Supplementary Material

DEFRA Emission Factor Toolkit

The DEFRA Emissions Factors Toolkit (EFT v8.0.1) (DEFRA, 2017) is used by local authorities in the United Kingdom (UK) to determine vehicle emissions. The EFT includes updated PM and NO_x emission equations and Euro 6 standards from the European Environment Agency (EEA) v5 COPERT (COmputer Program to calculate Emissions from Road Transport) emission factor model. To determine CO_2 and NO_x emission factors from the EFT, detailed hourly fleet composition from the ANPR data was inputted along with information on the region (not London), road type (A road), year (2016), traffic flow (corrected ANPR counts) and average speed (Figure S2). Euro class proportions for 2016 are automatically selected in the EFT and are based on the Department for Transport vehicle fleet composition projections (Base 2016) (DfT, 2016)

Supplementary Tables

Table S1: Number of licensed vehicles in the UK (units of 1000) at the end of 2016 (Source: DVLA/DfT https://www.gov.uk/government/collections/vehicles-statistics)

Vehicle Type	Petrol	Diesel	Hybrid Electric	Gas	Electric	Other	Total	% of total
Cars	18,825.2	12,574.3	322.3	36.0	34.1	0.4	31,792.3	82.8
Motorcycles							1,270.2	3.3
Light Goods Vehicles	130.5	3,745.3		7.8	5.3	0.8	3,889.7	10.1
Heavy Goods Vehicles		517.1					517.1	1.3
Buses and coaches							167.1	0.4
Other vehicles [1]							751.9	2.0
Total							38,388.2	

1. Includes rear diggers, lift trucks, rollers, ambulances, Hackney Carriages (i.e. taxis), three wheelers, tricycles and agricultural vehicles.

Table S2: Fraction of fuel type and vehicle type for the vehicles travelling through the Queensway tunnel from 06:00 to 19:59 (timestamp is the start of the hourly average). The last two columns show hourly mean $\Delta HONO/\Delta NO_x$ and $\Delta HONO/\Delta CO_2$ emission ratios measured at the sampling site inside the tunnel.

Start Hour	Diesel fraction	Non-Diesel fraction[a]	Car fraction[b]	LGV+HD fraction[c]	$\Delta HONO/\Delta NO_x$ (ppb/ppb)	$\Delta HONO/\Delta CO_2$ (ppb/ppm)
06:00	0.60	0.40	0.81	0.19	0.0071	0.033
07:00	0.58	0.42	0.82	0.18	0.0066	0.026
08:00	0.57	0.43	0.85	0.15	0.0075	0.027
09:00	0.62	0.38	0.83	0.17	0.0078	0.028
10:00	0.66	0.34	0.81	0.19	0.0091	0.033
11:00	0.63	0.37	0.81	0.19	0.0086	0.031
12:00	0.64	0.36	0.82	0.18	0.0088	0.029
13:00	0.61	0.39	0.84	0.16	0.0083	0.028
14:00	0.61	0.39	0.84	0.16	0.0079	0.026
15:00	0.58	0.42	0.87	0.13	0.0076	0.023
16:00	0.54	0.46	0.89	0.11	0.0078	0.021
17:00	0.54	0.46	0.93	0.07	0.0073	0.015
18:00	0.54	0.46	0.94	0.06	0.0095	0.021
19:00	0.56	0.44	0.94	0.06	0.0111	0.031

[a] petrol, biofuel and electric vehicles, [b] cars and other small vehicles (e.g. motor cycles), [c] light goods vehicles, heavy duty goods vehicles and buses

Supplementary Figures

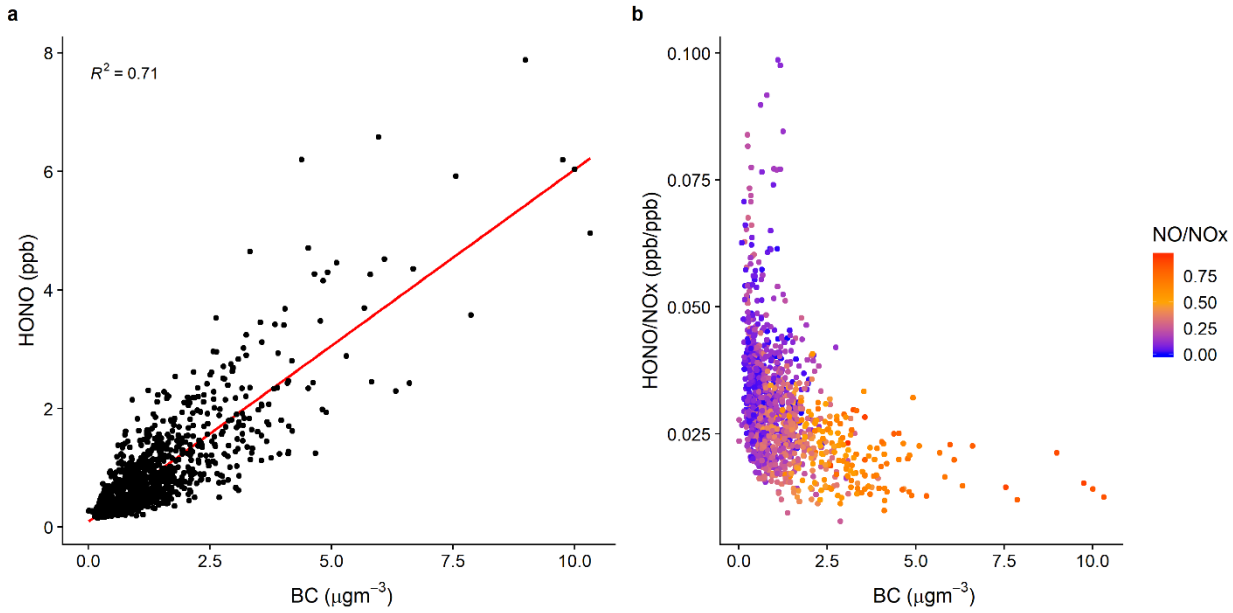

Figure S1: a) Hourly averaged BC vs HONO data taken at the North Kensington site in London in 2012 for the ClearfLo project (Bohnenstengel et al., 2015), b) BC vs HONO/NO_x coloured by the NO/NO_x ratio for the same period. High NO/NO_x ratios (yellow/red coloured points) are indicative of fresh pollution plumes and low chemical processing.

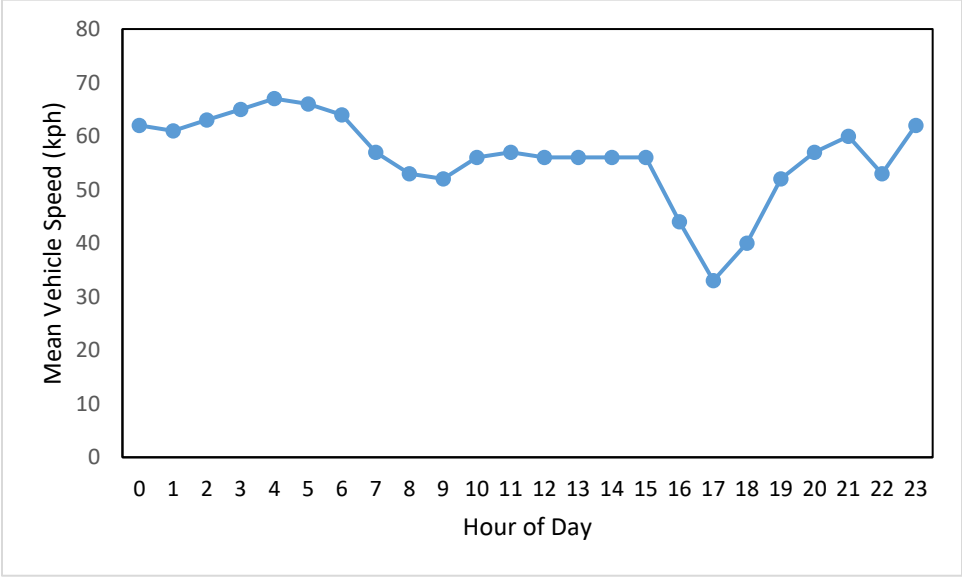

Figure S2: Hourly mean vehicle speed (kph) measured in the Southbound bore of the Queensway tunnel over 2015.

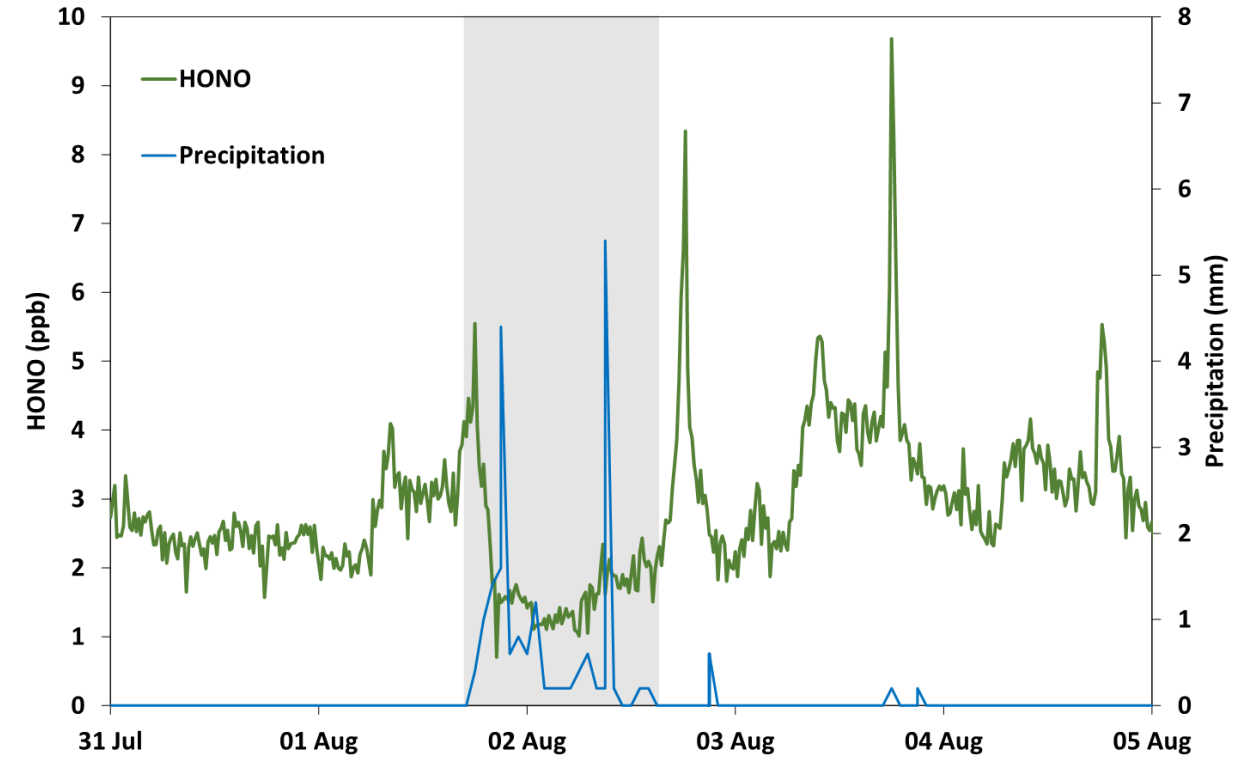

Figure S3:15 minute averaged HONO data sampled inside the tunnel (green) and hourly averaged precipitation data (blue) from the Winterbourne weather station from 29 Jul-08 Aug 2016. The shaded area represents the rainy period that was filtered from the final dataset (see main text for more details).

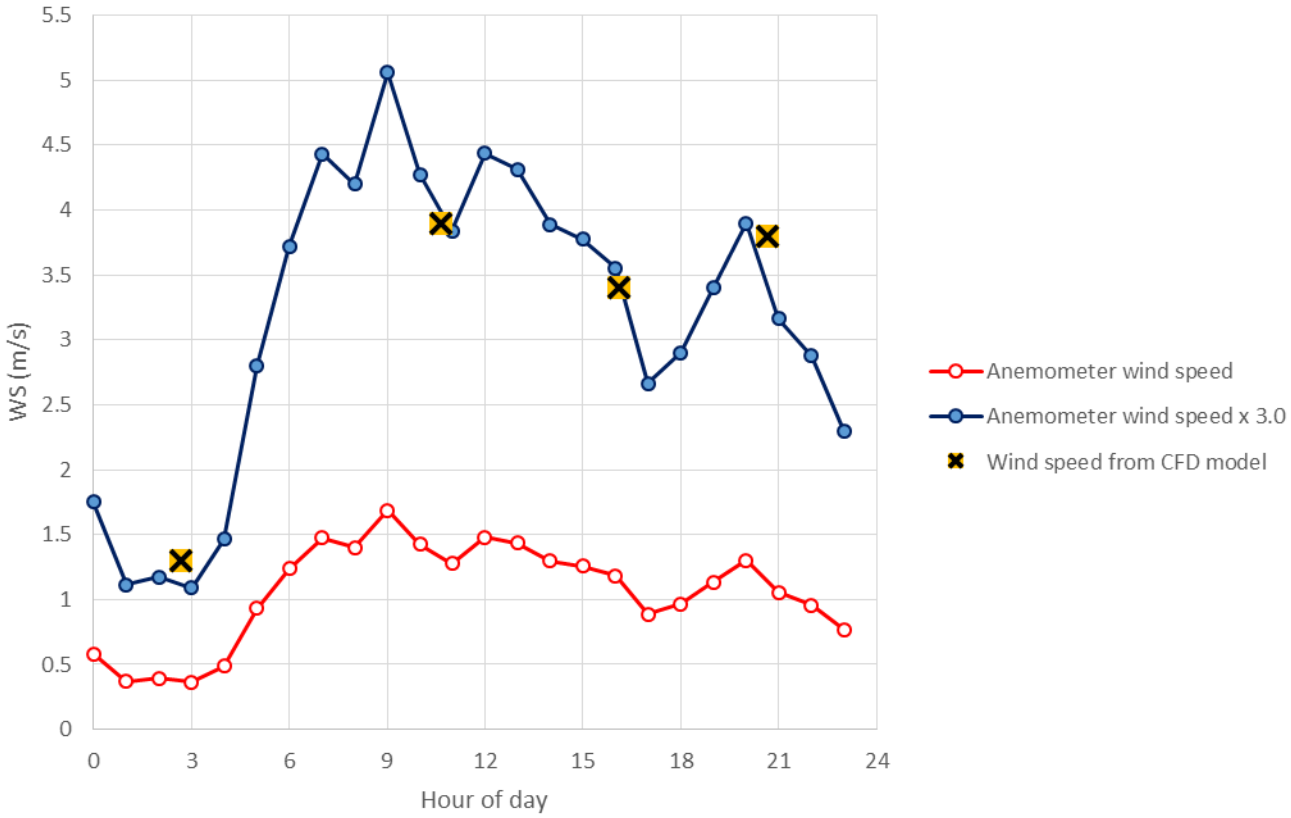

Figure S4:Diurnal profiles of wind speeds measured by the Kestrel anemometer (red time series) and wind speeds inferred from CFD modelling of CO_2 concentration profiles measured in the tunnel (black crosses on yellow background). The model wind speeds agree well with the anemometer wind speeds multiplied by a factor of 3.0 (blue time series). Note also the drop in wind speeds around 17:00-19:00 when traffic is congested inside the tunnel.

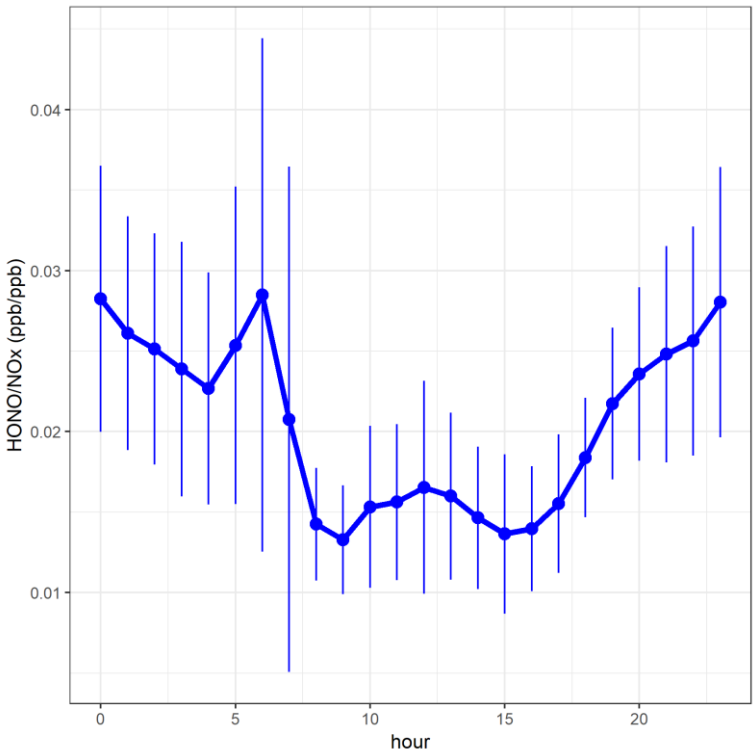

Figure S5: Average diurnal weekday HONO/NO_x cycle calculated from measurements taken at BAQS between March 18 and April 1, 2015.

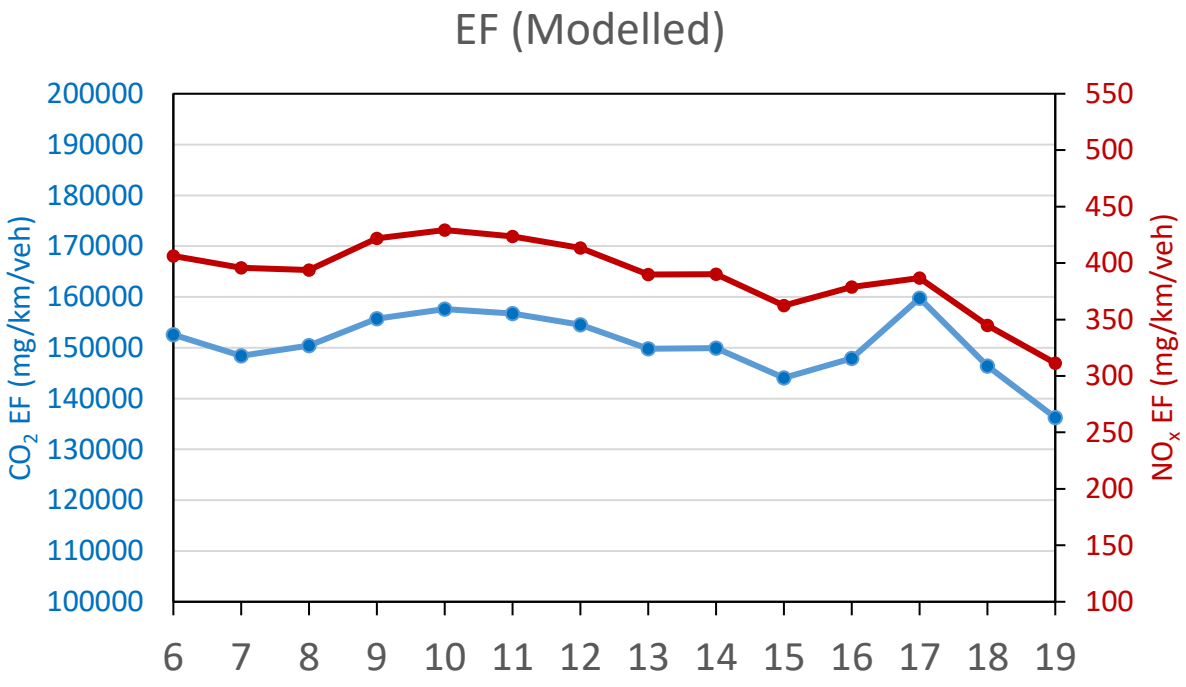

Figure S6: Hourly averaged CO_2 and NO_x emission factors calculated using the DEFRA Emissions Factors Toolkit (EFT v8.0.1) (DEFRA, 2017) for traffic flows in Queensway tunnel on weekdays from 06:00 to 19:00.

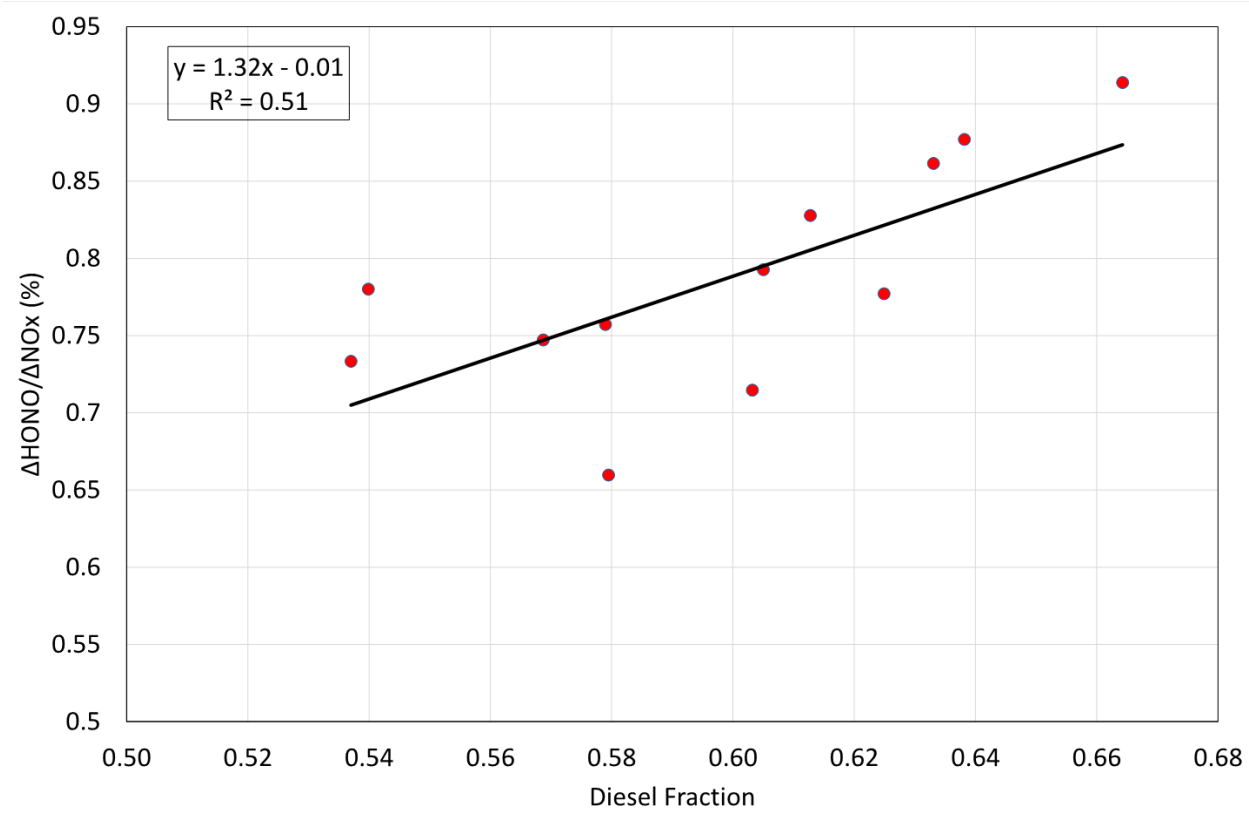

Figure S7: Linear regression analysis of hourly average diesel fraction vs ΔHONO/ΔNO$_x$ from 06:00 to 17:00.

References

Bohnenstengel, S.I., S.E. Belcher, A. Aiken, J.D. Allan, G. Allen, A. Bacak, T.J. Bannan, J.F. Barlow, D.C. Beddows, W.J. Bloss, A.M. Booth, C. Chemel, O. Coceal, C.F. Di Marco, M.K. Dubey, K.H. Faloon, Z.L. Fleming, M. Furger, J.K. Gietl, R.R. Graves, D.C. Green, C.S. Grimmond, C.H. Halios, J.F. Hamilton, R.M. Harrison, M.R. Heal, D.E. Heard, C. Helfter, S.C. Herndon, R.E. Holmes, J.R. Hopkins, A.M. Jones, F.J. Kelly, S. Kotthaus, B. Langford, J.D. Lee, R.J. Leigh, A.C. Lewis, R.T. Lidster, F.D. Lopez-Hilfiker, J.B. McQuaid, C. Mohr, P.S. Monks, E. Nemitz, N.L. Ng, C.J. Percival, A.S. Prévôt, H.M. Ricketts, R. Sokhi, D. Stone, J.A. Thornton, A.H. Tremper, A.C. Valach, S. Visser, L.K. Whalley, L.R. Williams, L. Xu, D.E. Young, and P. Zotter, 2015: Meteorology, Air Quality, and Health in London: The ClearfLo Project. *Bull. Amer. Meteor. Soc.*, **96**, 779–804, https://doi.org/10.1175/BAMS-D-12-00245.1

DEFRA, 2017. DEFRA, 2017: Emission Factors Toolkit for Vehicle Emissions. [WWW Document]. URL https://laqm.defra.gov.uk/review-and-assessment/tools/emissions-factors-toolkit.html (accessed 7.14.19).

DfT, 2016. Vehicle fleet composition projections (Base 2016) [WWW Document]. URL http://naei.beis.gov.uk/data/ef-transport.