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Air Quality Modelling for Ireland

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Air Quality Modelling for Ireland

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STRIVE Report

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EPA STRIVE PROGRAMME 2007-2013

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Executive summary

Air pollution is the primary environmental cause of premature death in the EU (European Commission, 2013) and the most problematic pollutants across Europe have consistently been oxides of nitrogen (e.g. nitrogen dioxide (NO_2)), particulate matter (e.g. PM_{10}, $PM_{2.5}$) and ozone (O_3). While measurements form an important aspect of air quality assessment, on their own they are unlikely to be sufficient to provide an accurate spatial and temporal description of the pollutant concentrations for exposure assessment and moreover they cannot provide information regarding future air quality. Annex XVI of 2008/50/EC requires member states to “ensure that up to date information on ambient concentrations of the pollutants covered” by the Directive are “made available to the public”. This information must include actual or predicted exceedances of alert and information thresholds and a forecast for the following day of which a model is an integral part. As a result, air quality models are increasingly required for public information, air quality management and research purposes. The primary objectives of this research fellowship were to develop a calibrated air quality forecast model for Ireland capable of predicting the Air Quality Index for Health (AQIH) in each of the air quality zones in Ireland and to model the spatial variation in concentrations on a national scale.

This research project has produced three different models for NO_2, PM_{10}, $PM_{2.5}$, O_3 and SO_2, all of which are available for further use. These are:

1. A hybrid point wise 48 hour forecast model;
2. Spatial model (WS-LUR) to produce annual mean maps of air pollution on a national scale;
3. Temporal WS-LUR model.

A comprehensive review of modelling systems carried out at the outset of this research fellowship, together with consideration for key EPA objectives, informed the direction of model development. This review is available as a separate EPA report. A priority within the EPA was to produce air quality forecasts based on the AQIH. The AQIH is based on point wise measurements and in order to extrapolate these measurements to the future, statistical modelling was deemed the most suitable. The advantages of this approach were that it could be developed from first principles specific to the area of interest and completely (avoiding any reliance on a third party to supply the model or apply licensing restrictions) and the associated speed of forecast computation. Forecasts are only useful if they can be computed and made available to the public relatively quickly. The accuracy of such methods also tends to be high and of low bias as they are developed site-specifically unlike large scale deterministic models that are often developed and tested in vastly differing domains. In particular, this method was capable of producing accurate point wise forecasts without the need for a detailed emissions inventory. At the project outset, the emissions inventory was not of sufficient spatial resolution to make realistic point wise forecasts in all air quality zones by deterministic means and it would have been an inefficient use of resources to base the development of forecasts on what was currently available.

Initial model development proceeded using time series analysis in conjunction with non-parametric kernel regression, with local meteorological parameters as predictor variables. A model validation study found that this technique produced accurate forecast of ozone and SO_2 but had a tendency to under predict peak NO_2

and $PM_{10/2.5}$ concentrations. An analysis of air mass history using the HYSPLIT model was carried out which revealed certain air masses (primarily easterly and re-circulated air) were responsible for most incidence of elevated concentrations. The results of this study were used to develop a HYSPLIT add-on for the forecast model which operates by forecasting air mass history in real time and invoking a different forecasting methodology depending on the region of origin of the air. The ability of the hybrid point wise model to predict daily maximum hourly NO_2, SO_2, 8 hourly ozone and daily average PM_{10} and $PM_{2.5}$ was demonstrated by comparing a full year of modelled data with measured data at each of the AQIH sites. Index of agreement values ranged from a low of 0.80 for SO_2 to 0.88 for NO_2 and ozone, while correlation coefficients ranged from a low of 0.69 for SO_2 to 0.82 for NO_2. Full results of this validation study are contained in a separate report.

In order to provide detail on the spatial variation of concentration levels across the country, land use regression (LUR) was recommended in the model review as the most suitable technique. This technique uses surrogate spatial indicators to explain the variation in concentration levels between monitoring points. Land cover data (CORINE), DTM output, road density information and population data are all factors that influence concentration levels and data that were broadly available. In contrast to most LUR studies, circular buffers were not used in the determination of spatial predictor variables. Rather, a novel sector based approach (WS-LUR) was adopted whereby variables were calculated within 8 pre-defined sectors representing the major wind directions around each monitoring site. This approach had a dual purpose. Firstly, it accounts for the direction of influence of emission sources on air quality in a given location. Traditional LUR assumes equal influence of emissions in the area surrounding a monitoring site regardless of wind direction. This approximation may be reasonable in highly urbanised areas where emissions sources are relatively uniform in the surrounding region. However, in this study the regression was applied on a national scale and prevailing winds coupled with clear directional influenced at air quality monitoring sites meant that WS-LUR is a superior option. The second advantage of this methodology is that it increases the effective number of data points available for the regression analysis, resulting in a more robust final equation.

In conjunction with research project (2013-EH-FS-7), a set of annual mean maps within a geographic information system (GIS) environment were created and validated for each of NO_2, PM_{10}, $PM_{2.5}$, O_3 and SO_2. These provide a highly relevant source of information regarding spatial variation in concentration levels on a national scale which can be used not only for exposure studies and general air quality assessment, but also as a tool to correlate emission sources and surrogates with air quality. A temporal WS-LUR model was developed for NO_2, Ozone and PM_{10} by including hourly meteorological data in conjunction with pre-specified spatial data as predictor variables. This model has the potential to provide fast, efficient national air quality forecast maps for Ireland with minimal computational requirements.

This project has achieved key EPA objectives and has produced a fully automated and operational air quality model which produced twice-daily forecasts of the AQIH in each air quality zone in Ireland. The stepwise approach chosen for model development allowed deliverables prior to completion of the project while minimising associated risks. The models developed as part of this fellowship form solid building blocks on which future air quality modelling studies in Ireland can be based.

1 Introduction

1.1 Overview

Air pollution as a societal concern is interlinked with other environmental, social, political and economic systems. Stakeholders for research in the field of air pollution may therefore come from diverse backgrounds but with a common interest in the impacts of air pollution. Air pollution is the primary environmental cause of premature death in the EU, accounting for ten times the toll of road traffic accidents (European Commission, 2013). The most problematic pollutants across Europe have consistently been oxides of nitrogen (e.g. nitrogen dioxide (NO_2)), particulate matter (e.g. PM_{10}, $PM_{2.5}$) and ozone (O_3), whilst polyaromatic hydrocarbons (PAHs) have been recently identified as pollutants of concern (Environmental Protection Agency, 2012) and proposed new EU metrics for black carbon (BC) are under discussion (EEA, 2013). In a recent review of the evidence on the health impacts of air pollution the World Health Organization (WHO) state that the previous causal link between $PM_{2.5}$ and adverse health impact in earlier guidelines has been strengthened by recent evidence (WHO, 2012). Both short and long term exposure to $PM_{2.5}$ were noted to result in adverse health impacts, even in long term exposure studies where exposure was below the current recommended WHO annual limit of $10ug/m^3$. Such findings highlight the need for the introduction of additional short term limit values for PM to account for the health impacts of short term but relatively high exposures, such as during commuting. These findings also further highlight the dangers of long term exposure to PM such as from domestic fuel use. In addition, this review of recent evidence also highlights the links between exposure to NO_2 and mortality/morbidity. Such exposure is noted to be particularly elevated near roads as a result of traffic emissions. As such, the health impact of outdoor air pollution continues to be a global concern among the scientific community for its impacts on human health, the environment and climate change.

As a member of the EU, Ireland is required to demonstrate compliance with a number of EU limit values encompassing NO_2, PM, Sulphur dioxide (SO_2), lead, benzene, carbon monoxide (CO), O_3, arsenic, cadmium, nickel and benzo(a)pyrene. Challenges facing Ireland in meeting its obligations under the EU directives include reductions of NO_x in traffic-impacted areas (noted to be a significant contributor to air pollution in Ireland (McGettigan et al., 2000) and targeted by plans such as the "Dublin Regional Air Quality Management Plan for Improvement In Levels Of Nitrogen Dioxide In Ambient Air Quality" (Dublin City Council et al.)), reduction of $PM_{2.5}$ by 10% between 2010 and 2020 (EU, 2008) and reducing emissions from domestic solid fuel systems which contribute to high levels of PM and PAHs in towns and cities (Environmental Protection Agency, 2012).

1.2 Air quality modelling and policy relevance

While measurements form an important aspect of air quality assessment, on their own they are unlikely to be sufficient to provide an accurate spatial and temporal description of the pollutant concentrations and as a result, models are often needed (Moussiopoulos, 1997). Government departments, agencies, and local authorities increasingly rely on air pollution models for decision making in relation to air quality, traffic management, urban planning, and public health and consequently, the community which uses these models is becoming larger and more diverse (Vardoulakis et al., 2002). EU Council Directive 2008/50/EC (CEU, 2008) states that "A combination of measuring and modelling techniques" may be used to assess ambient air quality where levels over a representative period are below a level lower than the limit value. The directive goes on to state that “the sole use of modelling or objective estimation techniques for assessing levels may be possible…..” where levels are below a specified level. The transposition of this into Irish law has resulted in the recognition of modelling as an assessment technique under the Air Quality Standards Regulations Act, 2002 (DEHLG, 2002) in certain circumstances, “where the levels of pollutants are below the lower assessment threshold, modelling or objective assessment techniques may be used solely to assess ambient air quality, except in agglomerations in the case of sulphur dioxide and nitrogen dioxide” and “where fewer than five years' data are available, measurement campaigns of short duration during the period of the year and at locations likely to be typical of the highest pollution levels may be combined with results obtained from emission inventories and modelling”. The use of modelling in other situations where concentrations may approach or exceed limit values is also emphasised in 2008/50/EC (CEU, 2008) which states that “Where possible modelling techniques should be applied to enable point data to be interpreted in terms of geographical distribution of concentrations. This could serve as a basis for calculating the collective exposure of the population living in the area”.

Air quality models are an important aspect of air quality management and have two primary functions in this regard. Firstly, they can be used to provide predictions of what the air quality is going to be like both in the near (48 hours) and more distant (years) future. Annex XVI of 2008/50/EC requires member states to “ensure that up to date information on ambient concentrations of the pollutants covered” by the Directive are “made available to the public”. This information must include actual or predicted exceedances of alert and information thresholds and a forecast for the following day of which a model is an integral part. The provision of air pollution forecasts requires the development of a suitable air quality model.

Their second major function of air quality modelling is to improve our knowledge regarding the spatial variation in pollutant concentrations and the identification of the concentration gradient and peak location. The need for caution when assessing air quality based on sampling networks alone and the importance of a spatial approach has long been emphasised (Muschett, 1981, Greenland and Yorty, 1985). Anticipating and managing changes in pollutant concentrations relies on an accurate representation of the current and future chemical state of the atmosphere. Numerical models capable of simulating the chemistry and transport of constituents in the atmosphere have, over the last number of years, been developed for the analysis and forecast of transboundary transport of photo-oxidant pollutants.

An air quality model (like any model) is a representation of reality in which a number of parameters are used to calculate a result and it can be conceptual, empirical or process oriented. The more physical processes that are included in the model, the more comprehensively it will generally be able to describe reality. However, increasing the inputs and processes leads to high demands on the quantity and quality of information needed to drive the models (European Environment Agency (EEA), 2011). Traditionally monitoring has been the primary means of assessing air pollution levels but it can only provide real time information at best and cannot provide the spatial coverage of modelling. However, it must be noted that model results are based on their input data and some models require extensive data that may be unreliable or difficult to obtain consistently. The accuracy of any model is dependent on the quality of the input data coupled with the ability of the model to represent real world chemical and physical responses.

1.3 Objectives

The overarching aim of this research was to develop an air quality model for Ireland that could be implemented to produce short term forecasts of the pollutants outlined in the CAFÉ Directive, particularly targeting NO_x, PM_{10}, $PM_{2.5}$, SO_2 and O_3. The model would furthermore be used to anticipate pollution episodes, to aid local and regional air quality management and for further research into population exposure. There were a number of specific objectives which included:

a) Review the applicability of current models and previous relevant studies relating to forecasting air quality levels in Ireland and internationally.
b) Assess the applicability of existing models to Ireland.
c) Participate in relevant EU initiatives on modelling.
d) Build, analyse and contribute to emissions data on a local and regional scale.
e) Develop a GIS-based statistical model to determine the spatial variation in background concentration levels of pollutants on a national scale at short and long temporal resolutions.
f) Develop a calibrated air quality forecast model for Ireland.

Such a model needs to be capable of being run routinely with minimum resource requirements. Routine air quality forecasts are of high importance from a public health, air quality management and scientific perspective. Densely populated areas and urban locations would benefit significantly from air quality forecasting as the population can be warned and emergency control measures adopted in advance of pollution episodes. These forecasts would necessarily be 24-72 hours in advance.

1.4 EU initiatives

Within Europe no single body has assumed formal responsibility for the development and use of particular models in specific circumstances. In contrast, the USA has adopted a more structured approach whereby the US Environmental Protection Agency cites regulatory models for specific uses. This means that model development has been structured, transparent and fully documented (Williams et al. 2011). Within the EU the development of models has mainly been driven by CLRTAP, the European Commission and the CAFÉ programme. The Directive sets out performance criteria which the model should satisfy but the choice of model is not indicated. As a result individual European countries have adopted a wide range of approaches to modelling their air quality depending on their main objectives, resources available and previous results. FAIRMODE is a concerted effort to bring together air quality modellers and harmonise modelling on a European scale. While models can be used to demonstrate compliance with EU limit values, no direction on which model to use is given by the Commission.

There are a number of EU initiatives whereby European air quality modellers run models for the whole European domain both in real time and retrospectively. The MACC/Copernicus programme generates ensemble forecasts on a twice daily basis which are made available to the public via an air quality map of Europe online. MACC-II has the overall objective of delivering reliable operational products and information that support research, European environmental policy and the development of user-specific downstream services.

1.5 AQIH

The air quality index for health (AQIH) was developed by the Air Quality Health Information Working Group comprising of members from the EPA, Met Eireann, the Department of the Environment and the HSE. The index is currently used by the EPA to provide information to the public about air quality in each of 6 air quality zones across Ireland in real time. It is a scale between 1 and 10 (Good to Poor) representing the level of air pollution. The index for each pollutant is calculated separately using set concentration ranges. The AQIH is the worst index of the five pollutants. Alongside the index itself additional information is provided regarding the health effects of air pollution and health advice to follow when using the AQIH. A priority for this fellowship as indicated by the EPA at the outset of this project was to produce forecasts of the AQIH. Since these forecasts are necessarily produced on a daily or twice daily basis for public information, practical considerations such as computing requirements, speed of computation and ease of operation influenced the direction of the work.

1.6 Context

This report provides an overview of major work areas completed as part of the research fellowship. The research project itself has produced some key, tangible outputs. Primarily, it has produced in the first fully operational air quality forecast model for Ireland. Model development has been carried out by the research fellow from first principles meaning that no licensing restrictions apply. The fellow has developed a modelling system that runs automatically twice daily to produce 24 and 48 hour forecasts of NO_2, PM_{10}, $PM_{2.5}$, SO_2 and ozone, and subsequently the AQIH. This work fulfils the key EPA requirement for an air quality forecast system that requires minimal resources to operate on a routine basis. The fellow has also developed a manual version of the model that can be run for any given date/time. In collaboration with research project (2013-EH-FS-7) national scale annual mean maps of background NO_2, PM_{10}, $PM_{2.5}$, SO_2 and ozone have been produced as part of that project. An hourly/daily national scale spatial model has also been developed.

This report provides a general overview of these major deliverables. It is not intended as a step by step guide on how to replicate the work but rather to introduce the tangible outputs that have been produced as part of this research fellowship. Details are provided in a number of publications and three detailed reports which were completed during the research fellowship. The publications are as follows:

- Donnelly, A., Naughton, O., Broderick, B. and Misstear, B. (2016). Short-Term Forecasting of Nitrogen Dioxide (NO2) Levels Using a Hybrid Statistical and Air Mass History Modelling Approach. *Environmental modelling and Assessment*. doi:10.1007/s10666-016-9532-4.
- Donnelly, A., Naughton, O., Misstear, B. and Broderick, B. (2016). Maximizing the spatial representativeness of NO2 monitoring data using a combination of local wind-based sectoral division and seasonal and diurnal correction factors. *Journal of Environmental Science and Health*, Part A, 51, 1003-1011.
- Donnelly, A., Misstear, B. and Broderick, B. (2015). Real time air quality forecasting using integrated parametric and non-parametric regression techniques. *Atmospheric Environment* (103), 53-65.
- Donnelly, A., Broderick, B. and Misstear, B. (2015). The effect of long-range air mass transport pathways on PM10 and NO2 concentrations at urban and rural background sites in Ireland:

Quantification using clustering techniques. *Journal of Environmental Science and Health*, Part A. (50), 647-658.

- Donnelly, A., Naughton, O., Broderick, B. and Misstear, B. (2015). Air quality forecasting using parametric and non parametric statistical modelling – Prediction of the air quality index for health in Ireland. *34th international Technical Meeting on Air Pollution Modelling and its Application*. 4th-8th May 2015, Montpellier.
- Naughton, O., Donnelly, A., Pilla, F., Misstear, B.D., Broderick, B. (in preparation). Ambient air quality mapping using continuous monitoring data land use regression
- Donnelly, A., Naughton, O., Broderick, B. and Misstear, B. (in preparation). A spatio-temporal model for mapping NO2 and ozone on a national scale at hourly resolution
- Naughton, O., Donnelly, A., Pilla, F., Misstear, B.D., Broderick, B. (in preparation). Mapping pointwise forecasts using novel interpolation techniques.

The reports are as follows:

- Donnelly, A., Misstear, B., Broderick, B. and Delaney, K. (2014). *Air Quality Modelling (2012-EH-FS-6) Interim Report: Review of modelling systems*. Prepared for the Environmental Protection Agency by Trinity College Dublin.
- Donnelly, A., Broderick, B. and Misstear, B. (2014). *Air Quality Modelling (2012-EH-FS-6) Interim Report: Validation of point wise air quality forecasting model*. Prepared for the Environmental Protection Agency by Trinity College Dublin.
- Donnelly, A., Broderick, B. and Misstear, B. (2015). *Air Quality Modelling (2012-EH-FS-6) Hybrid model validation study.* Prepared for the Environmental Protection Agency by Trinity College Dublin.

A brief overview of the model review is provided in the following section but for more detail the reader is referred to the full report. Section 3, which details the point wise model development, contains some reference to both the interim standard model validation study and the final hybrid model validation study but again, additional detail is contained in the reports referenced above.

2 Review of modelling systems

A model review was carried out to inform the development of an appropriate air quality forecasting model for Ireland. Air quality models have previously been developed for a range of different purposes often with distinct advantages and limitations. The aim of this review was to provide an overview of different modelling approaches. The success of any model is dependent on the availability of the necessary input data. This review was an important first step in developing a modelling system for Ireland. Various modelling techniques were analysed with consideration given to the resources and data available within Ireland at this time.

The range of individual modelling techniques discussed offer diverse and often unique advantages for a variety of purposes. An important aspect of modelling is determining which technique offers the best use of the resources and data that area available. The best model is not necessarily the most detailed or technically advanced and, fundamental to the success of a given model, is the availability at sufficient resolution of the necessary data to drive it. Since this work was carried out on a relatively restricted time frame is was important to tackle the relevant priorities within the EPA and ensure that the most urgent deliverables were made available within the shortest time frame.

A priority within the EPA was to produce air quality forecasts based on the Air Quality Index for Health (AQIH). The AQIH is based on point wise measurements and in order to extrapolate these measurements to the future, statistical regression and time series techniques were deemed the most suitable. The primary advantage of this approach was that it could be developed from first principles specific to the area of interest and completely removes the reliance on a third party to supply the model or apply licensing restrictions. Furthermore, a benefit of using such a method is the speed of computation. Forecasts are only useful if they can be computed and made available to the public relatively quickly. The accuracy of such methods also tends to be high and of low bias as they are developed site specifically unlike large scale deterministic models that are often developed and tested in vastly differing domains. In particular, this method was capable of producing accurate point wise forecasts without the need for a detailed emissions inventory. At the project outset, the emissions inventory was not of sufficient spatial resolution to make realistic point wise forecasts in all air quality zones by deterministic means, and it would have been an inefficient use of resources to base the development of forecasts on what was currently available.

In order to provide detail on the spatial variation of concentration levels across the country, land use regression (LUR) was recommended in the model review as a suitable technique. This technique uses surrogate spatial indicators to explain the variation in concentration levels between monitoring points. Land cover data (CORINE), DTM output, road density information and population data are all factors that influence concentration levels and data that were broadly available.

A stepwise approach was chosen for model development as this crucially allowed deliverables prior to completion of the project. This minimised risks and allowed the production of preliminary air quality forecasts at the end of the first year of the project. The fluid, stepwise approach adopted meant that the work completed by the EI fellow (2013-EH-FS-7) (who commenced at the end of year one) could be integrated efficiently into the current fellowship, making maximum use of resources. Details of each of the individual models developed are provided in sections 3, 4 and 5. The three primary models developed are as follows:

- Point wise forecast model (Section 3)
- Spatial mapping of air pollution (annual mean) (Section 4)
- Temporal LUR model (Section 5).

3 Point wise forecasting of the AQIH

3.1 Standard model

3.1.1 Overview

This section provides an introduction to the point wise forecast model used to predict daily maximum NO_2, SO_2 and 8 hour ozone and daily average PM_{10} and $PM_{2.5}$ at all of the sites used in the derivation of the Air

Quality Index for Health (AQIH). Necessary inputs are outlined and model outputs are illustrated. A full model validation report has been completed where details of model performance are examined.

The statistical forecast model provides point wise predictions of daily maximum NO_2, SO_2 and 8 hour ozone and daily average PM_{10} and $PM_{2.5}$ at all of the sites used in the derivation of the Air Quality Index for Health (AQIH). The model has been developed based on:

- Relationships which exist between air quality and meteorological parameters
- Long term trends in air quality levels
- Diurnal and seasonal cyclical variations at individual site types
- Persistence of concentration levels from one day to the next
- Air mass history and its relationship with NO_2 and $PM_{10/2.5}$ levels.

Aside from the actual predictions, the model provides useful information about concentration variations at each site in Ireland. This information is available for all pollutants and sites used in the AQIH. The data are available in tabular form with values for the following parameters at every site:

- Concentrations for all wind speed and wind direction combinations in both seasons
- Concentrations for each hour of the day at weekends in winter, weekdays in winter, weekends in summer and weekdays in summer (diurnal variation)
- Concentrations for each day of the year (seasonal variation)
- Weekday variations in concentration levels
- Trends in concentrations across sites
- Relationships between other meteorological parameters and concentration levels
- Air mass history forecast for background sites
- Back trajectory cluster analysis results for NO_2 and $PM_{10/2.5}$.

The model is trained for each site individually and ideally will be based on more than one year's data to enable long term trends to be captured. Typically the most recent 5 years of data have been used for model development at each site.

New sites can be added to the forecast model at any time provided there is sufficient monitoring data available on which to calibrate and train the model. Ideally monitoring would take place for a minimum of a year before the model was trained. However if necessary the model can be trained using 3 month (a season) of data and correction factors applied to develop an interim model. After a year the model would then be re-calibrated using the full data set. Where a site is discontinued (or moved), the model can continue to make predictions at that site while data are being collected at the new site.

3.1.2 Variation in concentrations with meteorological parameters

The variation in concentration levels of all pollutants shows clear correlation with wind speed and wind direction. It is important that these factors are not considered in isolation as there is generally important interaction between both wind factors. Figure 1 (left) shows a sample output (from Kilkitt) from the nonparametric model that is used to generate a wind speed/wind direction (WSWD) factor at each site (Donnelly *et al.,* 2010, 2011). This WSWD factor is used as an input to the forecast model and is calculated in operational mode using forecast meteorological data. This information can also be useful for source attribution or general analysis of air quality levels at certain sites.

Time series analysis is used to develop a diurnal and seasonal factor at each site which is also used as inputs to drive the forecast model. Concentrations vary between winter and summer months and between weekdays and weekends. An example is shown for Kilkitt in Figure 2 (left). There is a clear difference between weekdays and weekends and between summer and winter months. Considering firstly, weekday concentrations it can be seen that in winter there is a gradual increase in concentration levels throughout the day and a peak is reached in the evening at approximately 5pm. This peak is later in summer months due to longer hours of daylight and subsequently better mixing conditions. The delayed peak and gradual increase observed at this site is suggestive of a reasonably distant source or sources affecting the site. Concentrations at weekends are lower but still display the delayed peak.

In the same manner as for the diurnal variations, seasonal variations are calculated for each site. Figure 2 (right) shows the results for Kilkitt. At all sites, lower concentrations are observed during summer months (as expected). Sites with very local, dominant sources might be expected to display less percentage variation across seasons.

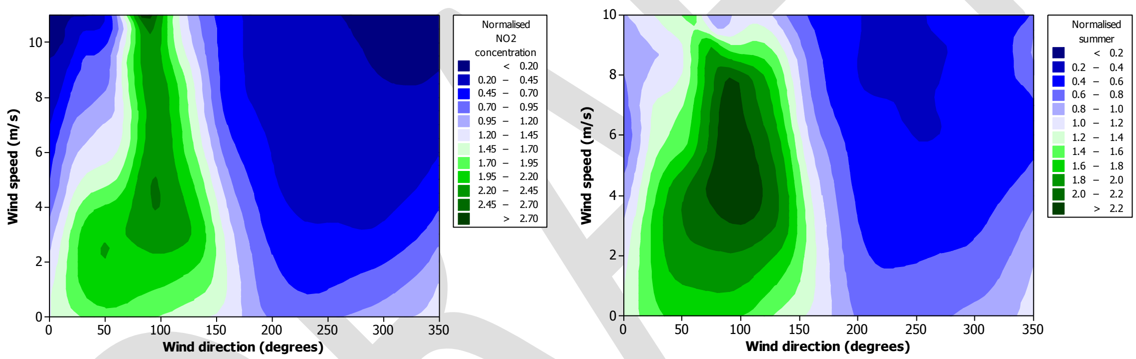


Figure 1 Variation of NO_2 concentrations at rural background site Kilkitt in winter (left) and summer (right) with wind speed and direction.

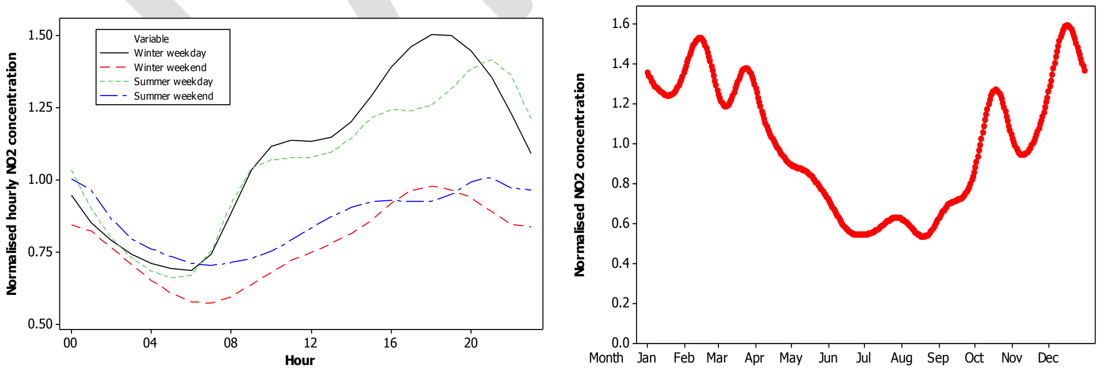


Figure 2 Diurnal (left) and seasonal (right) variation of NO_2 concentrations at rural background site Kilkitt

3.1.3 Model fitting

The model is developed using the variables outlined in Table 1 as predictor variables. The general form of the model is:

$$C = b_0 + \sum_{i=1}^{n} b_i\, x_i + \sum_{i=1}^{m} d_i\, y_i + \varepsilon$$

where C is the response variable (pollutant concentration), b_0 is the regression constant, the x_i are the meteorological predictor variables with coefficients b_i, and the y_i are the predictor variables output from the non-parametric and time series models with coefficients d_i. ε is the stochastic error associated with the regression.Technical modelling details including the derivation of the model y_i factors by advanced non parametric regression are outlined in detail in (Donnelly et al., 2015b).

Figure 1 outlines the general model development process. Measured pollutant concentrations together with wind speed, wind direction, day of the year and hourly information are fed into the non-parametric model. The $WSWD_f$, a seasonal factor and a diurnal factor are output from this model. These factors together with other meteorological data are fed into a multiple regression model as predictor variables, while measured pollutant concentration data are the response data. A first iteration model is then produced and this is assessed using a variety of techniques. Re-iteration continues until the model is accepted. This model was then validated using a separate validation data set.

A 6 month validation study was carried out on the standard point wise forecasting model and details in an associated report.

Table 2 shows key model performance statistics for each of the pollutants. In general it was found that the model predicted mean concentration variations well for all pollutants indicated by high Index of Agreement (IA) values. On occasion it can miss peak events (highlighted by reduced correlation coefficient (r) values). IA values are slightly lower for PM_{10} and $PM_{2.5}$ than for other pollutants indicting poorer point to point agreement. This is also the case for the correlation coefficient (r), although since this measure is very sensitive to outlier this is most likely due to under prediction of peak events by the model. FAC2 is considered one of the most robust measures of air pollution model performance (Borrego et al., 2011). It is the proportion of modelled values that lie within a factor of two of the measured value. It is recommended that an air quality model is considered acceptable if more than half of the model predictions lie within a factor of 2 of the observations and faulty if not – this FAC2 criterion was chosen here as it is a common metric in academic literature for assessing air quality model outputs (Derwent et al., 2010). This is 100% for ozone and is adequately high for NO_2, PM_{10} and $PM_{2.5}$ indicting no major false alarms. The value is poorer for SO_2 but this is thought to be due to the very low concentrations involved and a large number of measured zero values making the test unreliable.

Table 1 Predictor variables for standard point wise forecast model

Variable Category	Predictor Variable	Description
Model (Y_i)	$\boldsymbol{WSWD_f}$	wind speed, wind direction factor
	$\boldsymbol{S_f}$	Non-parametric seasonal factor
	$\boldsymbol{D_f}$	Non-parametric diurnal factor
	$\boldsymbol{TS_f}$	Time series forecast factor
Meteorological (X_i)	Temp	Hourly Temperature
	SunHr	Sunshine Hours
	RelHum	Relative Humidity
	AtmPres	Atmospheric Pressure
	StabilityCl	Stability Class
	NO_2h-24 NO_2h-48	Daily average NO_2 concentration at 24/48 hour lag
	NO_2max-24 NO_2max-48	Daily maximum NO_2 concentration at 24/48 hour lag
	O_3d-24 O_3d-48	Daily average O_3 concentration at 24/48 hour lag

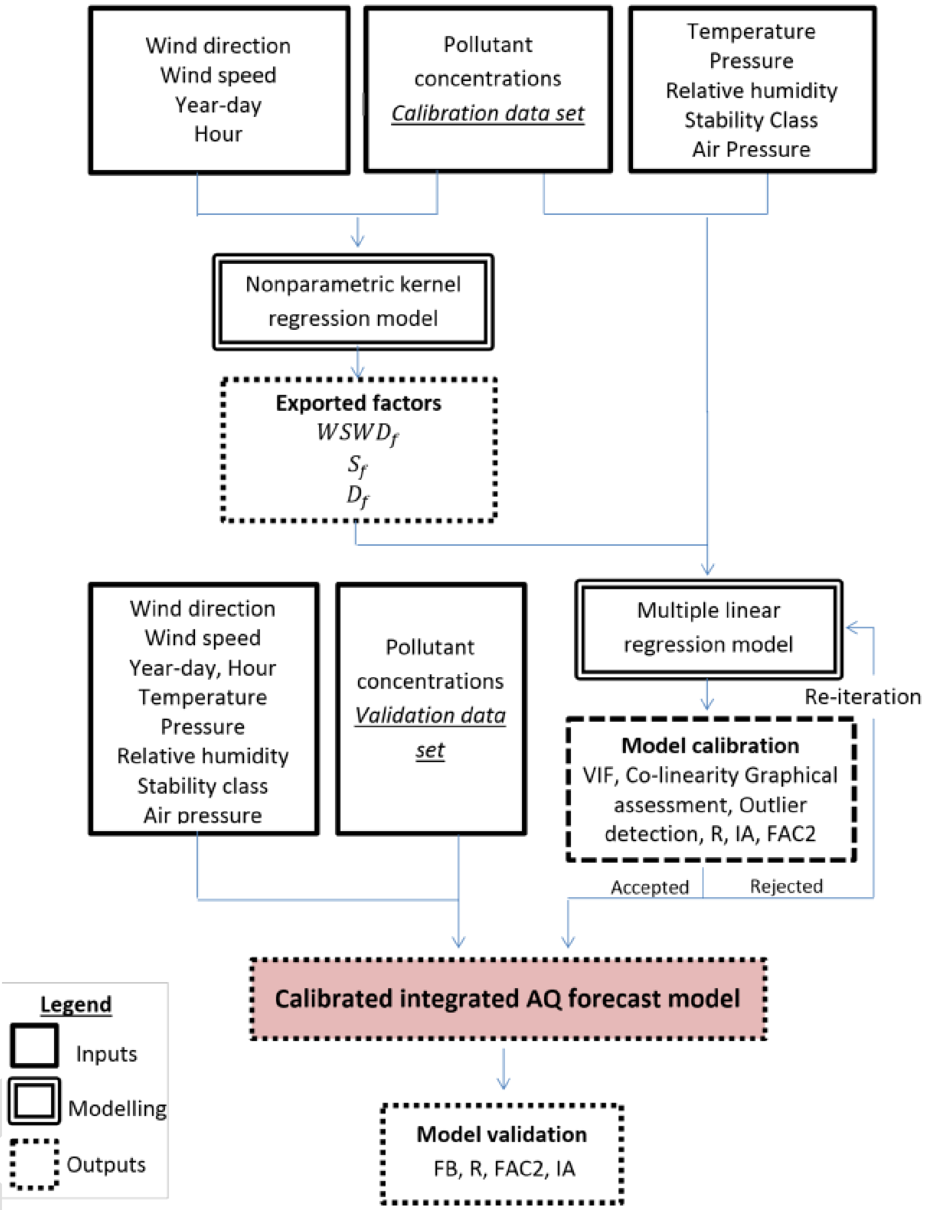


Figure 3 Methodology flowchart for standard point wise model development

Table 2 Statistical parameters for each pollutant September - March

	Ideal value	Ozone	NO_2	PM_{10}	$PM_{2.5}$	SO_2
R	*1*	0.85	0.74	0.46	0.50	0.79
FAC2	*1*	1.00	0.71	0.90	0.79	0.54
IA	*1*	0.91	0.84	0.66	0.69	0.86

3.1.4 Discussion

This model benefits from simplicity of the input data and requires very low computational resources to run making it ideally suited to providing fast and reliable real time air quality forecasts. The model validation study concluded that model performance for ozone and SO_2 was of a suitably high standard. Mean NO_2 and $PM_{10}/PM_{2.5}$ concentrations were also well predicted by the model. However, the under prediction of peak events for these pollutants warranted further investigation and led to an additional area of work being completed (air mass history modelling) and, ultimately, based on these results, the expansion of this point wise model to produce an improved hybrid point wise forecasting model.

3.2 Hybrid point wise forecast model

3.2.1 Analysis of air mass history and $NO_2/PM_{10/2.5}$ concentrations at urban and rural sites

The HYSPLIT model was developed by the US National Oceanic and Atmospheric Administration (NOAA)'s Air Resources Laboratory (ARL) and combines the Eulerian and Lagrangian approaches to track air mass movement (Draxler and Hess, 1998). While the model is capable and frequently used to calculate concentrations of pollutants, it is applied in this study to calculate back trajectories. With the HYSPLIT model, air mass paths from one region to another can be calculated and it can therefore be demonstrated whether or not the vector necessary for air pollutant transport is present (Anastasspoulos et al., 2004). When the model is run in back trajectory mode, the movement of a parcel of air can be calculated backwards in time from the receptor where concentrations were measured, allowing the origin of the pollution to be identified. Based on the results of the validation study of the standard forecast model, an analysis of air mass history in relation to concentrations was carried out with the objective of determining whether regional conditions could be contributing to peak NO_2 and PM events. The results are detailed in Donnelly et al. (2015a).

The specific aim of this work was to quantify the effects of various long range transport pathways on NO_2 and PM_{10} concentrations at a range of sites in Ireland and identify air mass movement corridors which may lead to incidences poor air quality for application in forecasting. The origin of and the regions traversed by an air mass 96h prior to reaching a receptor was modelled and k-means clustering is applied to create air-mass groups.

Trajectory cluster analysis was employed to group trajectories based on their three dimensional similarities and identify the primary meteorological pathways influencing the background site. This technique groups similar trajectories together, with the aim of minimising differences within clusters and maximizing the differences between clusters. It allows for the inclusion of re-circulated trajectories and trajectories with rapidly varying directionality. The hierarchical cluster method adopted in this study initially assumes that the number of clusters is equal to the total number of trajectories (N) and thus the spatial variance (SV) (the sum of the squared distances between end points of the clusters component trajectories and the mean of that cluster) is zero. In the first iteration, each combination of trajectory pairs is tested to compute the cluster SV. The total spatial variance (TSV) is then calculated by summing all of the clusters SV's. The two trajectories with the lowest SV are then combined into a single cluster, thus reducing the total number of clusters after the first iteration to N-1. Once paired, clusters remain together in subsequent iterations. In the second iteration, the clusters are either individual trajectories or the cluster of the initial pairing of trajectories. Again, every combination is assessed and the two clusters combined are those that result in the lowest increase in TSV. The iterations continue in this manner until the last two clusters are combined resulting in all N trajectories in one cluster. In the first number of iterations the TSV increases greatly as the number of clusters combined increases. Thereafter, it tends to increase gradually up to a point when it increases sharply, indicating that the clusters being combined are not very similar. A plot of TSV against the number of clusters will clearly indicate this change and suggest where clustering should be stopped.

Significant differences in air pollution levels were found between air mass cluster types at urban and rural sites as shown in Table 3. It was found that easterly or recirculated air masses lead to higher NO_2 and PM_{10} levels with average NO_2 levels varying between 124% and 239% of the seasonal mean and average PM_{10} levels varying between 103% and 199% of the seasonal mean at urban and rural sites. Easterly air masses are more frequent during winter months leading to higher overall concentrations. The span in relative concentrations between air mass clusters is highest at the rural site indicating that regional factors are controlling concentration levels. The methods used in this work were then applied to assist in modelling and forecasting air quality based on long range transport pathways and forecast meteorology without the requirement for detailed emissions data over a large regional domain or the use of computationally demanding modelling techniques.

Table 3 Average NO_2, NO_x/NO_2 ratio and PM_{10} expressed as a percentage of the seasonal mean at each sites for the major clusters

		Kilkitt			*Glashaboy*			*Ballyfermot*		
Winter	*Direction*	*NO_2*	*NO_x/NO_2*	*PM_{10}*	*NO_2*	*NO_x/NO_2*	*PM_{10}*	*NO_2*	*NO_x/NO_2*	*PM_{10}*
	East	160	96	154	124	105	135	149	124	147
	South west	47	101	56	59	92	81	49	85	51
	South west/West	78	105	77	80	102	65	84	96	75
	West	47	111	59	104	92	92	67	100	69
	North	78	93	71	107	100	99	113	105	112
Summer	*East/Re-circulated*	239	56	199	137	100	103	136	99	131
	South west fast	103	79	85	68	97	93	63	108	83
	South west Slow	123	104	113	82	96	87			
	West	36	141	84						
	North west	90	115	87	90	99	98	88	102	85
	North	49	96	57	90	97	98	99	99	84

3.2.2 Data partitioning at AQIH sites

Building on these results, 48 hour air mass back trajectories were calculated for two full calendar years (2011 and 2012) with hourly end-points located at Kilkitt and Claremorris monitoring stations. These two AQIH sites were assumed to represent background conditions for NO_2 and PM, respectively.

Resulting trajectories were divided into seasonal groups to account for known variability in both synoptic scale variations and air pollution levels between the winter (January – March), spring (April-June), summer (July – September) and autumn (October – December) periods. The trajectory duration was chosen as 48 hours because too short a duration may miss the actual source of the emissions and important path crossings while too long a run induces a large amount of uncertainty into the analysis and may produce misleading results. As an island with no nearby land mass to the west and south west and significant nearby land mass to the east and south, the appropriate trajectory duration may differ in Ireland than in land locked countries. A simple analysis of air masses and clusters should reveal the appropriate trajectory duration for a given country. Clustering was carried out on each seasonal group individually and the optimum number of clusters was chosen in each case by visual inspection of the TSV plots.

Results of the cluster analysis are shown in Figure 4. Six clusters were defined from January to March. These include a slow moving east/south easterly cluster and a moderate moving easterly cluster. These two clusters are associated with the highest NO_2 concentrations, (averaging 196% and 168% of the mean for this time period). From April to June only one easterly cluster is defined and NO_2 concentrations for these air masses average 161% of the mean for the time period. A similar result is observed between July and September where concentrations for the easterly cluster average 191% of the mean for the period. The easterly cluster results in average concentrations of 196% between October and December. During this time period Ireland is also frequently affected by slow moving northerly air masses representative of cold winter weather conditions. The defined cluster is of much shorter length in this season than in other seasons and its slow moving nature and its land track over parts of the UK result in average NO_2 concentrations of 153% of the mean for the time period.

Average concentrations for each cluster for NO_2, PM_{10} and $PM_{2.5}$ are displayed in Table IV, V and VI, respectively. After clustering, the variability in hourly NO_2 and daily PM between clusters was analysed and an analysis of variance (ANOVA) technique was applied to assess which cluster types led to increased concentrations. Data were partitioned into “high” background and “low” background groups at AQIH sites as illustrated by the shaded boxes in the tables.

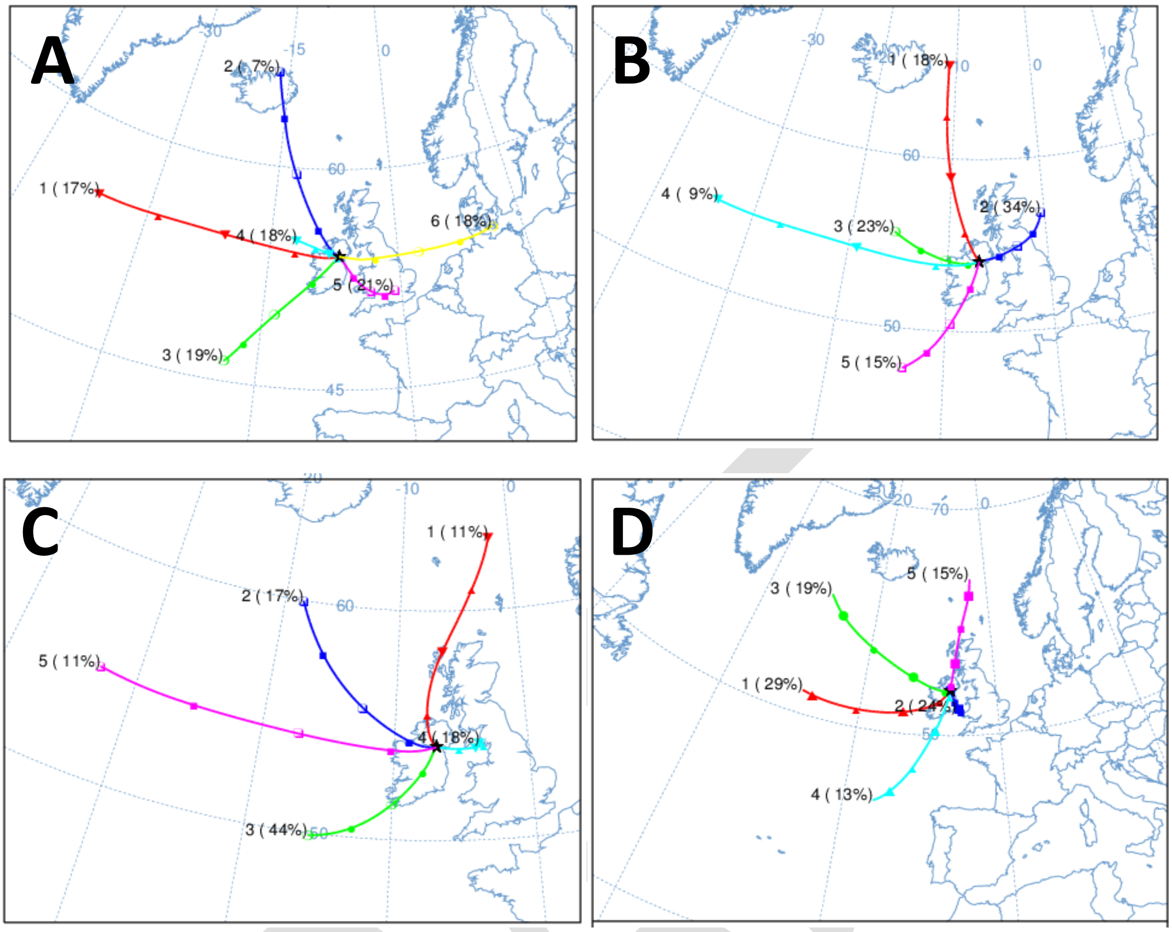


Figure 4 Cluster results from Kilkitt for 2012 and 2013 data Jan-Mar (A), Apr-Jun (B), Jul-Sep (C) and Oct-Dec (D)

Table IV NO_2 concentrations (ppb) for air mass clusters arriving at Kilkitt

Jan-Mar			Apr-Jun			Jul-Sep			Oct-Dec		
No.	Average NO_2	% of mean	No.	Average NO_2	% of mean	No.	Average NO_2	% of mean	No.	Average NO_2	% of mean
5	4.29	1.96	2	3.38	1.61	4	2.65	1.96	2	2.85	1.91
6	3.68	1.68	3	2.07	0.99	1	1.23	0.91	5	2.29	1.53
4	2.46	1.12	5	1.37	0.65	3	1.09	0.81	4	0.88	0.59
2	1.58	0.72	1	1.11	0.53	5	0.93	0.69	1	0.78	0.52
1	0.34	0.15	4	0.79	0.38	2	0.92	0.68	3	0.68	0.45
3	0.09	0.04	-	-	-	-	-	-	-	-	-

Table V PM_{10} concentrations (μg/m³) for air mass clusters arriving at Claremorris

Jan-Mar			Apr-Jun			Jul-Sep			Oct-Dec		
No.	Average PM_{10}	% of mean	No.	Average PM_{10}	% of mean	No.	Average PM_{10}	% of mean	No.	Average PM_{10}	% of mean
5	25.42	137	2	12.69	119	5	12.73	134	5	12.64	112
6	21.20	114	4	9.91	93	4	12.35	130	1	11.47	101
4	14.24	77	3	9.87	92	2	8.29	87	4	11.46	101
1	13.70	74	5	9.83	92	1	8.17	86	2	10.86	96
2	11.84	64	1	9.69	91	3	8.10	85	3	10.47	93
3	8.15	44									

Table VI $PM_{2.5}$ concentrations (μg/m³) for air mass clusters arriving at Claremorris

Jan-Mar			Apr-Jun			Jul-Sep			Oct-Dec		
No.	Average $PM_{2.5}$	% of mean	No.	Average $PM_{2.5}$	% of mean	No.	Average $PM_{2.5}$	% of mean	No.	Average $PM_{2.5}$	% of mean
5	19.52	180	2	8.49	139	4	7.16	157	5	7.89	141
6	17.58	162	3	5.27	86	5	**4.74**	104	2	5.88	105
4	8.78	81	5	5.05	83	2	3.95	87	3	5.17	92
1	5.51	51	4	5.00	82	1	3.79	83	4	4.86	87
2	5.18	48	1	4.42	72	3	3.75	82	1	4.77	85
3	3.66	34									

3.2.3 Model fitting

NO_2

The premise of the hybrid model is that pollution contributions are split into local and regional effects and different models are then developed for the "high" and "low" air mass groups. Two unique local NO_2 forecast models were developed for each condition.

- *Low Cluster Model*: This model forecasts NO_2 concentrations at all AQIH sites in Ireland for "low" air mass groups
- *High Cluster Model*: This model forecasts NO_2 concentrations at all AQIH sites in Ireland for "high" air mass groups. It is comprised of a "background" contribution and a "local" contribution. The forecast concentration at the local site on "high" days is thus the concentration computed by the "high" background model plus the concentration computed by the "high" local model.

The model fitting procedure and technical details are described in detail in Donnelly *et al (2016)*. The hybrid NO_2 model is developed using the same techniques to the standard model but, with the incorporation of the air mass history term, the modelled outputs now include:

- Forecasts for background locations in Ireland for high cluster time periods
- Forecasts at non-background locations due to local sources only for high cluster time periods

- Forecasts at all locations for non-high cluster time periods.

PM*10/2.5*

For $PM_{10/2.5}$ the same approach was also carried out and the model results were tested. However, it was found that due to the difficulty in defining a “background” location, that the results were not as strong. An alternative approach was therefore adopted, whereby the data were partitioned as above using the background monitoring data from Claremorris. A unique “high” cluster model was developed for Claremorris for high pollution days. Operationally, the results from this “high” forecast are compared to the results of the “basic” forecast at Rathmines and concentrations are other sites are multiplied by the difference to provide a better description of the regional influences on PM levels across Ireland.

Ozone/SO_2

Model fitting for both ozone and SO_2 did not make use of partitioned data but rather used total data sets. Analysis of air mass history in relation to concentrations indicated that the prediction of peak events would not be significantly improved through the inclusion of an air mass history term in the model for these pollutants.

3.2.4 Model operation

In operational model the model completes a number of tasks in sequential order to produce final forecasts of individual pollutants and subsequently the AQIH. These tasks are as follows:

- Download forecast meteorological data from the Met Eireann ftp server
- Download real time air quality data from all AQIH monitoring sites
- Download forecast hemispheric meteorological data from the NOAA server
- Run the HYsplit model in back trajectory mode for the next 48 hours
- Assess each forecast trajectory path and assign it to one of the pre-defined clusters
- If a high cluster is identified then run the “High” model for NO_2, PM_{10} and $PM_{2.5}$. Run the standard model for ozone and SO_2.
- If no high clusters are identified then run the standard model for all pollutants.

The model decision tree for NO_2 is illustrated by Figure 5. The model is written from first principles using visual basic code and presented using a Microsoft Excel Interface. Any model operational editing (such as adding a new site) must be done using Visual Basic code. Model coefficients for existing sites can be changed by adjusting the values in the model settings spreadsheet. However, this should be done with caution and only after a full calibration of response/predictor data. An SOP has been written as part of this research project to provide guidance of model updates and adjustments.

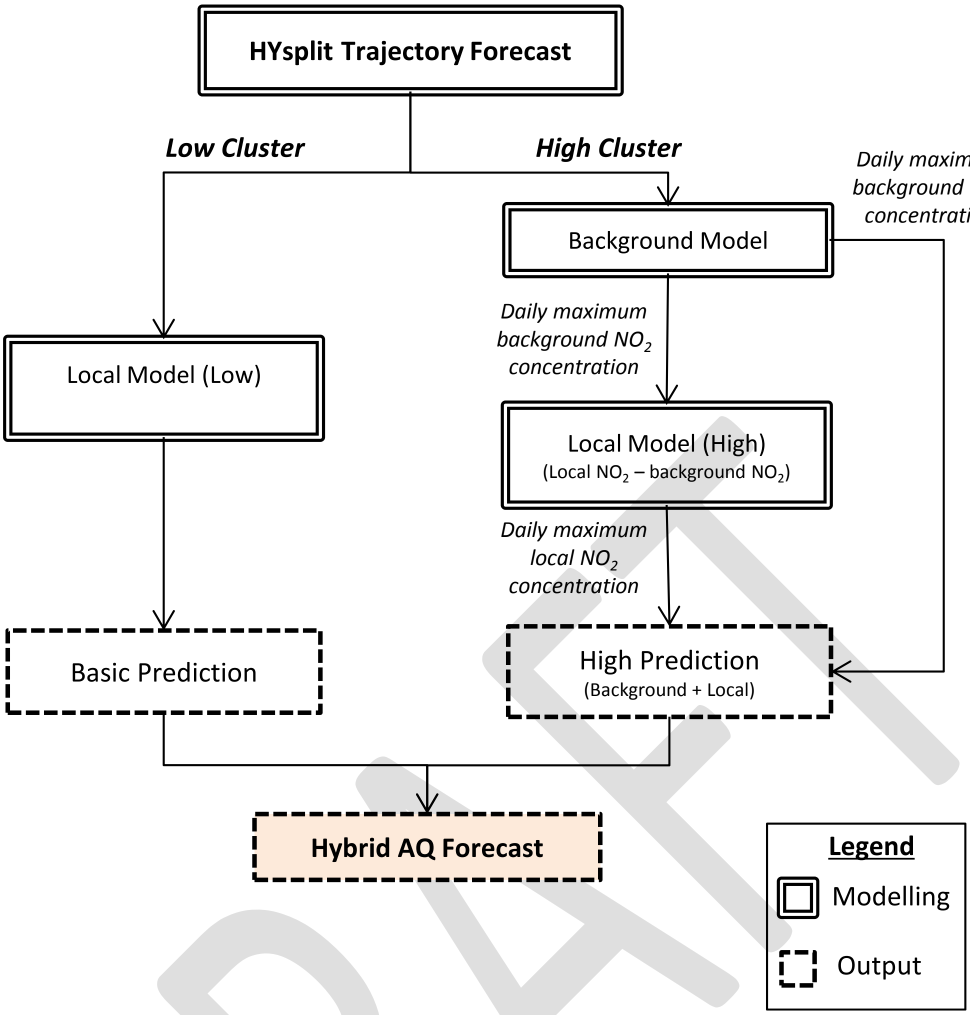


Figure 5 Hybrid model operational decision tree

3.2.5 Validation study

A comprehensive hybrid model validation study has been completed and is available as a separate report. The statistical forecast model was run daily for the time period January 2013 to December 2013 to provide 48 hour forecasts. This involved running the entire modelling system retrospectively in forecast mode. The model decision tree is shown in Figure 5. The HYsplit model was first run for every forecast day. Each trajectory was assessed and assigned to the relevant cluster. Based on this assignment either the standard or hybrid model was run for NO_2, PM_{10} and $PM_{2.5}$. The standard model was run for ozone and SO_2. 24, 48 and 3 day forecasts were produced by the model. These data were then compiled as time series for each of the pollutants. These time series have then been compared to observed time series data for the same time period. The observed data used in the comparison have been validated.

Descriptive modelled and measured statistics are shown in Table 7. The mean ozone value measured across sites is 70.25 µg/m^3. This is slightly under predicted by the model (64.46 µg/m^3). Mean NO_2 values are slightly over predicted by the model due to the shifting of the data distribution by incorporating the HYsplit term (17.4 µg/m^3 modelled compared to 13.5 µg/m^3 measured). Mean SO_2 concentrations are well predicted with an overall prediction of the average within 1 µg/m^3. The mean measured and modelled PM_{10} and $PM_{2.5}$

concentrations are very similar (<2 µg/m^3 of a difference for PM_{10} and <5 µg/m^3 for $PM_{2.5}$). Median values largely follow the same pattern as mean values and are well predicted by the model for all pollutants. There is some over prediction of the median SO_2 concentration which arises from the Kilkitt data where the model consistently over estimates the concentration. However in absolute terms the magnitude is very small.

In addition to the median and mean values the 90th 95th and 99.8th percentile values are assessed to examine the distribution of the data. These percentile values also largely correspond to the number of exceedances per year as defined by the EU limit values. For ozone, the 90th and 95th percentile are well mirrored by the modelled value. There is a small under prediction of the 99.8th percentile but this statistic is subject to a large degree of chance variation. The actual under prediction is less than 7% of the true value. NO_2 percentile values are slightly over predicted by the model (meaning a conservative estimate of the data distribution) but since the NO_2 values being considered are individual hourly values they are subject to a large degree of inherent variation. Nevertheless the model performs well at capturing the over distribution of the data. SO_2 predictions of percentile values are very good by the model with good agreement of the 99.8th percentile. The distribution of daily average PM_{10} and $PM_{2.5}$ concentrations is well captured across sites with some slight over prediction at high percentile values.

Table 8 show the statistical parameters for all modelled values. r values are good for gaseous pollutants. PM_{10} and $PM_{2.5}$ have lower r values but this can be attributed to a small number of outliers in the data set. Ideally FB should equal zero and a negative value indicates that there is some under prediction by the model. This was previously observed when using the standard model for both NO_2 and $PM_{10/2.5}$. The hybrid model has a very slight positive bias for PM_{10} and SO_2 (0.09 and 0.14, respectively) and a slight negative bias for ozone (-0.09). The negative bias in the ozone results may be due to some over smoothing in the model development at some sites. This is discussed in more detail in a later section. The positive bias is higher for $PM_{2.5}$, SO_2 and NO_2. This reflects a slight over prediction of the mean value. The Hysplit add on enables the model to forecast a greater number of high pollution events but it does also result in a slight shifting of the overall distribution of the data in a positive direction although this is not considered a significant issue. It is important to note that pollutant concentration distributions tend to be lognormal and therefore the linear measures of fractional bias (FB) and correlation coefficient can be disproportionality influenced by infrequently occurring high pollutant events.

FAC2 exceeds 70% for NO_2 and is close to 100% for Ozone. It is 93% for PM_{10} and 80% for $PM_{2.5}$. It is lowest for SO_2 but this is due to the very large number of measured zero values which the model cannot be within a factor of two of without perfect prediction (i.e. this measure fails for such conditions).

The index of agreement is a robust measure of the degree to which the measured value is accurately estimated by the model. It is a similar indicator to the correlation coefficient and also has an ideal value of 1 but unlike the correlation coefficient, the index of agreement measures the error of the modelled data rather than the direct correlation between variables and is not as sensitive to outlying data points. The index of agreement is very high for all pollutants (>=0.8) and compares favourably with values obtained in other air quality modelling studies (Zhang et al., 2012, Voukantsis et al., 2011, Kumar and Jain, 2010, Beelen et al., 2013a).

Figure 6 to Figure 10 show scatter plots of measured versus modelled concentrations (colour coded by AQIH site) for NO_2, PM_{10}, $PM_{2.5}$, ozone and SO_2. These plots together with time series plots and details validation statistics are discussed in detail in the model validation report. This is not repeated here for brevity but in general results were highly acceptable and comparable or better than results achieved by other forecast models. The hybrid model offered significant improvements over the standard model for prediction of NO_2, PM_{10} and $PM_{2.5}$ peak events.

Figure 11 illustrates the improvement that was obtained by incorporating the air mass history term into the model at Rathmines. The red symbols show that the standard model had a tendency to under predict at higher concentrations and could not account for concentrations over 45µg/m^3 at this site, in general. The hybrid model however results in a much stronger linear relationship between measured and modelled values across the entire range of concentrations.

The model was found to perform better at the urban sites than at the rural site. There are a number of reasons for this. Firstly, concentrations at the rural site are very low, which means that the monitoring instrument is often not sufficiently precise to measure near zero concentrations. As a result these values are estimated to be equal to zero or the nearest 0.01µg/m^3, which leads to an unnatural distribution within the data. Therefore, while the modelled data follow the measured data reasonably closely, the statistical tests do not account for this lack in precision and indicate poorer results. Secondly, rural sites are less impacted by local anthropogenic activities which tend to be repetitive and cyclical (e.g. rush hour traffic). Since emissions travel a greater distance prior to reaching the rural monitoring site, there is more opportunity for dispersion and transformation of pollutants. While this results in lower NO_2 concentrations, it also leads to more variability in concentration levels (albeit at much lower total concentrations).

Table 7 Modelled versus measured descriptive statistics

	Ozone (µg/m^3)			NO_2 (µg/m^3)			SO_2 (µg/m^3)			PM_{10} (µg/m^3)			$PM_{2.5}$ (µg/m^3)		
	24 hr forecast	*>=3 day forecast*	*Monitored*	*24 hr forecast*	*>=3 day forecast*	*Monitored*	*24 hr forecast*	*>=3 day forecast*	*Monitored*	*24 hr forecast*	*>=3 day forecast*	*Monitored*	*24 hr forecast*	*>=3 day forecast*	*Monitored*
Mean	64.46	67.55	70.25	17.4	17.93	13.53	7.45	7.18	6.46	18.1	17.07	16.52	15.6	14.1	11.50
Median	64.43	69.50	69.90	13.9	14.27	10.3	5.82	4.81	4.52	14.4	14.55	14.43	8.2	8.98	8.2
90th percentile	86.70	88.30	93.87	35.9	36.7	30.7	13.94	16.61	11.97	32.9	30.3	28.1	37.2	33.64	23.8
95th percentile	91.90	90.33	99.40	43.5	42.71	35.9	19.78	21.93	19.57	39.1	36.59	34.45	50.9	42.56	33.3
98th percentile	98.63	90.51	105.21	51.5	52.08	44.0	25.76	26.36	28.4	46.2	43.6	42.84	63.1	56.47	44.98

Table 8 Statistical parameters for each pollutant

		NO_2		**PM_{10}**			**$PM_{2.5}$**			**SO_2**		**Ozone**	
	Ideal value	24 hour forecast Hybrid	>= 3 day forecast Hybrid	24 hour forecast Standard	>= 3 day forecast Standard	>= 3 day forecast Hybrid	24 hour forecast Hybrid	>= 3 day forecast Standard	>= 3 day forecast Hybrid	24 hour forecast	>= 3 day forecast	24 hour forecast	>= 3 day forecast
FB	*0*	0.26	-0.278	0.09	0.02	-0.03	0.30	-0.05	-0.2	0.14	0.13	-0.09	-0.11
R	*1*	0.82	0.80	0.72	0.46	0.62	0.74	0.50	0.647	0.69	0.47	0.82	0.80
FAC2	*1*	73.4	70	93	90	93	79	79	80	77	79	99.5	99.1
IA	*1*	0.88	0.83	0.84	0.66	0.70	0.84	0.69	0.77	0.80	0.59	0.88	0.82

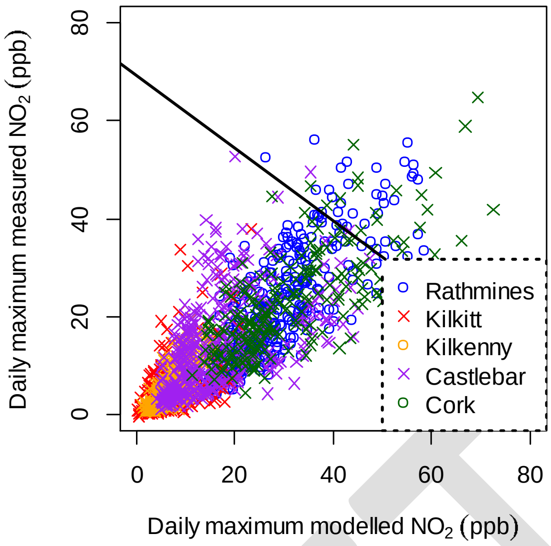


Figure 6 Measured versus modelled daily maximum NO_2 concentrations

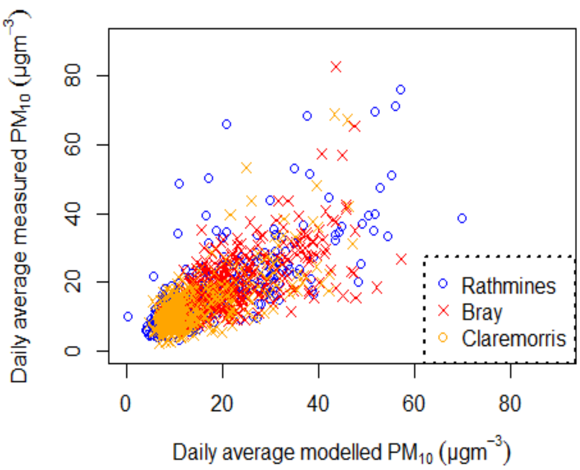


Figure 7 Modelled versus measured daily average PM_{10} concentrations for 24 hour forecasts

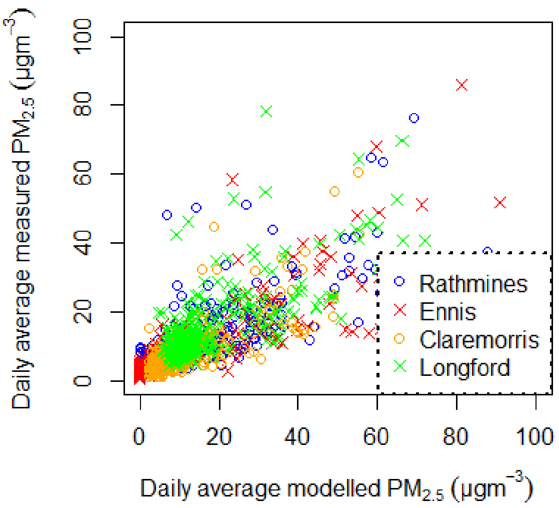


Figure 8 Modelled versus measured daily average $PM_{2.5}$ concentrations for 24 hour forecasts

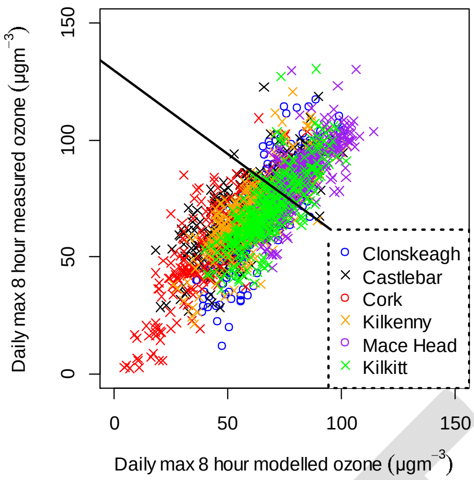


Figure 9 Modelled versus measured daily maximum 8 hourly O_3 concentrations for 24 hour forecasts

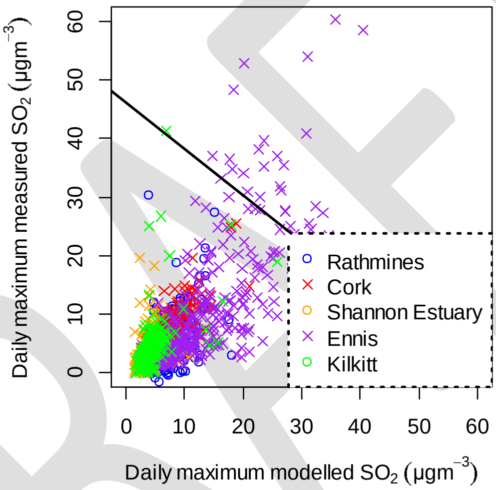


Figure 10 Modelled versus measured daily maximum hourly SO_2 concentrations 24 hour forecasts

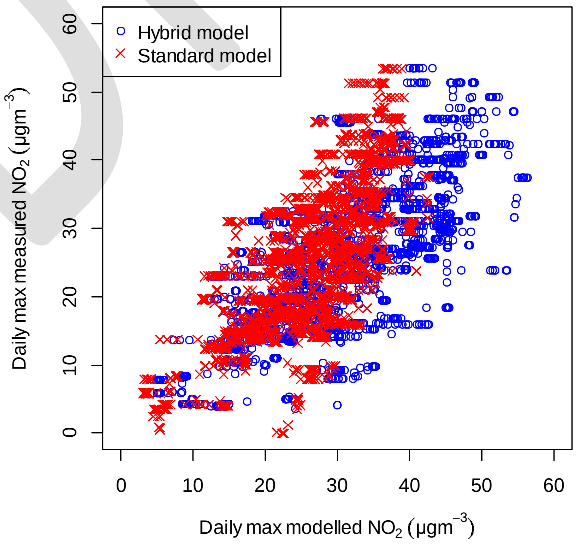


Figure 11 Observed NO_2 concentrations (rolling 24 hour maximums) at Rathmines showing improvement in hybrid model over standard model

3.3 Discussion

Real time air quality forecasting has become an area of much interest in recent years and various deterministic and statistical techniques have been used to produce functional forecasts. Numerous studies have used statistical techniques to develop air quality forecasts. Techniques adopted include multiple linear regression (Genc et al., 2010, Vlachogianni et al., 2011), ARIMA modelling (Zhang et al., 2012, Kumar and Jain, 2010), neural networks (Moustris et al., 2010, Voukantsis et al., 2011, Feng et al., 2011), nonlinear regression (Singh et al., 2012, Donnelly et al., 2015b), Kalman filtering (Hoi et al., 2010) and various combinations of these (Zhang et al., 2012, Voukantsis et al., 2011). However, most of these methods suffer from the disadvantage that they cannot capture the contribution of distant weather dependant sources and regional air mass movement. While deterministic models can account for air parcel history, they are computationally intensive, require detailed emissions inventories over the modelled domain and have a high operational cost (Zhang et al., 2012) which can make them unsuitable for real time air quality forecasting in many situations. Furthermore, many applications of real time air quality forecasting only require predictions at certain locations and in such instances, the processing required by deterministic models to provide detailed spatial variations may be a misuse of resources. As noted by Zhang *et al*. (2012) statistical models often have a better capability for describing complex site specific variations in concentrations than deterministic models, often with a higher accuracy than deterministic models.

The hybrid model developed as part of this research fellowship represents a novel way to forecast air quality routinely and accurately with minimal resource requirements. The hybrid model incorporates the advantages of the standard statistical model outlined in Donnelly *et al*. (Donnelly et al., 2015b) and combines it with the (open-source) deterministic HYSPLIT model. This allows regional effects to be included in the forecasts without the need for a complex deterministic and computationally demanding air quality model to be used. Hybrid model advantages include:

- Requires only simple input data
- Minimises model selection error by combining various statistical methods
- Low bias
- Ability to forecast cyclical and anthropogenic effects without the need for an emissions inventory
- Ability to describe complex site specific variations while including the effects of regional weather patterns
- Speed of computation
- Ease of operation.

3.4 International model application

A key underlying assumption in this hybrid approach is that the transboundary contribution to air quality at the background site is representative of that at the forecasting site. The geographic location, prevailing climatic conditions and relatively low urbanisation characteristic of Ireland make this a reasonable assumption to make in the case presented here. However, when applying the method internationally it should be considered that some areas may be influenced by heavy urbanisation, industrialisation or more complex regional air mass transport and care should be taken in the selection of appropriate background site to ensure representativeness. Multiple background sites may thus be required when applying the model across a national monitoring network, with parallel trajectory forecasts necessary to enable model selection and forecasting. Due to the low computational resources of the statistical model and the ease with which trajectory forecasts can be produced this does not represent a substantial increase in resource requirements and so this approach remains a viable option for producing fast and reliable real-time air quality forecasts in regulatory environments where resource availability is low.

3.5 Summary

- A fully operational air quality forecast model has been produced. The model runs in an automated manner to produce twice daily 48 hour forecasts of NO_2, SO_2, Ozone, PM_{10} and $PM_{2.5}$ at AQIH sites in Ireland.
- Incorporation of an air mass history parameter has resulted in a large improvement in the prediction of NO_2 and particulate matter concentrations.
- The model is quite conservative in its PM_{10} and $PM_{2.5}$ predictions. There is a slight positive bias at all sites as a result of the methodology used to account for regional air mass movement and pollutant transport. PM is one of the most difficult pollutants to model due to its wide range of anthropogenic and natural sources. Therefore, a conservative estimate accompanied by some specialist interpretation is considered to be the best means of producing a forecast. When the AQIH is forecast to be poor, air mass history and other conditions relating to PM concentrations should be examined in conjunction with the value given by the AQ model to produce a final forecast.
- During the next model calibration/training, ozone should be trained using hourly data. The use of 8 hour averages as the response variable has resulted in some over smoothing of the data.
- SO_2 values are very low at most sites. There is some over prediction by the model at Kilkitt but the very low values involved make this a relatively insignificant issue.
- Statistical parameters for SO_2 are stronger than initial model training would have suggested.
- Air quality forecasts should be made using a combination of:
  - Numerical output directly from the model
  - Assessment of forecast local meteorological conditions
  - Assessment of regional air mass movements as forecast by the Hysplit model which is built into the operational air quality model
  - Consideration of any other unusual events or conditions (e.g. volcanoes or Saharan dust episodes).
- The model should be re-calibrated using up to date validated air quality and meteorological data on an annual basis. This process will ensure that air quality trends at individual sites are well captured and any new sources in an area are identified and included in the model.
- However, it should be noted that if major changes in the emission source were to occur (such as a sudden increase in road traffic volume), the model would require re-calibration.
- In the case where new air quality monitoring sites are used for the derivation of the AQIH, the model can be used to continue to forecast at the old site until a full year of data are available at the new site. This will avoid any break in forecasts within any one air quality zone. Once at least a full year of data are available, the model should be re calibrated and updated to include this new site within the model architecture.

This model has been brought to full operational model as part of this research project and has been set up to run in a completely automated manner to provide daily forecasts of the AQIH in each zone in Ireland. The model can also be operated manually (retrospectively or in forecast mode) for any date/time where appropriate meteorological data are available.

4 Land use regression modelling – Annual mean maps

4.1 Introduction

The application of land use regression techniques provides the opportunity to produce high resolution maps of background air pollution on a national scale. Linear regression methods have frequently been employed in air quality modelling in the past (Briggs et al., 1997, Shi and Harrison, 1997, Robeson and Steyn, 1990). Konovalov et al. (2009) found that in applying model output statistics to the CHIMERE model using both linear regression and non-linear neural networking, there was no significant difference between the performance of PM_{10} forecasts carried out by each method.

Stedman et al., (1997b) developed maps of NO_x and NO_2 concentrations across the UK using an approach which involved a number of methods, one of them being regression. Firstly concentrations from monitoring stations which were representative of concentrations over areas >20km were directly interpolated. Thereafter the impact of local NOx emissions (<20km from the monitoring sites) were estimated using a box modelling approach incorporating surrogate statistics. At the time of the study emissions data for the UK were available at a resolution of 10km by 10km. Since it was noted that NOx and NO_2 concentrations vary at a much finer spatial scale than this emissions from major roads and the percentage of urban and suburban landcover were used as surrogate statistics rather than the emissions data. This could be applicable to Ireland since emission data are not currently available at a 1km resolution. The development and derivation of these maps is discussed in detail in a number of papers and reports (Stedman et al., 1997a, Stedman et al., 1997b, Abbott and Stedman, 1999, Abbott and Vincent, 1999, Stedman, 1998, Stedman and Bush, 2000, Kent et al., 2006, Stedman et al., 2007). They have been developed from a combination of emission estimates from the UK National Atmospheric Emissions Inventory (NAEI) and measurements from the national air monitoring networks (Stedman and Bush, 2000). Variable degrees of agreement between measured and modelled values were found with correlation coefficients (R^2 values) ranging from 0.33 to 0.78 for various pollutants (Stedman and Bush, 2000). It can be argued that these maps are limited in their usefulness, particularly in the area of exposure analysis as they only provide an annual mean value and no indication of shorter term values. There is also a substantial risk of double counting the source in certain cases.

Beelen *et al*. (2009) developed maps for the European Union for NO_2, PM_{10} and O_3 at 1km resolution using ordinary kriging, universal kriging and land use regression techniques. They cited the need for detailed input data together with the need for powerful computing facilities (for large areas and fine resolution) as a limitation in approaching air quality modelling through the use of dispersion models. They found that universal kriging performed the best of the three techniques with R^2 values ranging from 0.45 and 0.7.

Vienneau *et al*. (2010) note that land use regression has the potential to produce maps of air pollution on a national and European scale in a relatively simple manner to inform policy and as a basis for risk management. In their study they developed LUR models for both Great Britain and The Netherlands for NO_2

and PM_{10}. They found that the performance of models based on common data was only slightly worse than models optimised with local data. However, they advise the need for caution in transferring models across different study areas.

The spatial modelling carried out under this research project builds on many of these previous studies but incorporates a novel means of accounting for variability in prevailing wind directions and orientation of land use types in relation to receptors.

4.2 Sector based LUR

In contrast to previous LUR approaches, the approach adopted in this project did not use circular buffers. Rather, a sector based technique was used whereby the land area affecting air quality at a given site is dependent on wind direction.

The basis of LUR mapping is a multiple linear regression which uses summaries of spatial variables in the vicinity of the monitoring point. In general spatial indicators are calculated within circular buffers of varying radii around the monitoring point and the most significant used as predictor variables in the regression equation. However, circular buffers effectively apply equal weights to emission sources around a receptor, irrespective of the prevailing meteorological conditions and the relative positions of receptor and source. This limitation may be minor when LUR is applied within a local region, but when used on a national scale as required by the project, varying regional wind patterns may lead to poor model performance. Pollutant concentrations can show significant asymmetry depending on wind conditions, as demonstrated in figure 1, where the variance in NO_2 concentrations with wind speed and direction can be seen at both urban Figure 12: left) and rural sites (Figure 12: right).

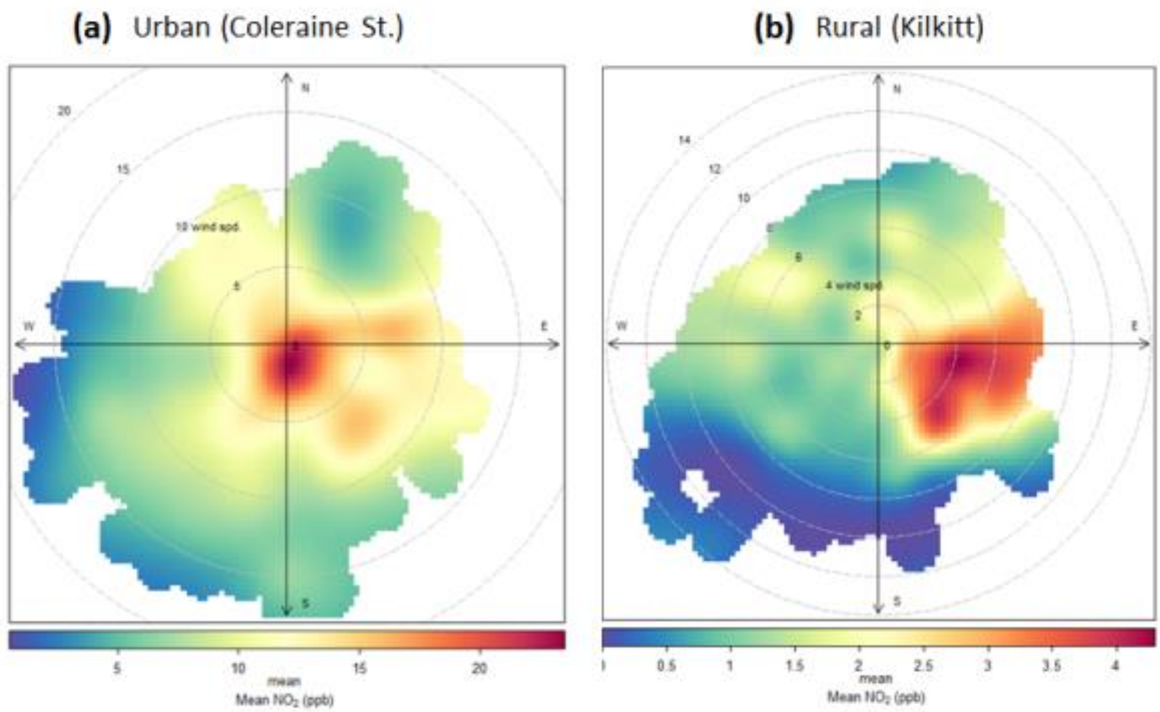


Figure 12 Polar plots of NO_2 concentration at urban site at an urban site (Coleraine St, left) and a rural background site (Kilkitt, right)

As regional prevailing wind conditions can vary substantially and thus impact the applicability of ordinary LUR techniques on a national scale, a novel LUR methodology was devised which incorporates wind effects using angular sectors or “wedges”. The 360° wind field is discretised into a set of eight wind sectors; average pollutant concentrations and predictor variables are then calculated for each sector and used in the LUR process. The use of continuous monitoring data from the national network, rather than short-term passive monitoring, allows the calculation of average concentrations within each wind sector. However, as prevailing wind directions vary geographically, seasonally and diurnally and hence a biased sectoral average may be obtained in some instances. For example, if a wind direction was more frequent during winter than summer months the raw sector average would be excessively high. Consequently, a non-parametric regression correction method has been applied to remove diurnal and seasonal bias from the data prior to sector averaging. There are four key steps involved in the wind sector land use regression (WS-LUR) model development and mapping process:

1. Calculate annual average pollutant concentrations within each wind sector at each monitoring site using a combination of hourly meteorological inputs and continuous monitoring data.
2. Generate predictor variables from geospatial datasets for each directional sector within a GIS environment
3. Select predictor variable for LUR equation using a supervised stepwise approach
4. Calculate key predictor variables on a national scale at a fine resolution and weight using interpolated local wind frequency.

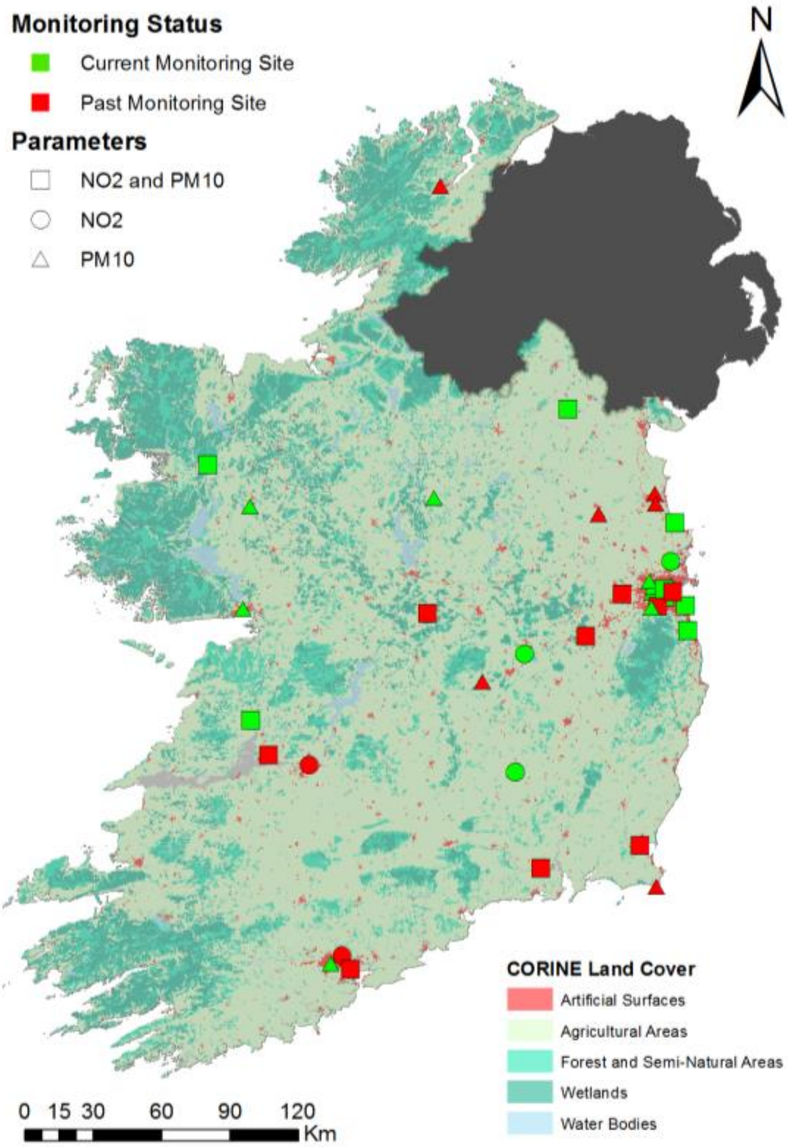


Figure 13 ***Monitoring site locations***

4.3 Correction factors

A correction method was developed to improve the representativeness of the monitoring data used in the sector based LUR and to account for uneven weighting of data from each season that may arise within different sectors. A brief overview of the correction method is provided here; a comprehensive explanation is provided in Donnelly et al. (2015).

Continuous monitoring data were used to calculate eight defined wind sector means for each station. The division of a concentration time series at a point into eight sectors maximises the number of data points available for the LUR; however, it also reduces data points available for long-term mean value calculation. Diurnal and seasonal concentration variations may lead to a biased annual sector average estimate when calculated from sub-annual datasets. Concentrations tend to be higher in winter months, than summer months (in Ireland) and, for example, if data within a sector was comprised 20% from winter and 80% from summer months, an unrealistically low value for the annual average would be obtained. Consequently, a short term correction factor (S_t) was applied to remove bias from the concentration data prior to sector averaging. Direct averaging of these data within each wind sector will still not necessarily result in reasonable predictions of the long run mean due to seasonal differences in wind direction frequencies and other external forcing factors such as variation in sunshine hours and stability conditions. L_t correction factors have been developed to apply to the data post binning. Figure 14 illustrates the procedure for correcting the raw data from a given monitoring site.

The removal of concentration fluctuations due to meteorological and seasonal factors allows the isolation of external forcing factors and thus improved quantification of spatial variability in concentration levels using spatial descriptors and subsequently a more robust LUR model. This is illustrated by an improvement in the correlation between pollutant concentrations and spatial emissions indicators as shown in Figure 15 for NO_2.

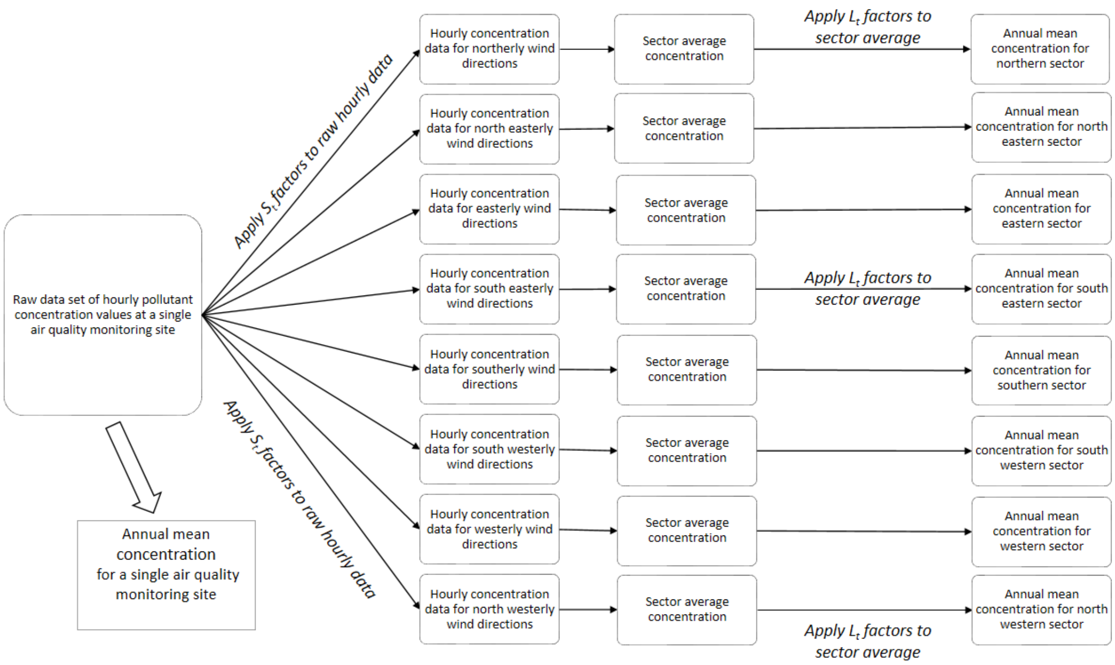


Figure 14 Application of correction factors flow chart

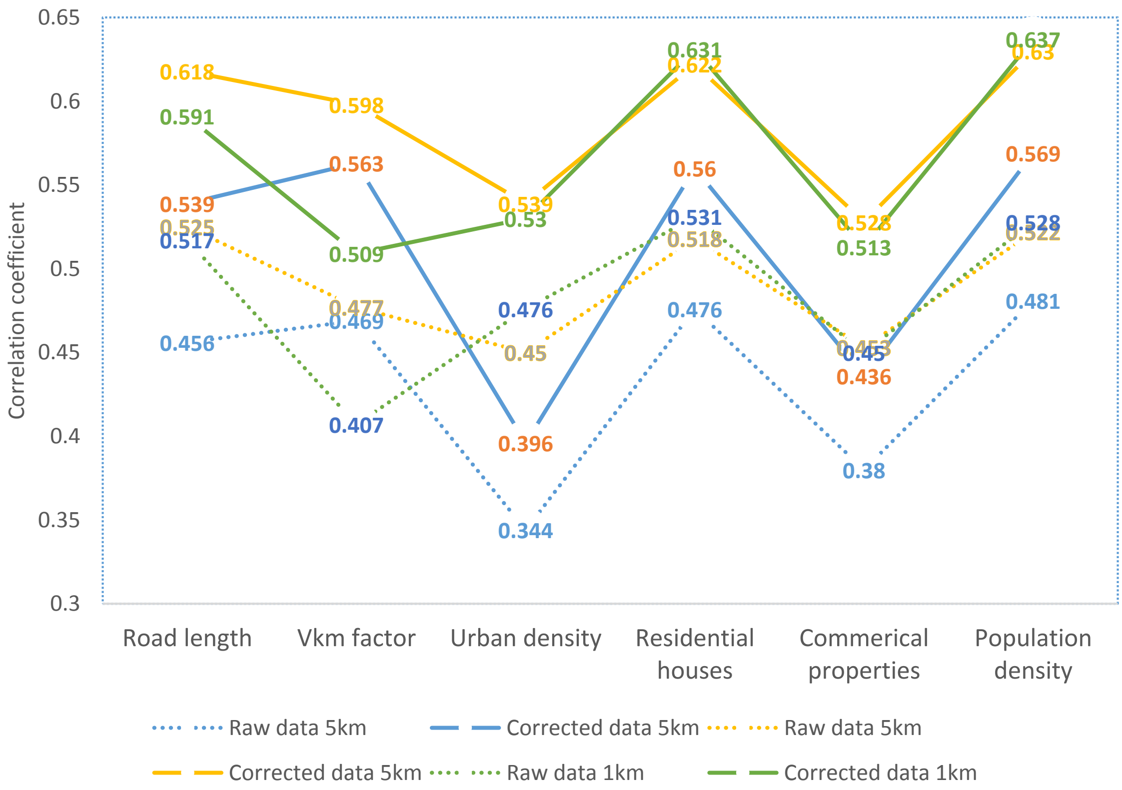


Figure 15 Improvement in correlation with land use variables using raw (dotted lines) and corrected NO_2 data (solid lines)

4.4 Predictor data

Geospatial predictor variables (Table 9) were calculated within each sector from nationally available, spatially homogeneous datasets for all sites using the ArcGIS 10.0 software package (ESRI, 2011). Eight circular buffers of variable radii were defined around each monitoring site, ranging from 25 metres to 5 kilometres, and further subdivided into eight 45° wind direction sectors (e.g. N, NE, E, etc.) (Figure 16 (a)). Residential and commercial property variables were derived using geographical coordinates from GeoDirectory (Figure 16(b)). Traffic network and flow data were obtained from the National Traffic Model (NTM), part of the National Transport Model (NTpM) developed by the National Roads Authority (NRA) (Figure 16(c)). Road length variables were calculated within each sector for each road category, and the length of each major road link passing through the sector was multiplied by the link AADT to give annual Vehicle km (Vkm). Due to the high correlation between NO_2 concentration and traffic parameters across the range of buffer radii, a weighted Vkm ($Vkm_{weighted}$) parameter was developed. The weighting applied to each sector is related to the inverse of the distance of the sector from the monitoring point, with the closest sector (i.e. 25m) carries the highest weighting. The inverse-distance weighted Vkm factor was calculated as:

$$Vkm_{invw} = \frac{1}{r_0} \times Vkm_0 + \sum_{i=1}^{N} \frac{1}{r_i} \times (Vkm_i - Vkm_{i-1})$$

Where $i = 0$ to $i = N$ represent each of the sectors considered, r is the distance from the monitoring point to the centre of a given sector, where Vkm_i is the sum of the Vkm in a given sector, i. In this case 8 sector sizes are considered, 25m, 50m, 100m, 250m, 500m, 1km, 2km and 5km.

Population and residential combustion data were derived from Census data and spatially disaggregated on the basis of residential property locations, whereby average household statistics were calculated within each Census Small Area (SA) using the total number of occupied residential properties within the SA. In each instance predictor variables were calculated by summing totals (e.g. total road length, total population etc) within each sector.

Land cover variables were derived from CORINE (COoRdination of INformation on the Environment) land cover data for the year 2006 the European Environment Agency (EEA). Following the methods outlined in Vienneau et al. (Vienneau et al., 2010) and Beelen et al. (Beelen et al., 2013b) the 44 land cover classes in CORINE were regrouped into six (High density residential, low density residential, industry, port, urban green, and semi-natural and forested areas) as well as additional land use class representing areas of sea and open ocean. Predictor variables were determined by calculating the area of each of these land use groups within each sector.

Large point source pollutant emissions were derived from the Pollutant Release and Transfer Register (PRTR), operated by the EPA. Point emission totals were assigned to each sector based on PRTR point locations and the annual emissions during the year (or years) for which monitoring data were available in the sector. The list of predictor variables and sector radii is provided in Table 9.

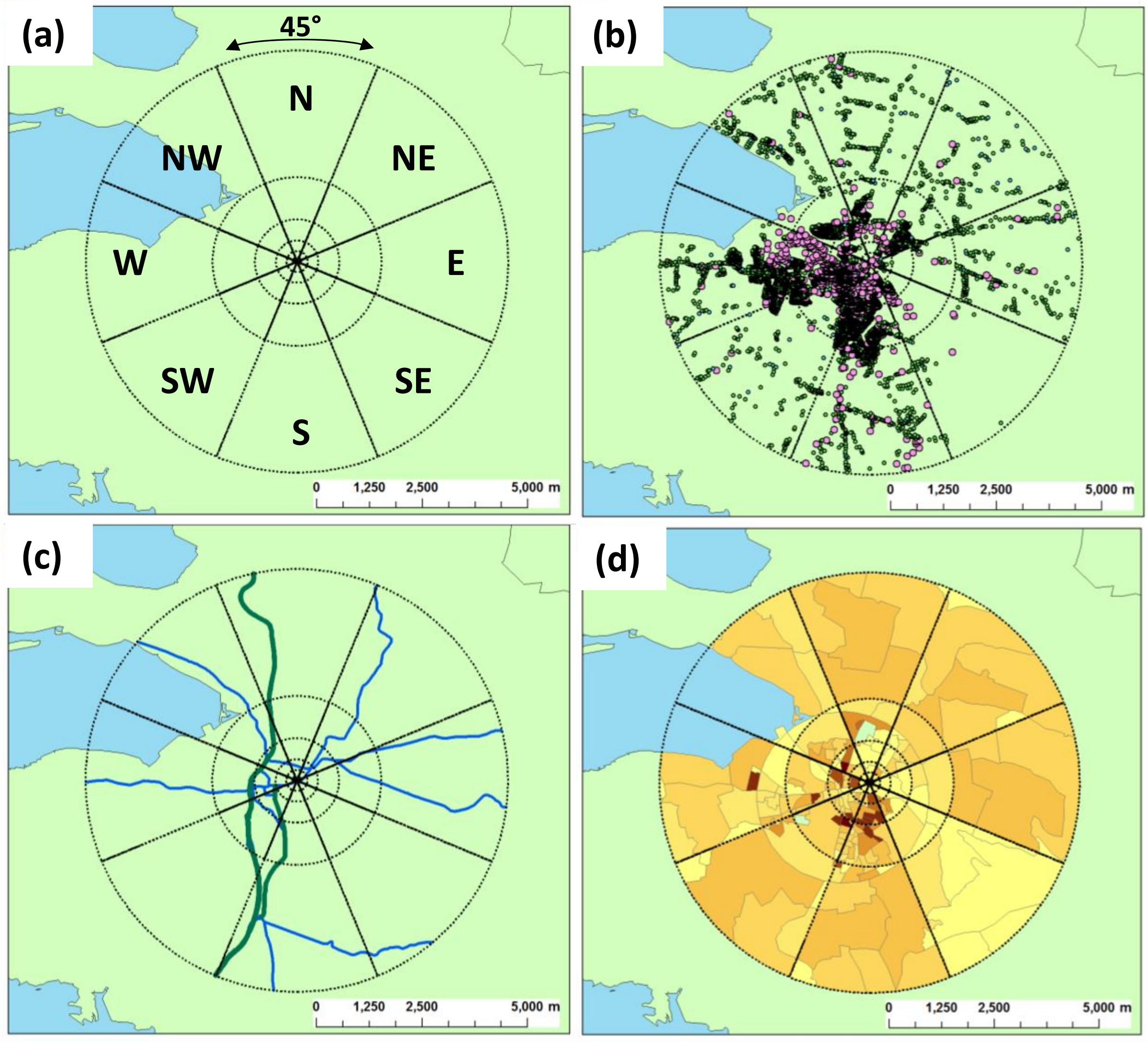


Figure 16***: (a) Wind direction sectors, (b) residential and commercial properties, (c) major roads and (d) solid fuel combustion***

Table 9*: Predictor variables with variable names, units, and sector size*

Category	*Units*	*Sector Size (m)*	*Subcategory*	*No. of Variables*
Road Length	Km	25, 50, 100, 250, 500, 1000, 2000, 5000	All roads National road Regional road Local road Major road	56
Proximity to Road	Km^{-1}, Km^{-2}	N/A	Nearest road Nearest major road	8
Traffic Flow	Vehicle Km	25, 50, 100, 250, 500, 1000, 2000, 5000	N/A	8
Weighted Traffic Flow	Vehicle Km	N/A	Inverse distance Gaussian	2
Land Cover	Hectares	25, 50, 100, 250, 500, 1000, 2000, 5000	High density residential Low density residential Industry Port Urban green Semi-natural and forested Natural Sea/Ocean	64
Population Density	$Persons/km^2$	25, 50, 100, 250, 500, 1000, 2000, 5000	N/A	8
Property Density	No. properties	25, 50, 100, 250, 500, 1000, 2000, 5000	Residential Commercial	16
Residential Heating	Properties per heating type	25, 50, 100, 250, 500, 1000, 2000, 5000	Solid Gas Electricity Oil	32
Household Cars	Cars	25, 50, 100, 250, 500, 1000, 2000, 5000	N/A	8
Proximity to Coast	Km	N/A	N/A	1
Point Source (PRTR)	Kg	25, 50, 100, 250, 500, 1000, 2000, 5000	N/A	8
Elevation	m	N/A	N/A	1
Wind Speed	m/s	N/A	N/A	1

4.5 Model fitting

Selection of the most appropriate explanatory variables within suitable sector sizes is important for defining final model performance. Variable selection was carried out using a supervised stepwise approach. Firstly each predictor variable was assigned a plausible direction of effect and univariate regression analyses were carried out for all predictor variables. The model with the highest adjusted R^2 having an appropriate slope as predefined by the direction of effect was considered as the start model. Additional predictor variables are then added consecutively to the model and maintained if the following three conditions are met:

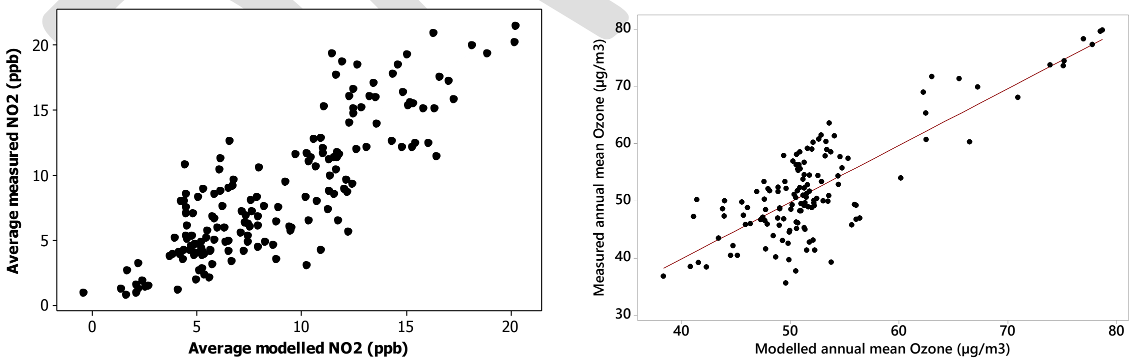
1. The R^2 value increases by at least 1%
2. The direction of effect of the new variable is as a priori defined
3. The direction of effect of previously included variables does not change

The large number of predictor variables examined meant that many of them were correlated. The variance inflation factor (VIF) was used to assess how much the variance of an estimated regression coefficient increases if predictors are correlated; it is equal to 1 if no factors are correlated. Variables with high VIF were removed from the model ensuring that each variable removed is redundant in the explanation of concentration. The set of predictor variables giving the highest adjusted R^2 value which conformed to a priori defined directions of effect were selected for inclusion in the final model. As a final step, variables with a p-value of greater than 0.05 were removed from the model.

Standard diagnostic tests for ordinary least squares regression were carried out. These included assessing residuals for heteroscedasticity and normality. Residuals were also analysed for influential or controlling observations or outlying data points. In a small number of instances this led to removal of certain data points after detailed investigations of the baseline data. Iterations cease and the final model is defined when residual diagnostics prove satisfactory.

4.6 Modelled versus measured values

A leave one out process was used to assess the annual mean maps. Scatter plots of measured versus modelled (Leave one out) values are shown in Figure 17. Results were strongest for NO_2. This is due to a combination of a well-represented monitoring network and the good description of NO_2 spatial variation by traffic related spatial variables.



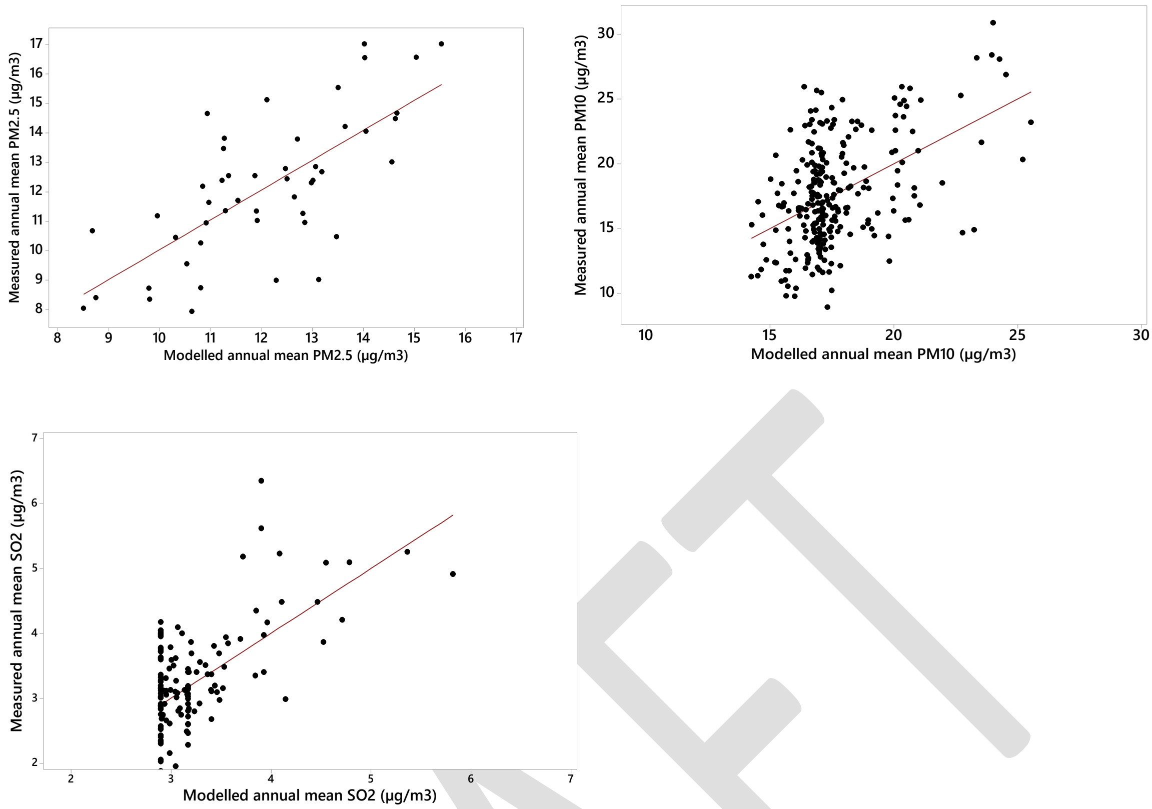


Figure 17 Annual mean LUR modelled versus measured concentrations for NO_2, Ozone, PM_{10}, $PM_{2.5}$ and SO_2

4.7 Results and applications

The output from this modelling work is a set of annual mean maps for each of NO_2, PM_{10}, $PM_{2.5}$, Ozone and SO_2. These maps are shown in Figure 18 to Figure 21. The NO_2 maps shows the dominant influence of traffic emissions on national (and urban) NO_2 concentrations. PM_{10} shows increases near coastal regions and also in regional towns due to the effects of solid fuel burning. $PM_{2.5}$ increases near major roads due to fine particulate emissions. As expected, ozone shows increases near coastal regions and decreases in heavily traffic areas where there are elevated NO emissions. The SO_2 map was limited by the number of monitoring stations available within each air quality zone. However, the clear influence of the coal ban zones can be observed in the final map.

These annual mean maps can be used for a variety of purposes:

- Direct analysis of air quality anywhere in Ireland
- For Assistance in determining appropriate areas to locate future air quality monitors (minimise monitor placement bias)
- Personal exposure studies.

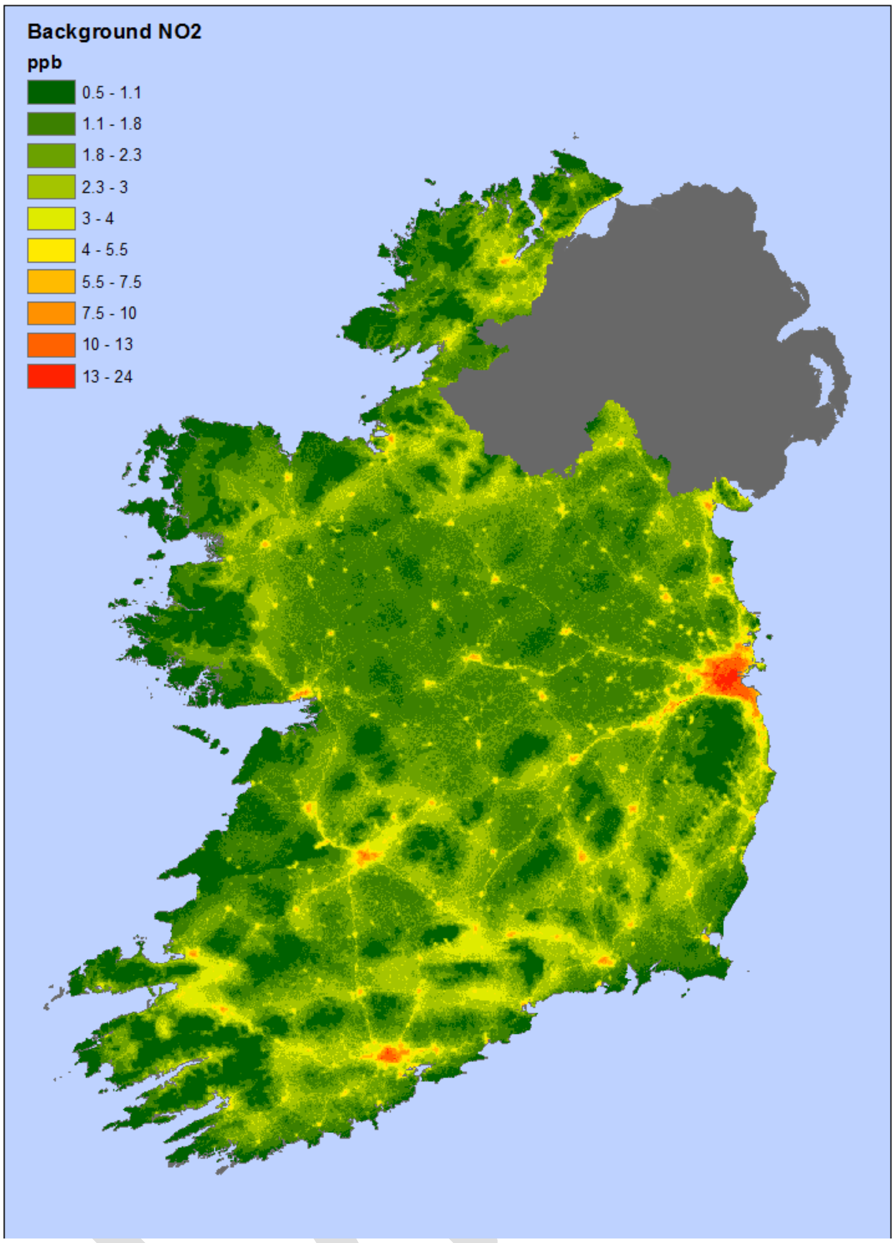


Figure 18 NO_2 map (National)

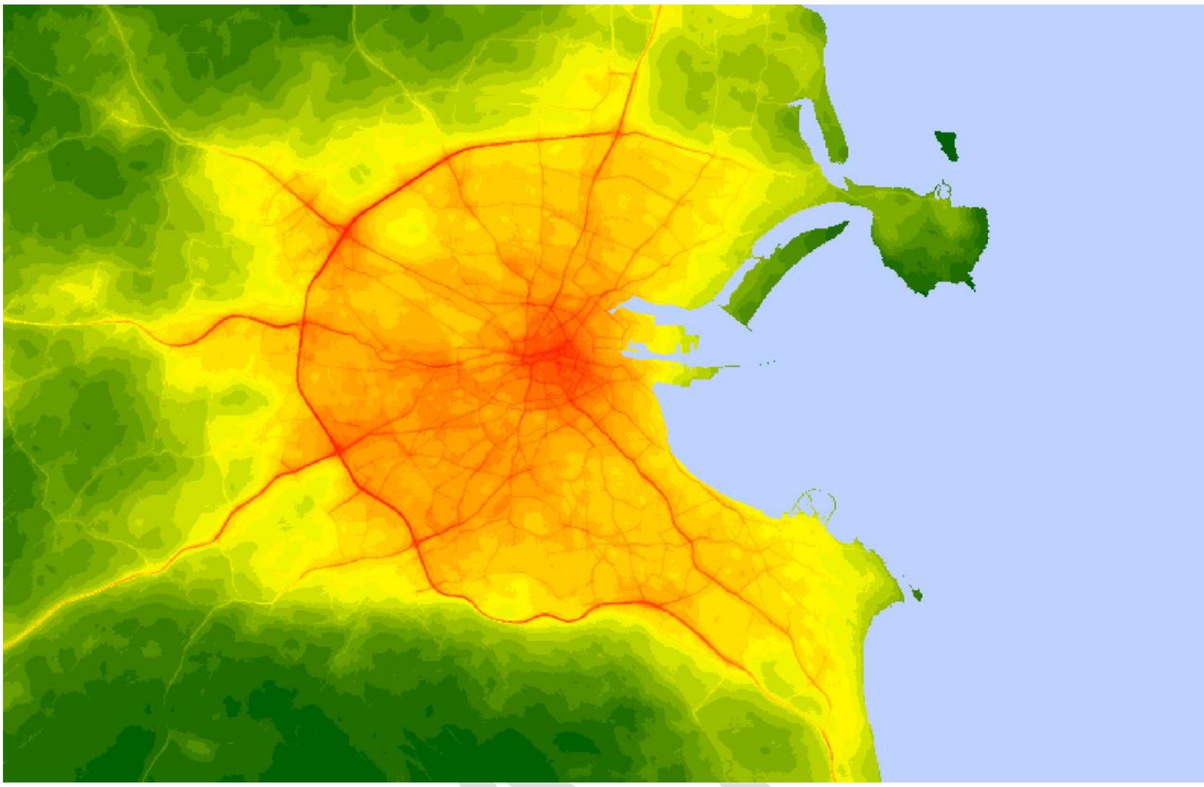


Figure 19 NO_2 map (Dublin)

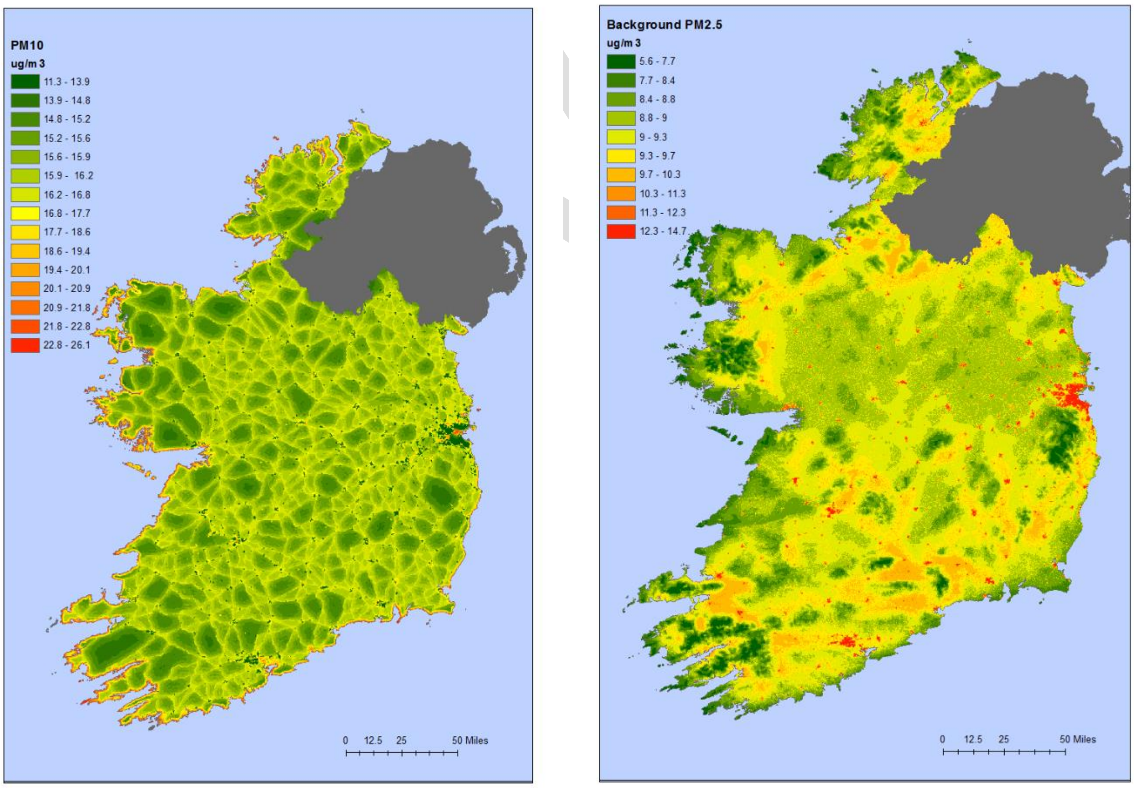


Figure 20 PM_{10} and $PM_{2.5}$ maps

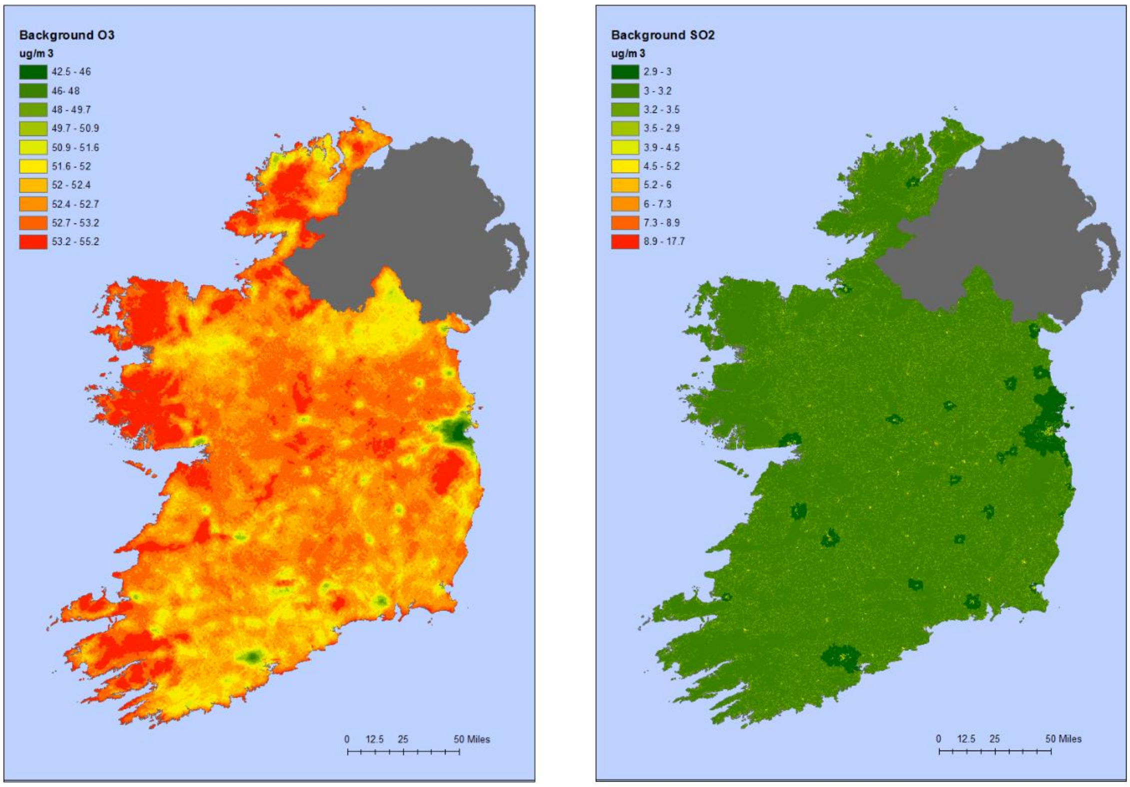


Figure 21 Ozone and SO_2 maps

5 Hourly LUR modelling

5.1 Overview of methodology

LUR models generally aim to explain spatial variation in concentrations and do not include a temporal aspect. Some studies have, however, attempted to model both temporal and spatial variation using a LUR base. Mölter et al. (2010) modelled annual concentrations of PM_{10} and NO_2 for Manchester between 1996 and 2008 using a LUR model from 2005. This model was temporally recalibrated and also made use of some temporal values of predictor variables and temporal trends. Dons et al. (2013) tested two methods of incorporating a temporal resolution into their model of black carbon in Flanders, Belgium. In the first approach they used 48 dummy variables for weekday and weekend hours (R^2 of 0.44) and in the second approach developed independent hourly models (R^2 between 0.07 and 0.8). Chen et al. (2010) included a temporal aspect to their LUR model of NO_2 and PM_{10} in Tianjin region in China by establishing four separate models, one for the heating season and one for the non-heating season for each pollutant. R^2 values ranged between 0.49 for PM_{10} in the non-heating season and 0.4 for NO_2 in the hearing season.

Saraswat et al. (2013) developed spatiotemporal models for $PM_{2.5}$ and black carbon (BC) in New Delhi by using a combined spatial monitoring campaign and data from a fixed continuous monitoring site. LUR data was sampled one site at a time and at each site measurements were collected for 1-3 hours during each time period (separate models were developed for morning and afternoon hours). The pollutant concentration was assumed to be associated with a multiplicative combination of a background temporal component (fixed site) and the spatial components. They assumed that the temporal component was spatially invariant and the spatial components were temporally invariant. $PM_{2.5}$ model fits of 85% and 73% were obtained for the morning and afternoon models, respectively. Su et al. (2008) developed a source area LUR for predicting hourly NO_2 concentrations in Vancouver from land use types and hourly wind speed, wind direction and cloud cover. They interpolated hourly meteorological data from 19 regulatory continuous monitoring stations for 116 passive samplers to create a source area LUR model. They compared these results to a source area LUR created from the 19 continuous monitoring stations and those from a regular LUR. Estimated concentrations for the hourly model were aggregated back to seasonal averages. They concluded that when variability in seasonal concentrations is present the source area LUR provides stronger results than the regular LUR.

In the current research project a novel model for forecasting spatially resolved hourly or daily concentrations and also tackles two of the main limitations of LUR. Firstly the issue of wind direction and area of influence when using circular buffer zones is addressed through the use of "sectors" within which predictor variables are defined and calculated (as introduced in section ????? for the annual mean LUR). In operational model the appropriate "sector" will vary with local wind direction. Secondly, the temporal resolution of the LUR is greatly improved through the inclusion of hourly meteorological data and seasonal factors as predictor variables.

Using the methods developed in this research project hourly concentrations can be mapped on a national scale and forecasts of daily average and daily maximum concentrations can be made across the country with minimal computational requirements.

5.2 Model Development

5.2.1 General model fitting

The WS-LUR model uses the same spatial predictor variables (where significant) as presented in Section 4 and Table 9. In addition the following temporally varying predictor variables are used:

- $WSWD_f$ (Wind speed/direction factor as introduced in Section 3)
- S_f (Seasonal factor as introduced in Section 3)
- Weekday/weekend dummy variable
- Hourly temperature
- Hourly precipitation
- Hourly atmospheric pressure
- Hourly relative humidity
- Sunshine hours
- Hourly stability class

The premise of the WS-LUR is that a separate prediction equations are developed for different environmental conditions and/or time periods. A first step is to identify appropriate methods of partitioning the data so that robust models can be developed. The grouping allows a unique prediction of air quality to be made at any location in Ireland for any hour where the appropriate meteorological data are available. Model development proceeded following these steps:

1. Generate an hourly data set of measured concentrations at air quality monitoring sites and associated hourly meteorological factors for two full calendar years (2011 and 2012)
2. Assign a weekday/weekend dummy variable and S_f to each data point
3. Generate predictor variables from geospatial datasets for each directional sector within a GIS environment
4. Assign relevant spatial predictors to each hourly data point at each monitoring site
5. Divide the data set into pre-identified environmental groups
6. Select appropriate spatial and temporal predictor variables for each dataset using a supervised stepwise approach
7. Merge to form a single model for all hours/seasons
8. Complete mapping process by calculating key predictor variables on a national scale at a fine spatial resolution and hourly temporal resolution for the validation year (2012)
9. Validate results by comparing to measured daily average and daily maximum concentrations for the same time period.

5.2.2 NO_2

NO_2 exhibits strong seasonal and diurnal variations the magnitude of which vary significantly on a national scale. Therefore 48 separate regression equations were developed (one for each hour in each season) to feed into the WS-LUR model.

5.2.3 $PM_{10/2.5}$

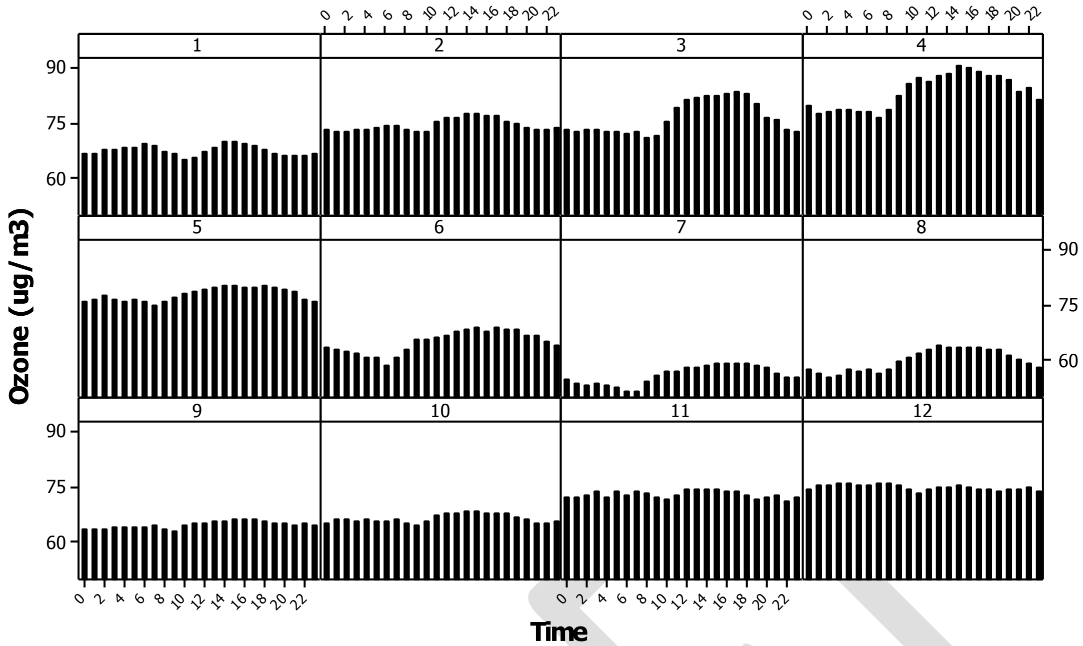
$PM_{10/2.5}$ concentrations exhibit nonlinear changes in concentration with wind speed and temperature. Therefore this method divides the training data into 4 separate temperature classes and four temperature wind speed classes. A separate regression equation is trained for each of these meaning a total of 16 regression equations. In operational mode, the appropriate regression equation can be chosen based on the daily average wind speed and temperature that is forecast on a given day. The classes were chosen based on the first quartile, median and 3rd quartile of the total temperature and wind speed data sets. The bands are shown in Table 10.

Table 10 Variable classes for PM_{10} and $PM_{2.5}$

Variable class	*Temperature (°C)*	*Wind speed (m/s)*
1	<6.35	<2.87
2	<9.76	<4.18
3	<13.15	<5.81
4	>=13.15	>=5.81

5.2.4 Ozone

In developing the WS-LUR for ozone the data were separated into three (spatially distinct) groups. They are a coastal group, a rural/suburban group and a group comprising of the Dublin and Cork air quality zones. The data were first grouped seasonally based on similarities between monthly values. They were then grouped for regression based on diurnal variations within each of the identified seasons and the three spatial location types. Figure 22 shows the diurnal variations at the coastal sites of Valentina and Mace Head split by month.



Panel variable: Month

Figure 22 Diurnal variation in Ozone at Mace Head and Valentia split by month

5.3 Results

The model was fitted for NO_2, Ozone and PM_{10} as discussed above. Hourly (or daily) modelled values were compared to monitored data for each of the pollutants. Standard statistical measures were used to assess model performance. The results are shown in Table 11. Scatter plots of measured versus modelled values are shown in Figure 23, Figure 24 and Figure 25. Results confirmed that the WS-LUR model is a useful and efficient means of forecasting air quality on a national scale in Ireland. There is a fair degree of scatter in the PM_{10} plot and the detailed analysis of PM carried out suggested that future spatio-temporal modelling of PM might be carried out using an interpolation method on the hybrid point wise forecasts in conjunction with the annual mean LUR maps.

Figure 26 shows a sample output of the Dublin region for two different times of the day on the 10th of August 2014. A clear difference can be observed between the midday and evening concentrations. This is the effect of rush hour traffic emissions. Clear definition can be observed around the road network, in particular the M50 and arterial routes highlighting the large contribution of traffic emission to overall NO_2 concentrations.

Table 11 Statistical performance measures for the WS-LUR model

	NO_2 (Daily average)	NO_2 (Daily maximum)	Ozone (Daily average 8 hour value)	Ozone (Daily maximum 8 hour value)	PM_{10} (Daily average)
FAC2	95.69%	93.20%	98%	98%	94%
R	0.84	0.77	0.664	0.665	0.60
Mean fractional bias	-1%	-1%			-5%
Index of agreement	0.91	0.86	0.787	0.794	0.73

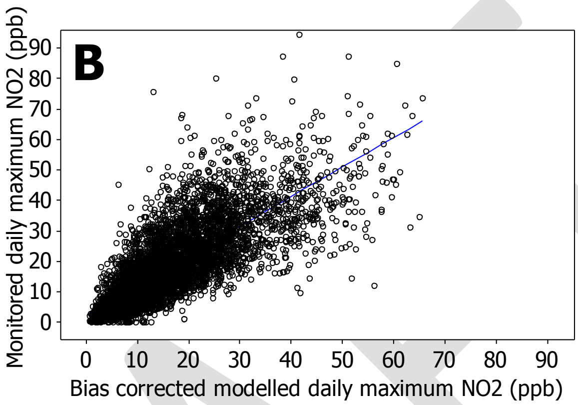


Figure 23 WS-LUR modelled versus measured NO_2 data

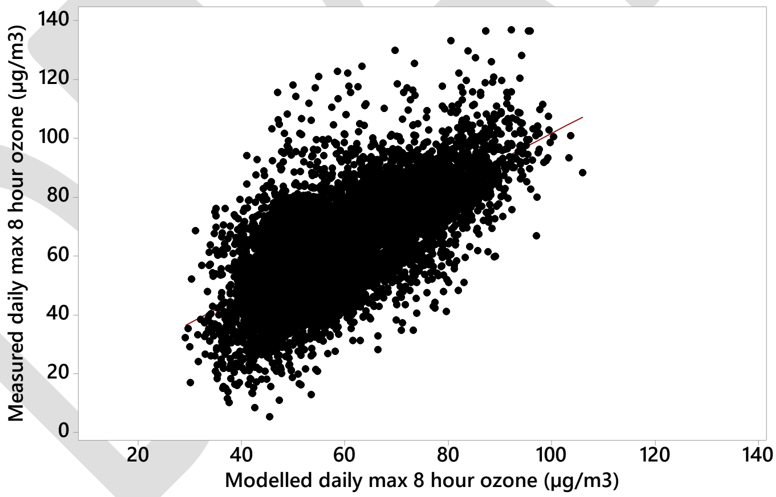


Figure 24 WS-LUR modelled versus measured Ozone data

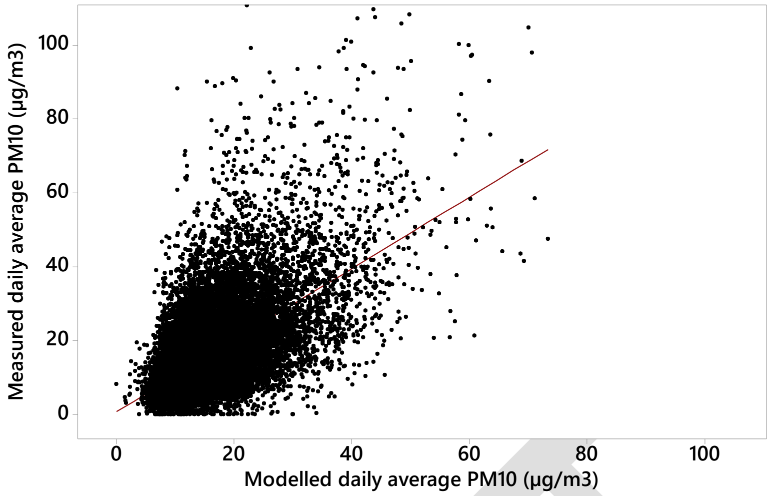


Figure 25 WS-LUR modelled versus measured PM_{10} data

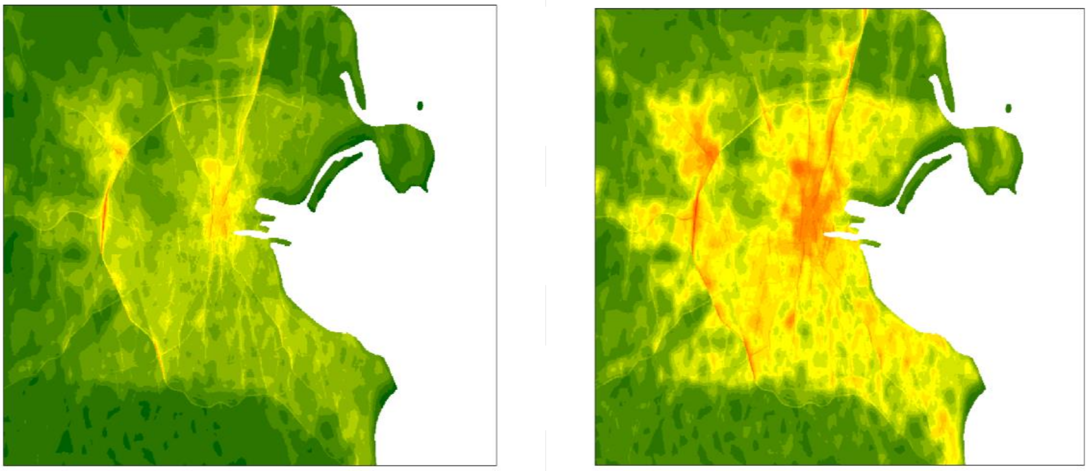


Figure 26 Sample output from the WS-LUR model for NO2 Dublin on the 10th August 2014 at 1pm (A) and 7pm (B)

6 Urban air quality survey

An urban monitoring campaign was carried out to provide additional information on the spatial variation of concentrations across urban and rural areas. This task was a joint work package between the air quality modelling fellow and the emissions inventory fellow (2013-EH-FS-7). The objective of the work is to develop linkages between air pollution levels at a fine spatial scale in Dublin and other spatial parameters.

70 NO_2/SO_2 diffusion tubes and 59 Ozone diffusion tubes were deployed at pre-specified locations around Dublin. The sites were chosen using the annual mean maps detailed in Section 4. It was necessary that the locations cover primary land use classes as well as the full range in concentrations over the Dublin region. Guidelines laid out in the ESCAPE study manual (European Study of Cohorts for Air Pollution Effects, 2008) were also followed in choosing site locations. This manual details site selection, site characterization,

temporal aspects, LUR model development and potential predictor variables in exposure assessment studies. The locations of the monitoring sites are shown in Figure 27.

The diffusion tubes were all deployed within 24 hours of each other and the GPS coordinates and time of deployment recorded. They were each left out for a period of 2 weeks between the 10/06/2015 and the 24/06/2015. The time of collection of each tube was recorded and any missing or damaged tubes were noted. The tubes were placed in sealed plastic bags and returned to the laboratory for analysis along with a number of blank tubes for corrective purposes. Final concentrations of NO_2, SO_2 and ozone at each location are provided as a dataset output from this project.

The diffusion tube survey results provide a valuable resource regarding spatial variation in concentration levels in the Dublin region. A second round of sampling will be carried out as part of Emission Inventory fellowship (2013-EH-FS-7) to capture additional information on the seasonal variation in concentration levels. Thereafter, finely resolved spatial data provided will be used together with the results from the monitoring campaign to carry out geo-statistical modelling of the Dublin area as part of the Emission Inventory fellowship. A model framework has been developed as part of the current fellowship which will upon which urban modelling will be based. This modelling will be carried out using the in-house statistical model developed as part of this fellowship and short term average values obtained will be converted to annual mean concentrations based on seasonal factors developed during the first two years of the fellowship. The methodology will be documented and a framework will be developed for model building and meteorological forecast integration.

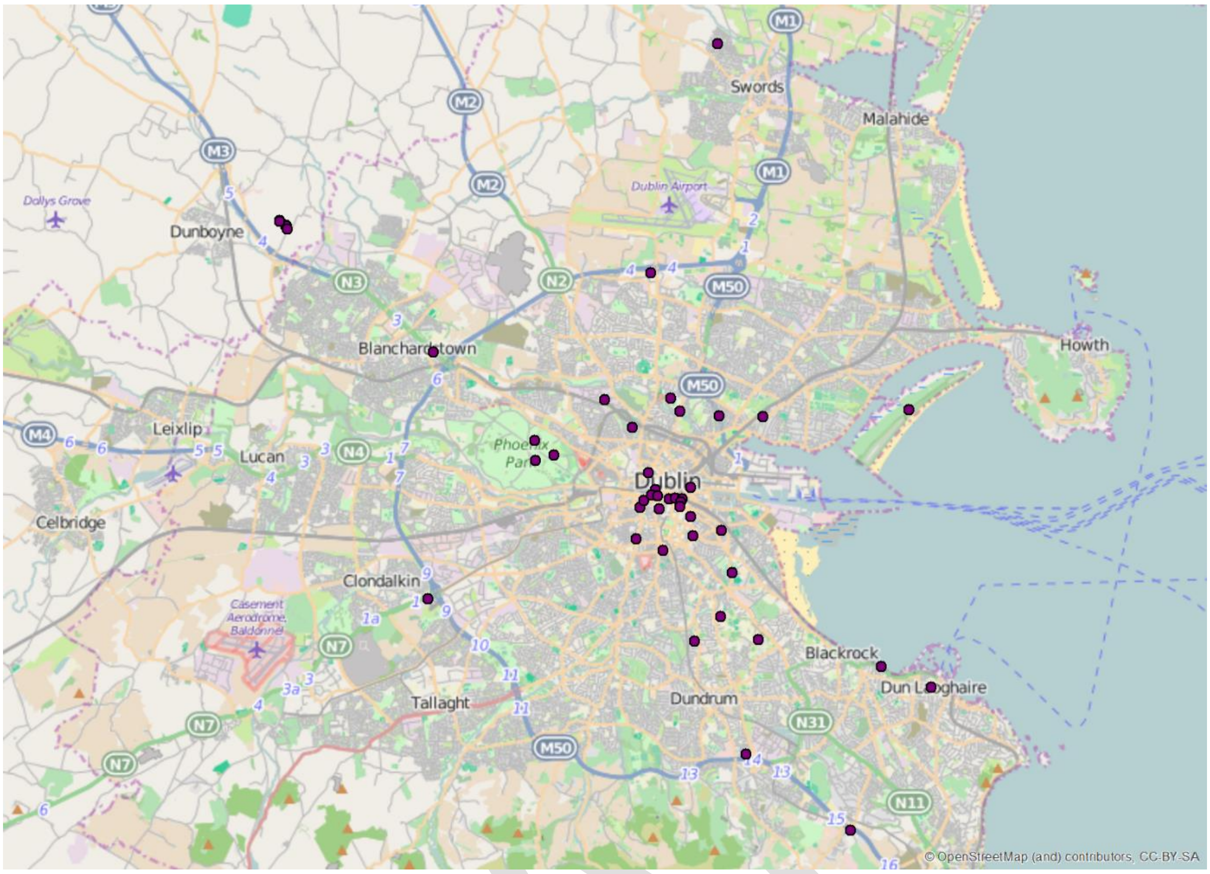


Figure 27 Diffusion tube monitoring site locations

7 Conclusions

A suite of air quality models has been developed as part of his research project to achieve key EPA objectives. The models provide the following information and operational capability:

- Automated twice daily 48 hour pointwise forecasts of NO_2, SO_2, $PM_{10/2.5}$, Ozone and he AQIH
- Automated twice daily forecasts of the origin of air reaching Ireland (using HYsplit)
- Annual mean maps of NO_2, SO_2, $PM_{10/2.5}$, Ozone
- National scale spatial model to predict hourly NO_2, 8 hourly average ozone and daily average PM_{10}

Air quality model development has necessarily been a stepwise process making maximum use of available resources. A statistical approach to provide point wise forecasts was adopted which used historical monitoring data to train the model in the absence of a detailed emissions inventory. Inclusion of regional transport of emissions improved model efficiency. Air mass history modelling was carried out and a HYsplit add-on was developed for the model. A validation study was carried out by comparing 12 months of modelled data to monitored data from the same period. Some general conclusions from this study were:

- Model validation statistics show good correlation between measured and modelled values and indicate a level of performance equal to or better than that generally expected from air quality models.

- Overall IA values of 0.88, 0.84, 0.84, 0.80 and 0.88 are achieved for NO_2, PM_{10}, $PM_{2.5}$, SO_2 and ozone, respectively.
- Overall r values of 0.82, 0.72, 0.74, 0.69 and 0.82 are achieved for NO_2, PM_{10}, $PM_{2.5}$, SO_2 and ozone, respectively.
- Overall FAC2 values of 73%, 93%, 79%, 77% and 100% are achieved for NO_2, PM_{10}, $PM_{2.5}$, SO_2 and ozone, respectively.
- Incorporation of an air mass history parameter resulted in a large improvement in the prediction of NO_2 and particulate matter concentrations.
- The model is quite conservative in its PM_{10} and $PM_{2.5}$ predictions. There is a slight positive bias at all sites as a result of the methodology used to account for regional air mass movement and pollutant transport. PM is one of the most difficult pollutants to model due to its wide range of anthropogenic and natural sources. Therefore, a conservative estimate accompanied by some specialist interpretation is considered to be the best means of producing a forecast. When the AQIH is forecast to be poor, air mass history and other conditions relating to PM concentrations should be examined in conjunction with the value given by the AQ model to produce a final forecast.
- During the next model calibration/training, ozone should be trained using hourly data. The use of 8 hour averages as the response variable has resulted in some over smoothing of the data.

The stepwise approach adopted in model development allowed outputs before completion of the final study and achieved a key EPA objective of producing forecasts of the Air Quality Index for Health 24 and 48 hours in advance.

Mid-way through the original research fellowship the EPA funded an additional fellowship concerned with development of an emissions inventory for Ireland. This research was closely linked with the air quality modelling fellowship and influenced the direction of the work. Air quality modelling results were required to feed into the emissions inventory development to ensure that the most appropriate surrogate data are used, while the emissions inventory work provided spatial datasets for the development of the national annual mean maps.

Using air quality data from the national ambient air quality monitoring network and spatial predictor data a LUR technique was used to model air quality on a national scale. A novel technique was employed whereby the air quality data were spilt into 8 wind dependent sectors and corrected for short term fluctuations. Spatial predictor variables were also defined using the same sector based approach. This had the effect of maximising the number of data points available for the regression while also improving the description of spatial emissions/air quality relationships. The outputs from this modelling work were annual mean maps of NO_2, PM_{10}, $PM_{2.5}$, SO_2 and ozone for the base year of 2012. A temporal aspect was introduced to the above model by including short term meteorological predictor variables and seasonal and diurnal factors in addition to spatial variables. The resulting model has the ability to forecast hourly NO_2, 8 hourly average ozone and daily average PM_{10} at any location in Ireland.

This work highlighted some bias in the monitoring network, particularly in the case of ozone and $PM_{10//2.5}$. A significant coastal influence was observed on PM_{10} but a lack of sufficient monitoring data meant that this could not be fully quantified. However, an approximation was developed and this is an area recommended for further work. Validation of the model using a “leave one out” procedure showed that the model performs excellently for NO_2 and is of a suitably high standard for $PM_{10/2.5}$, SO_2 and ozone to be used for studies of spatial variation in concentrations across Ireland. Insufficient detail in background concentration data is frequently cited as the reason for poor results in local and urban modelling studies. The national scale model outputs from the present project have high relevance as inputs (background concentrations) into more detailed urban modelling studies.

A detailed study was carried out into incidences of high PM_{10} and $PM_{2.5}$ across Ireland. The following events (together) are likely to lead to high PM and should be associated with a PM alert system:

- Low wind speed (<3m/s)
- Low temperature (<6 degrees)
- High pressure (>1020mbar)
- Shallow boundary layer (<500m)
- Stable conditions
- Low/no precipitation.

This fellowship has produced a number of key tangible outputs as detailed in section 8. The suite of models developed should form the building blocks for future modelling work, which is necessarily an iterative process. While a direct output from this work has been a fully automated air quality forecast model it should be noted that user knowledge and interpretation of model outputs are important in all air quality modelling work and the importance of developing a knowledge base in this area should not be underestimated. It is recommended that air quality forecasts should be made using a combination of:

- Numerical output directly from the model
- Assessment of forecast local meteorological conditions
- Assessment of regional air mass movements as forecast by the HYsplit model which is built into the operational air quality model
- Consideration of any other unusual events or conditions (e.g. volcanoes or Saharan dust episodes)
- Expert judgement.

8 Recommendations for future work

Recommendations for future work concerning model maintenance and development are as follows:

- The point wise model should be recalibrated using up to date validated air quality and meteorological data on an annual basis. This process will ensure that air quality trends at individual sites are well captured and any new sources in an area are identified and included in the model.
- In the case where new air quality monitoring sites are used for the derivation of the AQIH, the model can be used to continue to forecast at the old site until a full year of data are available at the new site. This will avoid any break in forecasts within any one air quality zone. Once at least a full year of data are available, the model should be recalibrated and updated to include this new site within the model architecture.
- The national annual mean maps and temporal LUR models should be recalibrated every two years (or when new and significant spatial data become available).

- Recalibration of the national annual mean maps and temporal LUR models should also be carried out in the case of significant changes in spatial characteristics within a given zone or region.

The work carried out as part of this fellowship highlighted a number of areas which require, or would benefit from, further research. These are as follows:

- Further work is required to ensure that the national ambient air quality monitoring network has sufficient spatial coverage across the range of pollutant concentrations. Outputs from the current fellowship can be used to assist in determining spatial coverage. Using the national scale maps, the total area within each AQ zone is calculated. The concentrations within each zone as indicated by the model are plotted as cumulative distribution plots. This is shown by the 4 curves in Figure . The concentrations at each of the AQ monitoring sites in the national network are then overlaid as points on these curves. Ideally the monitoring sites should cover the full range of concentrations shown by the distributions for each zone. These distribution curves show a clear lack of monitoring stations at the upper levels of the curve (trafficked sites in the case of NO_2) in zone B and C. Since the national scale model has been developed using the current AQ monitoring network, an iterative process would be necessary whereby a new model is developed after the AQ review and the process repeated.

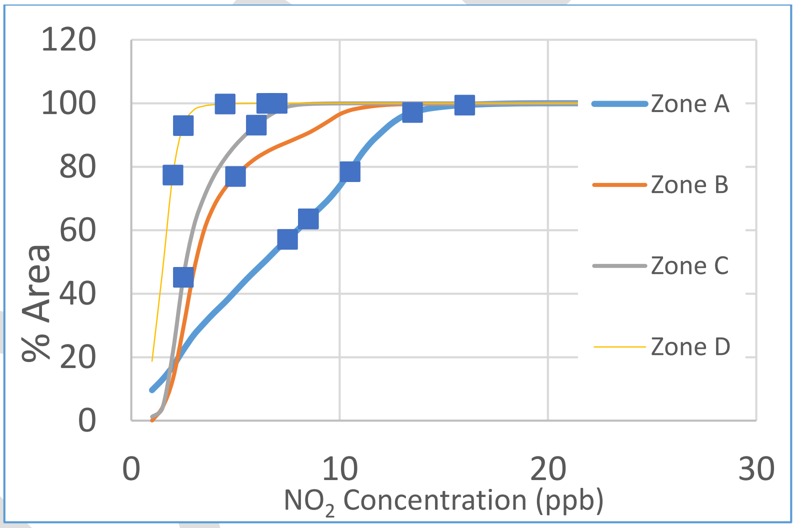


Figure 28 Cumulative distribution curves with concentrations at each monitoring site superimposed

- PM_{10} was found to display significant coastal influence. Bias in monitor placement in the national ambient network meant that an approximation had to be used in the present fellowship to quantify this. A research study should be carried out that quantifies $PM_{10/2.5}$ concentrations at set distances from the coast (up to 10km) during onshore, off shore and variable winds during different meteorological conditions and seasons. $PM_{10/2.5}$ coastal fall off curves should be developed for different meteorological classes.
- PM_{10} and $PM_{2.5}$ were found to display significant temporal variation due to both natural and anthropogenic effects. Further research is required which links source apportionment work with forecasting work. Quantifiable outputs from source apportionment could potentially be applied within a forecast environment to provide improved predictions of PM on a national scale.

- Ozone was found to be significantly higher in coastal regions but the small number of coastal sites made this difficult to quantify. Ozone monitoring should be carried out in coastal regions using either passive or active techniques to improve the quantification of high ozone events.
- The hybrid pointwise model developed as part of this fellowship was found to produce excellent 48 hour forecasts of the AQIH at pre-specified locations. A novel new area of work is the interpolation of these forecasts on a national scale. The number of monitoring sites is too limited to perform a direct interpolation; however, the integration of hybrid point wise model with the spatial model provides an innovative methodology for producing fast, resource efficient forecasts on a national scale at hourly resolution. Pointwise forecast would first be developed for every monitoring site in Ireland. The technique would involve background stripping the point wise forecasts in real time by subtracting the background concentration as provide by the annual mean maps from the concentration forecast at each location. These local forecasts are then interpolated using appropriate techniques. The background concentration can then be added back on nationally to provide real time national scale forecast maps. This model would be based in an GIS environment and could be fully automated.

References

ABBOTT, J. & STEDMAN, J. 1999. Dispersion modelling and mapping studies for review and assessment of PM 10. *A Report produced for the Department of the Environment, Transport and the Regions*.

ABBOTT, J. & VINCENT, K. 1999. Annual average sulphur dioxide concentration maps derived by dispersion modelling. Department of the Environment, Transport and the Regions.

ANASTASSPOULOS, A., NGUYEN, S. & XU, X. 2004. On the use of the HYSPLIT model to study air quality in Windsor, Ontario, Canada. *Environmental Informatics Archives* [Online], 2.

BEELEN, R., HOEK, G., PEBESMA, E., VIENNEAU, D., DE HOOGH, K. & BRIGGS, D. J. 2009. Mapping of background air pollution at a fine spatial scale across the European Union. *Science of the Total Environment,* 407**,** 1852-1867.

BEELEN, R., HOEK, G., VIENNEAU, D., EEFTENS, M., DIMAKOPOULOU, K., PEDELI, X., TSAI, M.-Y., KÜNZLI, N., SCHIKOWSKI, T. & MARCON, A. 2013a. Development of NO< sub> 2</sub> and NO< sub> x</sub> land use regression models for estimating air pollution exposure in 36 study areas in Europe–The ESCAPE project. *Atmospheric Environment,* 72**,** 10-23.

BEELEN, R., HOEK, G., VIENNEAU, D., EEFTENS, M., DIMAKOPOULOU, K., PEDELI, X., TSAI, M.-Y., KÜNZLI, N., SCHIKOWSKI, T., MARCON, A., ERIKSEN, K. T., RAASCHOU-NIELSEN, O., STEPHANOU, E., PATELAROU, E., LANKI, T., YLI-TUOMI, T., DECLERCQ, C., FALQ, G., STEMPFELET, M., BIRK, M., CYRYS, J., VON KLOT, S., NÁDOR, G., VARRÓ, M. J., DĖDELĖ, A., GRAŽULEVIČIENĖ, R., MÖLTER, A., LINDLEY, S., MADSEN, C., CESARONI, G., RANZI, A., BADALONI, C., HOFFMANN, B., NONNEMACHER, M., KRÄMER, U., KUHLBUSCH, T., CIRACH, M., DE NAZELLE, A., NIEUWENHUIJSEN, M., BELLANDER, T., KOREK, M., OLSSON, D., STRÖMGREN, M., DONS, E., JERRETT, M., FISCHER, P., WANG, M., BRUNEKREEF, B. & DE HOOGH, K. 2013b. Development of NO2 and NOx land use regression models for estimating air pollution exposure in 36 study areas in Europe – The ESCAPE project. *Atmospheric Environment,* 72**,** 10-23.

BORREGO, C., MONTEIRO, A., PAY, M., RIBEIRO, I., MIRANDA, A., BASART, S. & BALDASANO, J. 2011. How bias-correction can improve air quality forecasts over Portugal. *Atmospheric Environment*.

BRIGGS, D. J., COLLINS, S., ELLIOTT, P., FISCHER, P., KINGHAM, S., LEBRET, E., PRYL, K., VAN REEUWIJK, H., SMALLBONE, K. & VAN DER VEEN, A. 1997. Mapping urban air pollution using

GIS: a regression-based approach. *International Journal of Geographical Information Science,* 11**,** 699-718.
CEU 2008. DIRECTIVE 2008/50/EC OF THE EUROPEAN PARLIAMENT AND OF THE COUNCIL of 21 May 2008 on ambient air quality and cleaner air for Europe. *In:* COMMISSION, E. (ed.). Official Journal of the European Union.
CHEN, L., BAI, Z., KONG, S., HAN, B., YOU, Y., DING, X., DU, S. & LIU, A. 2010. A land use regression for predicting NO 2 and PM 10 concentrations in different seasons in Tianjin region, China. *Journal of Environmental Sciences,* 22**,** 1364-1373.
DEHLG 2002. Air Quality Standards Regulations 2002
DERWENT, D., FRASER, A., ABBOTT, J., JENKIN, M., WILLIS, P. & MURRELLS, T. 2010. Evaluating the performance of air quality models. *Report prepared for the UK Department for Environment, Food and Rural Affairs, Issue,* 3.
DONNELLY, A., BRODERICK, B. & MISSTEAR, B. 2015a. The effect of long range air mass transport pathways on PM10 and NO2 concentrations at urban and rural background sites in Ireland: Quantification using clustering techniques. *Journal of Environmental Science and Health Part A,* 50**,** 647-658.
DONNELLY, A., MISSTEAR, B. & BRODERICK, B. 2015b. Real time air quality forecasting using integrated parametric and non-parametric regression techniques. *Atmospheric Environment,* 103**,** 53-65.
DONNELLY, A., NAUGHTON, O., BRODERICK, B. & MISSTEAR, B. 2016. Short-Term Forecasting of Nitrogen Dioxide (NO2) Levels Using a Hybrid Statistical and Air Mass History Modelling Approach. *Environmental Modeling & Assessment***,** 1-11.
DONS, E., VAN POPPEL, M., KOCHAN, B., WETS, G. & PANIS, L. I. 2013. Modeling temporal and spatial variability of traffic-related air pollution: Hourly land use regression models for black carbon. *Atmospheric Environment,* 74**,** 237-246.
DRAXLER, R. & HESS, G. 1998. An overview of the HYSPLIT_4 modelling system for trajectories, dispersion, and deposition. *Australian Meteorological Magazine,* 47**,** 295-308.
DUBLIN CITY COUNCIL, SOUTH DUBLIN COUNTY COUNCIL, FINGAL COUNTY COUNCIL & DUN LAOGHAIRE-RATHDOWN COUNTRY COUNCIL Dublin Regional Air Quality Management Plan for improvements in levels of Nitrogen Dioxide in ambient air quality.
ENVIRONMENTAL PROTECTION AGENCY 2012. Irelands Environment - An assessment. Johnstown Castle.
ESRI 2011. ArcGIS Desktop: Release 10. Redlands, CA: Environmental Systems Research Institute.
EUROPEAN COMMISSION 2013. COMMUNICATION FROM THE COMMISSION TO THE EUROPEAN PARLIAMENT, THE COUNCIL, THE EUROPEAN ECONOMIC AND SOCIAL COMMITTEE AND THE COMMITTEE OF THE REGIONS. Brussels.
EUROPEAN ENVIRONMENT AGENCY (EEA) 2011. The application of models under the European Union's Air Quality Directive: A technical reference guide.
EUROPEAN STUDY OF COHORTS FOR AIR POLLUTION EFFECTS 2008. ESCAPE Study manual. Institute for Risk Assessment Sciences, Utrecht University, The Netherlands.
FENG, Y., ZHANG, W., SUN, D. & ZHANG, L. 2011. Ozone concentration forecast method based on genetic algorithm optimized back propagation neural networks and support vector machine data classification. *Atmospheric Environment,* 45**,** 1979-1985.
GENC, D. D., YESILYURT, C. & TUNCEL, G. 2010. Air pollution forecasting in Ankara, Turkey using air pollution index and its relation to assimilative capacity of the atmosphere. *Environmental monitoring and assessment,* 166**,** 11-27.
GREENLAND, D. & YORTY, R. A. 1985. The Spatial Distribution of Particulate Concentrations in the Denver Metropolitan Area. *Annals of the Association of American Geographers,* 75**,** 69-82.
HOI, K., YUEN, K. & MOK, K. 2010. Optimizing the performance of Kalman filter based statistical time-varying air quality models. *Global NEST J,* 12**,** 27-39.

KENT, A., STEDMAN, J., BOWER, J., MANNING, A., HARRISON, M. & DERWENT, R. D. 2006. Studies of episodes of high and low background air pollution. Bristol: UK Environment Agency.

KONOVALOV, I., BEEKMANN, M., MELEUX, F., DUTOT, A. & FORET, G. 2009. Combining deterministic and statistical approaches for PM< sub> 10</sub> forecasting in Europe. *Atmospheric Environment,* 43**,** 6425-6434.

KUMAR, U. & JAIN, V. 2010. ARIMA forecasting of ambient air pollutants (O3, NO, NO2 and CO). *Stochastic Environmental Research and Risk Assessment,* 24**,** 751-760.

MCGETTIGAN, M., O'DONNELL, C. & TONER, P. 2000. Draft national air quality monitoring programme. Johnstown Castle Wexford: Environmental Protection Agency.

MÖLTER, A., LINDLEY, S., DE VOCHT, F., SIMPSON, A. & AGIUS, R. 2010. Modelling air pollution for epidemiologic research–Part II: Predicting temporal variation through land use regression. *Science of the total environment,* 409**,** 211-217.

MOUSSIOPOULOS, N. 1997. *Ambient Air Quality, Pollutant Dispersion and Transport Models*, Office for Official Publications of the European Communities.

MOUSTRIS, K. P., ZIOMAS, I. C. & PALIATSOS, A. G. 2010. 3-Day-ahead forecasting of regional pollution index for the pollutants NO2, CO, SO2, and O3 using artificial neural networks in Athens, Greece. *Water, Air, & Soil Pollution,* 209**,** 29-43.

MUSCHETT, F. D. 1981. SPATIAL DISTRIBUTIONS OF URBAN ATMOSPHERIC PARTICULATE CONCENTRATIONS*. *Annals of the Association of American Geographers,* 71**,** 552-565.

ROBESON, S. M. & STEYN, D. G. 1990. Evaluation and comparison of statistical forecast models for daily maximum ozone concentrations. *Atmospheric Environment. Part B. Urban Atmosphere,* 24**,** 303-312.

SARASWAT, A., APTE, J. S., KANDLIKAR, M., BRAUER, M., HENDERSON, S. B. & MARSHALL, J. D. 2013. Spatiotemporal land use regression models of fine, ultrafine, and black carbon particulate matter in New Delhi, India. *Environmental science & technology,* 47**,** 12903-12911.

SHI, J. P. & HARRISON, R. M. 1997. Regression modelling of hourly NOx and NO2 concentrations in urban air in London. *Atmospheric Environment,* 31**,** 4081-4094.

SINGH, K. P., GUPTA, S., KUMAR, A. & SHUKLA, S. P. 2012. Linear and nonlinear modeling approaches for urban air quality prediction. *Science of the Total Environment,* 426**,** 244-255.

STEDMAN, J. & BUSH, T. 2000. Mapping of nitrogen dioxide and PM10 in the UK for Article 5 Assessment. Abingdon: The Department of the Environment, Transport and the Regions, The National Assembly for Wales, The Scottish Executive and the Department of the Environment in Northern Ireland.

STEDMAN, J. R. 1998. Revised High Resolution Maps of Background Air Pollutant Concentrations in the UK: 1996. *AEA Technology, National Environmental Technology Centre. Report.* Environment Agency for England and Wales.

STEDMAN, J. R., CAMPBELL, G. W. & VINCENT, K. J. 1997a. Estimated High Resolution Maps of Background Air Pollutant Concentrations in the UK in 1994. AEA Technology, National Environmental Technology Centre. Environment Agency for England and Wales.

STEDMAN, J. R., KENT, A. J., GRICE, S., BUSH, T. J. & DERWENT, R. G. 2007. A consistent method for modelling PM10 and PM2.5 concentrations across the United Kingdom in 2004 for air quality assessment. *Atmospheric Environment,* 41**,** 161-172.

STEDMAN, J. R., VINCENT, K. J., CAMPBELL, G. W., GOODWIN, J. W. L. & DOWNING, C. E. H. 1997b. New high resolution maps of estimated background ambient NOx and NO2 concentrations in the UK. *Atmospheric Environment,* 31**,** 3591-3602.

SU, J. G., BRAUER, M., AINSLIE, B., STEYN, D., LARSON, T. & BUZZELLI, M. 2008. An innovative land use regression model incorporating meteorology for exposure analysis. *Science of the total environment,* 390**,** 520-529.

VARDOULAKIS, S., FISHER, B. E. A., GONZALEZ-FLESCA, N. & PERICLEOUS, K. 2002. Model sensitivity and uncertainty analysis using roadside air quality measurements. *Atmospheric Environment,* 36**,** 2121-2134.

VIENNEAU, D., DE HOOGH, K., BEELEN, R., FISCHER, P., HOEK, G. & BRIGGS, D. 2010. Comparison of land-use regression models between Great Britain and the Netherlands. *Atmospheric Environment,* 44**,** 688-696.
VLACHOGIANNI, A., KASSOMENOS, P., KARPPINEN, A., KARAKITSIOS, S. & KUKKONEN, J. 2011. Evaluation of a multiple regression model for the forecasting of the concentrations of NO x and PM 10 in Athens and Helsinki. *Science of the total environment,* 409**,** 1559-1571.
VOUKANTSIS, D., KARATZAS, K., KUKKONEN, J., RÄSÄNEN, T., KARPPINEN, A. & KOLEHMAINEN, M. 2011. Intercomparison of air quality data using principal component analysis, and forecasting of PM 10 and PM 2.5 concentrations using artificial neural networks, in Thessaloniki and Helsinki. *Science of the Total Environment,* 409**,** 1266-1276.
WHO 2012. Health effects of black carbon, Joint WHO/UNECE Task Force on Health Aspects of Air Pollutants under UNECE's Long-Range Transboundary Air Pollution Convention (LRTAP). World Health Organization, Regional Office for Europe, Copenhagen.
ZHANG, Y., BOCQUET, M., MALLET, V., SEIGNEUR, C. & BAKLANOV, A. 2012. Real-time air quality forecasting, part I: History, techniques, and current status. *Atmospheric Environment,* 60**,** 632-655.