Optimisation of a Numerical Model to Simulate the Dispersion and Chemical Transformations Within the Oxides of Nitrogen/Ozone System as Traffic Pollution Enters an Urban Greenspace

Dewi Komalasari¹ · Zongbo Shi¹ · Roy M. Harrison^{1,2}

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Abstract

Urban greenspace has many health benefits, including cleaner air than the surrounding streets. In this study, a detailed exercise has been conducted to measure concentrations of NO/NO₂/NO_x and O₃ within an urban greenspace, the University of Birmingham campus, using continuous analysers, as well as transects of NO₂ measured with diffusion tubes. Concentrations have been simulated using the ADMS-Roads model which has been optimised initially using NO_x concentrations for traffic emissions on surrounding roads, background concentrations, and meteorological data considering four candidate sites. Optimisation for prediction of NO₂ shows the critical importance of the NO₂:NO_x ratio in traffic emissions, for which a derivation from atmospheric measurements is consistent with a value derived from optimisation of the model fit to roadside data. After optimisation, the model gives an excellent fit to continuous data measured at roadside. Comparison of model predictions with transects of NO₂ across the greenspace also show generally good model performance. The incorporation of dry deposition processes for the nitrogen oxides into the model leads to a reduction of less than 1% in predicted concentrations, leading to the conclusion that the cleaner air within urban greenspace is primarily the result of dispersion rather than deposition processes.

Keywords Nitrogen oxides · Ozone · Urban greenspace · Dispersion model · Diffusion tubes · ADMS

1 Introduction

Numerous studies have demonstrated the health benefits of urban greenspace (Richardson and Mitchell 2010; Kondo et al. 2018). These are largely associated with the psychological benefits associated with open air and nature, and the physical benefits accruing from activity and exercise. A further benefit derives from the lower air pollutant concentrations in urban parkland. As many urban parks are located close to heavily trafficked roads, a key issue is the dispersion of traffic pollution into parks, and the gradient in concentrations away from the road. In this context, the pollutants

of greatest concern are likely to be particulate matter and nitrogen dioxide. In the case of the former, the roadside increment above background tends to be quite small due to the large regional background, while for NO₂, the regional background is less substantial and a large increment exists at roadside (Harrison et al. 2021). The decay of this roadside increment as air advects into the park is a key issue in determining human exposure, and in urban design to minimise use of more polluted locations for exercise. Research on PAH, for example, has shown that sporting walkways adjacent to roads are subject to traffic pollution (Alghamdi et al. 2021).

Road traffic is a major source of NO_x, which comprises both nitric oxide (NO) and nitrogen dioxide (NO₂). Only the latter is considered toxic and a risk to public health. Emissions from traffic comprise mostly NO, but over recent years there has been a substantial increase in the proportion of NO₂ to around 25% of NO_x, fleet averaged, which more recently has been declining (Carslaw et al. 2019). NO₂ is also formed from the oxidation of NO by ozone (O₃),

✉ Roy M. Harrison
r.m.harrison@bham.ac.uk

¹ School of Geography, Earth and Environmental Sciences, University of Birmingham, Edgbaston, Birmingham B15 2TT, UK

² Department of Environmental Sciences/Center of Excellence in Environmental Studies, King Abdulaziz University, PO Box 80203, Jeddah 21589, Saudi Arabia

typically leading to an increasing $\text{NO}_2:\text{NO}_x$ ratio with distance from the road.

Numerical modelling of traffic-generated NO_2 concentrations requires optimisation of a number of separate factors. These include the NO_x source term which depends upon the traffic flow, the mix of vehicle types, and the mean speed of the vehicles. If hourly data are to be calculated, a diurnal variation of the emissions has to be input. It is necessary to specify the proportion of NO_2 in the exhaust gases (fleet average), and the model utilised in this work (ADMS-Roads) estimates the subsequent conversion of NO to NO_2 and hence the atmospheric $\text{NO}_2:\text{NO}_x$ ratios. ADMS also models the dispersion of NO_x using an advanced Gaussian formulation which requires the input of meteorological data, explicitly the wind speed and direction, temperature and relative humidity, and cloud cover, used to estimate atmospheric stability. There may be a need to optimise the wind data. The influence of dry deposition on concentrations was also evaluated using the model. Most studies of roadside dispersion of pollutants use Gaussian plume formulations, although data-driven (Artificial Intelligence) approaches are now available for use in situations, unlike this study, in which a large pre-existing air quality training dataset is available (Li et al. 2020; Vairo et al. 2020; Samal et al. 2020). There are also other situations, such as transfer of hydrocarbons from refineries which require high quality model simulations to estimate ground-level exposures (Milazzo et al. 2017; Ancione et al. 2021).

In this study, hourly measurements were made of NO, NO_2 and O_3 at a roadside location by a major highway, and downwind in the centre of an area of greenspace. These were supplemented by diffusion tube measurements of 14- to 21-day average nitrogen dioxide at multiple locations at different distances from the highway. The sensitivity to the various input variables was evaluated and the model optimised to simulate the distribution of NO_2 concentrations in the predominantly downwind direction. This paper provides a case study of model construction and optimisation.

2 Experimental

2.1 Site Location and Sampling Sites

The site location is shown in Fig. 1. This domain is in a suburban area to the south of the centre of the city of Birmingham, UK, a city of 1.1 million population in a contiguous conurbation of 2.5 million.

All air sampling was conducted within the University of Birmingham campus. The BROS (roadside), EROS and Biosciences (background) sites were equipped with continuous analysers for oxides of nitrogen and ozone. NO, NO_2 , and NO_x were measured using chemiluminescence analysers,

type 42i from Thermo Scientific, and 42C type from Thermo Environmental Instruments. These were calibrated each week with NO calibration gas at multiple concentrations and intercompared one with the other periodically. Calibration and sensitivity adjustments were made according to BS EN 14211:2012. O_3 was measured using UV absorption analysers, type 49i from Thermo Scientific. These are based upon a photometric principle and should not need calibration but were checked periodically for drift using ozone generated from a Pen-ray lamp, and adjusted according to protocol EN 14625:2012. The analyser inlets were at a height of 3.2 m above ground-level, and for the BROS site at a horizontal distance of 3.6 m from the nearest vehicles. The continuous data were collected from 2 to 11 February 2018. Subsequently, continuous data were collected at the University Biosciences building.

NO_2 was also measured in this study using diffusion tubes of the Palmes type. This type of diffusion tube consists of a plastic tube, two stainless steel grids (#100 mesh) with absorbent reagent and two end caps, one of which is removed during air sampling. NO_2 in the ambient air is captured by the triethanolamine (TEA) absorbent (20% in water) which coats the grid at the inner end of the tube. After exposure, nitrite (NO_2^-) which is collected on the grids is extracted and reacted with reagents (sulfanilamide and *N*-(1-naphthyl)-ethylenediamine dihydrochloride (NEDD)) to form a purple solution. The intensity of the colour of the purple solution is measured by a spectrophotometer and the concentration of nitrite in the sample is obtained using calibration from standard nitrite solutions. Then, the average ambient concentration of NO_2 can be calculated (Bryan and Grisham 2007; Patton and Kryskalla 2011; Targa and Loader 2008).

Preparation and analysis of diffusion tubes were conducted in a laboratory at the University of Birmingham (UK). The method used for the preparation of the sample solutions and the standard nitrite solutions for the UV-Vis analysis is taken from Targa and Loader (2008). NO_2^- measurements were performed by UV-Vis analysis, using a Spectrometer Jenway 6800, scanning the wavelength from 530 to 550 nm, and taking the optimum absorbance at 540 nm. Temperature-dependent diffusion coefficients were calculated according to Targa and Loader (2008) and used to convert nitrite concentrations to NO_2 in air, using also the dimensions of the tube. Diffusion tubes were deployed in triplicate at each site and the mean concentration used, after elimination of outlier data according to relative standard deviation (RSD) as recommended by Targa and Loader (2008) using a criterion of 15% to identify outliers. Additionally, since diffusion tubes are well known to be subject to bias, a bias correction was calculated using tubes exposed alongside the continuous analysers by regression of the diffusion tube data upon the automatic analyser data. The tubes

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Fig. 1 The study domain. The red lines indicate roads, with the largest source being the heavily trafficked A38 road which runs almost east–west near the bottom of the map. Sampling sites are indicated

by blue squares. The automatic analyser stations at BROS, EROS and Biosciences are labelled, and the other sampling sites which had diffusion tubes for NO_2

were deployed at a height of 2.6–3.2 m. The modelling was carried out on diffusion tube deployments from 18 March to 15 April 2018, 15 April–13 May 2018, 21 October–20 November 2018, 20–28 November 2018 and 28 November–6 January 2019.

2.2 Model Construction and Optimisation

A modelling study of the dispersion process was carried out using ADMS-Roads (version 4.1) to simulate the processes of dispersion and deposition of pollutants. The model is a new generation Gaussian plume model, meaning that atmospheric stability is categorised by use of two parameters, the boundary layer depth and Monin–Obukhov length rather than the Pasquill–Gifford stability categories. Dispersion under convective meteorological conditions uses a skewed Gaussian distribution. The full details of the mathematical formulation are available from the model developers (CERC 2021a). The model has been extensively validated against real world datasets (CERC 2021b), although others have argued that the Gaussian formulation can be improved by reformulation (Chen and Broday 2020). It was selected as it is widely used internationally and performs well when compared with other models with similar capabilities (CERC 2021b). The model includes a chemistry module which accounts for conversions within the NO/NO₂/O₃ system. This is a complex reaction system with many interactions (Sillman 1999). The rapid photochemistry in the roadside environment can be described more simply, and ADMS-Roads uses the Generic Reaction Set of eight reactions proposed by Venkatram et al. (1994) to simulate the transformations within the system. The meteorological data used as input in the model were obtained from the nearest weather stations. Following the recommendation of the model developers to assume level terrain if slopes were less than 1 in 10, no terrain corrections were applied. The model validation was performed initially using the observation data collected from the NO_x and O₃ automatic analysers during the period from 2 to 11 February 2018 at BROS and EROS. The NO₂ concentrations from diffusion tubes measured at 14 points in the University of Birmingham site were then compared with the modelling results.

2.2.1 Source Strength

In ADMS-Roads, the pollutant sources are divided into road sources and industrial sources. For the road sources, the road elevation, width, and canyon height are inputted. The pollutant emissions can be set manually or can be estimated by the software based on the input for the traffic flows, which includes average speed in km/hour and the vehicle count/hour in each vehicle category. The emission factor (g km⁻¹) and emission rate (g km⁻¹ s⁻¹) are automatically calculated

by the model based on the input data. The EFT (Emission Factors Toolkit) was used to calculate the emission rates at the location for a specific year, vehicle speed, road type (e.g., urban, rural or motorway) in each vehicle category (e.g., light duty, LDV, and heavy-duty vehicles, HDV).

In the model, roads are designated as line sources. Line sources near the receptors are included explicitly in the model, and roads far from the receptors are considered only through their contribution to the background (Heist et al. 2013). Emissions rates are derived using data on vehicle speed, vehicle type, number of vehicles, etc. Traffic flow data were generated from the Department for Transport (<https://roadtraffic.dft.gov.uk/local-authorities/141>) based upon vehicle count (Average Annual Daily Flow—AADF) for the roads with data available. Each road was divided into segments in terms of road width (m) and speed in kilometres per hour (km h⁻¹), shown in Fig. 1. A list of road links appears in Table S1. The diurnal variation of traffic flows was taken from TRA0307 (Figure S1), vehicle traffic distribution data from the Department for Transport, UK. These differ for weekdays, Saturdays and Sundays and are shown in Table S2.

There are eight roads identified, as shown in Fig. 1 and Table S1, and only two locations have the Dept. of Transport AADF data, ID numbers 81576 and 81577. For roads with no AADF data, the result from manual one-hour observation data on vehicles count was used, after extrapolation to 24 h based upon the diurnal profiles in TRA0307. The average hourly vehicle flows for the different roads appear in Figure S2. The speed of vehicles near traffic lights was set in the model at the lowest speed, 5 km h⁻¹, and sensitivity analysis of this value was conducted. In other locations, vehicle speeds were measured by manually timing vehicles over fixed distances. A default value of primary NO₂:NO_x in the fleet average exhaust emissions of 23.8% (molar) was adopted initially and varied subsequently in a sensitivity test.

2.2.2 Meteorology

The meteorological data should ideally be generated from the nearest weather station, and data for wind direction, wind speed, temperature, and relative humidity (RH) were taken from the nearest weather station, which is Winterbourne No. 2 (1.0 km from BROS) when available, or when unavailable, other sites at Coundon (27.0 km), Coleshill (16.5 km) or Elmdon (12.7 km) had data available. Cloud cover data were only available from the Coleshill weather station.

For the surface roughness, a value of 0.5 m was selected which corresponds to parkland and suburban areas. A sensitivity study was conducted in which roughness lengths of 1.0 m (recommended for cities and woodland) and 1.5 m (large urban areas) were also modelled.

2.2.3 Background Concentrations

Background concentrations of pollutants were obtained from the nearest air quality monitoring station, which was the EROS site (when available), Biosciences site, or Birmingham, Acocks Green, a national network (AURN) urban background site. Data from the EROS site were preferred as this site is within the model domain and shown by modelling not to be greatly affected by emissions from the local highways. The Acocks Green site is 7 km from EROS, and a comparison of data showed that it typically measured higher concentrations than EROS, especially for wind sectors with a northerly component.

2.3 Statistical Evaluation of the Model

Pollutant concentrations from on-site measurements were compared to concentrations from the modelling results during the specific sampling time. Fractional bias (FB), normalised mean square error (NMSE) and correlation coefficient (R) are statistics used to evaluate model performance (Briant et al. 2013; Chang and Hanna 2004; Dédéle and Miškinytė 2015; Heist et al. 2013; Hirtl and Baumann-Stanzer 2007; Hood et al. 2018; Owen et al. 1999):

$$\text{Fractional bias, } FB = \frac{(\bar{C}_o - \bar{C}_m)}{0.5(\bar{C}_o + \bar{C}_m)}, \quad (1)$$

$$\text{Normalised mean square error, } NMSE = \frac{(\overline{C_o - C_m})^2}{\bar{C}_o \bar{C}_m}, \quad (2)$$

$$\text{Correlation, } R = \frac{1}{n-1} \sum_{i=1}^n \left(\frac{C_o - \bar{C}_o}{\sigma C_o} \right) \left(\frac{C_m - \bar{C}_m}{\sigma C_m} \right), \quad (3)$$

where, C_o is the observed value of pollutant concentration; C_m is the modelled value of pollutant concentration; overbar C_o is the average of observed values of pollutant concentration; overbar C_m is the average of modelled values of pollutant concentration; σC_o is the standard deviation of observed concentrations and σC_m is the standard deviation of modelled concentrations.

2.3.1 Fractional Bias (FB)

FB is mean systematic bias, reviewing the mean difference between the observed and modelled values, and the ideal value of FB is zero (Chang and Hanna 2004; Heist et al. 2013; Hood et al. 2018). A negative FB means the model

over-estimates the measured data, and positive FB means the measured data is under-estimated (Hirtl and Baumann-Stanzer 2007).

2.3.2 Normalised Mean Square Error (NMSE)

NMSE measures the overall deviations, reviewing the systematic and random errors. The ideal value of NMSE is zero (Chang and Hanna 2004; Heist et al. 2013; Hood et al. 2018).

2.3.3 Coefficient of Correlation (R)

Coefficient of correlation demonstrates the strength of correlation and linear relationship between the observed and predicted data (Chang and Hanna 2004; Heist et al. 2013; Hood et al. 2018). The range of R is between -1 and 1 . The value near -1 or 1 indicates there is a strong relationship between the modelled and the observed data.

2.3.4 Coefficient of Determination (R^2)

Coefficient of determination represents the total variation of data that one variable can predict in the other variable's data, in range value $0-1$.

3 Results and Discussion

The flow chart for the model design and optimisation is shown in Fig. 2. The model was set up initially to estimate hourly average concentrations which could be compared with the concentrations measured with the continuous analysers at the BROS and EROS sites. There was a sequence of optimisation through sensitivity studies.

3.1 Meteorological Data

The first task was to select meteorological data, and data were available from four sites within the local region. An intercomparison of data showed a good agreement between all weather stations for wind direction, temperature and RH, but not for wind speed. When available, this was taken from Winterbourne No. 2, which is the closest weather station to the sampling area, at a distance of only 1.0 km from the BROS sampling location. A sensitivity study showed a good fit between modelled and measured NO_x as evidenced by FB and NMSE when using wind data from this site. Wind data for other sites gave low estimates of concentration, with Coundon the closest fit. For periods when data were unavailable for Winterbourne No. 2, data from the Coundon station were adopted after correction to Winterbourne No. 2 equivalent values

Fig. 2 Flow chart of model construction and optimisation

using a regression equation derived from 30 months (January 2016–June 2018) of simultaneous data from the two sites:

$$y = 1.5101x + 0.5464 \text{ m s}^{-1}$$

in which y is Coundon and x is Winterbourne wind speed.

The sensitivity study was run with roughness lengths of 1.0 m and 1.5 m applied to both the meteorological station and field site data to compare with the baseline simulation of 0.5 m, and applied to the periods of 15 April–13 May 2018 and 20–28 November 2018. In both cases of larger roughness lengths, the model predicted higher concentrations of NO_x within 10 m of the road, and reduced concentrations subsequently. The near road increase in NO_x concentration was up to 3.5% at 3.6 m for a 1.0 m roughness length and a decrease of generally < 1% beyond 10 m. The corresponding figures for a roughness length of 1.5 m were +6.5% at 3.6 m and generally < -2% at > 10 m downwind distance. We therefore conclude that the model is not strongly sensitive to the choice of roughness length.

3.2 Background Concentration

The next sensitivity test was to the background concentration which forms a major part of the measured concentration at the BROS site. As pollutant concentrations decline quite sharply with distance from a road, it was decided to test the use of EROS as a background as this is in the centre of the greenspace and well separated from any road. The model was run using EROS data as background, and the concentrations predicted for EROS showed low FB and NMSE indicating a good model fit and confirming that the road network surrounding the greenspace had a low impact at this location. It was, therefore, decided to use EROS concentrations as the background for modelling periods when such data were available. For most of the periods with diffusion tube measurements, this was not the case and data from another site had to be selected. In most cases, this was the Biosciences site within the University campus, but not directly on the transect from BROS to EROS (see Fig. 1).

Data were also available from one national network (AURN) urban background site within Birmingham, Acocks Green. This showed generally higher concentrations than those measured at the EROS and Biosciences sites.

3.3 Vehicle Emissions

The next input to be optimised was the vehicle speeds. These are derived from measured data, except in the vicinity of road junctions, where average speeds are hard to measure. For these locations, model runs were conducted with minimum average vehicle speeds of 5, 8 and 10 km h⁻¹, and judging from the low FB and NMSE in the prediction of NO_x concentrations at BROS seen in Table 1, the best fit was for a minimum vehicle speed of 10 km h⁻¹ which was adopted for further runs. The model was then run to predict NO₂ concentrations at BROS and showed a substantial over-estimation, by about 20%. This led to a sensitivity study in which the NO₂:NO_x ratio was varied, taking values of 10, 15 and 20%, all below the default value of 23.8% in the model. A ratio of 15% gave the lowest values of FB and NMSE (Table 2) and a regression of modelled (*y*) versus measured (*x*) NO₂ concentrations gave:

$$y = 0.9957x + 0.723 \mu\text{g m}^{-3} \quad R^2 = 0.7745.$$

A comparison of modelled with measured data appears in Fig. 3 and shows that the model now captures the main features of the variations in nitrogen dioxide.

The best fit for NO₂:NO_x ratio can be tested with real data. NO₂:NO_x ratio can be estimated by calculating the relative concentration of oxidant per unit NO_x, (NO₂ + O₃)/NO_x, at near road and background sites (Carslaw et al. 2016;

Jenkin 2004). This is based upon the concept that although there may be oxidation of NO, and hence an increase in NO₂, this is exactly compensated in molar terms by a decrease in O₃, and hence levels of O_x are conserved in the background air, and any increment in O_x between the roadside and downwind site is due to the primary emission of NO₂ (Table 3).

ΔO_x is due to the primary emission of NO₂, as the time-scales are too short for significant formation of ozone from photochemistry. Concentration changes (Δ) between EROS (taken as representative of the background) and BROS were:

$$\Delta\text{O}_x = 3 \text{ ppb}$$

$$\Delta\text{NO}_x = 22 \text{ ppb}$$

$$\text{Primary NO}_2 = \frac{\Delta\text{O}_x}{\Delta\text{NO}_x} \times 100\% \quad (4)$$

$$\text{Primary NO}_2 = (3/22) \times 100\% = 13.6\% \approx 14\%.$$

The result suggests that the primary NO₂ level is approximately 14%, and highly consistent with that estimated from optimisation of the model.

Table 3 shows that concentrations of ozone changed little between the BROS and EROS sites, with just a small decline due to reaction with NO, and hence model results are not shown for this pollutant, although it was a key input to the modelling.

3.4 Dry Deposition

Thus far, the model had run without inclusion of dry deposition, a reasonable approximation due to the relatively short distances of travel. Deposition processes were however

Table 1 Model evaluation of different vehicle minimum speeds

Setting minimum vehicle speed (km h ⁻¹)	Pollutant	Measured mean (μg m ⁻³)	Modelled mean (μg m ⁻³)	SD measured	SD modelled	FB (fractional bias)	NMSE	<i>R</i>	<i>R</i> ²
5	NO _x BROS	69.01	76.88	46.62	58.28	-0.11	0.012	0.78	0.61
8	NO _x BROS	69.01	67.81	46.62	49.82	0.02	0.000	0.80	0.64
10	NO _x BROS	69.01	65.32	46.62	47.41	0.05	0.003	0.80	0.64

Table 2 Model evaluation of different setting NO₂:NO_x ratio

Setting NO ₂ :NO _x ratio (%)	Pollutant	Measured mean (μg m ⁻³)	Modelled mean (μg m ⁻³)	SD measured	SD modelled	FB (fractional bias)	NMSE	<i>R</i>	<i>R</i> ²
10	NO ₂ BROS	28.95	27.74	15.61	14.16	0.04	0.002	0.89	0.79
15	NO ₂ BROS	28.95	29.55	15.61	15.55	-0.02	0.000	0.88	0.77
20	NO ₂ BROS	28.95	31.37	15.61	16.97	-0.08	0.006	0.87	0.76

NO₂:NO_x ratio Graph of NO₂ observation and NO₂ modelled at BROS

Fig. 3 Measured and modelled concentrations of NO₂ using different proportions of primary NO₂ in the model

included as a sensitivity investigation using the values of deposition velocity for NO and NO₂ available as a default in ADMS-Roads. Deposition of O₃ was not accounted for as the ADMS model does not allow inclusion of deposition for secondary components. Deposition velocities in the ADMS-Roads model were set at 0.0015 m s⁻¹ for NO₂, and 0.00015 m s⁻¹ for NO as recommended by CERC in normal runs. As a test of sensitivity to dry deposition, the deposition velocity of NO₂ was varied between 0.00067 and 0.005 m s⁻¹ based upon a survey of literature.

Using as input data the period of automatic analyser measurements from 2 to 11 February 2018, the largest reduction in NO₂ concentration was 1.45% when dry deposition was implemented at the maximum.

3.5 Modelling the Long-Term Data

Diffusion tube samplers were deployed over periods of one or more weeks, and hence were not suitable for direct comparison with hourly data from the model. The hourly resolution in the

Table 3 Mean pollutant concentrations at BROS and EROS sites

Pollutant	Mean concentration $\pm$ uncertainty (ppb)		Δ (ppb) [BROS] – [EROS]
	BROS	EROS	
NO	20 ± 1	2 ± 0.3	18 ± 3
NO ₂	14 ± 1	10 ± 0.3	4 ± 0.3
NO _x	34 ± 2	12 ± 1	22 ± 2
O ₃	17 ± 1	18 ± 1	-1 ± 0.1
O _x (NO ₂ + O ₃)	31 ± 3	28 ± 2	3 ± 0.3

Uncertainty is expressed by the standard error of the mean, with that for the difference combining the uncertainties in both means

model is needed to account for diurnal changes in emissions and meteorology, and therefore, the hourly model outputs were averaged over the full period of a diffusion tube deployment by selecting the long-term (LT) option on the output tab in the model to obtain the average model outputs for each receptor. When available, background estimates from the EROS site were used, and in other cases, these were taken from the Biosciences site within the campus, or the national network (AURN) station at Acocks Green.

A typical result from the modelling is shown in Fig. 4 which compares the model output with the diffusion tube measurements. It is from the period 15 April–13 May when the winds were predominantly south-westerly, which advects pollution from the road across the campus green space. The model fit to the data is generally good, with a regression of:

$$y = 0.82x + 5.12 \mu\text{g m}^{-3}; \quad R^2 = 0.71,$$

Fig. 4 Transect of modelled and measured NO₂ (15 April–13 May 2018) **Fig. 5** Transect of measured and modelled NO₂ (20–28 November 2018)

where y is the modelled concentration and x is the measurement. The absolute differences between the model and measurements are for most sites $< 2 \mu\text{g m}^{-3}$, and at their greatest $< 5 \mu\text{g m}^{-3}$. Hence the large intercept of $5.1 \mu\text{g m}^{-3}$ is rather surprising, but results from the large extrapolation from the data to the y -axis.

During the period 20–28 November, the winds were predominantly from the north-east, and hence, the pollution from the major A38 highway was mainly carried away from the greenspace. The results shown in Fig. 5 show an elevation in measured concentrations at the sites furthest from the A38 (between EROS and site I) indicating a likely underestimate of emissions from the roads to the north and east of our site, from which only manual traffic measurements were available. In this case, the regression fit also showed an appreciable intercept:

$$y = 1.01x - 3.57 \mu\text{g m}^{-3}; \quad R^2 = 0.47,$$

where y is the modelled value and x is the measurement.

3.6 Concentration Decay

Both modelled and measured concentrations of NO₂ concentrations declined from roadways to background locations within a distance of around 100 m. This result is similar to the findings of other researchers who have found NO₂ concentrations decrease to near background within 150–200 m from a road (Gilbert et al. 2003; Karner et al. 2010; Ducret-Stich et al. 2013).

4 Conclusions

There are few such tests of model set-up and optimisation relating to a complex road layout of this kind, and few relating to pollutant dispersion into greenspace. Several of the input variables are fixed, but many require optimisation. Traffic volumes derive from direct measurements, as do most traffic speed data. However, the main roadside sampling location was close to a road junction, and as mean traffic speeds in such a location were not easy to measure, optimisation of the speed close to road junctions was important. In addition to getting the emissions right, good input meteorological data are crucial. Using data from a nearby station is obviously advantageous, and fortunately this was available for most of the time. A notable feature was that wind speeds at the nearby Winterbourne station were lower by about 35% than those measured at the best of the other sites with available data. This is presumably due to the drag of urban structures leading to reduced wind speeds within the urban area. When the wind speed data from a site outside the city were used, notably better predictions were obtained if the wind speeds were reduced to equivalent Winterbourne speeds. The choice of background air quality data is crucial, as incorrect data will cause a bias irrespective of the quality of the dispersion model. Optimisation of all of these variables was conducted using measured NO_x concentration data. NO_x is a conserved species over the short timescales and hence is a good test of the quality of the dispersion characteristics of the model.

Another key input is the traffic-emitted $\text{NO}_2:\text{NO}_x$ ratio. This was optimised independently by tuning the model performance for NO_2 , once the performance for NO_x was shown to be good. One benefit of the experimental design was that the measured data allowed an independent estimate of this ratio, which emerged to be very close to that derived by optimising the model. This is an important variable, likely to change further as the vehicle fleet evolves with time.

Modelling the full transect across the greenspace proved more challenging for the model than the single site study. However, the model showed good skill, with most absolute model values close to those measured. Extrapolation of the regression to give an intercept could, however, be quite misleading due to the clustering of data points in a relatively small area of the graph. Overall, the results give confidence that with care taken over inputs, the model is highly skillful.

Another key area of investigation was the extent to which dry deposition influences measured concentrations within the greenspace. The model results give a clear indication that the decline in concentrations towards the centre

of the greenspace is predominantly the result of dispersion rather than deposition processes. This is consistent with the findings of Jeanjean et al. (2016) and Xing and Brimblecombe (2019) who both concluded that the deposition process influenced concentrations far less than dispersion. However, there are scenarios where dry deposition may be more significant, for example on the scale of an entire city (Tiwari and Kumar 2020), or if the greenspace contained trees which would enhance the deposition process.

Supplementary Information The online version contains supplementary material available at <https://doi.org/10.1007/s41748-021-00262-1>.

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Declarations

Conflict of interests The authors do not have any competing financial interests.

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