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New Directions: Should road vehicle emissions legislation consider primary NO_2? ☆

Vehicle emissions legislation in Europe has progressively developed over the years to set increasingly stringent limits for emissions of key pollutants. The legislation has also become more sophisticated. Early vehicle emissions legislation was concerned only with emissions of CO and total hydrocarbons (THC) from petrol passenger cars and light vans. More recently, limits have been set for NO_X, CO, THC and PM_{10} for passenger cars, light vans and heavy-duty vehicles (see http://europa.eu.int/comm/environment/air/legis.htm#transport for more information). As the legislation has developed, the drive cycles over which vehicles are tested have also become more complex and have increasingly aimed to replicate typical or "real-world" driving conditions. Furthermore, increased account has also been taken of actual ambient conditions by including an element of the vehicle test that is undertaken at low temperatures when vehicle emissions can be higher. The legislation has therefore developed to match more closely specific environmental problems of concern.

Over more or less the same period, legislation controlling concentrations in ambient air has also developed. Specifically, the First Air Quality Daughter Directive (1999/30/EC) has placed a new emphasis on air quality assessments in urban areas. In the UK, recent predictions of the annual mean NO_2 concentration in London for 2010 show that exceedences of the annual mean limit value of $40\,\mu g\,m^{-3}$ can be expected close to roads in London (Air Quality Expert Group, 2003, Nitrogen Dioxide in the UK, Department for Environment, Food and Rural Affairs, draft report, December 2003). As a consequence, there has been an increased interest in the prediction of annual mean NO_2 concentrations in the near-road environment. In addition, policy makers are interested to know more about the contribution made by different emission sources to exceedences of air quality limits since this is a prerequisite if effective plans are to be developed to reduce emissions and to meet the limits.

There have been relatively few vehicle emissions studies that have partitioned between NO and NO_2 and that quantify the typical levels of primary NO_2 expected in the exhausts of different vehicle types under different modes of operation. There has been even less consideration given to the effect on ambient concentrations that primary NO_2 emissions have. An analysis of data from a monitoring site situated in a busy street canyon in central London has highlighted that primary NO_2 emissions could make a significant contribution to observed concentrations of NO_2 (Carslaw and Beevers, 2003, Investigating the Potential Importance of Primary NO_2 in a Street Canyons, submitted to Atmospheric Environment, December 2003). Table 1 shows how predicted concentrations of NO_2 are likely to be partitioned between primary and secondary NO_2 and by the contribution made by petrol and diesel vehicles. These calculations were made using simple $NO–NO_2–O_3$ chemistry. In Table 1, the NO_2 derived through $NO–O_3$ chemistry in the street canyon i.e. the secondary NO_2, has been apportioned based on the relative emissions of NO_X from petrol and diesel vehicles. Account has been taken of a range of primary NO_2 emission levels expressed as a % by vol. of the total emission of NO_X. First, it was assumed that primary NO_2 emissions from petrol and diesel vehicles were zero. Second, both the petrol and diesel primary NO_2 emissions were set to 5% by vol., which is the level often quoted as being typical in the literature. Finally, the best estimate of primary NO_2 emissions from petrol and diesel vehicles at this location was used: 12.7% for diesel vehicles and 0.6% by vol. for petrol vehicles.

There are two important issues highlighted by the results in Table 1. First, if the primary NO_2 emissions were zero, predicted NO_2 concentrations would be considerably less than observed concentrations (35.1 vs. 47.4 ppbv). Primary NO_2 emissions are therefore important in their own right in contributing to ambient NO_2 concentrations at this location. Second, the relative importance of petrol and diesel vehicles is also noteworthy. Diesel vehicles are estimated to be responsible for 85% of the road contribution of observed

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Table 1
Influence of different ratios of primary NO_2:NO_X assumed for petrol and diesel vehicles at the Marylebone Road street canyon site and estimated source apportionment for 1999 (ppbv)

Vehicular primary NO_2 (% by volume)	Background	Primary NO_2		Secondary NO_2		Total
		Petrol	Diesel	Petrol	Diesel	
Observed	24.0	—	—	—	—	47.4
0%	24.0	0	0	4.0	7.1	35.1
5% (petrol and diesel)	24.0	2.9	5.1	3.9	7.0	42.9
12.7% (diesel), 0.6% (petrol)	24.0	0.3	13.6	3.4	6.1	47.4

NO_2 compared with only 68% of total NO_X concentrations. Diesel vehicles are therefore proportionately more important in terms of their contribution to ambient NO_2 than ambient NO_X, which is important since ambient limits are set for NO_2 and not NO_X.

In London, as in many other major cities, there are other developments that make the issue of primary NO_2 potentially more important. By 2005 for example, all Transport for London buses will be converted to have continuous diesel particulate filters (CDPF) (The Mayor's Air Quality Strategy, September 2002, The Greater London Authority, London). The particle filters used operate by converting NO to NO_2 and use the NO_2 to help oxidize the particles. These filters tend to produce higher proportions of NO_2:NO_X in the exhaust cf. the same vehicle without a CDPF. However, there are relatively few measurements of primary NO_2 from vehicles fitted with CDPF systems. Nevertheless, a large-scale study conducted by the Danish Road Safety and Transport Agency completed in 2002 on 120 HGVs (including 85 buses) using CDPF from several manufacturers showed that primary NO_2 increased from approximately 5% without a CDPF to 15–20% by vol. with a CDPF (Danish Road Safety and Transport Agency, 2002, Large Scale Project with Particulate Filters on Heavy-duty Vehicles in Odense, report available from http://www.fstyr.dk/sw1140.asp). Although there are technologies available to reduce both NO_X and NO_2 from vehicles exhausts, there is currently no legislation that specifically aims to control primary NO_2 emissions.

Because there are comparatively few measurements of primary NO_2 from road vehicles, the spatial and temporal variation in emissions in urban areas is not well understood. There is also little information available on the specific contribution that primary NO_2 emissions make towards observed ambient concentrations in urban areas close to roads. It appears important therefore that more work be undertaken to quantify the emissions of primary NO_2 in urban areas and that detailed emissions inventories are developed. If, following this work, it is shown that primary NO_2 emissions are significant in their own right to urban concentrations of NO_2, as appears to be the case in the street canyon study; there might be a case for legislation to control vehicular emissions of primary NO_2.

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