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A Mathematical Approach to Enhance the Performance of Air Pollution Models

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Additional information is available at the end of the chapter

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Abstract

The main objective of this chapter is to introduce a mathematical method for enhancing the correctness of the output results of air pollution dispersion models via the calibration of input background concentrations. For developing this method, an air pollution model was set up in ADMS-Roads for a study area in the City of Nottingham in the UK. The method was applied iteratively to the input background concentrations, which effectively reduced the error between calculated and monitored air pollution concentrations on both the annual mean and hourly levels. The inclusion of the traffic flow profiles of the modeled road network reduced further the error between the hourly, but not the annual mean, calculated and monitored concentrations. The application of the calibration approach to the model in ADMS-Roads was compared to the use of grid air pollution sources for the same model in ADMS-Urban. In terms of the accuracy of the model results, the calibration approach was better than using grid sources on the annual mean level and was much better on the hourly level. Compared to the use of grid sources in ADMS-Urban, the use of the calibration approach in either ADMS-Roads or ADMS-Urban can significantly reduce the air pollution model runtime.

Keywords: calibration, validation, background concentrations, modeling, air pollution

1. Introduction

Modeling 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 programs, including local transport plans [1]. As the majority of national air quality standards are in the form of annual mean and hourly objectives [2], this requires accurate annual mean and hourly air quality predictions.

The results of air pollution dispersion modeling should be accurate enough to provide reliable air quality predictions. Recent air pollution dispersion modeling 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 minimized [3–7].

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 [8]. 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.

Ref. [9] 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 modelers should avoid the application of such an adjustment factor to the model results [10]. Instead, CERC advised that various details of the model set up, such as input data and modeling options, should be adjusted until the calculated results fit the monitored concentrations.

Ref. [11] 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 [12].

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 [13].

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 oxidized NO_2 proportion, is recommended [10]. 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 [10].

Ref. [14] 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 [15]. The calibration of input emission rates slightly reduced the error (by 6.46%) between daily calculated and monitored PM_{10} concentrations over 8 days. This implies a nonsignificant 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 [16, 17].

Therefore, this chapter 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 modeling; namely, macrocalibration, macrovalidation, microcalibration, and microvalidation. The background concentrations are some of the most important input data to the broad variety of air pollution dispersion models [18]. 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 chapter, 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 modeled 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.

2. Set-up of the air pollution model

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 [19]. Dunkirk AQMA is an urban study area in the city of Nottingham, as shown in **Figure 1**, with NO_2 levels exceeding the permissible levels [20]. Therefore, NO_2 was selected as the modeled 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.

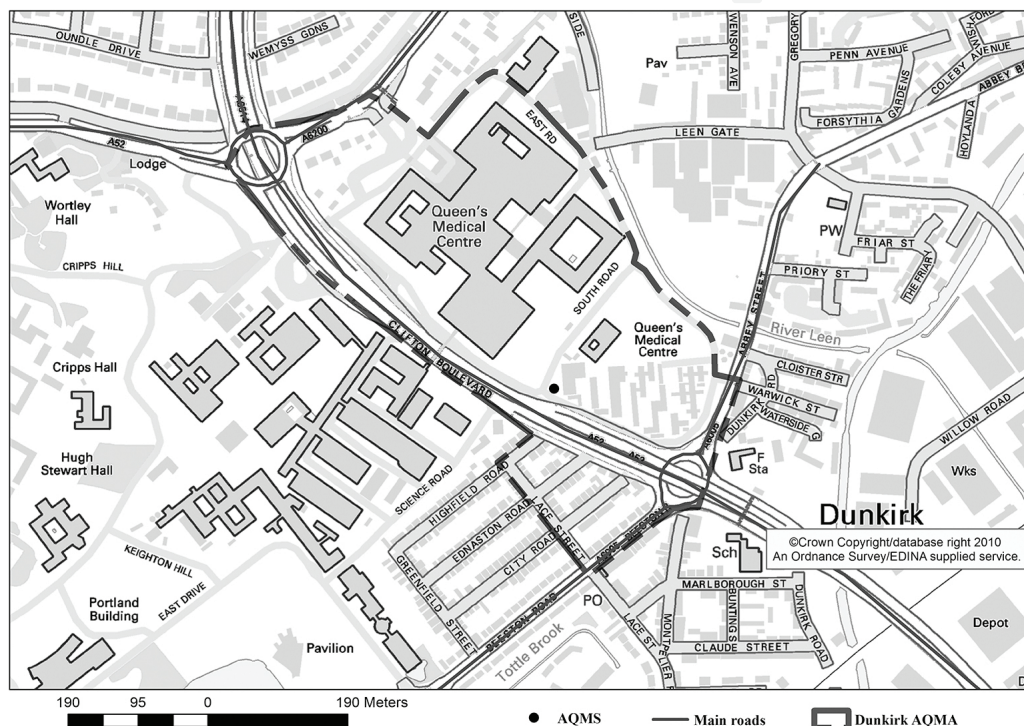


Figure 1. The Dunkirk AQMA.

Note that 2006 was selected as the modeling 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 **Figure 1**, and the relevant significant industrial air pollution sources. The Nottingham Watnall Weather Station [21] provided the 2006 hourly sequential meteorological data which included surface temperature, wind speed at 10-m 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 **Figure 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 5 min 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 5-min traffic counts, using the following mathematics:

For each day, the 5-min flow data was automatically aggregated to yield hourly flow data. Let f_{ijk} be the total traffic flow in both directions in hour i of day j of month k , and let N_k be the number of days in month k , such that $i = 0, \dots, 23$, $j = 1, \dots, N_k$ (where $N_k = 28, 29, 30$ or 31 as appropriate), and $k = 1, \dots, 12$.

Therefore,

$$\text{AADT (vehicles / hour)} = \frac{\sum_{k=1}^{12} \sum_{j=1}^{N_k} \sum_{i=0}^{23} f_{ijk}}{\left[\sum_{k=1}^{12} N_k \right] \times 24} \quad (1)$$

$$\text{Monthly average}_k \text{ (vehicles / hour)} = \frac{\sum_{j=1}^{N_k} \sum_{i=0}^{23} f_{ijk}}{N_k \times 24} \quad \forall k, k = 1, \dots, 12 \quad (2)$$

$$\text{Monthly factor}_k = \frac{\text{Monthly average}_k}{\text{AADT}} \quad \forall k, k = 1, \dots, 12 \quad (3)$$

Let p_k , q_k , and r_k be the number of weekdays, Saturdays, and Sundays, respectively, in month k , such that $p_k + q_k + r_k = N_k \quad \forall k, k = 1, \dots, 12$. Therefore, the Hourly Average _{i} (vehicles/hour):

$$\text{For weekdays (if } j \text{ denotes weekdays)} = \frac{\sum_{k=1}^{12} \sum_{j=1}^{p_k} f_{ijk}}{\sum_{k=1}^{12} p_k} \quad \forall i, i = 0, \dots, 23 \quad (4)$$

$$\text{For Saturdays (if } j \text{ denotes Saturdays)} = \frac{\sum_{k=1}^{12} \sum_{j=1}^{q_k} f_{ijk}}{\sum_{k=1}^{12} q_k} \quad \forall i, i = 0, \dots, 23 \quad (5)$$

$$\text{For Sundays (if } j \text{ denotes Sundays)} = \frac{\sum_{k=1}^{12} \sum_{j=1}^{r_k} f_{ijk}}{\sum_{k=1}^{12} r_k} \quad \forall i, i = 0, \dots, 23 \quad (6)$$

Hence, there are $3 \times 24 = 72$ different day-related hourly average traffic flows; so, correspondingly, there are 72 hourly factors, such that:

$$\text{Hourly factor}_i = \frac{\text{Hourly average}_i}{\text{AADT}} \quad \forall i, i = 0, \dots, 23 \quad (7)$$

Therefore, the full traffic flow data processing output for each main road was:

- 24 hourly factors for weekdays, in order, from hour 0 to hour 23.
- 24 hourly factors for Saturdays, in order, from hour 0 to hour 23.
- 24 hourly factors for Sundays, in order, from hour 0 to hour 23.
- 12 monthly factors for the 12 months, in order, from January to December.

Lack of data from some detectors for some time periods during the year 2006 had to be addressed. If the corresponding traffic data was available for another year, then that was used, factored using traffic data from the nearest detectors, for that other year and 2006. Steps were taken in the code to avoid zero division in factoring the traffic data of that other year. If the corresponding traffic data from another year was not available, then 2006 traffic data from the nearest available detectors were used. 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 [22], 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 [19]. Modeling these atmospheric reactions was necessary to get accurate NO_2 results, so NO_x and O_3 were modeled in addition to NO_2 . However, using this chemical scheme requires inputs for NO_2 , 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 [10].

3. 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 O₃ 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.

Δ background		Calculated concentrations	Target concentrations
Run 1			
NO ₂	0	26.25	35.29
NO _x	0	42.19	67.60
O ₃	0	44.23	31.00
Run 9			
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			
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			
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			
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			
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			
NO ₂	+17.18	43.56	35.29
NO _x	+25.42	67.61	67.60
O ₃	-16.71	28.69	31.00

Table 1. Macrocalibration development stages of the rural background concentrations.

3.1. Macrocalibration and macrovalidation

The term macrocalibration in this chapter 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 macrovalidation 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 in Section 2, it was decided to calibrate NO_x and O_3 , in addition to NO_2 , background concentrations. A trial and error approach was adopted to macrocalibrate the hourly sequential rural background concentrations until the above-mentioned macrocalibration criterion was achieved. This approach comprised 22 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 macrocalibration iteration, the values in the “ Δ background” 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 macrocalibrated background concentrations file. Another computer logic was applied to this file in order to preserve the fact that NO_x is $\text{NO} + \text{NO}_2$. Hence, for every hour in the macrocalibrated background concentrations file, if $\text{NO}_2 > \text{NO}_x$, then $\text{NO}_2 = \text{NO}_x$.

After each iteration of the macrocalibration, the macrovalidation 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 macrocalibrated background concentrations.

The results of the final macrocalibration run were used to derive Eqs. (8), (9), and (10), which could be used to evaluate directly the background concentration adjustment values, required to macrocalibrate the Dunkirk AQMA air pollution model, without the trial and error approach:

$$\Delta \text{NO}_{2 \text{ background}} = \frac{(\overline{\text{NO}}_{2 \text{ monitored}} - \overline{\text{NO}}_{2 \text{ uncalibrated}})}{9.2} \times 1.48, \quad (8)$$

where $\overline{\text{NO}}_{2 \text{ monitored}}$ is the annual mean of monitored NO_2 concentrations and $\overline{\text{NO}}_{2 \text{ uncalibrated}}$ is the annual mean of calculated NO_2 concentrations using the rural background concentrations.

$$\Delta \text{NO}_{x \text{ background}} = \overline{\text{NO}}_{x \text{ monitored}} - \overline{\text{NO}}_{x \text{ uncalibrated}}, \quad (9)$$

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.

$$\Delta O_3 \text{ background} = \frac{(\bar{O}_3 \text{ monitored} - \bar{O}_3 \text{ uncalibrated})}{(-13.22)} \times (-5.40), \quad (10)$$

where $\bar{O}_3$ monitored is the annual mean of monitored O_3 concentrations and $\bar{O}_3$ uncalibrated is the annual mean of calculated O_3 concentrations using the rural background concentrations.

3.2. Microcalibration and microvalidation

The term microcalibration in this chapter 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 microcalibration extends the macrocalibration as shown in **Figure 2**. The microvalidation was

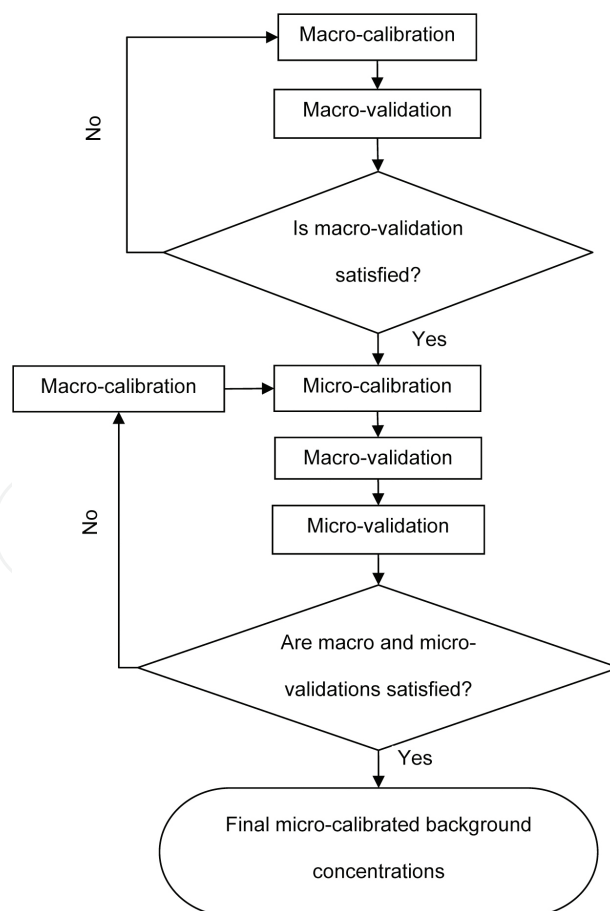


Figure 2. Calibration and validation process for rural background concentrations.

undertaken by comparing statistically two one-dimensional arrays of the 2006 calculated and monitored hourly sequential NO₂ concentrations at the AQMS. The statistical approach to compare these two arrays depended on the definition of them. If these two arrays were to be defined as two samples of two bigger populations, statistical tests would be the best approach to compare statistically the two bigger populations [23]. However, if these two arrays represented the two populations to compare, statistical tests would not be suitable and descriptive statistics would be the convenient statistical approach to compare these two populations.

Therefore, careful consideration was given to define correctly the two arrays of calculated and monitored 2006 hourly NO₂ concentrations at the AQMS, concluding that these two arrays should be defined as two populations, not as two samples. The reason was that these two arrays of concentrations did not comprise NO₂ concentrations from any year other than 2006, or averages over any time period other than an hour. Therefore, a hypothesis that these two arrays are two samples of two bigger populations that may extend over many years of time, or comprise air pollution concentrations calculated or monitored over a diversity of averaging times, was invalid. Consequently, Pearson correlation coefficient (r) and the root mean square error (RMSE) were used to compare the two populations. Further details about these two descriptive statistics are given in [7, 24, 25]. The slope of the regression line through the origin was also used to compare the two populations of hourly calculated and monitored concentrations.

Linear regression through the origin was used because it was already known that the perfect relationship between hourly calculated and monitored concentrations is $y_i = x_i$ without a constant, where y_i and x_i were the calculated and monitored NO₂ concentrations for hour i at the AQMS, respectively. The value of i ranged from 1 to 8760 which was the total number of hours in the year 2006. The linear regression analysis was undertaken for three cases, uncalibrated versus monitored, macrocalibrated versus monitored, and microcalibrated versus monitored, concentrations. In all these three cases, the independent variable was the monitored concentrations.

The comparison between the calculated and monitored hourly NO₂ concentrations at the AQMS was undertaken by the comparison between the slope of the best fit line through the origin and 1.0, the slope of the perfect relationship. The magnitude and sign of the difference between the slope of the best fit line through the origin and 1.0 indicated the tendency of calculated NO₂ concentrations to underestimate or overestimate the 2006 monitored NO₂ concentrations on the micro, hourly, level. Moreover, the slope of the regression best fit line through the origin was used for the graphical representation of the linear approximation of the actual relationship between calculated and monitored hourly NO₂ concentrations at the AQMS, after each stage of the calibration process.

The Dunkirk AQMA air pollution model was run with the uncalibrated rural background concentrations file to output the 2006 calculated hourly NO₂ 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 NO₂ concentrations at the AQMS, as shown in **Figure 3**. Then, the model was run with the macrocalibrated background concentrations file, corresponding to run 23 in **Table 1**, to output the 2006 calculated hourly NO₂ concentrations

at the AQMS. This was for the microvalidation after the macrocalibration of the rural background concentrations as shown in **Figure 4**.

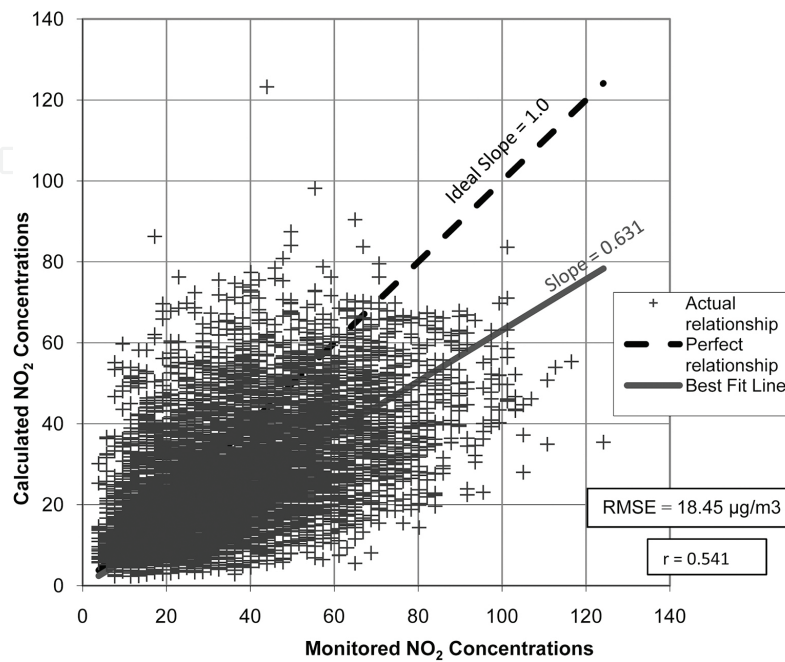


Figure 3. Scatter diagram of hourly NO₂ concentrations at the AQMS before any calibration.

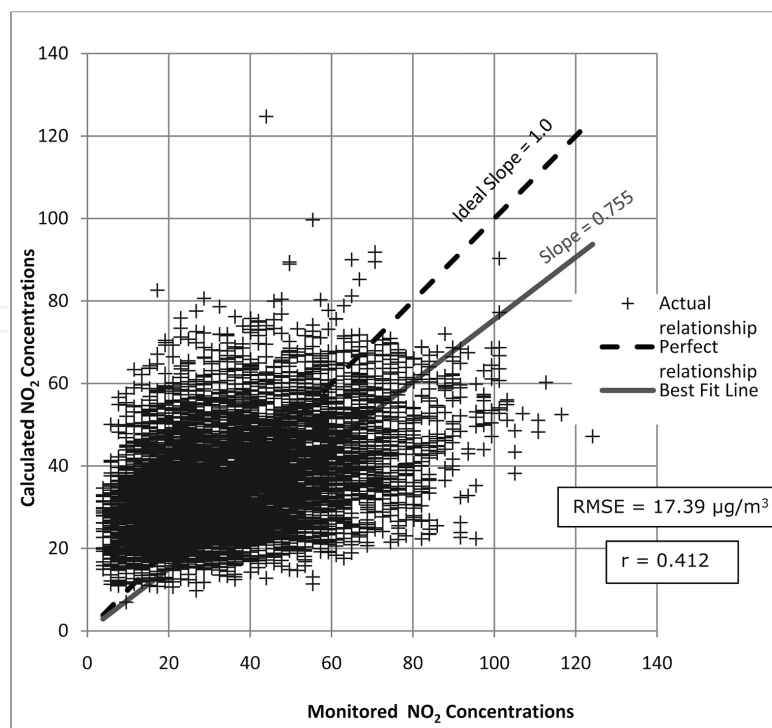


Figure 4. Scatter diagram of hourly NO₂ concentrations at the AQMS after macrocalibration.

Pearson's correlation coefficients were calculated as 0.541 before any calibration, and then as 0.412 after the macrocalibration, as shown in **Figures 3** and **4**. The slight decline in Pearson's correlation coefficient after the macrocalibration implied that the macrocalibration slightly decreased the degree of linearity of the actual relationship between the calculated and monitored hourly NO₂ concentrations at the AQMS. Hence, the macrocalibration 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 macrocalibration, as shown in **Figures 3** and **4**. The slight decline in the RMSE after the macrocalibration implied that the macrocalibration slightly lowered the difference between the calculated and monitored hourly NO₂ concentrations. Therefore, the macrocalibration not only improved the NO₂ predictions of the model on the macro, annual mean, level but also slightly improved the NO₂ 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 NO₂ concentrations at the AQMS was calculated as 0.631 before any calibration, and then as 0.755 after the macrocalibration, as shown in **Figures 3** and **4**. Although the results of the macrocalibration, corresponding to run 23 in **Table 1**, very slightly overestimated the 2006 annual mean of monitored NO₂ concentrations at the AQMS, the slope of the best fit line through the origin after the macrocalibration was less than 1.0. This indicated that, after the macrocalibration, the model generally underestimated the monitored NO₂ concentrations at the AQMS on the micro, hourly, level. However, the slight increase in the slope of the best fit line after the macrocalibration implied that the macrocalibration slightly reduced the tendency of the model to underestimate the monitored hourly NO₂ concentrations at the AQMS. This, together with the reduction in the RMSE after the macrocalibration, confirmed the slight improvement of the NO₂ predictions of the model, after the macrocalibration, on the micro, hourly, level.

To improve further the NO₂ predictions of the model on the micro level, the idea of microcalibration was developed. This idea depended on the modification of Eqs. (8), (9), and (10) in order to generate three one-dimensional arrays for ΔNO₂ background, ΔNO_x background, and ΔO₃ background as follows:

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

where $\Delta hNO_{2 \text{ background } i}$ is the adjustment value for the rural NO₂ background concentration for the hour i . $NO_{2 \text{ monitored } i}$ is the monitored hourly NO₂ concentration for the hour i . $NO_{2 \text{ uncalibrated } i}$ is the calculated hourly NO₂ concentration for the hour i using the uncalibrated rural background concentrations. $NO_{2 \text{ macro } i}$ is the calculated hourly NO₂ concentra-

tion for the hour i using the macrocalibrated background concentrations. The value of i ranged from 1 to 8760, which was the total number of hours in the year 2006:

$$\Delta NO_{X \text{ background } i} = NO_{X \text{ monitored } i} - NO_{X \text{ uncalibrated } i}, \quad (12)$$

where $\Delta hNO_{X \text{ background } i}$ is the adjustment value for the rural NO_X background concentration for the hour i . $NO_{X \text{ monitored } i}$ is the monitored hourly NO_X concentration for the hour i . $NO_{X \text{ uncalibrated } i}$ 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:

$$\Delta O_3 \text{ background } i = \frac{(O_3 \text{ monitored } i - O_3 \text{ uncalibrated } i)}{(O_3 \text{ macro } i - O_3 \text{ uncalibrated } i)} \times (-5.4), \quad (13)$$

where $\Delta hO_3 \text{ background } i$ is the adjustment value for the rural O_3 background concentration for the hour i . $O_3 \text{ monitored } i$ is the monitored hourly O_3 concentration for the hour i . $O_3 \text{ uncalibrated } i$ is the calculated hourly O_3 concentration for the hour i using the uncalibrated rural background concentrations. $O_3 \text{ macro } i$ is the calculated hourly O_3 concentration for the hour i using the macrocalibrated background concentrations. The value of i ranged from 1 to 8760, which was the total number of hours in the year 2006.

The three one-dimensional arrays of $\Delta NO_2 \text{ background } i$, $\Delta NO_X \text{ background } i$ and $\Delta O_3 \text{ background } i$ calculated by Eqs. (11), (12), and (13), were added to the arrays of the uncalibrated hourly sequential rural background concentrations of NO_2 , NO_X , and O_3 , respectively. Hence the microcalibrated background concentrations file was created based on the above three equations. However, running the model with these microcalibrated 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 microcalibrated 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 microcalibration based on Eqs. (11), (12), and (13) was the use of the macrocalibrated hourly concentrations in these equations. As discussed before with regard to **Figure 4**, the hourly calculated concentrations of the macrocalibrated model were not precise enough. The macrocalibrated model of the Dunkirk AQMA was validated only on the macro, annual mean, level. Therefore, instead of using $NO_2 \text{ macro } i$ and $O_3 \text{ macro } i$, the macrocalibrated calculated hourly NO_2 and O_3 concentrations, it was decided to alter two of the three equations for the microcalibration of the rural background concentrations, using the macrocalibrated annual mean NO_2 and O_3 concentrations, so that:

Case description	Receptor name	Annual mean NO _x		Annual mean NO ₂		Annual mean O ₃		NO ₂ RMSE	NO ₂ RMSE	NO ₂ RMSE
		Calculated	Monitored	Calculated	Monitored	Calculated	Monitored	before calibration	after macrocalibration	after microcalibration
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

Table 2. Microcalibration development stages of the rural background concentrations.

$$\Delta NO_{2 \text{ background } i} = \frac{(NO_{2 \text{ monitored } i} - NO_{2 \text{ uncalibrated } i})}{(\overline{NO}_{2 \text{ macro}} - \overline{NO}_{2 \text{ uncalibrated}})} \times \Delta NO_{2 \text{ macro background}}, \quad (14)$$

where $\Delta NO_{2 \text{ background } i}$ is the adjustment value for the rural NO_2 background concentration for the hour i . $NO_{2 \text{ monitored } i}$ is the monitored hourly NO_2 concentration for the hour i . $NO_{2 \text{ uncalibrated } i}$ 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. $\overline{NO}_{2 \text{ macro}}$ is the annual mean NO_2 concentration calculated using the macrocalibrated background concentrations. $\overline{NO}_{2 \text{ uncalibrated}}$ is the annual mean NO_2 concentration calculated using the uncalibrated rural background concentrations. $\Delta NO_{2 \text{ macro background}}$ is the macrocalibration adjustment value for the rural NO_2 background concentrations, as given in the column headed “ Δ background” in **Table 1**:

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

where $\Delta O_{3 \text{ background } i}$ is the adjustment value for the rural O_3 background concentration for the hour i . $O_{3 \text{ monitored } i}$ is the monitored hourly O_3 concentration for the hour i . $O_{3 \text{ uncalibrated } i}$ 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. $\overline{O}_{3 \text{ macro}}$ is the annual mean O_3 concentration calculated using the macrocalibrated background concentrations. $\overline{O}_{3 \text{ uncalibrated}}$ is the annual mean O_3 concentration calculated using the uncalibrated rural background concentrations. $\Delta O_{3 \text{ macro background}}$ is the macrocalibration adjustment value for the rural O_3 background concentrations, as given in the column headed “ Δ background” in **Table 1**.

A VBA computer program was written in MS-Excel in order to automate the generation of the three hourly sequential one-dimensional arrays for $\Delta NO_{2 \text{ background}}$, $\Delta NO_{x \text{ background}}$, and $\Delta O_{3 \text{ background}}$ using Eqs. (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: $\Delta NO_{2 \text{ background } i} = \Delta NO_{2 \text{ macro background}}$ for the hours of missing hourly NO_2 concentrations, $\Delta NO_{x \text{ background } i} = \Delta NO_{x \text{ macro background}}$ for the hours of missing hourly NO_x concentrations, and $\Delta O_{3 \text{ background } i} = \Delta O_{3 \text{ macro background}}$ for the hours of missing hourly O_3 concentrations.

The VBA computer program applied Eqs. (14), (12), and (15) along with the macrocalibration results of run 23 in **Table 1** to generate the microcalibrated 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 **Figure 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 macrocalibration approach, it was necessary to undertake additional runs of ADMS-Roads, beyond run 23, as shown in **Table 1**.

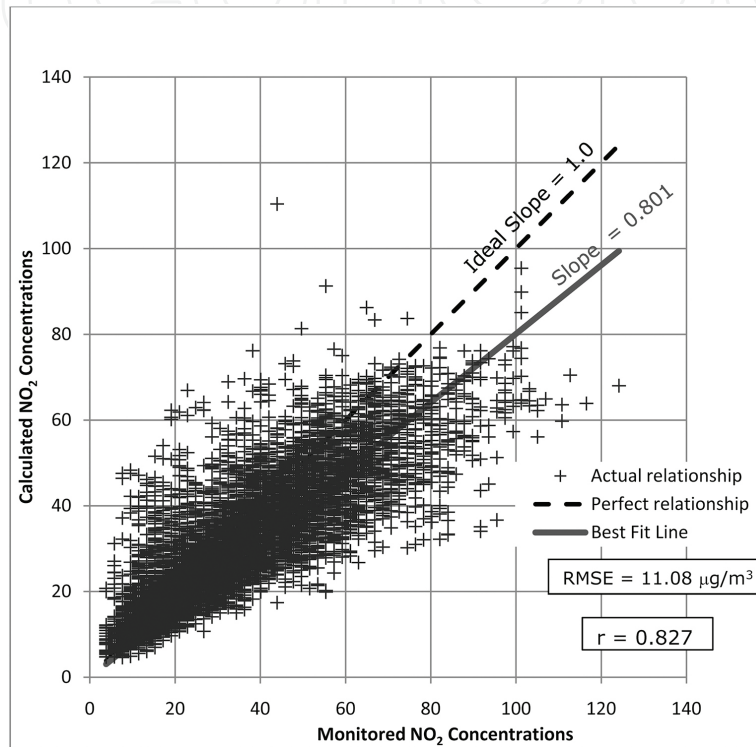


Figure 5. Scatter diagram of hourly NO_2 concentrations at the AQMS after the microcalibration based on run 23.

The background concentrations of these additional macrocalibration 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 underestimated, by these runs, named A–D in **Table 1**. Consequently, after the “normal” microcalibration underestimation of the annual mean of monitored NO_2 concentrations and the “normal” microcalibration overestimation of the annual mean of monitored O_3 concentrations, the microcalibration runs based on the results of these additional macrocalibration 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 microcalibrated model on the macro level, but also further improved the results on the micro level as shown in **Table 2** and **Figure 6**. Therefore, the microcalibrated background concentrations obtained by Eqs. (14), (12), and (15), based on the macrocalibration results of run D, were considered the final microcalibrated background concentrations.

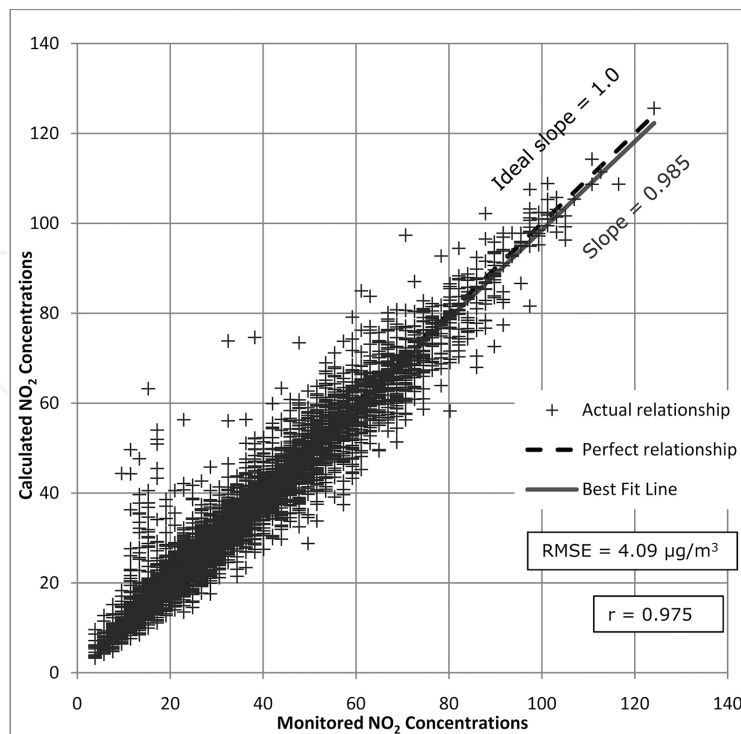


Figure 6. Scatter diagram of hourly NO_2 concentrations at the AQMS after the microcalibration based on run D.

The microcalibration 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 **Figures 5 and 6**. A thorough investigation was undertaken in order to identify the reason for such unexpected behavior of the microcalibration 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 microcalibration 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 microcalibration 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 microcalibration iterations was masked by the increase in calculated NO_2 concentration due to the high NO_x background concentration.

4. Impact of traffic profiles on the macro- and microvalidation

As mentioned in Section 2, the hourly and monthly traffic flow profiles were considered in the set-up of the air pollution model by use of a special text file, a FAC file. The impact of the traffic profiles on the macro and micro levels was investigated by turning off this FAC file in the final microcalibrated version of the Dunkirk AQMA model, corresponding to run D in **Table 2**. The exclusion of the traffic profiles did not have a significant impact on the calculated annual mean NO_2 , NO_x and O_3 concentrations as shown in **Table 2**. Therefore, it was concluded that the consideration of the traffic profiles in the air pollution model was not important for the macrovalidation.

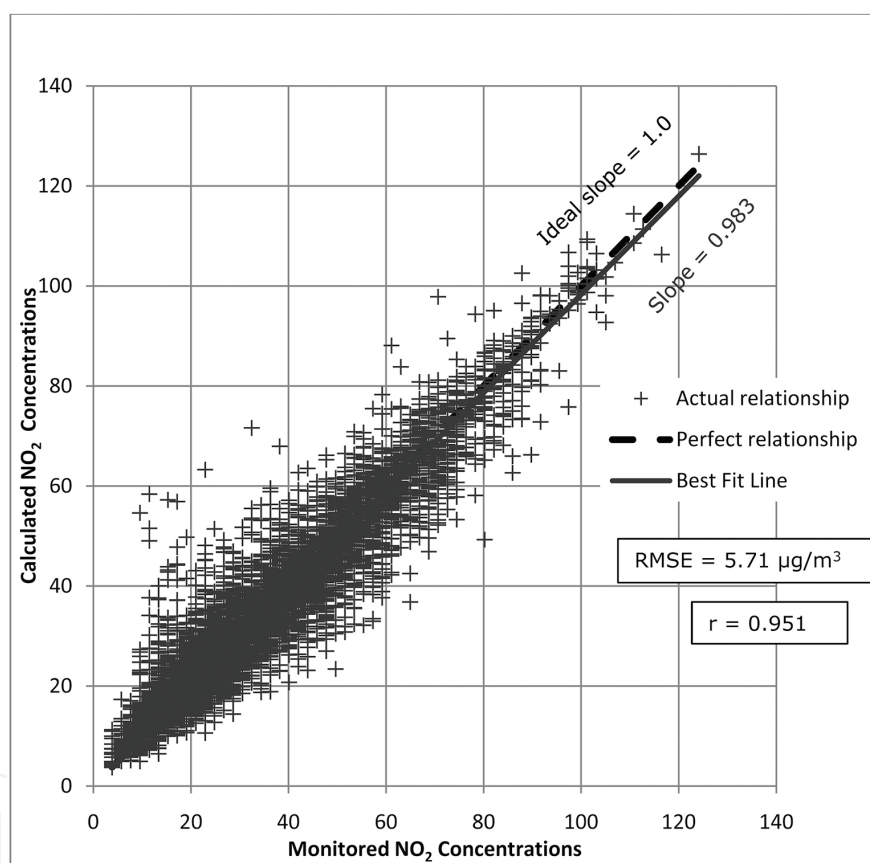


Figure 7. Scatter diagram of hourly NO_2 concentrations at the AQMS after the microcalibration based on run D without a FAC file.

On the other hand, the exclusion of the traffic profiles slightly worsened the hourly calculated NO_2 concentrations as shown in **Figure 7**. This was indicated by the higher RMSE, the lower r , and the slightly lower slope of the best fit line through the origin, without a FAC file in **Figure 7** compared to with a FAC file in **Figure 6**. Therefore, it was concluded that the incorporation of the traffic profiles in the air pollution model could further improve the microvalidation by reducing the RMSE between the calculated and monitored hourly NO_2 concentrations by 28.4%.

5. The calibration of background concentrations versus the use of grid sources

Grid air pollution sources are used in ADMS-Urban to model residual, poorly defined or diffused emissions in urban areas, such as the emissions from domestic heating sources and minor roads [26]. This enables ADMS-Urban to model emissions from sources that are not defined explicitly in the air pollution model. Therefore, Nottingham City Council uses grid sources in ADMS-Urban to compensate for the difference between rural and urban background concentrations [8]. However, the capability to model emissions from such air pollution sources is only available in ADMS-Urban, not in ADMS-Roads. Hence, the Dunkirk AQMA air pollution model was set up in ADMS-Urban, with the Rochester rural background concentrations, and this time with a grid source. The air pollution emissions of the grid source were obtained from the UK National Atmospheric Emissions Inventory (NAEI).

The ADMS-Urban model was run to output the 2006 annual mean concentrations of NO_2 , NO_x , and O_3 at the AQMS as shown in **Table 3**. Comparing **Table 2** with **Table 3**, the calculated annual mean NO_2 , NO_x , and O_3 concentrations from the ADMS-Roads model, with microcalibrated background concentrations, were closer to the corresponding annual means of monitored concentrations than were the calculated annual means from the ADMS-Urban model, with a grid source and rural background concentrations. This indicated that the ADMS-Roads model, with microcalibrated background concentrations only, was more precise than the ADMS-Urban model, with a grid source and rural background concentrations, on the macro level.

Case description	NO_2 annual mean, $\mu\text{g}/\text{m}^3$		NO_x annual mean, $\mu\text{g}/\text{m}^3$		O_3 annual mean, $\mu\text{g}/\text{m}^3$	
	Calculated	Monitored	Calculated	Monitored	Calculated	Monitored
ADMS-Urban with CRS	37.65	35.29	69.31	67.6	35.18	31.0
ADMS-Urban with CRS with trajectory model	37.77	35.29	69.31	67.6	35.07	31.0

Table 3. Monitored versus calculated annual mean concentrations at the AQMS by ADMS-Urban.

The 2006 hourly NO_2 concentrations calculated by the ADMS-Urban model were compared to the 2006 hourly monitored NO_2 concentrations at the AQMS as shown in **Figure 8**. Hence, comparing **Figure 6** with **Figure 8**, the results of the ADMS-Urban model, with a grid source and rural background concentrations, gave a much higher RMSE than did the results of the ADMS-Roads model, with microcalibrated background concentrations only. In addition, the results of the ADMS-Urban model gave a much lower r , and a lower slope of the best fit line through the origin, than did the results of the ADMS-Roads model, with microcalibrated background concentrations only. Therefore, the results of the ADMS-Roads model, with microcalibrated background concentrations only, were much closer to the 2006 hourly NO_2

concentrations monitored by the AQMS than were the results of the ADMS-Urban model, with a grid source and rural background concentrations. This indicated that the ADMS-Roads model, with microcalibrated background concentrations only, was much more precise than the ADMS-Urban model, with a grid source and rural background concentrations, on the micro level.

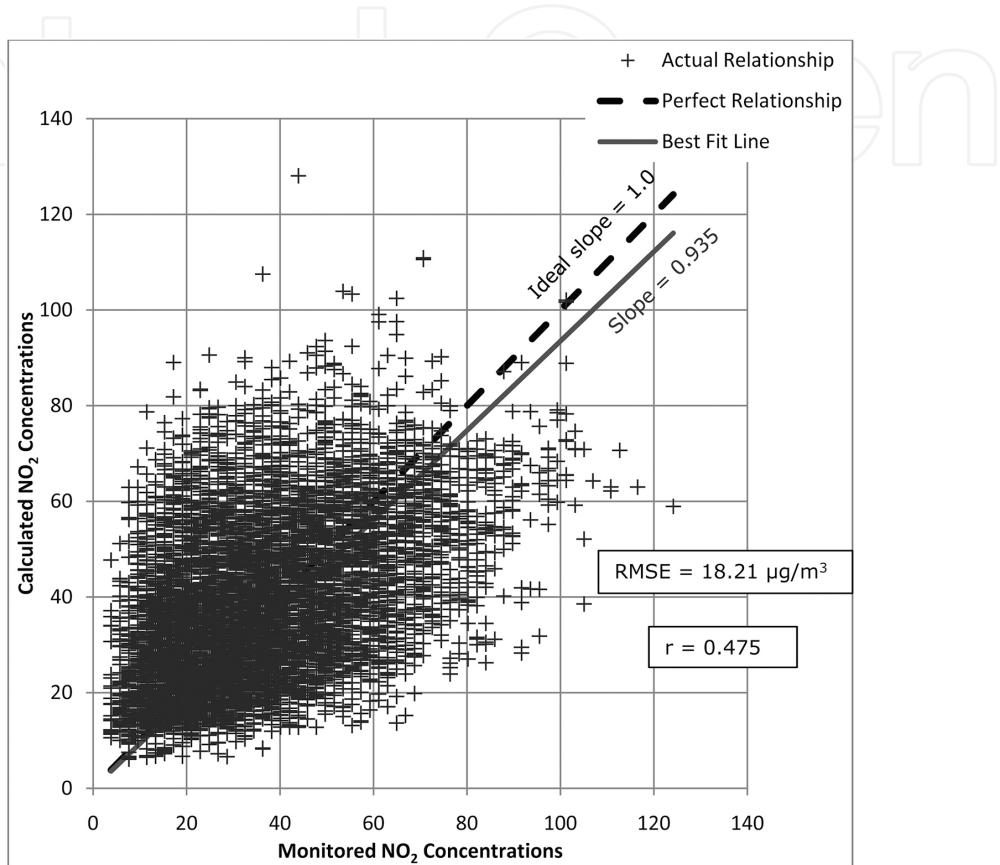


Figure 8. Scatter diagram of monitored versus calculated hourly NO₂ concentrations at the AQMS by ADMS-Urban.

Comparing **Table 1** (run 23) with **Table 3**, the calculated annual mean NO₂, NO_x, and O₃ concentrations from the ADMS-Roads model, with macrocalibrated background concentrations only, were closer to the corresponding annual means of monitored concentrations than were the calculated annual means from the ADMS-Urban model, with a grid source and rural background concentrations. This indicated that the ADMS-Roads model, with macrocalibrated background concentrations only, was more precise than the ADMS-Urban model, with a grid source and rural background concentrations, on the macro level.

In respect of the 2006 hourly NO₂ concentrations, comparing **Figure 8** with **Figure 4**, the results of the ADMS-Urban model, with a grid source and rural background concentrations, gave a slightly higher RMSE than did the results of ADMS-Roads, with macrocalibrated background concentrations only. Both the ADMS-Urban model and the macrocalibrated ADMS-Roads model generally underestimated the 2006 hourly monitored NO₂ concentrations which was indicated by the best fit line through the origin having a slope of less than

1.0 in both **Figures 8** and **4**. However, the slope of the best fit line in the ADMS-Urban case (in **Figure 8**) was closer to 1.0 than was the slope of the best fit line in the macrocalibrated ADMS-Roads case (in **Figure 4**). Therefore, the tendency of the ADMS-Urban model, with a grid source and rural background concentrations, to underestimate the hourly monitored NO₂ concentrations was less than that of the ADMS-Roads model, with macrocalibrated background concentrations only.

Continuing the comparison of **Figure 8** with **Figure 4**, the results of ADMS-Urban, with a grid source and rural background concentrations, gave a slightly higher r than did the results of ADMS-Roads, with macrocalibrated background concentrations only. This implied that the ADMS-Urban model slightly increased the degree of linearity of the actual relationship between the calculated and monitored hourly NO₂ concentrations at the AQMS. Hence, the actual relationship between the calculated and monitored hourly NO₂ concentrations was slightly closer to the perfect straight line relationship in the case of the ADMS-Urban model than it was in the case of the macrocalibrated ADMS-Roads model. The RMSE, r , and the slope of the best fit line through the origin indicated that the ADMS-Roads model, with macrocalibrated background concentrations only, was almost as precise as the ADMS-Urban model, with a grid source and rural background concentrations, on the micro level.

The trajectory model of CRS can be used along with a grid air pollution source in ADMS-Urban to adjust the background concentrations in the main model domain, the model application area, on the basis of the grid source emissions [26]. The trajectory model uses the grid source domain, which is usually larger than the main model domain. Then, the trajectory model increases the background concentrations within the nested main model domain, to take account of the emissions in the larger grid source domain. This converts the rural background concentrations within the model application area to urban background concentrations before ADMS-Urban actually starts its calculations of the air pollution concentrations. Therefore, it was decided to investigate the impact of running the ADMS-Urban model with the trajectory model of CRS on the annual mean and hourly calculated air pollution concentrations at the AQMS.

Running the ADMS-Urban model with the trajectory model of CRS did not significantly change the calculated annual mean NO₂, NO_x, and O₃ concentrations at the AQMS from the calculated annual means of these concentrations using the CRS only, as shown in **Table 3**. In addition, comparing **Figure 9** with **Figure 8**, running the ADMS-Urban model with the trajectory model of CRS did not significantly change the RMSE, r , or the slope of the best fit line through the origin of the actual relationship between the hourly calculated and monitored NO₂ concentrations at the AQMS. Therefore, it was concluded that using the trajectory model of CRS for running ADMS-Urban did not provide any significant improvement to running ADMS-Urban with the CRS only, on either the macro or the micro level. Therefore, using the trajectory model of CRS did not change the results of comparing the ADMS-Urban model, with rural background concentrations and a grid source, to the ADMS-Roads model, with either macro- or microcalibrated background concentrations.

In terms of the model runtime, running ADMS-Urban with a grid source, rural background concentrations and either the CRS or the trajectory model of CRS required 44 min to calculate

the annual mean and hourly concentrations of NO_2 , NO_x , and O_3 at a single output receptor point, the site of the AQMS. On the other hand, running ADMS-Roads with the CRS and either the macrocalibrated or microcalibrated background concentrations required 9 min to calculate the annual mean and hourly concentrations of NO_2 , NO_x , and O_3 at the same output receptor point, the site of the AQMS, on the same computer. Therefore, compared to running ADMS-Urban, using ADMS-Roads with the background concentrations calibration technique not only improved the air quality predictions of the air pollution model on the macro and micro levels, but it also saved 35 min of the model runtime for each output receptor point. This saving in the model runtime, when related to an output grid with a large number of receptor points, constitutes a significant reduction in the air pollution model runtime.

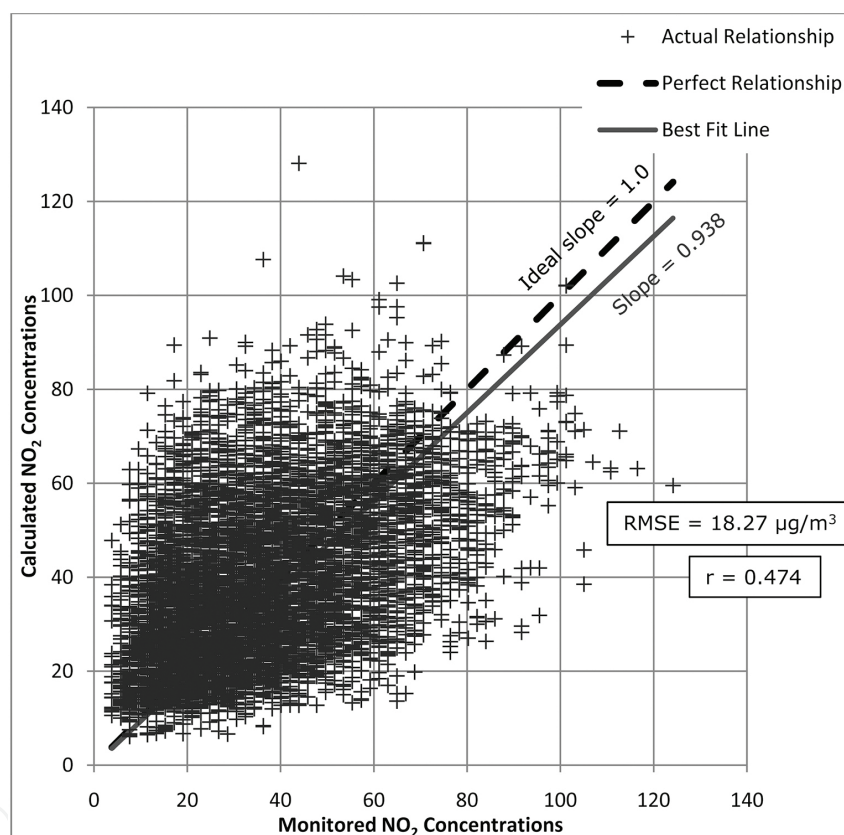


Figure 9. Scatter diagram of monitored versus calculated hourly NO_2 concentrations at the AQMS by ADMS-Urban with the trajectory model of CRS.

6. Conclusions and recommendations

The mathematical algorithm implemented by VBA computer programming in Section 2 was necessary for the processing of large files of primary traffic flow count data that were recorded every 5 min for all of the year 2006. The computer program outputs for each main road in the Dunkirk AQMA were the AADT flow, and the hourly and monthly traffic profiles for the air

pollution model. The application of this computer program significantly reduced the processing time and effort, which may allow an increase in the number of road links that can be modeled in air pollution dispersion models. This improves the model accuracy, and thus increases the reliability of air quality predictions.

The application of the VBA computer program also helps to avoid the potential human errors that may arise during the manual processing of large files of traffic flow input data, which may further increase the reliability of air pollution dispersion models. The high resolution of the primary traffic flow data for which the program can start the processing makes this computer program suitable for a broad range of other road links with similar or less traffic flow data resolution.

The macrocalibration 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 microcalibration Eqs. (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 investigation is required into the adaptation of the macrocalibration and microcalibration equations for modeling 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 macrocalibration, and one equation for the microcalibration, of the input background concentrations.

For the hours with missing monitored air pollution concentrations, the microcalibration equations were unusable. This was addressed by using the macrocalibrated background concentrations for these hours, as discussed in Section 3.2. As the macrocalibrated background concentrations give less precise calculated concentrations on the hourly level (see Sections 3.2 for details), 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 macrocalibrated 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 microcalibration of the rural background concentrations of these hours, based on the meteorological data and the microcalibrated 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% (see Section 4 for details). 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 microvalidation 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.

In terms of the error between the annual means of calculated and monitored NO₂ concentrations, using ADMS-Roads with only the macro- or microcalibrated background concentrations was more accurate than using ADMS-Urban with a grid source and rural background concentrations. Moreover, in terms of the error between the hourly calculated and monitored NO₂ concentrations, using ADMS-Roads with only the microcalibrated background concentrations was much more accurate, although slightly less accurate with only macrocalibrated background concentrations (see Section 5 for details). Using the trajectory model of CRS in ADMS-Urban did not significantly change the error between the monitored and calculated concentrations otherwise obtained, and so effectively did not change the comparative results between using ADMS-Roads and ADMS-Urban.

Replacing the grid source with either the macro- or microcalibrated background concentrations can save up to 35 min of the model runtime for each output receptor point. This saving in the model runtime, when related to an output grid with a large number of receptor points, constitutes a significant reduction in the air pollution model runtime. The microcalibration mathematical equations did not require any input data to start the iterations, apart from the monitored air pollution concentrations. In comparison, the grid air pollution sources require precise input data for the air pollution emissions which may impede their usage in air pollution modeling of areas without a precise emissions inventory.

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