements, were reduced by 30% to approximate improved fuel efficiency resulting from electric power train utilisation (Farren et al., 2021).

The fuel-specific emission factors from remote sensing measurements were then combined with the corresponding modelled fuel consumption values to generate time-specific emission factors (mass of pollutant emitted per unit time in $g\,s^{-1}$). Next, the time-specific emission factors were aggregated and mapped to 1 Hz drive cycle data obtained from portable emission measurement system (PEMS) measurements, undertaken by the UK Department for Transport. The PEMS data contained 4243 km of 'real-world' urban, rural, and motorway driving, with individual test lengths ranging from 70.4 to 78.1 km. Further description of these drive cycles, as well the full details of this approach are described extensively in Davison et al. (2020).

Distance-based emission factors were calculated as the sum of the modeled 1 Hz time-specific emission factors divided by the total distance over 58 drive cycles. The output of this method was a distance-based emission factor for each vehicle model year and fuel type derived directly from on-road remote sensing measurements, reflecting a wide range of real driving conditions. The method has been evaluated and compared with the UK national inventory, which demonstrated carbon/energy balance within 1.5% when calculating total UK passenger car emissions (Davison et al., 2021).

Table 1
Statistical summary of the remote sensing data set and subsets.

Characteristic	All	CO & HC	NH_3	NO_x
# measurements	602280	321073	194544	281188
Diesel (%)	45	45	48	46
Gasoline (%)	51	52	50	52
FHEV (%)	3	2	2	2
PHEV (%)	0.5	0.3	0.2	0.3
EV (%)	0.5	0	0	0
Euro 2 (%)	2	1	1	1
Euro 3 (%)	13	11	12	9
Euro 4 (%)	24	23	25	22
Euro 5 (%)	30	33	35	33
Euro 6 (%)	31	32	27	35
Mean speed ($km\,h^{-1}$)	36.1	36.04	36.13	36.26
Mean acceleration ($km\,h^{-1}\,s^{-1}$)	1.22	1.33	1.44	1.36
Mean vehicle specific power (kW t^{-1})	8.73	8.49	8.49	8.49
Mean temperature (°C)	14.54	14.37	14.61	14.14

2.4. Timeline construction

To evaluate UK passenger car emissions trends, a distance-based emission factor timeline was constructed as a function of vehicle model year for each measured pollutant. Model year was defined as the year in which a passenger car was first registered in the UK, obtained from the vehicle technical information. Remote sensing measurements were grouped by model year before the ratio of different fuel types was calculated for each group (excluding electric vehicles which do not produce exhaust emissions). This information was combined with the corresponding individual fuel type distance-based emission factor data to generate an average $g\,km^{-1}$ value for each vehicle model year.

A major advantage of this approach is that it enables retrospective analysis of multi-decade emission trends predating the remote sensing measurement period (2012 to 2022). In this work, the data contained sufficient vehicle measurements to construct emissions timelines for diesel and gasoline vehicles with model years between 1998 to 2022, gasoline FHEV vehicles with model years between 2006 to 2022, and gasoline PHEV vehicles with model years between 2015 to 2022. The exact number of measurements available for each model year and fuel type can be found in the supporting information. For the model years 2020, 2021, and 2022, no valid NH_3 measurements were reported as a result of an instrument calibration fault. To complete the emission factor data, the mean of the NH_3 distance-based emission factor for the model years 2017, 2018, and 2019 (Euro 6 vehicles) was calculated and applied to the missing period.

2.5. Scenario development

The NO_x emission impact of the promotion and decline of diesel passenger cars in the UK was assessed through the development of three scenarios, for each of which a NO_x emission factor timeline was constructed. 'Business as Usual' (BAU) was defined as the baseline scenario, and average NO_x emission factors were derived from unmodified remote sensing emissions and fuel type composition data. A 'No Diesel Promotion' (NDP) scenario was constructed in which the fuel type composition data was adjusted such that the ratio of diesel and gasoline passenger cars remained unchanged from the mean value of the years 1998 to 2000. In this case unmodified individual fuel type emission factor data were used, such that any differences in the scenario NO_x emission timelines were a direct result of the fuel type composition changes. Similarly, a 'No Diesel Demise' (NDD) scenario was constructed in which only the post 2015 model year fuel type composition data was adjusted such that the ratio of diesel and gasoline vehicles remained unchanged from the mean value of the years 2013 to 2015. A visualisation of the adjustments made to fuel type composition as a function of model year for each scenario is presented in Fig. S1. Only the ratio of diesel and gasoline vehicles was adjusted in the NDP and NDD scenarios, the proportion of the total of these fuel types combined (diesel + gasoline) to the total proportion of hybrid vehicles (gasoline FHEV + gasoline PHEV) was unchanged, as were the individual proportions of each hybrid vehicle type.

2.6. Lifetime emission calculation

To further understand and contextualise the average NO_x emission factor values developed for each scenario, the data were combined with new passenger car registration and average annual mileage information. Total lifetime NO_x emissions from all passenger cars registered between 1998 to 2023 were calculated for each scenario, such that they could be dis-aggregated into annual values and compared.

First, the average operational life span of a passenger car in the UK was assumed to be 14 years (SMMT, 2022). Because the data for each scenario contained information for vehicles registered between 1998 to 2022, the time period over which lifetime NO_x emissions were calculated was 1999 (the year all 1998 model vehicles had been registered), to 2036 (the year all 2022 model vehicles had reached the end of their assumed lifespan). For each vehicle model year between 1998 and 2022 in each scenario a total fleet NO_x emission factor (kt km^{-1}) was calculated to be the product of the corresponding average passenger car NO_x emission factor (g km^{-1}) and the number of new annual passenger car registrations. From here, these values were multiplied by annual average total passenger car mileage data (km) for the years 1999 to 2036 yielding total NO_x emissions for each year (kt). Finally, the total NO_x emission values for each year between 1999 to 2036 were combined to produce an overall NO_x emission value (kt) for each year between 1999 to 2036 for each scenario. Summing all annual overall NO_x amounts within the three scenarios yielded a total lifetime NO_x emission value for each.

Annual new passenger car registration data were obtained from the DfT (Department for Transport, 2022b) and Northern Ireland Statistics and Research Agency (Northern Ireland Statistics and Research Agency, 2022), and annual average total passenger car mileage information was acquired from the National Travel Survey (Department for Transport, 2022a). For the years 2023 – 2036 the annual average total passenger car mileage was assumed to be the mean of the years 2017, 2018, and 2019, which preceded the COVID-19 pandemic, during which travel restrictions reduced annual passenger car mileage.

3. Results and discussion

3.1. Vehicle emission trends

Passenger car distance-based (g km^{-1}) emission factors for CO, HC, NH_3, and NO_x are shown by fuel type and model year in Fig. 2. The range of calculated emission factors are consistent with those reported in the literature (Davison et al., 2020). For the pollutants that are regulated during passenger car Type Approval, the calculated emission factors align closely with the corresponding range of Euro standard emissions limits displayed in Table 2. It should be noted that the trend in NO_x emissions from diesel cars includes the effect of hardware and software fixes on UK vehicles in the wake of the Dieselgate scandal. The effect of these fixes, which are implicit in the remote sensing measurements, were calculated to be a 30–36% reduction in NO_x affecting 1.6 and 2.0 litre Volkswagen Group diesel passenger cars (Grange et al., 2020).

Overall, a steady decrease in emission factors with increasing model

Table 2
European passenger car (Euro) standard emissions limits from Euro 1 ('E1') to Euro 6 ('E6').

	Date	Distance-specific emission (g km^{-1}) CO	HC	HC + NO_x	NO_x	PM	(# km^{-1}) PN
Gasoline							
E1	1992	2.72	—	0.97	—	—	—
E2	1996	2.2	—	0.50	—	—	—
E3	2000	2.30	0.2	—	0.15	—	—
E4	2005	1.00	0.1	—	0.08	—	—
E5	2009	1.00	0.1	—	0.06	0.005	—
E6	2014	1.00	0.1	—	0.06	0.005	6×10^{11}
Diesel							
E1	1992	2.72	—	0.97	—	0.140	—
E2	1996	1.00	—	0.90	—	0.100	—
E3	2000	0.64	—	0.56	0.50	0.050	—
E4	2005	0.50	—	0.30	0.25	0.025	—
E5	2009	0.50	—	0.23	0.18	0.005	6×10^{11}
E6	2014	0.50	—	0.17	0.08	0.005	6×10^{11}

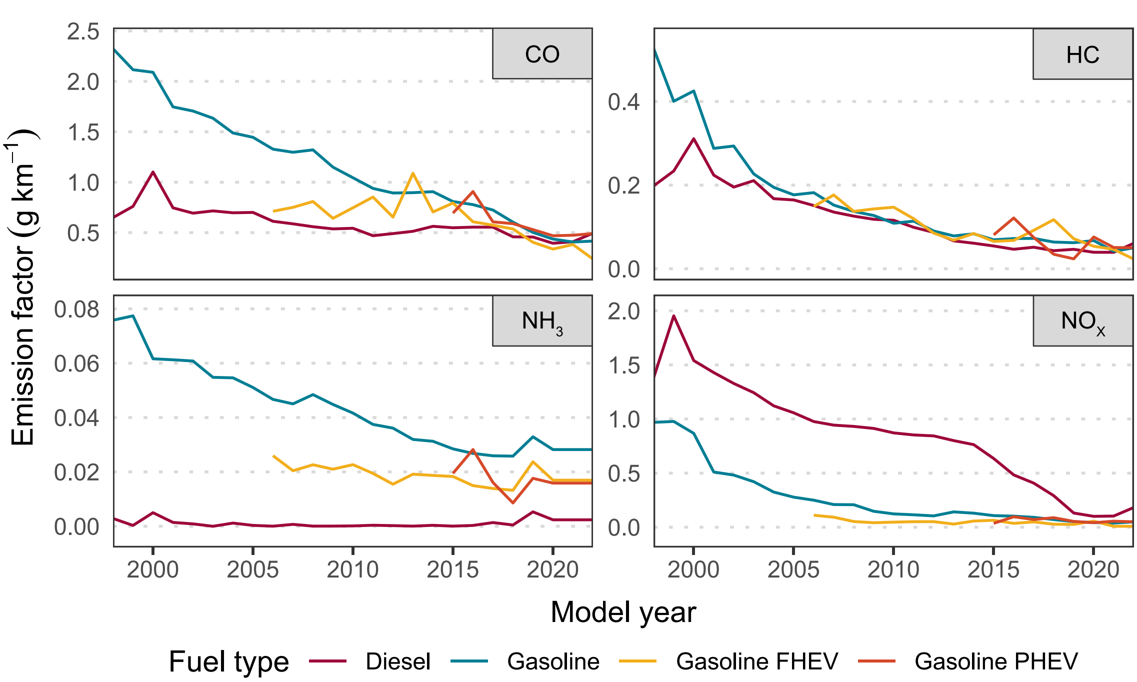

Fig. 2. Passenger car distance-based emission factor values (g km^{-1}) for diesel, gasoline, gasoline FHEV, and gasoline PHEV vehicles as a function of model year (1998 to 2022), for CO, HC, NH_3, and NO_x.

year is observed for all pollutants and fuel types. This trend is primarily attributed to continually improving internal combustion engine and exhaust aftertreatment technology, driven by increasingly stringent emission standards and testing methods. It is, however, important to consider that earlier-model passenger cars are older at the time of measurement, and the deterioration of these vehicles is likely to partially influence this observed trend. Vehicle model years before 2012 (the beginning of the remote sensing measurement period) were measured at an age that is equal to the difference between the measurement date and that of the vehicle's registration. Whilst age is not a direct measure of accumulated vehicle usage and deterioration, these variables can be assumed to be correlated. The effect of passenger car deterioration on pollutant (CO, NH_3, NO_x) emission factors has been previously derived from remote sensing data and reported in the literature (Davison et al., 2022). It is likely that the impact of the reported deterioration rates on the results of this work would be small. However, it is important to acknowledge the contribution of this factor towards the trends seen in Fig. 2, and recognize the limitations of the data. For passenger cars registered after 2012, the remote sensing measurement period spans their entire operational lifetime (up to and including 2022), and thus the data set contains measurements of these vehicles across a range of ages and conditions, reducing the influence of deterioration on the results.

The diesel emission factor values differ considerably from those of gasoline across all pollutants in Fig. 2, with the disparity being particularly prominent for NH_3 and NO_x. Gasoline NH_3 emissions were 0.025 to 0.078 $g\,km^{-1}$ higher than those of diesel fuel, which can be attributed to NH_3 formation in the three-way catalytic converter systems fitted exclusively to gasoline fuelled passenger cars (Tagliaferri et al., 1998; Heeb et al., 2006). The increase in the NH_3 emission factors for the 2019 model year is likely a statistical result arising from the low number of measurements for 2019 vehicles, owing to the missing ammonia remote sensing data from 2020 on-wards (Section 2.4). Conversely, NO_x emission factor values for diesel were 0.057 to 0.926 $g\,km^{-1}$ higher than those of gasoline. The rapid decline in the diesel NO_x emission factor from 0.635 $g\,km^{-1}$ in 2015 to 0.182 $g\,km^{-1}$ in 2022 (71.4%) corresponds to the introduction of NO_x specific exhaust after-treatment systems (SCR and LNT), adopted for diesel passenger cars to meet Euro 6 NO_x limits and on-road vehicle emission testing methods; see Fig. 1.

From the remote sensing data it is also possible to summarise information about changes in the fuel composition of a given property of the passenger car fleet as a function of model year. Fig. 3 presents the change in vehicle fuel type share from 1998 to 2022. In both the left combined plot panel and the right diesel plot panel the rise and subsequent decline of diesel fuel with increasing passenger car model year is apparent. From the model year 2001 to 2015 the share of new diesel registrations increased from 24.3% to 54.2%, before decreasing to 16.1% for the model year 2022.

The fuel-share statistics are generally consistent with statistics from the UK Department of Transport, which report the share of new diesel passenger car registrations to be 17.8%, 48.2%, and 11.5% for the model years 2001, 2015, and 2021 (the most recent year for which data are available) respectively (Department for Transport, 2022c). A distinct benefit of the remote sensing data is the direct measurements of vehicles in-use. Only passenger cars that are being actively driven will be measured by remote sensing, and so the resulting data set reflects the on-road vehicle fleet, as opposed to the total registered vehicle fleet represented in the DfT statistics. Diesel cars have higher annual mileage compared with gasoline cars, so their relative share is inflated in the on-road vehicle fleet compared to DfT statistics (Department for Transport, 2022a). However, remote sensing measurements are typically collected in urban areas, where there are fewer diesel vehicles compared to rural and motorway, which would result in a conservative estimate of diesel car numbers.

Gasoline passenger car fleet composition trends generally oppose those described for diesel fuel. However, for later model years the introduction of alternative fuel sources (FHEV, PHEV, and electric) results in a rate of gasoline share increase that is less than the rate of diesel share decrease. Whilst electric and hybrid passenger cars operating in full electric mode do not produce exhaust emissions they are still captured by the remote sensing camera equipment, and all vehicles observations are preserved in the composition data set regardless of emissions measurement validity. This approach is essential for ensuring that the reported trends are representative of the on-road vehicle fleet, and enables robust non-emissions related information such as that presented in Fig. 3 to be derived from over 600,000 on-road vehicle observations.

For each vehicle model year and pollutant, the average of the distance-based emission factors for each fuel type was calculated, weighted by the fuel type composition data. The solid black lines in Fig. 4 show these values as a function of model year for each pollutant, and present a combination of the previously discussed emission factor and fleet fuel type composition results. The shaded areas beneath the black lines display the contribution towards the average value from each

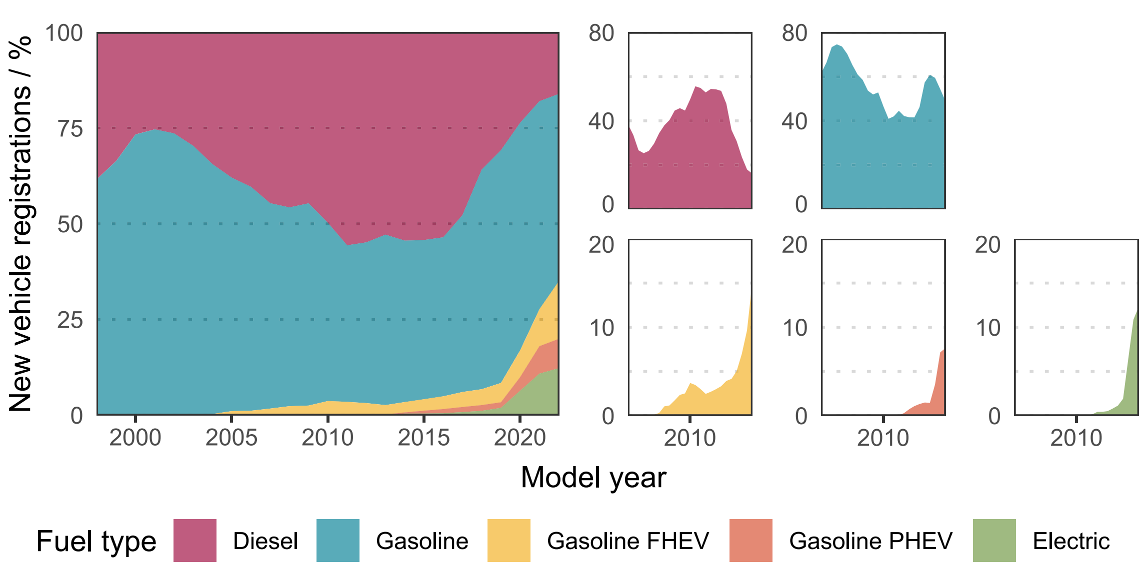

Fig. 3. Passenger car fuel type composition (%) as a function of vehicle model year (1998–2022). The combined total fuel type composition is displayed on the left and separate contributions from each fuel type are shown on the right.

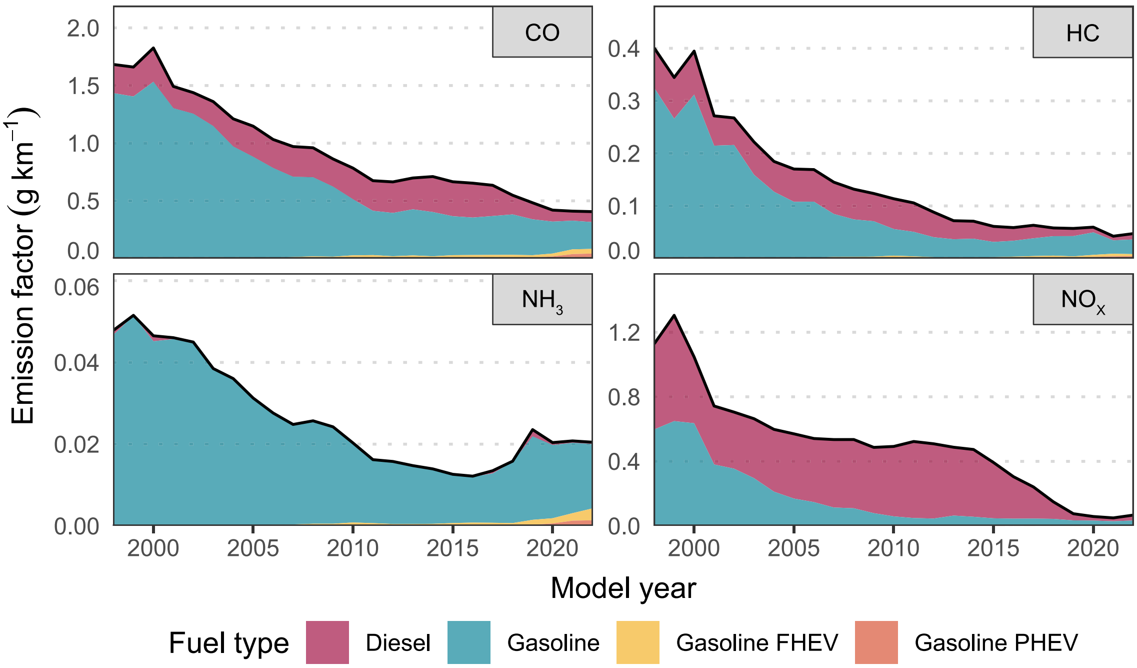

Fig. 4. Average passenger car distance-based emission factor values ($g\,km^{-1}$) as a function of model year for CO, HC, NH_3, and NO_x. The solid black line represents the average distance-based emission factor values and the shaded areas represent each fuel type's contribution towards the average.

fuel type, and assist in elucidating the driving factors behind the observed trends.

Similar results are shown for both the CO and HC timelines in Fig. 4; a general decrease in the average emission factor values of these pollutants is seen with increasing vehicle model year, resulting primarily from the decrease in the emissions of all fuel types seen in Fig. 2. Additionally, the relative contribution towards the average from gasoline fuel increases from the model year 2015 onwards, corresponding to the period of the demise of diesel fuel in which the relative share of gasoline passenger cars in the fleet increased. For NH_3, the average emission factor value is dominated by the contribution from gasoline vehicles for all model years, and the increase seen from the model year 2017 onwards can be attributed to the increase in the relative share of gasoline passenger cars in the fleet.

Diesel fuel contributions generally dominate the NO_x emissions following a period of growth between the model years 2001 to 2008. During this time the rate of decrease of diesel and gasoline NO_x emission factors was similar (66.1% and 59.3% respectively, Fig. 2), and thus this growth can be explained by the increase in the relative share of diesel passenger cars resulting from the promotion of diesel fuel. Another consequence arising from the increase in diesel vehicle share is the stagnation in the rate of the average NO_x emission factor decrease with increasing model year, seen between 2005 to 2014 in Fig. 4. During this period, despite both diesel and gasoline NO_x emission factors decreasing by 27.8% and 53.2% respectively, the average NO_x emission factor only decreased by 17.0%, because of the relative shift towards an increased proportion of newly registered diesel passenger cars. The implications of these results, which have been derived from on-road vehicle measurements, are very important: the rate of decrease of the average passenger car NO_x emission was diminished over a decade of vehicle model years, as a direct consequence of the promotion of diesel fuel in the UK.

Following this period of NO_x emission factor stagnation, the rapid decrease (from 0.149 $g\,km^{-1}$ to 0.067 $g\,km^{-1}$, 83.0%) between model years 2015 to 2022 seen in Fig. 4 is a result of the combination of two factors: a decrease in diesel vehicle NO_x emission factors resulting from the introduction of new exhaust after-treatment technology, and a decrease in relative diesel passenger car share due to reduced diesel fuel use. During this time, in addition to the decreasing average NO_x emission factor, the diesel contribution fell from 88.0% in 2015 to 49.9% in 2022. From the information reported, it is difficult to apportion the relative impact of reduced NO_x emissions and reduced use of diesel fuel.

3.2. Scenario comparison

To disentangle the influences of improving vehicle technology and changes in the proportion of vehicle fuel types, Section 3.2 explores three modelled scenarios and further evaluates the NO_x consequences of the promotion and demise of diesel fuel in the UK.

A NO_x emission factor timeline was produced for each scenario, shown in Fig. S2. Differences in the timeline average NO_x emission factor values between scenarios can be attributed entirely to the changes in fuel type composition, considering that the individual fuel type emissions data used was identical. This approach effectively enables the evaluation of NO_x in modelled situations where the development of vehicle technology is consistent with BAU, but either the promotion (NDP) or the demise (NDD) of diesel fuel in the UK did not occur. Fig. 5 shows the NO_x timelines for each scenario through two comparisons: BAU and NDP (left plot panel), and BAU and NDD (right plot panel).

The difference between the BAU and NDP scenarios (purple shaded region) represents the reduction in NO_x emission factors that could have been achieved had diesel fuel not been promoted in the UK, whereas the difference between the BAU and NDD scenarios (orange shaded region) represents the increase in NO_x emission factors that may have been reported had the demise of diesel fuel not transpired. Whilst the differences between scenarios within each comparison are a direct result of the modifications made to the fleet fuel type compositions to reflect different events, the disparity in the magnitude of these differences (and the sizes of the shaded regions) can be explained by the individual fuel NO_x distance-based emission factors shown in Fig. 2. For the BAU/NDP comparison, the difference in fuel type composition reflect the promotion of diesel and occur between model years 2001 to 2015, when diesel fuel NO_x emission factors are high. Conversely, for the BAU/NDD comparison, which reflects the demise of diesel, the difference in fuel type composition occurs between the model years 2015 to 2022, when diesel fuel NO_x emission factors are considerably lower due to effective

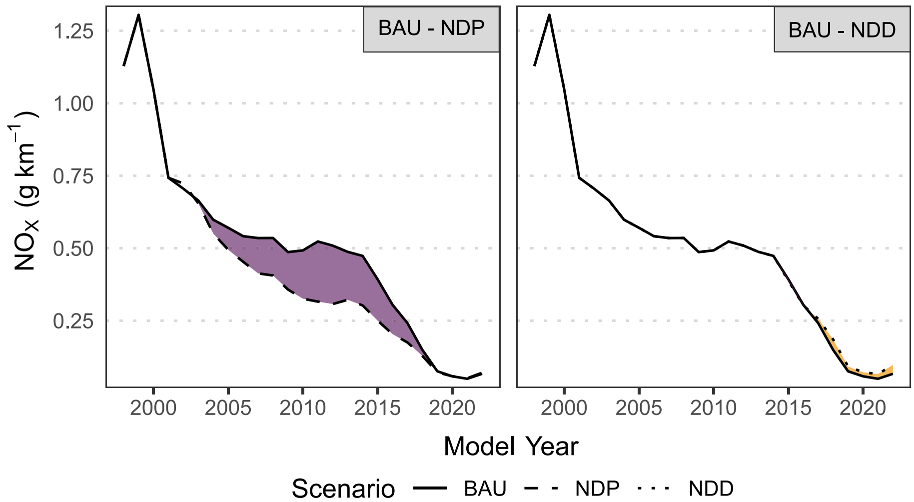

Fig. 5. Average passenger car distance-based emission factor values ($g\,km^{-1}$) as a function of model year for each scenario. The purple shaded section represents the difference between the 'Business as Usual' and 'No Diesel Promotion' (BAU-NDP) scenarios. The orange shaded section represents the difference between the 'Business as Usual' and 'No Diesel Demise' (BAU-NDD) scenarios.

aftertreatment systems. Therefore, the fuel type composition differences between BAU and NDP have a larger influence than those between BAU and NDD, and the variation in the resulting average NO_x emission factors seen in Fig. 5 is greater.

It is important to acknowledge that the development of vehicle technology is not entirely independent of events which influence vehicle fleet composition. The occurrence of Dieselgate is likely to have expedited the uptake of NO_x specific after treatment systems, resulting in post 2015 NO_x emission factors for a NDD scenario being higher than calculated in Fig. 5. However, the exact impact of Dieselgate on the development of vehicle technology has not been quantified, and was not considered in this analysis.

To further quantify and compare the relative NO_x impacts of the promotion and demise of diesel fuel, the estimated total lifetime NO_x emissions from all UK passenger cars with model years 1998 to 2022 was calculated for each scenario. These values were found to be 5480 kt, 4759 kt, and 5521 kt for BAU, No Diesel Promotion, and No Diesel Demise respectively. Fig. 6 presents this information as the difference between the two modelled scenarios and BAU, disaggregated into an annual time-series (lower panel) and cumulative total (upper panel). The difference between the BAU and NDP scenarios (purple shading) represents the additional NO_x emission as a result of the promotion of diesel, whereas the difference between the BAU and No Diesel Demise scenarios (orange shading) represents the reduction in NO_x emissions as

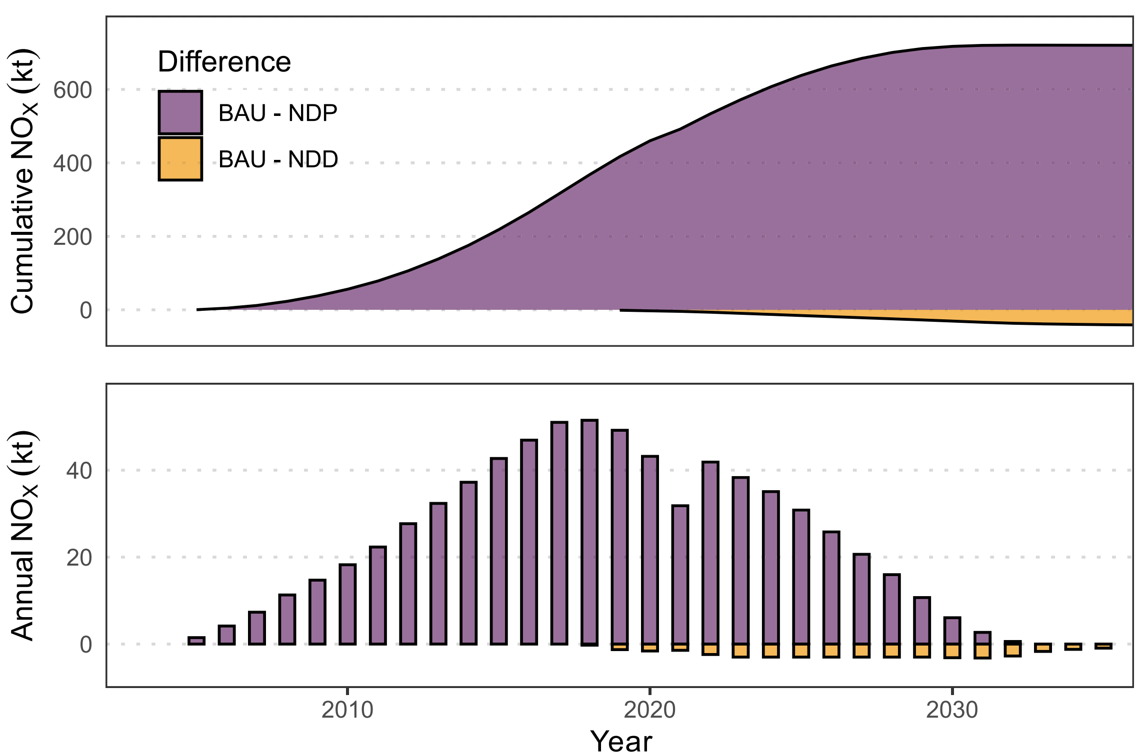

Fig. 6. Cumulative total and annual lifetime NO_x emission difference from passenger cars registered between 1998–2022 for the 'Business as Usual' and 'No Diesel Promotion' (BAU-NDP) scenarios (purple), and the 'Business as Usual' and 'No Diesel Demise' (BAU-NDD) scenarios (orange).

a result of the demise of diesel. The decrease in annual NO_x emissions for the years 2020 and 2021 is due to lower annual mileage values, arising from the travel restrictions imposed during the COVID-19 pandemic.

The cumulative effect over three decades of promoting diesel fuel on total emissions of NO_x is calculated to be 721 kt. To put this total into perspective, it is equivalent to 7.38 times the total NO_x emitted by all passenger cars in the UK in 2021 (NAEI, 2023). Indeed, it is greater than the total UK emissions of *all NO_x sources in 2021* of 681.8 kt.

The change in the total emission of NO_x is the principal impact that the growth in diesel use in passenger cars has had on air pollutant emissions. However, diesel vehicles are known to be important contributors to directly emitted (primary) NO_2. The use of diesel oxidation catalysts and particle filters (see Fig. 1) led to large increases in directly emitted NO_2 emissions, making an important contribution to ambient NO_2 concentrations (Carslaw et al., 2019). While the direct NO_2 emissions have not been estimated, the growth of diesel fuel use through the early 2000s would have led to considerably higher direct NO_2 emissions. The principal impact of directly emitted NO_2 is the contribution to local scale ambient NO_2 concentrations, which has been a major issue over the past two decades in Europe. Therefore, without diesel fuel promotion and growth, there would have been an additional benefit beyond the reduction in total NO_x emissions.

While the move away from diesel fuel in the wake of Dieselgate resulted in a small benefit in terms of NO_x emissions of 41 kt, there have been other consequences that could be important. As shown in Fig. 3, a consequence of a move away from diesel was increases in the use of gasoline, gasoline hybrids and BEV. The relative increase in gasoline and gasoline hybrids vehicles led to a reduction in NO_x emissions but an increase in NH_3, as shown in Fig. 4. Increased emissions of NH_3 would be expected to lead to increased concentrations of $PM_{2.5}$ through the formation of ammonium nitrate. The net impact of a reduction of NO_x emissions and an increase in emissions of NH_3 on concentrations of $PM_{2.5}$ is not known and would require a chemical transport model to establish effects on ambient $PM_{2.5}$ at a European scale. Nevertheless, the demise of diesel leading to increased vehicular NH_3 emissions is an unexpected outcome that requires further investigation.

4. Conclusions

The strong growth in diesel car use in the UK and Europe over the past two decades has been one of the most significant and profound factors influencing air quality from street level to a European scale. While there were likely benefits from policies encouraging diesel fuel from the perspective of carbon emissions, this benefit came at a significant cost to air quality, and in particular, urban air quality. It is clear that climate and air quality impacts of different policies should be considered together. Vehicle technologies that aim to reduce important air pollutants such as NO_x have changed considerably over the past three decades. The emissions from gasoline passenger cars have been effectively controlled (and further-refined) since the introduction of the three-way catalyst in 1992 in the UK. The situation for the control of air pollutants from diesel cars has evolved from very little control in the 1990s to the more recent use of complex, but effective aftertreatment systems such as selective catalytic reduction.

This work provides the first detailed evaluation and quantification of the impact that diesel car growth has had on the emission of important air pollutants. A novel and important component of the work is the use of extensive in-use vehicle emission measurements from comprehensive remote sensing measurements. These measurements underpin the quantification of 'real-world' emissions of pollutants including NO_x and NH_3. We identify two principal changes to diesel car use over the past two decades: the steady growth of diesel fuel use from the early 2000s for climate mitigation reasons and, the rapid decrease in diesel fuel use following the Dieselgate scandal in 2015. These two periods coincided with very different levels of NO_x emissions control from diesel passenger cars.

The finding that the promotion of diesel fuel use in passenger cars resulted in an excess emission of NO_x of 721 kt puts the impacts of these policies into context. This level of emission is over 7 times the total passenger car emissions of NO_x in the UK in 2021 and even exceeds UK total emissions of NO_x for the same year. Emissions of this magnitude were the result of the growth of diesel fuel use coinciding with a time when the control of diesel NO_x emissions was poor. Conversely, the rapid decrease in diesel fuel use has had a much lower impact than might have been expected (41 kt reduction in NO_x) because it occurred at a time when aftertreatment systems used to control diesel NO_x were effective.

The full implications of such a large excess NO_x penalty are not known but are wide in nature. The most direct effect would have been related to urban concentrations of NO_2, which have proved to be highly challenging to control across Europe since the early 2000s. Exceedances of air quality limits of NO_2 have been most strongly associated with diesel NO_x emissions and also the direct emission of NO_2, which is much higher from diesel–fueled vehicles compared with gasoline. Considering 2018 for example (the year with maximum NO_x contribution shown in Fig. 6), our results show that if diesel cars had not been promoted there would have been close to 20% reduction in road transport NO_x emissions. A decrease in emissions of NO_x (and directly emitted NO_2) would likely have meant that many locations in Europe would not have exceeded legal air quality limits for NO_2.

However, because emissions of NO_x are directly linked to a wide range of other air quality problems such as the formation of $PM_{2.5}$ and O_3, the impacts of diesel car growth would have extended well beyond urban air quality problems. An indication of the impacts of excess NO_x emissions can be gained from damage cost estimates that aim to quantify the human health and environmental impacts in terms of a financial amount; so-called externalities. The damage costs for NO_x include, for example, the impacts on human health associated with NO_x forming fine particulate matter ($PM_{2.5}$), which is estimated through the use of dispersion modelling and dose–response functions. In the UK for example, the damage cost estimates for NO_x are estimated to be £8,148 per tonne of NO_x (Defra, 2023; Brichby, 2023). Given the total excess NO_x emission estimate of 721 kt, the total damage cost is estimated to be £5.875 billion.

Finally, an unintended consequence of the rapid move away from diesel car use post-Dieselgate is the growth in the use of gasoline and gasoline hybrid vehicles. The remote sensing data used in the current study suggests that the net effect is an increase in emissions of NH_3 associated with three-way catalysts. As gasoline vehicles age, emissions of NH_3 increase, Farren et al. (2021) which suggests that NH_3 emissions in future will be higher than expected. Because NH_3 results in the formation of $PM_{2.5}$ and is strongly linked with problems associated with nitrogen deposition, this issue is of concern and deserves more attention.

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.

Data availability

Data will be made available on request.

Acknowledgements

We express our thanks to Ricardo and the Department of Chemistry at the University of York for funding the studentship of Sam Wilson. We would like to thank Adam Vaughan, Stuart Young, and Will Drysdale from the University of York for the collection of data using the FEAT instrument. Ricardo's remote sensing field team (Ben Fowler, Tom Green, Les Phelps, Sam Copsey, Paraic Marry, Sion Carpenter, and

Susannah Telfer) are thanked for collecting data using the Opus RSD5000.This study includes data from Sheffield which has been obtained from the CONOX database Borken-Kleefeld et al. (2018).

Appendix A. Supplementary material

Supplementary data associated with this article can be found, in the online version, at https://doi.org/10.1016/j.envint.2023.108330.

References

Anenberg, S., Miller, J., Henze, D., Minjares, R., 2019. A global snapshot of the air pollution-related health impacts of transportation sector emissions in 2010 and 2015. URL: www.theicct.org.
Bell, J., Honour, S.L., Power, S.A., 2011. Effects of vehicle exhaust emissions on urban wild plant species. Environ. Pollut. 159 (8–9), 1984–1990.
Bishop, G.A., Stedman, D.H., 1996. Measuring the emissions of passing cars. Accounts of chemical research 29 (10), 489–495.
Borken-Kleefeld, J., Bernard, Y., Carslaw, D., Åke Sjödin, Alt, G.-M., Fuente, J.D.L., Mcclintock, P., Gentala, R., Hausberger, S., Jerksjö, M., 2018. Contribution of vehicle remote sensing to in-service/real driving emissions monitoring-CONOX Task 3 report Commissioned by the Federal Office for the Environment (FOEN), Switzerland with contributions from James Tate. URL: www.ivl.se.
Brichby, D., 2023. Air Quality damage cost update 2023 – FINAL Report. Tech. rep., Ricardo Energy & Environment, accessed: 2023–08-01.
Burgard, D.A., Bishop, G.A., Stadtmuller, R.S., Dalton, T.R., Stedman, D.H., 2006. Spectroscopy applied to on-road mobile source emissions. Applied spectroscopy 60 (5), 135A–148A.
Cames, M., Eckard, H., 2013. Critical evaluation of the european diesel car boom - global comparison, environmental effects and various national strategies. Environmental Sciences. Europe 25.
Carslaw, D.C., Rhys-Tyler, G., 2013. New insights from comprehensive on-road measurements of NO_x, NO_2 and NH_3 from vehicle emission remote sensing in London. UK. Atmospheric Environment 81, 339–347.
Carslaw, D.C., Farren, N.J., Vaughan, A.R., Drysdale, W.S., Young, S., Lee, J.D., 1 2019.. The diminishing importance of nitrogen dioxide emissions from road vehicle exhaust. Atmos. Environ. X 1.
Commission, E., 2016. Official Journal of the European Union. Amending Regulation (EC) No 692/2008 as regards emissions from light passenger and commercial vehicles (Euro 6).
Comune di Milano, 2023. Mobility: Area B. https://www.comune.milano.it/aree-tematiche/mobilita/area-b, Accessed: 2023-10-18.
Davison, J., Bernard, Y., Borken-Kleefeld, J., Farren, N.J., Hausberger, S., Sjödin, Å., Tate, J.E., Vaughan, A.R., Carslaw, D.C., 2020. Distance-based emission factors from vehicle emission remote sensing measurements. Sci. Total Environ. 739, 139688.
Davison, J., Rose, R.A., Farren, N.J., Wagner, R.L., Murrells, T.P., Carslaw, D.C., 2021. Verification of a national emission inventory and influence of on-road vehicle manufacturer-level emissions. Environ. Sci. Technol. 55, 4452–4461, 4.
Davison, J., Rose, R.A., Farren, N.J., Wagner, R.L., Wilde, S.E., Wareham, J.V., Carslaw, D.C., 2022. Gasoline and diesel passenger car emissions deterioration using on-road emission measurements and measured mileage. Atmos. Environ.: X 14, 100162.
Defra, 2023. Air quality appraisal: damage cost guidance Updated 2 March 2023. https://www.gov.uk/government/publications/assess-the-impact-of-air-quality/air-quality-appraisal-damage-cost-guidance#damage-costs, accessed: 2023-07-31.
Department for Transport, 2022a. Vehicle Mileage and Occupancy NTS0901, Accessed: 2023-03-19. URL: https://www.gov.uk/government/statistical-data-sets/nts09-vehicle-mileage-and-occupancy.
Department for Transport, 2022b. Vehicle Licensing Statistics Data Tables VEH0150. https://www.gov.uk/government/statistical-data-sets/vehicle-licensing-statistics-data-tables, Accessed: 2023-03-19.
Department for Transport, 2022c. Vehicle licensing statistics data tables veh1153. Accessed:2023–05-04. URL: https://www.gov.uk/government/statistical-data-sets/vehicle-licensing-statistics-data-tables.
Dornoff, J., Rodriguez, F., 2019. Gasoline versus diesel: Comparing CO2 emission levels of a modern medium size car model under laboratory and on-road testing conditions. https://theicct.org/publication/gasoline-versus-diesel-comparing-co2-emission-levels-of-a-modern-medium-size-car-model-under-laboratory-and-on-road-testing-conditions/, Accessed: 2023-10-18.
Ebi, K.L., McGregor, G., 2008. Climate change, tropospheric ozone and particulate matter, and health impacts. Environ. Health Perspect. 116 (11), 1449–1455.
European Commission, 2022. Emissions in the automotive sector. https://single-market-economy.ec.europa.eu/sectors/automotive-industry/environmental-protection/emissions-automotive-sector_en, Accessed 2023-06-07.
European Council, 2008. Directive 2008/50/EC of the European Parliament and of the Council of 21 May 2008 on ambient air quality and cleaner air for Europe.
European Environment Agency, 2015. NEC Directive reporting status 2015. https://www.eea.europa.eu/publications/nec-directive-status-report-2015/nec-directive-reporting-status-2015, Accessed: 2023-06-07.
European Environment Agency, 2022. NEC Directive reporting status 2022. Accessed: 2023-06-07. URL: https://www.eea.europa.eu/publications/national-emission-reduction-commitments-directive-2022/national-emission-reduction-commitments-directive.
European Union Law, 1991. Council Directive 91/441/EEC of 26 June 1991 amending Directive 70/220/EEC on the approximation of the laws of the Member States relating to measures to be taken against air pollution by emissions from motor vehicles. https://eur-lex.europa.eu/legal-content/EN/ALL/?uri=CELEX%3A31991L0441, Accessed 2023-06-07.
European Union Law, 2016. Commission Regulation (EU) 2016/646 of 20 April 2016 amending Regulation (EC) No 692/2008 as regards emissions from light passenger and commercial vehicles (Euro 6) (Text with EEA relevance). https://eur-lex.europa.eu/eli/reg/2016/646/oj, Accessed 2023-06-07.
Farren, N.J., Davison, J., Rose, R.A., Wagner, R.L., Carslaw, D.C., 10 2021.. Characterisation of ammonia emissions from gasoline and gasoline hybrid passenger cars. Atmos. Environ. X 11.
Grange, S.K., Grange, S.K., Farren, N.J., Vaughan, A.R., Davison, J., Carslaw, D.C., 2020. Post-dieselgate: Evidence of noxemission reductions using on-road remote sensing. Environ. Sci. Technol. Lett. 7, 382–387, 6.
Hausberger, S., 2003. Simulation of Real World Vehicle Exhaust Emission, Vol. 82. Technische Universität Graz, Austria.
Heeb, N.V., Forss, A.-M., Brühlmann, S., Lüscher, R., Saxer, C.J., Hug, P., 2006. Three-way catalyst-induced formation of ammonia—velocity-and acceleration-dependent emission factors. Atmos. Environ. 40 (31), 5986–5997.
HM Treasury, 1998. Consultation on reform of Vehicle Excise Duty to ensure a cleaner environment. https://webarchive.nationalarchives.gov.uk/ukgwa/20090211145014/http:/www.hm-treasury.gov.uk/d/ConsultationOnReform.pdf, Accessed 2023-06-07.
HM Treasury, 2000. Budget 2000. https://www.gov.uk/government/uploads/system/uploads/attachment_data/file/265498/hc346.pdf, Accessed 2023-06-07.
Hoek, G., Krishnan, R.M., Beelen, R., Peters, A., Ostro, B., Brunekreef, B., Kaufman, J.D., 2013. Long-term air pollution exposure and cardio-respiratory mortality: A review. Environ. Health 12.
ICCT, 2017. Real-driving emissions test procedure for exhaust gas pollutant emissions of car and light commercial vehicles in europe. international council on clean transportation. https://theicct.org/publication/real-driving-emissions-test-procedure-for-exhaust-gas-pollutant-emissions-of-cars-and-light-commercial-vehicles-in-europe/.
Kroll, J.H., Ng, N.L., Murphy, S.M., Flagan, R.C., Seinfeld, J.H., 2005. Secondary organic aerosol formation from isoprene photooxidation under high-nox conditions. Geophys. Res. Lett. 32 (18).
NAEI, 2023. UK Informative Inventory Report (1990 to 2021). https://uk-air.defra.gov.uk/assets/documents/reports/cat09/2303151609_UK_IIR_2023_Submission.pdf, accessed: 2023-08-03.
Ngatia, L., Grace III, J.M., Moriasi, D., Taylor, R., 2019. Nitrogen and phosphorus eutrophication in marine ecosystems. Monitor. Marine Pollut. 1, 1–17.
Northern Ireland Statistics and Research Agency, 2022. Driver and Vehicle Agency Statistics. https://www.nisra.gov.uk/statistics/travel-and-transport/driver-vehicle-agency-statistics, Accessed: 2023-03-19.
Opus Inspection, 2023. Remote sensing device. https://www.opusrse.com/technology/remote-sensing-device/, Accessed: 2023-03-07.
République Français, 2022. Which vehicles can drive in Paris and the Greater Paris metropolis? https://www.service-public.fr/particuliers/actualites/A14948?lang=en, Accessed: 2023-10-18.
Rushton, C.E., Tate, J.E., Shepherd, S.P., Carslaw, D.C., 2018. Interinstrument comparison of remote-sensing devices and a new method for calculating on-road nitrogen oxides emissions and validation of vehicle-specific power. J. Air Waste Manage. Assoc. 68 (2), 111–122.
Selman, M., Greenhalgh, S., et al., 2010. Eutrophication: sources and drivers of nutrient pollution. Renew. Resour. J. 26 (4), 19–26.
SMMT, 2022. Automotive Sustainability Report - Average Vehicle Age. https://www.smmt.co.uk/industry-topics/sustainability/average-vehicle-age, Accessed: 2023-05-30.
Sullivan, J., Baker, R., Boyer, B., Hammerle, R., Kenney, T., Muniz, L., Wallington, T., 2004. Co2 emission benefit of diesel (versus gasoline) powered vehicles. Environ. Sci. Technol. 38 (12), 3217–3223.
Tagliaferri, S., Köppel, R., Baiker, A., 1998. Influence of rhodium-and ceria-promotion of automotive palladium catalyst on its catalytic behaviour under steady-state and dynamic operation. Appl. Catal. B: Environ. 15 (3–4), 159–177.
Transport for London, 2023. Ultra Low Emission Zone. https://tfl.gov.uk/modes/driving/ultra-low-emission-zone, Accessed: 2023-06-07.
Twigg, M.V., 2005. Controlling automotive exhaust emissions: Successes and underlying science. Philos. Trans. Roy. Soc. A: Math., Phys. Eng. Sci. 363 (1829), 1013–1033.
UK Goverment Asset Publishing Service, 2018. Road fuel consumption and the UL motor vehicle fleet. https://assets.publishing.service.gov.uk/government/uploads/system/uploads/attachment_data/file/812622/Road_fuel_consumption_and_the_UK_motor_vehicle_fleet.pdf, Accessed 2023-06-07.
UK Government, 2023. Uk vehicle excise duty rates. https://www.gov.uk/vehicle-tax-rate-tables/print, Accessed: 2023-02-17.
University of Denver, 2023. What is FEAT? https://digitalcommons.du.edu/feat/#:text=What%20is%20FEAT%3F,to%20as%20a%20remote%20sensor, Accessed: 2023-03-07.
Vehicle Certification Agency, 2022. Vehicle Type Approval. https://www.vehicle-certification-agency.gov.uk/vehicle-type-approval/, Accessed 2023-06-07.

WHO, 2021. World Health Organization. Air Quality Guidelines — Update 2021. WHO Regional Office for Europe, Copenhagen, Denmark.
Westerholm, R., Christensen, A., Rosén, Å., 1996. Regulated and unregulated exhaust emissions from two three-way catalyst equipped gasoline fuelled vehicles. Atmos. Environ. 30 (20), 3529–3536.
Zheng, M., Reader, G.T., Hawley, J.G., 2004. Diesel engine exhaust gas recirculation—-a review on advanced and novel concepts. Energy Convers. Manage. 45 (6), 883–900.