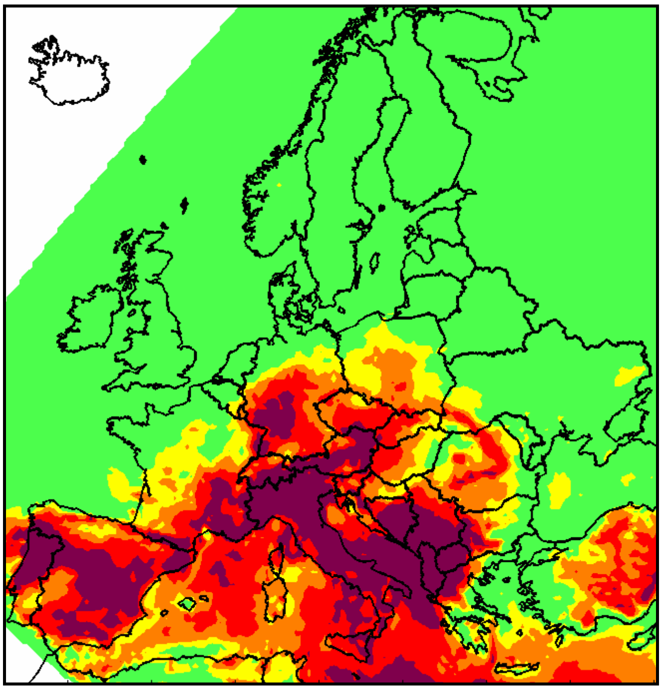
European scale exceedance mapping for PM10 and ozone based on daily interpolation fields



ETC/ACC Technical Paper 2007/8

January 2008

Final version

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European Topic Centre on Air and Climate Change

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Cover picture

Probability of the exceedance of the EU target value for ozone in the year 2005. Regions in red have a probability of more than 60% that they are in exceedance. Those in purple have a probability of more than 80%. Picture extracted from figure 20 in the report.

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Executive summary

This is the second of two reports written by the European Topic Centre for Air Quality and Climate Change as part of the delivery to the European Environmental Agency within the task 5.3.3.1 (2007). Within this task methodologies are being developed for the spatial mapping, on a European scale, of air quality indicators related to the European directives on air quality. These maps are constructed using air quality monitoring data, atmospheric chemical transport model calculations and other supplementary land use and meteorological data. The methods used for the mapping involve kriging, multiple linear regression and a combination of these.

In the first of these two reports focus is given to the further refinement and production of air quality indicator maps for Europe in 2005 using the methodologies established in previous reports, as well as the further assessment of uncertainty in the maps. For these mapping techniques annual statistics are used to create a single map of the appropriate air quality indicator.

In this, the second report, focus is given to the use of daily statistical data, i.e. creating maps on a daily basis, and combining these daily maps to derive the air quality indicators. For the current report the indicators assessed are a) the annual mean PM_{10} concentration, b) the number of days in exceedance of the limit value of 50 μgm^{-3} for PM_{10} and c) the number of days in exceedance of the target value of 120 μgm^{-3} for the maximum daily 8 hour running mean for ozone.

A number of interpolation methods are tested and the use of a variety of supplementary datasets, including both the EMEP and the LOTOS-EUROS chemical transport models, is assessed. The results of the daily mapping methods are compared to those from the annual mapping in regard to the statistical performance of the methods as well as the methods available for determining uncertainty.

It is found that residual kriging after multiple linear regression is the best performing interpolation methodology for all indicators on both a daily and an annual basis. This is in accordance with earlier studies using annual statistics. In addition, the use of daily mapping is generally found to perform better than the annual maps for the majority of years and indicators studied. Both of the chemical transport models applied give similar results but the LOTOS-EUROS model is found to perform best for PM_{10} with the EMEP model performs best for ozone.

As a result of this study it is recommended to use daily mapping methods when the number of exceedance days are to be mapped. This provides a physically consistent methodology for determining the exceedances and also provides an improved methodology for assessing the uncertainty of the maps. If annual statistics are to be used for mapping daily exceedances then it is recommended to use the percentile value, i.e. the n'th highest daily concentration, as the indicator rather than the number of exceedance days.

When annual means are to be mapped both the assessment and uncertainty maps using the daily and annual methods give very similar results, though the daily mapping is slightly better. Mapping of these indicators can be carried out using the annual statistics only.

1 Introduction

This is the second of two reports written by the European Topic Centre for Air Quality and Climate Change as part of the delivery to the European Environmental Agency within the task 5.3.3.1 (2007). Within this task methodologies are currently being developed for the spatial mapping, on a European scale, of air quality indicators related to the European directives on air quality.

In the first of these two reports (Horalék et. al., 2008) focus is given to the further refinement and production of air quality indicator maps for Europe using annual statistics from observational data, air quality models and relevant supplementary data and builds upon earlier reports from Horalék et. al. (2005; 2007).

In this second report the focus is on the use of daily statistical data, i.e. creating interpolated fields on a daily basis and combining these fields to derive the air quality indicators. This report builds upon the methodology described in Horálek et al. (2007) chapter 6 wherein the methodology was shown to improve interpolation of annual mean and number of exceedance (NOE) days for PM_{10}, when compared to the use of annual statistics, for the year 2003. In this current study the methodology is further extended and assessed.

1.1 Aim and scope of the report

This study will address the following points in regard to the daily mapping of concentration fields.

- A comparison of a number of different interpolation methods, including an alternative optimised weighting of kriging and model regression fields
- A comparison of the years 2003, 2004 and 2005 for PM_{10}
- Application of the methodologies to ozone exceedances for 2005
- A comparison of the EMEP and LOTOS-EUROS models for PM_{10} and ozone
- A comparison of the use of daily or annual statistics
- Presentation of maps of concentrations, as well as uncertainty and probability of exceedance maps
- Draw conclusions concerning best or most preferable methods and provide recommendations for future work

In the methodology employed only AirBase stations (AirBase, 2007) classified as rural background have been applied and the maps are made using a 25 x 25 km resolution grid, as described in Horálek et al. (2007) chapter 6. Two air quality models are used; the Unified EMEP model (Simpson et al., 2003) and the LOTOS-EUROS model (Schaap et al., 2007). This is in contrast to previous studies where on the EMEP model was available.

Interpolations of daily mean and annual mean data are applied to determine the following:

- PM_{10} exceedances of annual mean limit value (annual mean concentrations > 40 μgm^{-3}) and daily mean limit value (days with daily mean concentrations > 50 μgm^{-3}) for the 3 year period 2003-2005 are assessed
- Ozone exceedances of target value (days with a maximum 8 hour running mean > 120 μgm^{-3}) for the year 2005 are assessed

1.2 Interpolation methods

A number of different interpolation methods are tested and applied, though not all are applied on all data, and statistically analysed for both PM_{10} and ozone. These methods include the following:

1. Model and regression
    a. Pure model

    b. Linear regression of the model with observations
    c. Multiple linear regression of the model and altitude with observations
2. Kriging methods on station observations
    a. Ordinary kriging using fitted variograms
    b. Ordinary kriging using optimised variograms
    c. Lognormal kriging using fitted variograms
    d. Lognormal kriging using optimised variograms
3. Residual kriging methods
    a. Residual kriging of the observations after model regression (1.b) using fitted variograms
    b. Residual kriging of the observations after model regression (1.b) using optimised variograms
    c. Residual kriging of the observations after model and altitude regression (1.c) using fitted variograms
    d. Residual kriging of the observations after model and altitude regression (1.c) using optimised variograms
4. Weighted combination of model regression and observations kriging fields
    a. Weighted combination of fitted ordinary kriging (2.a) with regression (1.b or 1.c)
    b. Weighted combination of optimised ordinary kriging (2.b) with regression (1.b or 1.c)

1.3 Methodology

The above interpolation methods are applied to both

1. The EMEP model
2. The LOTOS-EUROS model

The methods are compared using

1. Daily interpolations to determine annual mean concentration and NOE days
2. Interpolation using annual statistics of annual mean and NOE days

An analysis is carried out of the results using cross-validation statistics of

1. Root mean square error (RMSE)
2. Mean absolute error (MAE)
3. Coefficient of determination (R^2)
4. Regression intercept and slope

These statistics are applied to:

1. Daily mean concentrations (PM_{10}) or daily maximum 8 hour running mean (O_3) for the entire assessment year
2. Annual mean concentrations
3. NOE days

Results are presented in bar charts and in tables for all years. Maps showing the results, and their uncertainties, are presented for the year 2005 only.

2 Summary description of the interpolation methodologies

In this section the methodologies applied are briefly described. For more details concerning these methodologies the reader is referred to Horálek et al. (2007), particularly chapter 6.

1.a. Pure model: This is based on the model results themselves with no further changes. Linear spatial interpolation of the model is used to determine model concentrations at monitoring sites

1.b. Linear regression of the model with observations: This methodology takes model calculations at monitoring sites and carries out a linear regression analysis. The determined regression coefficients, slope and intercept, are further used to create new maps. To avoid unrealistic results the following 2 criteria are applied for calculation of the regression coefficients. If the intercept is found to be < 0 the regression is reanalysed by setting the intercept to 0. If the coefficient of determination (R^2) is < 0.1 then the regression slope is set to 1 and the intercept calculated. The first is to avoid the possibility of negative values occurring in the regression model and the second is to avoid poorly defined regression fits.

1.c. Multiple linear regression of the model and altitude with observations: This methodology takes model concentrations and altitude at monitoring sites and carries out a multiple linear regression analysis against observations. The determined regression coefficients are further used to create new maps, using mean altitude from topographic maps of Europe at 25 km resolution. To avoid unrealistic results the following 3 criteria are applied. The regression slope for altitude must be positive for O_3 and negative for PM_{10}. If the intercept is found to be < 0 the regression is reanalysed by setting the intercept to 0. Altitude is only used in the multiple regression if the correlation coefficient for altitude regression is > 0.05, otherwise only model regression with observations is used subject to the criteria stated in method 1b.

2.a. Ordinary kriging of observations using fitted variograms: This is the standard ordinary kriging methodology where the variogram parameters of nugget, sill and range are determined by fitting all the available observational data with a spherical variance model. Only the 50 nearest stations are used in the interpolation and the maximum allowable range for the fit is 1000 km.

2.b. Ordinary kriging of observations using optimised variograms: This methodology is similar to 2.a. above but optimises the kriging parameters of nugget:sill ratio and range to obtain the minimum cross-validation RMSE. This is achieved by mapping the nugget:sill ratio in steps of 0.1, from 0 – 1, and the range using 100 km steps, from 100 km to 1000 km. Of the set of 100 variograms produced the variogram that provides the minimum cross-validation RMSE is used for the interpolation.

2.c. Lognormal kriging of observations using fitted variograms: This is the same as method 2.a above but the observations are first transformed using their natural logarithms

$$C_{\log} = \ln(C) \tag{1a}$$

before kriging and then back transformed using

$$C = \exp(C_{\log} + 0.5\sigma^2) \tag{1b}$$

as described in Cressie (1993). In the above back transformation σ^2 is taken to be the predicted kriging variance at that point in space.

2.d. Lognormal kriging of observations using optimised variograms: This is the same as method 2.b above but the observations are first transformed using their natural logarithms, equations 1a and 1b. The optimisation for cross-validation is made on the concentrations, not the logarithm.

3.a. Residual kriging of observations after model regression using fitted variograms: This method uses fitted ordinary kriging (2.a) to interpolate the residual (observations minus regression) determined at the monitoring sites. The regression used here is 1.b as described above.

3.b. Residual kriging of observations after model regression using optimised variograms: This method uses optimised ordinary kriging (2.b) to interpolate the residual (observations minus regression) determined at the monitoring sites. The regression used here is 1.b as described above.

3.c. Residual kriging of observations after model and altitude regression using fitted variograms: This method uses fitted ordinary kriging (2.a) to interpolate the residual (observations minus regression) determined at the monitoring sites. The regression used here includes altitude (1.c) as described above.

3.d. Residual kriging of observations after model and altitude regression using optimised variograms: This method uses optimised ordinary kriging (2.b) to interpolate the residual (observations minus regression) determined at the monitoring sites. The regression used here includes altitude (1.c) as described above.

4.a. Weighted combination of fitted ordinary kriging with regression: As an alternative to residual kriging a weighted combination of the fitted kriging (2.a) and the model regression (and/or altitude) is made. The weighting is based on the Bayesian principle that given the uncertainty (σ) in two values, in this case observed $O(x,y)$ and modelled $M(x,y)$ concentration fields, it is possible to find the optimal combination of these $C(x,y)$ using

$$C(x,y) = \frac{\sigma_O^2 M(x,y) + \sigma_M^2 O(x,y)}{\sigma_O^2 + \sigma_M^2} \qquad (2)$$

The uncertainty in the observational field (σ_O) is given by the square root of the kriging spatial variance and the model regression uncertainty (σ_M) is described by an error model defined as

$$\sigma_M(x,y) = \sqrt{\sigma_A^2 + \sigma_R^2 M(x,y)^2} \qquad (3)$$

where σ_A is an absolute uncertainty and σ_R a relative uncertainty. These model uncertainty parameters are difficult to determine so they are determined, in a similar way to the optimised kriging, by finding the minimum cross-validation RMSE by mapping in σ_A and σ_R space. This is done by stepping through the range of σ_R = 0 – 4 and σ_A = 0 – 4 x *mean*(M) in steps of 0.1 and 0.1 x *mean*(M) respectively. The values of σ_A and σ_R that provide the minimum cross-validation RMSE is used to determine the weighting.

4.b. Weighted combination of optimised ordinary kriging with regression: This is the same as method 4.a described above but uses the optimised kriging fields (2.b) instead of the fitted kriging fields.

5. Assessment of the spatial variability from observations: In addition to the above interpolation methods an assessment is made of the spatial variability of the observations to indicate how this will affect the statistical analysis. This variability will reflect the uncertainty related to spatial representativeness. It is assessed by taking the mean, centred at each of the grid squares, of all observations that are within a 25 km radius of that grid centre. When more than 1 observation is available within this radius then the associated error is calculated, treating the mean of the grid as if it was the interpolation value. In this way similar statistics can be made as for the other interpolation methods and these should give an indication of the ‘best’ possible statistics that could be achieved by the interpolation. Though the grid resolution is 25 x 25 km a larger spatial region covering a 50 km diameter has been chosen for this assessment since only 4 – 6 station pairs lie within a 25 km diameter area of each other (for PM_{10}), which would not provide a representative sample. Choosing a larger area will tend to overestimate the spatial variability but it will still provide indicative quantitative information.

3 Results of the interpolation methods for daily mean PM_{10} concentrations

In this section results are presented of the following

- A comparison of the daily interpolation methods described in Section 2
- A comparison of the years 2003, 2004 and 2005
- A comparison the EMEP and LOTOS-EUROS models
- A comparison of the use of daily or annual statistics
- An assessment of the daily and annual variability of the best interpolation method
- Presentation of maps and an assessment of uncertainty

3.1 Comparison of daily statistical methods for the EMEP and LOTOS-EUROS models

In this section bar graphs showing the RMSE of the daily mean, correlation coefficient (R^2) of the daily mean and the RMSE of the NOE days are presented for the different methods described in Section 2 and compared for the two air quality models used. The statistics of RMSE and R^2 are calculated using all the daily means for all the available monitoring stations. The NOE days are calculated for the available stations only. The results are shown in figures 1 - 3 for the 3 year period. The tabulated results can be found in Annex I.

The following comments and conclusions can be made concerning the methods applied

1a: Both models show poor correlation and a significant negative bias (see ANNEX table A.1) for all the years. The EMEP model generally provides the best correlation and the LOTOS-EUROS model the lowest RMSE.

1b: Linear regression of the model results removes a large part of the bias, reducing RMSE and increasing correlation. After regression the LOTOS-EUROS model performs slightly better than the EMEP model indicating that it is capturing the spatial distribution, if not the magnitudes, of the concentrations better.

1c: Linear regression of both the model and altitude provides slightly improved results for the daily mean values but no significant reduction in the NOE day RMSE.

2a-b: Optimised kriging of the observations provides a clear improvement on the fitted kriging in regard to the daily mean values, but less reduction in the NOE day RMSE, and significantly better results than for the model regression methods (1b and 1c).

2c-d: Lognormal kriging of the observation concentrations does not lead to significant improvement of the daily mean PM_{10} concentrations and actually increases the RMSE for the NOE days for most years. Only for the year 2005 does lognormal kriging improve the RMSE. As in ordinary kriging the optimisation of the variogram, instead of the use of the fitted variogram, improves the results.

3a-b: Residual kriging was only carried out using the linear regression of the model, not including the altitude. This was done due to the small, or lack of improvement in the case of NOE days, in the results when including altitude in the regression. Optimised residual kriging can be seen to give the best results of any of the interpolation methods and this is true for both daily mean and NOE day calculations. The LOTOS-EUROS model provides a slightly better interpolation than does the EMEP model for all years.

4a-b: The weighted combination of kriging and linear regression model provides daily mean statistics only second to the optimised residual kriging (3b), when the optimised kriging is used (4b). However, it performs significantly worse in regard to the NOE days, no better than the optimised kriging, on which it is based. On a daily basis the degree to which the model and observational fields are weighted

(equation 2) varies in both time and space. For some of the days the model is given significant weighting whilst for other days the combined field is based almost exclusively on the kriging field.

5. The calculated spatial variability helps to indicate the best possible statistic achievable for the interpolation. This is based however on a limited sample of stations that cannot be used as an absolute limit. Even so it can be seen that the interpolations are approaching this limit for all years and it is reasonable to ask how much improvement can still be achieved through further refinement of the interpolation method.

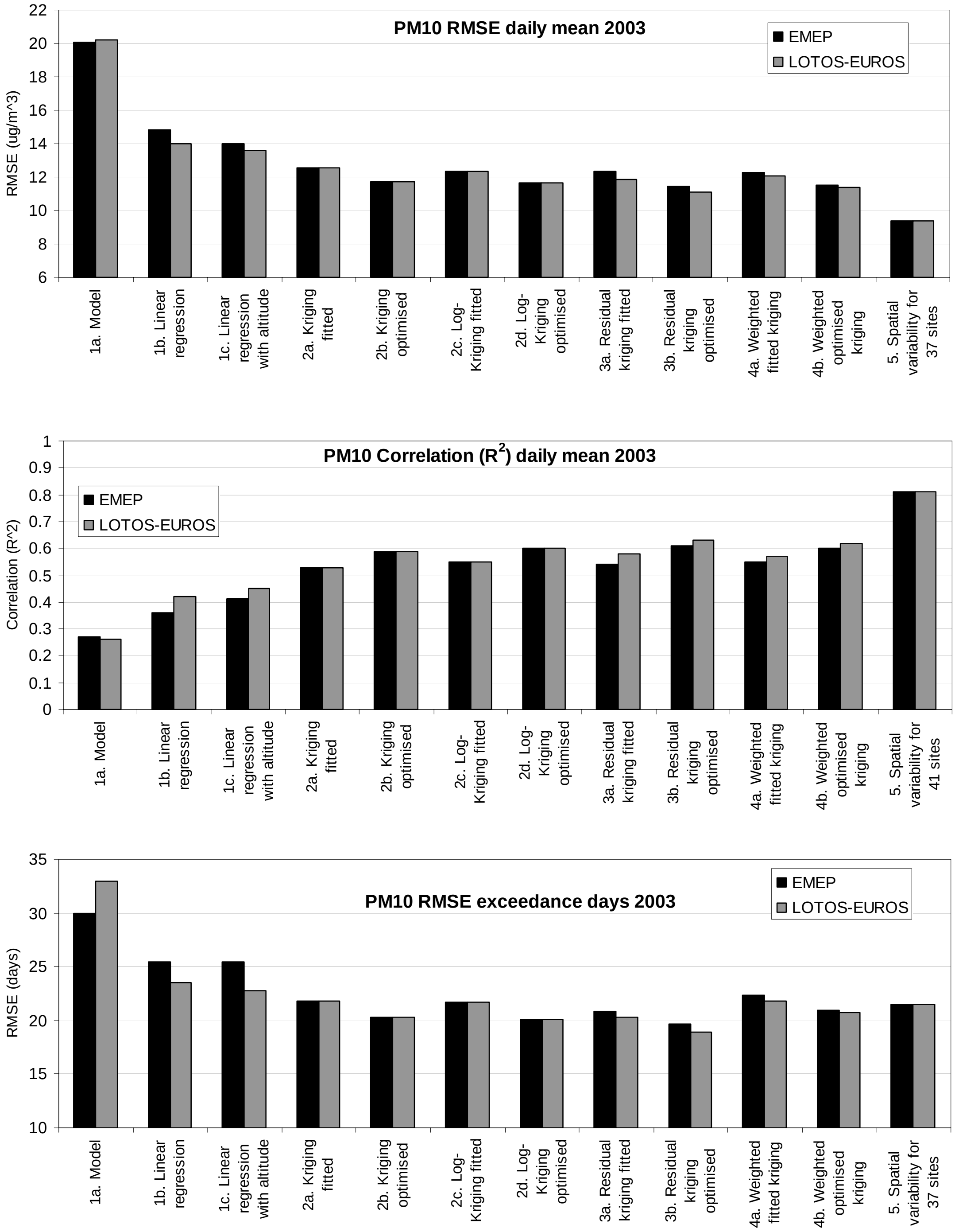


Figure 1. Graphs showing the cross-validation daily mean concentration RMSE, daily mean correlation and NOE days RMSE for PM_{10} using the interpolation methodologies described in the text for the year 2003. Shown are the results of the comparison between the EMEP and LOTOS-EUROS models. Results for the kriging interpolations and spatial variability are the same for both models.

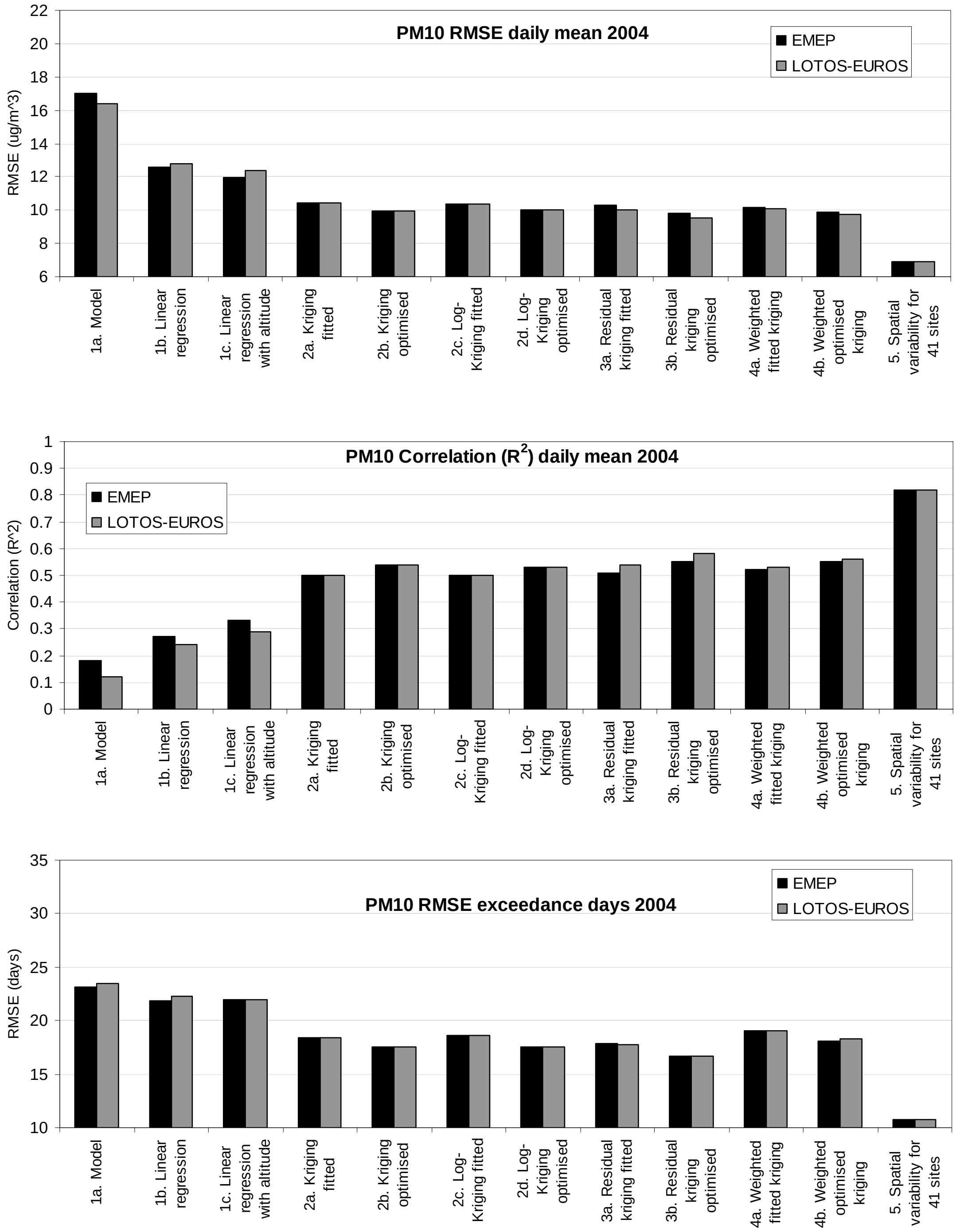


Figure 2. Graphs showing the cross-validation daily mean concentration RMSE, daily mean correlation and NOE days RMSE for PM_{10} using the interpolation methodologies described in the text for the year 2004. Shown are the results of the comparison between the EMEP and LOTOS-EUROS models. Results for the kriging interpolations and spatial variability are the same for both models

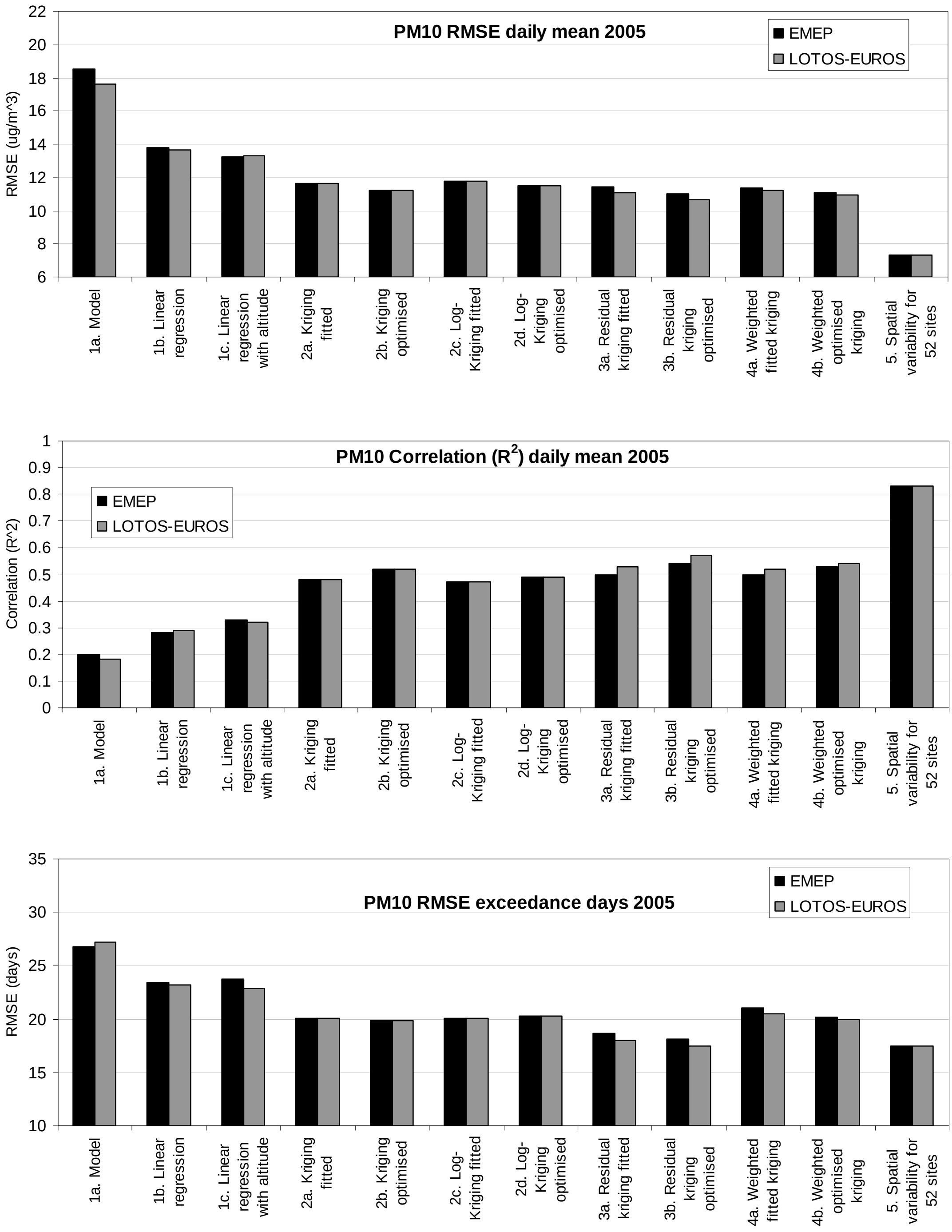


Figure 3. Graphs showing the cross-validation daily mean concentration RMSE, daily mean correlation and NOE days RMSE for PM_{10} using the interpolation methodologies described in the text for the year 2005. Shown are the results of the comparison between the EMEP and LOTOS-EUROS models. Results for the kriging interpolations and spatial variability are the same for both models

3.2 Comparison of the interpolation methods using daily and annual statistics

In this section bar graphs showing the RMSE of the annual mean, correlation coefficient of the annual mean and the RMSE of the NOE days are presented that compare the results using the daily statistics (section 3.1) with the calculations using annual statistics (Annex table A.2). The aim is to see if significant improvement is obtained for calculating annual mean and NOE day values when using the daily interpolations in comparison to direct interpolation of these annual statistics. This is presented for the LOTOS-EUROS model only, which was shown in section 3.1 to provide the best interpolation results. The EMEP model provides very similar relative results and is not presented. The results are shown in figures 4 - 6.

In order to make the comparison a selection is made on the stations so that only stations with a daily temporal coverage >75% are used in the comparison. The statistics are then recalculated based on these stations only.

The following comments and conclusions can be made concerning the methods applied.

1a: The model results are necessarily the same for both annual and daily statistics. They show, as previously mentioned, a large negative bias (Annex table A.2) which is reflected in the RMSE.

1b: Linear regression of the model removes a large part of the bias. The resultant statistics are very similar for both the daily and annual statistics in regard to the annual mean but for 2004 and 2005 the NOE days RMSE is significantly less using the annual statistics.

1c: Linear regression using both the model and altitude provides slightly improved results. The resultant statistics are very similar for both the daily and annual statistics. The most significant improvement occurs in the annual mean correlation coefficient but there is little improvement in the NOE days.

2a-b: In section 3.1 a significant improvement was found when using optimised kriging instead of fitted kriging for the daily interpolations. A less significant improvement is found when using the annual statistics. In regard to optimised kriging, the annual means are best interpolated using the daily statistics. However, in regard to NOE days the best interpolation method is using the annual statistics.

2c-d: Log-normal kriging of the observation concentrations gives slightly lower RMSE for the years 2003 and 2004, when compared to ordinary kriging, but gives poorer results for 2005. This is the case for both the daily and the annual statistics.

3a-b: Residual kriging was only carried out using the linear regression of the model, not including the altitude. This was done due to the small, or lack of, improvement in the case of NOE days, in the results when including altitude in the regression. Optimised residual kriging can be seen to give the best results of any of the interpolation methods. For the annual mean concentrations the use of daily interpolations gives better results than the annual interpolations. However, the results are mixed for the NOE days. In 2003 the best result is found using the daily interpolations, for 2005 the best result is found using the annual interpolation. In 2004 both interpolation methods give very similar results.

4a-b: The weighted combination of kriging and linear regression model provides annual mean results second only to the optimised residual kriging. However, it performs significantly worse in regard to the NOE days, no better than the optimised kriging, on which it is based. This methodology also provides better estimates of NOE days for all the years using the annual statistics, rather than the daily ones.

5. The calculated spatial variability helps to indicate the best possible statistic achievable for the interpolation. This is based however on a limited sample of stations that cannot be used as an absolute limit. Even so it can be seen that the interpolations are approaching this limit for all years and it is questionable how much improvement can still be achieved through further refinement of the interpolation method.

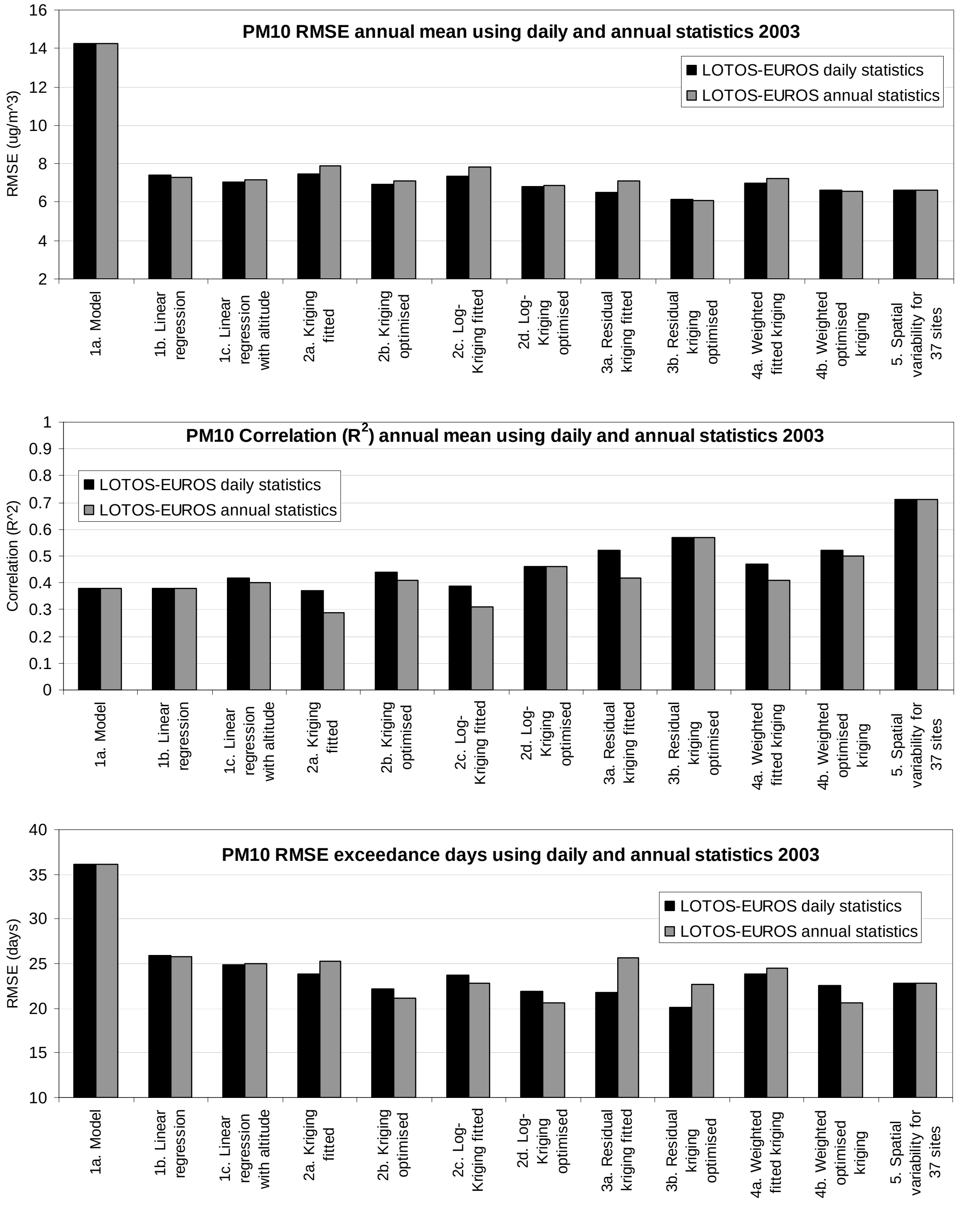


Figure 4. Graphs showing the cross-validation RMSE, correlation and NOE days for PM_{10} (number of days with daily mean concentrations > 50 μgm^{-3}) using the interpolation methodologies described in the text for the year 2003. Shown are the results for the comparison between interpolations using daily or annual statistics. Only results using the LOTOS-EUROS model are shown. Results from the model are the same in both cases.

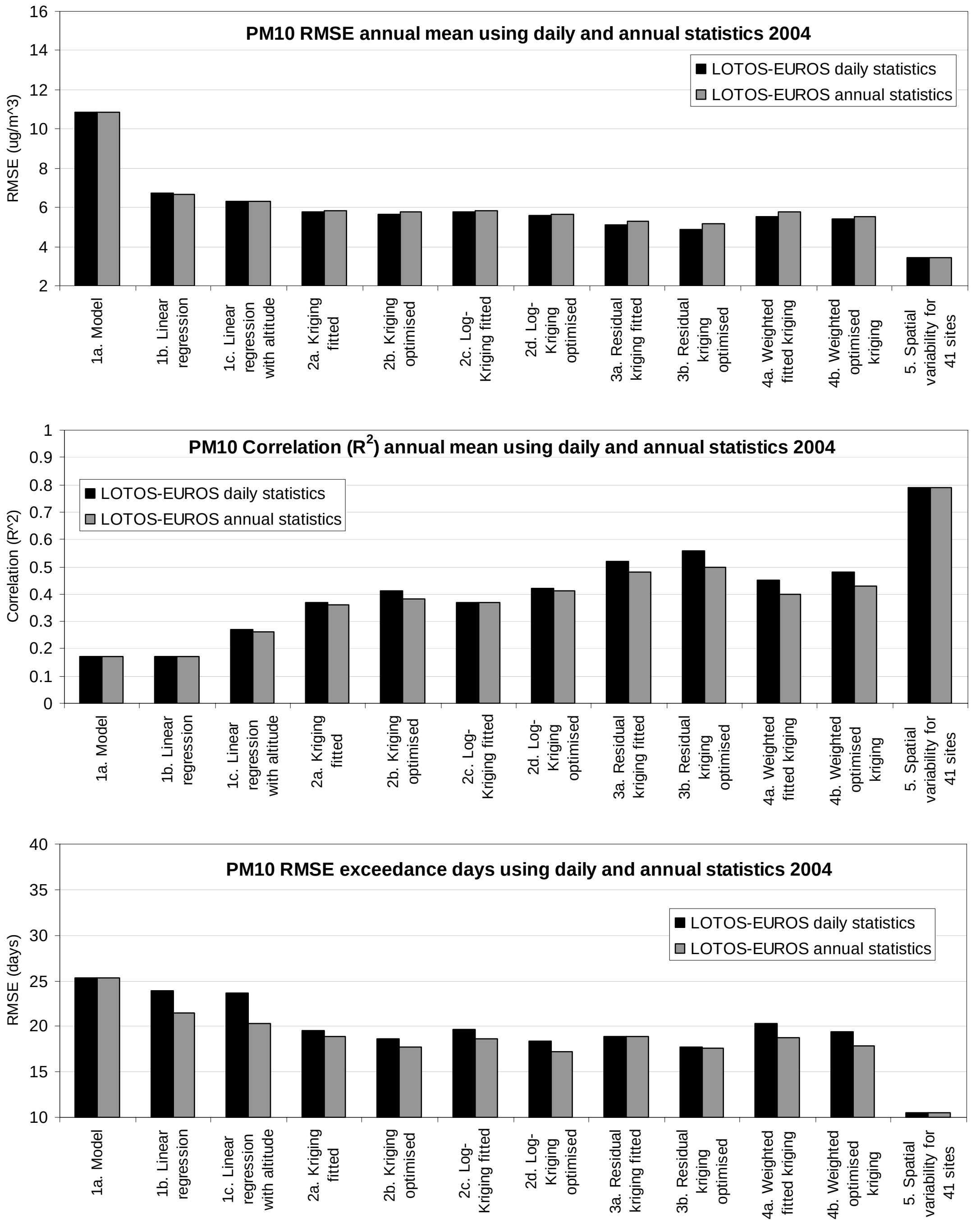


Figure 5. Graphs showing the cross-validation RMSE, correlation and NOE days for PM_{10} (number of days with daily mean concentrations > 50 μgm^{-3}) using the interpolation methodologies described in the text for the year 2004. Shown are the results for the comparison between interpolations using daily or annual statistics. Only results using the LOTOS-EUROS model are shown. Results from the model are the same in both cases.

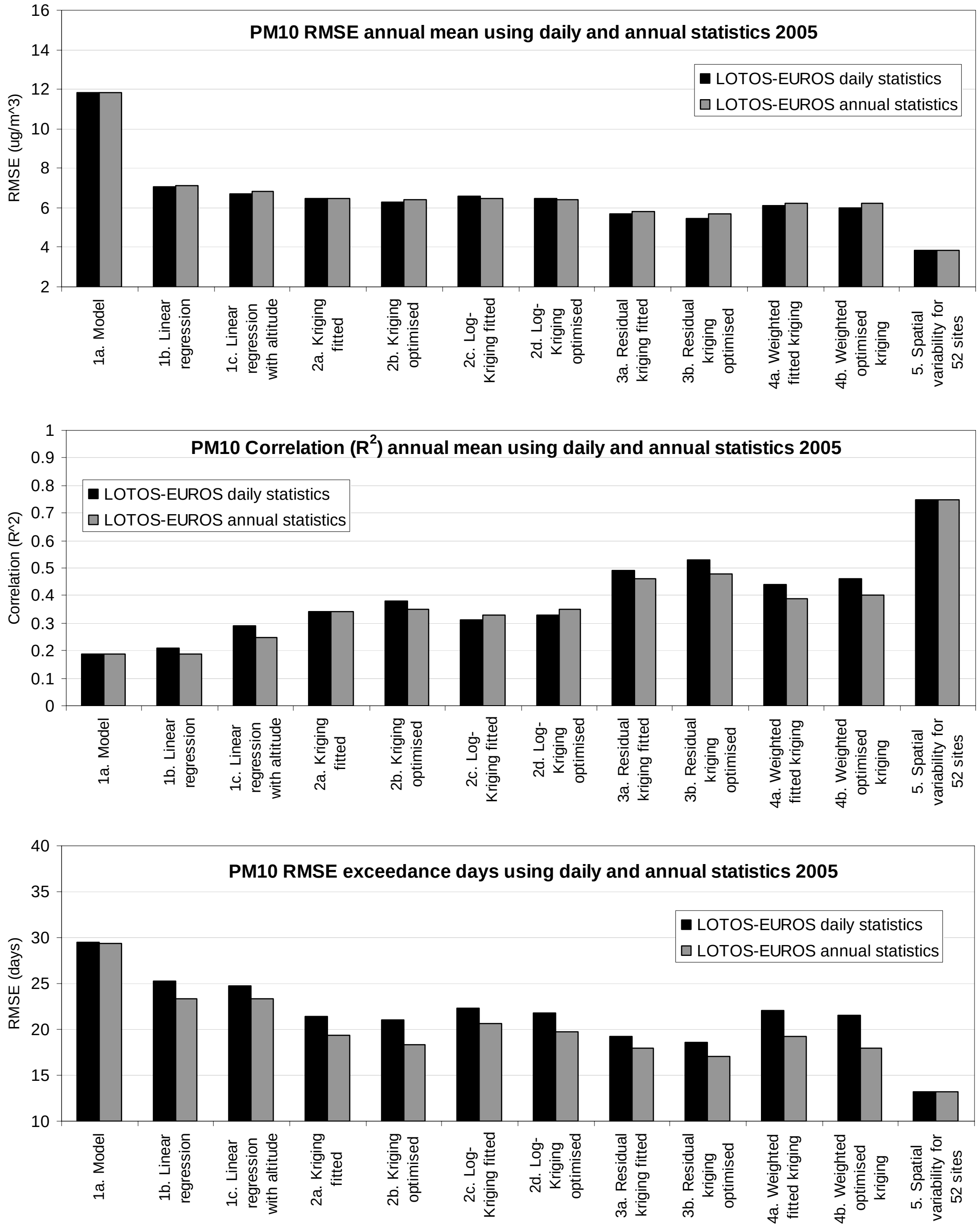


Figure 6. Graphs showing the cross-validation RMSE, correlation and NOE days for PM_{10} (number of days with daily mean concentrations > 50 μgm^{-3}) using the interpolation methodologies described in the text for the year 2005. Shown are the results for the comparison between interpolations using daily or annual statistics. Only results using the LOTOS-EUROS model are shown. Results from the model are the same in both cases.

3.3 Daily and inter-annual variability of the residual kriging method

In sections 3.1 and 3.2 the RMSE and correlation coefficient are presented for a number of different interpolation methods using 2 different air quality models and using both daily and annual statistics. In this section we briefly look at the variability, from day to day and year to year, of these statistics for the 'best' method, that being optimised residual kriging after regression using the LOTO-EUROS model and daily statistics (method 3.b).

In figure 7 the daily variability of the normalised cross-validation RMSE, normalised with the mean of the observations on that day, is presented for the years 2003 – 2005. As can be seen all years follow a very similar trend where the normalised RMSE is largest during the winter and lowest during the summer. During the winter the spatial variation of the daily mean observations is larger than during the summer (Horalék, 2007, chapter 6) indicating the increased heterogeneity of PM_{10} during the winter period.

There are also a number of peaks in the daily data. This is known, at least for the case in 2003 on day 150, to be the result of poor quality observational data. In that case there were 3 days in a row at a particular station with a concentration of exactly 500 μgm^{-3}. As indicated in Section 2, no quality control of the AirBase data has been undertaken in this study.

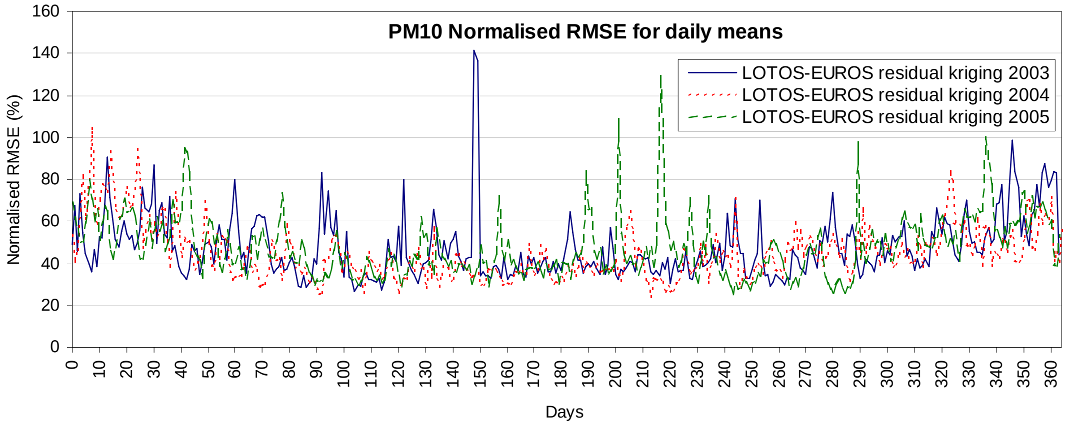


Figure 7. Plot showing the normalised cross-validation RMSE (%) for the years 2003 – 2005 using residual kriging with linear regression (3b) and the LOTOS-EUROS model. Each day is normalised using the mean observed concentrations from all stations on that day.

It is also interesting to compare the inter-annual variation of the statistics to see how the interpolation is performing for the various statistical parameters from year to year. These statistics are shown in table 1 for both the daily mean concentrations and the annual mean concentrations, where once again only the results from the residual kriging (method 3.b) are shown. The year 2003 has the highest mean concentrations and also highest RMSE but when the RMSE is normalised with the mean concentrations we find that for each year the interpolation is giving quite similar results. This is the case for both the daily mean and annual mean statistics.

Table 1. Comparison of cross-validation statistics for the years 2003 – 2005 using method 3b (optimised residual kriging with the LOTOS-EUROS model) based on daily interpolations. Shown are the statistics for the daily mean concentrations and the annual mean concentrations.

Statistic	**2003**	**2004**	**2005**
Observed mean concentration, all stations (μgm^{-3})	23.5	19.9	21.4
Average number of stations available for interpolation	167	181	209

Daily mean concentration cross-validation statistics			
Daily mean RMSE (μgm^{-3})	11.1	9.5	10.6
Daily mean normalised RMSE %	47.2	47.7	49.5
Daily mean correlation (R^2)	0.61	0.55	0.54
Annual mean concentration cross-validation statistics			
Annual mean RMSE (μgm^{-3})	6.1	4.9	5.5
Annual mean normalised RMSE %	25.9	24.6	25.7
Annual mean correlation (R^2)	0.57	0.56	0.53

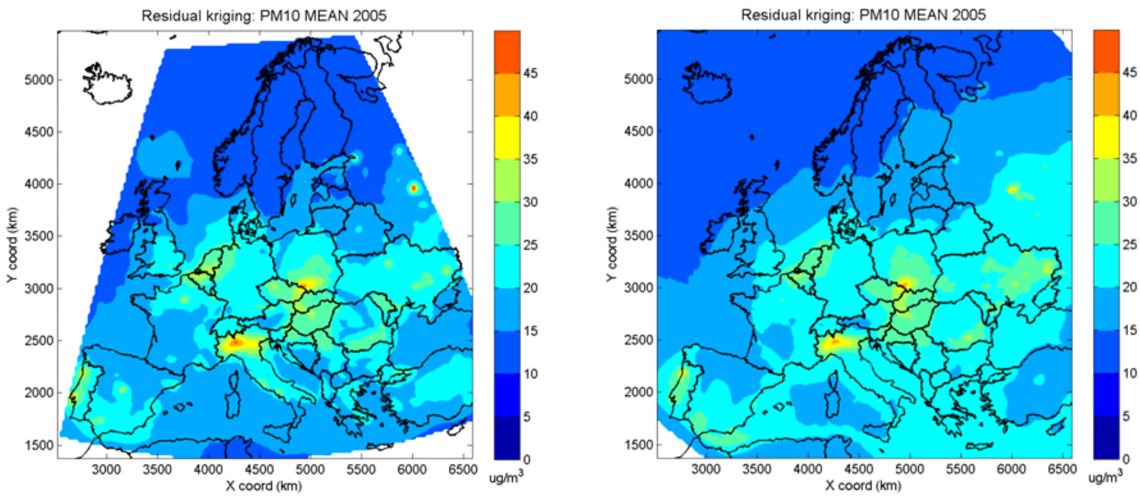
In conclusion we can state:

- The 'best' interpolation method (optimised residual kriging with linear regression, 3b) is found to give consistent statistical performance from year to year.
- Within each of the 3 year periods studied this method also consistently shows higher normalised RMSE in the winter and lower RMSE in the summer reflecting the increased heterogeneity of the spatial distribution of PM_{10} during the winter period.

3.4 Maps of annual mean and number of exceedance days

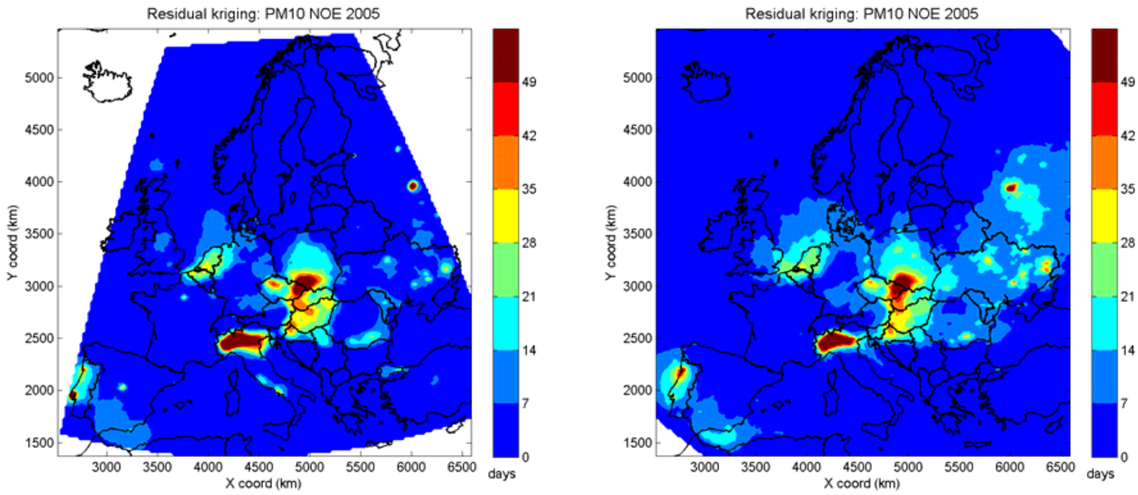
Maps showing the annual mean concentration of PM_{10} and the NOE days for the year 2005 are presented here. These maps have been determined using the daily interpolations and the optimised residual kriging methodology with linear regression of the model (3.b), selected as best interpolation method. Maps created using both the EMEP and the LOTOS-EUROS model are presented. Along with these maps uncertainty maps are also presented.

Visually the differences between the maps are small. The main differences occur in the regions far from observations, e.g. the far eastern side of Europe. In these areas the models provide most of the information.



LOTOS-EUROS EMEP

Figure 8. Maps showing the calculated rural background annual mean concentration of PM_{10} in 2005, as calculated using the optimised residual kriging method (3b) described in the text. Left is the LOTOS-EUROS model, right the EMEP model. All areas above 40 μgm^{-3} (orange and above) are in exceedance.

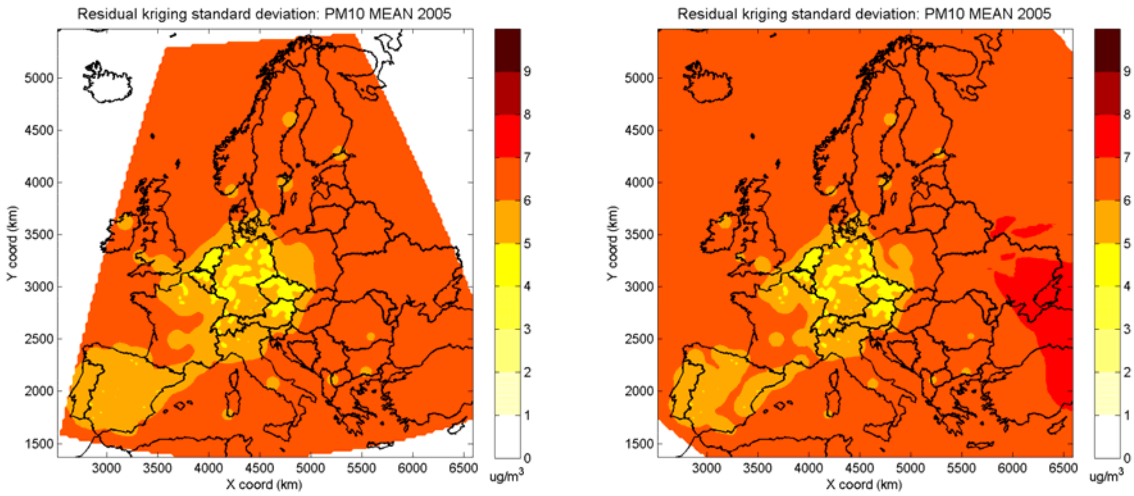


LOTOS-EUROS EMEP

Figure 9. Maps showing the calculated rural background NOE days of PM_{10} in 2005, as calculated using the optimised residual kriging method (3b) described in the text. Left is the LOTOS-EUROS model, right the EMEP model. All regions above 35 days (orange and above) are in exceedance.

3.5 Maps of uncertainty in the annual mean and number of exceedance days

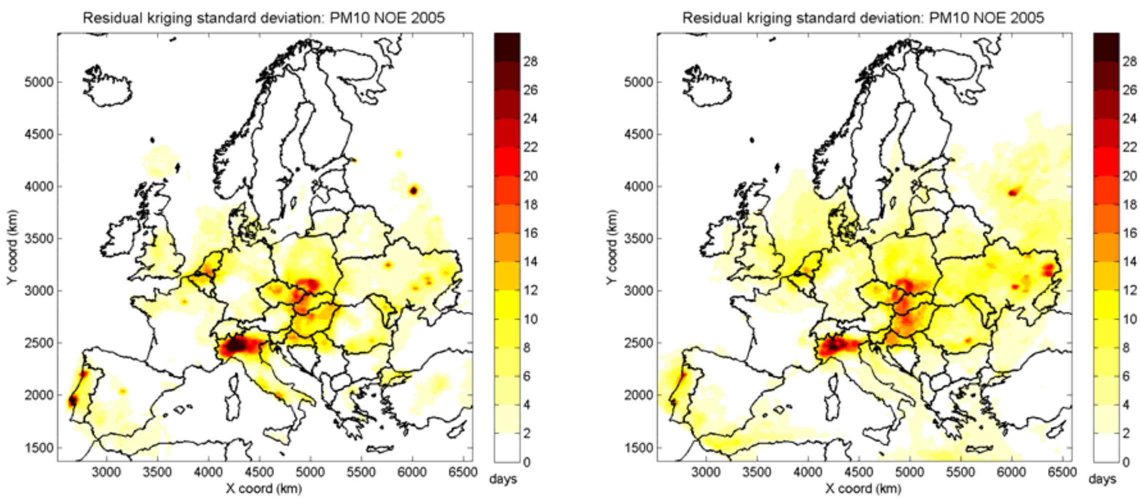
The uncertainty maps have been created using the methodology described in Horálek et al. (2007), chapter 6. The residual kriging variance field is used to determine the daily mean uncertainty. To further determine annual mean uncertainty from these daily mean uncertainties the temporal covariance must be taken into account. For 2005 the temporal covariance factor, required to determine the annual mean uncertainty from daily mean uncertainty, is determined to be F_{cv}=0.205. For the annual mean uncertainty both models give very similar spatially distributed uncertainties (figure 10), in the range 4 – 7 μgm^{-3}, with only slightly higher values for the EMEP model. As expected with the use of residual kriging the uncertainty is highest far from stations, reflecting the variogram sill value, and lowest in areas close to stations, reflecting the variogram nugget value. It is worth noting that the spatial variance, calculated with data from 52 stations, is 3.8 μgm^{-3} (indicating the lower end of the spatial uncertainty) and that the cross-validation RMSE is 5.5 μgm^{-3} (indicating an average spatial uncertainty). These values are thus consistent with the residual kriging uncertainty fields as indicators of uncertainty.



LOTOS-EUROS EMEP

Figure 10. Maps showing the estimated uncertainty in the rural background annual mean concentration of PM_{10} in 2005, as calculated using the optimised residual kriging method (3b) described in the text. Left is the LOTOS-EUROS model, right the EMEP model.

Uncertainty estimates for the NOE days are calculated based principally on the annual mean uncertainty fields, which indicates spatial model and representativeness bias in the interpolation. The method for calculating the uncertainty field is described in Horálek et al. (2007), chapter 6. Briefly, this method involves perturbing the daily mean concentration fields each day with ± the annual mean standard deviation, shown in figure 10. This results in a ± deviation from the total NOE days. The absolute maximum of these two deviations from the calculated NOE days is then used as representative for the standard deviation of the NOE days. Uncertainty in the NOE value can be as high as 30 days or more, e.g. in the Po valley region, though the NOE days in this region can be larger than 70 days. In marginal areas where the NOE days are close to the limit number of 36 days, such as in Eastern Europe, uncertainty can be in the range of 8 – 24 days. This is to a large extend the result of the spatial variability of the monitoring, which for 2005 is calculated to be typically around 13 days, using the 52 stations available.



LOTOS-EUROS EMEP

Figure 11. Maps showing the estimated uncertainty in the rural background NOE days of PM_{10} in 2005, as calculated using the optimised residual kriging method (3b) described in the text. Left is the LOTOS-EUROS model, right the EMEP model.

In regard to the mapping of uncertainty for the NOE days it is worth noting that the use of daily interpolations (figure 11) leads to a quite different uncertainty field than the one determined for the annual mean interpolations. In the case of the annual interpolations the observed NOE days are interpolated directly, using the residual kriging method, and the uncertainty is defined by the kriging variogram and its parameters of nugget and sill. The resultant uncertainty field will look similar to the annual mean uncertainty fields, as shown in figure 10.

To demonstrate the difference between the daily and annual interpolation uncertainty fields the uncertainty using both methods is plotted as a function of NOE days for all grid points in figure 12. For the daily uncertainty determination (figure 12 left) there is a clear dependence of the uncertainty on the number of NOE days up to around 50 days whereafter the scatter becomes larger. As described above calculation of uncertainty using this method perturbs the daily mean concentration fields with the annual mean uncertainty to determine if the daily mean could be in exceedance or not. This results in a small likelihood of a daily exceedance when the daily mean concentrations are low (i.e. low NOE days) and increasing likelihood of exceedance for higher concentrations (i.e. high NOE days). However, once the daily mean concentrations become significantly higher than the limit value then this perturbation has decreasing effect on the uncertainty of the exceedance and so the uncertainty will flatten out at higher exceedance levels. This seems to be a realistic description of the NOE day uncertainty. For the annual interpolation (figure 12 right) the range of uncertainty is dependent only on the kriging parameters of nugget and sill and is completely independent of the actual number of exceedances. This seems to be an unrealistic description of the uncertainty.

As a final point in regard to the use of daily or annual interpolations for the NOE days it is interesting to note that the maximum NOE days found in the maps using daily interpolations is 145 days. For the annual interpolation this is 372 (not included in figure 12), occurring in the hotspot region of Moscow. Two points should be noted concerning this. a) The value is physically impossible and b) there are no observations in this region and so the result is based on the regression model only. The use of regression in the model, with an intercept of 15 days and a slope of 4.3, leads to this unrealistic value, even though the cross-validation RMSE for the NOE days is lowest for the annual interpolations.

The above points indicate that caution must be taken when using the statistical assessment of some of these mapping methods. More consideration is required than only the statistical results when assessing ‘best’ methods.

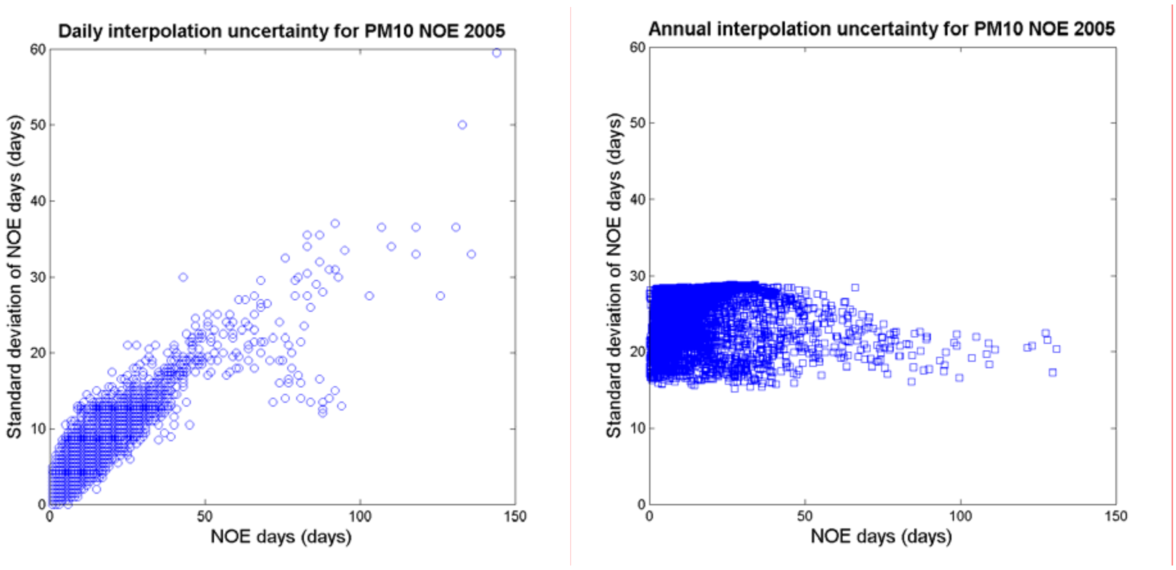


Figure 12. Scatter plots showing the calculated uncertainty in NOE days for PM_{10} as a function of the NOE days for the year 2005 using the residual kriging method (3b) and the LOTOS-EUROS model. Shown are the results using the daily interpolation (left) and the annual interpolation (right).

3.6 Maps of the probability of exceedance for the number of exceedance days

In addition to the concentration, or NOE day, maps and their associated uncertainty maps it is possible to combine these maps to create a probability of exceedance (POE) map. This is achieved by treating the uncertainty as having a normal Gaussian distribution in order to describe the probability distribution function (PDF). This is written analytically as

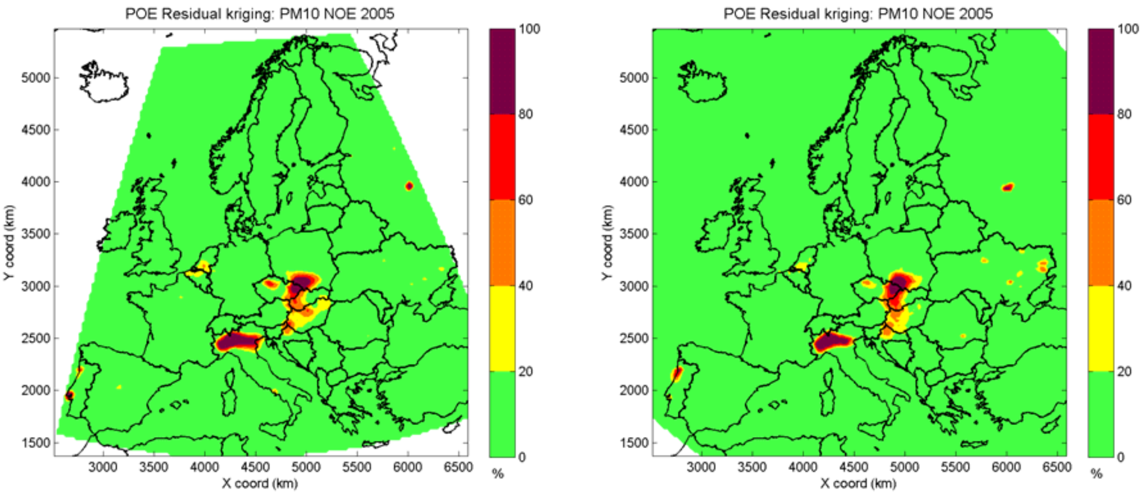
$$PDF = \frac{1}{\sqrt{2\pi\sigma}} \exp\left(-\frac{(c - c_0)^2}{2\sigma^2} \right) \tag{4}$$

where c_0 is the concentration and σ is the uncertainty expressed as a standard deviation. The POE can be determined by integration of the PDF from the limit value (LV) to infinity by

$$POE = \int_{LV}^{\infty} PDF dc = \frac{1}{2} erfc\left(\frac{(LV - c_0)}{\sqrt{2}\sigma} \right) \tag{5}$$

where *erfc* is the complimentary error function. The POE will always be 50% when $LV = c_0$ but will increase, or decrease, dependent on the variance. As a result of the integration, equation 5, the *POE* will be equivalent to *85%* where $c_0 = LV + \sigma$ and *15%* where $c_0 = LV - \sigma$.

In figure 13 the POE for the NOE days is shown, expressed as a % probability. Such maps are in general similar to the NOE maps, figure 9, and are interesting for interpretation. E.g. in the Benelux region the NOE day maps do not show any exceedances. However, incorporation of the uncertainty into this indicates that there is still a 30 – 40 % chance of an exceedance in this area. This can be important information especially when it relates to the representativeness uncertainty as it can then be interpreted to mean that 30 – 40 % of all measurements within that region will show exceedances.



LOTOS-EUROS EMEP

Figure 13. Maps showing the estimated probability of exceedance (POE) in the rural background NOE days of PM_{10} in 2005, as calculated using the optimised residual kriging method (3b) described in the text. Left is the LOTOS-EUROS model, right the EMEP model.

4 Results of the interpolation methods for daily maximum 8 hour running mean O_3 concentrations

In this section results are presented with the following aims

- A comparison of the daily interpolation methods described in section 2
- A comparison of the EMEP and LOTOS-EUROS models
- A comparison of the use of daily or annual statistics
- Presentation of maps and an assessment of uncertainty

4.1 Comparison of daily statistical methods for the EMEP and LOTOS-EUROS models

In this section bar graphs showing the RMSE of the daily maximum 8 hour running mean (MAX8HR), correlation coefficient (R^2) of the MAX8HR and the RMSE of the NOE days (days with MAX8HR > 120 μgm^{-3}) are presented for the different methods described in section 2 and compared for the two air quality models used. The statistics of RMSE and R^2 are calculated using all the daily MAX8HR values for all the available monitoring stations, see ANNEX table A.3 for more statistics. The NOE days are calculated for the available monitoring stations only. The results are shown in figure 14. The following comments and conclusions can be made concerning the methods applied.

1a: Both models show fairly good correlation (0.5), significantly better than for PM_{10}. The EMEP model shows the lowest RMSE for the daily MAX8HR as well as for the NOE days.

1b: Both models show a relatively low bias, certainly when compared to PM_{10}. Linear regression of the model removes this, reducing RMSE and increasing correlation. After regression both models show similar improvements but the linear regression does not significantly effect the NOE days.

1c: Linear regression using both the model and altitude provides improved results for both the daily MAX8HR and the NOE days.

2a-b: There is only a slight improvement when applying optimised kriging, as apposed to fitted kriging. This indicates that the fitting methodology is providing near to optimum results. Both kriging methods provide better results than the model regression.

3c-d: Residual kriging was carried out using the linear regression of the model and altitude. Methods 3a and 3b, using just regression of the model, are not included as they do not perform as well and do not provide any useful comparative results. Optimised residual kriging can be seen to give the best results of any of the interpolation methods though this is only slightly better than fitted residual kriging. The EMEP model provides a slightly better interpolation than does the LOTOS-EUROS model for this year.

4a-b: The weighted combination of kriging and linear regression with model and altitude provides daily MAX8HR results only second to the optimised residual kriging. However, it performs significantly worse in regard to the NOE days, no better than the optimised kriging, on which it is based.

5. The calculated spatial variability helps to indicate the best possible statistic achievable for the interpolation. For ozone this is based on a sample of 148 stations, which will provide a fairly good indication of the spatial variability. There is clearly still room for improvement in the interpolations in regard to RMSE though there is little to be gained in regard to improvement in correlation.

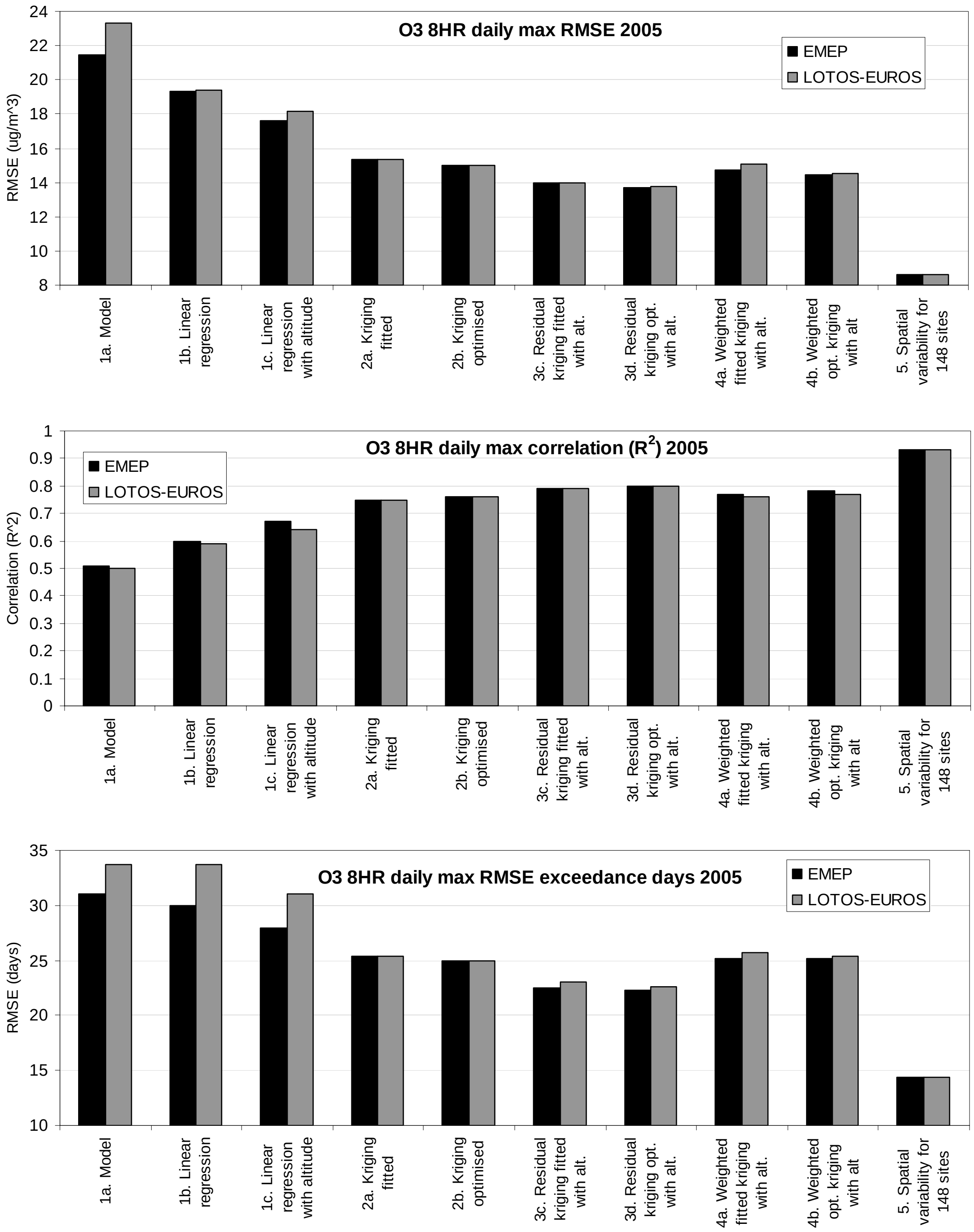


Figure 14. Graphs showing the cross-validation daily MAX8HR concentration RMSE, daily MAX8HR correlation and NOE days RMSE for O_3 using the interpolation methodologies described in the text for the year 2005. Shown are the results of the comparison between the EMEP and LOTOS-EUROS models. Results for the kriging interpolations and spatial variability are the same for both models

4.2 Comparison of the interpolation methods using daily and annual statistics

In this section graphs showing the RMSE and correlation coefficient of the NOE days are presented that compare the results using the daily statistics (section 4.1) with the calculations using annual statistics. Only results for the NOE days are presented as this is the directive related indicator we are interested in mapping. The aim is to see if significant improvement is obtained in calculating the NOE days when using the daily interpolations in comparison to direct interpolation of this as an annual statistic. In figure 15 the comparison between the daily and annual interpolation methods is shown separately for both the EMEP and LOTOS-EUROS models. In figure 16 the results using only the annual interpolations are shown for both models, to provide a direct comparison between the models.

In order to make the daily and annual comparison a selection is made on the stations so that only stations with >75% daily temporal coverage are used in the comparison. The statistics are then calculated based on these stations only.

The following comments and conclusions can be made concerning the methods applied.

1a: The model results are the same for both annual and daily statistics. Both models give low correlation for the NOE days. The LOTOS-EUROS model shows a correlation close to 0. In spite of the high correlation on the daily MAX8HR, figure 14, the NOE is poorly correlated with the model.

1b: Linear regression of the model reduces the RMSE for both models but this is most pronounced using the annual statistics.

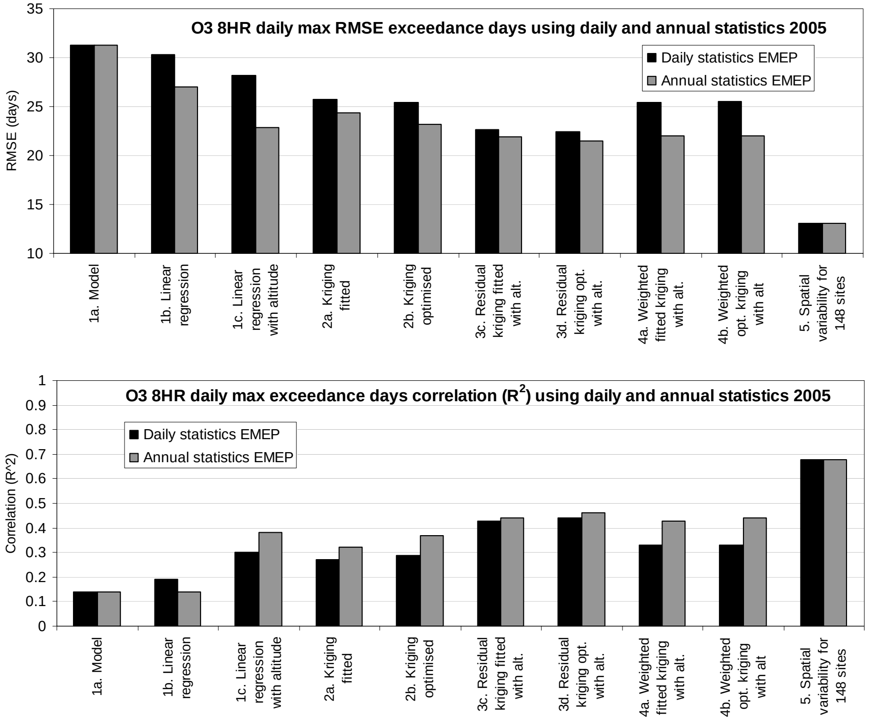
1c: As in the linear regression with the model the inclusion of altitude as a regression variable improves the results for both RMSE and correlation. Once more the interpolation using annual statistics is significantly better than using the daily statistics.

2a-b: Unlike the case for PM_{10}, kriging of the ozone parameter does not produce NOE day fields that are much improved over the regression model. In fact when using the annual statistics the EMEP model with regression provides a better representation of the NOE day fields than does the kriging.

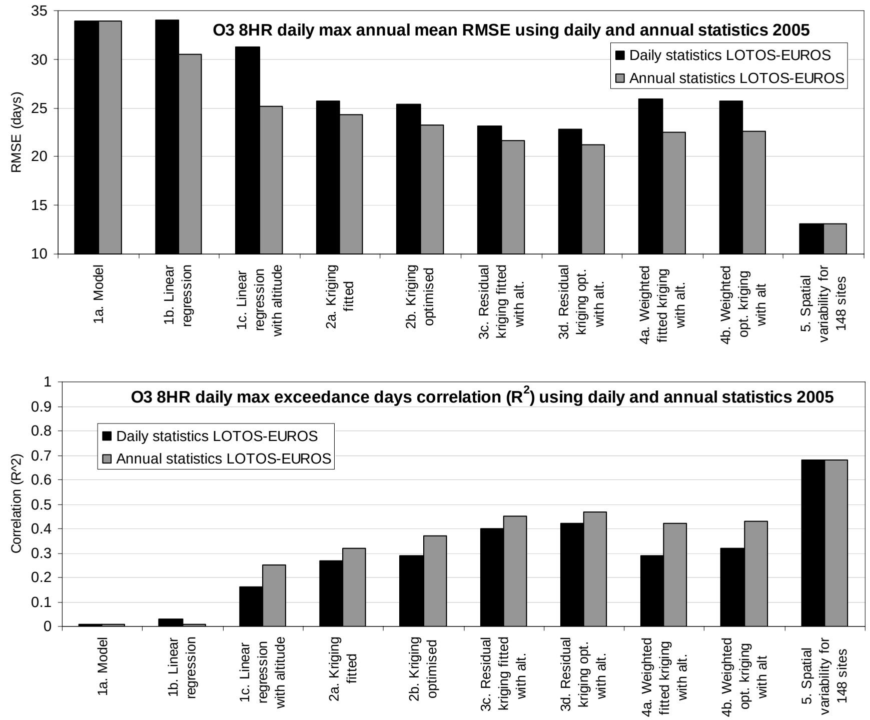
3c-d: Once more optimised residual kriging provides the lowest RMSE and the highest correlation of all the methods. Fitted residual kriging gives only a slightly poorer result indicating that the fitted variogram parameters are near to optimal. For both models the use of annual statistics gives the best results. The EMEP model provides the best statistical results though these are only slightly better than those from the LOTOS-EUROS model.

4a-b: The weighted combination of kriging and linear regression provides NOE day results second only to the optimised residual kriging when the annual interpolations are used. They do not provide results any better than kriging when the daily interpolations are used.

5. The calculated spatial variability helps to indicate the best possible statistic achievable for the interpolation. Using RMSE as an indicator for uncertainty this implies that the spatial representativeness uncertainty accounts for slightly more than half of the ‘best estimate’ uncertainty.

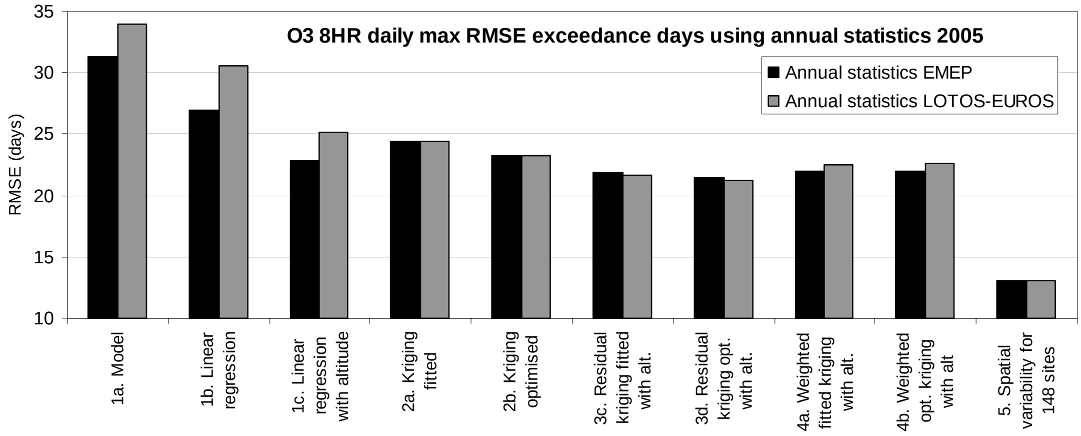


EMEP



LOTOS-EUROS

Figure 15. Graphs showing the cross-validation NOE days RMSE and correlation for O_3 using the interpolation methodologies described in the text for the year 2005. Shown are the results of the comparison between the use of daily interpolations or annual statistics for the EMEP model (left) and the LOTOS-EUROS model (right).



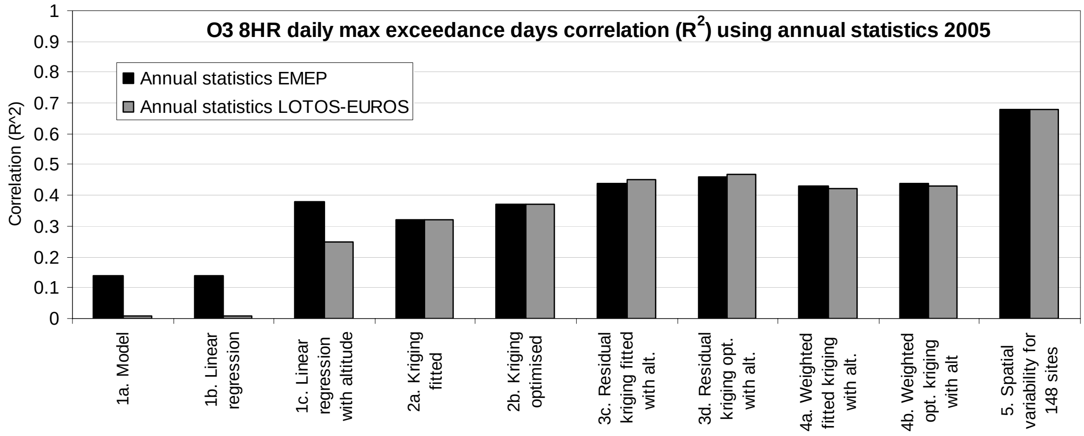
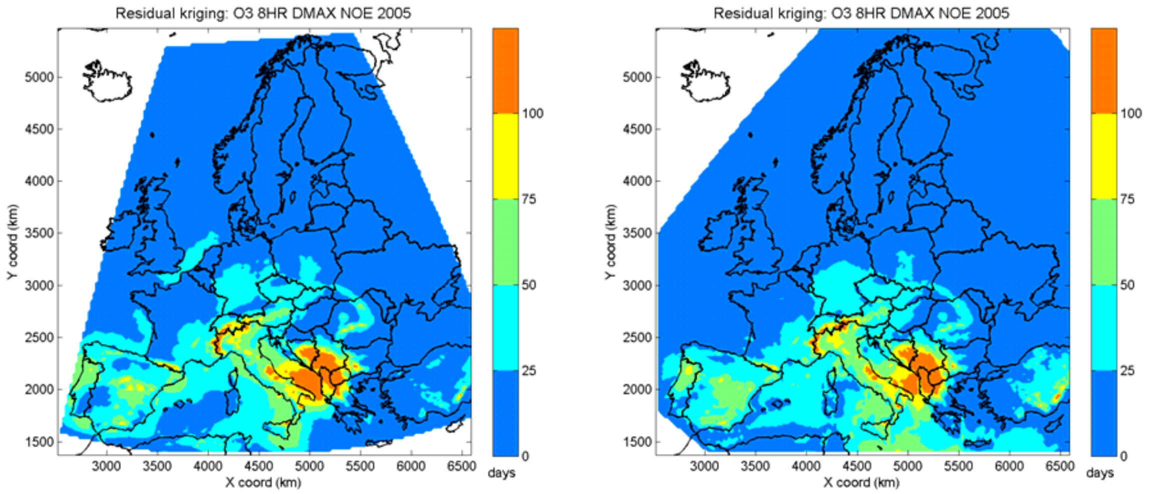


Figure 16. Graphs showing the cross-validation NOE days RMSE and correlation coefficient for ozone using the interpolation methodologies described in the text for the year 2005. Shown are the results of the comparison between the EMEP and LOTOS-EUROS models for the annual statistics only to clearly show the differences between the models for the annual statistics.

4.4 Maps of the number of exceedance days

Maps showing the NOE days of ozone (number of days when the daily maximum 8 hour running mean exceeds 120 μgm^{-3}) for the year 2005 are presented here. These maps have been determined using the daily statistics and the fitted residual kriging methodology with linear regression of the model and altitude (3.c). Though the best performance was found to be methodology 3.d this is computationally expensive to carry out with the current optimisation routine for the entire grid domain. As noted in section 4.2 there is only a small statistical difference between methods 3.c and 3.d and the maps will be visually difficult to distinguish. Maps created using both the EMEP and the LOTOS-EUROS model are presented. As to be expected the exceedance maps, figure 17, for the two models are very similar. The majority of southern Europe is seen to be in exceedance of the directive 26 day target.

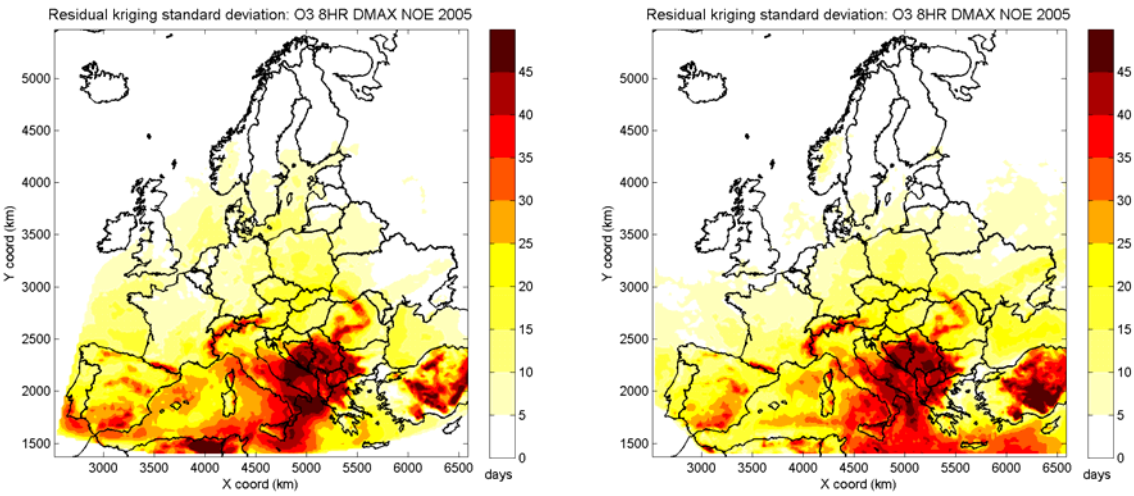


LOTOS-EUROS EMEP

Figure 17. Maps showing the calculated rural background NOE days for ozone in 2005, as calculated using the fitted residual kriging method (3c) described in the text. Left is the LOTOS-EUROS model, right the EMEP model. All areas above 26 days exceed the target value.

4.5 Maps of the uncertainty for the number of exceedance days

Maps showing the uncertainty in the NOE days of ozone (number of days when the daily maximum 8 hour running mean exceeds 120 μgm^{-3}) for the year 2005 are presented in figure 18. These maps have been calculated using the methodology outlined in section 3.5. In this case the spatial covariance factor, required to assess annual mean uncertainty, is found to be F_{CV} = 0.298.



LOTOS-EUROS EMEP

Figure 18. Maps showing the estimated uncertainty in the rural background NOE days for ozone in 2005, as calculated using the fitted residual kriging method (3c) described in the text. Left is the LOTOS-EUROS model, right the EMEP model.

In section 3.5 the difference between the daily and annual interpolation uncertainty assessment methods are described and explained for PM_{10}. A very similar situation occurs for ozone MAX8HR exceedances. To demonstrate this the uncertainty in the NOE days is plotted as a function of NOE days for both the daily and annual methods (figure 19). As for PM_{10} the daily uncertainty assessment method (figure 19 left) shows a clear increase in uncertainty with NOE days and a levelling off, and even decrease, for NOE days > 60 days. The annual uncertainty method (figure 19 right), based on the kriging parameters of nugget and sill, shows no such dependence.

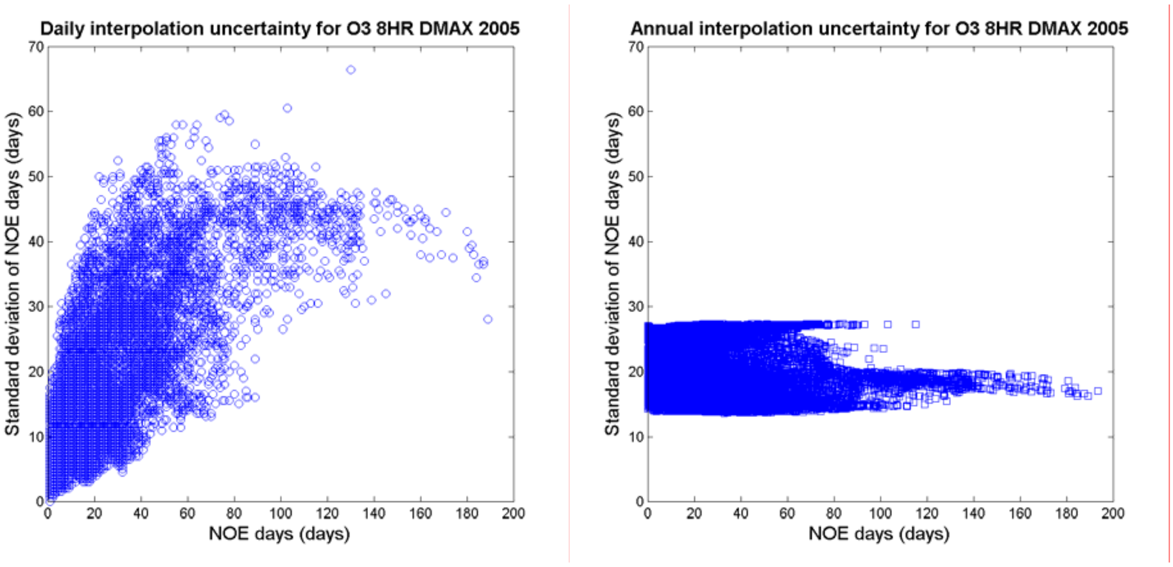
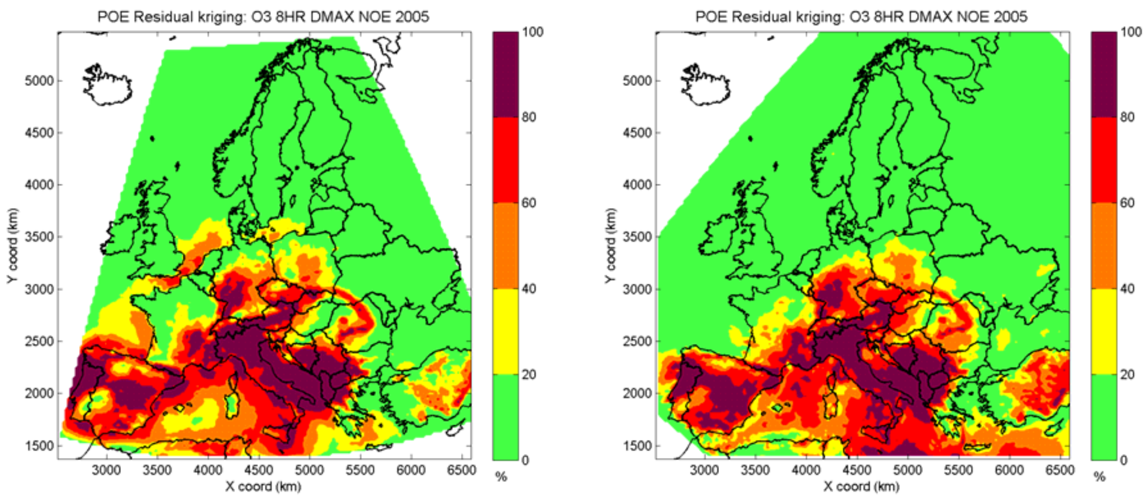


Figure 19. Scatter plots showing the calculated uncertainty in NOE days for ozone as a function of the NOE days for the year 2005 using the residual kriging method (3.c) and the LOTOS-EUROS model. Shown are the results using the daily interpolation (left) and the annual interpolation (right).

4.6 Maps of the probability of exceedance for the number of exceedance days

Maps showing the POE for the NOE days of ozone (number of days when the daily maximum 8 hour running mean exceeds 120 µgm^{-3}) for the year 2005 are presented in figure 20. These maps have a similar structure to the NOE day maps presented in figure 17. However, they do provide extra information especially in regard to spatial representativeness uncertainty. E.g. in figure 17 the entire island of Sicily is seen to be in exceedance. The uncertainty, however, in this region is large enough, around 30 days (figure 18), that the POE lies between 60 – 80%. This indicates a significant likelihood, 20 – 40%, that an independent measurement of ozone on the island would not be in exceedance. The opposite is also true. For a country such as Poland the assessment maps (figure 17) show a relatively small area in exceedance but the POE for the majority of Poland is greater than 20%. Such POE maps can thus provide more information than just the assessment maps.



LOTOS-EUROS EMEP

Figure 20. Maps showing the estimated probability of exceedance in the rural background NOE days for ozone in 2005, as calculated using the fitted residual kriging method (3c) described in the text. Left is the LOTOS-EUROS model, right the EMEP model.

5 Summary and conclusions of the study

In this section we summarise and present recommendations based on the results obtained here for both PM_{10} and ozone. The summary and conclusions are made in regard to the use of daily or annual statistics for the interpolation as well as the use of the EMEP or LOTOS-EUROS models.

5.1 Summary of PM_{10} results

From the assessment carried out here the following conclusions can be made:

1. The interpolation method consistently providing the best cross-validation RMSE and correlation is the optimised residual kriging method using regression of the model (3.b). The weighted kriging method (4.b) was also found to perform well, second to the residual kriging method.
2. The LOTOS-EUROS model consistently provides the best cross-validation RMSE and correlation for the optimised residual kriging method.
3. In regard to annual mean concentrations the interpolation of daily, rather than annual, statistics consistently provided slightly better statistical results, when using the optimised residual kriging method.
4. In regard to the NOE days the interpolation of daily, rather than annual, statistics provided mixed results. In 2003 daily statistics were best but in 2005 annual statistics were found to be best.
5. There is a certain amount of variability between years leading to some years giving different statistical results, in regard to best methods, to other years.
6. Most exceedances of the EU directives are the result of exceedances in the daily mean limit value requirement, rather than the annual mean.
7. A more acceptable description of uncertainty for the NOE days is obtained using the daily interpolation method, rather than the annual interpolation of NOE days.
8. Typical uncertainties are well reflected in the cross-validation RMSE. For the PM_{10} annual mean, using the optimised residual kriging method, this was consistently found to be around 25% of the average observed concentration for the 3 years studied. The spatial variance, due to sub-grid variability, was found to be around 15% or more of the total uncertainty.
9. Typical uncertainties for NOE days based on the RMSE were found to be up to 130%, when compared to the average number of observed exceedance days. This was found, however, to be at the same level as the variability in the spatial representativeness on a 25 x 25 km grid. In this case it appears that the spatial representativeness of the NOE dominates the uncertainty in the interpolation, but due to the small number of samples available, < 50 stations are used in assessing the spatial variability, it is not possible to be more concrete concerning the total contribution of spatial representativeness to the total interpolation uncertainty. However, it does appear to be very significant.

5.2 Recommendations from the PM_{10} results

The following recommendations are made for future applications

1. Optimised residual kriging using regression of the model is the recommended method as it consistently provides the best statistical results for both annual means and NOE days.
2. Improving the regression, through the use of other regression parameters, will ultimately lead to improvements in the residual kriging as well. As noted here, but not applied in this current study, altitude provides some improvement to this. Other land use characteristics, or even more directly high resolution emission estimates, may also provide similar improvements to the multiple regression and hence the residual kriging. It was noted during this study, for example, that there was a significant correlation (R^2=0.14) between the log of the population density and the annual mean concentrations.
3. There is only a very small benefit in using daily statistics for interpolating the annual mean concentrations and this is likely due to the inclusion of extra stations on a daily basis. The resulting maps, both for mean concentrations and uncertainty, are also very similar (Horálek et al.; 2007chapter 6). It is recommended for future applications, when assessing annual mean concentrations, to use annual statistics only.
4. There was no clear statistical improvement in the mapping of NOE days, results are mixed for the 3 years investigated, when using the daily or annual interpolation methods. However, the use of daily interpolations for assessing the NOE days is recommended since it a) provides a physically consistent method for assessing NOE days and b) it provides more realistic assessment and uncertainty maps.
5. If annual statistics are to be used for the directive relating to the NOE days then the methodology currently used by Horálek et al (2005, 2007) of interpolating the 36'th highest daily mean concentration is recommended. However, this interpolation method does not provide any information on the number of days an area is in exceedance, which may be a useful quantity for the users of such maps.
6. If annual statistics are to be used for the interpolations of the PM_{10} indicators then an improved methodology for determining spatial uncertainty is required. The current assumption of stationarity in the fields is not properly fulfilled and the resulting uncertainty maps, based on single values for the kriging variogram parameters, is an unrealistic assessment of the uncertainty.
7. Though the LOTOS-EUROS model provided the better result in this study it would be premature to select it to the exclusion of the EMEP model. Development of both models is currently ongoing and improvements in one model may change the results found here. Both models should be used in future work and their combined use should be investigated.
8. As part of future spatial interpolation studies, both experimental and operational, it is recommended to try to quantitatively assess, as is done here, the spatial representativeness of the observational data to get a clearer understanding of the source and limits of uncertainty in the interpolation.

5.3 Summary of ozone results

From the assessment carried out here the following conclusions can be made:

1. The interpolation method providing the best cross-validation RMSE and correlation is the optimised residual kriging method using multiple linear regression of the model and altitude (3.d). The weighted kriging method (4.b) was also found to perform well, second to the residual kriging method.
2. The EMEP model provided the best cross-validation RMSE and correlation with the monitoring data but there is little difference between the models when applied to the optimised residual kriging method. The performance of the models is much better for ozone than for PM_{10}.
3. In regard to the NOE days the interpolation of annual, rather than daily, statistics provided the best results for almost all interpolation methods. However, as found in the PM_{10} results, no firm conclusion can be made about the robustness of this result without further assessment of data from other years.
4. A more acceptable description of uncertainty for the NOE days is obtained using the daily interpolation method, rather than the annual interpolation of NOE days.
5. Typical uncertainties are reflected in the cross-validation RMSE. For the NOE days for O_3, using the optimised residual kriging method based on annual statistics, this was found to be 65% of the mean observed NOE days. However, an assessment of the spatial variability of the observations indicates that the representativeness uncertainty already accounts for 40% of the uncertainty, i.e. significantly more than half of the interpolation uncertainty is due to the inherent spatial representativeness uncertainty when using a grid of 25 x 25 km.

5.4 Recommendations from the ozone results

The following recommendations are made for future applications:

1. Either fitted or optimised residual kriging using multiple regression is the recommended method as it provides the best statistical results for both the daily and annual statistics.
2. Improving the regression, through the use of other regression parameters, will ultimately lead to improvements in the residual kriging as well. The use of altitude provides significant improvement. Other land use characteristics, or even more directly high resolution emission estimates, may also provide similar improvements to the multiple regression and hence the residual kriging.
3. From the cross-validation statistics there seems to be no clear benefit in using daily statistics for interpolating the NOE days. However, as in the case for PM_{10} the use of daily interpolations for assessing the NOE days is recommended since it a) provides a physically consistent method for assessing NOE days and b) it provides more realistic assessment and uncertainty maps.
4. If annual statistics are to be used for the directive relating to then number of exceedance days then the methodology currently used by Horálek et al (2007) of interpolating the 26’th highest daily maximum 8 hour running mean concentration is recommended. However, this interpolation method does not provide any information on the number of days an area is in exceedance, which may be a useful parameter for the users of such maps.

5. If annual statistics are to be used for the interpolations of the ozone indicators then an improved methodology for determining spatial uncertainty is required. The current assumption of stationarity in the fields is not properly fulfilled and the resulting uncertainty maps, based on single values for the kriging variogram parameters, is an unrealistic assessment of the uncertainty.

6. As part of future spatial interpolation studies, both experimental and operational, it is recommended to try to quantitatively assess, as is done here, the spatial representativeness of the observational data to get a clearer understanding of the source and limits of uncertainty in the interpolation.

5.5 Other aspects for discussion and improvement of the methodologies

During the course of this study and discussions during the year a number of possible improvements to the methodologies have been noted. These include the following points:

1. More thorough testing for the combination of urban and rural interpolations. This has not been addressed in this years reports.

2. The inclusion of land use, emission or population data at high resolution for the multiple regression. E.g. during this study it was noted that the logarithmic population density may also be used as a regression parameter for the rural scale. However, there is a limit as to how effective the use of such data in the regression can be. Many of the processes for PM_{10} and ozone are quite independent of land use, e.g. secondary organic aerosols. This probably works best for NO_2 as a pollutant, which is locally determined by emissions, but not for the other pollutants.

3. Further improvement of the uncertainty mapping methodologies is required especially dealing with the multiple linear regression and residual kriging. One possibility is the use of regionally determined variogram parameters for the interpolation.

4. There is an intrinsic assumption in the interpolation methodologies that monitoring data is the truth. Methods such as kriging and residual kriging that use monitoring data to predict other monitoring data will tend to give the best agreement with the observations. Other methods, such as the weighted combination of observations and models, make the assumption that some truth also lies in the models and so these are less likely to perform as well statistically as the direct kriging methods. e.g. if there is a bias in particular countries for PM_{10} measurements the kriging methods will still perform well statistically even though the truth may lie elsewhere. It is thus important to include in future work the uncertainty associated with the observations and to assess the effect of this on the interpolations and their assessment.

5. The sensitivity of the cross-validation statistical results that determine which method is ‘best’ needs to be properly assessed. As seen in this study for PM_{10} there are differences in the year to year assessment of NOE days as to which is the best method. Is this due to outliers in the datasets? Is it due to the statistical characteristics of the datasets?

6. The current assessment of the ‘best’ methods is statistically based. Other considerations will need to be taken into account, as described in Horálek et al.

(2007) chapter 9, to determine the best method. Are the statistical measures the only objective way of assessing the ‘best’ interpolation method? As an example, it was also found in this study for PM_{10} that though annual interpolations can give statistically better results than the daily interpolations for the NOE days, that the annual interpolations can be unrealistic in both the assessment and the uncertainty maps.

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Annex I: Tables of cross-validation statistics

Table A.1. Cross validation statistical parameters for the assessment of daily mean interpolations and number of exceedance days (NOE) of PM_{10} using the EMEP and LOTOS-EUROS models. Years 2003 – 2005. Regression parameters, slope and intercept, are determined using the interpolation results as the independent (X) variable.

2003

Cross-validation statistical results **PM_{10} EMEP 2003**	**1a.** Model	**1b.** Linear regression	**1c.** Linear regression with altitude	**2a.** Kriging fitted	**2b.** Kriging optimised	**2c.** Log-Kriging fitted	**2d.** Log-Kriging optimised	**3a.** Residual kriging fitted	**3b.** Residual kriging optimised	**4a.** Weighted fitted kriging	**4b.** Weighted optimised kriging	**5.** Spatial variability for 37 sites
Daily mean												
RMSE (μgm^{-3})	20.1	14.8	14.02	12.35	12.56	11.71	11.67	12.35	11.48	12.25	11.51	9.41
MAE (μgm^{-3})	14.0	9.2	8.73	7.51	7.65	7.28	7.14	7.48	7.10	7.46	7.14	4.36
Correlation (r^2)	0.27	0.36	0.41	0.55	0.53	0.59	0.60	0.54	0.61	0.55	0.60	0.81
Intercept (μgm^{-3})	13.5	1.3	0.5	0.1	-0.2	0.5	1.1	0.5	0.7	-1.3	-0.8	-0.0
Slope	0.89	0.95	0.98	0.98	1.0	0.97	0.97	0.98	0.98	1.06	1.03	1.03
NOE days												
RMSE (days)	30.0	25.4	25.4	21.7	21.8	20.3	20.1	20.8	19.7	22.3	20.9	21.5
MAE (days)	16.9	13.9	13.0	11.0	11.0	10.2	10.2	10.7	10.2	11.3	10.6	10.1

Cross-validation statistical results **PM_{10} LOTOS 2003**	**1a.** Model	**1b.** Linear regression	**1c.** Linear regression with altitude	**2a.** Kriging fitted	**2b.** Kriging optimised	**2c.** Log-Kriging fitted	**2d.** Log-Kriging optimised	**3a.** Residual kriging fitted	**3b.** Residual kriging optimised	**4a.** Weighted fitted kriging	**4b.** Weighted optimised kriging	**5.** Spatial variability for 37 sites
Daily mean												
RMSE (μgm^{-3})	20.2	14.0	13.59	12.35	12.56	11.71	11.67	11.89	11.13	12.08	11.36	9.41
MAE (μgm^{-3})	13.7	8.9	8.57	7.51	7.65	7.28	7.14	7.17	6.82	7.35	7.03	4.36
Correlation (r^2)	0.26	0.42	0.45	0.55	0.53	0.59	0.60	0.58	0.63	0.57	0.62	0.81
Intercept (μgm^{-3})	8.8	-0.2	0.2	0.1	-0.2	0.5	1.1	0.1	0.5	-1.8	-1.1	-0.0
Slope	1.34	1.00	0.99	0.98	1.0	0.97	0.97	1.0	0.98	1.08	1.05	1.03
NOE days												
RMSE (days)	33.0	23.5	22.8	21.7	21.8	20.3	20.1	20.3	18.9	21.8	20.7	21.5
MAE (days)	20.1	13.0	11.7	11.0	11.0	10.2	10.2	10.3	9.5	11.0	10.4	10.1

2004

Cross-validation statistical results **PM_{10} EMEP 2004**	**1a.** Model	**1b.** Linear regression	**1c.** Linear regression with altitude	**2a.** Kriging fitted	**2b.** Kriging optimised	**2c.** Log-Kriging fitted	**2d.** Log-Kriging optimised	**3a.** Residual kriging fitted	**3b.** Residual kriging optimised	**4a.** Weighted fitted kriging	**4b.** Weighted optimised kriging	**5.** Spatial variability for 37 sites
Daily mean												
RMSE (µgm^{-3})	17.04	12.56	11.99	10.38	10.42	9.98	10.04	10.30	9.80	10.19	9.85	6.91
MAE (µgm^{-3})	11.96	7.94	7.47	6.28	6.35	6.18	6.13	6.25	6.03	6.21	6.08	3.16
Correlation (r^2)	0.18	0.27	0.33	0.50	0.50	0.54	0.53	0.51	0.55	0.52	0.55	0.82
Intercept (µgm^{-3})	12.9	2.1	0.5	0.6	0.8	0.4	0.8	1.2	0.8	-1.0	-0.8	0.5
Slope	0.75	0.89	0.97	0.96	0.95	0.98	0.97	0.94	0.97	1.05	1.04	1.00
NOE days												
RMSE (days)	23.1	21.8	22.0	18.6	18.4	17.5	17.5	17.9	16.7	19.1	18.1	10.8
MAE (days)	11.8	10.5	10.4	8.4	8.4	8.1	8.1	8.1	7.6	8.7	8.2	5.2

Cross-validation statistical results **PM_{10} LOTOS 2004**	**1a.** Model	**1b.** Linear regression	**1c.** Linear regression with altitude	**2a.** Kriging fitted	**2b.** Kriging optimised	**2c.** Log-Kriging fitted	**2d.** Log-Kriging optimised	**3a.** Residual kriging fitted	**3b.** Residual kriging optimised	**4a.** Weighted fitted kriging	**4b.** Weighted optimised kriging	**5.** Spatial variability for 37 sites
Daily mean												
RMSE (µgm^{-3})	16.41	12.82	12.36	10.38	10.42	9.98	10.04	9.99	9.50	10.10	9.76	6.91
MAE (µgm^{-3})	10.91	8.18	7.78	6.28	6.35	6.18	6.13	6.00	5.78	6.11	5.98	3.16
Correlation (r^2)	0.12	0.24	0.29	0.50	0.50	0.54	0.53	0.54	0.58	0.53	0.56	0.82
Intercept (µgm^{-3})	12.5	3.0	1.8	0.6	0.8	0.4	0.8	0.9	0.5	-1.1	-1.0	0.5
Slope	0.65	0.85	0.91	0.96	0.95	0.98	0.97	0.96	0.99	1.06	1.06	1.00
NOE days												
RMSE (days)	23.5	22.3	22.0	18.6	18.4	17.5	17.5	17.8	16.7	19.0	18.3	10.8
MAE (days)	12.1	10.6	10.4	8.4	8.4	8.1	8.1	8.0	7.7	8.7	8.3	5.2

2005

Cross-validation statistical results **PM_{10} EMEP 2005**	**1a.** Model	**1b.** Linear regression	**1c.** Linear regression with altitude	**2a.** Kriging fitted	**2b.** Kriging optimised	**2c.** Log-Kriging fitted	**2d.** Log-Kriging optimised	**3a.** Residual kriging fitted	**3b.** Residual kriging optimised	**4a.** Weighted fitted kriging	**4b.** Weighted optimised kriging	**5.** Spatial variability for 37 sites
Daily mean												
RMSE (μgm^{-3})	18.54	13.77	13.25	11.74	11.66	11.22	11.50	11.43	11.00	11.39	11.07	7.32
MAE (μgm^{-3})	12.99	8.90	8.44	7.06	7.09	6.89	6.97	6.93	6.73	6.97	6.81	4.04
Correlation (r^2)	0.20	0.28	0.33	0.47	0.48	0.52	0.49	0.50	0.54	0.50	0.53	0.83
Intercept (μgm^{-3})	13.2	2.3	0.7	0.9	0.8	0.2	0.9	1.3	0.9	-1.2	-1.2	0.3
Slope	0.83	0.89	0.97	0.95	0.95	0.98	0.96	0.93	0.96	1.05	1.05	1.00
NOE days												
RMSE (days)	26.8	23.4	23.7	20.1	20.1	19.8	20.3	18.7	18.1	21.0	20.2	17.5
MAE (days)	14.9	12.6	12.5	10.9	10.9	10.6	10.9	10.4	10.1	11.2	10.8	9.8

Cross-validation statistical results **PM_{10} LOTOS 2005**	**1a.** Model	**1b.** Linear regression	**1c.** Linear regression with altitude	**2a.** Kriging fitted	**2b.** Kriging optimised	**2c.** Log-Kriging fitted	**2d.** Log-Kriging optimised	**3a.** Residual kriging fitted	**3b.** Residual kriging optimised	**4a.** Weighted fitted kriging	**4b.** Weighted optimised kriging	**5.** Spatial variability for 37 sites
Daily mean												
RMSE (μgm^{-3})	17.59	13.63	13.28	11.74	11.66	11.22	11.50	11.08	10.63	11.25	10.93	7.32
MAE (μgm^{-3})	11.78	8.80	8.47	7.06	7.09	6.89	6.97	6.66	6.46	6.86	6.72	4.04
Correlation (r^2)	0.18	0.29	0.32	0.47	0.48	0.52	0.49	0.53	0.57	0.52	0.54	0.83
Intercept (μgm^{-3})	11.8	2.0	1.3	0.9	0.8	0.2	0.9	0.8	0.5	-1.4	-1.5	0.3
Slope	0.82	0.91	0.94	0.95	0.95	0.98	0.96	0.96	0.98	1.07	1.07	1.00
NOE days												
RMSE (days)	27.2	23.2	22.9	20.1	20.1	19.8	20.3	18.0	17.5	20.5	20.0	17.5
MAE (days)	15.3	12.8	12.5	10.9	10.9	10.6	10.9	10.1	9.7	10.9	107	9.8

Table A.2. Cross validation statistical parameters for the assessment of annual mean concentrations and number of exceedance days (NOE) of PM_{10}. Shown are the results of daily interpolation, which uses daily interpolations to provide annual statistics, and annual interpolations, which use annual mean or NOE data from models and observations. Only the LOTOS-EUROS model is shown. Years 2003 – 2005. Regression parameters, slope and intercept, are determined using the interpolation results as the independent (X) variable.

2003

Cross-validation statistical results **DAILY INTERPOLATION PM_{10} LOTOS 2003**	**1a.** Model	**1b.** Linear regression	**1c.** Linear regression with altitude	**2a.** Kriging fitted	**2b.** Kriging optimised	**2c.** Log-Kriging fitted	**2d.** Log-Kriging optimised	**3a.** Residual kriging fitted	**3b.** Residual kriging optimised	**4a.** Weighted fitted kriging	**4b.** Weighted optimised kriging	**5.** Spatial variability for 37 sites
Annual mean												
RMSE (μgm^{-3})	14.24	7.39	7.07	7.36	7.44	6.95	6.82	6.50	6.14	6.98	6.60	6.61
MAE (μgm^{-3})	12.22	5.13	4.72	5.20	5.25	4.99	4.74	4.39	4.21	4.84	4.67	3.90
Correlation (r^2)	0.38	0.38	0.42	0.39	0.37	0.44	0.46	0.52	0.57	0.47	0.52	0.71
Intercept (μgm^{-3})	5.6	-6.4	-3.7	-6.6	-7.5	-3.9	-2.1	-4.9	-3.0	-10.1	-7.0	0.6
Slope	1.60	1.25	1.14	1.25	1.29	1.14	1.10	1.20	1.12	1.42	1.29	1.03
NOE days												
RMSE (days)	36.08	25.87	24.91	23.74	23.79	22.11	21.89	21.72	20.11	23.86	22.48	22.77
MAE (days)	21.45	13.44	12.50	11.97	11.98	11.23	11.03	10.69	9.80	11.93	11.32	10.51

Cross-validation statistical results **ANNUAL INTERPOLATION PM_{10} LOTOS 2003**	**1a.** Model	**1b.** Linear regression	**1c.** Linear regression with altitude	**2a.** Kriging fitted	**2b.** Kriging optimised	**2c.** Log-Kriging fitted	**2d.** Log-Kriging optimised	**3a.** Residual kriging fitted	**3b.** Residual kriging optimised	**4a.** Weighted fitted kriging	**4b.** Weighted optimised kriging	**5.** Spatial variability for 37 sites
Annual mean												
RMSE (μgm^{-3})	14.24	7.30	7.15	7.82	7.89	7.11	6.89	7.12	6.08	7.20	6.56	6.61
MAE (μgm^{-3})	12.22	4.96	4.76	5.35	5.48	5.05	4.86	4.60	4.13	4.84	4.47	3.90
Correlation (r^2)	0.38	0.38	0.40	0.31	0.29	0.41	0.46	0.42	0.57	0.41	0.50	0.71
Intercept (μgm^{-3})	5.6	0.0	0.0	-10.7	-9.3	1.2	3.5	-1.3	0.1	-6.4	-2.4	0.6
Slope	1.60	1.00	1.00	1.47	1.40	0.94	0.86	1.11	1.03	1.28	1.12	1.03
NOE days												
RMSE (days)	36.08	25.83	25.01	22.75	25.27	21.06	20.63	25.63	22.73	24.49	20.58	22.77
MAE (days)	21.45	16.87	15.77	14.42	15.64	12.89	11.23	15.75	13.71	15.26	12.74	10.51

2004

Cross-validation statistical results **DAILY INTERPOLATION PM_{10} LOTOS 2004**	**1a.** Model	**1b.** Linear regression	**1c.** Linear regression with altitude	**2a.** Kriging fitted	**2b.** Kriging optimised	**2c.** Log-Kriging fitted	**2d.** Log-Kriging optimised	**3a.** Residual kriging fitted	**3b.** Residual kriging optimised	**4a.** Weighted fitted kriging	**4b.** Weighted optimised kriging	**5.** Spatial variability for 37 sites
Annual mean												
RMSE (μgm^{-3})	10.88	6.75	6.28	5.79	5.79	5.62	5.58	5.09	4.88	5.54	5.39	3.46
MAE (μgm^{-3})	8.58	4.74	4.37	4.07	4.13	4.06	4.00	3.56	3.49	3.84	3.83	2.55
Correlation (r^2)	0.17	0.17	0.27	0.37	0.37	0.41	0.42	0.52	0.56	0.45	0.48	0.79
Intercept (μgm^{-3})	12.3	5.5	2.9	-2.1	-1.8	-2.3	-0.9	-2.1	-2.4	-6.1	-5.8	2.2
Slope	0.67	0.72	0.85	1.09	1.08	1.11	1.05	1.10	1.13	1.31	1.30	0.91
NOE days												
RMSE (days)	25.26	23.94	23.63	19.66	19.47	18.58	18.40	18.85	17.68	20.24	19.37	10.47
MAE (days)	13.36	11.70	11.39	8.49	8.86	8.44	8.19	8.39	8.05	9.23	8.83	5.69

Cross-validation statistical results **ANNUAL INTERPOLATION PM_{10} LOTOS 2004**	**1a.** Model	**1b.** Linear regression	**1c.** Linear regression with altitude	**2a.** Kriging fitted	**2b.** Kriging optimised	**2c.** Log-Kriging fitted	**2d.** Log-Kriging optimised	**3a.** Residual kriging fitted	**3b.** Residual kriging optimised	**4a.** Weighted fitted kriging	**4b.** Weighted optimised kriging	**5.** Spatial variability for 37 sites
Annual mean												
RMSE (μgm^{-3})	10.88	6.66	6.29	5.82	5.85	5.76	5.62	5.31	5.19	5.74	5.55	3.46
MAE (μgm^{-3})	8.58	4.66	4.40	4.05	4.11	4.11	3.99	3.66	3.67	3.95	3.86	2.55
Correlation (r^2)	0.17	0.17	0.26	0.37	0.36	0.38	0.41	0.48	0.50	0.40	0.43	0.79
Intercept (μgm^{-3})	12.3	-0.0	-0.0	-2.6	-1.2	1.7	0.9	-2.6	-1.1	-5.2	-2.3	2.2
Slope	0.67	1.00	1.00	1.13	1.05	0.91	0.97	1.13	1.07	1.26	1.12	0.91
NOE days												
RMSE (days)	25.26	21.45	20.35	18.59	18.93	17.71	17.19	18.86	17.64	18.73	17.81	10.47
MAE (days)	13.36	13.18	11.68	9.80	10.10	9.69	8.71	10.01	9.59	10.08	9.78	5.69

2005

Cross-validation statistical results **DAILY INTERPOLATION PM_{10} LOTOS 2005**	**1a.** Model	**1b.** Linear regression	**1c.** Linear regression with altitude	**2a.** Kriging fitted	**2b.** Kriging optimised	**2c.** Log-Kriging fitted	**2d.** Log-Kriging optimised	**3a.** Residual kriging fitted	**3b.** Residual kriging optimised	**4a.** Weighted fitted kriging	**4b.** Weighted optimised kriging	**5.** Spatial variability for 37 sites
Annual mean												
RMSE (μgm^{-3})	11.85	7.09	6.71	6.59	6.44	6.26	6.46	5.68	5.48	6.09	5.97	3.83
MAE (μgm^{-3})	9.65	5.41	5.12	4.80	4.66	4.59	4.71	4.12	3.96	4.45	4.40	2.78
Correlation (r^2)	0.19	0.21	0.29	0.31	0.34	0.38	0.33	0.49	0.53	0.44	0.46	0.75
Intercept (μgm^{-3})	12.2	3.5	1.8	-1.0	-2.2	-3.1	-0.8	-1.8	-2.3	-7.6	-7.7	2.3
Slope	0.76	0.83	0.90	1.03	1.09	1.14	1.04	1.08	1.11	1.35	1.36	0.90
NOE days												
RMSE (days)	29.44	25.27	24.79	22.34	21.47	21.08	21.76	19.19	18.64	22.06	21.54	13.26
MAE (days)	16.93	14.10	13.80	11.96	11.65	11.35	11.62	10.85	10.44	11.86	11.54	7.69

Cross-validation statistical results **ANNUAL INTERPOLATION PM_{10} LOTOS 2005**	**1a.** Model	**1b.** Linear regression	**1c.** Linear regression with altitude	**2a.** Kriging fitted	**2b.** Kriging optimised	**2c.** Log-Kriging fitted	**2d.** Log-Kriging optimised	**3a.** Residual kriging fitted	**3b.** Residual kriging optimised	**4a.** Weighted fitted kriging	**4b.** Weighted optimised kriging	**5.** Spatial variability for 37 sites
Annual mean												
RMSE (μgm^{-3})	11.85	7.12	6.84	6.46	6.45	6.39	6.39	5.83	5.69	6.23	6.22	3.83
MAE (μgm^{-3})	9.65	5.42	5.20	4.62	4.61	4.55	4.54	4.17	4.08	4.54	4.54	2.78
Correlation (r^2)	0.19	0.19	0.25	0.33	0.34	0.35	0.35	0.46	0.48	0.39	0.40	0.75
Intercept (μgm^{-3})	12.2	0.0	-0.0	0.1	0.0	-0.2	0.8	-1.3	-1.5	-5.2	-6.1	2.3
Slope	0.76	1.00	1.00	0.99	0.99	1.00	0.97	1.06	1.08	1.24	1.29	0.90
NOE days												
RMSE (days)	29.4	23.3	23.32	20.69	19.41	18.38	19.73	17.93	17.05	19.17	18.01	13.26
MAE (days)	16.9	16.1	16.06	12.88	12.08	11.80	11.24	11.78	11.29	12.37	11.89	7.69

Table A.3. Cross validation statistical parameters for the assessment of daily maximum 8 hour running mean interpolations and number of exceedance days (NOE) for ozone using the EMEP and LOTOS-EUROS models. Year 2005. Regression parameters, slope and intercept, are determined using the interpolation results as the independent (X) variable.

Cross-validation statistical results **OZONE EMEP 2005**	**1a.** Model	**1b.** Linear regression	**1c.** Linear regression with altitude	**2a.** Kriging fitted	**2b.** Kriging optimised	**3c.** Residual kriging fitted with alt.	**3d.** Residual kriging opt. with alt.	**4a.** Weighted fitted kriging with alt.	**4b.** Weighted opt. kriging with alt	**5.** Spatial variability for 148 sites
Daily mean										
RMSE (μgm^{-3})	21.48	19.31	17.64	15.36	15.02	13.94	13.70	14.71	14.45	8.63
MAE (μgm^{-3})	16.13	14.41	13.03	10.58	10.35	9.58	9.45	10.29	10.11	5.87
Correlation (r^2)	0.51	0.60	0.67	0.75	0.76	0.79	0.80	0.77	0.78	0.93
Intercept (μgm^{-3})	6.6	2.9	-0.1	1.5	1.2	0.8	0.9	-2.4	-2.3	-0.2
Slope	0.90	0.96	1.00	0.98	0.99	0.99	0.99	1.03	1.03	1.00
NOE days										
RMSE (days)	31.0	30.0	27.9	25.4	25.0	22.5	22.3	25.2	25.2	14.4
MAE (days)	19.5	18.4	17.1	13.9	13.8	12.3	12.3	14.1	14.1	9.4

Cross-validation statistical results **OZONE LOTOS 2005**	**1a.** Model	**1b.** Linear regression	**1c.** Linear regression with altitude	**2a.** Kriging fitted	**2b.** Kriging optimised	**3c.** Residual kriging fitted with alt.	**3d.** Residual kriging opt. with alt.	**4a.** Weighted fitted kriging with alt.	**4b.** Weighted opt. kriging with alt	**5.** Spatial variability for 148 sites
Daily mean										
RMSE (μgm^{-3})	23.29	19.43	18.18	15.36	15.02	13.98	13.77	15.08	14.53	8.63
MAE (μgm^{-3})	18.43	14.63	13.50	10.58	10.35	9.64	9.52	10.50	10.17	5.87
Correlation (r^2)	0.50	0.59	0.64	0.75	0.76	0.79	0.80	0.76	0.77	0.93
Intercept (μgm^{-3})	23.5	1.5	-0.4	1.5	1.2	0.8	1.0	-1.0	-2.2	-0.2
Slope	0.71	0.98	1.00	0.98	0.99	0.99	0.99	1.02	1.03	1.00
NOE days										
RMSE (days)	33.7	33.7	31.0	25.4	25.0	23.0	22.6	25.7	25.4	14.4
MAE (days)	23.0	22.1	19.8	13.9	13.8	12.6	12.5	14.3	14.2	9.4

Table A.4. Cross validation statistical parameters for the assessment of the number of exceedance days (NOE) of ozone. Shown are the results of daily interpolation, which uses daily interpolations to provide annual statistics, and annual interpolations, which uses annual NOE data from models and observations. Both the EMEP and LOTOS-EUROS models are shown. Year 2005. Regression parameters, slope and intercept, are determined using the interpolation results as the independent (X) variable.

EMEP

Cross-validation statistical results **DAILY INTERPOLATION OZONE EMEP 2005**	**1a.** Model	**1b.** Linear regression	**1c.** Linear regression with altitude	**2a.** Kriging fitted	**2b.** Kriging optimised	**3c.** Residual kriging fitted with alt.	**3d.** Residual kriging opt. with alt.	**4a.** Weighted fitted kriging with alt.	**4b.** Weighted opt. kriging with alt	**5.** Spatial variability for 148 sites
NOE days										
RMSE (days)	31.31	30.33	28.21	25.71	25.40	22.67	22.47	25.44	25.52	13.09
MAE (days)	19.64	18.75	17.41	14.16	14.09	12.46	12.42	14.36	14.36	9.15
Correlation (r^2)	0.14	0.19	0.30	0.27	0.29	0.43	0.44	0.33	0.33	0.68
Intercept (days)	19.4	12.5	12.5	10.4	8.5	6.1	5.8	6.8	6.6	-0.2
Slope	0.78	1.15	1.08	0.83	0.93	0.97	0.98	1.09	1.10	1.00

Cross-validation statistical results **ANNUAL INTERPOLATION OZONE EMEP 2005**	**1a.** Model	**1b.** Linear regression	**1c.** Linear regression with altitude	**2a.** Kriging fitted	**2b.** Kriging optimised	**3c.** Residual kriging fitted with alt.	**3d.** Residual kriging opt. with alt.	**4a.** Weighted fitted kriging with alt.	**4b.** Weighted opt. kriging with alt	**5.** Spatial variability for 148 sites
NOE days										
RMSE (days)	31.31	26.99	22.85	24.36	23.20	21.89	21.48	22.00	22.02	13.09
MAE (days)	19.64	18.26	13.84	13.87	13.67	12.19	12.01	12.73	12.81	9.15
Correlation (r^2)	0.14	0.14	0.38	0.32	0.37	0.44	0.46	0.43	0.44	0.68
Intercept (days)	19.4	0.0	-0.0	6.8	-2.9	3.6	1.0	-4.0	-7.4	-0.2
Slope	0.78	1.00	1.00	0.79	1.09	0.89	0.99	1.13	1.23	1.00

LOTOS-EUROS

Cross-validation statistical results **DAILY INTERPOLATION OZONE LOTOS 2005**	**1a.** Model	**1b.** Linear regression	**1c.** Linear regression with altitude	**2a.** Kriging fitted	**2b.** Kriging optimised	**3c.** Residual kriging fitted with alt.	**3d.** Residual kriging opt. with alt.	**4a.** Weighted fitted kriging with alt.	**4b.** Weighted opt. kriging with alt	**5.** Spatial variability for 148 sites
NOE days										
RMSE (days)	33.98	34.04	31.21	25.71	25.40	23.19	22.80	25.93	25.67	13.09
MAE (days)	23.12	22.44	20.12	14.16	14.09	12.77	12.68	14.56	14.45	9.15
Correlation (r^2)	0.01	0.03	0.16	0.27	0.29	0.40	0.42	0.29	0.32	0.68
Intercept (days)	28.0	24.6	18.7	10.4	8.5	7.6	7.0	9.0	6.9	-0.2
Slope	0.27	0.55	0.84	0.83	0.93	0.92	0.94	0.98	1.08	1.00

Cross-validation statistical results **ANNUAL INTERPOLATION OZONE LOTOS 2005**	**1a.** Model	**1b.** Linear regression	**1c.** Linear regression with altitude	**2a.** Kriging fitted	**2b.** Kriging optimised	**3c.** Residual kriging fitted with alt.	**3d.** Residual kriging opt. with alt.	**4a.** Weighted fitted kriging with alt.	**4b.** Weighted opt. kriging with alt	**5.** Spatial variability for 148 sites
NOE days										
RMSE (days)	33.98	30.51	25.20	24.36	23.20	21.62	21.22	22.53	22.63	13.09
MAE (days)	23.12	20.98	16.51	13.87	13.67	12.27	12.07	13.55	13.57	9.15
Correlation (r^2)	0.01	0.01	0.25	0.32	0.37	0.45	0.47	0.42	0.43	0.68
Intercept (days)	28.0	24.1	0.0	6.8	-2.9	3.2	0.2	-8.7	-12.9	-0.2
Slope	0.27	0.27	1.00	0.79	1.09	0.90	1.00	1.27	1.39	1.00