Changing primary NO$_2$ emissions and validation from tunnel studies


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**NOX emission legislation (LDV)**

- **70/220/EEC, 1971**
- **74/290/EEC, 1975**
- **77/102/EEC, 1977**
- **80/85/EEC, 1979**
- **83/351/EEC, 1984**
- **88/76/EEC, 1991**
- **88/436/EEC, 1992**
- **91/441/EEC, 1992**
- **98/69/EC, 2000**
- **98/69/EC, 2005**
- **98/69/EC, 2008**
- **99/45/EC, 1992**
- **94/12/EC, 1996**
- **98/69/EC, 2000**
- **98/69/EC, 2005**
- **98/69/EC, 2008**

**Diagram**

- **HC+NOx (g/km)**
- **Petrol**
- **Diesel**
Recent trends in background & roadside NO\textsubscript{X} and NO\textsubscript{2}
Primary NO$_2$ emission data sources

- TRAMAQ/HA primary NO$_2$ emission measurements (2002), TRL
- HGV TRAMAQ emission tests, TRL
- DfT funded measurement programmes post 2002, Millbrook, Ricardo, Perkins & Shell
- European measurements at TNO, EMPA & LAT
- TfL measurements (bus, taxis and LCV)
- Tunnel studies, TRL
NO$_2$ formation and importance of sampling protocol

- High temperature NO$_X$ (especially lean diesel)
- Exhaust after-treatment related NO$_2$
  - Oxidation catalysts
  - CRT
- Chemical NO$_2$ (reactions with ambient O$_3$, and free radicals, which can catalyse the oxidation of emitted hydrocarbons.
- Low temperature NO$_2$ (exhaust cool down in CVS, bag and sample lines)
- NO$_2$ decomposition (reactions in lines and bag with UV)
## TRL 2002 emission survey

- 18 vehicles which broadly covers the range of road vehicles used in the UK, comprising of:
  - 3 Small Cars, 3 medium cars, 3 large cars, 3 Vans, 3 HGVs and 3 Buses.
  - Each set of 3 comprised of a pre-Euro I, Euro I and Euro II vehicle.
  - Cars and Vans driven over a ECE 15 test, 2 urban, one rural and tests at 2 constant higher speeds resembling motorway driving

### Cars and Vans driven over 5 types of cycle
- ECE 15 test +EUDC
- Warren spring suburban & rural
- Warren spring rural and urban
- Warren spring motorway 90 & 113 kph
- Warren spring congested urban cycle

### HGV & buses
- Vehicle version of the 13-mode test for HGVs
- Vehicle version of the FIGE test for HGVs
- Millbrook London bus cycle for buses
Sampling techniques

- Raw unheated CLD
- Raw heated CLD
- Dilute heated CLD
- Dilute heated FTIR
- Raw heated FTIR
• Significant relationship between NO$_2$ and fuel type, cycle speed but not test temperature or sampling method.
• NO$_2$ from diesels are higher than petrol vehicles.
• Pre-Euro I vehicles emit highest NO$_2$ followed by Euro I then Euro II however NO$_2$/NO$_x$ ratio is independent of legislation.
• Insufficient data to modify %5 EF.
TRAMAQ HGV emissions (Euro I to Euro III)

Vehicle number

0.0 0.1 0.2 0.3 0.4 0.5 0.6
NO2/NOx ratio

LNG & oxidation catalyst

Particle trap & oxidation catalyst

LPG

Particle trap

Particle trap
The primary NO₂ percentage of some emerging diesel technologies plus data for Euro III petrol, diesel and GDI technology.
<table>
<thead>
<tr>
<th>Fuel</th>
<th>Legislative category</th>
<th>Sample size and technology</th>
</tr>
</thead>
<tbody>
<tr>
<td>Petrol</td>
<td>Euro 0</td>
<td>7 various technologies, carburator, multi/monopoint,</td>
</tr>
<tr>
<td></td>
<td>Euro 3</td>
<td>7 Port injection Three Way Catalyst</td>
</tr>
<tr>
<td></td>
<td>Euro 4</td>
<td>10 Port Injection Three Way Catalyst, 2 Gasoline Direct Injection</td>
</tr>
<tr>
<td>Diesel</td>
<td>Euro 0</td>
<td>7 diesel</td>
</tr>
<tr>
<td></td>
<td>Euro 1</td>
<td>7 diesel</td>
</tr>
<tr>
<td></td>
<td>Euro 2</td>
<td>7 diesel with oxidation catalyst</td>
</tr>
<tr>
<td></td>
<td>Euro 3</td>
<td>16 diesel with oxidation catalyst, 1 Fuel Borne Catalyst Diesel Particle Filter</td>
</tr>
<tr>
<td></td>
<td>Euro 4</td>
<td>2 diesel with oxidation catalyst, 1 Fuel Borne Catalyst Diesel Particle Filter, 3 Catalysed Diesel Particle Filter, 1D-KAT (combined oxidation /NOx storage catalyst and particle filter) all with oxidation catalyst</td>
</tr>
</tbody>
</table>
The NO and NO$_2$ emission of various technologies and legislative categories

Gense et. al. (2006)
The NO₂ percentage for various technologies and legislative categories

Gense et. al. (2006)
Tunnel validation studies

- Primary NO$_2$ EF validation study, funded by TRL, using inverse modeling
  - Hatfield Tunnel A1(M), 100,000 veh/day, summer & winter 2005.
  - Bell Common M25, 180,000+ veh/d, summer 2006.
  - Winter 2006?
Pollutant concentrations increase along the length of a tunnel as the emissions from the traffic accumulate. An average emission factor for a pollutant $i$ and all the traffic passing through a tunnel during a time period $t$ can therefore be derived as follows:

$$EF_i = \frac{(C_{i, exit} - C_{i, entrance}) \cdot v_{air} \cdot t \cdot A}{L \cdot N}$$

where:

- $EF_i$ = total emission factor for pollutant $i$ (g vehicle$^{-1}$ km$^{-1}$)
- $C_{i, exit}$ = concentration of pollutant $i$ at tunnel exit (g m$^{-3}$)
- $C_{i, entrance}$ = concentration of pollutant $i$ at tunnel entrance (g m$^{-3}$)
- $v_{air}$ = velocity of the air in the tunnel (m s$^{-1}$)
- $t$ = time duration of sampling (s)
- $A$ = tunnel cross-sectional area (m$^2$)
- $L$ = tunnel length (km)
- $N$ = number of vehicles passing during time $t$
Measurements in Hatfield tunnel

- North tunnel portal
  - Tunnel ‘entrance’ site
    - NO, NO₂, O₃
  - Tunnel ‘mid’ site
    - NO, NO₂, O₃
    - NOMAD met station
  - Tunnel ‘exit’ site
    - NO, NO₂, O₃

- South tunnel portal

Additional notes:
- Sampling sites
- Hard shoulder
- Walkway

Distance markers:
- 46m
- 550m
- 472m
- 50m
- 18m
- 6m
Measurements

- Measuring NO, NO₂, NOₓ, O₃, wind speed, wind direction, temperature, relative humidity, traffic flows, composition and speed

- Two tunnels: Summer & winter campaigns
The proportion of NO\textsubscript{x} emitted as primary NO\textsubscript{2} was estimated from three models, each involving a different assumption regarding the formation of NO\textsubscript{2}:

**Model M1**: In this model, it was assumed that O\textsubscript{3} was conserved. In other words, no NO\textsubscript{2} formation reactions occurred in the tunnel air, and all NO\textsubscript{2} measured at the tunnel middle and exit was derived solely from vehicle exhaust.

**Model M2**: In this model, it was assumed that NO\textsubscript{2} in the tunnel air was derived from both vehicle exhaust and via the reaction of NO with O\textsubscript{3}.

**Model M3**: In this model, it was assumed that, as well as the NO\textsubscript{2} production mechanisms used in model M2, NO\textsubscript{2} was also formed by the reaction of NO with O\textsubscript{2}. The two NO\textsubscript{2} formation reactions proceed at different rates, according to the concentrations of NO and O\textsubscript{3} (the concentration of oxygen could be assumed to be constant).

The emission factors for different vehicle categories were derived using multiple regression analysis.
## Traffic classification

<table>
<thead>
<tr>
<th>Class</th>
<th>Golden River class</th>
<th>Assumed equivalent emission category</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Motorcycle</td>
<td>Motorcycle</td>
</tr>
<tr>
<td>2</td>
<td>Car or light van (&lt;2.5m)</td>
<td>Car or small van (&lt;2.5 t GVW[1])</td>
</tr>
<tr>
<td>3</td>
<td>Car or light van with trailer</td>
<td>Light goods vehicle (2.5-3.5 t GVW)</td>
</tr>
<tr>
<td>4</td>
<td>Heavy van (≥5.2 m) or minibus</td>
<td>Rigid heavy goods vehicle (&gt;3.5 t GVW)</td>
</tr>
<tr>
<td>5</td>
<td>Rigid light goods vehicle (&lt;8.7 m)</td>
<td>Articulated heavy goods vehicle (&gt;3.5 t GVW)</td>
</tr>
<tr>
<td>6</td>
<td>Rigid medium goods vehicle (≥8.7 m)</td>
<td></td>
</tr>
<tr>
<td>7</td>
<td>Rigid goods vehicle with trailer</td>
<td></td>
</tr>
<tr>
<td>8</td>
<td>Articulated heavy goods vehicle</td>
<td></td>
</tr>
<tr>
<td>9</td>
<td>Bus or coach</td>
<td>Bus or coach (&gt;3.5 t GVW)</td>
</tr>
</tbody>
</table>

[1] GVW = gross vehicle weight
Hatfield Tunnel traffic characteristics

- Traffic flow (veh per hour)
- Traffic speed (km/h)
- Proportion of traffic flow (%)

*Bus/coach,
*Artic HGV,
*Rigid HGV,
*Large van,
*Cars, light vans (+trailer)*
Measured & modeled NO$_2$ & O$_3$
## Traffic emission factors

<table>
<thead>
<tr>
<th>Tunnel section</th>
<th>Model</th>
<th>NO\textsubscript{x} (g vehicle\textsuperscript{-1} km\textsuperscript{-1})</th>
<th>NO\textsubscript{2} (g vehicle\textsuperscript{-1} km\textsuperscript{-1})</th>
<th>%NO\textsubscript{2}</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>EF</td>
<td>95%</td>
<td>EF</td>
</tr>
<tr>
<td><strong>ENT-MID</strong></td>
<td>M1</td>
<td>0.930</td>
<td>0.083</td>
<td>0.125</td>
</tr>
<tr>
<td></td>
<td>M2</td>
<td>0.930</td>
<td>0.083</td>
<td>0.096</td>
</tr>
<tr>
<td></td>
<td>M3</td>
<td>0.930</td>
<td>0.083</td>
<td>0.096</td>
</tr>
<tr>
<td><strong>MID-EXIT</strong></td>
<td>M1</td>
<td>1.094</td>
<td>0.090</td>
<td>0.088</td>
</tr>
<tr>
<td></td>
<td>M2</td>
<td>1.094</td>
<td>0.090</td>
<td>0.088</td>
</tr>
<tr>
<td></td>
<td>M3</td>
<td>1.094</td>
<td>0.090</td>
<td>0.088</td>
</tr>
<tr>
<td><strong>Overall</strong></td>
<td>M1</td>
<td>1.006</td>
<td>0.086</td>
<td>0.108</td>
</tr>
<tr>
<td></td>
<td>M2</td>
<td>1.006</td>
<td>0.086</td>
<td>0.092</td>
</tr>
<tr>
<td></td>
<td>M3</td>
<td>1.006</td>
<td>0.086</td>
<td>0.092</td>
</tr>
</tbody>
</table>
Regression modeling for NO\textsubscript{x} and NO\textsubscript{2}

\[ E_{total} = (NCSV \cdot ECSV) + (NLGV \cdot ELGV) + (NRHGV \cdot ERHGV) + (NAHGV \cdot EAHGV) + (NAHGV \cdot EAHGV) + (NBC \cdot EBC) + c \]

where:

- \( E_{total} \) = the total hourly emissions from the traffic (the average emission factor per vkm multiplied by the total number of vehicles)
- \( NCSV \) = the number of cars and small vans per hour
- \( NLGV \) = the number of large vans per hour
- \( NRHGV \) = the number of rigid heavy goods vehicles per hour
- \( NAHGV \) = the number of articulated heavy goods vehicles per hour
- \( NBC \) = the number of buses and coaches per hour

and the variables were:

- \( ECSV \) = the emission factor for cars and small vans
- \( ELGV \) = the emission factor for large vans
- \( ERHGV \) = the emission factor for rigid heavy goods vehicles
- \( EAHGV \) = the emission factor for articulated heavy goods vehicles
- \( EBC \) = the emission factor for buses and coaches
- \( c \) = a constant
### Estimated primary NO₂ from Hatfield Tunnel

<table>
<thead>
<tr>
<th>Vehicle type</th>
<th>Emission factor (g vehicle km⁻¹)</th>
<th>NO₂/NOₓ (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>NOₓ</td>
<td>NO₂</td>
</tr>
<tr>
<td>Cars and small vans</td>
<td>0.18 (± 0.03)</td>
<td>0.03 (± 0.00)</td>
</tr>
<tr>
<td>Light goods vehicles</td>
<td>1.30 (± 0.35)</td>
<td>0.24 (± 0.05)</td>
</tr>
<tr>
<td>Rigid HGVs</td>
<td>3.49 (± 0.61)</td>
<td>0.43 (± 0.08)</td>
</tr>
<tr>
<td>Articulated HGVs</td>
<td>3.84 (± 0.97)</td>
<td>0.44 (± 0.12)</td>
</tr>
<tr>
<td>Buses and coaches</td>
<td>9.74 (± 7.97)</td>
<td>1.44 (± 1.02)</td>
</tr>
</tbody>
</table>
Summary

- The 5% primary NO₂ component of NOₓ would appear to be an underestimate. Measurements on Euro I to Euro IV technologies have indicated primary NO₂ proportions of between 8% and 80%.
- The emission data underlying the primary NO₂ emission factors are not robust, due to limited sample sizes and inconsistent sampling protocols.
- Diesels, lean-burn technologies and oxidation catalysts are a significant cause of increased fleet primary NO₂ emissions.
- Euro III petrol LDV range from 2% to 10%.
- Euro III diesel LDV range from 20% to 70%.
- Euro III diesel LCV range from 22% to 38%.
- Euro III buses equipped with CRT can exhibit primary NO₂ emissions of 45%.
- Tunnel measurements have proven to provide a useful tool in the estimation of vehicle emissions and primary NO₂.
- A default primary NO₂ value of 20% would be more appropriate.
- Given this increase in primary NO₂, associated with exhaust after-treatment technologies, there is a case to consider emission standards for primary NO₂. CARB have proposed standards limiting the primary NO₂ component to a maximum of 30% and 20% for 2007 and 2009, respectively.
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