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Data assimilation of ozone in the atmospheric transport chemistry model LOTOS ☆

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Abstract

Modelled ozone concentrations often differ from measured concentrations quite substantially, partly due to measurement errors, but mainly due to uncertainties in the model. Modelling studies would therefore benefit highly from more reliable model simulations. One way to achieve this is the application of data assimilation, a technique that uses measurement information within the model simulation in a way that is consistent with the model itself. This aim of this paper is to show that this is indeed one way to go with atmospheric transport chemistry models (ATCMs) by presenting results of data assimilation simulations of ozone with the model LOTOS. The assimilation technique used in this study is the Ensemble Kalman Filter. A simulation for a period of 4 weeks has been performed in which ground-level ozone measurements have been assimilated. The necessary noise input consisted of uncertainties in the emissions of NO_x, SO_x, VOC and CO in 17 groups of countries. The main conclusion is that it is possible to improve ATCM simulations of ozone by data assimilation, but that noise inputs other than emissions only are essential for the reduction of the differences between measured and modelled concentrations to acceptable margins.

Keywords: Data assimilation; Kalman Filter; Ozone; Transport modelling

1. Introduction

Atmospheric Transport Chemistry Models (ATCMs) simulating ozone (O_3) concentrations are not perfect. There are always differences between modelled and measured (ozone) concentrations, even though the underlying physical processes are, in the case of tropospheric ozone formation, quite well known. The discrepancies between model results and measurements put a limitation on the outcome of modelling studies. Therefore, these studies would benefit highly from more reliable model simulations that could be used as a starting point for scenario calculations, etc.

One way to try to reduce the gap between measured and modelled quantities is the application of data assimilation, which has already proved to be useful in various fields of application (see, for example, Verlaan and Heemink, 1996). In recent years, application of data assimilation has received increasing attention among scientists in the field of atmospheric chemistry and transport modelling (see, for example, van Loon and Heemink, 1997; Segers et al., 1998a,b; Elbern et al., 1997; Elbern and Schmidt, 2000; Proceedings of the SODA Workshop on Chemical Data Assimilation, 1999).

Data assimilation is a technique that combines two sources of information: the model and the available measurements. It assumes that both the model and the measurements are subject to errors. These errors or uncertainties need to be specified in statistical terms (see Section 2) in order to be used by the data assimilation technique. Specification of these errors is often far from trivial and plays a key role in being able to successfully apply data assimilation.

In this paper the results of a case study with the ATCM LOTOS are presented. An implementation of the Ensemble Kalman Filter (EnKF) (Evensen, 1997) is used. The emissions of NO_x, SO_x, VOC and CO in 17 groups of countries are taken as uncertain. No other model errors are taken into account yet. The reason to (first) consider the emissions is that their yearly and hourly values are not known very precisely and have

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a rather large potential impact on the simulated ozone concentrations. Officially reported emission totals (following a protocol) may not agree with the actual emissions. More important, though, is that even if total yearly emissions are correct, their temporal distribution may be incorrect because models like LOTOS use idealised time profiles that derive hourly emission input from yearly totals. For these reasons assuming the model errors to be linked with the emission input seems to be a logical starting point. In a later stage, of course, other sources of model errors need to be taken into account as well. For addressing the key question of this paper, however, the assumed model errors only by emission are sufficient.

In Section 2 some principles of data assimilation are discussed, followed by a short description of LOTOS in Section 3. In Section 4 the set-up of the case study and its results are presented. The paper concludes with a section on conclusions and recommendations.

2. Data assimilation

In this study the Kalman Filter approach is used. More specifically, an Ensemble Kalman Filter (EnKF) is applied. Other filter techniques can be used as well. In Elbern et al. (1997) and Elbern and Schmidt (2000) a 4D-var method is applied to a chemical transport model, showing that this method can also be applied successfully to non-linear atmospheric models. Other examples can be found in, for example, the Proceedings of the SODA Workshop on Chemical Data Assimilation (1999). It is beyond the scope of this study to discuss the advantages and drawbacks of the different methods. A short description of the main aspects of the Kalman Filter and its limitations is given below.

2.1. Kalman filtering

Instead of simply adjusting the state of the model at certain points where measurement information is available, the data assimilation technique tries to process the measurements in a physically consistent way, adhering to the "laws of the model". That means that variations in the model state can only result in different model states that could have been achieved by evolution in time of the model using different, (physically possible) input parameters, etc. In mathematical terms, this is achieved by an extension of the deterministic model to a stochastic model. Instead of a deterministic model of the form

$$x^{k+1}=f(x^k), \tag{1}$$

in which x^k denotes the state vector x of the model at time level k and f the model operator, we have the stochastic extension

$$x^{k+1}=f(x^k,w^k), \tag{2}$$

where w^k is the noise input vector. Each element of this vector represents an uncertainty in the model, to be specified by the modeller. As with most filter techniques, the EnKF assumes the distribution of the entries of the noise vector to be Gaussian, but not necessarily independent. The state vector x is not necessarily restricted to the concentrations of the species in all grid cells, but may contain other variables as well. In the work reported here emission correction factors are also part of the state vector. In that way the filter explicitly estimates these variables.

As with the model, the measurement uncertainty needs to be specified. A measurement vector y^k, representing a set of measurements at time level k, is written as

$$y^k=C^k x^k+v^k, \tag{3}$$

where C^k represents the operator that computes the modelled value from the model state at the measurement location. The vector v^k is the measurement error, again by the filter assumed to be Gaussian. It this study it is assumed that the measurement errors are uncorrelated in time and space.

The Kalman Filter uses the error description of the model to create an approximation of the covariance matrix for the entries of the state vector. Based on this matrix and the observations and their uncertainties, the Kalman Filter computes a best fit of the model with the observations.

The specific implementation chosen in this study is the Ensemble Kalman Filter (EnKF) (Evensen, 1997). An EnKF uses a random generator to produce noise inputs w^k in order to form an ensemble of state vectors. The filter is able to calculate the conditional mean of the state given the statistics of the state (specified by the ensemble) and the statistics of the measurements (specified by Eq. (3)).

2.2. Limitations of a Kalman Filter

The main limitation of the technique is the fact that it assumes errors to be Gaussian. In particular, model errors will not be Gaussian, even if the underlying uncertainties are Gaussian. For example, a standard deviation of 10% on the NO_x emission is assumed. The Kalman Filter evaluates the impact on the state vector by performing a computation with the NO_x emission increased by 10%. The assumption of a resulting Gaussian model error in fact means that the Kalman Filter assumes the same response of the model (but with opposite sign) when the NO_x emissions are reduced by 10%.

3. The model LOTOS

3.1. Short description

The Long Term Ozone Simulation (LOTOS) model is a Eulerian grid model that uses a 70×70 equidistant grid, covering the domain [10W–60E]×[35N–70N]. This domain is large enough to be able to simulate ozone concentrations in Europe for both episodic and long-term calculations. In the vertical direction there are three layers: the mixing layer and two layers above it. Depending on the mixing height, the top of the model domain is between 2 and 3 km. The height of the mixing layer is spatially varying and also varying in time. Hence in vertical direction the grid is irregular and not fixed. However, within one time step the grid is fixed. For output purposes, a fourth, diagnostic surface layer is present: using the concentrations in the mixing layer, the concentrations at a given height near the ground are computed using "subgrid-scale" considerations. This principle is also useful for data assimilation because the measurements are taken close to the ground. The concentrations close to the ground (for O_3 and in general for species which are deposited) can be up to, say, 30 percent lower than the average concentration in the mixing layer, especially during the night. The model includes the processes: horizontal and vertical advection and diffusion, dry and wet deposition and chemistry. The chemical mechanism used in LOTOS is the CBM-IV mechanism (Evensen, 1997; Gery et al., 1989; Morris et al., 1990). The meteorological input consists of 3-hourly fields with the horizontal winds, water vapour concentration, temperature, layer depth for the three model layers and of fields with the surface wind and temperature, cloud fraction and rain intensity. Unfortunately, the rain intensity fields only consist of a flag, indicating precipitation in the 3-hour interval or not. Hence, wet deposition can be computed in a very crude way. Since for ozone formation wet deposition only plays a minor role, wet deposition is not taken into account at all. Apart from meteorological input the model uses the following input: initial and boundary conditions (for each month) from the TNO-Isaksen 2D model (Poppe et al., 1996; Isaksen and Rodhe, 1978); the surface roughness length, z_0, and the surface resistances for each species that is deposited, derived from a landcover database, for each grid cell; anthropogenic NO_x, VOC, CO, CH_4 and SO_x surface emissions per grid cell; NO_x, and SO_x point sources; biogenic VOC emissions derived from a landcover database; a speciation of the VOC emissions into the CMB-IV species; time and temperature dependencies per emission source category.

For more details on the model, the interested reader is referred to Builtjes (1992) and Roemer (1996).

3.2. Model limitations

As has been stated in the introduction, knowledge of the model errors is crucial for a successful data assimilation simulation. Two different types of errors can be distinguished:

- input data errors: emission input, meteorological fields, land use data and surface resistance, the boundary conditions;
- modelling errors/conceptual limitations. In LOTOS the first modelling layer is the mixing layer. Emissions are assumed to be fully mixed in this layer instantaneously. This puts a limitation on the modelled results especially in area with dense emissions. Also, near-ground concentrations need to be computed from the mixing layer concentration, using a vertical concentration profile. For ozone, for instance, it is assumed that it is always in photostationary equilibrium with NO and NO_2 within this profile. This assumption is used in the computation of the deposition and for the estimate of the concentration at measurement height. Numerical accuracy plays a role as well. For example, because of the coarse horizontal resolution of the model transport calculations tend to be diffusive.

3.2.1. Consequences for data assimilation

As can be seen from the list of "model errors" in Section 3.2, there are many more sources of uncertainties than emission input only. In some way all of these uncertainties contribute to the difference between modelled and measured concentrations. The way they contribute needs to be quantified in order to take them into account or to neglect them in the case of very little impact. In this study, this has not been done. As has been said, only the emissions are taken as uncertain, because they are considered to be responsible for a large part of the difference between observed and simulated concentrations. As a consequence, the assimilation may find a good fit between the model and the measurements for the wrong reason. It may, for example, use smaller NO_x emissions instead of increasing the deposition velocity of NO and NO_2, simply because the assimilation is not allowed to do the latter. Therefore, it is clear that at least the resulting emission factors from the assimilation computations are only indicative.

4. Modelling studies

4.1. Set-up of the simulation

For the data assimilation simulation specification of the measurement and the model uncertainties are

required. As mentioned earlier, the model uncertainty is specified in terms of uncertain emissions only. For this purpose, 17 groups of countries are taken, see Table 1. In each area the NO_x, SO_x, VOC and CO emissions are considered uncertain, resulting in 68 noise parameters.

The emission emis_{ij}^k in a certain area i of a species j is calculated for each simulated hour k according to the formula:

$$\text{emis}_{ij}^k = \text{emis\_db}_{ij}^k \cdot (1.25)^{w_{ij}^k} \quad (4)$$

where w_{ij}^k is the corresponding element of the noise input and emis_db_{ij}^k denotes the original emission values from the emission database for the area i, species j and hour k. A noise element w_{ij}^k has a Gaussian distribution with zero mean and unit variance; the emissions are therefore assumed to vary around the deterministic emission with equal probability of being some percentage higher or lower (on average 25%). The noise elements are assumed to be uncorrelated for different species and areas, but are forced to correlate in time according to (omitting subscripts i and j)

$$\langle w^k w^{k+1} \rangle = \exp(-0.1). \quad (5)$$

This reflects the idea that although emissions are uncertain, they will not fluctuate from, say, +25% to −25% within 2 hours. Moreover, introducing coloured noise instead of white noise generally increases the computed variance of the state vector, because the propagation of a single ensemble member by the model is forced by a more or less constant deviation in the emissions. This gives the model the opportunity to build up substantial differences between individual ensemble members.

For each ozone measurement it is assumed that the standard deviation of the error is equal to 10% of the measured concentration with a minimum of 0.5 ppb and a maximum of 3 ppb. The motivation to take a 10% error is not that we consider ozone measurements to be that inaccurate. From statistical analysis (Tilmes and Zimmermann, 1998) it is known that in an area of the size of a LOTOS grid cell, approximately 60×60 km, the ozone concentrations may easily vary by 10% or more. Because we assimilate the measured concentrations into cell averages, a larger standard deviation of the measurements is justified for reasons of representiveness of the measurements for the grid cell area. Apart from the representiveness there is an instrumental error of 5 to 10% (see Tilmes and Zimmermann, 1998), so the actual standard deviation may be even larger than 10% of the measured concentration. However, specifying an even higher standard deviation would lead to smaller adaptations by the filter, since the standard deviations of the measurements are compared to those of the computed concentrations. Since only noise parameters are defined for the emissions, the "simulated" standard deviations are smaller than they would be if more noise parameters connected to other processes were defined. In order to prevent an imbalance between the modelled standard deviations and those of the measurements, the latter are bounded as indicated above. Such an imbalance would otherwise lead to only small adaptations of the simulated concentrations.

Time series of ozone measurements are available at 42 locations. From these 42 time series, 21 have been selected that are used in the assimilation. The other series are used for "diagnostic" purposes: do the simulated concentrations also improve by assimilation at locations other than measurement locations? Also for some other species, measurements series are available, which are only used for diagnostic purposes. The number of time series per country and per species are listed in Table 2.

4.2. Results of the simulation

In Fig. 1 a scatter plot is given of the average residues at the measurement locations of the ozone concentrations computed from the simulations with and without data assimilation. The residue is defined as the average of the

Table 2
Number of measurement series per country and per species used in this study

Country	O_3	NO_2	NO	SO_2
Netherlands	6	0	0	0
Germany	18	18	20	14
Austria	6	2	1	4
United Kingdom	8	1	2	2
Denmark	4	0	1	0

Table 1
Emission areas considered in this study

No.	Countries	No.	Countries	No.	Countries
1[	UK, Ireland	7	Russia	13	Italy
2	Germany	8	Poland	14	Bulgaria, Romania
3	Belgium, Netherlands, Luxembourg	9	Czechoslovakia	15	Yugoslavia, Albania
4	France	10	Hungary	16	Greece, Turkey
5	Denmark	11	Switzerland, Austria	17	sea
6	Norway, Sweden, Finland	12	Spain, Portugal		

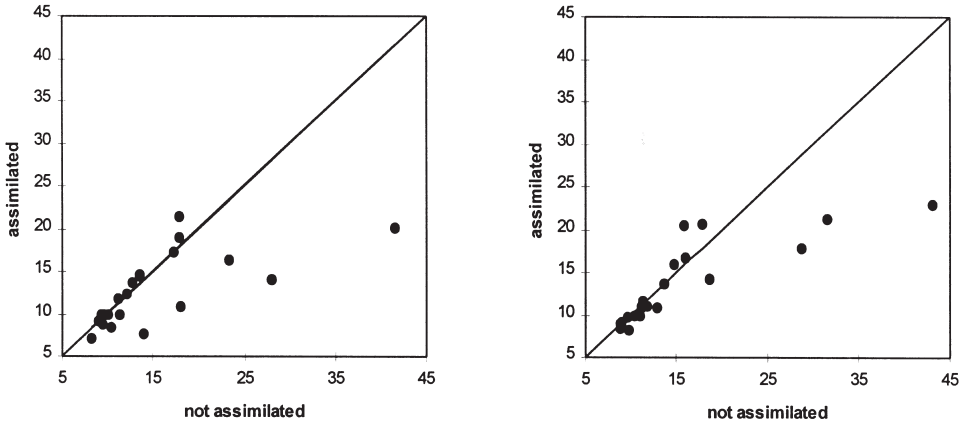

Fig. 1. Assimilated average residues versus not assimilated average residues (ppb) of ozone per station: *left*, assimilated stations; *right*, diagnosed stations.

absolute differences between the measured and modelled concentrations at all simulated hours for which measured values are available.

From Fig. 1 it can be concluded that when data assimilation is applied, in both the assimilated and the diagnosed stations the residues in general decrease with respect to the simulation without data assimilation, indicating a proper functioning of the data assimilation. It can also be seen from Fig. 1 that although the assimilation achieves a substantial decrease, the residues are still large. The mean concentrations and the average residues are given in Table 3, not only for ozone but also for a few other species.

It can be concluded that although the residues for ozone decrease by a significant amount, this is not the case for the other (only diagnosed) species. There is a slight improvement for NO, but the average residues for NO_2 and SO_2 are somewhat larger. The changes in NO_x (the sum of NO and NO_2), however, are very small in absolute value and also compared to the measured values. This may be surprising, since NO_x has a large impact on the ozone production. It should, however, be noted that the NO_x concentration is much more locally influenced than ozone and, therefore, not well reproduced by LOTOS due to the coarse horizontal and vertical resolution. In Table 3 two different results for SO_2 are presented. The main difference is that three German stations have not been taken into account in the "corrected" results, because the modelled concentrations at these locations were relatively high, probably because the spatial distribution of the SO_x emissions has not been corrected yet for installations that have been closed in Eastern Europe. Also the Austrian measurements (four stations) seem to be in ppm, so in the "corrected" results they have been multiplied by 1000. Note that omitting the German stations and correcting the Austrian measurements is only a cosmetic change, since the SO_2 measurements have not been assimilated in the model simulation. Examination of the residues and average concentrations of SO_2 reveals that the model generally overestimates the SO_2 concentrations. This should be expected, because wet deposition is neglected in the model, whereas it is an important sink of SO_2. It is also seen from Table 3 the simulated SO_2 concentrations are somewhat higher in the simulation with assimilation. One obvious reason is that the assimilation changes (increases) the SO_2 emissions in the system (see also below). It may want to do so because SO_2 influences the OH concentration which in turn plays a key role in the ozone production. Due to variation in other emissions, the OH concentration will already be changed, leading to a different oxidation rate of SO_2. These two effects

Table 3
Average concentrations and residues of some species

Component	No. stations	Average concentration			Average residues (ppb)	
		Measured	Not assimilated	Assimilated	Not assimilated	Assimilated
O_3 (assimilated stations)	21	35.4	36.4	30.7	15.2	12.7
O_3 (diagnosed stations)	21	33.6	36.6	31.2	15.6	13.5
NO	24	1.49	0.30	0.39	1.84	1.69
NO_2	20	5.39	1.97	1.92	4.21	4.24
SO_2	19	1.52	5.21	5.38	4.13	4.37
SO_2 ("corrected")	16	2.06	3.27	3.44	2.94	2.97

may counteract each other, so that the percentage increase in SO_x emission does not need to be equal to the percentage rise in the SO_2 concentration. These considerations on the residues and concentrations of SO_2 are very instructive, because they clearly show that the assimilation may do things for the wrong reason. Although the modelled SO_2 concentrations are already too high, they are further increased by the assimilation. This would probably not have occurred had SO_2 measurements been assimilated as well. In the present simulation the room the EnKF has to vary the SO_x emissions is purely used to improve the ozone concentrations, regardless of the effect of the modifications on other concentrations. If SO_2 measurements were assimilated as well, the decrease in residues of ozone would probably have been smaller. This once more proves that other noise processes that are not taken into account yet are responsible for the gap between measured and modelled ozone concentrations.

Instead of examining residues it is worthwhile to look at time series at individual locations as well. Fig. 2 shows a concentrations plot at location Eibergen (NL).

From Fig. 2 it becomes clear that patterns in the measured time series are generally better followed by the simulation with assimilation. For example, the simulation without assimilation tends to overestimate the ozone concentrations at 8–9 and 13–14 August. This is clearly corrected in the simulation with assimilation. In both cases the measured values during the night-time are not well represented. This may be caused by a combination of two effects: (1) the night-time chemistry may be not very accurately described by the CBM-IV mechanism and (2) the mixing layer concept of the model puts a limit on the NO_x concentrations at night, thus preventing part of the O_3 titration.

The values of the noise parameters that act on the emissions give insight into the emissions that are used by the assimilation simulation. In Fig. 3 the average values of the resulting emission correction factors per source area (numbering according to Table 1) are given. For presentation purposes two values higher than 1.5 have been cut off. In areas 4 and 17, the correction factors for NO_x are 2.5 and 1.8, respectively. In general, the values of the average emission correction factors are close to one. Also the factors for areas relatively far from the measurement locations are different from one, reflecting that ozone concentrations at the measurement locations are influenced by precursors at a large distance.

As has been explained in Section 3.2.1 the emission correction factor can at most be considered indicative, since no other noise parameters have been used, representing uncertainty in other processes in the model. The fact that the values for the emission correction factors do not deviate much from one on average, and that residues are still relative large, indicates that the filter was able to blame the emissions for the large residues only to a limited extent. In other words, the results reported here confirm that other processes in the model are responsible for the residues as well.

5. Conclusions and recommendations

From the results reported in this paper the following conclusions are drawn:

- successful application of the (Ensemble) Kalman Filter in combination with an ATCM is possible;
- the uncertainty in the emission input is evidently one of the explaining factors, but certainly not the only one;
- using coloured noise instead of white noise seems to work well;
- the emission correction factors are indicative, not only because no other (noise) processes are taken into account yet but also because of conceptual limitations of the model itself and the fact that the filter assumes the impact of a change in the emission on the ozone concentration to be a linear.

The following recommendations are made:

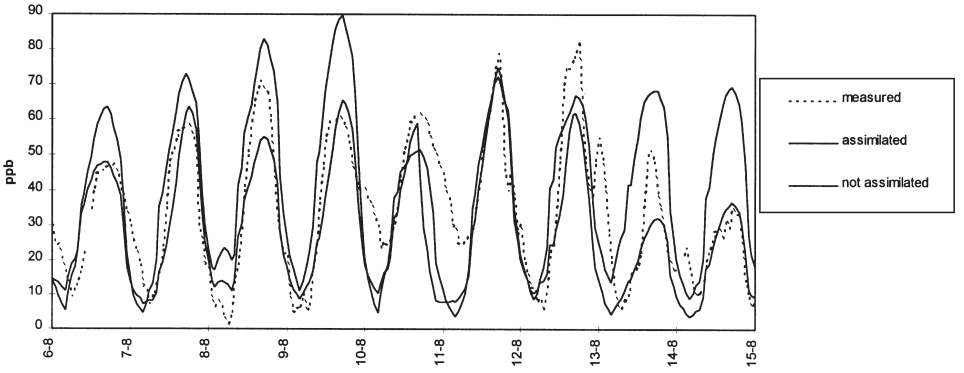

Fig. 2. Simulated and measured ozone concentrations at Eibergen (NL) in part of August 1997.

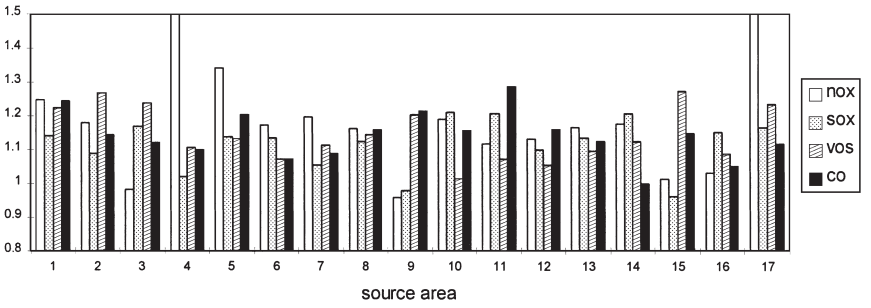

Fig. 3. Average emission correction factors per source area.

- other noise processes need to be taken into account; possible processes are deposition, vertical diffusion, mixing layer formulation, meteorological input;
- measurements of other species should be taken into account, in addition to model restrictions;
- measurements of emitted species at remote locations can be useful to get a better estimate for the emissions, if used in combination with other noise processes;
- modifications in the filter need to be implemented in order to take non-linear effects into account.

A paper on results of data assimilation simulations using additional noise input is planned for the near future.

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