A Novel Approach to Improve the Air Quality Predictions of Air Pollution Dispersion Modelling systems

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ABSTRACT: The aim of this research paper is the introduction of a novel mathematical approach to improve the accuracy of the results of air pollution dispersion models based on the calibration of input background concentrations. Using the Dunkirk area of the City of Nottingham in the UK as a case study, an air pollution model in ADMS-Roads was created for developing the mathematical approach. The iterative application of this approach to the input background concentrations effectively reduced the error between not only the annual mean, but also the hourly, calculated and monitored air pollution concentrations. The traffic flow profiles of the modelled road network were included in the air pollution model and their impact on the model results, after the application of the calibration approach, was investigated. The inclusion of the traffic flow profiles reduced further the error between the hourly, but not the annual means of, calculated and monitored concentrations.

Key words: Calibration, Validation, Background concentrations, Modelling and air pollution

INTRODUCTION

Modelling the air quality is a powerful technique that can be used to assess the ambient air quality against the mandatory air quality standards. In addition, it can be used to assess the effectiveness of the proposed Air Quality Action Plans (AQAPs) in improving the air quality within areas in which air pollution exceeds the national air quality standards. This technique can also be used as a tool to undertake a strategic air quality assessment for a wide range of plans and programmes, including local transport plans (NCC and NCC, 2006). As the majority of national air quality standards are in the form of annual mean and hourly objectives (AEA, 2010), this requires accurate annual mean and hourly air quality predictions.

The results of air pollution dispersion modelling should be accurate enough to provide reliable air quality predictions. Recent air pollution dispersion modelling research assesses the validation of air pollution models by the determination of the error between calculated and monitored air pollution concentrations. However, this recent research has not investigated potential sources of this error so that it can be minimised (Majumdar *et al.,* 2009; Cai and Xie, 2010; Ginnebaugh *et al.,* 2010; Jain and Khare, 2010; Parra *et al.,* 2010).

Nottingham City Council compared the monitored annual mean NO_2 concentrations at three continuous monitoring stations to the calculated concentrations by ADMS-Urban. The model overestimated the annual mean of monitored concentrations at the three sites (PCS, 2008). Therefore, the model results were multiplied by an adjustment factor, the average ratio of monitored to calculated annual mean concentrations at the three monitoring sites, to correct the annual mean results of the model. This might help to improve the annual mean results; however it did not improve the hourly calculated results of the model.

Namdeo et al. (2002) used the hourly predictions of ADMS-Urban and the hourly observations for the first half of 1993 to derive a multiplicative adjustment factor. The factor was applied to the air quality predictions for the second half of 1993 and the adjusted predictions were compared to the corresponding observations. This approach improved the long-term results over the second half of 1993; however it did not show how much improvement was achieved on the short-term level. In addition, Cambridge Environmental Research Consultants (CERC), the developers of ADMS software, have recommended that modellers should avoid the application of such an adjustment factor to the model

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results (CERC, 2009). Instead, CERC advised that various details of the model set-up, such as input data and modelling options, should be adjusted until the calculated results fit the monitored concentrations.

DEFRA (2009) stated that the NO_X (not NO_2) concentrations should be verified and adjusted if NO_2 results of the model disagree with the monitored concentrations. It also commented that "The adjustment of NO_X is often carried out on the component derived from local Road Traffic Emissions – the Road Contribution". This is because the source contribution is often small compared with the background contribution. Therefore, Nottingham City Council used this approach to verify the annual mean NO_2 results of ADMS-Urban (PCS, 2010).

ADMS-Urban was used to predict the annual mean road contribution NO_X concentrations. For each monitoring site, the annual mean background NO_X was estimated from the national background maps and subtracted from the monitored total NO_X. This resulted in the monitored annual mean road contribution NO_X which was compared to the results of ADMS-Urban for each monitoring site to derive an average adjustment factor. The results of ADMS-Urban were multiplied by this factor, and the adjusted results of NO_X were used, along with the background NO_2 concentrations, to derive the adjusted calculated total annual mean NO_2 concentrations by using the LAQM Tools – NO_X to NO_2 spreadsheet (DEFRA, 2010).

This approach did not eliminate the error between the calculated and monitored annual mean NO_2 concentrations. This is probably due to inaccuracy in the monitored annual mean road contribution NO_X, caused by inaccuracy in the estimation of the annual mean background NO_X from the national background maps. In addition, the simple NO_X to NO_2 spreadsheet is usually imprecise, and using a chemistry scheme to model the atmospheric chemical reactions of NO_X, and derive the oxidised NO_2 proportion, is recommended (CERC, 2009). Moreover, this verification approach is only suitable for the calculated annual mean concentrations and is not applicable to the short-term, e.g. hourly, concentrations (CERC, 2009).

Li et al. (2010) adjusted the air pollution model set-up by the calibration of emission rate inputs to the model through the application of a genetic algorithm. This was helpful to reduce the uncertainties existing in air pollution emission inventories such as those relevant to traffic emission factors (Belalcazar *et al.,* 2010). The calibration of input emission rates slightly reduced the error (by 6.46%) between daily calculated and monitored PM_{10} concentrations over eight days. This implies a non-significant reduction in the error

between hourly calculated and monitored concentrations over a large time period such as a full meteorological year. Furthermore, no validation was undertaken for the output results of the model, calculated using the calibrated emission rates, against monitored concentrations at monitoring sites independent of the calibration process. This process also required a very expensive computing time, due to the use of a genetic algorithm, which may extend to several weeks on a single PC before the actual running of the air pollution model, which may extend to several days to model the air pollution dispersion in a study area (Barrett and Britter, 2008; Barrett and Britter, 2009). Therefore, this research paper introduces a mathematical approach for adjusting the model set-up by the calibration of input background concentrations, in order to improve significantly the accuracy of the model results and reduce the computing time. This includes the introduction of four new concepts to the science of air pollution dispersion modelling; namely macro-calibration, macro-validation, micro-calibration and micro-validation. The background concentrations are some of the most important input data to the broad variety of air pollution dispersion models (Venegas and Mazzeo, 2006). They account for all emission sources that may affect the air quality in a model application area, and are not defined explicitly in the air pollution model. Therefore, a great uncertainty exists in input background concentrations which may vary for the same model according to the number of explicitly defined air pollution sources. Consequently, the calibration of input background concentrations is necessary to provide the appropriate background concentrations for a certain model set-up. It may also account for the uncertainties existing in input air pollution emission rates.

In the following sections of this paper, the set-up of the air pollution model of the Dunkirk area in Nottingham is described and the error between calculated and monitored air pollution concentrations is illustrated. Then, the different development stages of the calibration process are discussed, along with the reduction in the error after each stage. The impact of including the traffic profiles of the modelled road network on the error between calculated and monitored concentrations is explained. Finally, the calibration of background concentrations in ADMS-Roads is compared to the use of grid air pollution sources in ADMS-Urban.

MATERIALS & METHODS

As a study area, Dunkirk Air Quality Management Area (AQMA) was used to set-up an air pollution model in ADMS-Roads version 2.3 for the initial development

of the calibration approach. ADMS-Roads was developed by CERC (CERC, 2006). Dunkirk AQMA is an urban study area in the city of Nottingham, as shown in Fig. 1, with NO_2 levels exceeding the permissible levels (PCS, 2001). Therefore, NO_2 was selected as the modelled air pollutant as the majority of the available air pollution monitoring data, required to calibrate and validate the air pollution model, in and around the Dunkirk AQMA was NO_2 data.

2006 was selected as the modelling year of the air pollution model due to data availability for this year. The significant industrial air pollution sources relevant to the Dunkirk AQMA were identified and their emission rates were obtained from Nottingham City Council, which also provided the traffic speed data of the main roads in the Dunkirk AQMA. The emission sources defined explicitly in the air pollution model were the traffic on the main roads within, and close to, the Dunkirk AQMA, as shown in Fig 1, and the relevant significant industrial air pollution sources. The Nottingham Watnall Weather Station (MO, 2010) provided the 2006 hourly sequential meteorological data which included surface temperature, wind speed at 10-metre height above the ground surface, wind direction, precipitation, cloud cover and degree of humidity. The 2006 annual mean and hourly monitored NO_X, NO_2 and O_3 concentrations by the Air Quality Monitoring Station (AQMS), located in the Dunkirk AQMA as shown in Fig. 1, were provided by Nottingham City Council.

The traffic flow data of the main roads in the Dunkirk AQMA were obtained from Nottingham City Council in the form of the traffic count every five minutes collected automatically using detector loops embedded in the main roads. A Visual Basic for Applications (VBA) computer program was written in MS Excel in order to calculate automatically the 2006 Annual Average Daily Traffic (AADT) flow and the 2006 hourly and monthly traffic flow profiles from the five-minute traffic counts. The traffic flow profiles were compiled to a special text file, a FAC file, which was used in ADMS-Roads to reflect the hourly and monthly variations in the AADT flow on traffic air pollution emissions, so that for each hour, the traffic flow, used in the model to derive the traffic emissions, was the AADT flow $\times$ monthly factor $\times$ hourly factor. The 2003 DMRB traffic emission factors (DMRB, 2007), built-in in ADMS-Roads, were used to derive the traffic emission rates from the traffic flow and speed data.

The Chemical Reaction Scheme (CRS) was used to model the atmospheric conversion of NO_X to NO_2 due to a number of chemical reactions with background O_3 (CERC, 2006). Modelling these atmospheric reactions was necessary to get accurate NO_2 results, so NO_X and O_3 were modelled in addition to NO_2. However, using this chemical scheme requires inputs for NO_2,

Fig. 1. The Dunkirk AQMA
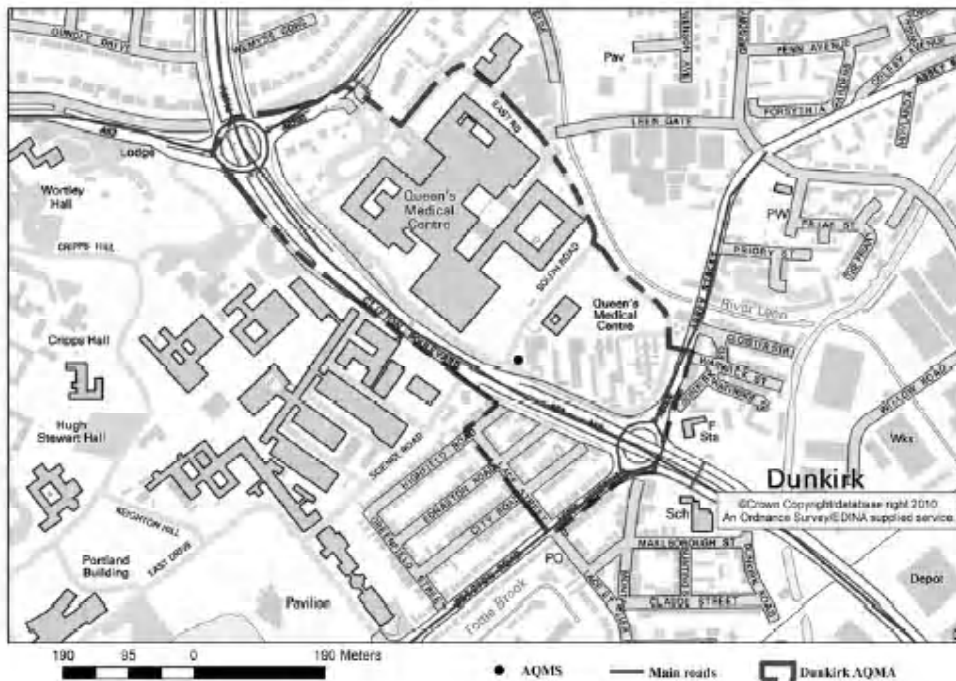
NO_X and O_3 background concentrations. Therefore, Nottingham City Council provided the 2006 hourly sequential NO_2, NO_X and O_3 concentrations monitored by the Rochester air quality monitoring station. This is a rural monitoring station remote from the Dunkirk AQMA and far from urban air pollution, and hence it was recommended to use its monitoring data as the input background concentrations to avoid double counting (CERC, 2009).

Calibration and validation of the background concentrations

An output receptor was defined in the air pollution model at the geographical location of the AQMS. With reference to Run 1 in Table 1, the calculated 2006 annual mean NO_X and NO_2 concentrations underestimated the monitored ones by 37.6% and 25.6% respectively at the AQMS. In addition, the calculated 2006 annual mean of O_3 concentrations overestimated the monitored one by 42.7% at the AQMS. This necessitated developing the set-up of the air pollution model by performing two operations. The first operation was the iterative calibration of the rural background concentrations so as to account for the urban background emissions, e.g. residual, poorly-defined or diffused emissions, from domestic heating sources and minor roads, in the Dunkirk AQMA. The second operation was the validation of the calculated air pollution concentrations after each iteration of the calibration process, in order to decide the final acceptable iteration of this process.

Table 1. Macro-calibration development stages of the rural background concentrations

run 1	Δ background	calculated concentrations	target concentrations
NO_2	0	26.25	35.29
NO_X	0	42.19	67.60
O_3	0	44.23	31.00
run 9	**Δ background**	**calculated concentrations**	**Target concentrations**
NO_2	+7.70	37.27	35.29
NO_X	+25.42	67.61	67.60
O_3	-12.60	28.99	31.00
run 23	**Δ background**	**calculated concentrations**	**Target concentrations**
NO_2	+1.48	35.45	35.29
NO_X	+25.42	67.60	67.60
O_3	-5.40	31.01	31.00
run A	**Δ background**	**calculated concentrations**	**Target concentrations**
NO_2	+7.02	36.89	35.29
NO_X	+25.42	67.61	67.60
O_3	-12.40	28.86	31.00
run B	**Δ background**	**calculated concentrations**	**Target concentrations**
NO_2	+10.12	38.73	35.29
NO_X	+25.42	67.61	67.60
O_3	-13.20	29.46	31.00
run C	**Δ background**	**calculated concentrations**	**Target concentrations**
NO_2	+14.55	41.64	35.29
NO_X	+25.42	67.61	67.60
O_3	-15.30	29.18	31.00
run D	**Δ background**	**calculated concentrations**	**Target concentrations**
NO_2	+17.18	43.56	35.29
NO_X	+25.42	67.61	67.60
O_3	-16.71	28.69	31.00

RESULTS & DISCUSSION

The term macro-calibration in this research paper refers to the adjustment of input background concentrations, so that the error between the annual means of calculated and monitored air pollution concentrations can be effectively reduced. The macro-validation was undertaken by the direct comparison between the calculated and monitored annual means of NO_X, NO_2 and O_3 concentrations at the AQMS. As calculated NO_2 concentrations were linked to calculated NO_X and O_3 concentrations through the atmospheric chemical reactions discussed above, it was decided to calibrate NO_X and O_3, in addition to NO_2, background concentrations which were subsequently entered to the air pollution model to calculate NO_2, NO_X and O_3 concentrations. A trial and error approach was adopted to macro-calibrate the hourly sequential rural background concentrations until the above mentioned macro-calibration criterion was achieved. This approach comprised 23 runs of the model, and involved changing the background concentrations manually every time. In Table 1, the results of an intermediate run (run 9), and the final macro-calibration run (run 23), are shown in order to illustrate the progress of this approach. For each macro-calibration iteration, the values in the Δbackground' field of Table 1 were added to every hour of the 2006 NO_2, NO_X, and O_3 rural background concentrations. However, adding these values to the original background concentrations file resulted in having many consecutive hours with a negative O_3 background concentration which raised an error and interrupted the model run. This technical problem was overcome by replacing the negative, invalid, O_3 background concentrations with zero in the macro-calibrated background concentrations file. Another computer logic was applied to this file in order to preserve the fact that NO_X is $NO + NO_2$. Hence, for every hour in the macro-calibrated background concentrations file, if $NO_2 > NO_X$, then re-adjust the macro-calibrated background NO_2 concentration so that $NO_2 = NO_X$. This assumed a zero NO background

concentration for violating hours in the macro-calibrated background concentrations file. After each iteration of the macro-calibration, the macro-validation was undertaken by comparing the calculated concentrations and the target concentrations in Table 1. The calculated concentrations were the 2006 annual means of calculated NO_2, NO_X, and O_3 concentrations and the target concentrations were the 2006 annual means of monitored NO_2, NO_X, and O_3 concentrations at the AQMS. Run 23 in Table 1 gave the least error between the calculated and target concentrations. Therefore, the background concentrations corresponding to this run were considered the final macro-calibrated background concentrations. The values corresponding to the final macro-calibration run in Table 1 were used to derive Equations 8, 9 and 10, which could be used to evaluate directly the background concentration adjustment values, required to macro-calibrate the rural background concentrations of the Dunkirk AQMA air pollution model, without the trial and error approach:

where $\overline{NO_{2\,monitored}}$ is the annual mean of monitored NO_2 concentrations, $\overline{NO_{2\,uncalibrated}}$ is the annual mean of calculated NO_2 concentrations using the rural background concentrations and 9.2 is the difference between the values of calculated annual mean NO_2 concentrations of run 23 (the final macro-calibration run) and run 1 (performed using the uncalibrated rural background concentrations) in Table 1.

where $\overline{NO_{x\,monitored}}$ is the annual mean of monitored NO_X concentrations and $\overline{NO_{x\,uncalibrated}}$ is the annual mean of calculated NO_X concentrations using the rural background concentrations.

$$\Delta NO_{2\,background} = \frac{\left(\overline{NO_{2\,monitored}} - \overline{NO_{2\,uncalibrated}}\right)}{9.2} \times 1.48, \qquad (8)$$

$$\Delta NO_{x\,background} = \overline{NO_{x\,monitored}} - \overline{NO_{x\,uncalibrated}}, \qquad (9)$$

$$\Delta O_{3\,background} = \frac{\left(\overline{O_{3\,monitored}} - \overline{O_{3\,uncalibrated}}\right)}{(-13.22)} \times (-5.40), \qquad (10)$$

where $\overline{O_{3\ monitored}}$ is the annual mean of monitored O₃ concentrations, $\overline{O_{3\ uncalibrated}}$ is the annual mean of calculated O₃ concentrations using the rural background concentrations and -13.22 is the difference between the values of calculated annual mean O₃ concentrations of run 23 (the final macro-calibration run) and run 1 (performed using the uncalibrated rural background concentrations) in Table 1.

The term micro-calibration in this research paper refers to the adjustment of input background concentrations so that the error between not only the annual means of, but also the hourly, calculated and monitored air pollution concentrations can be effectively reduced. The micro-calibration extends the macro-calibration as shown in Fig. 2. The micro-validation was undertaken by comparing statistically two one-dimensional arrays of the 2006 calculated and monitored hourly sequential NO_2 concentrations at the AQMS. Pearson Correlation Coefficient (r) and the Root Mean Square Error (RMSE) were used to compare the two arrays. Further details about these two descriptive statistics are given in Hanna et al. (1991; 1993) and Jain and Khare (2010). The slope of the regression line through the origin was also used to compare the two arrays of hourly calculated and monitored concentrations.

The Dunkirk AQMA air pollution model was run with the uncalibrated rural background concentrations file to output the 2006 calculated hourly NO_2 concentrations at the AQMS. This was carried out for the identification of the initial discrepancy, before any calibration, between the 2006 calculated and monitored hourly NO_2 concentrations at the AQMS, as shown in Fig. 3. Then, the model was run with the macro-calibrated background concentrations file, corresponding to run 23 in Table 1, to output the 2006 calculated hourly NO_2 concentrations at the AQMS. This was for the micro-validation after the macro-calibration of the rural background concentrations as shown in Fig. 4. Pearson's correlation coefficients were calculated as 0.541 before any calibration, and then as 0.412 after the macro-calibration, as shown in Fig.s 3 and 4. The slight decline in Pearson's correlation coefficient after the macro-calibration implied that the macro-calibration slightly decreased the degree of linearity of the actual relationship between the calculated and monitored hourly NO_2 concentrations at the AQMS. Hence, the macro-calibration slightly increased the drift of the shape of this actual relationship away from the perfect straight-line relationship. On the other hand, the values of the RMSE were calculated as 18.45 μg/m³ before the calibration, and then as 17.39 μg/m³ after the macro-calibration, as shown in Figs 3 and 4. The slight decline in the RMSE after the macro-calibration implied that the macro-calibration slightly lowered the difference between the calculated and monitored hourly NO_2 concentrations. Therefore, the macro-calibration not only improved the NO_2 predictions of the model on the macro, annual mean, level but also slightly improved the NO_2 predictions on the micro, hourly, level. The slope of the best fit line through the origin of the actual relationship between the calculated and monitored hourly NO_2 concentrations at the AQMS was calculated as 0.631 before any calibration, and then as 0.755 after the macro-calibration, as shown in Figs 3 and 4. Although the results of the macro-calibration, corresponding to run 23 in Table 1, very slightly overestimated the 2006 annual mean of monitored NO_2 concentrations at the AQMS, the slope of the best fit line through the origin after the macro-calibration was less than 1.0. This indicated that, after the macro-calibration, the model generally underestimated the monitored NO_2 concentrations at the AQMS on the micro, hourly, level. However, the slight increase in the slope of the best fit line after the macro-calibration implied that the macro-calibration slightly reduced the tendency of the model to underestimate the monitored hourly NO_2 concentrations at the AQMS. This, together with the reduction in the RMSE after the macro-calibration, confirmed the slight improvement of the NO_2 predictions of the model, after the macro-calibration, on the micro, hourly, level.

To improve further the NO_2 predictions of the model on the micro level, the idea of micro-calibration was developed. This idea depended on the modification of Equations (8), (9) and (10) in order to generate three one-dimensional arrays for ΔNO_2 background, ΔNO_X background and ΔO_3 background as follows:

where $\Delta NO_{2\ background\ i}$ is the adjustment value for the rural NO_2 background concentration for the hour i. $NO_{2\ monitored\ i}$ is the monitored hourly NO_2

$$\Delta NO_{2\ background\ i} = \frac{(NO_{2\ monitored\ i} - NO_{2\ uncalibrated\ i})}{(NO_{2\ macro\ i} - NO_{2\ uncalibrated\ i})} \times 1.48, \qquad (11)$$

$$\Delta NO_{X\ background\ i} = NO_{x\ monitored\ i} - NO_{x\ uncalibrated\ i}, \qquad (12)$$
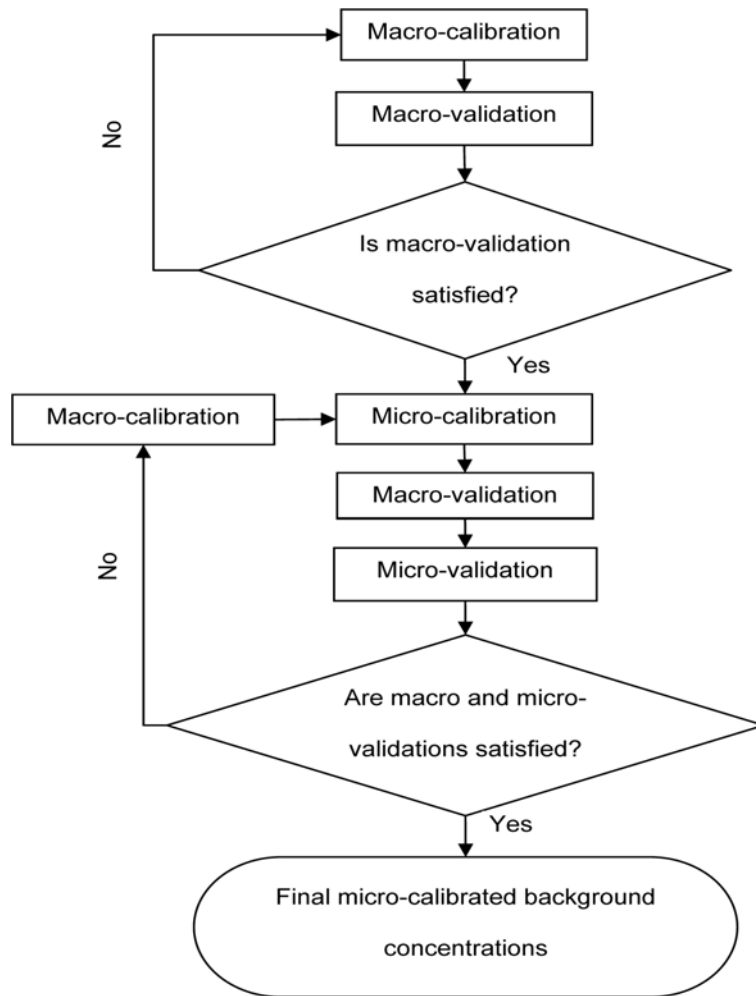
Fig. 2. Calibration and validation process for rural background concentrations
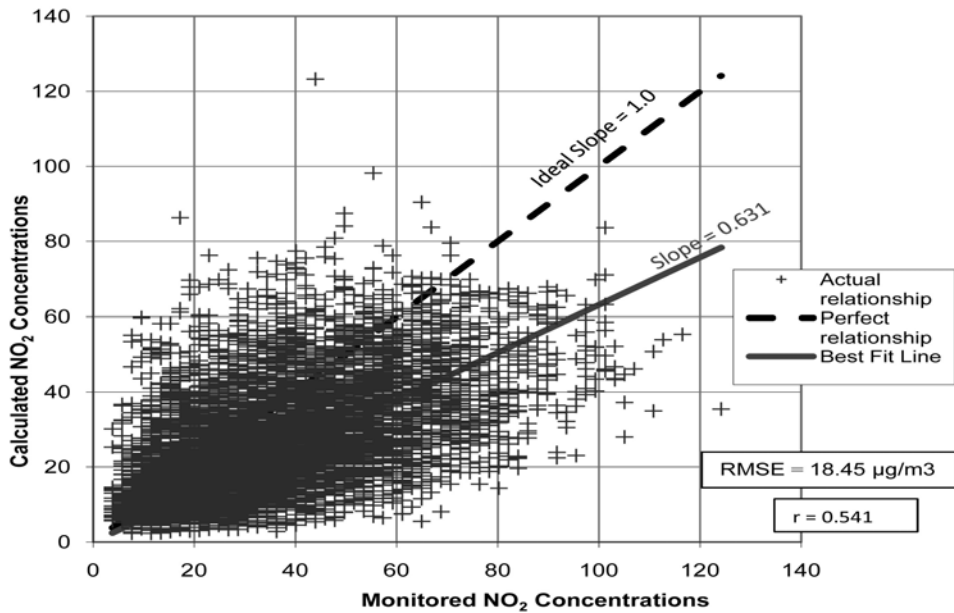

Fig. 3. Scatter diagram of hourly NO$_2$ concentrations at the AQMS before any calibration
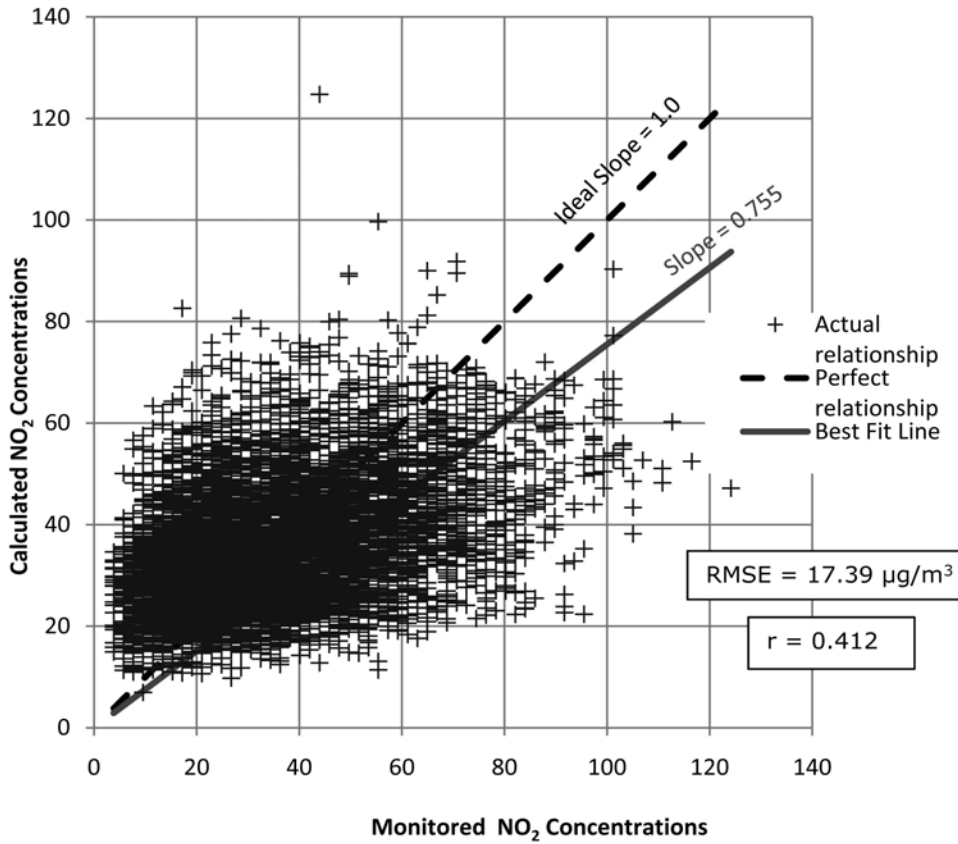
Fig. 4. Scatter diagram of hourly NO$_2$ concentrations at the AQMS after macro-calibration

concentration for the hour i. $NO_{2\ uncalibrated\ i}$ is the calculated hourly NO$_2$ concentration for the hour i using the uncalibrated rural background concentrations. $NO_{2\ macro\ i}$ is the calculated hourly NO$_2$ concentration for the hour i using the macro-calibrated background concentrations. The value of i ranged from 1 to 8760, which was the total number of hours in the year 2006. 1.48 is the macro-calibration adjustment value for the rural NO$_2$ background concentrations, as given in the column headed '**D** background' in Table 1 for run 23 (the final macro-calibration run).

where $\Delta NO_{X\ background\ i}$ is the adjustment value for the rural NO$_X$ background concentration for the hour i. $NO_{X\ monitored\ i}$ is the monitored hourly NO$_X$

concentration for the hour i. $NO_{X\ uncalibrated\ i}$ is the calculated hourly NO$_X$ concentration for the hour i using the uncalibrated rural background concentrations. The value of i ranged from 1 to 8760, which was the total number of hours in the year 2006.

where $\Delta O_{3\ background\ i}$ is the adjustment value for the rural O$_3$ background concentration for the hour i. $O_{3\ monitored\ i}$ is the monitored hourly O$_3$ concentration for the hour i. $O_{3\ uncalibrated\ i}$ is the calculated hourly O$_3$ concentration for the hour i using the uncalibrated rural background concentrations. $O_{3\ macro\ i}$ is the calculated hourly O$_3$ concentration

$$\Delta O_{3\ background\ i} = \frac{(O_{3\ monitored\ i} - O_{3\ uncalibrated\ i})}{(O_{3\ macro\ i} - O_{3\ uncalibrated\ i})} \times (-5.4), \tag{13}$$

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for the hour i using the macro-calibrated background concentrations. The value of i ranged from 1 to 8760, which was the total number of hours in the year 2006. -5.4 is the macro-calibration adjustment value for the rural O_3 background concentrations, as given in the column headed Δ background' in Table 1 for run 23 (the final macro-calibration run). The three one-dimensional arrays of $\Delta NO_{2\,background}$, $\Delta NO_{X\,background}$ and $\Delta O_{3\,background}$, calculated by Equations (11), (12) and (13), were added to the arrays of the un-calibrated hourly sequential rural background concentrations of NO_2, NO_X and O_3, respectively. Hence the micro-calibrated background concentrations file was created based on the above three equations. However, running the model with these micro-calibrated background concentrations resulted in the overestimation of the annual means of the monitored NO_2, NO_X and O_3 concentrations at the AQMS as shown in Table 2. In addition, using these micro-calibrated background concentrations increased the difference between the calculated and monitored hourly NO_2 concentrations on the micro, hourly, level. This was indicated by the large increase in the RMSE as shown in Table 2.

A possible reason for the large increase in the RMSE after the micro-calibration based on Equations (11), (12) and (13) was the use of the hourly concentrations calculated using the macro-calibrated background concentrations in these equations. As discussed before with regard to Fig. 4, the hourly calculated concentrations of the macro-calibrated model were not precise enough. The macro-calibrated model of the Dunkirk AQMA was validated only on the macro, annual mean, level. Therefore, instead of using $NO_{2\,macro\,i}$ and , the hourly NO_2 and O_3 concentrations calculated by the macro-calibrated model, it was decided to alter two of the three equations for the micro-calibration of the rural background concentrations, using the annual mean NO_2 and O_3 concentrations calculated by the macro-calibrated model, so that:

where $\Delta NO_{2\,background\,i}$ is the adjustment value for the rural NO_2 background concentration for the hour i.

$NO_{2\,monitored\,i}$ is the monitored hourly NO_2 concentration for the hour i. $NO_{2\,uncalibrated\,i}$ is the calculated hourly NO_2 concentration for the hour i using the uncalibrated rural background concentrations. The value of i ranged from 1 to 8760, which was the total number of hours in the year 2006. $\overline{NO_{2\,macro}}$ is the annual mean NO_2 concentration calculated using the macro-calibrated background concentrations, as given in the column headed 'calculated concentrations' in Table 1 for a particular macro-calibration run. $\overline{NO_{2\,uncalibrated}}$ is the annual mean NO_2 concentration calculated using the uncalibrated rural background concentrations, as given in the column headed 'calculated concentrations' in Table 1 for run 1. $\Delta NO_{2\,macro\,background}$ is the macro-calibration adjustment value for the rural NO_2 background concentrations, as given in the column headed Δbackground' in Table 1 for a particular macro-calibration run.

where $\Delta O_{3\,background\,i}$ is the adjustment value for the rural O_3 background concentration for the hour i. $O_{3\,monitored\,i}$ is the monitored hourly O_3 concentration for the hour i. $O_{3\,uncalibrated\,i}$ is the calculated hourly O_3 concentration for the hour i using the uncalibrated rural background concentrations. The value of i ranged from 1 to 8760, which was the total number of hours in the year 2006. $\overline{O_{3\,macro}}$ is the annual mean O_3 concentration calculated using the macro-calibrated background concentrations, as given

$$\Delta NO_{2\,background\,i} = \frac{(NO_{2\,monitored\,i} - NO_{2\,uncalibrated\,i})}{(\overline{NO_{2\,macro}} - \overline{NO_{2\,uncalibrated}})} \times \Delta NO_{2\,macro\,background} , \tag{14}$$

$$\Delta O_{3\,background\,i} = \frac{(O_{3\,monitored\,i} - O_{3\,uncalibrated\,i})}{(\overline{O_{3\,macro}} - \overline{O_{3\,uncalibrated}})} \times \Delta O_{3\,macro\,background} , \tag{15}$$

Table 2 .Micro-calibration development stages of the rural background concentrations

case description	receptor name	annual mean NOx		annual mean NO₂		annual mean O₃		NO₂ RMSE before calibration	NO₂ RMSE after macro-calibration	NO₂ RMSE after micro-calibration
		calculated	monitored	calculated	monitored	calculated	monitored			
based on equations (11), (12) and (13).	AQMS	73.37	67.60	37.90	35.29	39.43	31.00	18.45	17.39	117.83
based on equations (14), (12), (15) and run 23.	AQMS	68.46	67.60	31.58	35.29	34.99	31.00	18.45	17.39	11.07
based on equations (14), (12), (15) and run A.	AQMS	67.71	67.60	33.03	35.29	33.10	31.00	18.45	17.39	6.63
based on equations (14), (12), (15) and run B.	AQMS	67.55	67.60	33.96	35.29	32.52	31.00	18.45	17.39	5.11
based on equations (14), (12), (15) and run C.	AQMS	67.48	67.60	34.85	35.29	31.47	31.00	18.45	17.39	4.21
based on equations (14), (12), (15) and run D.	AQMS	67.47	67.60	35.19	35.29	30.96	31.00	18.45	17.39	4.09
based on equations (14), (12), (15) and run D with no FAC file.	AQMS	68.65	67.60	35.51	35.29	30.74	31.00	18.45	17.39	5.71

in the column headed 'calculated concentrations' in Table 1 for a particular macro-calibration run.

$\overline{O}_{3\ uncalibrated}$ is the annual mean O_3 concentration calculated using the uncalibrated rural background concentrations, as given in the column headed 'calculated concentrations' in Table 1 for run 1.

$\Delta O_{3\ macro\ background}$ is the macro-calibration adjustment value for the rural O_3 background concentrations, as given in the column headed Δ background' in Table 1 for a particular macro-calibration run.

A VBA computer program was written in MS Excel in order to automate the generation of the three hourly sequential one-dimensional arrays for $\Delta NO_{2\ background}$, $\Delta NO_{X\ background}$ and $\Delta O_{3\ background}$ using Equations (14), (12) and (15). For any hour in the year 2006, if either the calculated or monitored hourly concentration was missing, then the equation relevant to the type of missing concentration would not be usable. This was handled in the VBA computer program as follows:

$\Delta NO_{2\ background\ i} = \Delta NO_{2\ macro\ background}$ for the hours of missing hourly NO_2 concentrations, Δ

$NO_{X\ background\ i} = \Delta NO_{X\ macro\ background}$ for the hours of missing hourly NO_X concentrations, and $\Delta O_{3\ background\ i} = \Delta O_{3\ macro\ background}$ for the hours of missing hourly O_3 concentrations. The VBA computer program applied Equations (14), (12) and (15) using the values corresponding to run 23 in Table 1 to generate the micro-calibrated background concentrations file. Running the Dunkirk AQMA air pollution model with this background concentrations file significantly improved the RMSE, r and the slope of the best fit line through the origin as shown in Table 2 and Fig. 5. This indicated a significant improvement for NO_2 hourly predictions by the model when using this background concentrations file. However, the model with this background concentrations file underestimated the annual mean of monitored NO_2 concentrations, and overestimated the annual mean of monitored O_3 concentrations, at the AQMS as shown in Table 2. Hence, using the trial and error macro-calibration approach, it was necessary to undertake additional runs of ADMS-Roads, beyond run 23, as shown in Table 1.

The background concentrations of these additional macro-calibration runs were modified so that the annual mean of monitored NO_2 concentrations was deliberately overestimated, and the annual mean of monitored O_3 concentrations was deliberately
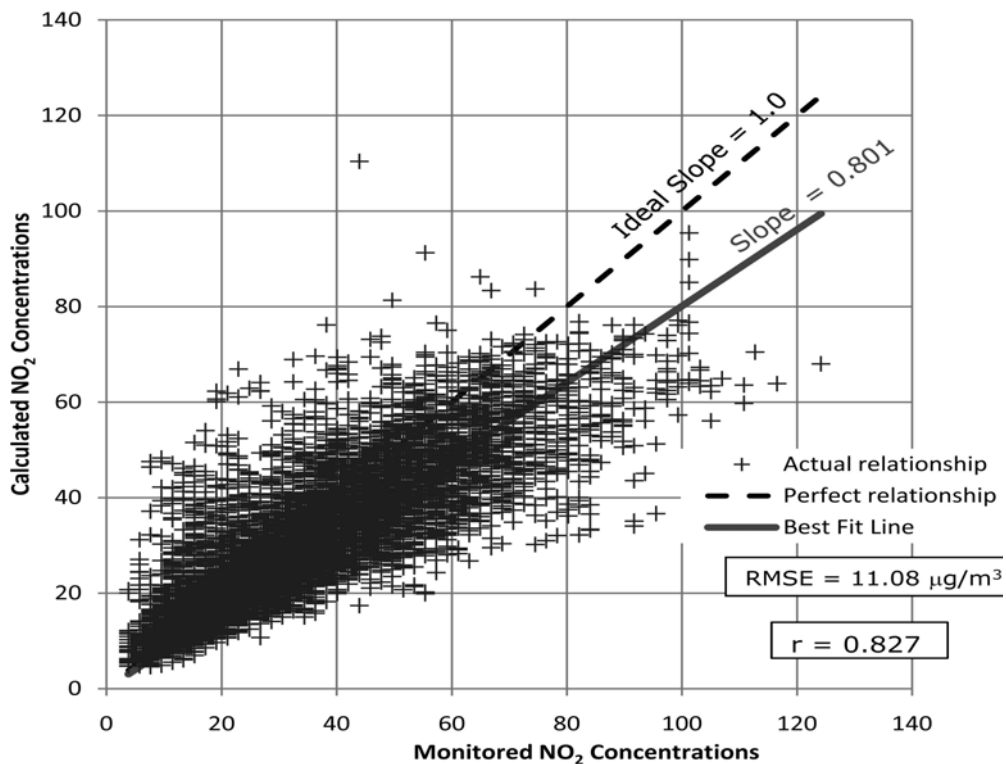

Fig. 5. Scatter diagram of hourly NO₂ concentrations at the AQMS after the micro-calibration based on Run 23

underestimated, by these runs, named A-D in Table 1. Consequently, after the 'normal' micro-calibration underestimation of the annual mean of monitored NO_2 concentrations and the 'normal' micro-calibration overestimation of the annual mean of monitored O_3 concentrations, the micro-calibration runs based on the results of these additional macro-calibration runs gave a good estimate of the annual means of both the monitored NO_2 and O_3 concentrations at the AQMS. This not only improved the results of the micro-calibrated model on the macro level, but also further improved the results on the micro level as shown in Table 2 and Fig. 6. Therefore, the micro-calibrated background concentrations obtained by Equations (14), (12) and (15), based on the values corresponding to run D in Table 1, were considered the final micro-calibrated background concentrations.

The micro-calibration development, from run 23 to run D, increased the error between the calculated and monitored NO_2 concentrations at a few hours, as implied by the comparison between the scatter in the overestimated points on the lower left side of Figs 5 and 6. A thorough investigation was undertaken in order to identify the reason for such unexpected behaviour of the micro-calibration process at these hours. A potential reason was the very high ratio of the monitored NO_X concentration to the monitored NO_2 concentrations, e.g. 7, which was accompanied by a

high monitored O_3 concentration at these hours. However, a high calculated NO_X concentration by the air pollution model was accompanied by high calculated NO_2 concentration and low calculated O_3 concentration at these hours. This suggested either imprecise model simulation of the actual atmospheric chemical reactions between NO_X and O_3 due to inaccurate input meteorological data or imprecise monitoring data at these hours. The high monitored NO_X concentration resulted in a high increase in the NO_X background concentration due to the micro-calibration at these hours. Such a high increase in the NO_X background concentration substantially increased the calculated NO_2 concentration, resulting in a big difference between the calculated and low monitored NO_2 concentrations at these hours. At some of these hours, for which the NO_2 concentration was underestimated before any calibration, the micro-calibration iterations increased the background NO_2 concentration in order to increase the calculated NO_2 concentration, which changed the NO_2 underestimation into an increasingly greater NO_2 overestimation. At the rest of these hours, for which the NO_2 concentration was overestimated before any calibration, the reduction in calculated NO_2 concentration due to the micro-calibration iterations was masked by the increase in calculated NO_2 concentration due to the high NO_X background concentration.

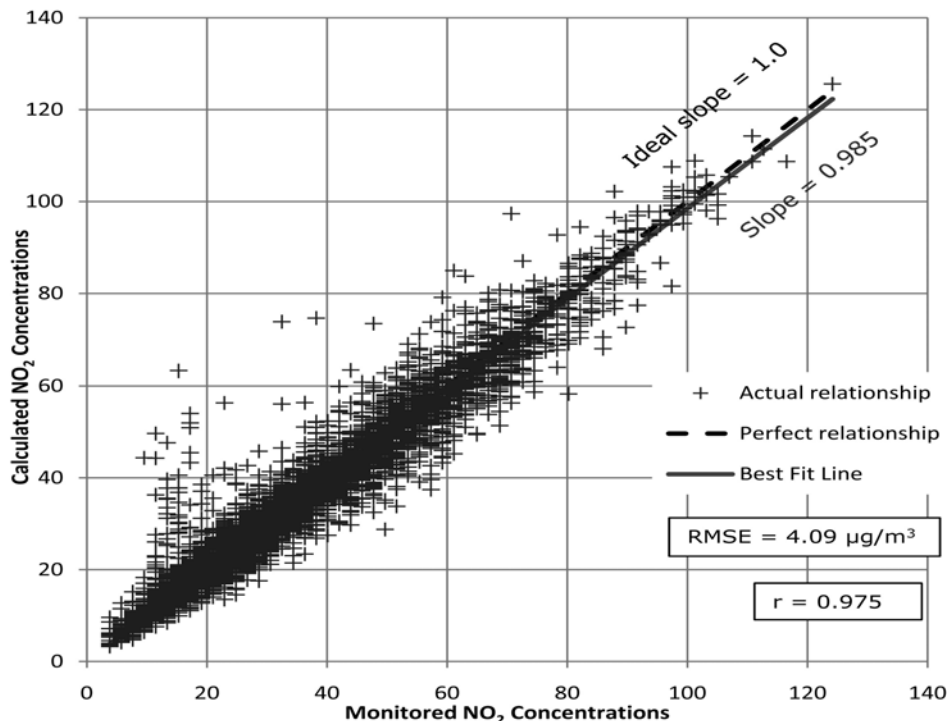**Fig. 6. Scatter diagram of hourly NO_2 concentrations at the AQMS after the micro-calibration based on Run D**

CONCLUSION

The macro-calibration of background concentrations reduced effectively the error between the calculated and monitored annual means of NO_x, NO_2, and O_3 concentrations. The iterative application of the micro-calibration equations (14), (12) and (15) to background concentrations reduced effectively the error between the calculated and monitored annual means of NO_x, NO_2, and O_3 concentrations, and also the error between the hourly calculated and monitored NO_2 concentrations. Further research is required to adapt the macro-calibration and micro-calibration equations for modelling the air pollution dispersion of inert pollutants, e.g. CO and PM. As chemical reactions will not be considered, the calibration equations may reduce to one equation for the macro-calibration, and one equation for the micro-calibration, of the input background concentrations. For the hours with missing monitored air pollution concentrations, the micro-calibration equations were unusable. This was addressed by using the macro-calibrated background concentrations for these hours. As the macro-calibrated background concentrations give less precise calculated concentrations on the hourly level, such a strategy may reduce the reliability of the number of exceedances and percentiles predicted by the air pollution model. Therefore, for the hours with missing monitored air pollution concentrations, further research is needed to investigate the impact of using the macro-calibrated background concentrations on the reliability of the predicted number of exceedances and percentiles by the air pollution model. In case of a significant adverse impact, further research is recommended into the micro-calibration of the rural background concentrations of these hours, based on the meteorological data and the micro-calibrated background concentrations of other hours with monitored concentrations. The inclusion of the hourly and monthly traffic profiles in the Dunkirk AQMA air pollution model did not have a significant impact on the error between the annual means of calculated and monitored concentrations. On the other hand, the inclusion of these traffic profiles did reduce the RMSE between the hourly calculated and monitored NO_2 concentrations by 28.4%. As the Dunkirk AQMA air pollution model did not include a large number of road sources, further research is recommended to investigate the impact of including the monthly and hourly traffic profiles on the micro-validation of an air pollution model that has a large number of road sources. This is to correlate between the number of road sources with traffic profiles in the air pollution model and the possible reduction in the RMSE between the hourly calculated and monitored NO_2 concentrations.

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