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*Supplement of*

## **Nitrous acid (HONO) emissions under real-world driving conditions from vehicles in a UK road tunnel**

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### *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<sub>x</sub> 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<sub>2</sub> and NO<sub>x</sub> 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 vehicle speeds (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>)**

<b>Vehicle Type</b>	<b>Petrol</b>	<b>Diesel</b>	<b>Hybrid Electric</b>	<b>Gas</b>	<b>Electric</b>	<b>Other</b>	<b>Total</b>	<b>% of total</b>
<b>Cars</b>	18,825.2	12,574.3	322.3	36.0	34.1	0.4	31,792.3	82.8
<b>Motorcycles</b>							1,270.2	3.3
<b>Light Goods Vehicles</b>	130.5	3,745.3		7.8	5.3	0.8	3,889.7	10.1
<b>Heavy Goods Vehicles</b>		517.1					517.1	1.3
<b>Buses and coaches</b>							167.1	0.4
<b>Other vehicles</b> <sup>1</sup>							751.9	2.0
<b>Total</b>							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  $\Delta\text{HONO}/\Delta\text{NO}_x$  and  $\Delta\text{HONO}/\Delta\text{CO}_2$  emission ratios measured at the sampling site inside the tunnel.**

Start Hour	Diesel fraction	Non-Diesel fraction <sup>a</sup>	Car fraction <sup>b</sup>	LGV+HD fraction <sup>c</sup>	$\Delta\text{HONO}/\Delta\text{NO}_x$ (ppb/ppb)	$\Delta\text{HONO}/\Delta\text{CO}_2$ (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

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

Supplementary Figures

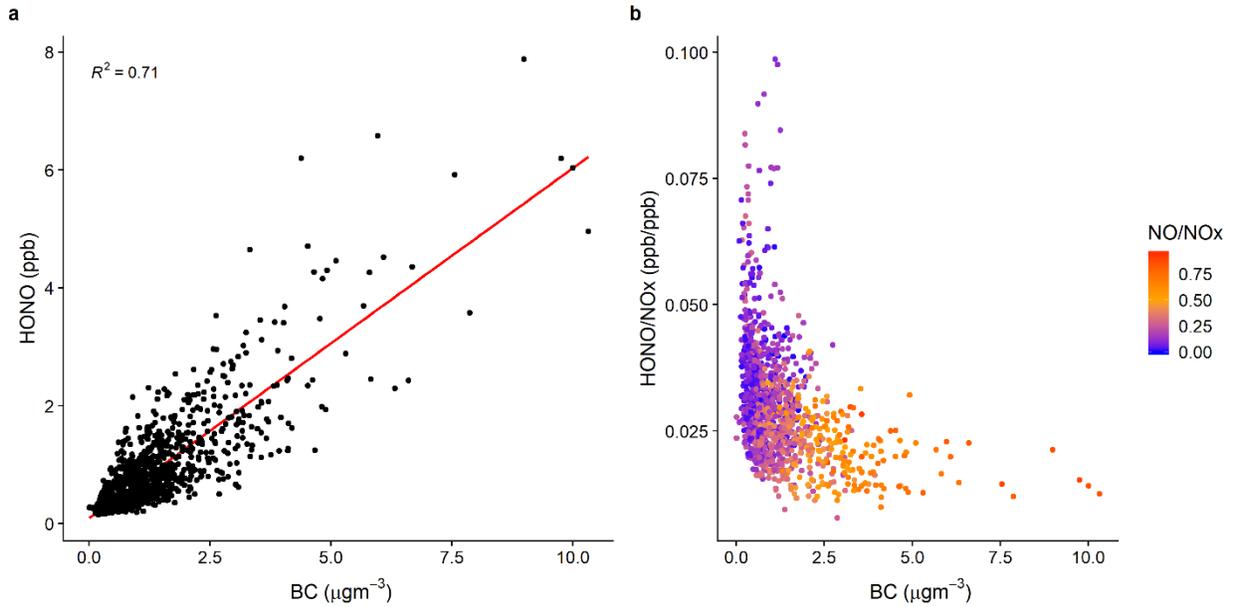


Figure S1: a) Hourly averaged BC vs HONO data taken at the North Kensington site in London in 2012 during the ClearLo project (Bohnenstengel et al., 2015), b) BC vs HONO/NO<sub>x</sub> coloured by the NO/NO<sub>x</sub> ratio for the same period. High NO/NO<sub>x</sub> ratios (yellow/red coloured points) are indicative of fresh pollution plumes and low chemical processing. There is no clear correlation observed here between BC and HONO/NO<sub>x</sub> for fresh pollution plumes. HONO formation via NO<sub>2</sub> conversion on BC, therefore, may not be a large source of HONO after emission from the vehicle exhaust.

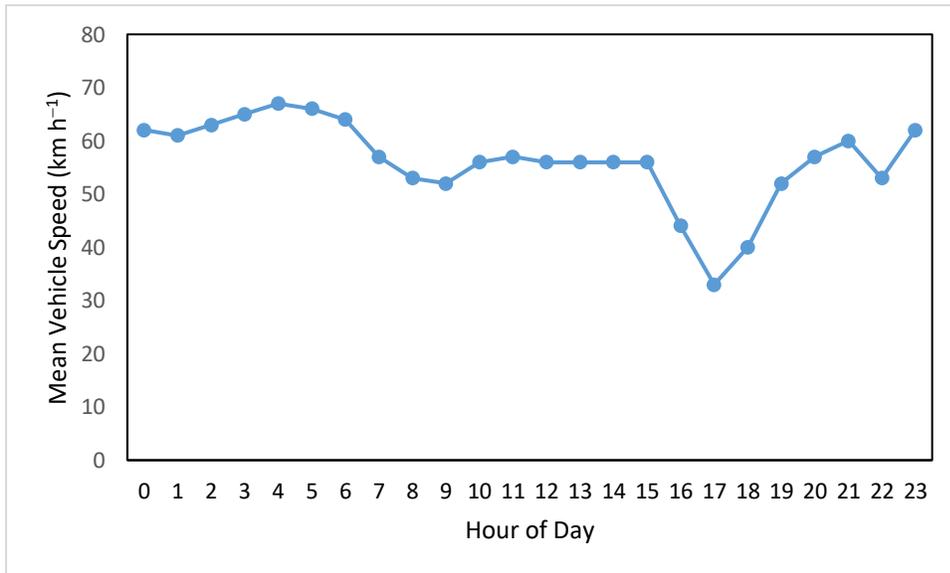
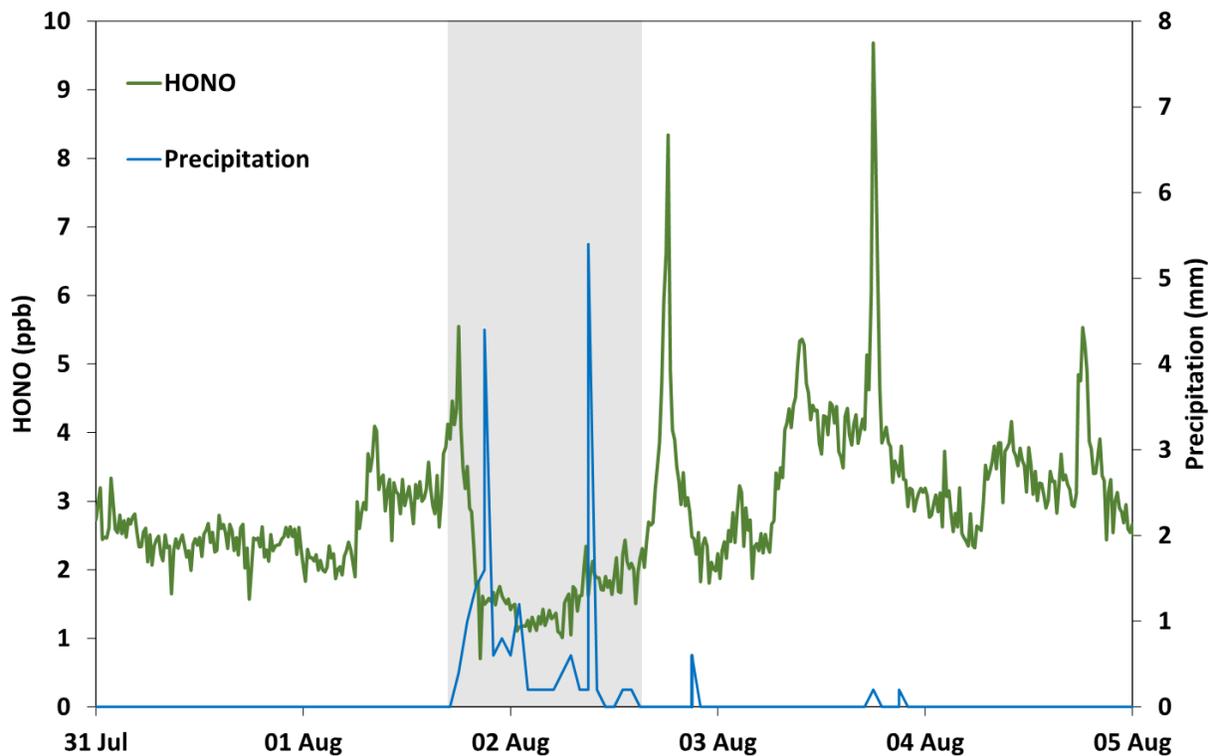
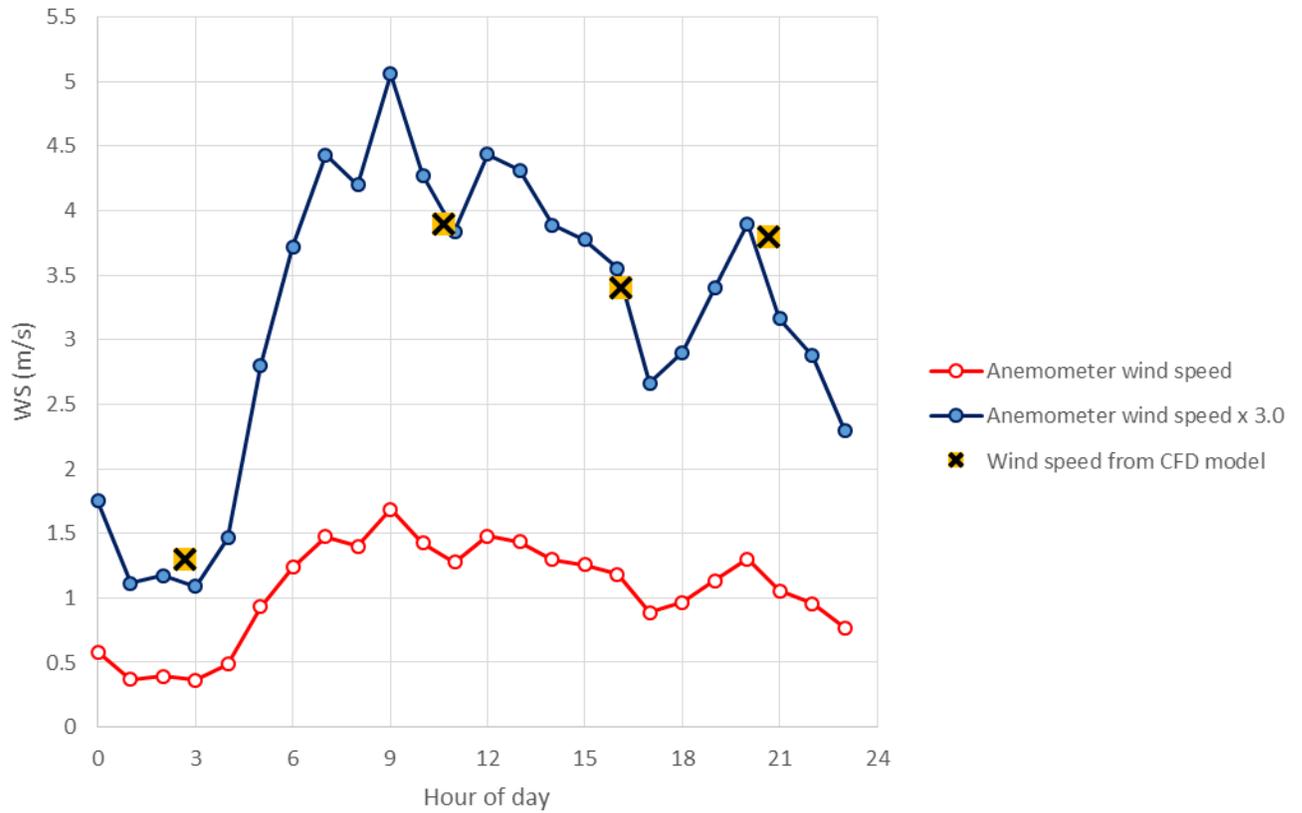


Figure S2: Hourly mean vehicle speed ( $\text{km h}^{-1}$ ) measured in the Southbound bore of the Queensway tunnel over 2015.



**Figure S3:** An extract of 15 minute averaged HONO data sampled inside the tunnel (green) and hourly averaged precipitation data (blue) from the Winterbourne weather station from 00:00 on 31 July to 00:00 on 05 Aug 2016. The shaded area represents the rain-affected period that was excluded 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<sub>2</sub> 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 to 19:00 when traffic flow is congested inside the tunnel.**

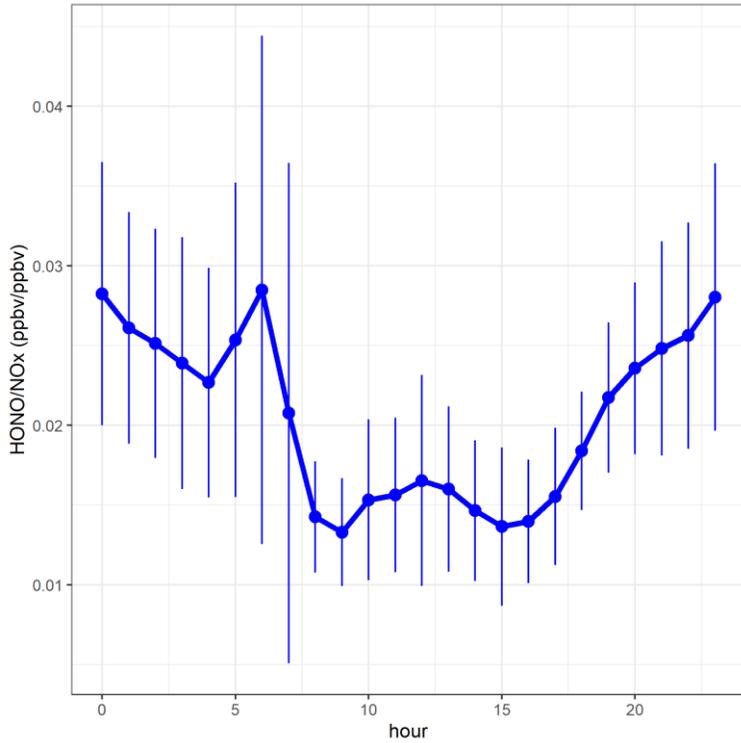


Figure S5: Average diurnal weekday HONO/NO<sub>x</sub> cycle calculated from measurements taken at BAQS between 18 March and 1 April, 2015. Error bars represent 1- $\sigma$  standard deviation of the mean.

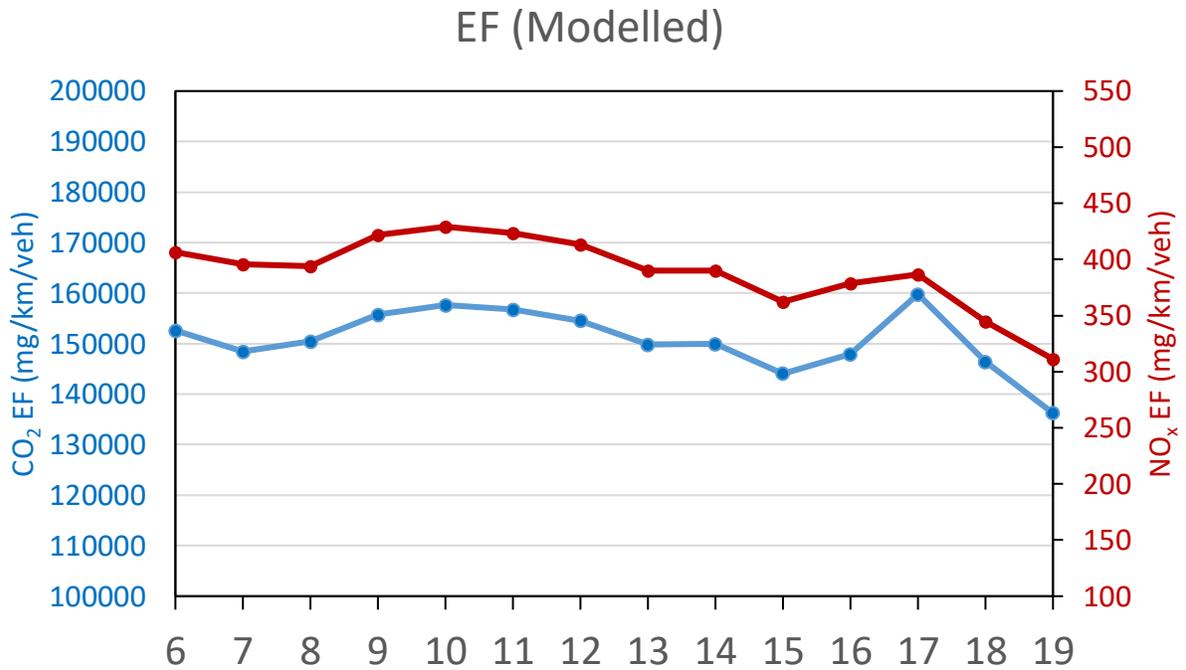
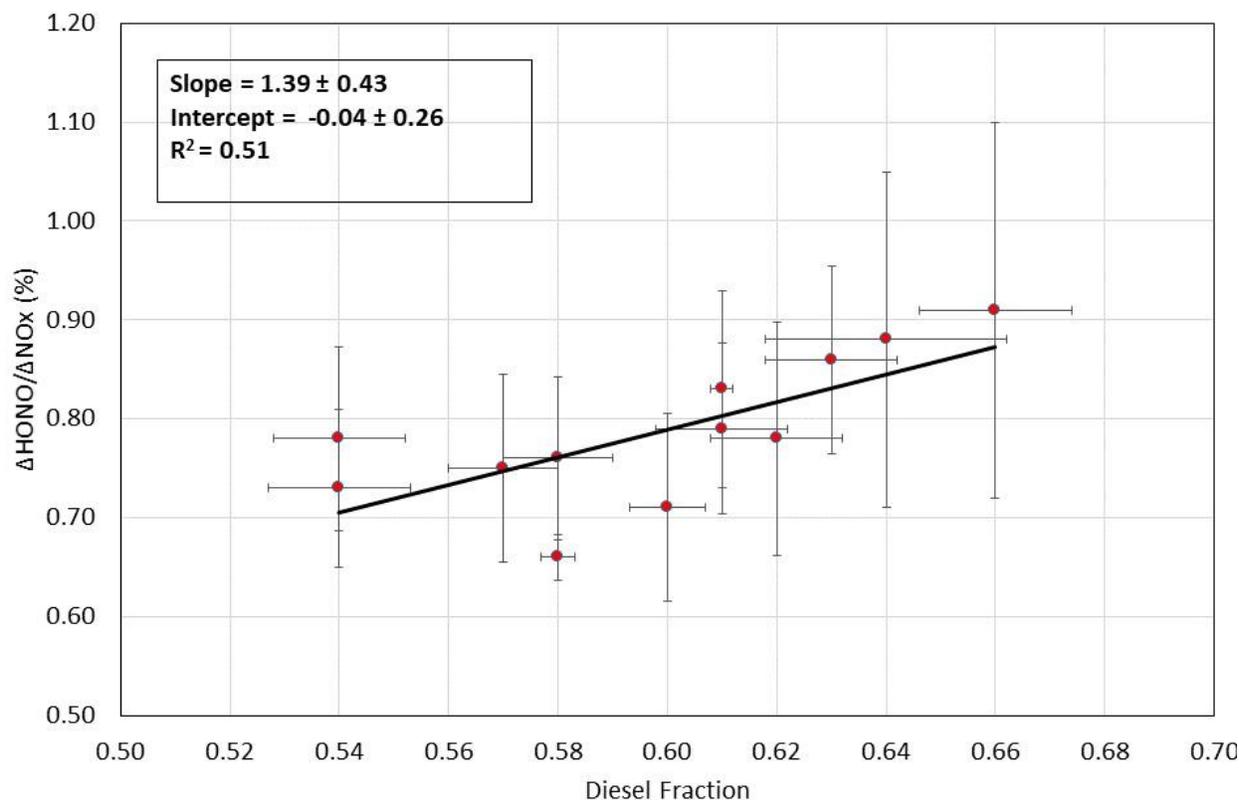


Figure S6: Hourly averaged CO<sub>2</sub> and NO<sub>x</sub> emission factors calculated using the DEFRA Emissions Factors Toolkit (EFT v8.0.1) (DEFRA, 2017) for traffic flows in Queensway tunnel on weekdays from 06:00 to 19:00.



**Figure S7: Linear regression analysis of hourly average diesel fraction vs  $\Delta\text{HONO}/\Delta\text{NO}_x$  from 06:00 to 17:00 based on the data from Table S2. Error bars represent 1- $\sigma$  standard deviation of the mean.**

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