A NOVEL METHOD FOR DEFINING HOURLY BACKGROUND NO_2 AND PM_{10} CONCENTRATIONS FOR USE IN LOCAL AIR QUALITY MODELLING STUDIES AND COMPARISON TO EXISTING PRACTISES

A. DONNELLY[1], B. BRODERICK[1] & B. MISSTEAR[1]
[1]Department of Civil, Structural and Environmental Engineering, Trinity College Dublin, Ireland.

ABSTRACT
The accuracy of air quality modelling studies is significantly influenced by the values adopted for background concentrations. In the absence of a reliable method of combining modelled and background concentrations, it has been common practice to sum the percentiles or annual means of each contribution to obtain a value for comparison with short-term limit values. This is often not appropriate as in many cases the meteorological conditions producing high concentrations from the source do not correspond to those resulting in high background concentrations. A novel method for predicting variable hourly background NO_2 and PM_{10} concentrations based on diurnal and seasonal variations and variation with wind speed and direction has been developed and compared to a baseline method. The variable method has been compared to commonly applied methods such as the annual mean or percentile method. Furthermore, the validity of a number of equations derived in the UK to add background concentrations to modelled stack contributions has been examined for Irish conditions. The equations allow a total percentile concentration to be predicted at a given receptor based on an annual mean background concentration and hourly modelled concentrations. A theoretical line source was modelled using Caline4 and corresponding meteorological data, and the addition equations applied using monitored background NO_2 and PM_{10} data. The methods were also tested for a point source, modelled using the Point source Gaussian plume equation. Baseline values were calculated by addition of the relevant hourly or daily background concentration to the modelled concentrations to produce a full year of total hourly or daily concentrations. Percentiles and annual mean values, and corresponding 95% confidence limits were calculated directly from this data set and concentrations predicted by each method assessed for agreement. The variable method was found to produce the best results for both NO_2 and PM_{10} when modelling a point and line source. Of the technical addition equations, the sum of squares method performed best for PM_{10} and NO_2. The annual mean and the percentile methods performed poorly in all instances producing very large under- and overestimations highlighting the importance of this research. It is anticipated that this novel method will produce significant improvements in the overall accuracy of local air quality modelling studies.
Keywords: background concentrations, dispersion modelling, limit values, modelled concentrations, NO_2, NO_x, *percentiles,* PM_{10}.

1 INTRODUCTION

Air quality dispersion models are frequently used in the assessment of air quality in urban and rural settings. Commonly used models provide a prediction of concentrations at a given receptor due to the source(s) being modelled. The background concentration must then be added on to the modelled output at the discretion of the air quality modeller. Recent research had shown that the values adopted for background concentrations can have a significant effect on the accuracy of the overall modelling study [1, 2]. In the absence of a reliable method of combining source and background concentrations, it has been common practice to sum the corresponding percentiles or annual means of each the background and the source concentrations for comparison to limit values, both to demonstrate compliance with limit values and in the development of environmental impact statements (EIS). This is often not appropriate as in many cases the meteorological conditions producing high concentrations from the line or point source do not correspond to those resulting in high background concentrations. Other common methods used include a number of technical equations derived in association with

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the UK Environment Agency [3–5] in an attempt to more accurately determine total concentrations. These equations produce a total percentile concentration using modelled annual mean and percentile concentrations and an annual mean background concentration as input. The validity of these equations for Irish conditions was examined by the authors in a previous study by comparison of predictions to baseline values [6]. Some of the methods (namely sum of squares method) were found to provide significant improvement over commonly applied methods such as the annual mean. However, all methods produced values outside the confidence limits and still required additional information such as corresponding background percentile values that are frequently unavailable, thus inducing further uncertainty into the analysis.

This paper builds on these results and examines a novel method of predicting hourly concentrations for an entire year or number of years. The method accounts for diurnal and seasonal variations and variation with wind speed and wind direction. The method is advantageous over common methods in that (while still requiring only a prediction of the annual mean background value) it provides predictions of values for comparison to short term limit values and also allows prediction of total hourly concentrations through direct addition of modelled and background values on an hourly basis.

The variable background method is introduced and its validity and applicability assessed. It is compared to common simple and technical methods. A theoretical line source was modelled using Caline4 and these equations applied using monitored hourly and daily NO_2 and PM_{10} data from two background sites in Ireland. Hourly and daily meteorological data was obtained from meteorological sites located close to the air monitoring sites. The equations were also tested for addition of background NO_2 concentrations to a modelled point source; modelled using the point source Gaussian plume equation. Baseline total concentrations were produced through hourly (or daily) addition of the modelled output to the background concentration. Percentile or annual mean concentrations were then calculated from the combined data set. The results produced by each of the simple methods were compared to these baseline total values.

2 METHODS

2.1 Monitoring sites

The Environmental Protection Agency (EPA) monitors air quality at 26 stations throughout Ireland [7]. Pollutants monitored include particulate matter, ozone, NO_x, SO_2, lead, CO, and benzene. The variable background method was derived using data from background sites in this network. It was therefore necessary to use an independent background site to validate the procedure. In the case of NO_2, data from Wexford site, a suburban monitoring station, have been used. This site is located on the eastern side of Wexford town (which has a population of 9,000) and is surrounded by a number of reasonably trafficked roads. Hourly NO_2 data are available from March 2005 to March 2006. The PM_{10} site is located at Heatherton Park in Cork city on the southern side. The site is similar to that at Wexford in that it is surrounded by a number of national roads with high traffic flow. Daily average data are available for 2006. Further to these two sites, NO_2 and PM_{10}were monitored from August 2008 to August 2009 at a rural background site at Clonee, Co. Meath providing an additional validation site.

For the purpose of this work, three meteorological stations were chosen to correspond with the air quality monitoring stations. The first station is Johnstown Castle in Co. Wexford and hourly data were obtained for use in the Wexford NO_2 modelling study. The second site at Cork Airport in Co. Cork was used in the Heatherton Park PM_{10} modelling study. Hourly wind direction, wind speed, and stability class data were obtained from each site. To correspond with the monitoring undertaken at Clonee in Co. Meath, data from Dublin Airport were used. Meteorological data at an hourly resolution was used as input to the model for both NO_2 and PM_{10}. In the development of the updated background concentrations, hourly data were used for NO_2 while the data were averaged to 24 hours for the derivation of PM_{10} data.

2.2 Dispersion models

The Caline4 model was used to model a theoretical line source and a point source Gaussian plume model was developed using visual basic and Microsoft excel as a user interface to model a theoretical point source.

2.2.1 Caline4

The Caline4 model is a line source model developed by the California Department of Transportation. Caline4 divides individual highway links into a series of elements from which incremental concentrations are calculated. The model is based on the Gaussian dispersion equation and the concentration at a receptor located downwind of the road is modelled using the crosswind finite line source Gaussian formulation. Caline4 computes receptor concentration as a series of incremental concentrations due to the contribution from several shorter lines sources each of length equal to the horizontal dispersion coefficient or a fraction of it. The model computes the concentration for a maximum of six segments within 3 times the horizontal dispersion coefficient of the receptor. In this work we have considered a single theoretical 1km link at each site with variable receptor locations and road orientations. Generalised composite emission factors were developed based on the National Atmospheric emissions inventory (NAEI) (http://www.naei.org.uk/datachunk.php?action=search) and assumed vehicle composition fleet data in Ireland [1]. The emission factor for NO_x was assumed to be 1.08 g/km and for PM_{10} it was assumed to be 0.04 g/km. Annual average daily traffic flows (AADTF) of between 1,000 and 200,000 veh/day were assumed.

2.2.2 Point source Gaussian plume model

The Gaussian plume model is a commonly used air pollution model based on the description of the three-dimensional concentration field generated by a point source under stationary meteorological and emission conditions [8]. For predicting NO_2 concentrations including a term for reflection at ground surface (based on the assumption that no pollutant is absorbed by the ground), the equation takes the following form,

$$c(x,y,z) = \frac{q}{2\pi\sigma_h\sigma_z\bar{u}} exp\left[-\frac{1}{2}\left(\frac{y}{\sigma_h}\right)^2\right]\left\{exp\left[-\frac{1}{2}\left(\frac{(z_s-H)}{\sigma_z}\right)^2\right] + exp\left[-\frac{1}{2}\left(\frac{(z+H)}{\sigma_z}\right)^2\right]\right\} \quad (1)$$

Where $c(x, y, z)$ is the concentration at point x, y, z; Q is the emission rate; σ_h and σ_Z are the horizontal and vertical standard deviations of the plume concentration spatial distribution (vertical and horizontal dispersion coefficients) calculated based on the downwind distance of the receptor form the source, d; $\bar{u}$ is the average wind velocity vector. H is the assumed height of emission of the plume, including allowance for plume rise. The reflection at ground surface term was not included for the modelling of particulate matter as it is assumed to be deposited on the ground when the plume reaches ground level.

Dispersion equations based on the Pasquill stability classes were used to derive σ_h and σ_z. These equations can be found in Masters [9]. As input, the model takes receptor point coordinates, emission rate data, hourly or daily wind speed, wind direction, and stability class data. As output, the model produces an equivalent set of concentrations at a given receptor. Emissions of between 0.5g/s and 100g/s were assumed for NO_x and that of between 0.5 g/s and 1000 g/s for PM_{10}.

2.3 Addition methods

The following section describes both commonly used addition methods and a selection of the simple methods developed by Abbott and Downing [5] and Stedman *et al.* [3, 4] which were found to be most accurate. Finally, the variable background method is presented. A more detailed examination of the accuracy of the technical methods for Irish conditions is presented in [6].

2.3.1 Baseline method (B)

The monitored background concentrations were added hour by hour in the case of NO_x and day by day in the case of PM_{10} to the modelled concentrations. The oxides of nitrogen emitted from stacks and exhausts are a combination of NO_2 and NO. In order to account for the limitation of NO_2 based on the conversion of NO to NO_2, it has been assumed using methods validated for Irish conditions in Ganguly and Broderick [10] that the total NO_2 concentration at the receptor is calculated as follows,

$$NO_{2(total)} = 0.22 NO_{x(road)} + NO_{2(background)} \tag{2}$$

Therefore, the output from the model has been multiplied by 0.22 to obtain a realistic NO_2 concentration at the receptor. This is the assumed modelled NO_2 data. The addition equations have been applied using this value and not requiring any information about background ozone concentrations. If ozone data are available ozone limiting methods can be applied as was carried out in [6].

Using each of these methods for NO_2 and direct daily addition for PM_{10}, relevant percentile values were subsequently calculated from the total data sets. This is assumed to be the true value. Hours where monitored data were missing or where the model was unable to calculate the concentration due to low wind speed were left out of the analysis.

Due to the non-normality of air pollution data a non-parametric method was used to estimate the percentile concentrations as described by Gilbert [11]. The n data are ordered to obtain sample order statistics. To estimate the percentile, the value of *x* corresponding to *p* is required. To estimate this value is calculated, and then the estimated percentile, is the order statistic. If this is not an integer then linear interpolation is used. In order to determine whether the differences between the baseline method results and the subsequent results obtained from the various methods are due to chance variation or systematic differences, confidence limits were calculated for the baseline percentile values. To estimate the lower and upper confidence limits for the percentile, the lower limit l and the upper limit u were computed. These values are order statistics and the corresponding value can easily be found by the same method as for calculating the percentile.

Where Z and α relate to the standard normal distribution.

$$l = p(n+1) - Z_{1-\frac{\alpha}{2}}[np(1-p)]^{1/2} \tag{3}$$

$$u = p(n+1) + Z_{1-\frac{\alpha}{2}}[np(1-p)]^{1/2} \tag{4}$$

2.3.2 Commonly applied methods

There are a number of methods commonly employed to add the background concentration to the modelled contribution. A conservative method is to add the relevant percentile for the modelled contribution to the relevant percentile for the background contribution (percentile method, (P)). In each case the subscript q refers to the percentile of interest.

$$T_q = S_q + A_q \tag{5}$$

Alternatively the annual mean background concentration (AM) or twice the annual mean background concentration (2AM) is added directly to the percentile modelled contribution.

$$T_q = S_q + A_m, 2A_m \tag{6}$$

2.3.3 90th percentile method [3]

The assumption underlying the 90th percentile method (9P) is that the stack contribution is considerably greater than the background contribution and that the latter can be conservatively estimated to be equal to the annual 90th percentile background concentration.

$$T_q = S_q + A_{90} \tag{7}$$

2.3.4 Sum of squares [3]

The sum of squares method (SS) uses the additive properties of the variances of the stack and background concentrations and assumes that the two concentrations are not correlated. It is also assumed that the concentration distributions are similar.

$$T_q^{No_2} = A_m + S_m + \sqrt{(S_q - S_m)^2 + (A_q - A_m)^2} \tag{8}$$

2.3.5 Annual mean and twice annual mean stack contributions [3]

The annual mean (AMS) and twice annual mean stack (2AMS) contribution methods assume that the stack contribution is small relative to the background concentration and high percentile events are mainly due to elevated background concentrations.

$$T_Q = S_M, 2S_m + A_q \tag{9}$$

2.3.6 Maximum annual mean and maximum twice annual mean [3]

The maximum annual mean (MAM) and maximum twice (M2AM) annual mean methods use the larger of the annual mean stack method and the annual mean method and the twice annual mean stack method and the twice annual mean method respectively.

2.3.7 Variable background method

A novel method for predicting a background concentration data set that varies with every hour or day and with day of the week or weekend over the course of an entire calendar year or years has been developed. The method can be expanded to include additional data if available but in this instance the most basic form of the method has been assessed. In keeping with the above addition methods the only additional information required is an annual mean background concentration. Based on data from all continuous urban and rural background monitoring sites in Ireland (aside from the baseline data sets), default diurnal variations in background NO_2 and PM_{10} concentrations were calculated as follows. Values in each data set were normalised and grouped into rural or urban sets, respectively. Weighted average variations were obtained and bootstrapping methods used to derive 95% confidence intervals about these variations. Typical diurnal variations were therefore available for both winter (Oct–Mar) and summer (Apr–Sep) at urban and rural sites respectively. Variations were divided into weekday and weekend variations as a known distinction in background levels exists. Fractional hourly NO_2 and PM_{10} values are then multiplied by the annual mean value to obtain typical diurnal variations. For a conservative estimate the upper confidence limit can be used. However, in this instance the mean values have been adopted. For NO_2, the diurnal variation must be accounted for since the short-term limit value is expressed as an hourly average. For PM_{10} this is not essential for limit value comparison since the short term limit value is a daily average. However, use of the default diurnal variation in PM_{10} will allow a better prediction of the individual hourly total concentrations (if modelling was carried out at hourly temporal resolution).

While it is often the case that some on-site monitoring is often carried out it has been assumed here that there are no continuous monitoring data. Default factors based on the variation of NO_2 and PM_{10} with wind speed and wind direction at a regional background site have been derived in previous study using non-parametric regression methods [12]. As such they represent the regional influences on background concentrations in Ireland, and cannot account for more local factors. While these factors are specific to Irish conditions following the methods presented in [12] using short-term (6-week) monitoring data from a regional background site in any country site specific factors can be developed. The site should be representative of regional and transboundary transport of emissions to any background site in the locality.

For each hour of the year (from which the meteorological data used as input to the model was obtained), the relevant default normalised PM_{10} or NO_2 concentration (subscript *d*) is assigned. Values are different for weekdays and weekends (subscript *w*) and for winter and summer months (subscript *s*). There are now 8,760 hourly values, one for each hour and these are denoted by $BG_{d,w,s}$. Each value is listed with the corresponding wind speed and wind direction for that hour and multiplied by the corresponding wind speed and wind direction factor. Thus, the normalised concentrations for a specific hour, $NomalisedBG_h$ is given by:

$$Nomalised\ BG_h = BG_{d,\,w,\,s} \times WS, WD_{factor} \tag{10}$$

Finally, every hourly value is multiplied by the derived annual mean value for the site to obtain the true hourly annual mean concentrations (). This is then an annual data set of hourly background concentrations. Each hourly value can be added directly to the corresponding hourly value from the modelled data set. This produces a total concentration data set composed of hourly means:

$$Hourly_{BG} = Nomalised\ BG_h \times Annual\ mean \tag{11}$$

In the case of NO_2, the 99.8th percentile concentrations can be calculated from this data set and compared to the limit value of 200 μg/m^3. Daily averages can be calculated directly from the TC PM_{10} data set and the 90.41st percentile value compared with the daily average limit value of 50 μg/m^3.

3 RESULTS

Caline4 and the point source Gaussian model (using a 50m stack) were run for NO_x and PM_{10} for a line and point source, respectively, for a duration corresponding to the monitoring period at each background site. In each case six receptors were placed in locations upwind, downwind, and at oblique positions relative to the prevailing southwesterly winds to capture differing conditions. For the line source, the models were also run six times at each location for NO_x and PM_{10}. A typical traffic flow was obtained and factored using variable AADTF and the model was run for each separate data set. For the stack emissions, the model was run six times using variable NO_x and PM_{10} emissions. Using the baseline data, the simple, technical, and variable background methods, the 99.8th percentile of total NO_2 concentrations and the 90.41st percentile of total PM_{10} concentrations were calculated for each emission rate, receptor location, and site. Results were normalised by dividing the relevant percentile value by the corresponding baseline percentile value. A number of measures were used to compare the performance of the various methods. First, the percentage of values predicted by each of the methods above the baseline value was noted. Second, the largest and smallest normalised values given by each of the methods were noted. Finally, the number of values falling within the confidence limits was noted.

3.1 Line source NO_2

The major advantage of the variable background method was found to be that it performs better than any of the simple or technical methods across all emission weightings and for various road/receptor orientations. This is shown in Table 1, which displays the results from the entire analysis. The variable background method produces the highest number of values within the 95% confidence limits at just fewer than 70%. The range in normalised values is also reasonable and the greatest underprediction is at 78% of the true value and most predictions are closer than this. The 90th percentile method produces the next highest number of values within the confidence limits. However, this method also produces a large underestimation of 55% of the true value. The sum of squares method is perhaps the best method notwithstanding the variable method. It produces 42% of values within the confidence limits and a small range in normalised values with no large underpredictions. However, it must be remembered that the sum of squares method makes assumptions about the 99.8th percentile of background hourly NO_2 concentrations that are rarely known and the uncertainty involved through such assumptions would induce a further and significant amount of uncertainty into the results.

The graphing of the normalised predictions of the 99.8th percentile of hourly NO_2 concentrations by the various addition methods for all receptor/road orientations highlights that the methods that are

Table 1: NO_2 line source results, Wexford. Including all road, receptor orientations.

Method	P	90P	TAM	AM	TAMS	MTAM	AMS	MAM	SOS	Updated BG
Number of values above baseline	48	9	8	0	0	8	0	0	9	12
Largest normalised value	1.43	1.07	1.05	0.99	0.98	1.05	0.96	0.99	1.08	1.06
Smallest normalised value	1.01	0.55	0.53	0.35	0.26	0.79	0.15	0.69	0.89	0.78
Percentage of values within 95% CL	13	48	44	15	2	46	0	15	42	69

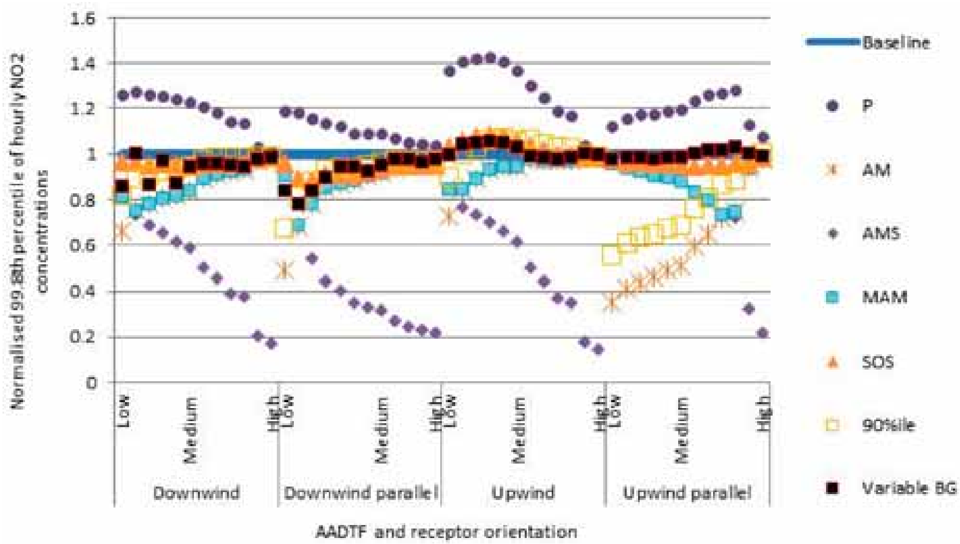

Figure 1: Normalised 99.8th NO_2 percentile values using selected addition methods at Wexford (line source).

commonly used in Ireland produce gross under- and overestimations of the true concentrations (Fig. 1). The variable background method would again appear to produce the most accurate prediction of the baseline 99.8th percentile. It is particularly relevant that reasonable predictions are observed for all road/receptor orientations (although upwind receptor locations were most favourable) and for all emission weightings. Barring the sum of squares method, all other methods performed poorly unless the modelled concentration was strongly dominant. A major advantage of the variable background method is that it is useful across all emissions weightings.

The annual mean method, which is often used, results in the underprediction of values by almost 70%. It performs poorest when the background concentration is high relative to the modelled source. The twice annual mean method is not shown here for clarity but, although slightly more conservative, produces similar erroneous predictions. The percentile method is conservative on all occasions as expected. However, the values are generally misleadingly high, particularly where the background is high relative to the modelled output. The maximum annual mean and the annual mean stack methods perform similarly to the annual mean and twice annual mean methods. The 90th percentile method provides some improvement on the aforementioned methods. However, it requires assumptions about the 90th percentile of background concentrations, which would induce a further uncertainty into the analysis and would most likely results in significant worsening of the results observed here.

In this case the variable background method was found to perform best when the receptor was located in the prevailing upwind direction or for upwind parallel sources, Table 2. This is because the N25 and the N11 are major roads located <3 km to the west of the site. The prevailing wind causes these roads to have an influence on concentrations at the Wexford monitoring station which is the assumed background in this study. Furthermore the monitoring station is located on the east of Wexford town itself. Therefore when the receptor is located on the prevailing downwind side of the hypothetical source there is superposition of the high hypothetical source events with the monitored high background events caused by the various sources in the vicinity. Although the

Table 2: NO_2 line source results, Wexford. Upwind and upwind parallel results only.

Method	P	90%ile	TAM	AM	TAMS	MTAM	AMS	MAM	SOS	Updated BG
Number of values above baseline	24	9	8	0	0	8	0	0	9	11
Largest normalised value	1.43	1.07	1.05	0.99	0.98	1.05	0.96	0.99	1.08	1.06
Smallest normalised value	1.01	0.55	0.53	0.35	0.26	0.84	0.15	0.74	0.95	0.98
Percentage of values within 95% CL	4	54	54	25	4	58	0	25	50	92

updated background values predict the average diurnal and seasonal variation well, they do not account for this superposition. This lack of accountability for high events is also observed in the sum of squares method which performs well for upwind data but poorly for downwind data. This is not, however, considered a major drawback of the variable background method as they are very accurate in predicting upwind concentrations, and in most instances, where background values are required such pre-existing sources are not present in the area. In any case, as a result of this analysis, it is recommended that in order to obtain an accurate 99.8th percentile of hourly NO_2 concentrations where the receptor is located in the downwind prevailing direction of a source additional to that being modelled, the annual mean updated background values should be increased by a factor of 1.5. The results for the upwind and upwind parallel situations only are considered to represent a truer background situation. In this instance, the range in normalised values predicted by the updated background values is very low and the greatest underprediction is at 98% of the true value. Furthermore 92% of values are predicted to be within 95% confidence limits about the baseline value. All other methods, aside from the sum of squares method are again shown to perform poorly.

Table 3 and Fig. 2 show the results from modelling a theoretical line source using Caline4 at Clonee and monitored data from 2009/2010 as the true background value. Results are, in general, similar to those at Wexford but the upwind/downwind disparity (due to the pre-existence of the source) is not observed. The variable method produces very large improvements over the commonly applied annual mean and twice annual mean methods which were frequently found to produce large underpredictions. The percentile method, while conservative, produces a large and unrealistic range in normalised values. Good predictions by the variable background method were observed for all receptor orientations and traffic flow levels. 91% of predictions are within the confidence limits and it is the best performing of all of the additions methods including the sum of sources method.

3.2 Line source PM_{10}

Table 4 shows the results for the 90.41st percentile of daily average PM_{10} concentrations at Heatherton Park in 2006 as predicted by the various simple and technical additions methods. The variable

Table 3: NO_2 line source results, Clonee.

Method	P	90%ile	TAM	AM	TAMS	MTAM	AMS	MAM	SOS	Updated BG
Number of values above baseline/58	58	8	0	0	9	9	1	1	29	33
Largest normalised value	1.52	1.02	0.99	0.97	1.07	1.07	1.02	1.02	1.12	1.08
Smallest normalised value	1.00	0.54	0.49	0.38	0.31	0.89	0.21	0.80	0.92	0.89
Percentage of values within 95% CL	21	72	59	31	17	76	12	43	86	91

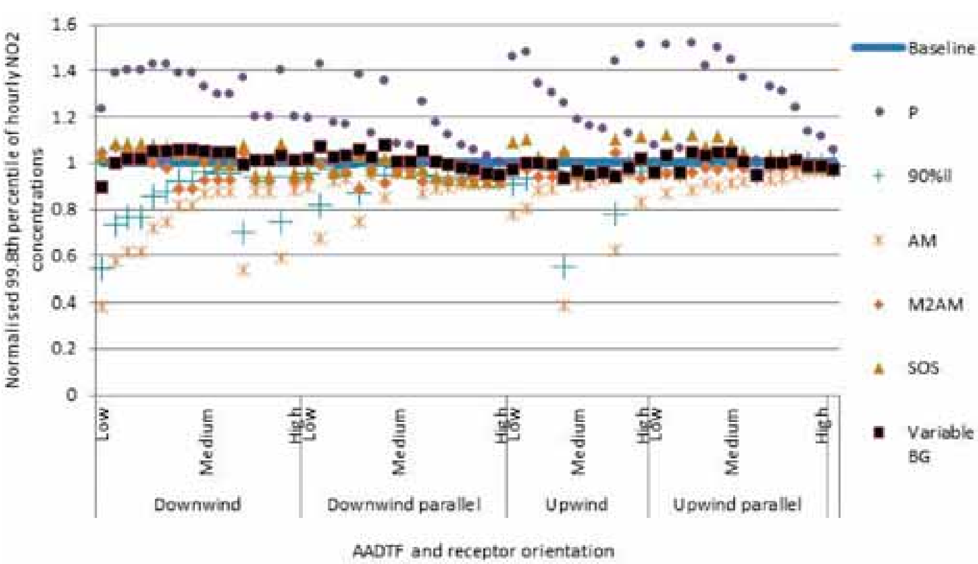

Figure 2: Normalised 99.8th percentile NO_2values using selected addition methods at Clonee (line source).

background method performs well producing 100% of values within 95% confidence limits and a small range of normalised values. The greatest underprediction is at 94% of the true baseline value. It is largely advantageous over the commonly applied annual mean method which produces large underestimations. The sum of squares method is the next best method after the updated background

Table 4: PM_{10} line source results, Heatherton Park.

Method	P	TAM	AM	TAMS	MTAM	AMS	MAM	SOS	Variable BG
Number of values above baseline	22	24	8	8	24	2	10	14	12
Largest normalised value	1.33	1.38	1.16	1.12	1.38	1.00	1.16	1.18	1.09
Smallest normalised value	1.00	1.04	0.56	0.64	1.04	0.36	0.82	0.94	0.94
Percentage of values within CL	92	71	54	92	71	38	92	100	100

method. However, as noted earlier when analysing the NO_2 results, the sum of squares method makes assumptions about the 90.41st percentile of daily background PM_{10} concentrations. Such values are not known and having to estimate them would induce a further large degree of uncertainty into the results. The commonly applied annual mean method produce just 50% of values within confidence limits and significant underestimations at up to 56% of the true value. The twice annual mean method is, of course more conservative but produces more values outside the confidence limits. The percentile method performs similarly.

Table 5 shows the results from a similar analysis applied by modelling a theoretical line source at Clonee and using the monitored PM_{10} data as the true background. Results are very similar to those at Glashaboy. Both the updated background method and the sum of squares method produce 100% of values within the confidence limits. The next best method is again the twice annual mean method. Although the updated method produces a relatively high number of underpredictions, none of these is major and the largest underprediction is in fact close to the true value at 96% of its magnitude.

Figure 3 displays the results for the 90.41st percentile of daily average PM_{10} concentrations at Heatherton Park in 2006 subdivided by emission weighting and road/receptor orientation. Figure 4 shows the results from Clonee. Both charts are similar and for all road type emission weighting, the variable background method produces very consistent values that are always close to the baseline method. The annual mean method is observed to produce very large underprediction of the true value and the percentile method is generally (although not always) conservative. The sum of squares method predicts similar values to the updated background method.

Unlike the case for NO_2 in Wexford the updated background values show no difference for the various orientations. It produces an accurate prediction of the true value for all situations. Unlike the NO_2 analysis the percentile of interest in the case of PM_{10} is the 90.41st percentile of daily averages. This statistic is less susceptible to unusual or chance events than the relevant statistic for NO_2 and there is therefore less opportunity for superposition of high events on a short-term basis. Therefore the updated background data that shows typical variation performs well in the determination of this value.

While the sum of squares and the updated background methods perform similarly well, the updated background method has the major advantage over the sum of squares method in that it require no assumptions about the 90.41st percentile of daily average background PM_{10} concentrations. The sum of squares method necessarily adds an extra degree of uncertainty through this approximation.

Table 5: PM_{10} line source results, Clonee.

Method	P	90%ile	TAM	AM	TAMS	MTAM	AMS	MAM	SOS	Statistics	Variable BG
Number of values above baseline	48	39	41	6	40	48	17	23	36	48	16
Largest normalised value	1.23	1.22	1.23	1.01	1.18	1.23	1.02	1.02	1.05	2.35	1.05
Smallest normalised value	1.00	0.98	0.99	0.50	0.81	1.00	0.43	0.85	0.98	1.00	0.96
Percentage of values within CL	56	56	56	31	77	56	63	94	100	8	100

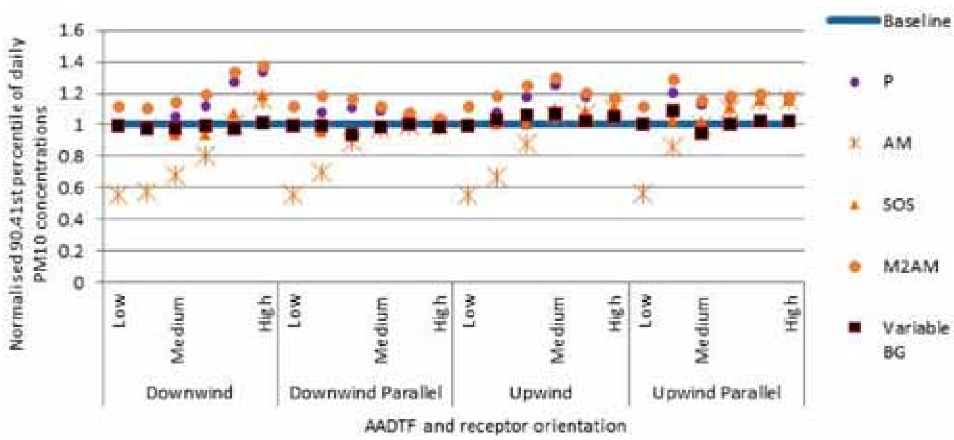

Figure 3: Normalised 90.41st percentile PM_{10} values using selected addition methods at Heatherton Park (line source).

3.3 Point source Gaussian plume model NO_2

Table 6 displays a comparison between the various methods for predicting the 99.8th percentile NO_2 concentration and their comparison with the actual 99.8th percentile NO_2 concentration calculated by direct addition of the background and modelled data sets at Wexford. The updated background method provides a good prediction in most instances and provides a significant improvement over the commonly applied annual mean and twice annual mean methods that require the same data (i.e. an annual mean value only). The percentile method, which is also commonly applied, produces large overestimation of the true value on most occasions. The sum of squares and the maximum twice annual mean methods provide the best approximations (apart from the updated

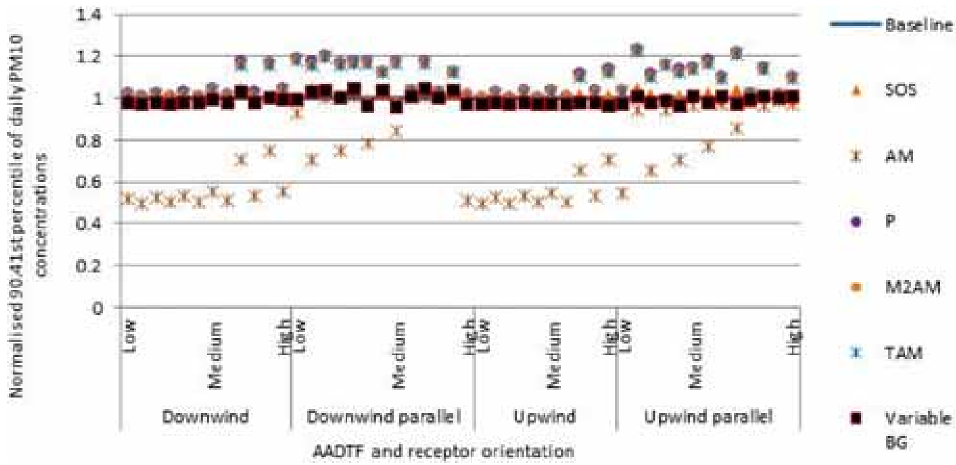

Figure 4: Normalised 90.41st percentile PM_{10} values using selected addition methods at Clonee (line source).

Table 6: NO_2 point source results, Wexford.

Method	P	TAM	AM	TAMS	MTAM	AMS	MAM	SOS	Updated BG values
Number of values above baseline	46	23	12	1	24	0	12	36	18
Largest normalised value	1.53	1.33	1.30	1.00	1.33	1.00	1.30	1.30	1.09
Smallest normalised value	0.97	0.49	0.31	0.12	0.89	0.11	0.79	0.88	0.92
Percentage of values within CL	13	40	45	17	57	15	60	64	66

background method) but require additional assumptions. The updated method produces the smallest range in normalised values whilst producing the highest number of values within the 95% confidence limits.

The variable background method performs well across all emission weightings and road/receptor orientations. However, as was observed for the line source, it performs more conservatively when the receptor is located upwind of the source. The reasons for this are similar to those discussed in relation to the line source, i.e. when the receptor is downwind of the source there is superposition of the emissions from the roads in the surrounding area with the hypothetical source leading to unusually high percentile concentrations. The effect is not as strong as was observed for the line source but nevertheless if the method is being applied to a situation where there is likely to be superposition of modelled and pre-existing sources, a conservative annual mean values is recommended to ensure that the maximum percentile value is established.

3.4 Point source PM_{10}

Results for the prediction of the 90.41st percentile of daily average PM_{10} concentrations at Heatherton Park due to the hypothetical line source and pre-existing background concentrations are shown in Table 7. The updated background values perform well producing 81% of predictions within 95% confidence limits. The span in normalised values is small and the greatest underprediction of the true value is by 5%. The sum of squares method performs best from the remaining methods producing 52% of values within confidence limits. The greatest underprediction is by 73%. However, there is a very large range in normalised values. The percentile method is not always conservative for predicting the 90.41st percentile because in many instances a low background and a high modelled concentrations combine (or vice versa) leading to significant changes in the distribution of the total data set. The annual mean and twice annual mean methods that are commonly applied produce large underestimations at 60% and 47% below the true value.

Splitting the data into two separate groups (due to some large overestimations dominating the Results) highlighted that the updated background values produce good estimates of the true percentile across all emission weightings and source/receptor orientations. Again the method tends to be more conservative for upwind receptor locations as was observed for NO_2 and for both species in the line source study. Aside from a number of very large overestimations that occur under specific conditions, the sum of squares methods performs reasonably well across emission weightings and for various receptor/source orientations. However, it does tend to underpredict on many occasions; if only slightly.

The very large overestimations that are observed from the sum of squares and annual means methods arise from the unusual situation whereby the annual mean modelled concentration is actually higher than the 90.41st percentile. This could occur when the receptor is located so that it is not

Table 7: PM_{10} point source results, Heatherton Park.

Method	P	TAM	AM	TAMS	MTAM	AMS	MAM	SOS	Updated BG values
Number of values above baseline/48	11	26	0	29	46	19	19	21	33
Largest normalised value	1.26	1.33	0.97	13.63	13.63	7.20	7.20	13.29	1.10
Smallest normalised value	0.71	0.79	0.40	0.53	1.00	0.30	0.87	0.93	0.95
Percentage of values within CL	21	44	10	33	25	40	50	52	81

generally influenced by the source. Unlike a line source, a particular receptor located near a point source will be affected to a very low degree for oblique winds. As the angle between the line from the source to the receptor and the wind direction tends to 90°, the downwind distance x tends to infinity and the distance y from the plume centre line becomes very large. This results in very small concentrations of pollutants for most wind directions. However, a number of high pollution events when the weather conditions are such as to cause the source to have a high impact on the concentrations at the receptor cause increases in the annual mean. These high events occur less than 35 times a year (corresponding to the 90.41st percentile) but act so as to cause a large increase in the annual mean concentration because the emission rate is very high. The updated background values are robust against this error. This situation does not tend to arise for the line source because a receptor located at any location relative to the road tends to be influenced by all wind directions in a 180° segment, i.e. the road acts similarly to a series of line sources so that relatively equal influence is observed for varying wind orientations. The same effect was not observed in the analysis of the 99.8th percentile of hourly NO_2 concentrations due to the point source because the annual mean value does not exceed or even approach this high percentile.

4 DISCUSSIONS

The methods used for determining the background concentration to use in air quality modelling study vary widely in Ireland and worldwide. Furthermore, assuming an accurate annual mean background value is known, or an estimate made, there is no clear procedure for adding this value on to the modelled value to develop a percentile concentration for comparison to short-term limit values. While the background concentration is often neglected (or assumed zero) in modelling study, its consideration often invokes the use of inaccurate or inappropriate methods.

NO_2 results showed improvement across all emission weightings and road/receptor orientations when the variable background method was used for both the line and point source modelling studies. In the case of the Wexford data, this is particularly impressive for a number of reasons. First, because the Wexford site is not a typical background site; there are peaks in the data that would not be present in a true background data set. Therefore in some cases the updated addition method underpredicts the true high percentile values. A large improvement (even over these results) would be expected for a more typical background site located some distance away from any one particular source background data set. In any case the variable method offers a strong improvement over the traditionally applied methods. Second, the sum of squares method, which performs the best out of the UK addition methods, requires information about the background NO_2concentrations which would not be reliably available in a typical study where the addition methods are required. As is the case for the updated background method, it requires information on the annual mean of the background concentrations. But furthermore, it requires the relevant percentile value of the background concentrations. It is the very fact that this is rarely available for the background data that this work was undertaken and therefore assumptions about its value induce a large amount of uncertainty into the analysis over what is presented here. In this instance it has been calculated based on the monitored background data set and as such can be considered 100% accurate (since it is being compared to the baseline data set derived from identical data). Other methods such as the straightforward percentile method also require knowledge of the relevant percentile. The results using the variable background NO_2 concentration on the Clonee data set are also very strong. The method produces over 90% of values within the confidence limits and performs better than any of the other methods and, in particular, is a large improvement over the commonly applied annual mean or percentile methods. Unlike the results from the Wexford site, the methods do not appear to perform differently for the various receptor orientations and AADTF values.

The prediction of the 90.41st percentile of daily average PM_{10} concentrations due to the line source and background concentrations at Heatherton Park is vastly improved through the use of the variable background values. In particular the methods can be applied across all emission weightings and for all road/receptor orientations. The sum of squares method also performs reasonably well but the assumptions required about the percentile of background concentrations are majorly limiting. The updated background method also performs well for the point source producing the most reliable results out of all the methods examined. In the case of the line source, some of the simple and technical addition methods were found to produce very large overestimations of the true percentile concentrations. Due to the large number of days on which the modelled concentration was zero (because of wind direction), the average modelled PM_{10} value is significantly higher than the 90.41st percentile. For each emission rate, there are a reasonable number of high pollutant events that push up the average value, while the 90.41st percentile remains low. As a result, the direct addition of modelled percentile to background percentile results in underestimation of the total concentrations for all but very low emissions (in which case the total 90.41st percentile is approximately equal to the background 90.41st percentile), whereas the sum of squares method produces very large overestimations because of the high annual mean modelled output. As the emission rate increases (in the different model runs), there is little change in the 90.41st percentile of modelled concentrations but the high pollutant events increase resulting in a higher annual mean PM_{10} concentration and a higher 90.41st percentile of total PM_{10} concentrations. At the Clonee site, the updated background method produced 100% of values within the confidence limits and, while the sum of squares method produces a similar result, it does not require any added information about the percentile values of the background concentration.

In both cases it is clear from this analysis that the use of the updated background values in preference to commonly applied methods such as the direct annual mean, twice annual mean, or percentile additions methods produces significant improvements. No extra knowledge is required to use the updated background values over these simple methods. The only assumption required is that of the annual mean and in any case that is required for any of the methods. Neither Heatherton Park nor Wexford monitoring data were used in the derivation of the updated background values and this therefore serves as an impartial validation of the values.

In general it was found that a greater degree of improvement in the prediction of the 99.8th percentile of hourly NO_2 concentrations was gained through use of the variable background method than in the 90.41st percentile of daily average PM_{10} concentrations (where in the case of the latter some of the simple methods were found to perform adequately). High background PM_{10} events have a greater tendency to occur more often than high (hourly) NO_2 events as PM_{10} concentrations are highly dependent on natural sources and processes. This makes the generation of a default data set for PM_{10} slightly more problematic. Nevertheless, the 90.41st percentile of daily PM_{10} concentrations has been remarkably well-predicted by the updated background data set. Although there are a number of underpredictions, none of them are large and the greatest underprediction was at 94% of the true value for both the line and point source. Of all the addition methods, the updated background values produced the highest number of values within the confidence limits in both modelling studies.

The variable background method can be further improved on in a case by case basis where short-term monitoring of the background concentrations can be carried out using a combination of the default values, the monitored data, and the nonparametric regression methods discussed in [12] to describe wind speed and direction variations. Such site specific results will further improve the accuracy of this novel method.

5 CONCLUSIONS

The updated background NO_2 and PM_{10} values have been validated using two independent and non-biased sites, neither of which was used in the derivation of the updated values. The new methods were found to provide large improvements over the commonly applied methods such as the annual mean and twice annual mean methods that require the same information for application. The methods also showed large improvements over the percentile method and the other UK derived technical methods such as the maximum annual mean, sum of squares, and stack methods, all of which require information about the relevant percentile of background values, which is rarely available in comparative modelling studies. The main instance where the methods were found to consistently produce slight underestimations of the true 99.8th percentile of hourly NO_2 concentrations was for receptors located downwind of the line source. This was attributed to superposition of the hypothetical source and pre-existing sources in the area. As a result, it has been recommended that in urban areas or areas where the background concentrations are influenced by one or more pre-existing sources, the annual mean value used in the updated method should be multiplied by a factor of 1.5 for downwind receptor locations. The same effect was not observed for NO_2 concentrations due to the point source whereby there was very little variation in performance of the methods for all receptor locations. In the case of PM_{10}, receptor location was not particularly important for either the point or line source in the performance of the updated methods. This is because the 90.41st percentile is a more robust statistic and the aforementioned superposition has less of an effect on its value.

Aside from the updated method, the sum of squares and ozone methods for predicting NO_2 concentrations provide the best results, providing a majority of values within the confidence limits and generally conservative results. The sum of squares method produced the best results for predicting total PM_{10} concentrations. However, overall results for PM_{10} were poorer than those for NO_2 and values showed a large range in magnitude. Such methods although not applicable for detailed air quality studies, may be useful for screening assessments of air quality in an area due to a modelled source. The major disadvantage of these methods is that they require information about the 99.8th and 90.41st percentiles of NO_2 and PM_{10} concentrations, respectively. The major advantage of the variable method is the simplicity of application and the fact that the only estimate required for its application is that of an annual mean background value. Any additional data such as on-site or representative monitoring, modelling, or meteorological variables can be incorporated into the background data set leading to an overall improvement in performance.

REFERENCES

[1] Broderick, B., Budd, U., Misstear, B., Jennings, S. & Ceburnis, D., *Validation of Air Pollution Dispersion Modelling for the Road Transport Sector under Irish Conditions, Project 2000-LS-6.3-M1*, Environmental Protection Agency: Johnstown Castle, Co. Wexford, 2004.

[2] Vardoulakis, S., Valiantis, M., Milner, J. & ApSimon, H., Operational air pollution modelling in the UK—Street canyon applications and challenges. *Atmospheric Environment*, **41(22)**, pp. 4622–4637, 2007. doi: http://dx.doi.org/10.1016/j.atmosenv.2007.03.039

[3] Stedman, J., Abbott, J., Willis, P. & Bower, J., *Review of Background Air Quality Data and Methods to Combine these with Process Contributions: Technical Modelling Aspects*, Environment Agency for England and Wales: Bristol, 2006.

[4] Stedman, J., Abbott, J., Willis, P. & Bower, J., *Review of Background Air Quality Data and Methods to Combine these with Process Contributions*, Environment Agency for England and Wales: Bristol, 2006.

[5] Abbott, J. & Downing, C., *The Addition of Background Concentrations to Modelled Contributions from Discharge Stacks*, Environment Agency for England and Wales: Bristol, 2000.

[6] Donnelly, A., Broderick, B. & Misstear, B., Assessing background air pollutant concentrations for modelling studies, evaluation of addition equations under Irish conditions. *Air Pollution XVIII*, p. 25, 2010.

[7] O' Leary, B., *Air Quality in Ireland 2007, Key indicators of Ambient Air Quality*, Environmental Protection Agency, Johnstown Castle, Co. Wexford, 2008.

[8] Zannetti, P., *Air Pollution Modeling: Theories, Computational Methods, and Available Software*, Van Nostrand Reinhold, New York, 1990.

[9] Masters, G., *Introduction to environmental engineering and science*, Prentice Hall Englewood Cliffs: NJ, 1991.

[10] Ganguly, R. & Broderick, B., Development of models for predicting roadside nitrogen dioxide concentrations and their evaluation against datasets measured in Dublin, Ireland. *Transportation Research Part D*, **14(4)**. pp. 231–239, 2009. doi: http://dx.doi.org/10.1016/j.trd.2008.12.005

[11] Gilbert, R., *Statistical Methods for Environmental Pollution Monitoring*, Wiley, New York 1987.

[12] Donnelly, A., Misstear, B. & Broderick, B., Application of nonparametric regression methods to study the relationship between NO2 concentrations and local wind direction and speed at background sites, *Science of the Total Environment*, **409(6)**, pp. 1134–44, 2011. doi: http://dx.doi.org/10.1016/j.scitotenv.2010.12.001