Chemical Data Assimilation for Air Quality Forecasting

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1 Introduction

Unlike the typical design of data assimilation for numerical weather forecasting, initial value optimisation by chemical data assimilation for air quality simulations were often considered as unessential, as errors in initial values were regarded as of vanishing impact. Rather, air surface interactions, especially emissions are a driving forcing factor, while, at the same time, of insufficient knowledge. This fact necessitates to generalise the air quality data assimilation problem to an inversion problem, which calls for a spatio–temporal, that is, model based technique. The paper presents a four–dimensional variational data assimilation implementation, along with a variety of assimilation examples.

2 Objectives of air quality data assimilation

The overarching aim of air quality data assimilation is to find a best estimate of the control parameters for those processes of the atmosphere, which govern the chemical evolution of biospherically relevant height levels. As in general data assimilation, where only sparse observations are available, we have to resort to modelled fields, in order to complement our lack of knowledge, where numerical models also serve as system constraints. In artificial intelligence parlance, system evolution constraints as induced by adopted formulation of differential equations is sometimes referred to as procedural knowledge (a model must be run), while model forecasts, as well as observations, are termed as 'declarative knowledge'.

As this does not differ from the problem of meteorological data assimilation, developments of similar methods are on the agenda of various research groups. Techniques range from attempts to use nudging to spatiotemporal methods like the four-dimensional variational data assimilation and Kalman filtering, the latter with different methods for complexity reduction.

However, there are a variety of aspects to be considered, which differ considerably from traditional atmospheric data assimilation. These include:

- The number of parameters per gridpoint: while there are 4 - 10 in meteorology, (depending on how complete the states of water are resolved by microphysical processes,) a state of the art air quality model prognoses more than 50 constituents in gas phase only. If aerosol dynamics and chemistry is included, this number is easily doubled.

- The completeness problem: with the above mentioned figures only the presumably most important constituents are enclosed. Especially hydrocarbons, referred to a volatile organic compounds (VOC) occur in a variety which cannot be accounted for in a complete way. Further, aerosol particles are even more different in size, shape, chemical composition and complexity of reactions.

- The spatial scale problem: generally, local air quality is forced by local emissions. This includes vertical transport as well. As a consequence, a scale gap should be bridged from simulating long range, even intercontinental transport of pollutants, down to a proper representation of emissions effectuated by point and line sources like stacks and streets. In practice, different chemical regimes prevail on short spatial scales. Sinks are acting by surface uptake from soil and vegetation, again imposing a much finer pattern as mesoscale meteorological features typically show.

- The surface interaction problem: emissions are not only a problem of scale, but also an issue of both impact on model results and paucity of knowledge. Consequently, emission rates must be considered as optimisation parameter. By the like arguments, deposition velocities could also be considered as optimisation quantity.

- The observation suite problem: in routine operation, legacy surface in situ observations are sparse and hampered by the representativity problem in populated areas. Anisotropic and heterogeneous formulations of the radii of influence are highly advisable (Hoelzemann et al., 2001). Ozone radiosonde network is even much sparser. Tropospheric satellite data are limited to very few species and often only given in terms of tropospheric columns. Available are nitrogen dioxide, elevated levels of sulphur dioxide and formaldehyde, mostly retrieved from GOME (ERS2 platform) or SCIAMACHY (ENVISAT) (e.g Eskes and Boersma, 2003, Heue et al., 2005). Also carbon monoxide soundings from MOPITT sensors are given (Deeter et al., 2003). First attempts of neural network retrieved ozone profiles are available (Müller et al., 2003). Finally, in situ observations made on board of commercial aircraft, for example in the frame of the MOZAIC activity (Thouret et al., 2000) can be assimilated. Other data sources are mainly restricted to spatially and temporally limited campaigns.

We seek for a data assimilation algorithm, which is able to combine the suit of heterogeneous observations, scattered in space and time, having variable spatial and temporal representativity with a sufficiently resolving air quality model system. This invokes the application space-time data assimilation algorithms preserving the BLUE property (Best Linear Unbiased Estimator) (Talagrand, 1998).

3 Theory

The problem description given above has clear implications for the data assimilation methodology to be selected. Chemistry–transport models (CTMs) do not passively accept external 3–dimensional state analyses, where no care is taken of chemical balance or naturally forced imbalances. The effect is to engender spurious relaxations toward some other chemical state. As a solution, models can contribute with their chemical kinetics as constraint to estimate both a balanced and most probable state or parameter values, at least theoretically providing for the BLUE property. Using data assimilation algorithms with the BLUE property, allow for hypothesis testing. There are two families

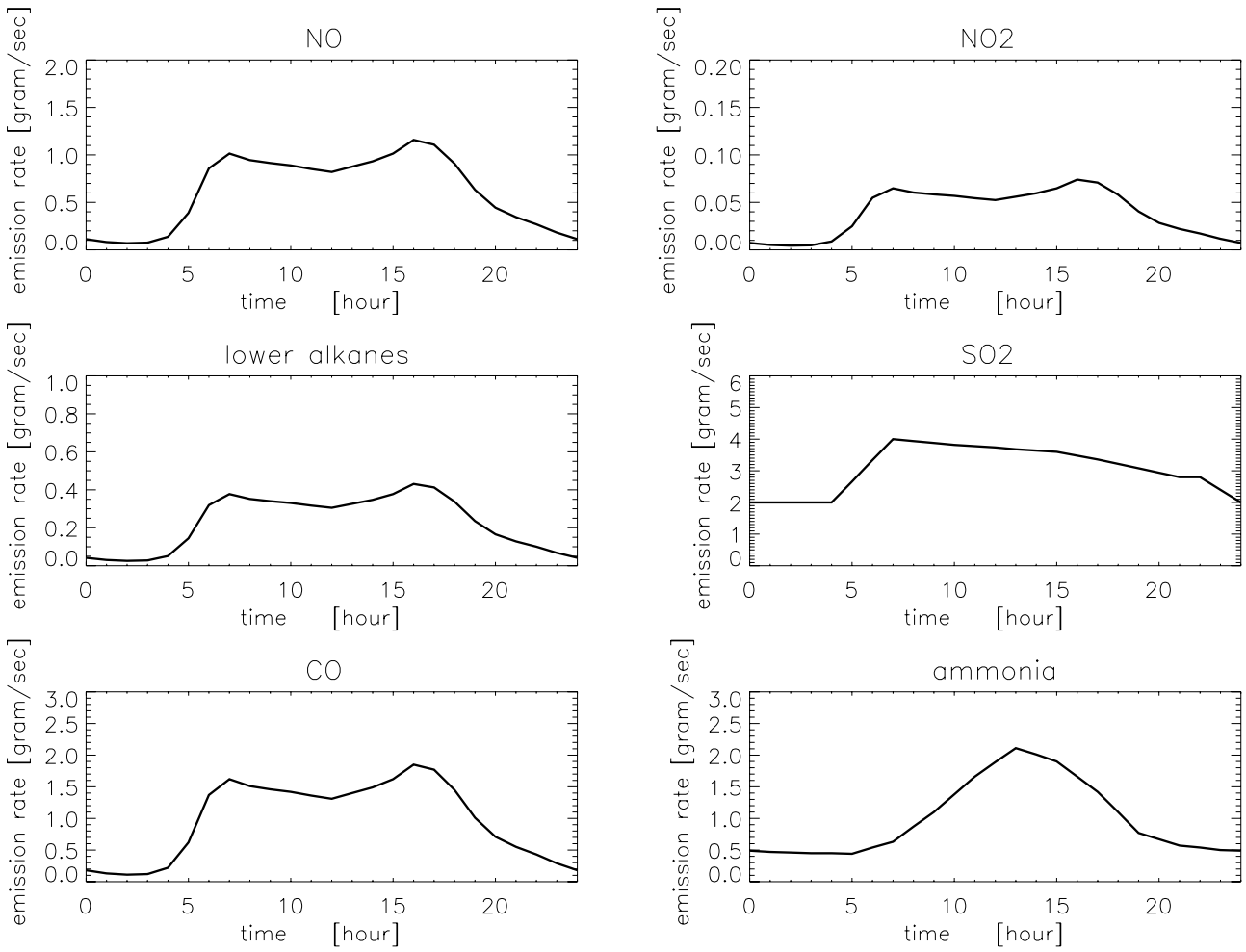

Figure 1: Diurnal profiles applied in the emission module for six of the 19 emitted species, the amplitude of which is taken as emission factor for optimisation.

of algorithms with this property in a spatio–temporal context: the four-dimensional variational data assimilation (4D–var), and Kalman filtering. Examples for spacio-temporal BLUEs applied in tropospheric chemistry in the case of 4D–var include Elbern and Schmidt (1999, 2001), with the EURAD model and Issartel and Baverel (2003) with POLAIR. Practical Kalman filtering can only be made with drastic complexity reduction measures. Most prominent techniques here are ensemble Kalman filtering and Reduced Rank SQuare root Kalman Filtering. Pioneering examples are given in van Loon et al,(2000) and Hanea et al., (2004).

It should be noted that 3–dimensional BLUE algorithm analyses like those from Optimal Interpolation, repeatedly ingested into a model, do not result in a four–dimensional BLUE analysis.

This can be formulated as follows: Deviations of the background chemical state $\mathbf{x}(t_0) - \mathbf{x}_b = \delta\mathbf{x}(t_0)$ and the emission inventory $\mathbf{e}(t_0) - \mathbf{e}_b = \delta\mathbf{e}(t_0)$ may be combined to define an incremental formulation of a cost function, objective function or distance function $\mathscr{J}$ as follows (see for example Elbern et

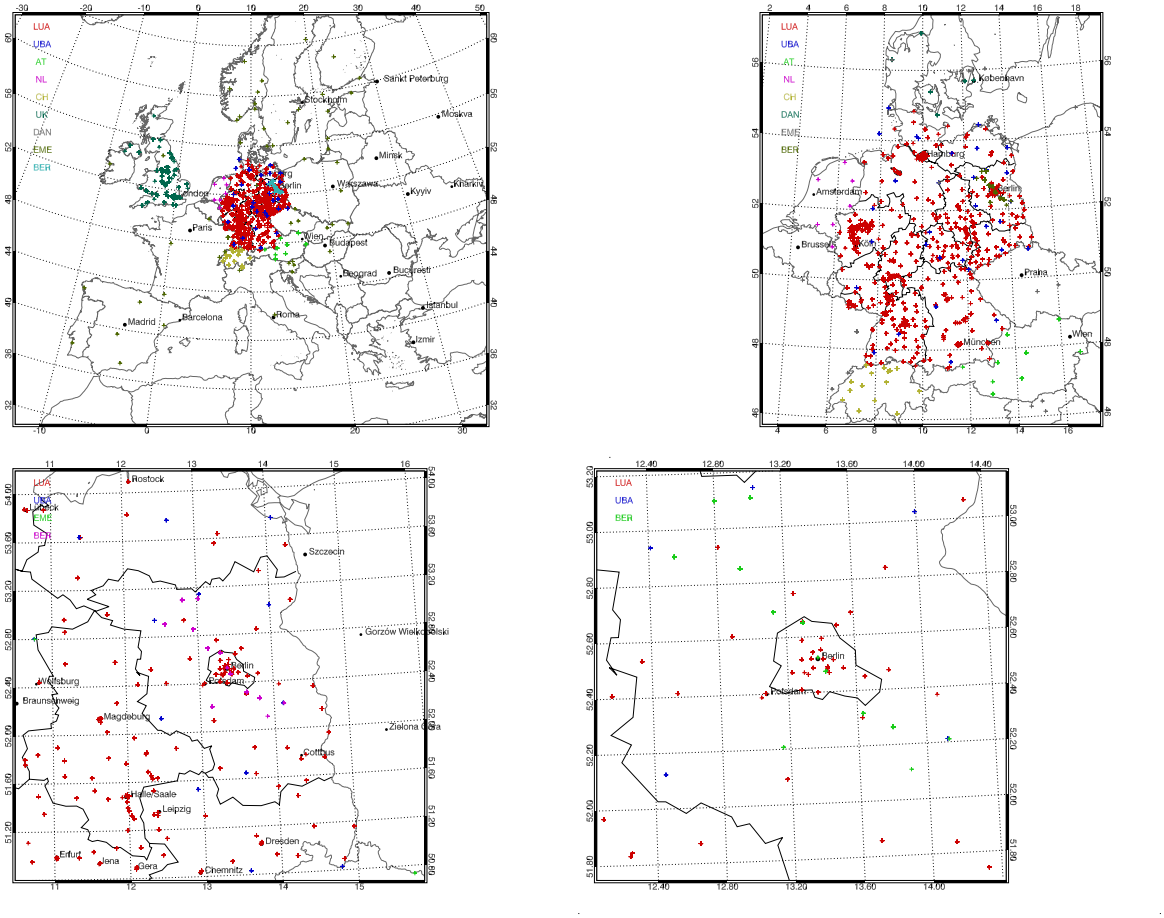

Figure 2: Nested integration domains with observation sites for the BERLIOZ case.

al, 2000 for a more detailed description):

$$\begin{aligned}\mathscr{J}(\delta\mathbf{x}(t_0),\delta\mathbf{e}) =& \frac{1}{2}(\delta\mathbf{x})^T\mathbf{B}^{-1}\delta\mathbf{x} + \frac{1}{2}\int_{t_0}^{t_N}(\delta\mathbf{e})^T\mathbf{K}^{-1}\delta\mathbf{e}dt + \\ & \frac{1}{2}\int_{t_0}^{t_N}(\mathbf{d}(t)-\mathbf{H}(t)\delta\mathbf{x}(t))^T\mathbf{R}^{-1}(\mathbf{d}(t)-\mathbf{H}(t)\delta\mathbf{x}(t))dt\end{aligned} \tag{1}$$

where $\mathscr{J}$ is a scalar functional defined on the time interval $t_0 \le t \le t_N$ dependent on the vector valued state variable $\mathbf{x}(t)$. $\mathbf{d}(t) := \mathbf{y}(t) - \mathbf{H}(t)\delta\mathbf{x}_b(t)$ is the observation minus model discrepancy at time t, when first guess initial values and emission inventory values are taken. The error covariance matrices are defined as follows: for the first guess or background values $\mathbf{B} \in \mathbb{R}^{N\times N}$ with N number of model variables, for the emission factors $\mathbf{K} \in \mathbb{R}^{E\times E}$ with E number of emitting grid points times emitted species, and of observation errors are denoted $\mathbf{R} \in \mathbb{R}^{M(t)\times M(t)}$, with $M(t)$ the number of available observations at time t. Operator $\mathbf{H}(t)$ calculates the model equivalent to each observation.

We want to determine the gradient of $\mathscr{J}$ with respect to the joint chemical state and emission rate variable $\mathbf{z} = (\delta\mathbf{x}, \delta\mathbf{e})^T$, and find as gradient $\partial\mathscr{J}/\partial(\delta\mathbf{x}, \delta\mathbf{e})^T$. The gradient of the cost function $\mathscr{J}$

then reads

$$\partial \mathscr{J} / \partial (\delta \mathbf{x}, \delta \mathbf{e})^T = -\mathbf{B}^{-1}(\delta \mathbf{x}(t)) - \sum_{t_0}^{t_N} \mathbf{M}^T(t_0, t)\mathbf{H}^T(t)\mathbf{R}^{-1}(\mathbf{d} - \mathbf{H}(t)\delta \mathbf{x}(t)) \\ - \sum_{t_0}^{t_N} \mathbf{K}^{-1}(\mathbf{e}_b(t) - \mathbf{e}(t)), \tag{2}$$

where $\mathbf{M}^T(t_0, t)$ denotes the adjoint (= transposed T) model operator, formally integration from time t backward in time to the initial time t_0. This optimisation problem can be solved by some quasi–Newton minimisation procedure, like L–BFGS.

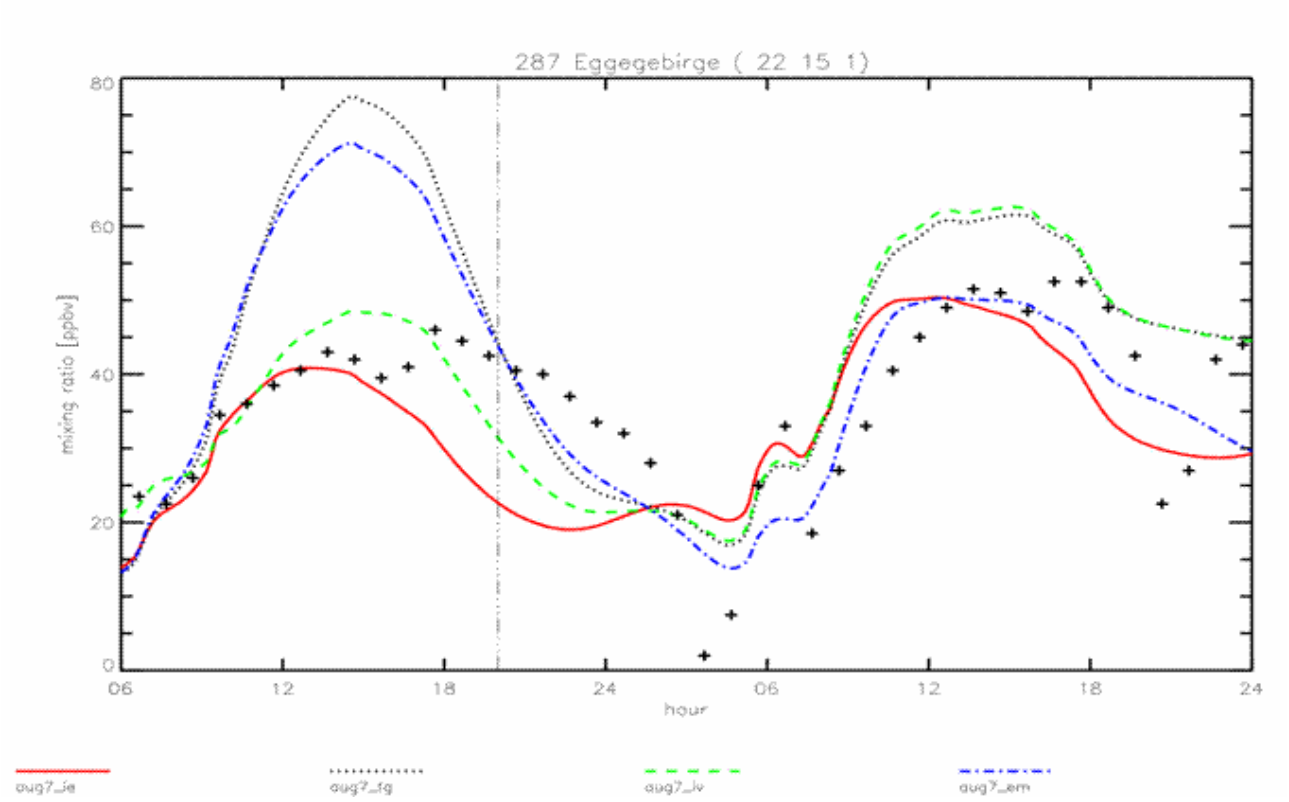

Figure 3: Assimilation results and performance at the ozone measurement site Eggegebirge for August 7 and 8, 1997. Black crosses: observations, left of vertical line at August 7, 20:00 taken for assimilation, later observations for forecast verification only. Reference forecast: black dotted line, initial value optimisation only: green dashed line, emission rate optimisation: blue dash-dotted line, joint emission rate / initial value optimisation: red bold line.

4 Implementation of a chemical 4D-var system

Model description

The EURAD CTM2 used in the following exposition is a comprehensive tropospheric Eulerian model operating on continental to local scale (Jakobs et al., 2005), presently also in an hemispheric mode (www.eurad.uni-koeln.de). The chemistry transport model calculates the transport, diffusion, and

gas phase transformation of about 60 chemical species with 158 reactions. The associated adjoint operators include the gas phase mechanism, the transport schemes and an implicit vertical diffusion scheme. The emission data in this study are taken from EMEP (co-operative programme for monitoring and evaluation of the long range transmission of air pollutants in Europe) and further processed as presented in Memmesheimer et al. (1995).

4D–var implementation

The variational chemistry data assimilation algorithm is composed by four components: (1) the forward model, (2) the adjoint of its tangent linear version, (3) the background error covariance matrix, and (4) the minimisation routine. Ozone records of about 400 surface observation sites were available for each day, most of which concentrated on a central European domain (Figure 2). Estimated observation accuracy, to be included in the observation error covariance matrix, is about 10% or at least 2 ppbv for ozone. An additional portion of the error is assumed to be due to the poor spatial representativeness as implied by a horizontal resolution of 54 km. This is a special problem for the use of urban and suburban measurement sites, although observations close to areas with elevated traffic load were omitted. (For more details see Elbern et al., 2000, and Elbern and Schmidt, 1999, 2001).

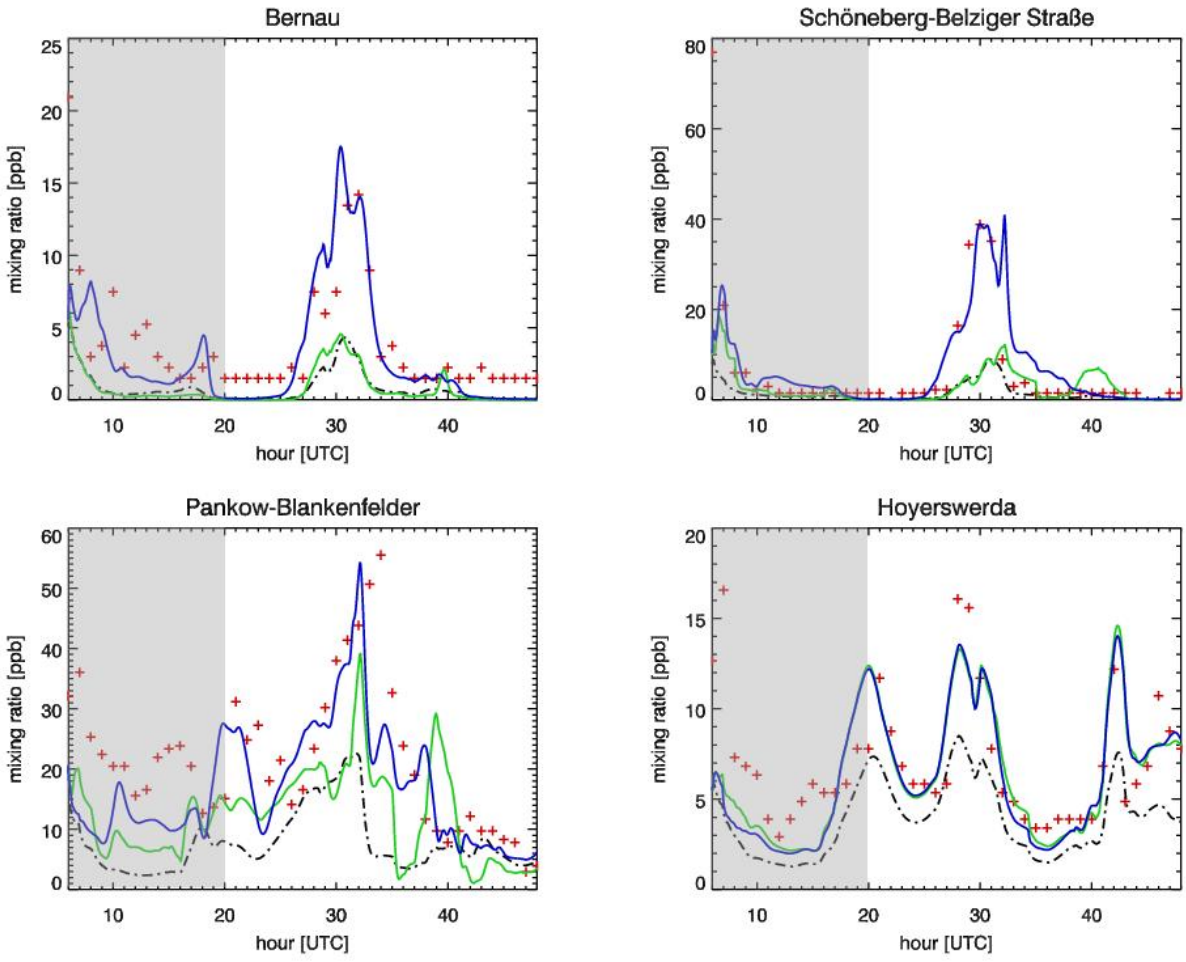

Figure 4: Time series for selected NOx stations (upper panel NO, lower panel NO2) on nest 2. Red crosses: observations, black line: no assimilation, green line: N1 assimilation (18 km resolution), blue line: N2 (6 km resolution) assimilation, grey shading: assimilation interval with assimilated observations, others: forecasted.

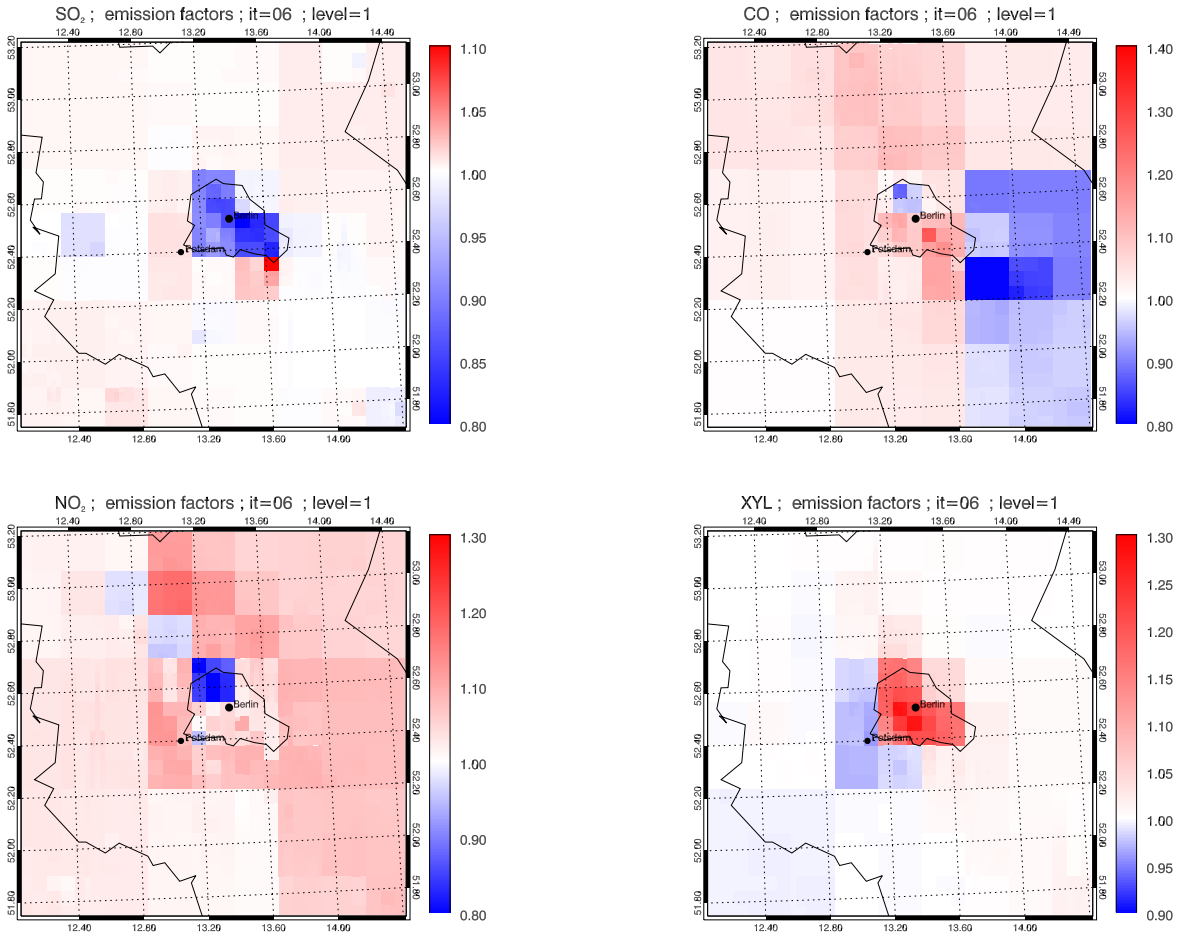

Figure 5: Optimised emission factors for the nest 3 area for SO_2, CO_2, NO_2*, and xylene. Red areas indicate the emission inventory to be increased, blue to be diminished by the colour coded factors. Coloured squares indicate the different nest levels of optimisation.*

5 Some Example Results

The 4D–var technique allows for the assimilation of a wide variety of data types, a cursory demonstration of which will be given. A careful estimation of the error of (spatial) representativity is however prerequisite for success. Specifically, model grid resolutions of about 50 km, widely used for continental scale integration domains, admit only for a limited number of species to be assimilated by point measurements. For example, quickly oxidizing point and line source emitted $NO_x \in \{NO, NO_2\}$ should only be assimilated by, rarely available, observation sites situated at background locations. In practice, gaseous constituent assimilation in coarse grid models mostly rests on ozone observations.

The selected coarse grid case study features a long lasting episode of elevated ozone levels over central Europe, which took place from 3 to 20 August 1997. The mesoscale meteorological simulations of this time span are made by MM5 which is restarted every 48 hours starting from August 1, 0000 GMT to August 20, 2400 GMT. Meteorological initial and boundary values were taken from ECMWF analyses. The model grid encompasses a greater European area, with 54 km horizontal grid resolution. The assimilation interval is selected to span the time from 06 to 20 UTC.

A coarse grid example

To demonstrate the feature of joint emission rate – initial value inversion, the semi–rural observation site "Eggegebirge" is presented, where both parameter types are of like importance for a two day forecast: the site has minor emission sources in its vicinity, but polluted air can be advected from within some 100 km distance. Figure 3 displays an unsatisfying skill of a prediction without assimilation, while pure initial value optimisation exhibits a good performance during the time of the assimilation window, the ensuing forecast on the second day relaxes toward the forecast without assimilation, clearly demonstrating a short "chemical memory" of the system. On the other hand, pure emission rate optimisation exhibits a clear positive impact at the second day, without significant improvements during the assimilation interval. This is due to the fact that the local emission rates are low, but also due to the delay caused by the oxidation time from precursors for ozone production. A joint emission rate – initial value inversion combines both positive impacts and clearly leads to the desired effect of forecast skill improvement.

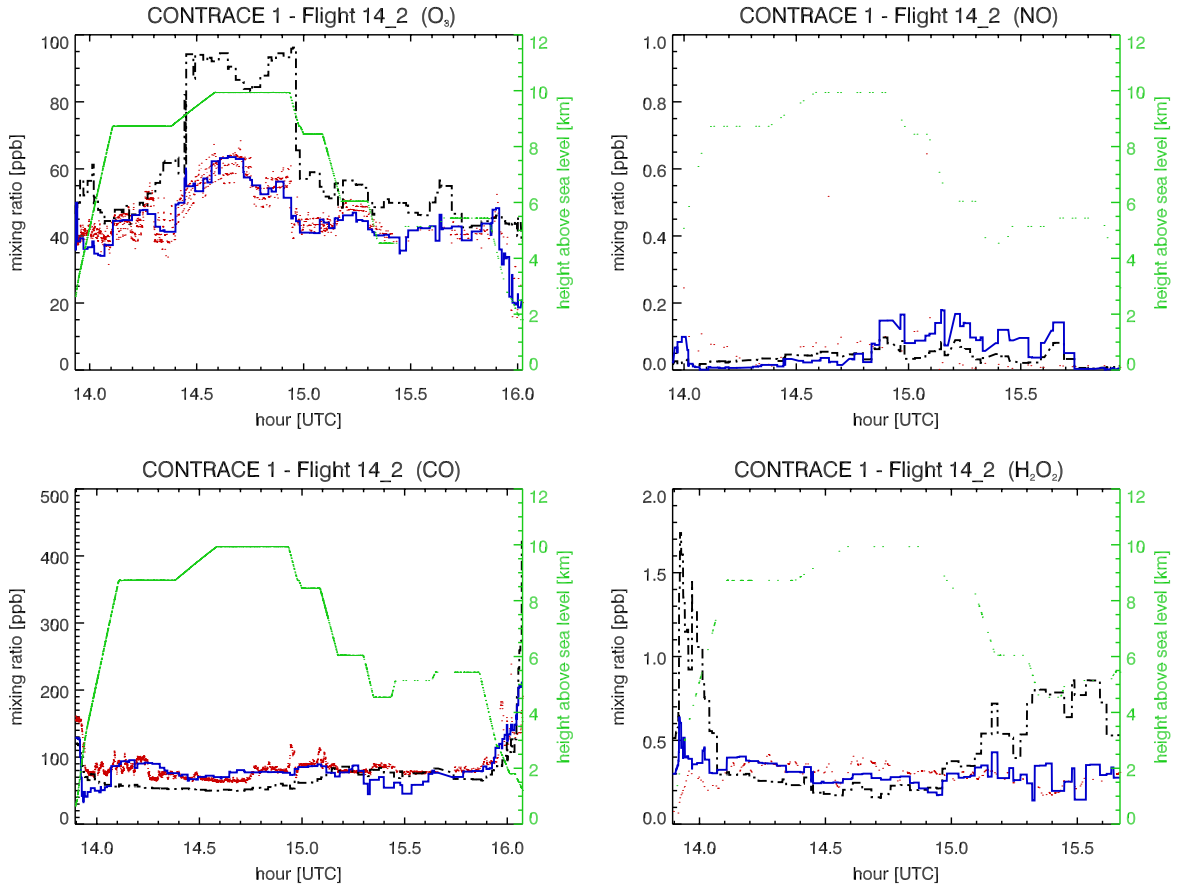

Figure 6: Time series for CONTRACE flight on Nov 14, 2001; measurements (red points), first guess (black dash-dotted line) and analysis (solid blue line). The green line, together with the right ordinate, indicates flight heights.

Nested application of 4D–var

As a matter of representativity of observations on coarse grids, for a reactive emitted species like NO_2 a success similar to ozone cannot be demonstrated. In order to also exploit measurements of those species, a nesting technique is implemented for adjoint modelling and applied to the 1998 urban plume campaign BERLIOZ around the metropolitan area of Berlin, Germany (Volz–Thomas

et al., 2003). Assimilated species are O_3, NO, NO_2, CO and SO_2 within an assimilation window of 14 hours, from 06 UTC to 20 UTC on July 20, 1998. The nesting procedure included a coarse grid simulation with horizontal grid size of 54 km and three recursively nested grids with a nesting ratio of three. Hence, there is a 2 km final resolution. The assimilation of NO_x surface observations proves difficult, as usually point and line emission sources imply only a small spacial scale of validity. Figure 4 demonstrates the assimilation performance for 4 measurement stations mostly within the greater Berlin area, as achieved by a 18 km resolution grid (nest 1) and a further nested 6 km resolution grid (nest 2) with joint emission rate and initial value optimisation. Clearly, a significantly improved performance of the forecast can be seen for nest 2, with observations beyond the assimilation interval. It can be concluded, that, under conditions given, a 6 km horizontal resolution allows for a satisfying exploitation of the suburban NO_x observation sites.

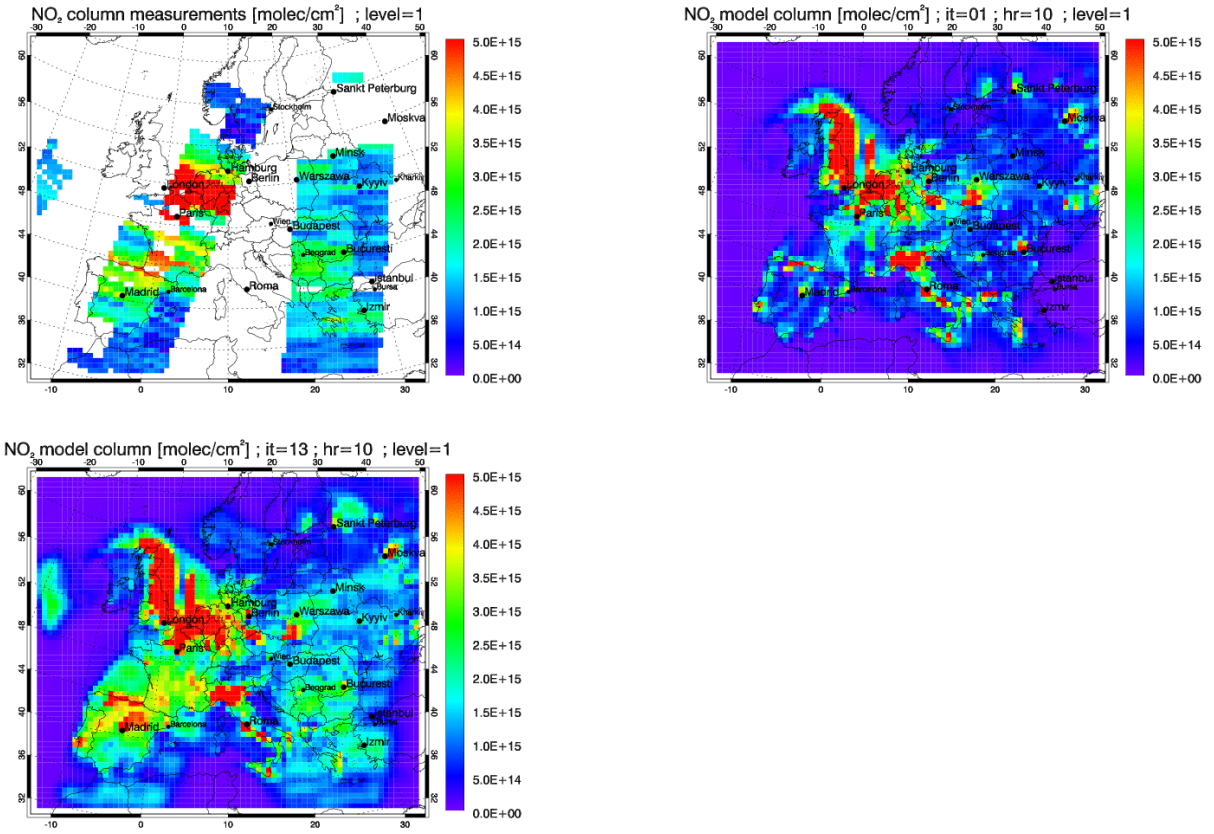

Figure 7: Assimilation of NO_2 tropospheric columns at 3.8.1997 (top panel). Simulation without assimilation for 10:30 UTC (top right panel), and post assimilation forecast for the same time and started at 06:00 UTC, Units are number of molecules/cm^2.

Emission rate estimates

As a unique feature, the adjoint calculus has the potential to optimise initial values as well as emission rates.

The impact of emission rate optimisation is demonstrated by Figure 5, which exhibits SO_2, CO, NO_2, and xylene optimisation factors over the integration domain of the finest (2km resolution) grid. The inversion process at each grid level hands over the result to the next finer grid, allowing for an increasingly better resolved emission estimate, provided the necessary observational density is given. As Berlin is mostly a large urban island within a more rural environment, sulfur emissions are confined

to the greater metropolitan area. The upper left panel of Figure 5 clearly indicates a nearly overall reduced emission rate over the densely populated area, probably indicating a moderately larger success of reduction efforts than estimated by the emission inventory. In the case of CO similar effects can only be claimed for the area east of Berlin.

While the effects vary moderately for NO_2, xylene, with an amplification factor of about 1.2, appears to be underestimated by the emission inventory. In all exhibited cases, the inversion results remain well within the error limits of the inventory. Emission rate optimisation of SO_2 and CO is based on concentration observations of these species. In the case of other emissions, which are rarely observed, inference can only rest on measured product constituents, most prominently ozone. This is the case for emission factor estimates of xylene, establishing an ill–posed problem. In this context it should be noted that short campaigns like BERLIOZ may be insufficient to build up reliable quantitative statistics for emission inversion, as underlying error covariance statistics need longer estimation times, with varying meteorological conditions.

Air borne data assimilation
The assimilation of air borne data can provide better evidence of the origin of air masses, especially the cleansing off polluted boundary layer air by cyclonic dynamics. The focus of the free troposphere CONTRACE flight campaign (Lawrence et al., 2003) is placed on updraft processes. The first CONTRACE episode with a special flight from Corsica to Munich, Germany, on Nov 14, 2001 has been selected for middle tropospheric assimilation, with warm conveyor belt features induced by cyclone dynamics. The vertical grid structure has been refined for the relevant height region, having now 26 layers. Horizontal grid size is 25 km. Assimilated observations of species aloft are O_3, NO, H_2O_2 and CO, while NO_2 and SO_2 at the surface only. Figure 6 shows forecast improvements for the data of the flight during the afternoon of Nov 14. Initial values have been optimised for midnight (00:00 UTC) to ensure a consistent chemical model state evolution over the day. Especially ozone and CO show very good performance results due to assimilation, while all assimilated species are improved. In the case of NO, occasional strong observation spikes indicate local lightning induced elevated concentration levels, which are neither simulated nor reproduced by the assimilation. The key difficulty here is to place strong convective clouds with lightning at the very correct position. The assimilation based analysis of the H_2O_2 radical exhibits comparably good results as well.

Tropospheric satellite data assimilation
Satellite retrievals from tropospheric height levels are an emerging issue in earth observation, although there is a limited number of species like SO_2, NO_2 and formaldehyde, which is presently amenable for retrieval. Moreover, in these cases data are presented in terms of tropospheric columns.

The conceptual flexibility of the variational technique must be invoked, where data, like tropospheric columns, are ingested, which do not have a direct correspondence to a model parameter. In the case of tropospheric columns, data are given in terms of molecules per cm^2. The model correspondence (operator **H** in the cost function) can then be calculated, along with its adjoint, and included in the algorithm. Observing some technical details of preconditioning (see Elbern and Schmidt, 2001), the optimisation procedure of the assimilation adapts the model column to the retrieval in consistency with the model. Figure 7 gives an example of the assimilation of NO2 tropospheric columns obtained from IFE, University Bremen. Spain and Tunisia are areas with visible discrepancies between retrievals and model, prior to assimilation. The final analysis then finds the differences largely removed.

6 Summary

The 4D–var data assimilation method proves useful for applications in air quality simulations. Looking for analogies with meteorological weather forecasting, problems resemble the challenges associated with low level humidity assimilation: strictly nonisotropic and inhomogeneous correlation lengths, especially in the boundary layer, frequent violation of the assumption of the tangent linear approximation, and significant violation of the perfect model assumption due to deficiencies in the knowledge of meteorological parameters. In addition, emission rates are at least as important as initial values, and henceforth to be included as optimisation parameter. For many regions, the deposition rates are also to be taken as optimisation parameter. With an enhanced set of optimisation parameters, the optimisation problem becomes more ill–posed and enhanced precision of estimates of the error covariance matrices is the only possibility to account for this problem. The most obvious way to account for this is operational run of the assimilation system, which allows for compilation of relevant statistics.

Acknowledgments
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