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Data assimilation with a regional scale model: homogenous regional background for city assessment

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1. Executive Summary

In the context of implementing the European air quality legislation and aiming to maintain the limit values for the regulated pollutants $PM_{10/25}$, O_3 and NO_2 cities strive to identify abatement measures to limit emissions from mobile and stationary sources. In this context it is of importance to distinct as precisely as possible between air quality concentrations caused by sources within the city and regional background concentrations. To assess the latter is much more difficult since the local authorities normally do not have enough information available (e.g. emission inventories) to assess the background concentrations through model runs. In addition, the models used often have a limited model performance, especially in the case of PM10. The determination of the regional background by observations is also problematic due to the fact that it is in general difficult to assess to which extent rural, background stations are influenced by the city emissions themselves, resulting in the problem of double counting.

The AIR4EU project has recognised that determining in an reliable way the regional background concentrations is one of the major drawbacks cities are facing and so AIR4EU has developed a method to prepare background concentrations through innovative data assimilation methods.

The method developed consists of the CTM LOTOS-EUROS coupled with the data assimilation method Ensemble Kalman Filtering. This system can combine observations of the species O3, PM10, 2.5 and NO2, and also of other species if wanted, with modelled concentrations of these species. The result are consistent air quality fields over Europe, on an hourly/daily averaged basis, and on a grid resolution currently of about 15 x 15 km2., and for time periods of years. Examples have been made for the year 2003 for PM10 and O3. These resulting fields have a reduced uncertainty relative to the uncertainty of the observations, or the uncertainty of the modelling separately.

In principle, because the system is operational, these background concentrations can be made available and can be used by city users and others

2. Case study description

2.1 Background

This case study is of a different character than the case studies performed for the cities of Rotterdam, Prague etc. This so-called EUROPE 1 Case Study has not been carried out in close contact with a city user, or another user. The aim of this “Case study” is to investigate whether it is possible to create coherent and consistent regional background concentrations which can be used by cities in their air quality assessment

The main motivation for applying data assimilation is to obtain a synergy effect towards improved assessment by combining air pollution dispersion models with observations as compared to just using them separately. For regional scale air quality assessments, this means primarily to combine a given regional scale air pollution dispersion model with available air quality observations

2.2 Aim and description

Cities need realistic information on the regional background concentration of air pollutants to assess the increment in pollution levels from the region to the city. The aim of this case study is to show the beneficial use of data assimilation to assess regional background concentrations on the European scale. Hence, we aim to provide consistent hourly modelled data over Europe using data assimilation.

2.3 Relevance to recommendations in Air4EU

In the report Air4EU-D 5.2: First recommendations for air quality assessment at regional/continental scales a number of recommendations have been formulated. In the case study described in this report the following recommendations have been addressed:

- Regional background for urban areas should be created by combining observations and modelling by passive as well as active data assimilation techniques.
- A procedure should be developed for operational data assimilation

In this study only active data assimilation is considered. Passive data assimilation is addressed in AIR4EU-CS Report D 7.1.13: Statistical combination of modelling an monitoring on the European scale.

The method described in this report is operational in the sense that city users and other users can use the determined air quality fields for 2003 as regional background. Air quality fields for other years can be made available also, provided that funding can be made available to produce these maps.

3. Methodology

3.1 Overview

Observations of air pollutants are irregularly distributed in space and time. Data assimilation allows the calculation of continuous fields in space and time from observations that are irregularly distributed. Data assimilation consists of making a best estimate of the state of the atmosphere on the basis of observations and a model prediction of the atmospheric state both of which have associated errors. Data assimilation basically defines a new atmospheric state by making a weighted average of the observed and modelled state in an intelligent and statistically sound way. Hence, if a model value is more uncertain than an observed value, more weight will be put on the observation, and the assimilated value will tend to get closer to the observed value and vice versa.

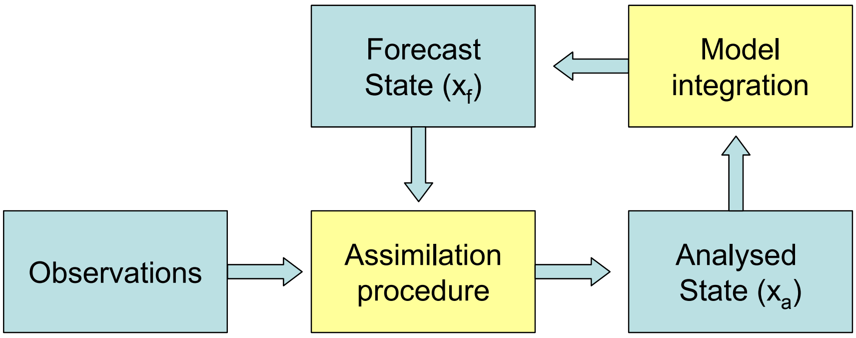

Figure A.3.1 Schematic representation of the data assimilation procedure.

In this study we used an ensemble Kalman filter to assimilate observations of O3 and PM10 within LOTOS-EUROS. The uncertainties involved with the modelled and observed values determine the weights that are put on the measured and calculated values. With an ensemble Kalman filter there is no need to specify the model uncertainties as they are determined by the range of modelled states of the ensemble members. Hence, the specification of the noise influences the weights and therewith results of the procedure. Below we describe the model, the generation of the ensemble and the measurements. The assimilation scheme is described in appendix A.

The assimilation experiments have been performed for the full year of 2003. Hence, all data discussed in this report are for 2003.

3.2 The LOTOS-EUROS model

LOTOS-EUROS (LE) is an Eulerian dispersion model for calculating atmospheric transport and deposition of air pollutants on a European scale. Below the main features of the LOTOS-EUROS model are described. For a detailed description we refer to Schaap et al. (2006).

Domain

The master domain of LOTOS-EUROS is bound at 35° and 70° North and 10° West and 60° East. The projection is normal longitude-latitude and the standard grid resolution is 0.50° longitude x 0.25° latitude, approximately 25x25 km. In this study we have used several domains within this master domain. The model code is structured such that zooming (up to a factor of 8) is possible. In the vertical there are three dynamic layers and an optional surface layer. The model extends in vertical direction to 3.5 km above sea level. The lowest dynamic layer is the mixing layer, followed by two reservoir layers. The height of the mixing layer is part of the diagnostic meteorological input data. The heights of the reservoir layers are determined by the difference between the mixing layer height and 3.5 km. Both reservoir layers are equally thick with a minimum of 50m. In some cases when the mixing layer extends near or above 3500 m the top of the model exceeds the 3500 m according to the abovementioned description. Simulations were performed with the optional surface layer of a fixed depth of 25 m. Hence, this layer is always part of the dynamic mixing layer. For output purposes the concentrations at measuring height (usually 3.6 m) are diagnosed by assuming that the flux is constant with height and equal to the deposition velocity times the concentration at height z.

Transport

The transport consists of advection in 3 dimensions, horizontal and vertical diffusion, and entrainment/detrainment. The advection is driven by meteorological fields (u,v) which are input every 3 hours. The vertical wind speed w is calculated by the model as a result of the divergence of the horizontal wind fields. The recently improved and highly-accurate, monotonic advection scheme developed by Walcek (2000) is used to solve the system. The number of steps within the advection scheme is chosen such that the courant restriction is fulfilled. Entrainment is caused by the growth of the mixing layer during the day. Each hour the vertical structure of the model is adjusted to the new mixing layer depth. After the new structure is set the pollutant concentrations are redistributed using linear interpolation.

The horizontal diffusion is described with a horizontal eddy diffusion coefficient following the approach by Liu and Durran (1977). Vertical diffusion is described using the standard Kz-theory. Vertical exchange is calculated employing the new integral scheme by Yamartino et al. (2004).

Chemistry

The LOTOS-EUROS model contains two chemical mechanisms, the TNO CBM-IV scheme (Schaap et al., 2005) and the CBM-IV by Adelman (1999). In this study we used the TNO CBM-IV scheme which is a modified version of the original CBM-IV (Whitten et al., 1980). The scheme includes 28 species and 66 reactions, including 12 photolytic reactions. Compared to the original scheme steady state approximations were used to reduce the number of reactions. In addition, reaction rates have been updated regularly. The mechanism was tested against the results of an intercomparison presented by

Poppe et al. (1996) and found to be in good agreement with the results presented for the other mechanisms. Aerosol chemistry is represented using ISORROPIA (Nenes et al., 1999).

Dry and wet deposition

The dry deposition in LOTOS-EUROS is parameterised following the well known resistance approach. The deposition speed is described as the reciprocal sum of three resistances: the aerodynamic resistance, the laminar layer resistance and the surface resistance. The aerodynamic resistance is dependent on atmospheric stability. The relevant stability parameters (u^*, L and Kz) are calculated using standard similarity theory profiles. The laminar layer resistance and the surface resistances for acidifying components and particles are described following the EDACS system (Erisman et al., 1994).

Below cloud scavenging is described using simple scavenging coefficients for gases (Schaap et al., 2005) and following Simpson et al. (2003) for particles. In-cloud scavenging is neglected due to the limited information on clouds. Neglecting in-cloud scavenging results in too low wet deposition fluxes but has a very limited influence on ground level concentrations (see Schaap et al., 2004a).

Meteorological data

The LOTOS-EUROS system is presently driven by 3-hourly meteorological data. These include 3D fields for wind direction, wind speed, temperature, humidity and density, substantiated by 2-d gridded fields of mixing layer height, precipitation rates, cloud cover and several boundary layer and surface variables. The standard meteorological data for Europe are produced at the Free University of Berlin employing a diagnostic meteorological analysis system based on an optimum interpolation procedure on isentropic surfaces. The system utilizes all available synoptic surface and upper air data (Kerschbaumer and Reimer, 2003). Also, meteorological data obtained from ECMWF can be used to force the model.

Emissions

The anthropogenic emissions used in this study are a combination of the TNO emission database (Visschedijk and Denier van der Gon et al., 2005) and the CAFE baseline emissions for 2000. For each source category and each country, we have scaled the country totals of the TNO emission database to those of the CAFE baseline emissions. Elemental carbon (EC) emissions were derived from (and subtracted from) the primary PM2.5 (PPM2.5) emissions following Schaap et al. (2004b). Hence, we use the official emission totals as used within the LRTAP protocol but we benefit from the higher resolution of the TNO emission database (0.25x0.125 lon-lat). The annual emission totals are broken down to hourly emission estimates using time factors for the emissions strength variation over the months, days of the week and the hours of the day (Builtjes et al., 2003).

In LOTOS-EUROS biogenic isoprene emissions are calculated following Veldt (1991) using the actual meteorological data. In addition, sea salt emissions are parameterised following Monahan et al (1986) from the wind speed at ten meter height.

Boundary conditions

The lateral boundary conditions used in this study were obtained by a simulation of the LOTOS-EUROS model with its full domain. Model top boundary concentrations were set to 0.8 ug/m3 for sulphate and ammonium was set to neutralise the sulphate. Other aerosol species were set to zero. Ozone concentrations are obtained from Logan (1999).

3.3 Ensemble generation

To create an ensemble of simulations for the kalman filter we have selected seven parameters to apply noise to (Table 3.1). We have selected the most important sources and some deposition values to add noise to. For O3 we specified the emissions of NOx, SOx and NMVOC to add noise to., as well as the dry deposition velocity. For PM the parameters were the emissions of SOx, NOx, NH_3, BC, PM2.5 and PM10 in combination with the dry deposition speed. The black carbon (BC) emissions were given the same noise as the primary PM2.5 since the BC fraction is better known than the absolute (total) emissions.

All noise factors were applied with a mean of 1 and a standard deviation of 0.25. Hence, the vast majority for the factors are within the range of 0.5 to 1.5. All noise factors were set after an assimilation step and held constant until the next assimilation step.

Parameter	O3	PM10
Emissions of NOx	x	x
Emissions of SOx	x	x
Emissions of NH3		x
Emissions of PM2.5 and BC		x
Emissions of NMVOC	x	
Dry deposition speed fine mode aerosol		x
Dry deposition speed of ozone	x	

Table 3.1. Noise parameters used to create an ensemble of model simulations for the assimilation of O3, NO2 and PM10

3.4 Measurements

Ozone

For assimilation in the model we use the ozone monitoring data from the EMEP network. There is a vast experience in measuring ozone in Europe. Moreover, within the EMEP protocol an extensive QA/QC procedure warrants high quality data. Hence, the uncertainty associated with ozone data are rather small. We use a fixed relative standard deviation of 10% in combination with an upper limit of 5 $\mu g/m^3$ following van Loon et al. (2001).

For validation we use the rural background ozone observations from the AIRBASE stations that report hourly values. It is important to note that the QA/QC procedures within AIRBASE are not as strict as within the EMEP network and that the classification of the stations is less unambiguous. Consequently, the quality of the data is expected to be somewhat less than for the EMEP data.

	O3	PM10
Assimilation set	EMEP	AIRBASE
Uncertainty (stdev)	10% (max = 5 ug/m3)	12.5% (max=10 ug/m3)

Validation set	AIRBASE	-

Table 3.2 Source and estimated uncertainty of the observational data to be assimilated in the model as well as the validation data set for all experiments

PM10

The EU reference method to measure PM_{10} concentrations is described in CEN standard EN 12341, adopted by CEN in November 1998 (CEN, 1998). It defines a PM_{10} sampling inlet coupled with a filter substrate and a regulated flow device. The mass collected on the filter is determined gravimetrically by means of a microbalance under well-defined environmental conditions. This is the reference method under the First Daughter Directive; it gives, by definition, the “correct” PM_{10} results.

However, for practical reasons (requirement of technique to be fully automatic), also other methods can be used if a Member State can demonstrate that it gives equivalent results or displays a consistent relationship to the reference method. In the latter case, results have to be corrected by a correction factor to produce results equivalent to the reference method. These correction factors can vary substantially in space and even seasonally. Differences between correction factors and the application itself hinder integration on a European scale of all PM_{10} data. An overview of correction factors used for PM_{10} data of 2002 in the AirBase database is given in Buijsman and de Leeuw (2004).

At this moment, the most commonly used techniques for measuring PM are gravimetry (i.e., the reference method), the beta-absorption technique and the Tapered Element Oscillating Microbalance (TEOM) technique.

- The gravimetry method is based on directly weighing the collected aerosols. Ambient air is pumped with a constant flow rate into a specially shaped inlet where particulate matter is separated into size fractions. The particulate matter is then collected on a filter and weighed in a temperature and humidity controlled environment.
- With the beta-absorption technique the amount of particles on the filter is determined by measuring the attenuation of a beam of beta-radiation (electrons) which are send through the filter. The attenuation is proportional to the mass of the aerosols on the filter.
- The TEOM makes use of the change in eigen-frequency of a tapered glass element that is connected to the filter. The change in eigen-frequency is determined by the mass of particles attached to the filter.

In Table 3.3, an overview is given of measurement methods used for PM_{10} and $PM_{2.5}$. Clearly, most PM_{10} measurements are performed with the beta-absorption technique. Measurement of PM mass concentrations are subject to considerable uncertainties mainly because of alterations of the air sample during the measurement process. Alteration of the air sample highly depends on the environmental conditions and composition of the particles. Loss of semi-volatile particles is the major problem. In most cases the results from beta-absorption instruments as well as the TEOM underestimate the concentration (e.g. Hitzenberger et al., 2004; Charron et al., 2004). The comparability of the PM mass measurements of these samplers has therefore been recognized as a major issue of concern (CAFE-WGPM, 2004).

Table 3.3 Measurement method and station type of the measurement locations.

	PM2.5	PM10
Measurement method		
Oscillating Microbalance	57	484
Gravimetry	15	284
Beta-absorption	12	604
Other/Unknown	4	244
Stationtype		
(Sub-)urban background	40	650
Rural background	8	187
Traffic	28	502
Other	12	277

In 2003, 28 European countries submitted their PM_{10} data to AirBase. Most stations only deliver daily averaged PM concentrations to Airbase. The meta-information in AirBase includes a description of the surroundings (rural, suburban of urban), the type of station (traffic, background, or other), the measurement method used, the altitude etc. We have only used the stations classified as rural background (see Fig 3.1). A number of elementary quality checks have been done on the AirBase data, such as removing data from stations with clearly erroneous latitude/longitude coordinates. A full quality check is recognised to be important but out of the scope of this feasibility study. A full quality check should include a detailed evaluation of the data quality and an assessment of the measurement uncertainty per (group of) stations. Here, we use a fixed relative standard deviation of 12% in combination with an upper limit of 10 $\mu g/m^3$.

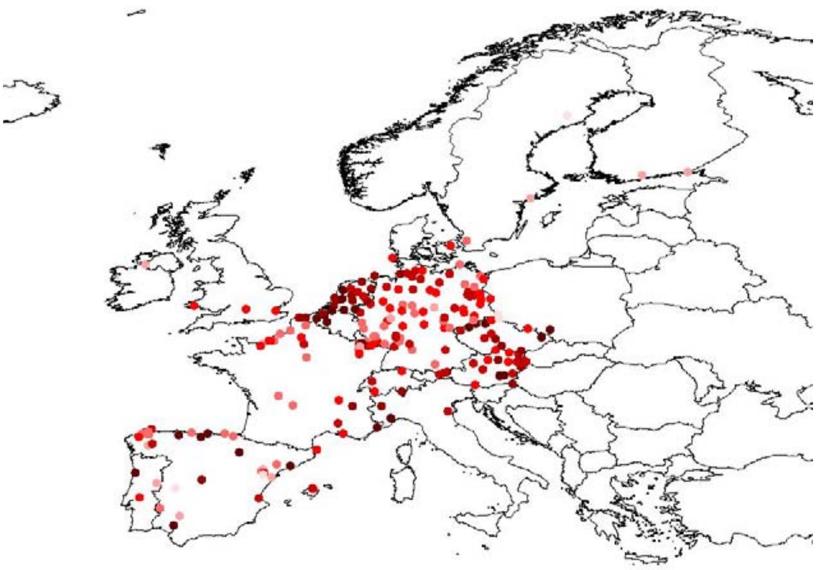

Figure 3.1 The regional AIRBASE stations measuring PM10 during 2003

3.5 Analysis of the results

The assimilation experiments are evaluated against observational data. For ozone the AIRBASE database provides a large data set of independent observations. For the other components, however, the data availability is low and all monitoring data are used for the assimilation. Only a few independent data sets were available to us. Hence, for PM10 we show the improvement at assimilation stations (see table 3.2).

The focus of the analysis of the simulations is on the correlation and RMSE values between the concentrations of the simulations and observations, as an indication of the change in temporal variability and absolute differences.

4. Results

4.1 Ozone

For ozone we have assimilated observations from the EMEP net work into the LOTOS-EUROS chemistry transport model. Below we present an overview of the results of the assimilation procedure.

4.1.1 Fields

In Figure 4.1 we present an example of the modelled and assimilated ozone distribution for july, 2003. The model predicts high ozone concentrations (>45ppb) over the Alpine region, Eastern Germany, Poland and the outflow areas over the North and Baltic seas. Minima are modelled over densely populated areas such as the Benelux and the UK. Furthermore, a large number of mega-cities such as Paris, London and Milan can be recognized in the modelled map.

LOTOS-EUROS tends to overestimate ozone during summer. Hence, the assimilation pulls the concentrations in the model down. The assimilated ozone field shows about 3-4 ppb less ozone than the modelled one. Also the patterns are affected. For example, the lower levels over northern Germany are incorporated due to the assimilation process. Also, the area with average levels above 50ppb is confined to the alpine region. Note that the influence of the assimilation is not only obvious close to the measurement stations but also in areas with few or no stations such as over the seas.

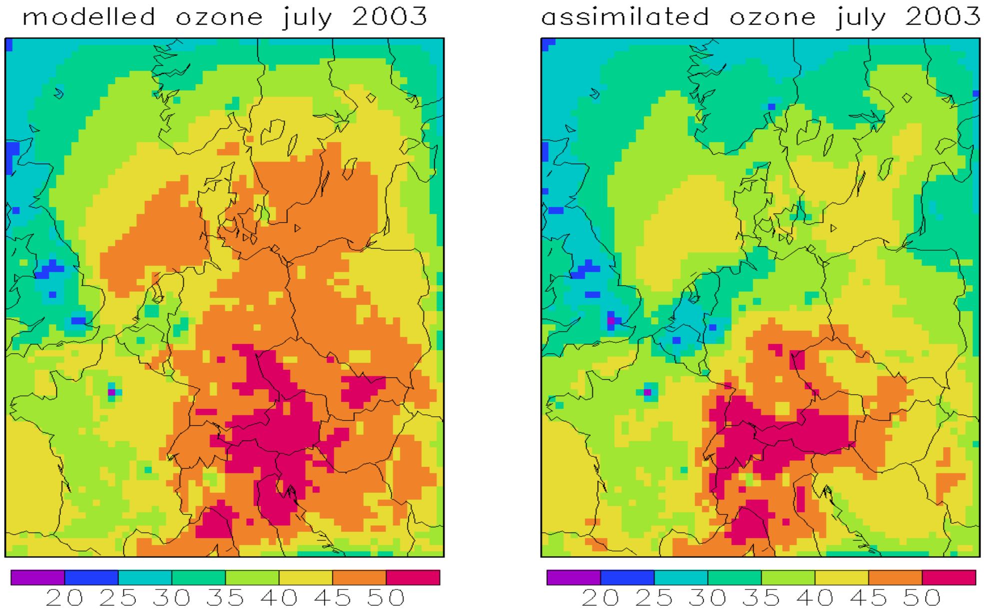

Fig 4.1 Modelled and assimilated ozone fields for July, 2003

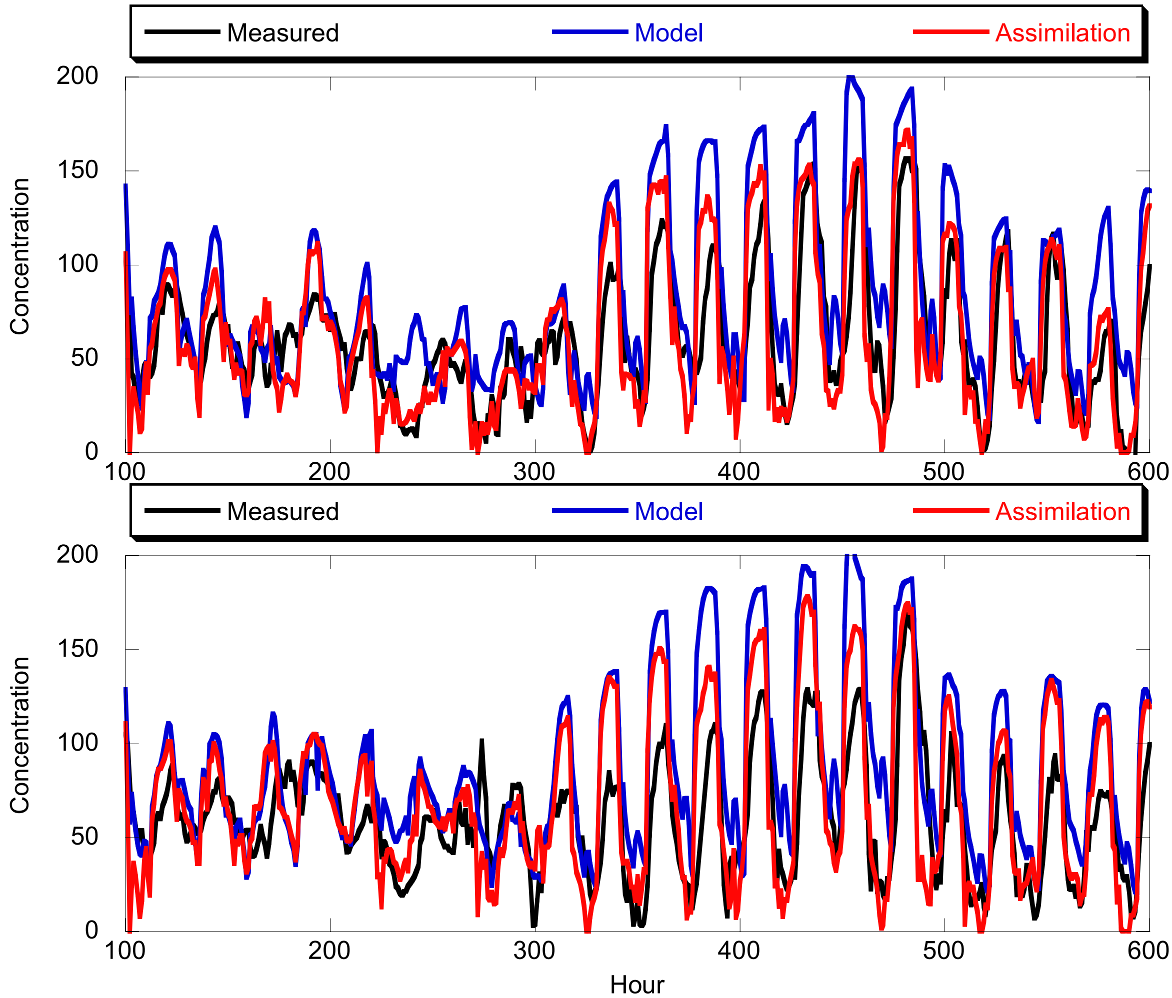

Fig 4.2 Time series of ozone ($\mu g/m^3$) for the Dutch stations Vredepeel (used in the assimilation) and Westmaas (validation station). An episode with an overestimation was selected to illustrate the influence of the assimilation.

4.1.2 Validation

The results of the assimilation experiment have been compared to independent observations of the AIRBASE network. In Figure 4.2 we show the influence of the assimilation procedure at an assimilation station, Vredepeel (NL), upper graph, and an validation station Westmaas (NL), lower graph. To illustrate the effect we chose an episode with a significant overestimation of the ozone concentrations in the Netherlands. Only two EMEP stations are used as assimilation stations in the Netherlands. At Vredepeel the assimilation pulls the concentrations towards the measured values causing a much better correspondence with the observations. At Westmaas, located close to Rotterdam, the assimilation experiment causes a better performance. The concentrations are effectively lowered by the assimilation, though not as much as at Vredepeel.

We have used all available (335 within the domain) rural background AIRBASE stations for comparison. The data are summarised in table 4.1 and shown per country in Figure 4.3. The assimilation significantly reduces the average residual and RMSE between model and observation, whereas the annual average is not affected as much. The better correspondence can be explained by the much higher temporal correlation in the assimilation experiment. The latter is evident and a measure for the successful application of the system to ozone.

Table 4.1 Statistical comparison between modelled and assimilated ozone concentrations at 335 AIRBASE stations for 2003.

	Measured	*Modeled*	*residu*	*RMSE*	*Correlation*
Model	82.1	83.7	26.5	33.3	0.51
Assimilation	82.1	79.8	22.5	28.8	0.65

The correlation between the modelled and observed ozone concentrations at AIRBASE sites is significantly lower than for EMEP sites. This is probably related to the less strict station classification and quality assurance within AIRBASE. Note that the AIRBASE data in principle give a better spatial coverage than the EMEP network. An attempt to clean the data from the AIRASE network and/or concisely assess the quality of the data would be very beneficial for validation and probably future assimilation experiments.

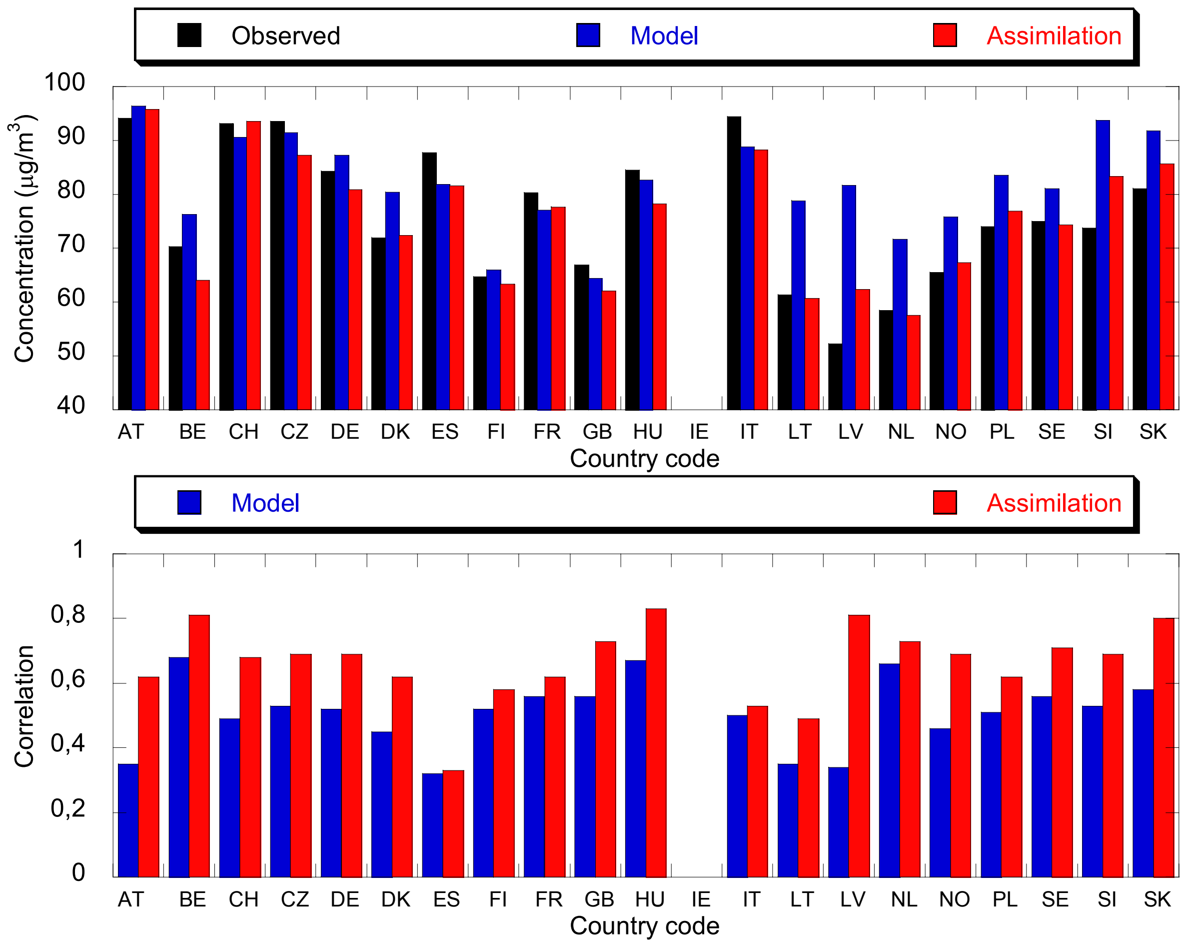

Fig. 4.3 Comparison of annual average ozone concentrations against measurements (a) and the correlation between model/assimilation and observations. All stations grouped per country

4.2 PM10

The application of the assimilation system to PM10 is hampered by a number of issues. One of these issues is a bias between modeled and measured PM10 concentrations. Current status of PM10 modelling is that the models underestimate the observed concentrations severely due to missing knowledge on processes and emissions to incorporate all components in the models. LOTOS-EUROS also shows this underestimation (see Figure 4.4). Hence, for PM10 a bias correction has been applied of a factor of 2. The application of this bias correction is a highly pragmatic approach.

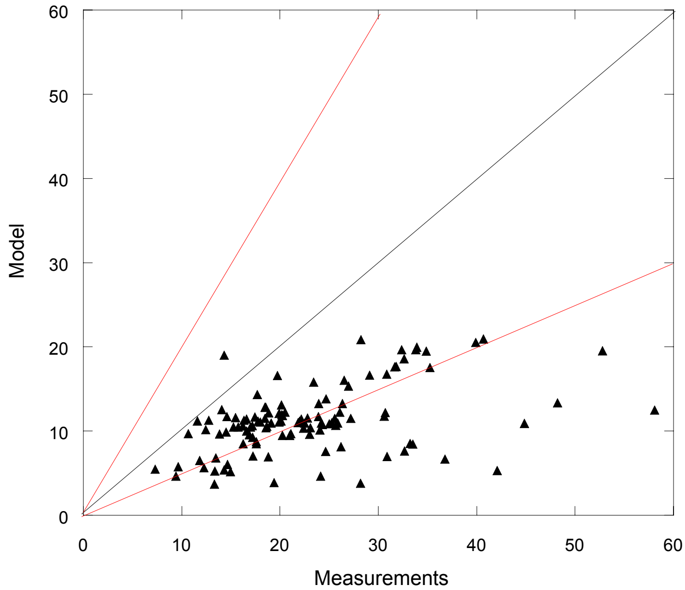

Figure 4.4 Comparison between measured and modelled PM10 concentrations in Europe for 2003

Another issue it that the observation network is inhomogeneous with respect to measurement methods and correction procedures. Hence, the data quality will be inhomogeneous as well. Furthermore, we expect that the different procedures per country or regions will affect the measured concentrations in a systematic way, causing discontinuity at borders. Consequently, the experimental design is flawed by the input data. Hence, we will present an example that shows that the system works but do not present an extensive results section.

In Figure 4.5 we show the PM10 concentration field for the spring season in which PM levels often show their maximum concentration. Through the bias correction applied in the procedure the PM levels in the assimilation experiment are about twice as high as in the model simulation. The assimilation changes the fine structures of the modelled distribution. For example, the Rhine valley in the north of Switserland pops up, northern Germany shows higher levels and the pattern over Denmark has changed with a more pronounced maximum around Copenhagen. The influence of the boundary conditions remains visible and may indicate that the boundary conditions may be a good candidate as a noise parameter in the system.

In Figure 4.6 we show the time series for the stations Vredepeel (NL10) and Kolummerwaard (NL09) in the Netherlands. For March and April two periods of high PM10 concentrations can be identified. The model captures these periods but underestimates the concentrations severely. The assimilation uses a bias correction of a factor 2, which already closes a large part of the gap, but does not affect the temporal behaviour. The assimilation clearly improves the temporal evolution as is clearly seen for the episodes in march, 2003. In short, as for ozone the assimilation improves especially the temporal correlation.

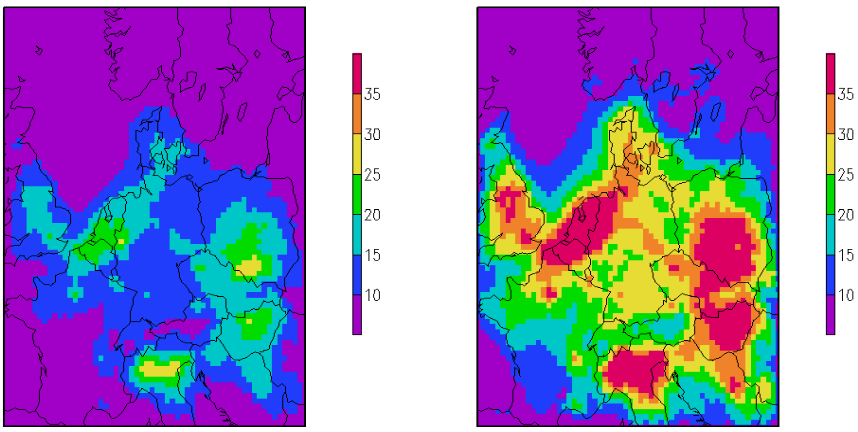

Fig 4.5 Modelled and assimilated PM10 fields for Spring, 2003

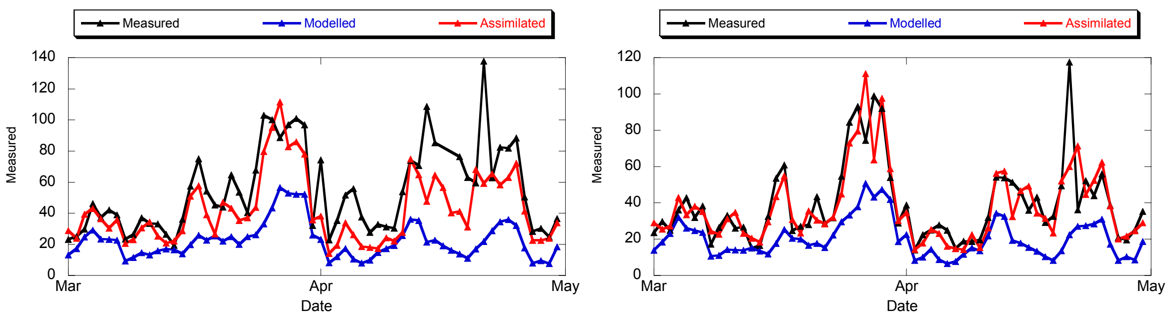

Figure 4.6 Time series of measured, modelled and assimilated PM10 at Vredepeel (left) and Kolummerwaard (right), the Netherlands. Both stations are assimilation stations.

5. Conclusion and discussion

5.1 Assessment of the case study

Within this case study the following products are available:

1. A system that is able to assimilate observations of O3, NO2 and PM10/2.5 from the EMEP and AIRBASE networks.
2. Regional background fields of O3 and PM10 with an hourly and daily temporal resolution for central Europe for 2003.

The validation of the results shows that the assimilated fields are in better agreement with independent observations compared to the stand alone model. Especially the temporal correlation is significantly improved by the assimilation. The produced air quality fields for O3 and PM10 are available for use as Regional Background Concentrations by City Users.

For use as boundary conditions one has to consider that the regional model also incorporates the emissions of the city itself. As a consequence, the cells in which the city is located are not usable for assessing boundary conditions. In addition, to properly account for recirculation of air one has to take the data at some distance of the city, preferably several 10s of Kilometres. The CITYDELTA exercise (Cuvelier et al., 2007) used an area of 300x300 Km2 centred around the city for the local scale modelling and obtained the boundary conditions from a regional model. From the experience obtained in this exercise we estimate that a distance of 100m Km from a large city would be ideal. Note that the consistency between emission data for the region and the city has to be warranted.

A similar issue plays a role when the derived fields would be used as the regional background for a city, to which the local contributions are added. In that case, the impact of the city emissions would be double counted. An option would be to use the model output from the second model layer, as we expect that the city influence is much lower for the mixing layer as a whole compared to the surface layer. However, the influence is still not negligible and for large cities the impact would still be highly significant. For large cities it may be possible to perform dedicated simulations in which the emissions of the city are removed to avoid the double counting.

Regional air quality models underestimate the observed PM10 concentrations. From a modelling point of view some underestimation can be expected since not all existing particle types are included in the model, and several sources are not very well known, e.g. fugitive dust, secondary formation of carbonaceous aerosols, and biomass burning sources. Previous evaluation of model results has shown that the concentrations of elemental and organic carbon are underestimated by a factor 2 (Schaap et al., 2004). Hence, the total PM10 concentration is underestimated. The resulting bias has been dealt with in the assimilation procedure in a very simple and pragmatic way. Ideally, the models should not show biases and should incorporate all relevant processes and sources. For PM10 this is not yet the case. To solve this issue a large effort is needed from the European community. Note that the underestimation would be lower for PM2.5 compared to PM10 as most of the missing mass is found in the course mode particles. Hence, assimilation of PM2.5 (when enough measurement come available) appears to be more promising than that for PM10.

The success of an assimilation experiment is determined by the quality of the input data, the model uncertainty and the correct implementation of the assimilation procedure. The model uncertainty is generated through an ensemble in which noise is added to key parameters. The noise specification is subjective and is based on the expertise of the modeler. Furthermore, the system requires detailed information about the quality of measurements. At present, the detailed information on uncertainty estimates for each measurement point is not available. For networks with a well defined measurement protocol the estimate of uncertainty may be performed in a generalized manner. Especially, for PM and its components these protocols are not well developed and hamper the assessment of data quality. At present, the AIRBASE data comprises a wealth of data on PM. An effort to screen these data in detail would be highly valuable for the assessment of the European PM distribution.

5.2 Improvements in assessment derived from case study

This study is not a case study in the sense that it is aimed to apply the AIR4EU recommendations to an assessment protocol of a European city. Hence, there is no assessment to compare the results to. In general, however, the use of data assimilation improves an assessment by definition. Assessing the background concentrations with measurements alone is not possible as one cannot approach a continuous coverage. Hence, one relies on modelling to make consistent regional background concentrations. By assimilation of observations the performance of the model is improved as we have shown above. Hence, besides the application for boundary conditions the application for a European wide air quality assessment benefits greatly from these fields.

5.3 Recommendations resulting from the case study

Based on the experience gained in this case study we recommend:

- To use the produced background conditions in combination with a high resolution urban model in which the city as well as the area around the city of about 100 Km is also represented.
- To try and improve the PM10 modelling (e.g. by incorporation of crustal matter and improved emission estimates).
- To attempt to clean the data from the AIRBASE network and concisely assess the quality of the data. This would be very beneficial for validation and probably future assimilation experiments.
- To launch a study to address the impact of the developed system on a cities air quality assessment using a detailed city scale model.

5.4 Suitability for implementation in other cities

The use of this system to provide regional background conditions to cities requires the use of a urban scale modelling system and requires a degree of experience of the application of models and assimilation techniques. Therefore, the approach seems to be suited for middle to large sized cities.

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Appendix A. Ensemble Kalman Filter

The first step in order to build the Ensemble Kalman Filter around LOTOS-EUROS is to embed the model and the available measurement in a stochastic environment:

$$x^{k+1} = f^k(x^k, w^k)$$
$$y^k = C^k x^k + v^k,$$

where the superscripts (k) denote the time-steps. The model state vector is denoted by *x* and the measurements by *y*. The function *f* denotes the non-linear model operator which apart from on the state vector acts on a white noise vector *w* with Gaussian distribution and diagonal covariance matrix *Q*. The measurement vector *y* is assumed be a linear combination of elements of the state vector and a random, uncorrelated Gaussian error *v* with (diagonal) covariance matrix *R*. The basic idea behind the ensemble filter is to express the probability function of the state in an ensemble of possible states $\{\xi_1, ..., \xi_N\}$, and to approximate statistical moments with sample statistics:

$$\hat{x} \approx \frac{1}{N}\sum_{j=1}^{N}\xi_j$$

$$P \approx \frac{1}{N-1}\sum_{j=1}^{N}\left(\xi_j - \hat{x}\right)\left(\xi_j - \hat{x}\right)^T$$

where the pair ($\hat{x}$,*P*) (expectation and covariance matrix) describe the probability of the state vector *x* completely if *x* has a Gaussian distribution. Since we are dealing with strongly non-linear models, it cannot be expected that *x* really has a Gaussian distribution. We assume however that the distribution is at least close to Gaussian so that the bulk of the statistical properties is captured by the pair ($\hat{x}$,*P*). The filter algorithm consists of three stages:

<u>initialisation</u>:
each ensemble member is set to the initial state:

$$\xi_j = x^0$$

<u>forecast</u>:
each ensemble member is propagated in time by the model, where the noise input w^k is drawn from a random generator with covariance Q;

$$\xi_j^f = f(\xi_j, w^k)$$

analysis:
given an (arbitrary) gain matrix K, each ensemble member is updated according to:

$$\xi_j^a = \xi_j^f + K(y + \nu - H^T \xi_j^f)$$

where v_j represents a measurement error, drawn from a random generator with zero mean and covariance *R*. The gain matrix *K* is given by the optimal gain matrix from the original Kalman Filter. In the original filter the Kalman gain was obtained by matrix multiplications in which the covariance matrix *P* is involved. Fortunately, the use of this matrix can be avoided, since this matrix is too large to store into memory. Instead, a square root *S* (such that $P=SS^T$) can be used. From the definition of *P* it can be seen that the columns s_i of such a square root can be defined by

$$s_i = \frac{1}{\sqrt{N-1}}\left(\xi_i - \hat{x}\right).$$

Note that the sample mean $\hat{x}$, and the matrix *S* completely define the ensemble and vice versa; it is therefore not necessary to store both *S* and the ensemble.

The analysis of the measurements y_j (entries of the vector *y*) can now be performed by the following sequential procedure (dropping the time index):

$$\begin{aligned}
h_j &= S_{j-1}^T c_j^T \\
a_j &= \left(h_j^T h_j + r_{jj}\right)^{-1} \\
b_j &= \left(1 + \sqrt{a_j r_{jj}}\right)^{-1} \\
k_j &= a_j S_{j-1} h_j \\
S_j &= S_{j-1} - b_j k_j h_j^T \\
x_j &= x_{j-1} + h_j\left(y_j - c_j x_{j-1}\right)
\end{aligned}$$

The index *j* is the iteration index. The starting values for the procedure are

$S_0 = S_f^{k+1}$ and $x_0 = x_f^{k+1}$.

After the analysis of all the measurements the final values for the state vector and (square root of) the covariance matrix have been obtained: $S^{k+1} = S_m$ and $x^{k+1} = x_m$. For a detailed description we refer to van Loon et al. (2000) and Evensen (1997).

The forecast step is the most expensive part of the algorithm, since for each ensemble member the model has to be evaluated one time. Typical ensemble sizes range from 10-100. If the number of measurements is limited (in order of hundreds), the total computation time involved with the ensemble filter is proportional with the ensemble size.

Random noise

In the model implementation used in this study, the noise parameters are part of the model state. Hence they are estimated by the filter as well. The noise parameters w_i can be interpreted as emission correction factors since the actual emission field E_j is estimated by the filter as

$$E_i \leftarrow E_j\ (1+w_i).$$

This approach has the disadvantage that there is no "memory" in the system: the w_i are uncorrelated in time; at a certain hour *t* the noise parameter may indicate an emission increase of 20% with respect to the original field, whereas it estimates a decrease of 20% at *t+1*. Such irregular behaviour can be prevented to a large extent by the use of coloured noise. However, in the present set-up we use the same noise factors for the period between retrievals.

Spatially limiting influence of measurements

For two reasons correlations between elements in the state vector arise which are unlikely to be correlated. Firstly, spurious correlations arise, mainly because the sample size is finite. Secondly, undesired correlations arise due to the choice of the noise processes. The noise processes to be introduced in this study are all acting on emission fields of various emitted compounds causing "instantaneous" correlations throughout the domain. For example the particulate matter concentration at hour *t* somewhere in The Netherlands becomes correlated with the particulate matter concentration in, say, the south of France, because noise was added to the NO_x emission field at hour *t*-1. Although this is exactly what should happen when defining noise in this way, such correlations are not realistic and should be somehow ignored by the filter. The noise processes is chosen this way because it is infeasible to subdivide the emission fields into a number of sub-domains on each of which a different noise parameter is acting. That would increase the dimension of the noise vector dramatically and hence the necessary ensemble size to capture the statistical properties.

One way to ignore unrealistic correlations over large distances is the use of a gain matrix which is only unequal to zero around the locations of observations. Such a gain matrix k may be formed using a covariance matrix which is an element wise product of the original sample covariance and a correlation function with local support. For a single scalar measurement, the resulting gain matrix is given by (omitting the subscripts):

$$k = I(\rho)Ph/(h^T Ph + r)$$

where $I(\rho)$ is a diagonal matrix; the diagonal elements are filled with a prescribed correlation between the corresponding grid cell and the grid cell of the measurement. Different choices for the values of ρ_i are possible. In this study we take

$$\rho_i = \exp(-0.5\ (r_i/L)^2) \qquad \text{for } r_i \leq 3.5\ L$$

and zero otherwise. r_i denotes the distance from the grid cell considered to the site of the analysed measurement and *L* denotes a length scale parameter, taken to be 100Km in this study.