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Letter

# Post-Dieselgate: Evidence of $NO_x$ Emission Reductions Using On-Road Remote Sensing

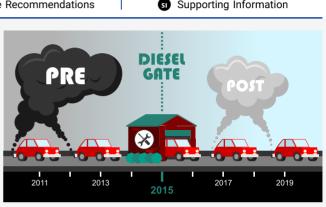
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 ABSTRACT: The Dieselgate scandal which broke in September
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**ABSTRACT:** The Disselgate scandal which broke in September 2015 demonstrated that vehicle manufacturers, such as the Volkswagen Group (VWG), engaged in software-based manipulation which led to vehicles passing laboratory-based emission testing limits but were far more polluting while being driven on roads. Using 23 000 on-road remote sensing measurements of light-duty Euro 5 diesel vehicles in the United Kingdom between 2012 and 2018, VWG vehicles with the "Dieselgate-affected" EA189 engine demonstrated anomalous NO<sub>x</sub> emission behavior between the pre- and post-Dieselgate periods which was not observed in other vehicle makes or models. These anomalous changes can be explained by voluntary VWG hardware and software fixes which have led to improved NO<sub>x</sub> emission control.



The VGW 1.6 L vehicles, with a simple hardware fix and a software upgrade, resulted in a 36% reduction in  $NO_{x^{1}}$  whereas the 2.0 L vehicles that required a software-only fix showed a 30% reduction in  $NO_{x}$  once controlled for ambient temperature effects. These results show that even minor changes or upgrades can considerably reduce  $NO_{x}$  emissions, which has implications for future emission control activities and local air quality.

# INTRODUCTION

Ambient nitrogen dioxide (NO<sub>2</sub>) concentrations around European roadside environments is a significant concern owing to the deleterious health effects this pollutant causes. In the past few decades, decreases in concentrations of  $NO_{2}$ , the regulated component of nitrogen oxides  $(NO_x)$ , have been achieved across Europe.<sup>1</sup> Despite this, hourly and annual legal limits imposed by the European Union (EU) 2008/50/EC Air Quality Directive are still breached by many countries.<sup>2,3</sup> This roadside NO<sub>x</sub> issue is not seen as a significant issue in other locations such as the United States (USA), Asia, Africa, or Oceania because it is driven primarily by diesel-powered vehicles. Europe has seen a much higher penetration of dieselpowered light-duty vehicles than other markets,<sup>4</sup> and these vehicles tend to emit greater amounts of NO<sub>x</sub> and NO<sub>2</sub> when compared to gasoline-fueled vehicles, primarily due to their lean combustion cycle.<sup>5</sup>

A discrepancy between laboratory and on-road  $NO_x$  emissions was first reported by the Center for Alternative Fuels, Engines & Emissions, West Virginia University, after a project investigating real-world  $NO_x$  emissions coordinated by the International Council on Clean Transportation (ICCT).<sup>6</sup> On September 18, 2015, the Volkswagen Group (VWG) was issued a Notice of Violation (NOV) by the U.S. Environmental Protection Agency (EPA) for the discovery of an illegal defeat device for vehicles powered by two liter diesel-fueled engines manufactured between 2009 and 2015.<sup>7</sup> VWG admitted to the

illegal defeat device in late September 2015, and the resulting political, legal, and environmental fallout is referred to as the "Volkswagen emissions scandal", "Dieselgate", or "emissions-gate". The impact of excess  $NO_x$  emissions has been extensively considered after September 2015, and legal actions are ongoing.<sup>8–11</sup> Despite Dieselgate's origin being the USA, the air quality consequences of excess  $NO_x$  emissions from light-duty diesel vehicles are much more relevant to Europe.<sup>4</sup>

The VWG defeat devices uncovered by Thompson et al.<sup>6</sup> interfered with NO<sub>x</sub> after-treatment systems, namely, selective catalytic reduction (SCR) and lean NO<sub>x</sub> traps (LNT). In the European market, these efficient after-treatment systems were not used on the equivalent vehicles because of the more lenient Euro 5 emission standards at the time of sale. Therefore, the European light-duty diesel-powered vehicles relied only on less effective Exhaust Gas Recirculation (EGR) for their NO<sub>x</sub> control. It would be expected, therefore, that any "fix" would result in less of an improvement in NO<sub>x</sub> emissions compared with vehicles in the USA. However, current knowledge of the

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effectiveness of any vehicle recalls and fixes in Europe on in-use  $NO_x$  emissions has not been established.

In the UK, different fixes were applied to the VWG 1598 and 1968 cc vehicles with the EA189 engine. In the case of 1598 cc engines, hardware and software fixes were required that consisted of a "flow transformer" fitted directly in front of the air mass sensor to stabilize the air flow and allow for more accurate detection of mass flow. In the case of the 1968 cc vehicles, only a software fix was required. All fixes in the UK were voluntary, but in other European Union (EU) member states such as Germany, France, and Austria, these recalls were mandatory.<sup>12</sup> These modifications are in stark contrast to the fixes required in the USA where replacement of NO<sub>x</sub> after-treatment systems were often required, along with particulate filters with strict durability requirements which had to be met.<sup>13</sup>

The primary objective of this work is to investigate lightduty Euro 5 diesel vehicles  $NO_x$  emissions for two distinct periods: before and after the Dieselgate scandal, hereafter referred to as pre- and post-conditions. Specifically, the principal aim is to quantify  $NO_x$  emission changes of affected VWG vehicles in comparison with emissions from other manufacturers. A comprehensive vehicle emission remote sensing data set collected in the UK is used to explore thousands of individual vehicle emission measurements for popular manufacturer groups.

# MATERIALS AND METHODS

**On-Road Remote Sensing.** The University of Denver's Fuel Efficiency Automobile Test (FEAT) remote sensing (RS) instrument was used to measure on-road vehicle  $NO_x$  emissions for all measurement campaigns.<sup>14</sup> The instrument measures ratios of NO and  $NO_2$  to  $CO_2$  in individual vehicle exhaust plumes, which can readily be expressed as fuel-dependent emission factors, for example, g kg<sup>-1</sup> of  $NO_x$ . Further, in-depth details about the FEAT RS instrument, measurement principles, monitoring setup, and calibration procedures can be found in previous publications.<sup>15–17</sup>

Measurements with the FEAT RS were taken in the north and south of England, UK, in 10 locations and between May 2012 and April 2018 (Figures SI 1 and 2; Table SI 1).<sup>16,18</sup> All data were processed to conform to a constrained data model resulting in a harmonized database.<sup>19</sup> The vehicle captures in the database were filtered to only contain type approval categories of M1 and N1 (passenger cars and light-duty vans, respectively) and Euro 5 diesel vehicles. These filters ensured that the vehicles which were in-service pre-Dieselgate were also in-service post-Dieselgate. The pre- and post-measurements were conducted under similar driving conditions. For example, the average Vehicle Specific Power (VSP) were 5.3 and 5.6 kW  $t^{-1}$ , respectively. The mean speed of the vehicles was 35.9 km  $h^{-1}$  with a minimum and maximum of 8 and 106 km  $h^{-1}$ . The same site was used in 76% of the post-measurements and 42% of the pre-measurements, providing consistency between the two periods.

For the VWG marques, Volkswagen, Audi, Škoda, and SEAT, the engines which displaced 1598 and 1968 cc were assumed to be of the EA189 engine family which were the focus of Dieselgate.<sup>20</sup> In the pre-period monitoring sessions, the EA189 engines would have been operating in their factory state, i.e., with any defeat devices active. However, by 2017 and 2018, many of these vehicles would have had fixes applied which involved new software and/or hardware to reduce their

on-road NO<sub>x</sub> emissions. With a random sample of 200 Volkswagen vehicles (only VW and not other marques) in the data set, it was found that 70% of passenger cars had this service action applied, while only 42% of vans had the fixes applied (as of mid-July, 2019). The self-service website VW provides was used for this process, and 149 cars and 51 vans were included in the sample.<sup>21</sup> The estimated proportion of updated vehicles is in good agreement with regular European Commission reporting of 70% as of March 2019 for all EA189 affected vehicles in the UK.<sup>12</sup>

Vehicle technical data such as make, model, engine size, date of manufacture, type approval category, mass, and Euro status were used to classify vehicles into "vehicle families" in a similar way described by Bernard et al.<sup>22</sup> Here, a vehicle family was defined as a combination of fuel type and Euro status (all vehicles were Euro 5 diesel, however), type approval category (passenger cars (M1) or vans (N1)), manufacturer group, and engine size (rounded to 0.1 L). There were a total of 142 vehicle families in the data set. Forty-four vehicle families were selected for ambient temperature modeling (discussed below) because 30 valid NO<sub>x</sub> measurements were required in each of the two time periods for the modeling process to occur (Table SI 2). For the makes or marques belonging to each manufacturer group, see Table SI 3.

Almost 23 000 valid  $NO_x$  emission measurements were analyzed with 13 600 measurements in the pre-Dieselgate period, while 9100 measurements were available from the post-Dieselgate period. The two monitoring periods were conduced in different mean temperature conditions with the pre-Dieselgate period being an average of 19.8 °C compared to 10.7 °C for the post-Dieselgate period (Figure SI 3).

Accounting for Ambient Temperature. Ambient temperature is an important driver of  $NO_x$  emissions for light-duty diesel vehicles<sup>17</sup> and to account for this effect, a Generalized Additive Model (GAM) was developed.<sup>23</sup> A GAM was chosen as a flexible modeling framework that can account for nonlinear responses, which are important in the case of ambient temperature and emission of  $NO_x$  and also interactions between covariates. The latter issue is important in the current context because the temperature response of  $NO_x$  emissions is not fixed but depends on the vehicle family being considered. A range of model formulations was developed, with the final model shown in eq 1:

$$E_{\text{NO}_x} = f(T_{\text{ambient}}) + f_{\text{vehicle}}(T_{\text{ambient}}) + d(\text{period}, \text{vehicle})$$
(1)

where  $E_{\text{NO}_x}$  is the fuel-specific emission of  $\text{NO}_x$ ,  $f(T_{\text{ambient}})$  is a smooth function of ambient temperature,  $f_{\text{vehicle}}(T_{\text{ambient}})$  are individual vehicle-specific smooth functions of ambient temperature, and d(period, vehicle) is the random effect interaction between period and vehicle. In this case, period takes the values for the pre- and post-Dieselgate periods. The models were developed for vehicle families with at least 200 measurements. This resulted in 26 vehicle families being included in the GAM modeling.

The model does not include a term for VSP because there were very similar mean VSP values in the pre- and post-Dieselgate monitoring periods. The model can be considered as a hierarchical GAM, as described in detail by Pedersen et al.<sup>24</sup> When using the models to control for ambient temperature, the mean temperature of the data set was used: 16 °C. Uncertainties, expressed as 95% confidence intervals in

Table 1. Descriptive Statistics for On-Road NO <sub>x</sub> Emissions for Three Manufacturer Groups' (BMW, GM, and VWG) Lig	ht-
Duty Euro 5 Diesel Vehicles for Pre- and Post-Dieselgate Time Periods <sup>4</sup>	

TAC	Man. group	Engine size (L)	Pre-Dieselgate (g kg <sup>-1</sup> )	Post-Dieselgate (g kg <sup>-1</sup> )	Delta (g kg <sup>-1</sup> )	Change (%)
M1	VWG	1.6	24.9 [23.9, 25.9]; $n = 591$	15.9 $[14.8, 17.0]; n = 437$	-9.0	-36.2
M1	VWG	2.0	17.0 [16.3, 17.7]; $n = 982$	13.3 [12.5, 14.1]; $n = 772$	-3.7	-21.8
M1	BMW	3.0	9.3 [8.7, 9.9]; $n = 390$	8.8 [7.6, 10.1]; $n = 144$	-0.4	-4.8
M1	BMW	1.6	12.2 [10.1, 14.3]; $n = 80$	12.1 [ 9.7, 14.4]; $n = 47$	-0.2	-1.2
M1	BMW	2.0	13.1 [12.4, 13.9]; $n = 694$	13.1 [12.2, 14.1]; $n = 422$	0.0	0.1
M1	GM	1.7	17.3 [15.2, 19.4]; $n = 139$	21.6 [18.9, 24.4]; $n = 75$	4.3	24.6
M1	VWG	3.0	16.5 [14.8, 18.1]; $n = 259$	25.6 [23.0, 28.1]; $n = 139$	9.1	55.1
M1	GM	2.0	16.1 [14.7, 17.6]; $n = 305$	31.2 [28.7, 33.7]; n = 191	15.0	93.2
N1	VWG	1.6	26.1 [24.7, 27.4]; $n = 252$	20.2 [18.4, 22.0]; $n = 160$	-5.9	-22.5
N1	GM	2.0	21.0 [19.0, 23.0]; $n = 200$	27.4 [24.7, 30.1]; $n = 133$	6.4	30.4
N1	GM	1.2	15.3 [12.7, 17.9]; $n = 48$	23.6 [19.4, 27.7]; $n = 60$	8.3	54.1
N1	VWG	2.0	17.2 [16.1, 18.4]; $n = 496$	26.4 [24.9, 27.9]; $n = 363$	9.2	53.1

<sup>a</sup>The summaries are ordered by type approval category (TAC) and mean change. The ranges represent 95% confidence intervals.

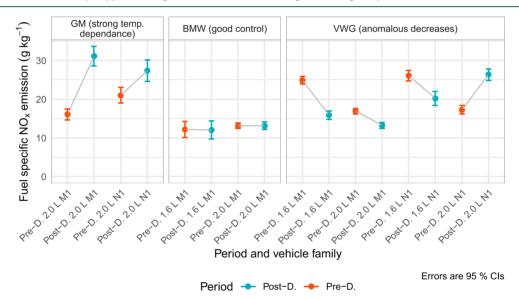


Figure 1. Mean  $NO_x$  emissions for 1.6 and 2.0 L Euro 5 diesel engines for three selected manufacturer groups for the pre- and post-Dieselgate (abbreviated as Pre-D. and Post-D.) time periods.

the mean for the predicted emissions were also obtained from the model.

# RESULTS AND DISCUSSION

Mean NO<sub>x</sub> Emission Changes. Mean NO<sub>x</sub> emissions for light-duty Euro 5 diesel vehicles showed considerably different responses across the two time periods when monitoring took place (pre- and post-Dieselgate) and across different vehicle families (Table 1). These simple before/after differences using the raw data show that in the case of passenger cars (M1), the VWG 1.6 L vehicles showed the greatest reduction in NO<sub>x</sub> of 36.2%. The second greatest reduction for passenger cars were the VWG 2.0 L vehicles with a NO<sub>x</sub> reduction of 21.8%. Similarly, for diesel vans (N1), the 1.6 L VWG vehicles had the greatest reduction in NO<sub>x</sub> of 22.5%. The results shown in Table 1 exhibit clear consistency between different manufacturer groups, as seen by the typically close proximity of different vehicle manufacturer models. For a larger table containing more manufacturer groups (a total of 44), see Table SI 4.

Table 1 also shows that  $NO_x$  emissions from the many vehicle families were higher in the second, post-Dieselgate,

monitoring period. This more  $NO_x$  polluting behavior can be explained by the low temperature  $NO_x$  emission penalty reported by Grange et al.<sup>17</sup> (with a similar data set), which results in higher  $NO_x$  emissions at lower temperatures.

Some vehicle families from a few manufacturer groups such as GM, RNA, and FCA, were found to emit approximately twice as much NO<sub>x</sub> in the cooler post-Dieselgate period compared to the pre-Dieselgate period (Table 1 and Table SI 4). The vehicle families which had consistently small changes to mean NO<sub>x</sub> emissions were BMW, Volvo, Tata, and PSA. Figure 1 shows 1.6 and 2.0 L engine sizes (very popular engine sizes in the UK) for three selected manufacturer groups demonstrating the different pre- and post-Dieselgate NOx emission behavior. GM vehicles demonstrated a strong ambient temperature response where NO<sub>x</sub> emissions were higher in the post-period owing to lower temperatures. BMW vehicles showed good NO<sub>x</sub> control where NO<sub>y</sub> emissions were low with little variation (when compared to other vehicle families) between the two time periods, despite the differences in ambient air temperatures for the two monitoring periods. VWG vehicles were anomalous however, with considerably lower NO<sub>x</sub> emissions for the post-Dieselgate period, with the notable exception of the 2.0 L N1 vehicles.

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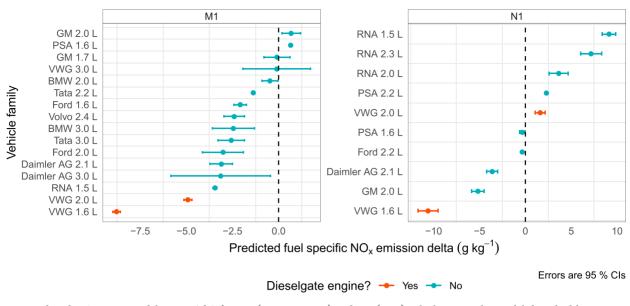


Figure 2. Predicted  $NO_x$  emission deltas at 16 °C for M1 (passenger cars) and N1 (vans) vehicles using the model described by eq 1.

All VWG vehicles, except for the N1 (vans) 2.0 L vehicle family, showed significantly *lower* mean NO<sub>x</sub> emissions for the post-Dieselgate compared to the pre-Dieselgate period (Table 1 and Figure 1). These decreases in mean NO<sub>x</sub> emissions for the post-Dieselgate periods are anomalous when considering the fleet as a whole and is interpreted as evidence that the VWG service actions, recalls, or fixes for vehicles embroiled in Dieselgate reduced on-road NO<sub>x</sub> emissions. Table 1 gives evidence that the VWG service actions have reduced on-road NO<sub>x</sub> emissions in the European market. There is evidence that the VWG fixes reduced fuel efficiency, and the Handbook Emission Factors for Road Transport (HBEFA) notes a reduction of 4% with limited data.<sup>25</sup> The fuel-specific emission factors presented in Table 1 and Table SI 4 are not adjusted for this potential effect.

It is important to note that the decrease in NO<sub>x</sub> emissions for the VWG Dieselgate vehicles would be an underestimation too, owing to only a proportion of vehicles being fixed between the two time periods ( $\approx$ 70%). The inconsistency concerning the VWG 2.0 L N1 vehicle family is interesting, however, and is most likely due to owners of these vehicles being less likely to submit their vehicle to be fixed because they are commercial vehicles. Without access to more detailed service actions by VWG, we cannot speculate further on what caused this inconsistency.

NO<sub>x</sub> Emissions and Ambient Temperature. To account for the strong ambient temperature dependency of NO<sub>x</sub> emissions for light-duty diesel vehicles, a GAM model (eq 1) was developed and used in a exploratory fashion for vehicle families with at least 200 measurements. Figure SI 4 shows smooth functions of NO<sub>x</sub> emissions against ambient temperature for the 26 vehicle families included in the modeling. For some manufacturers and engine sizes, there was a clear and strong temperature dependence on NO<sub>x</sub> emissions. Examples include GM and RNA vehicles. Conversely, there are some vehicle manufacturers and models that show very little if any temperature response, such as BMW. These results help explain some of the changes seen in Table 1 and Figure 1.

The model shown in eq 1 was used to predict  $NO_x$  emissions for specific vehicle models for the pre- and post-Dieselgate conditions for a *fixed* ambient temperature. The predictions were made for 16  $^{\circ}$ C, corresponding to the mean temperature of the full data set. The change in NO<sub>x</sub> emissions (post- minus pre-) is shown in Figure 2 for passenger cars (M1) and vans (N1).

For passenger cars, it is clear from Figure 2 that the two VWG vehicles with affected engines show the greatest reduction in NO<sub>x</sub> and that the reduction for the 1.6 L engine vehicle is considerably more than any other vehicle manufacturer or model. Compared with Table 1, the large increases in NO<sub>x</sub> seen, for example, for the GM 2.0 L vehicle of 15.0 g kg<sup>-1</sup> are dramatically reduced (to around 0.7 g kg<sup>-1</sup>, as shown in Figure 2) when temperature effects are accounted for. These results highlight the considerable importance of ambient temperature affecting emissions of NO<sub>x</sub> from some vehicle models and the need to control for temperature when quantifying changes in emissions.

There are few other studies that report the change in emissions owing to the software and hardware fixes implemented by the VWG in Europe. However, the ICCT summarizes tests from a few individual vehicles tested over a highway cycle.<sup>13</sup> The tests on two 1.6 L VW Golf vehicles showed a reduction of 55% and 28%. Smaller reductions were observed for the 2.0 L vehicles (six tests) with a median reduction of 12%. Even through these results are limited by sample size and tests over a variety of driving conditions, they are consistent with our findings that show the hardware and software fixes on the 1.6 L vehicles resulted in a greater improvement in the emissions of NO<sub>x</sub> compared with the software-only fix on 2.0 L vehicles.

On the basis of eight EA189-powered vehicles, HBEFA (version 4.1) gives updated Euro 5 emission factors for vehicles which have undergone software fixes.<sup>25,26</sup> NO<sub>x</sub> emissions were estimated to decrease by approximately 30%. Although this result is consistent with the analysis reported here, it does not differentiate between engine sizes or the different types of fixes applied.

The analysis in the current work shows that simple hardware and software fixes implemented on VWG vehicles after the Dieselgate scandal have led to improved  $NO_x$  emission control. Given the large share that these vehicles have in the UK market, the reductions in  $NO_x$  emissions are important to consider. Furthermore, given the wider popularity of these vehicles across Europe, the resulting emission reductions coincide with wider actions to reduce urban  $NO_x$  and  $NO_2$  concentrations through actions such as low emission or clean air zones. The "fix rate" across European countries for the EA189 engine varies widely from about 30% to over 90%,<sup>12</sup> which suggests there is scope for further  $NO_x$  reduction than has been achieved to date.

While these results are welcome from an air quality perspective, they also highlight the importance of other factors leading to excess emissions of NO<sub>v</sub>. It is clear, for example, that ambient temperature has a strong influence on light-duty vehicle NO<sub>x</sub> emissions, with the potential for substantially increased emissions under colder ambient temperatures. The analysis of emissions in the current work demonstrates that the effect of temperature varies widely among the different vehicle manufacturers and models, with some manufacturers demonstrating almost no low temperature penalty. For the vehicles with a strong temperature dependence, emissions of NO<sub>x</sub> can be increased beyond the changes brought about by the actions to fix VWG vehicles. In this respect, improved emission controls over a wider range of temperatures from more manufacturers would offer significant further reductions in NO<sub>x</sub> emissions in vehicle fleets throughout Europe.

## ASSOCIATED CONTENT

#### **Supporting Information**

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.estlett.0c00188.

Basic information for the 10 locations, sites, or roads where on-road remote sensing was conducted. The 44 vehicle families which were used in the analysis and the number of NO<sub>x</sub> measurements used for data analysis. The manufacturer groups which were included in the analysis along with their makes or marques. Descriptive statistics for on-road NO<sub>x</sub> emissions for 44 manufacturer groups' light-duty Euro 5 diesel vehicles for pre- and post-Dieselgate time periods. The three regions in England, United Kingdom, where on-road remote sensing was undertaken (source: naturalearthdata. com). Timeline and locations of when on-road remote sensing was undertaken. Air temperature distributions when on-road remote sensing monitoring was undertaken for two time periods. Generalized additive models (GAMs) predicting  $NO_x$  emissions based on air temperature for 26 vehicle families. (PDF)

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## Notes

The authors declare no competing financial interest.

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## REFERENCES

(1) Air Quality in Europe 2018; EEA Report No. 12/2018; European Environment Agency. https://www.eea.europa.eu//publications/air-quality-in-europe-2018 (accessed 24 April 2020).

(2) Exceedances of Air Quality Objectives Due to Traffic; European Environment Agency, 2016. http://www.eea.europa.eu/data-and-maps/indicators/exceedances-of-air-quality-objectives/exceedances-of-air-quality-objectives/exceedances-of-air-quality-objectives-9 (accessed 24 April 2020).

(3) Barnes, J. H.; Chatterton, T. J.; Longhurst, J. W. S. Emissions vs exposure: Increasing injustice from road traffic-related air pollution in the United Kingdom. *Transp. Res. D* **2019**, *73*, 56–66.

(4) Share of Diesel in New Passenger Cars; ACEA, European Automobile Manufacturers' Association, 2018. https://www.acea.be/statistics/article/Share-of-diesel-in-new-passenger-cars (accessed 24 April 2020).

(5) Heywood, J. B. Internal Combustion Engine Fundamentals; McGraw-Hill: New York, 1988.

(6) Thompson, G. J.; Carder, D. K.; Besch, M. C.; Thiruvengadam, A.; Kappann, H. K. *In-Use Emissions Testing of Light-Duty Diesel Vehicles in the United States*; Center for Alternative Fuels, Engines & Emissions, West Virginia University and International Council on Clean Transportation, 2014 https://theicct.org/publications/use-emissions-testing-light-duty-diesel-vehicles-us (accessed 24 April 2020).

(7) Brooks, P. A. VW Notice of Violation, Clean Air Act; U.S. Environmental Protection Agency, September 18, 2015. https://www.epa.gov/sites/production/files/2015-10/documents/vw-nov-caa-09-18-15.pdf (accessed 24 April 2020).

(8) Barrett, S. R. H.; Speth, R. L.; Eastham, S. D.; Dedoussi, I. C.; Ashok, A.; Malina, R.; Keith, D. W. Impact of the Volkswagen emissions control defeat device on US public health. *Environ. Res. Lett.* **2015**, *10*, 114005.

(9) Chossière, G. P.; Malina, R.; Ashok, A.; Dedoussi, I. C.; Eastham, S. D.; Speth, R. L.; Barrett, S. R. H. Public health impacts of excess  $NO_x$  emissions from Volkswagen diesel passenger vehicles in Germany. *Environ. Res. Lett.* **2017**, *12*, 034014.

(10) Anenberg, S. C.; Miller, J.; Minjares, R.; Du, L.; Henze, D. K.; Lacey, F.; Malley, C. S.; Emberson, L.; Franco, V.; Klimont, Z.; Heyes, C. Impacts and mitigation of excess diesel-related  $NO_x$  emissions in 11 major vehicle markets. *Nature* **2017**, *545*, 467–471.

(11) Dey, S.; Caulfield, B.; Ghosh, B. The potential health, financial and environmental impacts of dieselgate in Ireland. *Transport Plan. Techn.* **2018**, *41*, 17–36.

(12) State of Play of the Recall Actions Related to  $NO_x$  Emissions; European Commission, 2019. https://circabc.europa.eu/sd/a/ 8691fc9d-3924-4664-b407-6771d48d7a46/20190517 (accessed 24 April 2020).

(13) German, J. VW Defeat Devices: A Comparison of U.S. and EU Required Fixes; International Council on Clean Transportation, 2017. https://www.theicct.org/publications/VW-defeat-device-fixes-US-EU-comparison-dec2017 (accessed 24 April 2020).

(14) What's a FEAT?; Fuel Efficiency Automobile Test Data Center, University of Denver, 2011. http://www.feat.biochem.du.edu/ whatsafeat.html (accessed 24 April 2020).

(15) Bishop, G. A.; Haugen, M. J. The Story of Ever Diminishing Vehicle Tailpipe Emissions as Observed in the Chicago, Illinois Area. *Environ. Sci. Technol.* **2018**, *52*, 7587–7593.

(16) Carslaw, D. C.; Farren, N. J.; Vaughan, A. R.; Drysdale, W. S.; Young, S.; Lee, J. D. The diminishing importance of nitrogen dioxide emissions from road vehicle exhaust. *Atmos. Environ. X* **2019**, *1*, 100002.

(17) Grange, S. K.; Farren, N. J.; Vaughan, A. R.; Rose, R. A.; Carslaw, D. C. Strong Temperature Dependence for Light-Duty Diesel Vehicle  $NO_x$  Emissions. *Environ. Sci. Technol.* **2019**, *53*, 6587–6596.

(18) Carslaw, D. C.; Rhys Tyler, G. New insights from comprehensive on-road measurements of  $NO_{xy}$   $NO_2$  and  $NH_3$  from vehicle emission remote sensing in London, UK. *Atmos. Environ.* **2013**, *81*, 339–347.

(19) Grange, S. K. *emitr: Tools to Help with On-Road Vehicle Emission Data Analysis*, 2019. https://github.com/skgrange/emitr (accessed 24 April 2020).

(20) Technical Measures for the EA 189 Diesel Engines Affected Presented to the German Federal Motor Transport Authority; Volkswagen AG, Newsroom 11/25/2015. https://www.volkswagennewsroom.com/en/press-releases/technical-measures-for-the-ea-189diesel-engines-affected-presented-to-the-german-federal-motortransport-authority-1780 (accessed 24 April 2020).

(21) EA189 Service Action—VIN Check: A Self-Service Website, So That You Can Check If Your Vehicle Is Affected by the EA189  $NO_x$ Emissions Issue; Volkswagen, 2019. https://www.volkswagen.co.uk/ owners/emissionsinfo (accessed 24 April 2020).

(22) Bernard, Y.; Tietge, U.; German, J.; Muncrief, R. Determination of Real-World Emissions from Passenger Vehicles Using Remote Sensing Data: International Council on Clean Transportation and The Real Urban Emissions Initiative (TRUE), 2018. https://www.theicct.org/ publications/real-world-emissions-using-remote-sensing-data (accessed 24 April 2020).

(23) Wood, S. Generalized Additive Models: An Introduction with R, 2nd ed.; Chapman and Hall/CRC, 2017.

(24) Pedersen, E. J.; Miller, D. L.; Simpson, G. L.; Ross, N. Hierarchical generalized additive models in ecology: an introduction with mgcv. *PeerJ* **2019**, *7*, No. e6876.

(25) Notter, B.; Keller, M.; Althaus, H.-J.; Cox, B.; Knörr, W.; Heidt, C.; Biemann, K.; Räder, D.; Jamet, M. *HBEFA* 4.1—*Development Report*; INFRAS: Bern, 2019. https://www.hbefa.net/e/documents/ HBEFA41\_Development\_Report.pdf (accessed 24 April 2020).

(26) Matzer, C.; Weller, K.; Dippold, M.; Lipp, S.; Röck, M.; Rexeis, M.; Hausberger, S. *Update of Emission Factors for HBEFA*, Version 4.1, 2019. https://www.hbefa.net/e/documents/HBEFA41\_Report\_TUG\_09092019.pdf (accessed 24 April 2020).