Air4EU

Air Quality Assessment for Europe: from local to continental scale

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Basic data assimilation: application to the European scale

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1. Executive Summary

There are a number of methodologies available for combining monitoring and modelling data that will lead to the production of maps showing air quality. Many of these are complex applications involving data assimilation techniques that will require direct interaction between model calculations and observational data. The complexity of such methods limits there application to research institutes, making them less accessible to authorities that require such maps. A methodology already developed for the production of European maps based on annual mean model and monitoring statistics is applied in this case study. The assimilation method investigated uses linear regression analysis combined with residual kriging of the observed-modelled residual. The methodology is further applied to daily mean model fields and monitoring data to assess the differences between assimilation using annual or daily statistics for PM_{10}. The quality of the two different methods is assessed using the cross validation RMSE and by producing uncertainty maps. Maps showing the annual mean, the 36^{th} highest daily mean percentile value and the number of exceedance days for PM_{10} are produced using both methods

The result of the case study indicates that there are differences in the maps produced using either annual or daily statistics. The most significant difference occurs in regard to the maps showing the number of exceedance days. Differences in particular areas can be as high as 30 days. However, the uncertainty in both methods has also been shown to be of a similar order, 20 – 30 days, in large regions of Europe. It is concluded in the case study that assessment maps should produced using daily assimilated means rather than annual statistics. These maps contain more information, since observational data is often discarded from annual statistics when the temporal coverage is incomplete, and produce consistent maps in regard to percentiles and the number of exceedance days.

To demonstrate the improvements in the air quality maps, corresponding uncertainty maps are also produced. The construction of such maps aids in visualising the uncertainty in the assessment maps however methodologies for producing these maps still require further refinement. The case study supports a number of recommendations concerning combining monitoring and modelling on the European scale.

2. Case study description

2.1 Background

There are a number of methodologies available for combining monitoring and modelling data. Many of these are complex applications involving data assimilation techniques that require direct interaction between model calculations and observed data. Examples of these are variational methods, e.g. 4D var, Kalman filters and other particle filter methods, see the cross cutting issue report on data assimilation (Air4EU – M5) for references to current applications and references. There are also other methods that can be applied ‘offline’ where precalculated model fields can be adjusted using monitoring data. These methods do not require interaction with the model and make use solely of the resulting fields. Examples of such methods are 3D var, optimal interpolation, regression modelling and various kriging methods. these methods are easier to implement without having to interact with complex models and some of these can be implemented using standard software.

As part of Air4EU these simpler methods are also explored in two case studies; this case study, which looks at a particular application on the European scale, and the Prague case study (Air4EU – CS D7.1.6) that looks at a variety of methods on the urban scale. In addition the Paris case study (Air4EU – CS D7.1.7) also applies one of these types of methods. Other applications using similar statistical methods, such as kriging and regression, include Kassteele et al. (2006), Horálek et al. (2005), Denby and Flicstein (2005) and Blond et al. (2003).

This case study is based on a methodology that has been applied to the production of European maps of air quality by Horálek et al. (2005) as part of the European Topic Centres (ETC/ACC) work on spatial mapping of pollutants for the European Environmental Agency (EEA). Maps of various ozone and PM10 indicators were produced using a combined method of multiple linear regression (using supplementary data such as EMEP model, altitude and a number of meteorological parameters) and kriging of the residual fields. In that study rural background maps were combined with urban background maps to produce European maps of air quality at 10 km resolution. During the study a number of different regression and kriging methods were tested.

In Horálek et al. (2005) the methodology was applied to annual statistics, i.e. maps were created for PM10 based on the annual mean and the 36th highest daily mean concentrations, that address legislated indicators according to the EU daughter directives (EC, 1999). In this case study the same methodology is applied to daily mean fields and the assessment is based on the analysis of the daily assimilation rather than on the annual statistics.

2.2 Aim and description

The aim of this case study is to apply and test the basic data assimilation methodology developed by Horálek et al. (2005) with a higher temporal resolution (daily) rather than the previously applied (annual) temporal resolution. Assimilated maps using both temporal resolutions will be compared and their uncertainty estimated using the cross-validation root mean square error (RMSE) and by comparison of uncertainty maps.

Rural background maps of Europe for PM10 will be produced using both annual and daily temporal resolutions and showing the following indicators

1. Annual mean PM_{10} concentration
2. 36th highest daily mean PM_{10} concentration
3. The number of exceedance (NOE) days of $PM_{10,}$ i.e. days when daily mean $PM_{10} > 50\ \mu g \cdot m^{-3}$

Conceptually differences in calculated fields for the two methods should occur for the following reasons:

- The regression and residual kriging parameters will be defined on a daily basis so that the resultant indicator field will be constructed differently.
- The number of stations available on a particular day will vary. Stations with coverage < 75%, excluded from the annual calculations, can contribute on a daily basis.

The aim thus of this case study is to assess the importance of these differences and their advantages and disadvantages

2.3 Relevance to recommendations in Air4EU

Air4EU intends to provide recommendations on spatial assessment for a range of users including city and European authorities. If data assimilation techniques are to be implemented by these authorities then they must be operationally applicable and, unless research institutes active in data assimilation are engaged in the assessment, they must also be based on accessible technologies. For this reason it is important to explore and recommend methodologies for data assimilation that can lead to improved spatial assessment using basic and available methods.

This case study tests a particular methodology that can be applied to regional scales. It will lead to recommendations concerning the use of this method for air quality assessment as well as exploring methodologies for displaying the uncertainty in these fields.

3. Methodology

As previously mentioned the methodology applied is based on the work of Horálek et al. (2005) and entails the use of linear regression to produce concentrations fields and then kriging the remaining residual error to include spatial interpolation space. In that previous study a number of supplementary data sources were used for the linear regression. In this study, mainly to aid analysis, only the most important supplementary data source, that being the Unified EMEP model, is included in the regression analysis. The technique is applied here to rural background PM10 data for the year 2003.

In this report we will first examine the statistical, regression and kriging parameters on a day-by-day basis to see how these vary with time (section 4.1). We will then go on to look at the results for the three different PM_{10} indicators mentioned above (sections 4.2 – 4.4) including a discussion on uncertainty mapping (section 4.5) and conclude with a discussion of the results and uncertainty (section 5).

Throughout the comparison use is made of the cross validation RMSE to assess the quality of the interpolations.

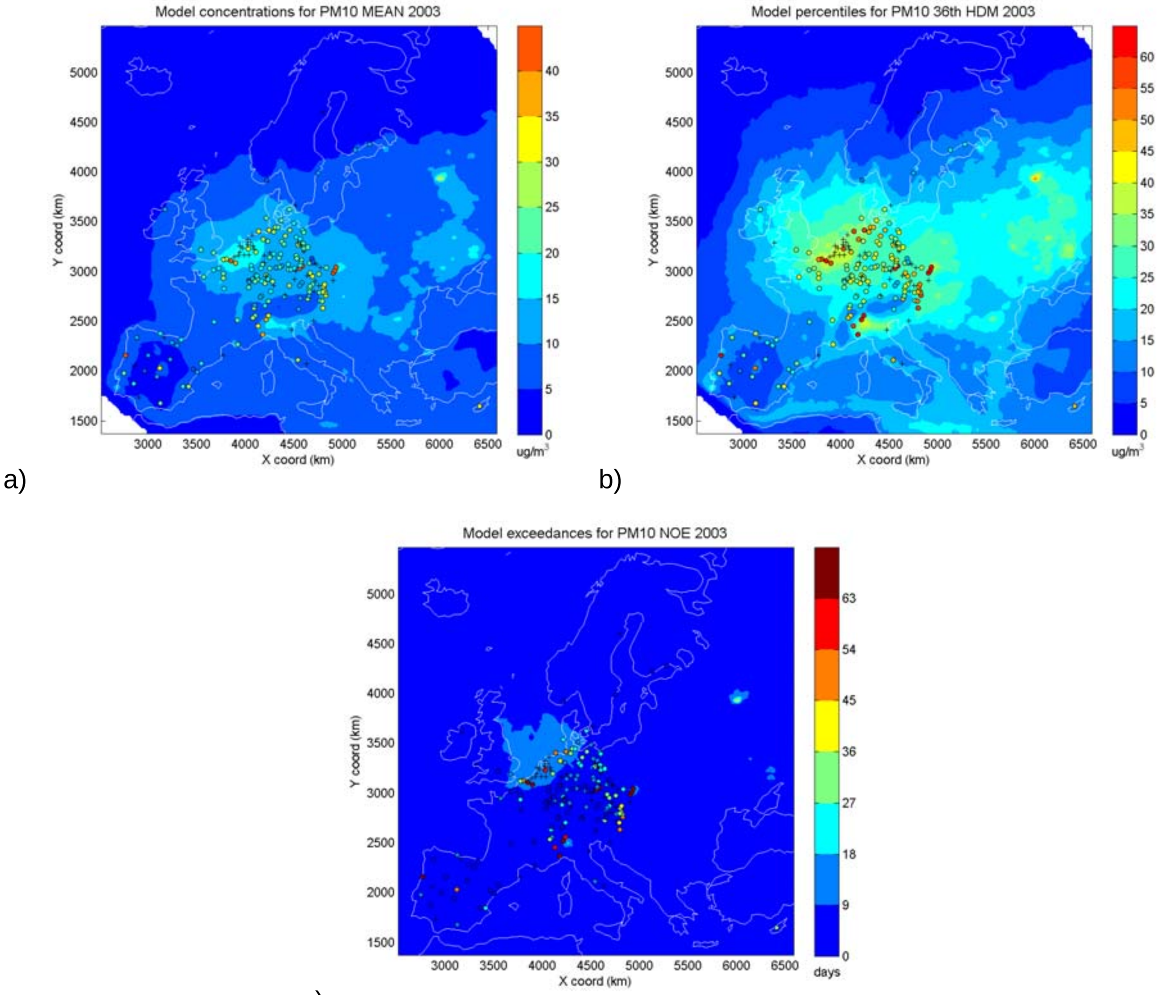
3.1 The observations

Observations used for the study have been extracted from both the AirBase (AirBase, 2005) and EMEP (EMEP, 2005) databases. All available PM10 stations providing daily mean concentrations, and classified as being rural background stations, have been included. In total 203 stations have been used. Stations with availability > 75% (151) have been used for the calculation of annual statistics, needed for the annual assimilation method. All stations are used, if available, for the daily mean assimilations. On average 161 stations are available every day. More discussion concerning the observations can be found in section 4.1. Figure 3.1 shows the station positions and their values for the 3 indicators analysed in this study.

3.2 The Unified EMEP model

The Unified EMEP model version rv2_1_2 (Simpson et al., 2003) has been used for the linear regression analysis of PM10. The model produces concentration fields of PM10 on a 50x50 km grid for all of Europe. In a recent intercomparison study for the years 1999 and 2001 (van Loon et al., 2004) the model, like many other European scale models, was shown to significantly underestimate PM10 concentrations with a relative bias of -45%. The underestimation is, most likely, caused by the large uncertainty in the modelling and measurement of secondary organic aerosols (SOA), and by the missing emission sources such as wind blown dust and resuspension. Despite this large bias the spatial distribution of PM10 concentrations represented by the EMEP model is expected to give valuable information to their spatial distribution.

For this case study daily mean PM10 concentrations as calculated by the EMEP model are used. When these concentrations are compared to observations bilinear interpolation is used from the 4 nearest model grids. Examples of the calculations for 2003 for the 3 indicators are shown in figure 3.1. THese figures clearly show the underestimation of all 3 indicators under discussion



a)

b)

c)

Figure 3.1: Unified EMEP model fields showing a) annual mean PM10 (ug/m^3), b) 36th percentile of the daily mean PM10 concentration (ug/m^3) and c) the number of exceedances (days) of the daily mean limit value. Filled contours indicate model fields, circles and crosses the observations. Stations with coverage > 75%, used in the annual interpolation, are shown with values as circles. Other stations used in the daily interpolation with coverage >25% are shown as crosses., and observed, circles with values, annual mean concentrations (ug/m^3) for PM10 in Prague 2003. Model field as calculated by the ATEM model.

3.3 Assessment of the results

In order to assess the results a number statistical parameters are used. Central to the assessment, as well as the determination of uncertainty, is the use of cross validation as a comparative test. Cross validation involves carrying out the assimilation using all the observations except one. The excluded observation is compared to the calculated one and this process is rotated around all the observations. To give a single value assessment of the quality of the assimilation method the RMSE of the cross-validated errors is used.

3.4 Assimilation method

The methodology applied in the study is described in the following sections. It consists of firstly fitting the model to the observations using a linear regression analysis and then using ordinary kriging of the residuals to interpolate the remaining errors in space.

3.4.1 *Linear regression*

Linear regression is used to produce regression models that relate model calculated concentrations to observed quantities. It is also possible to include other relevant fields, such as altitude, emissions, etc. in multiple regressions but in this study we will apply the regression model solely to the model calculated fields. Linear regression produces a result that minimises the RMSE between the regression model and the observations based on just 2 global parameters. This is written as

$$M_{LR} = a + bM \tag{3.1}$$

where M is the model field M_{LR} is the resulting linear regression field and a and b are the regression coefficients. In effect the entire model field is rescaled by the factor b (slope) and adjusted absolutely by the factor a (intercept). The intercept parameter a tells us something about the background concentrations but this will only be reliable when there is a high correlation in the regression.

It is also possible to apply linear regression in a limited fashion. e.g. when there is low correlation between modelled and observed values then simply correcting for the mean bias is more appropriate. This is the same as fixing the regression slope at $b = 1$ allowing an absolute the background value to vary.

Linear regression can be appropriate for models that have intrinsic bias in

- Total emissions
- Dispersion model formulations
- General estimates of wind speeds
- Background levels

However, linear regression is global in nature and will not help improve results when the variations are the result of:

- Local errors in emission factors
- Spatial variation in meteorological fields
- Local effects in wind, dispersion or emissions near monitoring sites

3.4.2 *Ordinary kriging and residual kriging*

Kriging, in all its forms, is an often used interpolation method in the geosciences. It revolves around the assumption that there is spatial correlation between points in space that is related to the distance between the points, i.e. the closer the points are then the more correlated, the further away the less correlated. This spatial correlation is described by a spatial variance function, called the semi-variogram, that describes spatial variance as a function of distance, figure 3.2. This function can be used to interpolate to any point in space when observations are available. The interpolation is carried out by weighting the nearby measurement points so that the variance at the interpolation point is minimised. In other words, given the assumed nature of the spatial variance function the value given to the interpolated point is statistically the most likely one. Defining the semi-variogram is thus critical to the method and should in principle be based on fits to the measured spatial variance. The weighting method is defined below where M_{OK} is the result of the ordinary kriging, λ_i is the weighting parameter and $O(x_i, y_i)$ is the observation *i* at position *x* and *y*.

$$M_{OK}(x,y) = \sum_{i=1}^{n} \lambda_i O(x_i, y_i) \tag{3.2}$$

In this study only ordinary kriging is applied. This type of kriging is most often used. It separates itself from other kriging methods in that it requires the sum of the weights to be equal to 1. This leads to particular properties including that the interpolated field, far from the observations, approaches the mean of these observations. For a description of kriging methods one is referred to various books on the subject such as Webster and Oliver (2001) and Cressie (1993)

Ordinary kriging assumes some form of stationarity to the field in as much that there are no spatial trends present. This is often not the case and so a number of methods for accounting for spatial trends, or ‘external drift’, can be applied. The method used here is to assume the model regression field represents the general character of the field, i.e. the drift, and to apply kriging to the residuals (also termed innovation), i.e observations minus regression model concentrations. This has been shown in other studies (Horálek, 2005; Blond et al., 2003) to be an effective method for improving mapped concentration fields on regional scales. The kriged field then becomes:

$$M_{OK\_RES}(x,y) = \left[\sum_{i=1}^{n} \lambda_i \big(O(x_i, y_i) - M_{LR}(x_i, y_i) \big) \right] + M_{LR}(x,y) \tag{3.3}$$

One of the major problems with kriging of the residual is that the semi-variogram is less well defined since much of the covariance is removed in the subtraction of the trend. Since it is often poorly defined the semi-variogram parameters can be determined by minimising the cross validation RMSE.

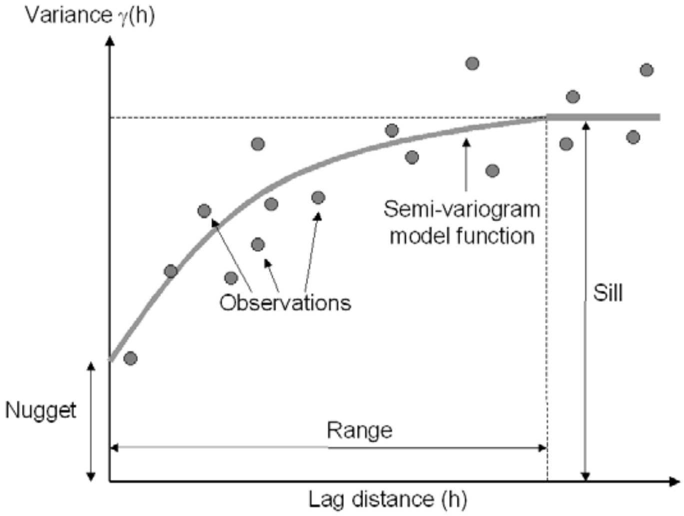


Figure 3.2: Showing the various parameters and terms used to describe the kriging semi-variogram function and how such a function can be fitted to the observed variance.

3.5 Uncertainty mapping

A number of methods can be considered when mapping the uncertainty, dependent on the method used to produce the map and on the parameter that should be shown. In this study methods for presenting uncertainty of particular parameters will be explored.

3.5.1 Uncertainty fields based on residual kriging variance

The uncertainty in the residual kriged fields can be visualised using the kriged variance calculated at each interpolated point. This variance is interpreted to be equivalent to the square of the standard deviation and is based on the form of the semi-variogram model used for the kriging. The nature of the semi-variogram, that of increasing variance with distance from observations, means that uncertainty maps based on kriging will show lowest uncertainties around the observations, increasing to the sill value far from the observations.

The disadvantage of this method, and generally with the kriging concept, is that the semi-variogram and its parameters are assumed to be universally applicable everywhere. Thus the variance associated with the semi-variogram is based more on the mean values of the fields. This means that, for instance in the case of annual mean, the kriged variance will be as large in areas of high concentrations as those with low concentrations. This is not likely to be the case as there will be some dependency of the uncertainty as a function of absolute concentration.

3.5.2 Uncertainty fields based on the temporal covariance matrix and residual kriging variance

Direct use of the kriging variance field can be made when no temporal dimension is involved. If the total uncertainty is to be established based on the accumulated uncertainty from daily calculations then the temporal covariance of the assimilated fields must be taken into account. This methodology is described below and applied in section 4.5.1.

To estimate the total variance for the annual mean, when it is based on the sum of daily mean values, it is not sufficient to simply ‘add up’ the daily variance fields calculated from the residual kriging, since there is a certain amount of correlation between concentration fields from day to day. A more extensive analysis is thus required. To estimate the total variance the temporal covariance matrix must be calculated since it is this that represents the correlations between all the days of the year. Mathematically it is useful to decompose the total variance into the sum of the variances and covariances, noting that in terms of the covariance matrix the variances are the diagonal terms and the covariances are the off diagonal terms. If we wish to calculate the total variance of the mean of a parameter *X,* based on the individual variances *Var(X)* then the variance can be written as

$$Var\left(\frac{1}{n}\sum_{i=1}^{n}X_i\right)=\frac{1}{n^2}\sum_{i=1}^{n}Var(X_i)+\frac{1}{n^2}\sum_{i=1}^{n}\sum_{j=1,j\neq i}^{n}Cov(X_i,X_j) \qquad (3.4)$$

$$=\frac{1}{n}\sum_{i=1}^{n}Var(X_i)\left[\frac{1}{n}+\frac{\frac{1}{n^2}\sum_{i}^{n}\sum_{j\neq i}^{n}Cov(X_i,X_j)}{\frac{1}{n}\sum_{i=1}^{n}Var(X_i)}\right] \qquad (3.5)$$

$$=\frac{1}{n}\sum_{i=1}^{n}Var(X_i)[F_{cv}]$$

The first term on the right hand side of equation 3.4 represents the on-diagonal terms of the covariance matrix and the second term the contribution from the off-diagonal terms. When there is no correlation between the elements of X, in this case days of the year, then this second term is 0 and the variance of the mean can simply be determined by the first term on the right hand side. In that case it would be possible to use the daily determined kriging variance to represent $Var(X)$ and simply divide by n^2, where n is the number of days. However, there is quite high correlation in the concentration fields and this must be accounted for by estimating the other covariance terms.

The above equation is rewritten, equation 3.5, to simplify interpretation. In its final form the total variance is simply the mean variance of all the days multiplied by a covariance factor F_{cv}. This factor represents approximately the ratio of the mean off-diagonal terms with the mean on-diagonal terms. Thus when the days are completely correlated with one another this factor approaches 1. When they are totally uncorrelated they approach n^{-1}. Writing the equations in this form allows us to estimate F_{cv} by creating the temporal covariance matrix, and using the daily kriged variance fields to represent $Var(X_i)$. The individual elements of the covariance matrix are estimated by calculating the variance between the interpolated fields from day to day. In this case the covariance matrix elements are calculated using the residual kriging fields at the positions of the observational stations, instead of the entire model domain, since these are likely to give the most representative results. The covariance matrix created thus contains 365 x 365 elements, representing the covariance of every day with every other day of the year.

3.5.3 Uncertainty fields for exceedance calculations

If the number of daily exceedances is to be mapped, based on the annual statistics, then this can be done by using the residual kriged variance. This, as previously mentioned, may be a poor indicator of the uncertainty since the uncertainty is as large in areas with very low numbers of exceedances as in areas with high numbers of exceedances.

When calculating the uncertainty of the NOE days, based on daily means, consideration must be made for the uncertainty in each of the daily mean exceedances. Though statistically it is possible to assess this it is strongly influenced by other aspects such as spatial representativeness and spatial model bias. In this case a more pragmatic approach is required than simply trying to assess the kriging variance directly. In this case individual daily means are perturbed by the uncertainty in the annual mean fields, interpreted to represent model bias and representativeness uncertainty. The perturbed daily mean values are then used to establish the uncertainty in the NOE days. This methodology is outlined in section 4.5.2 along with the resulting uncertainty map

3.5.5 Cross validation of the observational mean and the RMSE

As previously described, cross validation and its RMSE is used throughout this case study to assess the quality of the assimilation methods. RMSE is defined as

$$RMSE = \sqrt{\frac{1}{n}\sum_{i=1}^{n}\left(M_i - O_i\right)^2} \tag{3.6}$$

where M is the modelled value (be it directly from the model, from the regression analysis of from the residual kriging), O is the observed concentration and n is the number of observations available.

From a statistical perspective the following note is also made:

The simplest ‘model’ that can be used to describe the concentration field is simply the mean of all the observations. Taking the RMSE of this simple ‘model’ is equivalent to calculating the standard deviation (SD) of the observations. Any assimilated field should be able to improve on the observational SD as a method for estimating the concentration field.

4. Results

In the following sections the results of the case study are presented starting with an interpretation of the daily assimilation, comparison of the annual and daily methods and concluding with uncertainty analysis and mapping.

4.1 Daily variation of interpolation parameters

For each day the daily mean observed and modelled data are analysed and displayed in figures 4.1-4.5. The analysis includes the following parameters

- Daily mean of observed and modelled concentrations
- Daily standard deviation (SD) of observed and modelled concentrations
- Regression parameters of intercept, slope and regression coefficient (R^2)
- Kriging parameters of range and nugget:sill ratio
- Cross validation RMSE for
    - The EMEP model
    - Model regression
    - Kriging only
    - Residual kriging

4.1.1 Station availability

For the year 2003, 203 PM_{10} stations were available from the AirBase database with data coverage > 25%. Of these, only 151 stations with a coverage > 75% are used in the annual statistics. On a daily basis however the number of stations used for the interpolation varies. This is shown in figure 4.1. The number of stations used each day is, on average, 166.

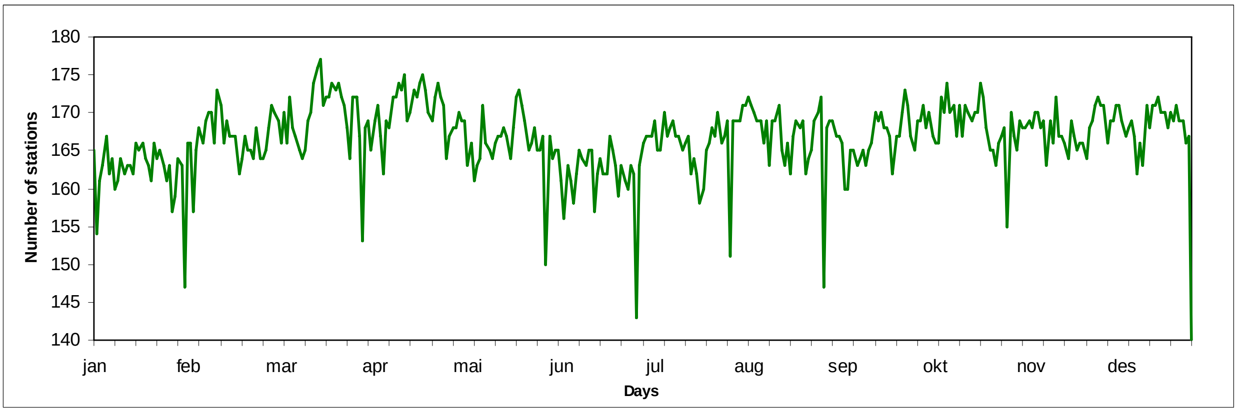


Figure 4.1 Plot showing the number of stations used for the daily interpolations, for the year 2003.The number of stations with coverage > 75% is 151. Number of stations with coverage>25% is 203. Average number of stations used each day is 166

4.1.2 Daily mean and standard deviation comparison

The daily mean (mean of all available stations for that day) is plotted in figure 4.2-top for both the observed and modelled (interpolated to monitoring sites) concentrations. There is a clear bias between monitoring and modelling with an average model bias of -12.2 μg·m^{-3}, or a relative bias of around –50%. The EMEP model is clearly underestimating PM_{10} concentrations throughout the model domain but the daily variation is well correlated, R^2=0.64, suggesting that there are missing sources (e.g. secondary particle formation, soil dust, etc.) but the effect of meteorology is well represented.

The daily SD (SD of all available stations for each day) is plotted in figure 4.2-bottom for both the observed and modelled (interpolated to monitoring sites) concentrations. The SD shows a similar bias to the mean concentrations, however when the normalised SD is calculated then both model and observations show very similar relative variance, of 59 % and 65 % respectively. This indicates that the EMEP model is capturing much of the variation inherent in the system even though concentrations are too low.

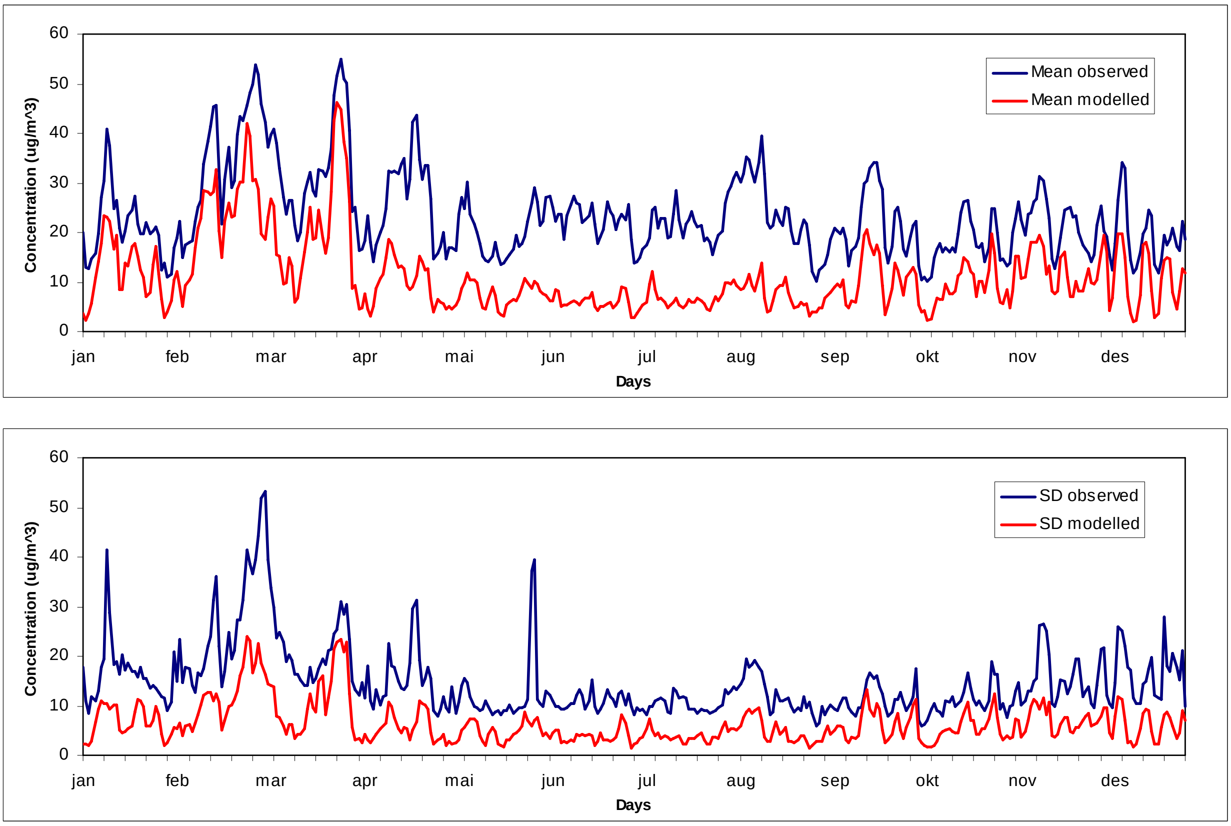


Figure 4.2. Plot of the daily mean concentrations (top) and SD (bottom) for all stations used in the daily interpolation, blue, along with the corresponding model concentrations, red, for the year 2003.

4.1.3 Regression parameters

For each day the model is fitted using linear regression with the resulting regression parameters of intercept, slope and correlation coefficient (R^2). Note that, to avoid spurious regressions when correlation is poor, the linear regression model was substituted by a simple bias correction when R^2 < 0.1. These parameters are plotted in figure 4.3. Only occasionally does the intercept fall below 0, with an average for the entire period of 12.0 μg·m^{-3}, c.f. model bias of –12.2 μg·m^{-3}. This reflects the general bias already known in the EMEP model in regard to PM_{10}.

The slope of the linear regression varies from day to day but is rarely below 0.5 or above 2.0. The correlation coefficient is larger than 0.1 for 63% of the time and so normal linear regression is applied for the majority of cases. The average slope of all the days is 1.10.

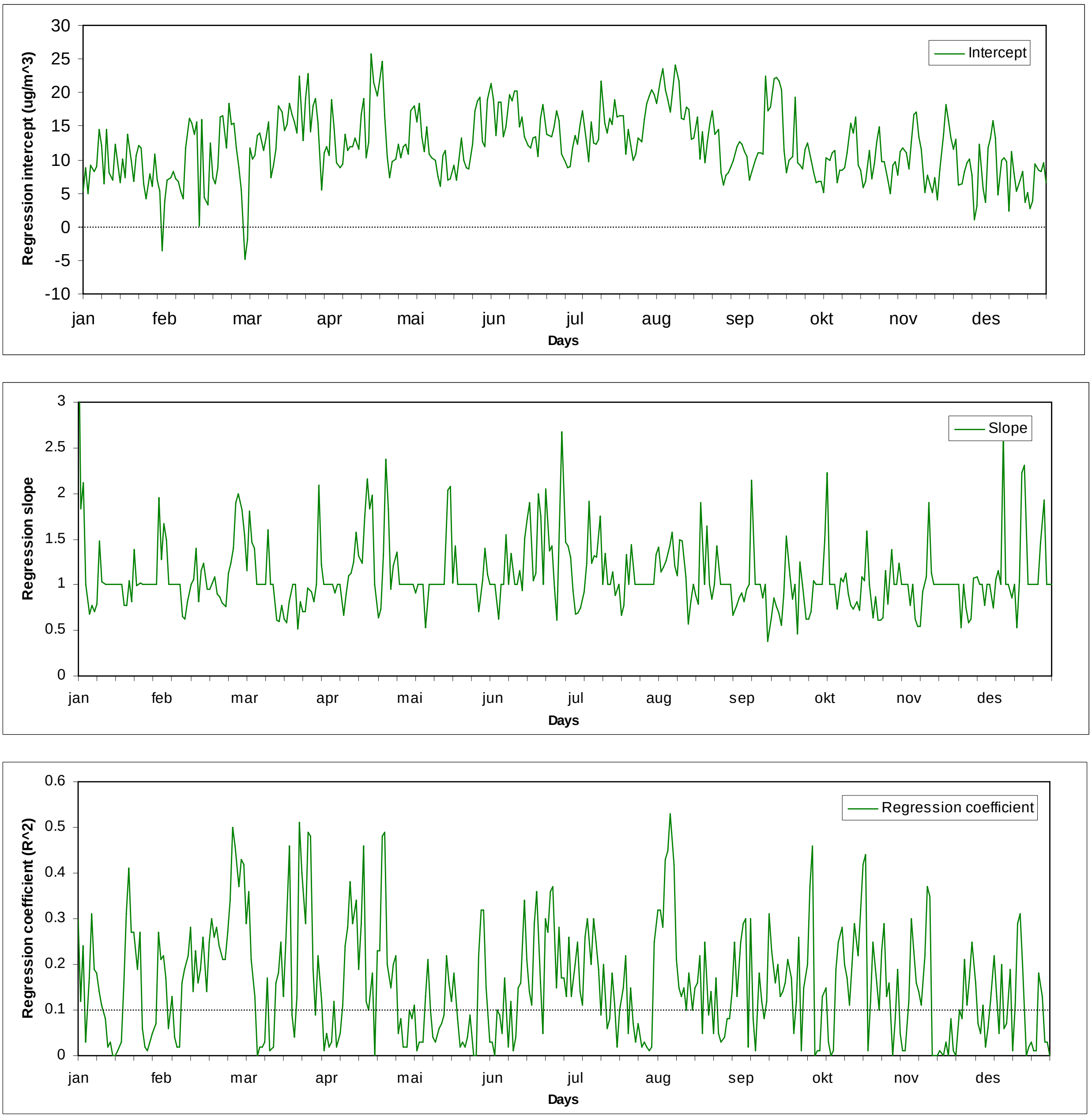


Figure 4.3 Plot showing the daily variation of the regression parameters used in the linear regression model fit of the modelled concentrations to the observed concentrations for the year 2003. Intercept (top), slope (middle) and correlation coefficient (bottom).

4.1.4 Kriging parameters

For each day the residual, observed concentration minus regression model concentration, is interpolated to create the final interpolation map. For comparison purposes the direct kriging of the observed concentrations is also carried out. The kriging parameters are determined by an automatic routine that minimises the cross validation RMSE in regard to range and nugget:sill ratio. This routine tests range values, from 100 km to 1000 km in steps of 100 km, as well as nugget:sill ratios, from 0 to 1 in steps of 0.1, to find the parameters that minimise the cross validation RMSE. In addition to this routine it is also possible to directly fit the semi-variograms with a spherical model for which the range is fixed at 300 km, a value found to be optimal (see section 4.2) for annual mean interpolations. The sill is specified in both cases by this fit.

The nugget:sill ratio and range, figure 4.4 top and bottom, indicate the dependence of spatial variance on the lag distance. Short ranges indicate small scale spatial variance and small nugget:sill ratios indicate strong local covariance. If the variogram model has a high nugget:sill ratio then this means there is little spatial covariance between observations. If the range is very small, independent of the nugget:sill ratio, then there is also very little spatial covariance over the region. When this is the case the kriging methodology does little more than produce the spatial average of the stations used for the kriging interpolation at that point.

From figure 4.4-top it can be seen that there are a significant number of days with nugget:sill ratios of intermediate values, indicating that kriging is indeed providing spatial interpolations. It is interesting to note that the nugget:sill ratios for residual and pure kriging follow very similar temporal developments. This indicates, as will be discussed later, that the spatial covariance of the residual and the observations are quite similar.

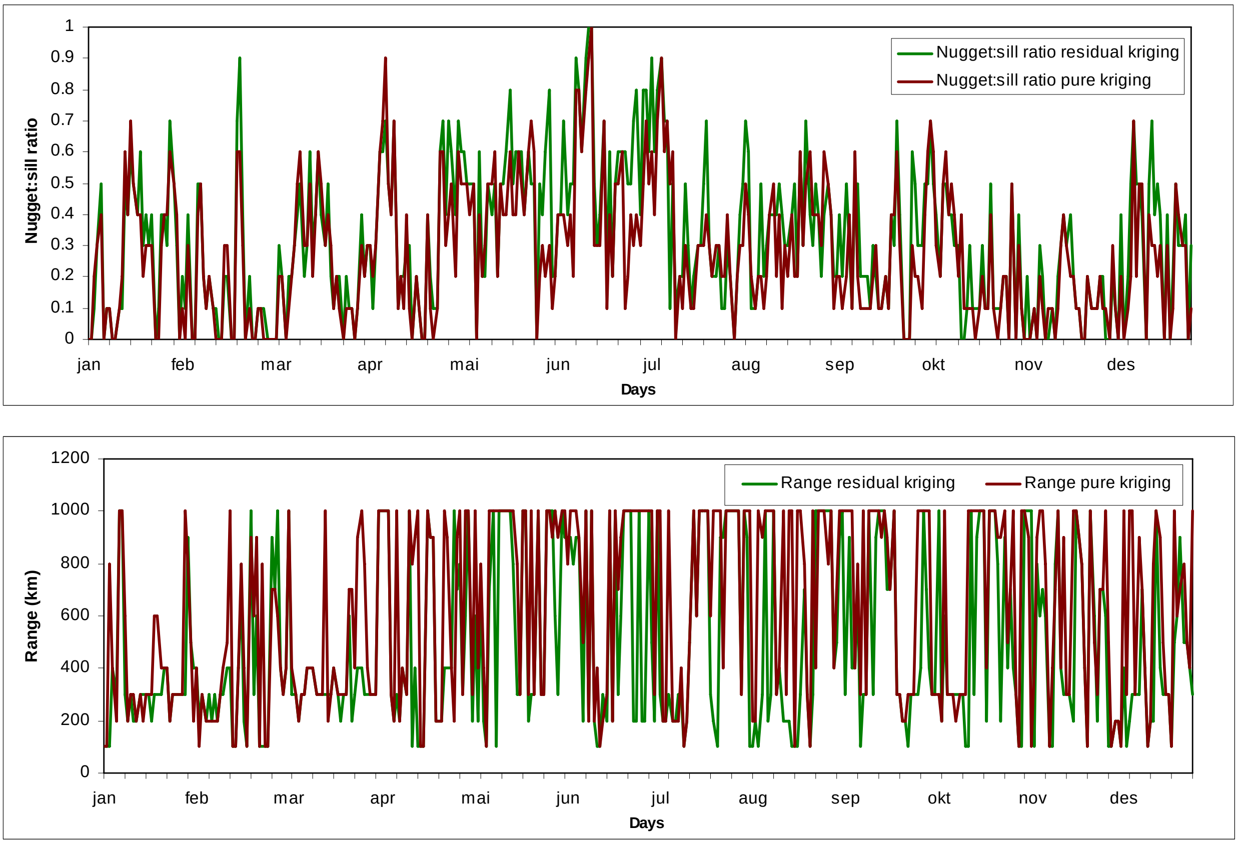
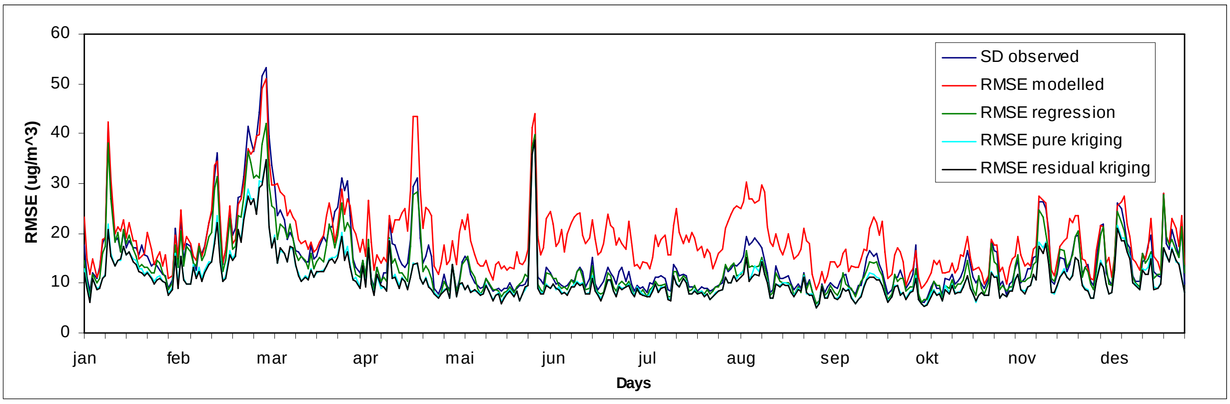


Figure 4.4 Plots of the kriging parameters determined on a daily basis: nugget:sill ratio (top) and range (bottom). The parameters are determined by minimising the cross validation RMSE for each day. Shown are the kriging parameters for both residual kriging, green, and pure kriging (direct kriging of observations), brown.

4.1.5 Cross validation RMSE

The RMSE has been calculated on a daily basis for the 4 cases of model, regression model, pure kriging and residual kriging. The resulting graph is shown in figure 4.5 (top), along with a cut out for the month of April (figure 4.5, bottom) to show the results in more detail. Included in the graph is also the observed SD for reference. This is included since any interpolation should improve upon this value as it indicates the RMSE of the simplest model available, that being a model where the entire concentration field is equal to the mean of the observations.



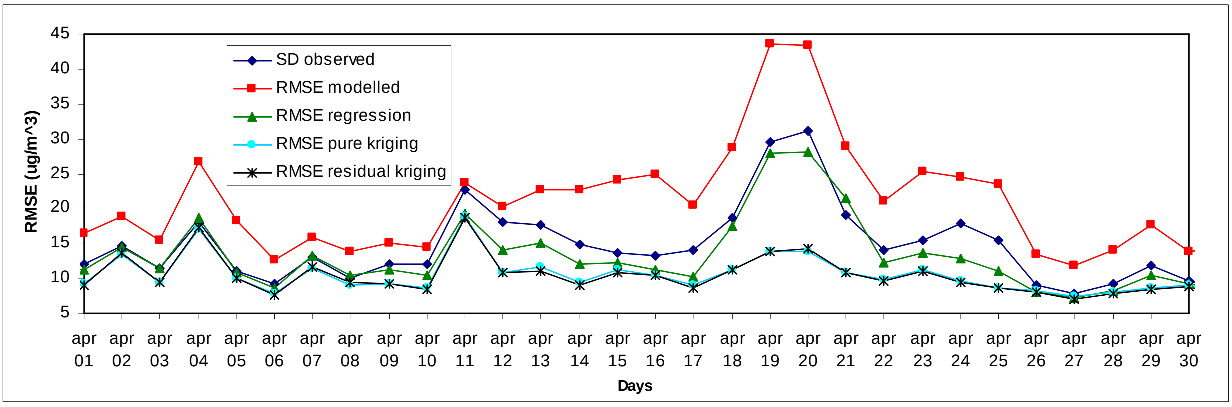


Figure 4.5 Plots for the entire year (top) and for April only (bottom) of the cross validation RMSE determined on a daily basis for model, regression model, pure kriging and residual kriging. Included is the observed SD for comparison.

The modelled RMSE is clearly the largest, mainly due to the bias in the EMEP model for PM_{10}. Regression removes this bias and for the most part improves on the observed SD. However, pure kriging generally produces RMSEs that can be significantly less than the regression models. There is only a minimal improvement when kriging of the residual is applied.

The results are summarised in table 4.1 below, which gives the total RMSE for all stations and all days. These values indicate that kriging is dominant in providing the best interpolation on the daily scale. The improvement obtained through residual kriging is small.

Table 4.1 Summary table showing the total cross validation RMSE on a daily basis. Normalisation of RMSE (NRMSE) is carried out using the total mean observed concentration = 23.48 $\mu g \cdot m^{-3}$

Interpolation method	Cross validation	Cross validation

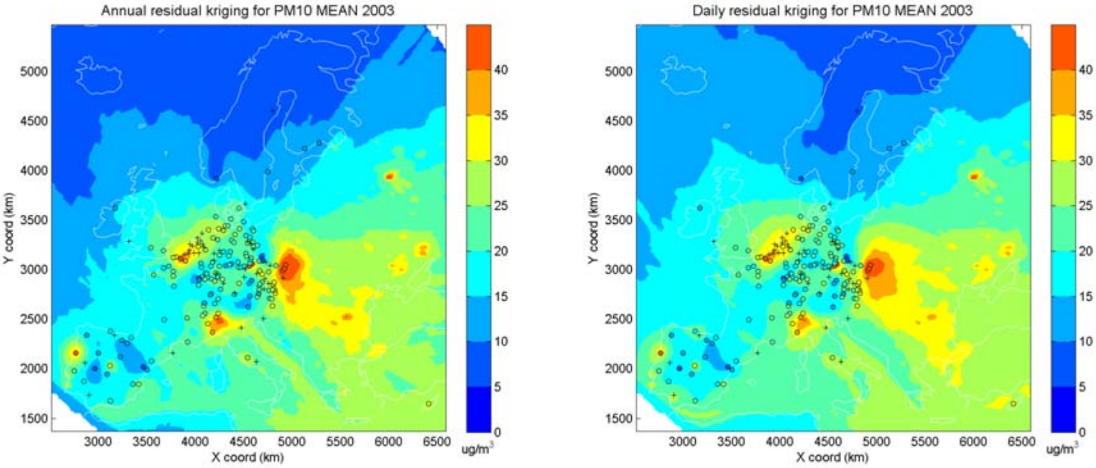
	RMSE ($\mu g \cdot m^{-3}$)	NRMSE (%)
Observed SD	16.4	70
EMEP model	20.0	85
Regression model	14.8	63
Pure kriging	12.1	52
Residual kriging	11.8	50

Since the interpolation, when using regression or kriging, is generally unbiased the RMSE given in table 4.1 can be directly interpreted in terms of the global uncertainty in the interpolation, expressed in terms of standard deviation when a Gaussian distribution is assumed. Even when using the best interpolation method the normalised RMSE (NRMSE) is still 50% indicating that the global uncertainty in the daily mean concentration fields of rural background PM_{10} is quite high.

4.2 Comparison of annual mean fields using daily and annual statistics

We will now compare the calculated annual mean fields derived from daily and annual statistics using residual kriging of the model regression. The two fields are calculated in essentially the same manner except that for daily statistics the resultant field is given by the mean of the interpolated fields whilst for annual statistics the resultant field is the interpolation of the observed means.

Figure 4.6 shows the annual field, the daily field and the difference field (annual – daily) respectively. Differences over most of Europe are of the same order or smaller than the estimated uncertainty, based on the RMSE shown in table 4.2. However, it should be noted that the comparison is complicated by the RMSE minimisation methodology applied to determine the kriging parameters. This minimisation is applied to the annual means only once and the kriging parameters obtained strongly influence the resulting interpolation. To avoid this complication the map shown in figure 4.6 (top-left) was determined using the average nugget:sill ratio and range taken from the daily fits, i.e. 0.31 and 250 km respectively. If this was not done then the automatic RMSE minimalisation routine would have provided a nugget:sill ratio of 0 and a range of 100 km resulting in a significantly different plot but with an insignificant improvement in RMSE.



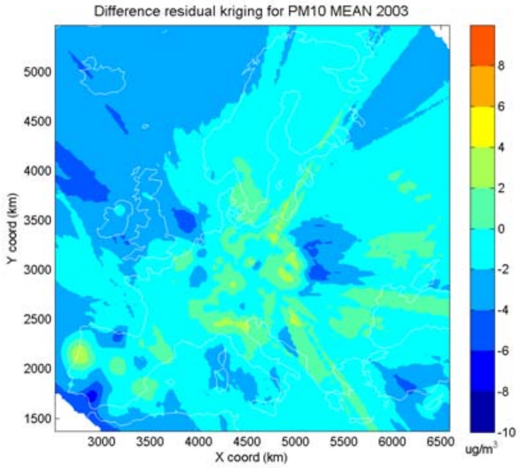


Figure 4.6. Showing the interpolated field for annual mean PM_{10} ($\mu g \cdot m^{-3}$) in 2003 based on annual statistics (top-left), daily statistics (top-right) and the difference (annual – daily) between the 2 fields (bottom). The interpolation method used is the residual kriging of model regression and the grid resolution of the interpolation is 25 x 25 km. Also included are the stations used in the interpolations. Stations with coverage > 75%, used in the annual interpolation, are shown with values as circles. Other stations used in the daily interpolation with coverage >25% are shown as crosses.

Table 4.2 summarises the results in terms of the cross validation RMSE and NRMSE. The annual mean field produced using residual kriging after regression produces the lowest RMSE. The RMSE is slightly lower when using daily statistics for all cases.

Table 4.2 RMSE of annual and daily statistics. The NRMSE is calculated using the annual limit value = 40 $\mu g \cdot m^{-3}$ to indicate the relative uncertainty at this value.

	Annual		**Daily**	
Interpolation method	RMSE ($\mu g \cdot m^{-3}$)	NRMSE (%)	RMSE ($\mu g \cdot m^{-3}$)	NRMSE (%)
Observed SD	9.26	23	9.26	23
EMEP model	14.4	36	14.4	36
Regression model	8.31	21	8.26	21
Pure kriging	7.07	18	6.93	17
Residual kriging	6.74	17	6.56	16

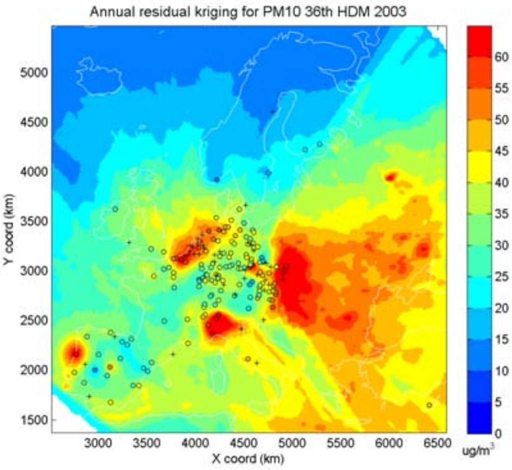
Conclusion

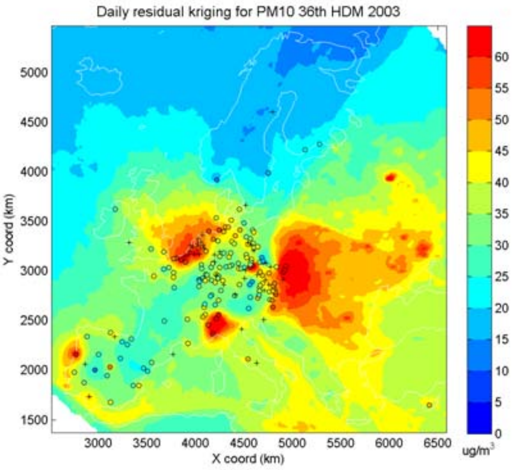
Though there are some differences between the annual mean concentration fields for PM_{10} produced using annual and daily statistics, these differences are of the same order, or smaller, than the estimated uncertainty in the methods when appropriate kriging parameters are used for the annual interpolation. Thus the small improvement obtained using daily data must be weighed against the extra data handling requirements to achieve them. However, use of daily interpolations must be seen as a more robust method in terms of estimating the appropriate kriging parameters than the use of a single interpolation for the annual statistics. In addition the inclusion of more stations, on a daily basis, can only help to improve the resulting mean fields.

4.3 Comparison of percentile fields using daily and annual statistics

We will now compare the calculated annual percentile fields derived from daily and annual statistics using residual kriging of the model regression. The two fields are calculated in essentially the same manner except that for daily statistics the resultant field is given by the 36^{th} highest daily mean of the interpolated fields whilst for annual statistics the resultant field is the interpolation of the observed 36^{th} highest daily mean.

Figure 4.7 shows the annual percentile field, the daily percentile field and the difference field (annual – daily) respectively. Differences over most of Europe are of the same order or smaller than the estimated uncertainty, based on the RMSE shown in table 4.3. However, as in the case of the annual mean, the comparison is sensitive to the choice of kriging parameters. In a similar fashion the top-left map of figure 4.7 was determined using the average nugget:sill ratio and range taken from the daily fits, i.e. 0.31 and 250 km respectively. If this was not done then the automatic RMSE minimalisation routine would have provided a nugget:sill ratio of 0 and a range of 100 km resulting in a quite different plot and a more substantial difference between the daily and annual fields.





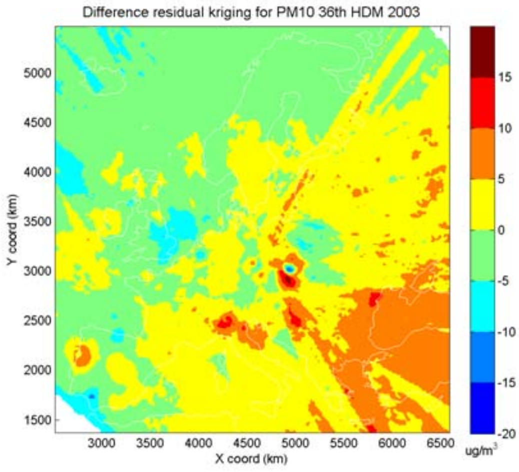


Figure 4.7. Showing the interpolated field for the 36th *highest daily mean concentration of* PM_{10} *(*$\mu g{\cdot}m^{-3}$*) in 2003 based on annual statistics (top-left), daily statistics (top-right) and the difference (annual – daily) between the 2 fields(bottom). The interpolation method used is the residual kriging of model regression and the grid resolution of the interpolation is 25 x 25 km. Stations with coverage > 75%, used in the annual interpolation, are shown with values as circles. Other stations used in the daily interpolation with coverage >25% are shown as crosses.*

Table 4.3 summarises the results in terms of the cross validation RMSE and NRMSE, where the RMSE has been normalised with the daily limit value for human health of 50 $\mu g{\cdot}m^{-3}$. The percentile field produced using residual kriging after regression produces the lowest RMSE. The RMSE is slightly lower when using daily statistics for all cases.

Table 4.3 RMSE of annual and daily statistics for the 36th *highest daily mean. The NRME is normalised using the limit value of 50 ug/m3 to indicate the relative uncertainty at the threshold value.*

	Annual		**Daily**	
Interpolation method	RMSE (ug/m3)	NRMSE (%)	RMSE (ug/m3)	NRMSE (%)
Observed SD	17.4	35	17.4	35
EMEP model	22.8	46	22.8	46
Regression model	16.0	32	16.1	32
Pure kriging	13.2	26	12.6	25
Residual kriging	12.6	25	12.0	24

Conclusion

Though there are some differences between the percentile fields for PM_{10} produced using annual and daily statistics, these differences are of the same order, or smaller, than the estimated uncertainty in the methods when appropriate kriging parameters are used for the annual interpolation. However, use of daily interpolations must be seen as a more robust method in terms of estimating these kriging parameters than the use of a single interpolation for the annual statistics. In addition the inclusion of more stations, on a daily basis, can only help improve the resulting percentile fields.

4.4 Comparison of number of exceedance days fields using daily and annual statistics

We will now compare the calculated number of exceedance days (NOE) derived from the daily and annual statistics using residual kriging of the model regression. The NOE and the 36th highest daily mean are two methods for describing the exceedance of limit values but they differ in that the percentile field is more continuous, having no cut-off value, whilst the NOE on the other hand is calculated using such a threshold value. This tends to lead to a more discontinuous field.

The interpolation using daily and annual statistics differ in that for daily statistics each daily mean concentration field is assessed as to whether it has exceeded the limit value of 50 $\mu g{\cdot}m^{-3}$. The exceedance field is then summed over the entire year. When using annual statistics the number of observed exceedance days, in combination with those calculated with the EMEP model and regression, are interpolated. This leads to complications since the EMEP model, due to its bias, rarely exceeds the threshold value leading to little improvement through regression. In addition, because there are many regions with no exceedance days, the residual kriging or pure kriging interpolations can lead to negative values of the exceedance days when the interpolation is carried out based on annual statistics.

Figure 4.8 shows the annual based NOE field, the daily based NOE field and the difference field (annual – daily) respectively. Differences between the two methods can be quite large but are of the order of the estimated uncertainty, based on the RMSE shown in table 4.5 and the uncertainty maps given in figure 4.10. It is important to note that even the best RMSE for the NOE days is still 20 days indicating a large uncertainty in the spatial mapping of this parameter.

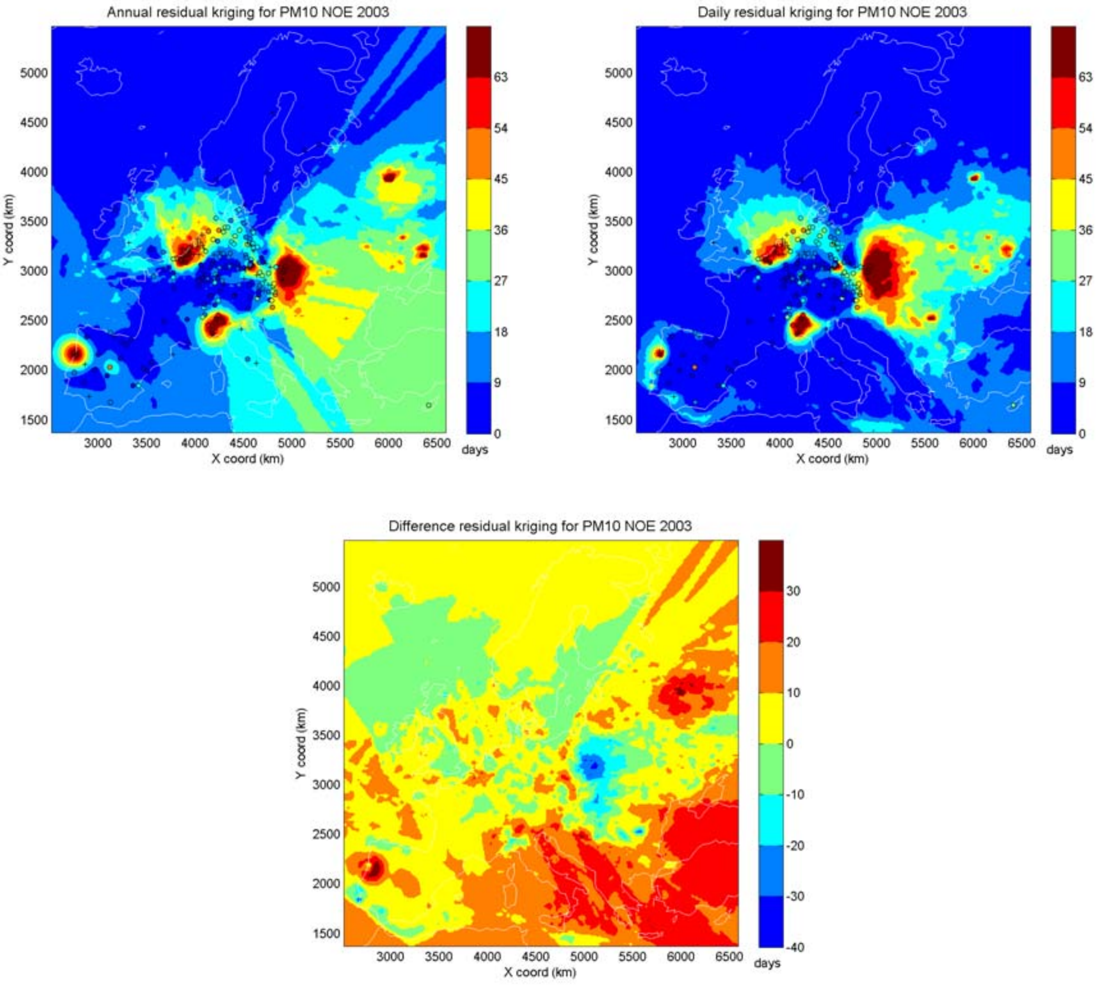


Figure 4.8. Showing the interpolated field for NOE days for PM_{10} (days) in 2003 based on annual statistics (top-left), daily statistics (top-right) and the difference (annual – daily) between the 2 fields (bottom). The interpolation method used is the residual kriging of model regression and the grid resolution of the interpolation is 25 x 25 km. Stations with coverage > 75%, used in the annual interpolation, are shown with values as circles. Other stations used in the daily interpolation with coverage >25% are shown as crosses.

Table 4.5 summarises the results in terms of the cross validation RMSE and NRMSE, normalised with the threshold value of 36 days. The NOE field produced using residual kriging after regression produces the lowest RMSE. In this case, as opposed to the mean and percentile fields, the RMSE is slightly lower when using annual statistics, though it cannot be considered to be significant. Also improvement of the RMSE of NOE through regression with the Unified EMEP model is less than that found in then annual mean and percentile calculations, table 4.4. Residual kriging is only a small improvement on the pure kriging method. This is true for both the annual and daily statistics.

Conceptually it is expected that the best method for creating NOE day fields is by the use of daily statistics, as has been found for the annual mean and percentile calculations, see tables 4.2 and 4.3. The cross validation RMSE is, however, not improved using daily statistics when interpolating the NOE, table 4.4. However it is important to note that there is a direct spatial correspondence between the percentile and NOE maps produced when daily statistics are used. This guarantees that the contour representing the limit value, for the case of percentile maps, and the contour representing the number of allowed exceedance days, for the NOE maps, are concurrent in space. This is not the case when interpolating the percentiles and NOE using annual statistics, since the two indicators are interpolated independently of each other. This can be seen when comparing the annual based percentile and NOE interpolations, figures 4.7-top-left and 4.8-top-left, where the limit value contour of 50 $\mu g \cdot m^{-3}$ is not spatially coincident with the 36 NOE days contour.

Table 4.4 RMSE of annual and daily statistics for the number of exceedance days. The NRMSE is normalised with the limit threshold of 36 days to indicate the relative uncertainty around this threshold.

	Annual		**Daily**	
Interpolation method	RMSE (days)	NRMSE (%)	RMSE (days)	NRMSE (%)
Observed SD	29.2	81	29.2	81
EMEP model	32.6	91	32.6	91
Regression model	26.1	73	27.8	77
Pure kriging	21.1	59	21.8	61
Residual kriging	20.8	57	21.0	58

Conclusion

The differences between the NOE fields for PM_{10} produced using annual and daily statistics, are of the same order as the estimated uncertainty in the methods and as such it cannot be considered as significant. Even though the cross validation RMSE is almost the same for the annual interpolation the use of daily interpolations must be seen as a more robust method for mapping NOE for the following reasons

- It is more robust in estimating the appropriate kriging parameters than the use of a single interpolation for the annual statistics
- There is consistency with the percentile maps
- The number of observations used is higher
- Kriging of a non-continuous function, as is the case of NOE using annual statistics, is not recommended

4.5 Uncertainty analysis and mapping

A discussion concerning the uncertainty in the two different methods is required. Throughout this chapter and this report the cross validation RMSE has been used to indicate the quality of the interpolation. This is a useful and well established parameter, though several other parameters may also have been used. This parameter, however, does not give spatial information on the uncertainty of the maps but provides a more global indication of the mapping quality. To indicate the spatial uncertainty use can be made of the variance field produced from the residual kriging interpolation, whether it be based on annual or daily statistics. The spatial uncertainty can be represented by the square root of this variance which is indicative, if a normal distribution is assumed, of the standard deviation (SD_{krig}).

The sill values of the residual kriging interpolation, determined automatically by fitting a spherical model to the residual semivariogram, should thus be close to, but most likely slightly larger than, the regression model RMSE. This is because the regression model RMSE is equivalent to the residual standard deviation (SD_{res}) and the sill represents variances that are generally larger than this value, given that smaller variances are found for shorter lag distances. This is found to be the case, as shown in table 4.5 for the interpolation based on annual statistics. The cross validation RMSE should then be less than the sill value since it represents stations both close to and far from other stations. This is indeed the case, as shown in table 4.5.

Table 4.5 Cross validation RMSE for residual kriging, regression model RMSE and SQRT(sill) for the annual based residual interpolations

Interpolation indicator	Cross validation RMSE	Regression model RMSE (SD_{res})	SQRT(sill)
Mean ($\mu g \cdot m^{-3}$)	6.7	8.3	8.4
Percentile ($\mu g \cdot m^{-3}$)	12.6	16.0	16.3
NOE days (days)	20.4	26.1	26.3

The use of the single globally valid semivariogram model in the kriging interpolation also leads to a rather homogenous spatial view of the uncertainty, particularly far from stations, since it then becomes independent of local concentrations levels. This is likely not to be an appropriate representation of the true spatial uncertainty in the maps.

Despite this, the residual kriging standard deviation field (SD_{krig}) may be used to indicate the spatial uncertainty for the annual based statistics but this becomes more complicated when using daily statistics and when estimating the NOE days, as will be further discussed in section 4.5.1 and 4.5.2.

4.5.1 Uncertainty in the annual mean when using daily statistics

The methodology for calculating the spatial uncertainty of the annual mean concentration, based on the accumulated daily mean values, is described in section 3.5.2. It requires calculation of the temporal covariance matrix to estimate of the parameter $F_{cv.}$ This parameter indicates the ratio of the mean off-diagonal elements (covariances) to the on-diagonal ones (variances). Making the calculation in equation 3.5 provides

$F_{cv} = 0.239$

indicating a substantial contribution to the total variance from the covariance terms. This factor is then used to scale the sum of the kriged variance estimates, taken from the daily interpolations, to estimate the total variance of the daily averaged fields. The results of this calculation, along with the kriged standard deviation field calculated using annual statistics, are shown in figure 4.9. The uncertainty calculated in this way is generally slightly lower for the daily statistical method, but only by a value of around 1 $\mu g \cdot m^{-3}$. It is possible to see reduced uncertainty, in the daily based method, at stations that are not represented in the annual calculations.

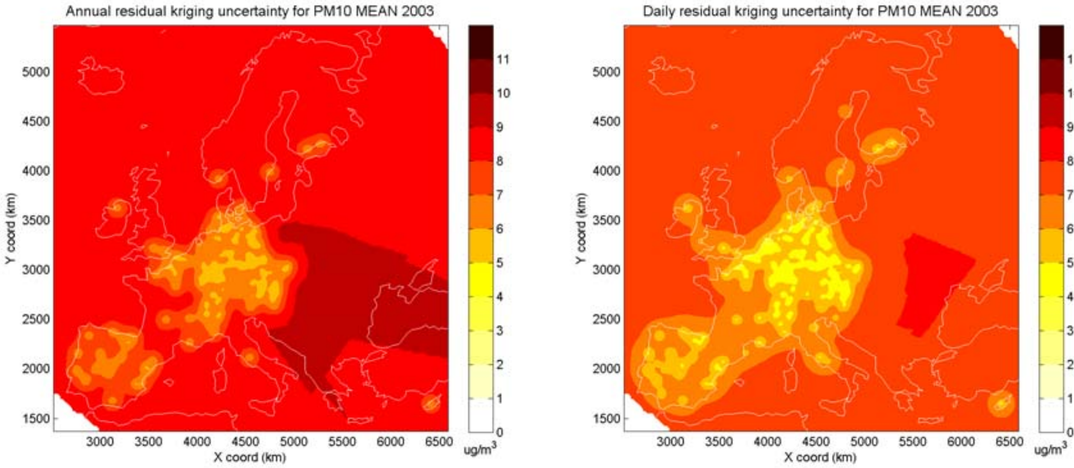


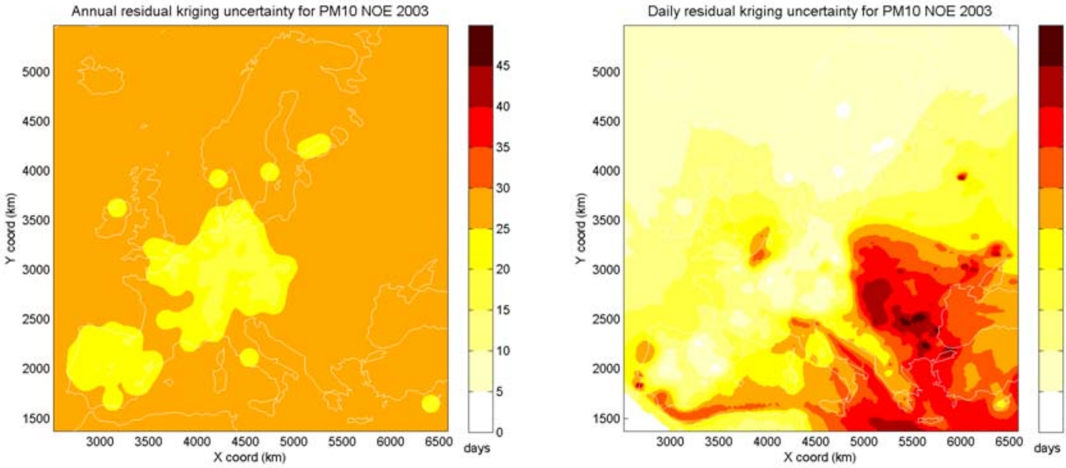
Figure 4.9 Showing the uncertainty maps (SD_{krig}) for the annual mean PM_{10} concentrations based on (left) annual statistics and (right) daily statistics. See text for detailed description of their derivation. The annual mean concentration maps related to these can be found in figure 4.6.

4.5.2 Uncertainty in the exceedance fields when using daily statistics

In order to compare the spatial uncertainty in the number of exceedance (NOE) days another, in this case more pragmatic, approach is required. From a statistical perspective the expected number of exceedances is the sum of the probability of exceedance for every individual day. It is in principle possible to calculate the probability of exceedance for every day if we know the probability density distribution of the daily mean concentration. If the probability density function is represented by a normal distribution we can use the residual kriging variance to represent the standard deviation and thus calculate the probability of exceedance for each day. If this is done in this fashion, then the final uncertainty in the NOE days will tend to be very small, since it assumes each probability to be uncorrelated (which is shown in section 4.5.1 not to be the case) and it does not take into account the question of spatial representativeness and bias.

Instead, the uncertainty in the expected number of exceedance days is calculated by adding up the individual probabilities of exceedance, as described above, but in addition adding and subtracting the annual mean variance, as shown in figure 4.9, to represent the representativeness and model error. The uncertainty in NOE days is then interpreted as being the maximum deviation, in number of days, from the plus and minus calculations. For example, at one spatial point the annual standard deviation is calculated to be 5 $\mu g \cdot m^{-3}$ and the expected NOE days at that point is calculated to be 20 days. By adding and subtracting 5 $\mu g \cdot m^{-3}$ from the daily mean concentrations used in the probability calculation it is found that adding gives 29 exceedances and subtracting gives 15 exceedances. The uncertainty in the NOE days is then given as 9 days. Using this methodology accounts for the threshold nature of exceedances, giving low uncertainty in NOE days when the daily mean values are well below the limit and giving high uncertainty in NOE days when the annual mean bias is uncertain and the daily mean values are close to the limit value.

The results of this calculation, as well as the kriged standard deviation field for NOE days calculated using annual statistics, are shown in figure 4.10. The annual uncertainty map gives a much more homogenous interpretation of the uncertainty, showing reduced uncertainty only in regions near observations. Such kriging is not actually suitable for mapping of this type when a threshold value is involved, and overestimates the uncertainty in areas where low numbers of exceedances occur. The daily based uncertainty map, on the other hand, shows low uncertainty in areas with low numbers of exceedances and in regions close to observations. High uncertainty is estimated in areas without observations and with large numbers of exceedances.


Annual residual kriging uncertainty for PM10 NOE 2003

Daily residual kriging uncertainty for PM10 NOE 2003

Figure 4.10 Showing the uncertainty maps (SD_{krig}) for the NOE days for PM_{10} based on (left) annual statistics and (right) daily statistics. See text for detailed description of their derivation. The NOE maps related to these can be found in figure 4.8.

4.5.3 Comments on the kriging semivariogram

The question also arises as to the applicability of the kriging assumptions and semivariogram models. One of the assumptions is that there is spatial correlation, as a function of lag distance, of the parameter to be interpolated. This is at the heart of the variance models used in the kriging methodology. Though this methodology has been fruitful in many geosciences, its application for air quality mapping is still open to debate. Investigation of the empirical semivariograms used in this study indicates a limited dependence of the variance with lag distance, figure 4.11, though this may vary from year to year. This brings into discussion the spatial representativeness of the observations since even at small distances the variance of the observations is quite high. Even if kriging methods are applied, the form of the semivariogram model should also be assessed. In this report the spherical model has been applied, based on initial sensitivity tests carried out in Horálek et al. (2005), but investigation of the daily mean variograms used in this study implies that other models may give better fits to the data on a daily basis, such as power law models. Alternative methods, eg. Blond et al. (2003), for determining the spatial variation of the covariance field may be more appropriate than the lag distance dependent method used in kriging. Such methods establish spatial covariance relationships based on analysis of a set of temporal data and creating functional relationships with model calculations.

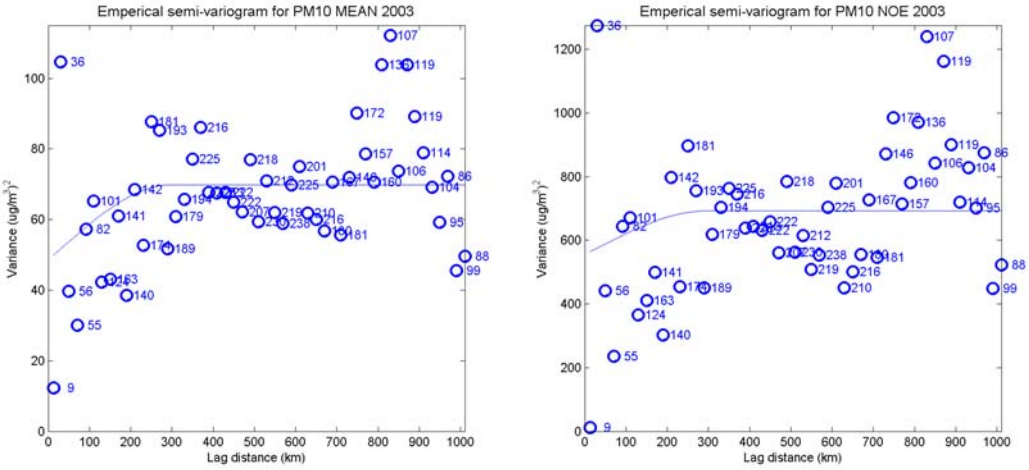


Figure 4.11 Showing the annual statistics residual semivariogram, and estimated sill (solid line), for annual mean PM$_{10}$ (left) and NOE days PM$_{10}$ (right), for the year 2003. The numbers indicate the number of station pairs used to calculate the variance.

4.5.4 Conclusions on uncertainty mapping

The estimated uncertainty in the annual mean rural PM$_{10}$ values, of 8 $\mu g{\cdot}m^{-3}$ or less (figure 4.9), should be considered quite reasonable, considering that spatial representativeness may contribute significantly to this value. Uncertainty in the percentile concentration and NOE days however, must be considered to be quite high, especially in areas where no measurements are available. The spatial variation of uncertainty in NOE days is also highly variable due to its threshold nature (figure 4.10). In many areas of Europe the uncertainty in NOE is too large to make the assessment useful for policy implementation. However, it must be noted again that representativeness is an important aspect of this uncertainty. The maps produced should indicate the mean concentration, or NOE, in a 25 x 25 km grid. Clearly there is a large variability within such a grid, as is indicated in figure 4.11 and the uncertainty discussed here will include that. This means, for example, that a significant part of the uncertainty in the NOE will not lie with the interpolation itself but with the variation of the observed concentration within the gridded region. More effort is thus required to assess this particular aspect of the interpolation uncertainty.

Further to this it should be noted that the use of kriging, or residual kriging, to interpolate NOE days using annual statistics is not recommended as it does not deal well with the threshold nature of this parameter and does not provide uncertainty maps that give a suitable spatial representation. If annual statistics are to be used to map exceedances then the 36th percentile concentration field is recommended, though this does not provide all the information that may be required for assessment. The use of daily means, on which the NOE and percentiles are based, is recommended when spatially interpolating the NOE days.

5. Conclusion and discussion

5.1 Assessment of the case study

This case study has studied a number of aspects connected to a basic data assimilation method, that being residual kriging using regression. It has extended the use of the method from annual statistics to its application to daily means for assessment of PM10 exceedances on the European scale. Both the annual mean and the percentile limit have been assessed. The two methods have been quantatively compared using the cross validation RMSE and the uncertainty has been spatially represented through the use of uncertainty maps. Methods for producing the uncertainty maps have been discussed and these differ according to the indicator and the method used.

The use of daily mean data has a number of advantages over the use of annual statistics. These are:

1. The number of observation days used is larger when using daily statistics than annual statistics when a limit on the allowable temporal coverage is applied to the stations.
2. The quality of the maps is as good as, and generally better than, maps produced using annual statistics, based on the cross validation RMSE.
3. There is consistency between the percentile and NOE fields.
4. The maps are more robust in regard to the use of automatic routines for defining interpolation parameters used in the kriging interpolation.
5. Future improvements in the interpolation based on multiple regression with other supplementary data, e.g. meteorological parameters, will be better represented on a daily basis.

The following disadvantages also exist:

1. The data and calculation requirements are significantly higher for the daily than for the annual statistics
2. The reduction in uncertainty may be small in regard to other possible improvements in the interpolation methodology, particularly for the case of annual means, that would be less data intensive
3. The interpretation of uncertainty mapping is more complex

The results of the case study indicates that the uncertainty in the assessment maps varies spatially and is dependent on the method used. For instance the use of the kriging variance as a spatial uncertainty indicator probably requires further refinement since it is based on the kriging semi-variogram model which is assumed to be applicable throughout the assessment domain. Special attention must also be given to the uncertainty assessment of the number of exceedances since this is based on a discrete limit value. The method applied to assess its spatial uncertainty when using daily mean values gives quite different results to the method used to assess the exccedance days when using the annual statistics. There is still a need for further refinement of the uncertainty mapping methods applied in this study.

Based on the cross validation RMSE the uncertainty in the mapped daily mean concentrations is around 12 $\mu g{\cdot}m^{-3}$. The resulting uncertainty in the annual mean concentrations is around 7 $\mu g{\cdot}m^{-3}$. The uncertainty in the number of exceedance days is found to be around 20 days. These uncertainties are the result of not just model uncertainty but also of spatial representativeness. I.e. on the spatial scales addressed (25 km) the variability of point measurements within that space is of a similar order as these values. Model improvements through process descriptions may improve on the bias in the models, which is significant for PM10, but they may not improve significantly on the assimilated uncertainty due to the spatial representativeness. Increased spatial resolution may help to address this issue.

It was shown that the kriging variograms, used to define the spatial covariance for the residual kriging assimilation, are not well defined. Alternative methods, eg. Blond et al. (2003), for determining the spatial variation of the covariance field may be more appropriate than the lag distance dependent method used in kriging. Such methods establish spatial covariance relationships based on analysis of a set of temporal data and creating functional relationships with model calculations.

The methods presented here can be assessed using standard software applications available to city and European authorities and so represent operational methods for improving the spatial assessment of pollutants.

5.2 Improvements in assessment derived from case study

This case study has brought improvements in the following areas.

1. Has furthered assessed a basic data assimilation method and applied it at higher temporal resolutions to assess the importance of this method
2. Has clearly demonstrated that the combination of monitoring and modelling data leads to improved assessment maps
3. Has discussed and suggested methods for producing uncertainty maps attached to these concentrations fields and has produced uncertainty maps to help visualise the improvement obtained using the differing assimilation methods
4. Has assessed the improvement through the cross validation RMSE

5.3 Recommendations resulting from the case study

A number of recommendations resulting from this case study can be identified.

1. It is recommended to carry out the assimilation on the same temporal scale for which the limit values apply. For the case of PM10 the assimilation should be carried out on daily mean concentrations rather than on annual statistics of the percentiles.
2. Basic assimilation methods such as linear regression, scaling or correction of bias using observations can significantly improve model assessment and their associated uncertainty.
3. Pure kriging of observational data alone can give statistically better results than models but the uncertainty in these is very high, especially far from the observations. This uncertainty far from stations is clearly reflected in the kriging parameter of sill and uncertainty maps should be used to indicate this.
4. Estimates of uncertainty and/or uncertainty maps should be co-produced with the assessment map to give a clear indication of their validity
5. Standard deviation is a useful parameter for indicating spatial uncertainty.
6. Effort should be made to understand the sources and form of uncertainty so that the best possible uncertainty map can be produced.

5.4 Suitability for implementation on other scales

The methodologies described in this case study have been applied to the European scale. They have also been applied to the regional scale (Blond et al., 2003) and to the urban scale for Prague (Air4EU – CS D7.1.6). As was shown in the Air4EU Prague case study the application of the residual kriging method is limited by the spatial scale and spacing of the stations. When concentrations vary significantly on scales smaller than the station spacing then residual kriging will not be a useful method. In general though the use of regression analysis is applicable on all scales, as long as the model and observations are spatially representative of the same scales.

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