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# Quantifying local traffic contributions to $NO_2$ and $NH_3$ concentrations in natural habitats

Sally R. Gadsdon [1], Sally A. Power*

*Division of Biology, Imperial College London, Silwood Park Campus, Ascot, Berkshire SL5 7PY, UK*

*$NO_x$ and $NH_3$ concentrations exceed vegetation critical levels up to 20 m from roadsides, both at ground level and in tree canopies.*

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ABSTRACT

$NO_2$ and $NH_3$ concentrations were measured across a Special Area for Conservation in southern England, at varying distances from the local road network. Exceedances of the critical levels for these pollutants were recorded at nearly all roadside locations, extending up to 20 m away from roads at some sites. Further, paired measurements of $NH_3$ and $NO_2$ concentrations revealed differences between ground and tree canopy levels. At "background" sites, away from the direct influence of roads, concentrations were higher within tree canopies than at ground level; the reverse pattern was, however, seen at roadside locations. Calculations of pollutant deposition rates showed that nitrogen inputs are dominated by $NH_3$ at roadside sites. This study demonstrates that local traffic emissions contribute substantially to the exceedance of critical levels and critical loads, and suggests that on-site monitoring is needed for sites of nature conservation value which are in close proximity to local transport routes.



## 1. Introduction

Ecosystems are being threatened by the pressures and consequences of modern living. Increasing pollution, encroachment of the built environment and rising visitor pressure are having harmful effects on natural areas (Woodin and Farmer, 1993; Lee and Caporn, 1998; Haskins, 2000). The emissions from road vehicles contribute a significant proportion to the total UK pollution emissions, especially for nitrogenous pollutants (Dore et al., 2008) with oxides of nitrogen ($NO_x$, 32% of UK emissions) generated as a by-product of fossil fuel combustion and ammonia ($NH_3$, 3% of UK emissions) being emitted by catalytic converters (Sutton et al., 2000). Roads are often the nearest source of pollution to natural areas. Therefore, quantifying the pollutants associated with road traffic is an important step in understanding the effects of air quality on adjacent ecosystems.

Nitrogen is an essential plant macronutrient and, as such, nitrogen-containing pollutants (e.g. nitrogen dioxide ($NO_2$) and $NH_3$) can be detoxified and assimilated in low concentrations by plants, therefore contributing to plant nitrogen budgets (Caporn et al., 2000; Sheppard and Leith, 2002). However, where concentrations are high nitrogen can be detrimental to plant vitality (Wellburn, 1990; Pearson and Stewart, 1993) and the deposition of these pollutants can alter ecosystem functioning (Boxman et al., 1995; Green, 2005). Effects of high concentrations of gaseous nitrogen pollutants on plants include leaf injury caused by cellular damage (Srivastava and Ormrod, 1984; Wellburn, 1990), changes in growth (Bender et al., 1991; Honour et al., 2009), and increased tissue nitrogen concentrations (Caporn et al., 2000; Sheppard and Leith, 2002). The latter effect can have secondary consequences such as increased herbivory (Port and Thompson, 1980; Flückiger and Braun, 1999; Hartley and Mitchell, 2005) and changes in phenology (Power et al., 1998). Effects on plant physiology, biochemistry and/or phenology can result in changes at the plant community and ecosystem level. For example, plant species composition has been found to change in relation to roadside proximity in various habitat types, with effects being seen up to 230 m away (Angold, 1997; Bernhardt-Römermann et al., 2005; Bignal et al., 2007). Furthermore, nitrogen-driven changes in soil chemistry and the composition and activity of the soil microbial community have been linked with shifts in nutrient cycling, with detrimental effects on ecosystem functioning (Green, 2005). As well as emissions of atmospheric pollutants, road transport is frequently associated with elevated levels of dusts, heavy metals and salts, and changes in the microclimate and hydrology of adjacent habitats (Spellerberg, 1998; Farmer, 2000).

The current European guidelines for protecting terrestrial vegetation from air pollution damage are in the form of critical levels (WHO, 2000). Critical levels are derived from exposure–response

* Corresponding author. Tel.: +44 207 594 2318; fax: +44 207 594 2339.
*E-mail addresses:* sally.gadsdon@cityoflondon.gov.uk (S.R. Gadsdon), s.power@imperial.ac.uk (S.A. Power).
[1] Current address: Epping Forest, City of London, The Warren, Loughton, Essex IG10 4RW, UK.

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experiments on plants, typically carried out in greenhouses or open top chamber fumigation systems, and values are set in order to protect the most sensitive plant elements of an ecosystem (Cairns, 1992). Currently, the critical level for $NO_x$ is 30 μg $m^{-3}$ (WHO, 2000); the critical levels for $NH_3$ are 1 μg $m^{-3}$ for lichens and bryophytes and 2–4 μg $m^{-3}$ for higher plants (UNECE, 2007).

At sites of nature conservation value there is a need to identify areas where pollution levels exceed critical thresholds and thus represent a threat to plant health and ecosystem integrity. This information can then feed into habitat management policy and planning. Areas of air pollution concern are often identified through air quality models; these use measurements taken from monitoring networks, together with meteorological data, to estimate pollutant concentrations. However, in the UK, modelled values for national maps are typically at a relatively low spatial resolution (e.g. 5 km for $NO_x$) and do not include contributions from more local sources of pollution, e.g. heavily used local roads. While higher resolution models are available for environmental impact assessments, availability of data from such models is limited in terms of geographic cover, and modelled values are no substitute for on-site measurements. There is, therefore, a clear need to examine the extent to which modelled pollutant data under-represent actual concentrations in areas of high nature conservation importance, particularly in those sites which experience emissions associated with a dense or heavily trafficked road network. Furthermore, given that pollution monitoring is mainly carried out at ground level, there may be differences between measured concentrations and those experienced within canopies, where gaseous pollutant uptake is focussed for trees.

Passive sampling techniques are commonly used for field measurements and are based on diffusion tubes with a chemical absorbent at one end which cumulatively absorbs the pollutant. Whilst these are unable to provide information on the occurrence of high concentrations reached over short periods of time, e.g. during peak flows of traffic, they do provide a good estimate of mean pollutant exposure, at high spatial resolution. Diffusion tubes were used to measure spatial patterns in concentrations of two key air pollutants, $NO_2$ and $NH_3$, within a site of high nature conservation importance in southern England.

In order to evaluate the potential contribution of traffic-derived pollution to the local pollution climate, the study aimed to: 1) quantify the effect of proximity to local roadsides on annual mean concentrations of $NO_2$ and $NH_3$ in natural habitats and to compare concentrations and deposition with modelled data; 2) investigate the spatial variation in concentrations of $NO_2$ and $NH_3$ between ground level and within the tree canopy. This is important in terms of assessing the potential impact of locally derived pollution on woodland ecosystems because, although most diffusion tube monitoring takes place approximately 2 m above the ground (Targa et al., 2008), the majority of tree leaves are typically located 10–25 m above ground. This enables more informed judgement about the exceedance of critical levels of key gaseous pollutants within tree canopies. Collectively, the results of this study contribute to an assessment of the impact of the local road network on air pollution levels at a site of high nature conservation value, and of the potential for differences in pollutant exposure between ground and canopy levels in a woodland ecosystem.

## 2. Methods

### 2.1. Site details

Epping Forest is a 2500 ha site of national and international conservation importance (Site of Special Scientific Interest (SSSI), Special Area of Conservation (SAC)) owned and managed by the City of London Corporation, located 18 km NE of London. The majority of the land is relic wood-pasture with scattered grasslands and dry and wet heathlands, and there is current concern that poor air quality is affecting the health of these habitats (Natural England, 2007a). The prevailing wind across the area is south-westerly (London Air Quality Network, 2008) with the site, therefore, being located downwind of London. There are several major roads running through Epping Forest: Major ("A") roads typically carry 17–24,500 vehicles per day, local ("B") roads have 10–23,000 vehicles per day, whilst minor roads carry 500–1750 vehicles per day. Furthermore, two of the UK's busiest motorways run either through (M25) or close to (M11) the site.

### 2.2. Spatial variation in $NO_2$ and $NH_3$ concentrations

Palmes-type diffusion tubes were used to measure $NO_2$ (Bush et al., 2001). These were produced and analysed by Gradko International (Winchester, Hampshire, UK) and used a 50% triethanolamine (TEA)/water preparation (see Targa et al., 2008 for quality assurance/control methods). In May 2004, triplicate tubes were installed at all sites and changed monthly over a period of 12 months. Alpha samplers were used to monitor $NH_3$ concentrations and were produced and analysed by the Centre for Ecology and Hydrology (CEH), Edinburgh (UK) (see Tang and Sutton (2004) for details of both the method and the quality assurance/control protocols). Ammonia monitoring began in August 2004 using duplicate samplers changed every two months, over a 12-month period. Across the Forest, $NO_2$ was measured at 32 locations and $NH_3$ was measured at 14 locations.

There were three types of monitoring sites: roadside (R) (0–4 m from the kerb); intermediate (I) (20–252 m from the roadside); and background (B) (>275 m from the roadside). The majority of sites were located in woodland. Diffusion tubes were mounted to various features, at heights of 1.5–3.0 m above the ground; positioning of tubes followed the guidance given in the UK $NO_2$ Diffusion Tube Network instruction manual as far as possible (Targa et al., 2008).

Spatial patterns of these two pollutants were investigated further by comparing concentrations at ground level to those measured within the tree canopy, using additional diffusion tubes at a further twenty sites, distributed across the Forest. One $NH_3$ sampler and two $NO_2$ tubes (from the same suppliers stated above) were placed a) at 2 m above ground level and b) 2 m below the top of the canopy of an oak tree (*Quercus robur* L., circumference range 180–300 cm) for a single month in February 2006; study trees were 12–23 m tall, and were without foliage at this time of year. The use of $NO_2$ and $NH_3$ tubes during the 2004–2005 monitoring period showed that the variance between duplicate $NH_3$ tubes and triplicate $NO_2$ tubes was low, permitting the use of a single $NH_3$ and duplicate $NO_2$ tubes for the subsequent ground level/canopy comparison. Thirteen sites for ground–canopy comparison were positioned away from the direct influence of the road, more than 100 m from the kerbside (='non-roadside site') on internal woodland edges. Seven comparison sites were located 2–30 m from the nearest road ('road margin sites') also on internal or roadside woodland edges; the total of 20 sites formed a north–south transect through Epping Forest, towards central London. Distance from central London was measured as the distance to Hyde Park Corner (grid reference TQ283798) using MapInfo (version 8.0).

For the 2004–2005 and the February 2006 monitoring, sample sites were located in both upwind and downwind positions in relation to local roads, with no systematic control of location in relation to the prevailing wind direction.

### 2.3. Quantification of nitrogen deposition

At sites where both $NO_2$ and $NH_3$ concentrations were measured, nitrogen deposition was calculated. The annual mean concentrations for $NO_2$ and $NH_3$ from the 2004–2005 monitoring were used to estimate the nitrogen deposition from these sources using deposition velocities appropriate for the pollutants and habitat types ($NO_2$ to woodland 0.00120 $ms^{-1}$, to heathland 0.00077 $ms^{-1}$; $NH_3$ to woodland 0.0360 $ms^{-1}$, to heathland 0.01905 $ms^{-1}$, Smith et al., 2000). Whilst it is recognised that the deposition velocity for $NH_3$ depends on its concentration (Flechard and Fowler, 1998), a single, habitat-specific deposition velocity (listed above) was used to estimate deposition rates for this gas in this study. Estimates of total nitrogen deposition at the site included data supplied by the Centre for Ecology and Hydrology (Edinburgh, UK) for ammonium and nitrate aerosols, nitric acid and wet deposition of ammonium and nitrate, averaged between 2002 and 2004.

### 2.4. Statistical analysis

$NO_2$ and $NH_3$ data were analysed by general linear modelling in R (version 1.7.1, R Development Core Team, 2004). Linear regression was used to evaluate the relationships with distance from London, while non-linear regression was used for road transect data. Values were log transformed where appropriate and subsequently back-transformed for presentation. The significance of differences in concentrations of $NO_2$ and $NH_3$ at intermediate and background sites was determined using a *t*-test. Differences in concentrations of $NO_2$ and $NH_3$ between canopy and ground levels were tested through paired *t*-tests.

## 3. Results

### 3.1. Spatial patterns in concentrations of $NO_2$ and $NH_3$

Annual mean concentrations of $NO_2$ and $NH_3$ at each site are shown in Table 1. Where concentrations exceeded the appropriate

**Table 1**
Annual mean concentrations of $NO_2$ and $NH_3$ (μg $m^{-3}$, ±SEM values) at the Epping Forest pollution monitoring sites. – = this pollutant was not monitored at this site. Concentrations that exceed the critical level are highlighted in bold. *Site names beginning with: R = roadside (0–4 m from the roadside); I = intermediate (20–252 m from roadside); B = background (>275 m from roadside).

| Site* | Distance from roadside (m) | $NO_2$ | $NH_3$ |
|---|---|---|---|
| R1 | 4.0 | **63.2 ± 4.04** | **3.21 ± 0.28** |
| R2 | 1.4 | **39.3 ± 1.51** | **2.68 ± 0.07** |
| R3 | 1.3 | **57.7 ± 1.27** | **6.12 ± 0.05** |
| R4 | 0.4 | **40.5 ± 1.26** | **2.79 ± 0.11** |
| R5 | 0.5 | 23.9 ± 0.87 | – |
| R6 | 0.2 | 25.4 ± 1.44 | – |
| R7 | 1.0 | **37.9 ± 1.50** | **2.37 ± 0.15** |
| R8 | 1.0 | **33.5 ± 0.87** | **2.96 ± 0.04** |
| R9 | 1.0 | **36.0 ± 1.90** | **3.67 ± 0.12** |
| R10 | 0.8 | – | **7.89 ± 0.18** |
| I1 | 190 | 24.0 ± 0.82 | – |
| I2 | 135 | 25.2 ± 1.04 | – |
| I3 | 251 | 25.1 ± 0.91 | – |
| I4 | 20 | **32.0 ± 1.33** | – |
| I5 | 90 | 23.8 ± 0.70 | – |
| I6 | 25 | 28.2 ± 1.15 | – |
| I7 | 100 | 25.6 ± 0.71 | – |
| I8 | 20 | **31.3 ± 1.25** | **1.38 ± 0.10** |
| I9 | 100 | 24.7 ± 0.96 | 0.62 ± 0.03 |
| I10 | 68 | 22.9 ± 0.75 | 0.63 ± 0.03 |
| I11 | 35 | 20.7 ± 0.89 | – |
| I12 | 115 | 22.9 ± 0.82 | – |
| I13 | 25 | 25.6 ± 0.77 | 0.65 ± 0.02 |
| I14 | 100 | 23.2 ± 0.95 | 0.54 ± 0.02 |
| I15 | 131 | 23.0 ± 0.44 | – |
| I16 | 172 | 22.2 ± 0.58 | – |
| I17 | 33 | 22.7 ± 0.71 | – |
| I18 | 252 | 23.4 ± 0.76 | – |
| I19 | 207 | 24.9 ± 0.88 | – |
| B1 | 443 | 21.2 ± 0.97 | – |
| B2 | 795 | 19.9 ± 0.45 | 0.44 ± 0.01 |
| B3 | 690 | 20.0 ± 0.76 | – |
| B4 | 281 | 19.2 ± 0.60 | – |

critical level, values are highlighted in bold in Table 1. An exceedance of the $NO_x$ critical level was measured at all except two of the roadside sites, as well as at two sites 20 m from the road. The concentration of $NH_3$ exceeded the 1 μg $m^{-3}$ critical level for lichens at eight roadside sites and one site 20 m from a road. Two roadside sites also exceeded the 2–4 μg $m^{-3}$ critical level of $NH_3$ for higher plants.

Fig. 1a shows the attenuation in $NO_2$ concentrations with distance from the road, with levels falling to background values after around 250 m. The concentration of $NO_2$ at the furthest "intermediate" sites (177–252 m) was still raised above levels at background sites ($p < 0.001$). The average difference between concentrations at sites 177–252 m from roads and those more than 280 m away was 4.25 μg $m^{-3}$.

As with $NO_2$, concentrations of $NH_3$ rapidly attenuated with distance from the roadside (Fig. 1b). Concentrations around 100 m from the road were still elevated by approximately 0.12 μg $m^{-3}$ above background concentrations (795 m from the road, $p = 0.04$).

There was a statistically significant increase in within-canopy $NO_2$ concentrations with increasing proximity to London ($r^2 = 0.399$, $p = 0.02$, Fig. 2a). The same trend was seen at ground level, although this was not statistically significant ($r^2 = 0.416$, $p = 0.06$). Within-canopy $NH_3$ concentrations did not change significantly in relation to distance from London ($r^2 = 0.251$, $p = 0.09$); however, the concentrations at the two sites furthest from the city were subsequently determined to be influenced by another source of ammonia (a local sewage works) (Fig. 2b). When these sites were excluded from the data set, the regression of $NH_3$ concentrations against distance from London became highly significant, at both within-canopy ($r^2 = 0.798$, $p < 0.001$) and ground levels ($r^2 = 0.552$, $p = 0.03$).

### 3.2. Comparison of within-canopy and ground-level concentrations of $NO_2$ and $NH_3$

The precise height of the canopy diffusion tubes above ground level, which was dependent on the height of the tree, did not affect the concentrations of $NO_2$ or $NH_3$ (data not shown). At sites more than 100 m away from roads, within-canopy concentrations of $NO_2$ and $NH_3$ were significantly higher than concentrations at ground level ($NO_2$ average difference 2.62 μg $m^{-3}$ ± 0.41 μg $m^{-3}$: $p < 0.045$, $NH_3$ average difference 0.32 ± 0.03: $p < 0.001$; Table 2, Fig. 3a and b, respectively). At the road margin sites, for both $NO_2$ and $NH_3$, ground-level concentrations were generally higher than those measured within the canopy (average differences $NO_2$: 6.59 ± 1.88 μg $m^{-3}$ and $NH_3$: 0.61 ± 0.26 μg $m^{-3}$), although these differences were not statistically significant.

### 3.3. Quantification of nitrogen deposition

Table 3 summarises estimates of nitrogen deposition for those sites where both $NO_2$ and $NH_3$ measurements were made. At the roadside sites, deposition to vegetation was dominated by nitrogen in the dry deposited form. Exceedances of the upper limit of the critical load for nitrogen for woodlands (temperate forest 10–20 kg N $ha^{-1}$ $yr^{-1}$) were found at all except one site; the lower limit of the critical load for nitrogen for heathland (lowland dry heathland 10–20 kg N $ha^{-1}$ $yr^{-1}$, wet heathland 10–25 kg N $ha^{-1}$ $yr^{-1}$), a habitat represented in fragments throughout the Forest, was also exceeded (Achermann and Bobbink, 2003).

## 4. Discussion

### 4.1. Spatial variation in concentrations of $NO_2$ and $NH_3$

Most roadside sites and two of those 20 m away from the roadside exceeded the critical level for $NO_x$ by their concentrations of $NO_2$ alone. Concentrations of NO were not recorded systematically in this study, so have not been taken into account in evaluation of $NO_x$ critical level exceedance. Roadside NO:$NO_2$ ratio values measured in London have been in the range of 4–5:1 (Buckingham et al., 1997; Honour, 2003). Limited nitric oxide (NO) monitoring carried out in Epping Forest during 2004–2005 (data not shown) indicated roadside ratios of NO:$NO_2$ ranged from 0.41 to 0.77 within 1–4 m of the road (Gadsdon, 2007). Nitric oxide can, therefore, be expected to contribute a potentially significant addition to the total roadside concentration of $NO_x$, with the likelihood that all roadside sampling points, and many at 20 m away exceed $NO_x$ critical levels when even a small contribution from NO is included. Over 4% of the Forest SAC occurs within 20 m of a major or locally important road and can, therefore, be expected to experience detrimental effects of locally derived $NO_x$ pollution.

Exceedance of the recently revised critical level of $NH_3$ for lichens and bryophytes (1 μg $m^{-3}$) was found at all roadside sites and one site 20 m away from the road. In a recent survey of lichen species distributions in Epping Forest (the study site), James and Davies (2006) found that roadside trees had a lower lichen species diversity and a higher proportion of species associated with eutrophic conditions, compared to trees on internal woodland edges, away from the influence of roads. Roadside $NH_3$ concentrations ranged from 2.68 to 7.89 μg $m^{-3}$, therefore also exceeding critical levels for higher plants (2–4 μg $m^{-3}$, UNECE, 2007).

The attenuation in $NO_2$ and $NH_3$ concentrations away from the road edge has been found to be relatively steep in several studies.

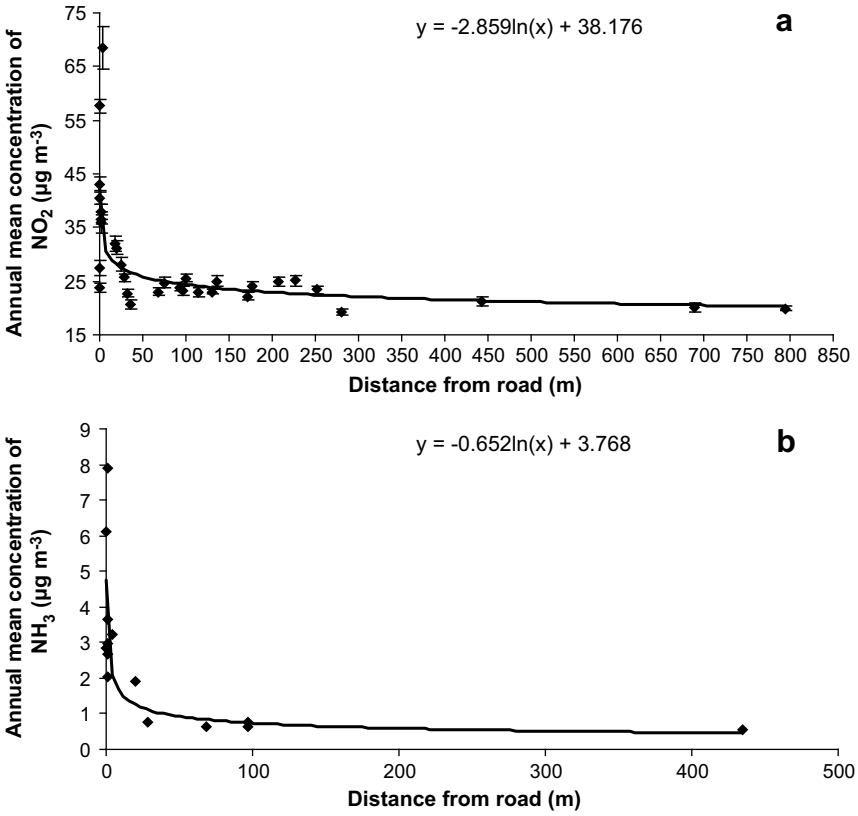


**Fig. 1.** a) Annual mean $NO_2$ concentrations (May 2004–April 2005) and b) annual mean $NH_3$ concentration (August 2004–July 2005) with distance from the nearest road throughout Epping Forest. Error bars represent ± 1 SEM.

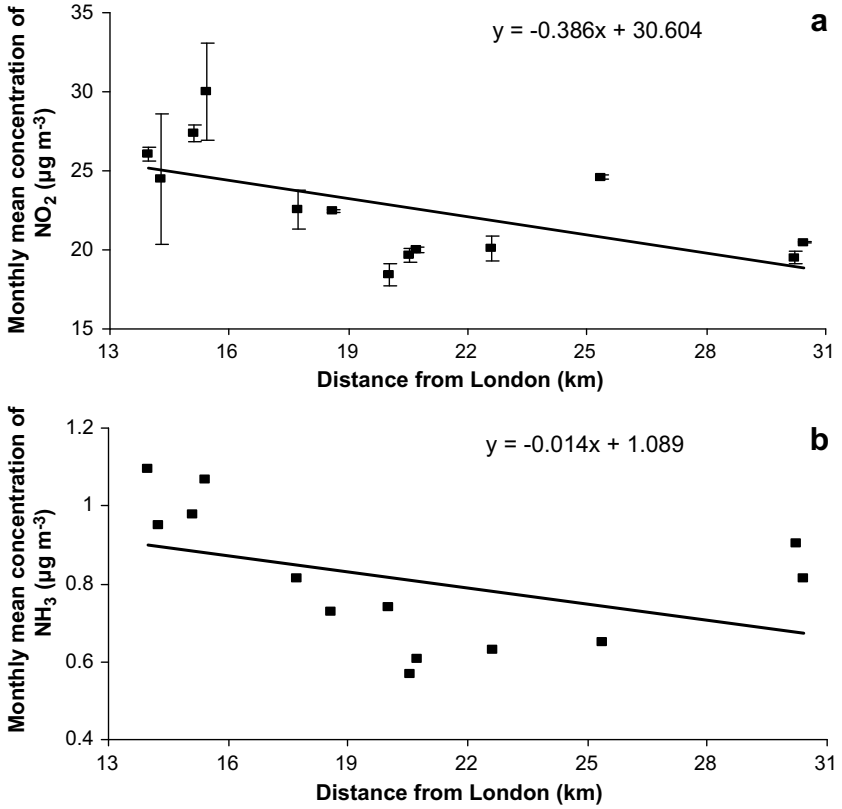


**Fig. 2.** a) Monthly mean $NO_2$ concentrations and b) monthly mean $NH_3$ concentrations in February 2006 measured in the canopies of oak trees, in relation to distance from London. Error bars represent ± 1 SEM.

**Table 2**
Concentrations of $NO_2$ and $NH_3$ (μg $m^{-3}$) at within-canopy and ground levels at a range of sites across Epping Forest in February 2006. *Tubes/sampler vandalised/stolen.

| Site | Distance from roadside (m) | Within-canopy $NO_2$ | Ground-level $NO_2$ | Within-canopy $NH_3$ | Ground-level $NH_3$ |
|---|---|---|---|---|---|
| 1 | 3 | 23.5 ± 0.4 | 33.8 ± 1.5 | 0.68 | 2.18 |
| 2 | 5 | 32.2 ± 1.4 | 44.6 ± 1.3 | 1.27 | 2.93 |
| 3 | 11 | 20.8 ± 0.5 | 26.5 ± 0.8 | 0.99 | 1.15 |
| 4 | 16 | 20.0 ± 0.7 | 22.6 ± 1.9 | 0.64 | 0.73 |
| 5 | 22 | 19.7 ± 1.6 | 26.2 ± 0.2 | 0.67 | 1.20 |
| 6 | 22 | 26.9 ± 1.2 | 37.3 ± 1.7 | 1.37 | 1.79 |
| 7 | 25 | 23.0 ± 0.4 | 23.3 ± 0.6 | 0.77 | 0.67 |
| 8 | 141 | 27.3 ± 0.5 | 25.2 ± 0.3 | 0.98 | 0.54 |
| 9 | 147 | 24.5 ± 4.1 | * | 0.95 | 0.64 |
| 10 | 151 | 24.6 ± 0.1 | 20.0 ± 3.3 | 0.65 | 0.31 |
| 11 | 153 | 30.0 ± 3.1 | * | 1.07 | 0.26 |
| 12 | 264 | 19.5 ± 0.4 | 17.7 ± 0.2 | 0.90 | 0.45 |
| 13 | 272 | 26.1 ± 0.5 | 25.5 ± 1.4 | 1.10 | * |
| 14 | 323 | 22.4 ± 0.1 | 19.8 ± 0.1 | 0.73 | 0.36 |
| 15 | 348 | 22.5 ± 1.2 | 21.5 ± 0 | 0.81 | 0.69 |
| 16 | 373 | 20.5 ± 0.1 | 17.9 ± 1.0 | 0.81 | 0.52 |
| 17 | 585 | 20.0 ± 0.1 | 16.1 ± 1.1 | 0.61 | 0.25 |
| 18 | 644 | 20.1 ± 0.8 | * | 0.63 | * |
| 19 | 670 | 18.4 ± 0.7 | 15.5 ± 0.8 | 0.74 | 0.34 |
| 20 | 684 | 19.7 ± 0.4 | 15.7 ± 1.0 | 0.57 | 0.41 |

Cape et al. (2004) measured a 90% fall off in concentrations above background levels by 15 m for $NO_2$ and 10 m for $NH_3$ at a variety of road types and habitats in Scotland (240–85,623 vehicles per day). The results from the current study suggest that $NO_2$ concentrations were elevated above background levels up to approximately 250 m away from local roads. Other studies have recorded concentrations higher than background levels up to 200 m (185,000 vehicles per day, Gilbert et al., 2003) and 1000 m (24,000 vehicles per day, Glasius et al., 1999) away from heavily trafficked motorways.

As expected, concentrations of $NO_2$ at canopy level increased nearer to London. Harrison et al. (2006) found that $NO_x$ concentrations were, on average, 140% higher 12 km downwind of a major urban area in the West Midlands (UK), compared to similar distances upwind. The location of nature conservation sites in relation to prevailing wind direction and urban conurbations is likely, therefore, to affect the balance between local and background contributions to pollutant concentrations, with potential implications for local pollution management policy. In the current study, $NH_3$ concentrations were also highest at the sites closest to central London. Although relatively few studies have looked at the relationship between urban proximity and $NH_3$ concentrations, one such study in Rome found that $NH_3$ concentrations at an urban

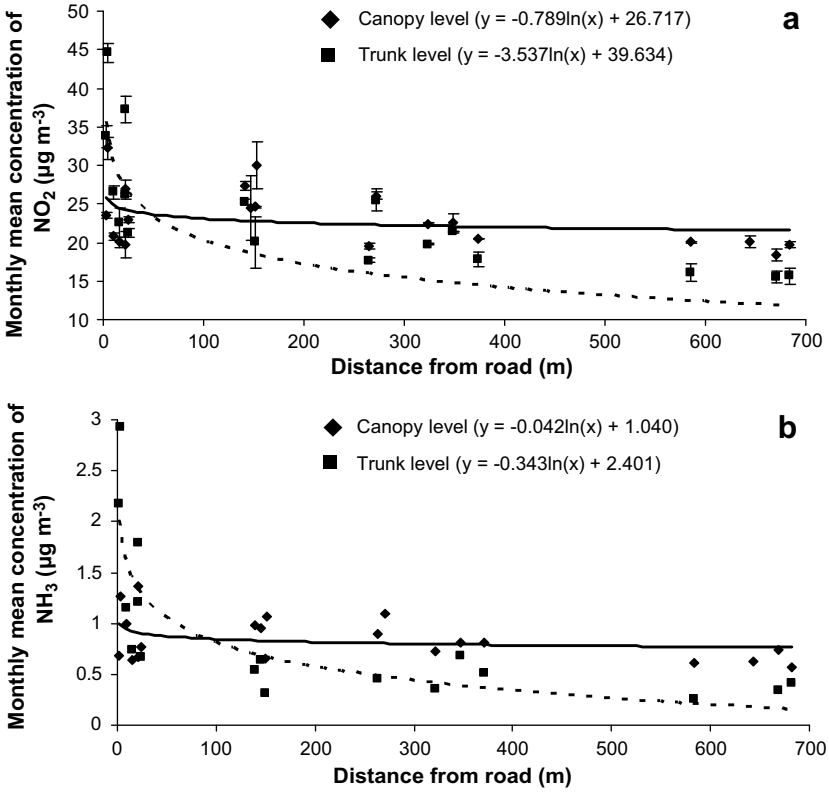


**Fig. 3.** a) Monthly mean $NO_2$ concentrations and b) monthly mean $NH_3$ concentrations in February 2006 measured at within-canopy (solid line) and trunk level (dotted line) with distance from the nearest road. Some of the canopy points do not have a trunk level concentration due to vandalism. Error bars represent ± 1 SEM.

**Table 3**
Estimates of N deposition (kg N $ha^{-1}$ $yr^{-1}$) for a twelve-month period in 2004–2005 to sites across Epping Forest, and estimated average N deposition (kg N $ha^{-1}$ $yr^{-1}$) to each site given by the APIS[a] website for the period 2003–2005. §Site names beginning with: R = roadside (0–4 m from the roadside); I = intermediate (20–252 m from roadside); B = background (>275 m from roadside).

| Site§ | N deposition from $NO_2$ | N deposition from $NH_3$ | Total N deposition[c] | Predicted N deposition from APIS |
|---|---|---|---|---|
| R1[b] | 4.69 | 15.88 | 33.10 | 18.3 |
| R2[b] | 2.96 | 13.26 | 27.42 | 17.5 |
| R3 | 6.67 | 57.22 | 88.24 | 36.4 |
| R4 | 4.68 | 26.09 | 55.12 | 36.4 |
| R7 | 4.38 | 22.16 | 50.89 | 36.4 |
| R8 | 3.87 | 27.67 | 55.89 | 36.4 |
| R9 | 4.61 | 34.31 | 62.82 | 36.4 |
| I3[b] | 4.6 | 1.90 | 18.22 | 17.5 |
| I8 | 3.62 | 12.90 | 40.87 | 36.4 |
| I9[b] | 4.40 | 0.90 | 15.28 | 17.5 |
| I10 | 2.65 | 5.89 | 32.89 | 36.4 |
| I13 | 2.96 | 6.08 | 33.39 | 36.4 |
| I14 | 2.68 | 5.05 | 32.08 | 36.4 |
| B2 | 2.30 | 4.11 | 30.76 | 36.4 |

[a] Values from www.apis.ac.uk.
[b] These sites are short vegetation habitats (grassland or heathland) located within the Forest area.
[c] Total includes data on the average deposition of ammonium and nitrate aerosols, nitric acid, wet deposition of ammonium and nitrate between 2002 and 2004 at a 5 $km^2$ resolution, provided by CEH, Edinburgh.

traffic station were 5 times higher than at an urban background site and higher still than levels measured at a rural site 20 km from the city (Perrino et al., 2002).

### 4.2. Comparison of within-canopy and ground-level concentrations of $NO_2$ and $NH_3$

Away from roadsides, concentrations of $NO_2$ and $NH_3$ were higher within the canopy than at ground level; $NO_2$ and $NH_3$ were up to 25% (3.99 μg $m^{-3}$) and 140% (0.35 μg $m^{-3}$) higher within the canopy, respectively. However, at most of the road margin sites this pattern was reversed. At these sites, ground-level concentrations of $NO_2$ and $NH_3$ were up to 43% (10.2 μg $m^{-3}$) and 220% (1.5 μg $m^{-3}$) higher, respectively, than levels within the tree canopy, reflecting the strong influence of vehicle emission sources at street level. Interestingly, those sites with the highest concentrations around the trunk were not necessarily those with the greatest concentrations of $NO_2$ and $NH_3$ in the canopy, although concentrations at the ground and within the tree canopy were highly correlated. Furthermore, there was very little decrease in canopy level concentrations of either pollutant away from the road. Similar differences in concentrations of other pollutants between ground level and tree canopy level have been reported elsewhere. Lovett and Lindberg (1992) measured concentrations of nitric acid vapour through canopies of *Quercus* and *Acer* species and found that the levels in the upper canopy were 32% higher than at ground level. Ozone concentrations have also been reported to be 45–89% higher in the upper canopy compared to ground level (Krzyzanowski, 2004).

These results highlight the fact that considerable differences can exist in pollutant concentrations between ground level and within woodland canopies. These differences are likely to vary when the canopy is in leaf because of the modifying effect on wind patterns around the canopy and pollutant uptake by leaves (Hosker and Lindberg, 1982; Lovett and Lindberg, 1992). Nevertheless, when trees are in leaf, large parts of the outer canopy would still be directly exposed to the air passing over it and any conclusions about the exposure of trees to gaseous pollutants which are based on ground-level measurements may be underestimated at sites away from the direct influence of roads.

### 4.3. The contribution of road traffic to nitrogen deposition at adjacent habitats

The concentrations of $NO_2$ and $NH_3$ measured in this study were used to calculate their contribution to nitrogen deposition inputs to woodland habitats. At the roadside sites, deposition was 10–58 kg N $ha^{-1}$ $yr^{-1}$ above background levels; these values exceed the critical nitrogen loads for woodland and heathland habitats just from dry deposition, without including other atmospheric nitrogen inputs. $NO_2$ and $NH_3$ contributed between 52 and 72% to the total nitrogen deposition, with the majority attributed to $NH_3$ close to road edges. Although some uncertainty must be associated with the use of a single (not concentration-dependant) deposition velocity for $NH_3$, it nevertheless highlights the importance of this pollutant in the evaluation of traffic impacts on adjacent habitats. Ammonia is currently not one of the pollutants listed in the UK Air Quality Strategy (DETR, 2000) and it is often excluded from environmental impact statements because of the lack of statutory requirement for its inclusion. Even though there are still uncertainties in the modelling of this pollutant, efforts need to be made to make this one of the suite of pollutants regularly assessed otherwise the calculations of nitrogen deposition to habitats may continue to be underestimated. The contribution of vehicles to overall rates of nitrogen deposition alongside moderately trafficked roads will be greatest at the roadside but are likely to extend at least 100 m (further for $NO_2$) into natural areas, as shown by the elevation of concentrations of $NO_2$ and $NH_3$ above background levels at this distance from local roads in the current study.

Table 3 shows values derived from the UK Air Pollution Information System website (APIS, www.apis.ac.uk) for each of the sites where deposition estimates were made. The APIS modelled values for the background and intermediate sites were all within ±15% of the nitrogen deposition values estimated in this study. However estimates based on field measurements at roadside sites were 40–142% higher than APIS modelled values. This provides a relatively high level of confidence in the ability of models to predict concentrations of gaseous pollutants at sites which are not strongly influenced by roads. However, it highlights the fact that modelled data based on interpolation of measurements at a relatively low spatial resolution (i.e. >1–5 km) are not able to reflect local hotspots of pollution which can be considerable. At the study site, critical levels of nitrogen are exceeded throughout the Forest, although the degree of exceedance is higher with increasing proximity to local roads. The level of exceedance is such that detrimental effects on the structure and functioning of adjacent ecosystems can be expected, although these were not assessed in the current study. Many sensitive nature conservation sites in the UK and Europe are located close to roads. The greater the extent to which a site is fragmented by local road networks, the greater the potential involvement of traffic-derived

pollutants in the exceedance of critical pollutant thresholds, and the greater the need for on-site measurement to determine actual concentrations and contributions.

## 5. Conclusion

The $NO_2$ and $NH_3$ measurements carried out at the study site have demonstrated the value of on-site monitoring for identifying local pollution hotspots and providing reliable information on exceedances of critical levels for selected pollutants. The profile of traffic emissions continues to change with, for example, increases in primary $NO_2$ emissions (Carslaw, 2005), and lower emissions of $NH_3$ associated with second generation catalytic converters (Dore et al., 2008). However, with predicted increases in vehicle numbers in the future (DETR, 2005), traffic-derived pollutants are likely to make an increasing contribution not just to the exceedance of critical levels, but also of critical nitrogen loads to adjacent habitats. These exceedances are very likely to have long term ecological implications for adjacent habitats including, for example, increased dominance of plant species associated with higher levels of nitrogen availability (Angold, 1997; Truscott et al., 2005).

The study also highlights differences between ground level and within-canopy pollutant concentrations, such as have been identified previously for both ozone and nitric acid (Lovett and Lindberg, 1992; Krzyzanowski, 2004); this should be considered when evaluating the exposure levels of woodland trees and associated canopy-dwelling organisms such as epiphytic lichens.

Whilst the relatively poor air quality measured at Epping Forest cannot be directly linked to the poor health of its habitats (Gadsdon, 2007), exceedance of $NO_x$ and $NH_3$ critical levels and nitrogen critical loads has been demonstrated. Over 7% of the area covered by Sites of Special Scientific Interest (SSSI) in England is thought to be in unfavourable condition due to air pollution (Natural England, 2007b), yet air quality issues are not consistently considered in statutory assessments of site condition. On-site monitoring is, therefore, recommended for sites of high nature conservation importance to identify local pollution issues and to enable air quality to be integrated into site condition assessments and, where appropriate, management plans.

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