Articles, Letters and other Contributions
for the EURASAP Newsletter since 2008
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HARMO 11
11th International Conference on Harmonisation within Atmospheric Dispersion Modelling for Regulatory Purposes
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Queens' College, University of Cambridge
www.harmo.org

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ITM29
29th NATO/SPS International Technical Meeting on Air Pollution Modelling and its Application
24 – 28 September 2007
CESAM, Department of Environment and Planning, University of Aveiro, Aveiro, Portugal
www.dao.ua.pt/itm/29th

European association for the science of air pollution

CONTENTS

EDITORIAL

Dear EURASAP members,

In 2007 EURASAP gave Travel grants to Agnese Osite (Latvia), Hans Orru (Estonia) and Vladimir Fuka (Czech Republic) to participate in the UAQ2007 in Cyprus and to Maria Prodanova (Bulgaria) for the 11th Harmonisation conference in Cambridge, UK. EURASAP Awards for best paper and presentation (oral or poster) by young scientists were given at the 29th ITM in Aveiro, Portugal to Sergey Napelenok (EPA, USA), Andy Delcloo (Belgium) and Marina Astitha (Greece). Sergey Napelenok and Andy Delcloo submitted articles on their work to the EURASAP Newsletter which you may read in issues 64 and 65.

The EURASAP Committee had a meeting on September 26, 2007 in Aveiro, Portugal and came with a number of decisions:

Eurasap will fund in 2008/2009

- Harbour Air Quality and Climate conference, Rotterdam, May 2008.
- The 12th Harmonisation Conference in Cavtat, Croatia, October 6-9, 2008.
- The 30th ITM conference, San Fransisco, USA May 2009.

Jan Kretzschmar has retired from VITO and on this occasion retired from the EURASAP Committee as well. Jan Kretzschmar suggested Clemens Mensink from VITO to join the Committee (VITO is one of the regular EURASAP corporate members), which was accepted by the Committee. The President of EURASAP Peter Builtjes invited Clemens Mensink to join the EURASAP Committee and he accepted. Clemens was also kind to submit with Guido Cossemans an article on VITO for the EURASAP Newsletter with a special view on Jan Kretzschmar's activities (Newsletter issue 64).

The EURASAP Newsletter Issue 64 (ISSN 1026-2172) is the last printed newsletter. After 22 years of newsletter mailing, EURASAP will follow the modern way of publishing: starting from Newsletter 65 we will have online

publication only (as described in the letter by the President and the Chairman of EURASAP, issue 64). The online newsletter is registered as online publication - ISSN 2070-2582, Key title: Newsletter (European Association for the Science of Air Pollution. Online), Abbreviated key title: Newsletter (Eur. Assoc. Sci. Air Pollut., Online), *Parallel title: EURASAP newsletter*. The parallel title will continue the numbering of issues under the new ISSN. As requested by EURASAP members the online newsletter will be in a format easy to download and print. There was an enquiry in 2006 through the newsletter on this topic. The new form is in line with new communication facilities and will save more funding for travel support of young scientists for different conferences and EURASAP workshops organisation.

Finally, the duties of Dr Ekaterina Batchvarova (NIMH, Sofia, Bulgaria) as newsletter editor ended in 2007. Dr. Zvjezdana Klaic (Zagreb University, Zagreb, Croatia) is the new EURASAP newsletter editor starting in 2008.

Therefore Issues 64 and 65 are edited by both of us

Ekaterina Batchvarova and Zvjezdana Klaic

We kindly invite you to continue to use the EURASAP Newsletter for exchange of information and discussions and point out that it continues to be internationally recognized publication.

The online registration of the EURASAP Newsletter is:
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Letter

Aveiro, 11th January 2008

Dear EURASAP members,

This letter is to inform you about forthcoming changes concerning EURASAP membership fees, and the EURASAP newsletter.

As you know, the membership fee for individual members in Europe is € 40. No fee is required in case your personal or social circumstances prevent you from paying the normal fee, especially recognising the countries in economic transition.

However, those who have the opportunity to pay as Corporate members are very welcome and will help to stabilize the EURASAP financial situation (the small fee of 250 €, for most of the institutions, will allow that you and your colleagues do not need to pay as individual members).

From 1 January 2008 we will not send the newsletter anymore by post, but we will distribute the newsletter by e-mail. We will, as normal, distribute at least 4 newsletters per year. The newsletter will be send directly by e-mail to all paying members, and to the members who informed us that they are in special circumstances as explained above. With a delay of 6 months, the newsletters will then be put on our open web-page.

We do hope that you will all become paying members, as far as possible. This will enable us to maintain giving travel funds especially to younger scientists to attend workshops co-organised by EURASAP, and to make hopefully interesting newsletters with information about air pollution projects and events throughout Europe. And please contribute to the newsletter with new articles!

So, please fill in the membership form as soon as possible, and send your subscription fee.

Best regards

Carlos Borrego and Peter Builtjes

Scientists' Contributions

JAN KRETZSCHMAR: A TALE ON AIR QUALITY MODELING

Guido Cosemans and Clemens Mensink
VITO, Belgium

Last summer, Jan Kretzschmar, one of the founding members of EURASAP, has retired from VITO and from EURASAP Committee. He can look back on 35 years of experience in air quality monitoring and modelling. In this contribution, we would like to recall some of the pioneering work which was carried out by him.

The beginning

Jan Kretzschmar obtained a masters in civil engineering (electricity and on nuclear sciences) and a PhD on microwaves before he was hired in 1972 by the Belgian Nuclear Energy Research Centre SCK/CEN. His task was to make the Centre's nuclear know-how and facilities suitable for the study of ambient air pollution in urban and industrial regions in Belgium. Among this know-how and facilities were the Bultynck-Malet stability classification scheme and dispersion parameters, a 124 m high meteorological tower with continuous registration of 12 wind- and temperature gradient parameters year-round, the BR1 nuclear research reactor with unique facilities for Neutron Activation Analysis and an IBM mainframe computer – able to do 60 K integer instructions per second.

Jan had, apparently, everything in operation in no time, in order to compute the impact of the SO_2 emissions from two new 153 m high stacks at an oil refinery in Antwerp, using the first version of the Immission Frequency Distribution Model IFDM. In the mean time he participated in setting up the Belgian network for monitoring heavy metals in ambient air, measuring daily concentrations of Pb, Zn, Cd, Ba, V, Ni, Mn, Cu, Cr and Fe in 15 monitoring sites, and in organising mini-max and random SO_2 monitoring campaigns in most industrialized areas in Belgium. The publications given in references [1-4] illustrate some of his activities in these early days

Modelling starts with verification

There are no publications on emission inventories, but when our group recently moved to an other building, we found lots of mail between Jan's early staff and industrial plant managers containing, among others, letters on the fuel types used in these plants and their sulphur content. This demonstrates the large effort to complete the data sets needed for air quality modelling. (It might be noted here that at that time the city of Antwerp had a very good industrial emission inventory, and that the epidemiology group of the Ministry of Public Health in Brussels (IHE) operated flawlessly a network of monitoring sites, yielding daily SO_2 and black smoke concentrations at several hundred locations in urban, rural and industrial sites in Belgium since 1968.)

The Belgian National R&D Program Environment-Air (1975-1981) laid the firm basis for some of the air quality management tools still in use in Belgium today. The construction of emission inventories was centralized at the University of Ghent (prof. R. Dams). Jan could expand his team with some newly graduated engineers, one of them being Guido Cosemans.

Jan has always been a firm advocate to summarise air pollution data not only by the annual average, but also by the Cumulative Frequency Distribution (CFD) from median over the 95 and 98-th percentiles up to the maximum value of the data set. So an air quality model, such as his own IFDM, had to be able to reproduce the CFD of the actual air pollution levels at a given site, as provided by the local air quality network. This 'reproduction' guaranteed that the emission inventory used was relevant for the region and free of gross errors, and that model output was indeed relevant for the pollution situation studied, so that computed changes of air pollution due to changes in the emissions could also be trusted to be realistic.

In the National R&D Program, Jan and his co-workers showed that the IFDM methodology, using the hundreds of industrial and urban (SO_2) sources typical for large industrial and urban regions, in combination with a times series of hourly meteorological data over one year, could reproduce the annual averages and CFD's of the measured daily (Antwerp, 1972-1975) and half-hourly (Gent, 1978) SO_2-concentrations. Once this was demonstrated, IFDM was used to compute air pollution over the entire industrial region with a spatial resolution of 1 km by 1 km or finer, to evaluate the cost-effectiveness of alternative emission granting and emission abatement strategies.

In this whole process, model verification was and is not an isolated activity that has to be carried out once during the development of a model, but model verification turned out to be rather an essential part of the quality control on an integrated set of activities, data and tools (emission inventory, measured concentration, meteorological data and air quality model). Some of this experience is given in references [5-10].

Sub-model verification

It is interesting to see how Jan has tackled the intrinsic quality of the different components in his IFDM model. His first point of interest was the plume rise formula to be used. In the early days of air quality modelling, dozens of plume rise formula were around, predicting plume rises for a comparable situation that differed by a factor 5 to 20. Using the first pocket calculator with *scientific* functions that could replace a slide rule, the HP-35, Jan must have evaluated all these formulas. Combining his findings with the validated IFDM results on the Antwerp industrial stacks, he selected the Stumcke II formula and later on in 1992, the Briggs 1975 formula to be used in IFDM.

The influence of the turbulence typing scheme on the calculated concentrations was examined thoroughly by Jan and his team between 1978 and 1984 (see also references [11-14]). One reason to do this is the fact that Belgium is surrounded by countries with nuclear energy facilities located close to the Belgian border. Governments, being concerned about the dangers of population exposure in case of accidental releases from such facilities, were demanding a reliable assessment of the impact. In order to test this, Jan conducted a series of ground-level released SF6 dispersion experiments over a (Belgian) rural terrain. It turned out that for this set of experiments, the use of the Bultynck-Malet scheme, originally derived for non-buoyant emissions through a 69 m high stack over the park-like landscape of the SCK/CEN terrains, performed -for near-ground emissions over west-European flat rural terrain, equally good as Pasquill for the average concentration over all measurements (both having a

bias of 20%) – but the standard deviation between the individual observations and predictions by Bultynck-Malet were much smaller than those given by Pasquill. The results of the SF6 dispersion experiments have been reported in the EUR 9385 EN [14].

New insights by further verification

During the R&D-program, the pollutant of main concern was SO_2, being emitted in large quantities by hundreds of industrial stacks, and in minor quantities by every household: there was almost no natural gas in Belgium then, so space heating was by either oil or coal. Moreover, air masses coming from industrialized regions in Germany and Poland were already loaded with SO_2. This "background" concentration was poorly understood at that time, since regional or meso-scale models were hardly existing. Consequently, some people doubted if the IFDM model prediction of the impact of a single stack emission could be validated correctly. Then someone suggested to concentrate the know-how of all the teams in the Belgian R&D-program on Environment-Air on a small antimony smelter in the north of Belgium. The smelter had three tall stacks. The nearest antimony smelter was more than a thousand kilometres away, so any trace of antimony found in the air or on the vegetation, had to come from that smelter. This became a very interesting two year exercise, with an on-site meteorological tower, a perfectly designed monitoring network with 10 sampling sites for antimony in ambient air, some 50 deposition gauges, in stack monitoring of emissions, and every instrumentation that that could be deployed by some 30 research groups. Interestingly enough, the IFDM model using the on-site meteorological data and the in stack emissions measurements, predicted ambient air concentrations of antimony that were, close to the stacks, ten to hundred times lower than measured. If IFDM was correct, then most of the antimony found in the network originated from fugitive emissions from the smelter building – mainly condensation aerosol with an aero-dynamical diameter less that 4 μm. This finding was confirmed at the end of the study, when the size distribution and morphology of the dust particles, sampled at different distances from the stacks, was revealed. Fly ash and some stack-specific antimony crystals were found only in air samples at the remote sites. (Note: ore handling was not a source of emission because the plant used moist ores packed in plastic bags). This study was then further oriented towards the determination of these fugitive emissions, using SF6-releases on the work floor in combination with reverse modelling. Besides the results on fugitive emissions, an important finding was also the value of the dry deposition velocity for this kind of condensation aerosol (see references [15-19].

The techniques and know-how developed during this case study, especially the determination of fugitive sources by means of reverse modelling, are still successfully used in Belgium to help non-ferro plants to meet the ambient air quality standards for heavy metals.

During the last two years that the Belgian network for heavy metals in ambient air was operated by SCK/CEN, the network was reconfigured to observe the plume of the antimony factory leaving Belgium at places as far away from the plant as possible. These antimony measurements were used to demonstrate that a puff/segmented plume version of IFDM was able to model transport and dispersion of antimony over distances of several hundreds of kilometres.

Model verification in a European context: the Harmonization Initiative.

From 1984 on, Jan occupied positions where his management skills, rather than his scientific skills, were challenged. When in 1988, the Belgian federal government decided to move all non-nuclear activities from the SCK/CEN to the regional Flemish authorities, Jan is one of the architects to streamline the new institute VITO (Flemish Institute for Technological Research), starting its activities in 1991. He creates a new division, where the focus is on environment, energy and sustainable development, trying to give authorities and industries a solid advice and practical solutions based on sound science and state of the art technology. Air pollution modelling is one of the topics for which he provides new opportunities, e.g. in developing expertise on regional and urban scale modelling. But at the same time Jan continues his crusade for good quality local-scale models. This time also at a European level.

As national models still give very different results when applied to the same source, using similar meteorological data for sites located near the border between two countries, Jan is eager to participate in the Initiative on Harmonization of Atmospheric Dispersion Models, to chair the COST 710

action on pre-processing of meteorological data for dispersion modelling and to host the third (1994) and fourth (1996) workshop of the Initiative on Harmonization (see references [20-26]). Jan did this in such way that the workshops, initially intended to be a series of three meetings of short duration to demonstrate the superiority of the Next Generation models over the Pasquill generation of models, grew out into a series of much appreciated conferences, where, as we all hope, experiences and know-how are exchanged leading to models whose output agrees as good as possible with reality for the correct reasons.

Jan retired from VITO, but does this mean that he is resting now? Not at all! He started his own consulting company* and further enjoys his hobby, namely… working!

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A meeting of "Expert group C on the Atmospheric Dispersion of Fission products Following a Reactor accident", which was a subgroup of the "Indirect action Research programme on the Safety of Thermal Water reactors" (EU research programme) was held in Grenoble 7 to 9 march 1983.

One can see Jan Kretzschman on the first row second from right to left.

Helen ApSimon, Sven-Erik Gryning and Peter Builtjes were participating in this project as well. Later, in 1986 this core group lead by Helen ApSimon created EURASAP.

The fourth workshop on "Harmonisation within Atmospheric Dispersion Modelling for Regulatory Purposes" was held May 6-9, 1996 in Oostende, Belgium.

The principal organizer of workshop was VITO, Belgium. Jan Kretzschmar was the host and organized with his group the workshop so well, that it became a series of International Conferences.

Scientists' Contributions

DEVELOPING A METHOD FOR RESOLVING NO_x EMISSION INVENTORY BIASES USING DISCRETE KALMAN FILTER INVERSION, DIRECT SENSITIVITIES, AND SATELLITE-BASED NO_2 COLUMNS

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Abstract
An inverse method was developed to integrate satellite observations of atmospheric pollutant column concentrations with specie concentrations and direct sensitivities predicted by a regional air quality model in order to discern biases in the emissions of the pollutant precursors. Using this method, the emission fields were analyzed using a "top-down" approach with an inversion performed by Discrete Kalman Filter (DKF) and direct sensitivities calculated using the Decoupled Direct Method in 3D (DDM-3D) embedded in the Community Multiscale Air Quality (CMAQ) model. The system was tested through an experiment focusing on NO_2 concentrations and emissions of NO_x in the southeastern United States. The method reproduced the expected NO_x emission fields from initially perturbed starting values. Responses to different parameters in the system, including assumptions for uncertainties in the emission fields and satellite observations, were also tested. The method is readily extendable to other pollutants.

1. INTRODUCTION

Current regional air quality models rely on well-developed emission inventories with high spatial and temporal resolution. While much work has been done in the development of such inventories, uncertainties still exist. At the same time, retrieval techniques for satellite data have improved and several datasets are available for observations of NO_2, CO, and some hydrocarbons recorded by several satellites in orbit.

A method was developed for using satellite NO_2 column observations to check for biases in current emission inventories of NO_x with Discrete Kalman filter (DKF) inversion and sensitivities calculated by the Decoupled Direct Method in three dimensions (DDM-3D), which had been previously integrated (Cohan et al., 2005; Napelenok et al., 2006) into the Community Multiscale Air Quality (CMAQ) model (Byun and Schere, 2006). The method was tested using a pseudodata scenario representing hypothetical satellite observations. A base-case CMAQ simulation acted as the true representation of the relationship between NO_x emissions and NO_2 column concentrations in the domain. Ground-level NO_x emissions were then adjusted in pre-defined geographic regions within the modeling domain to mimic possible biases. Finally, the inverse procedure was applied to attempt to arrive back at the base-case emissions taking into account uncertainties in transport and chemistry. The method has proved to converge robustly at the correct solution in only a few iterations for various spatially distributed emission biases.

Integration of satellite observations of NO_2 with regional air quality modeling efforts can potentially reduce uncertainty in emission inventories. Retrieval algorithms for NO_2 column densities have been developed for several satellites, including GOME (Richter and Burrows, 2002), SCIAMACHY (Sioris et al., 2004), and more recently, OMI (Bucsela et al., 2006). Inverse modeling of NO_x emissions has been applied previously, but typically on a global scale (Martin et al., 2003; Muller and Stavrakou, 2005) with some efforts on a continental scale (Quelo et al., 2005; Konovalov et al., 2006). In finer scale NO_2 inverse modeling, the difficulties arise from the importance of resolving the nonlinearities in chemistry and transport, which are overcome in this exercise with the aid of direct sensitivities.

The Discrete Kalman Filter inversion driven by direct sensitivities developed here is a robust and computationally efficient procedure for improving the capabilities of regional air quality models through inclusion of the increasing wealth of satellite observations. This method can also be readily extended to other species with sufficient satellite, ground-based, or elevated observations.

2. METHOD

2.1 Discrete Kalman Filter

Inverse modeling of the NO_x emissions field was performed using Discrete Kalman Filter. DKF is an optimization technique used to estimate discrete time series and states that are governed by sets of linear differential equations. It has seen frequent use in inverse modeling of emissions on both the global scale and regional scales for various gaseous and particulate species (Hartley and Prinn, 1993; Chang et al., 1996; Haas-Laursen et al., 1996; Gilliland et al., 2003). Since chemical transport models also parameterize nonlinear processes, the linearity assumption is overcome by applying DKF iteratively. This method is also attractive for inverse modeling, because it allows for the use of uncertainty information in both the emissions fields and the observed pollutant values. A brief overview of DKF is presented here, while more detailed explanation is available elsewhere (Gilliland and Abbitt, 2001).

DKF evolves the emission vector, $\overline{E}_t$, according to the following:

$$\overline{E}_{t,k+1} = \overline{E}_{t,k} + \mathbf{G}_{t,k}\left(\overline{\chi}_t^{obs} - \overline{\chi}_t^{\mathrm{mod}}\right) \quad (1)$$

At iteration *k+1* and time *t*, the emissions vector is altered based on the gain matrix, $\mathbf{G}_{t,k}$, and the difference between the vectors of observations, $\overline{\chi}_t^{obs}$, and modeled values, $\overline{\chi}_t^{\mathrm{mod}}$. The gain matrix is defined in terms of the matrix of partial derivatives of the change in concentration with respect to emissions, $\mathbf{P}_t$, the matrix of the covariance of the error in the emissions field, $\mathbf{C}_{t,k}$, and the noise matrix, $\mathbf{N}_t$, such that:

$$\mathbf{G}_{t,k} = \mathbf{C}_{t,k}\mathbf{P}_t^T\left(\mathbf{P}_t\mathbf{C}_{t,k}\mathbf{P}_t^T + \mathbf{N}_t\right)^{-1} \quad (2)$$

The covariance of error matrix also evolves with subsequent iterations according to:

$$\mathbf{C}_{t,k+1} = \mathbf{C}_{t,k} - \mathbf{G}_{t,k}\mathbf{P}_t\mathbf{C}_{t,k} \quad (3)$$

The covariance functionally determines the degree to which the emissions vector is allowed to deviate from its initial values. As iterations are progressed, the covariance is reduced according to Eq. (3) and subsequent differences between $\overline{E}_{t,k+1}$ and $\overline{E}_{t,k}$ are smaller in a mathematically stable system. In this application, the initial covariance of the error in the integrated emissions estimates, $\mathbf{C}_{t,k=0}$, was based on an estimate of the normalized uncertainty in the emissions, U_E, according to the following:

$$C_{jj} = \left(U_{E,j}\cdot E_j\right)^2 \quad (4a)$$

$$C_{jk,j\neq k} = \left(0.1\cdot\frac{U_{E,j}+U_{E,k}}{2}\cdot\frac{E_j+E_k}{2}\right)^2 \quad (4b)$$

Similarly, the noise matrix was based on the estimated normalized uncertainties in the observations, U_{obs}, according to:

$$N_{jj} = Max\left[0.5, \left(U_{obs,j}\cdot\chi_{j,t}^{obs}\right)^2\right] \quad (5a)$$

$$N_{jk,j\neq k} = 0.0 \quad (5b)$$

Theoretically, the noise matrix, $\mathbf{N}_t$, can account for both errors in observations, as it does here, and also errors in the modeling system. The minimum value of 0.5 (10^{15} molecules/cm^2)^2 was imposed to prevent mathematical instability.

2.2 Decoupled Direct Method in 3D

The relationship between precursor emissions and resulting pollutant concentrations was represented using sensitivities calculated using the Decoupled Direct Method in 3D. DDM-3D is an efficient and convenient way to calculate responses in the outputs of an air quality model to perturbations in various combinations of input parameters (Dunker, 1981; Yang et al., 1997). DDM-3D propagates sensitivities using some of the same algorithms that are in place to solve the atmospheric diffusion equation:

$$\frac{\partial C_i}{\partial t} = -\nabla\left(uC_i\right) + \nabla\left(K\nabla C_i\right) + R_i + E_i \quad , \qquad (6)$$

where C_i is the concentration of species i, u is the fluid velocity, K is the diffusivity tensor, R is rate of chemical generation, and E is the emissions field. An analogous equation is developed to calculate sensitivities:

$$\frac{\partial S_{ij}}{\partial t} = -\nabla\left(uS_{ij}\right) + \nabla\left(K\nabla S_{ij}\right) + J_i S_{ij} + E_i^{'}, \qquad (7)$$

where J_i is the ith row vector in the Jacobian matrix J, which represents the chemical interactions between species ($J_{ij} = \partial R_i / \partial C_j$), and S_i is defined as the change of a pollutant i in space, $\bar{x}$, and time, t, in respect to a perturbation in some model parameter, (emission rate, initial condition, etc.):

$$S_{ij}\left(\bar{x},t\right) = \frac{\partial C_i\left(\bar{x},t\right)}{\partial p_j}. \qquad (8)$$

Calculating both pollutant concentration and sensitivity fields using one model is of great convenience for inverse modeling applications where longer averaging periods are sometimes necessary to assure adequate geographical coverage of the observation data. Implementation of DDM-3D for the CMAQ model has been evaluated previously for both gaseous and particulate species and has been shown be suitable for producing NO_2 sensitivities to NO_x emissions as compared to discrete difference sensitivity methods (Cohan et al., 2005; Napelenok et al., 2006).

3. PSEUDODATA ANALYSIS

To evaluate the inverse modeling system, a pseudodata scenario was developed in a sample domain centered on the southeastern United States. Emissions source regions were defined based on similar spatial emissions patterns of ground-level NO_x during the summer months of 2004 and included the urban areas of Memphis, TN; Nashville, TN; Birmingham, AL; Atlanta, GA; and Macon, GA, as well as the rural areas approximately covered by the states of Tennessee, Mississippi, Alabama, and Georgia (Figure 1). A 144 km wide margin was left around the source regions in order to completely resolve sensitivity fields originating from the defined regions.

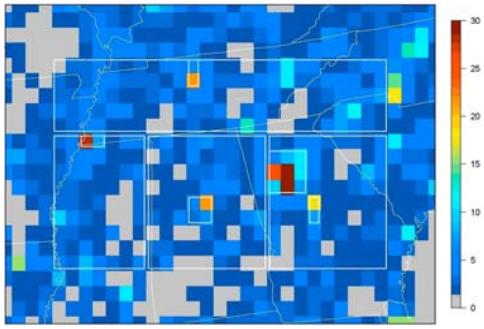

Figure 1. Source region definitions and average ground-level hourly NO_x emissions (moles/sec) on August 1, 2004

CMAQ with DDM-3D simulated base concentration fields of NO_2 and sensitivities to NO_x emissions from each source region. Sensitivities to the emissions from the surrounding "border" region and to the boundary conditions were also calculated and were found to have negligible impact on NO_2 column densities in the inner region (Figure 2).

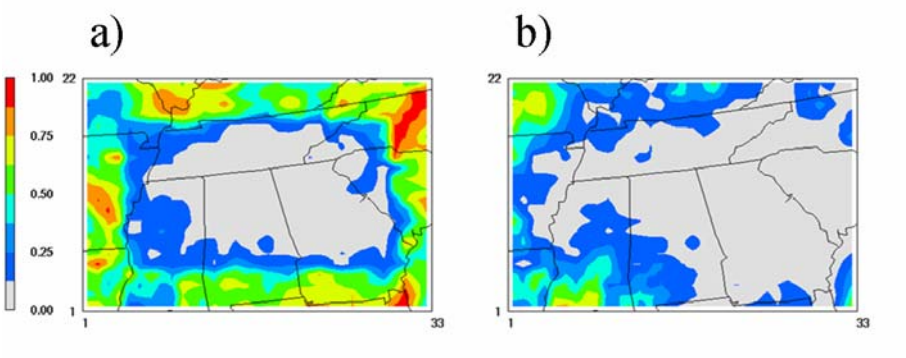

Figure 2. Fractional contribution of total NO_2 column sensitivity to NO_x emissions only from 144km "border" region and b) boundary conditions. The total NO_2 column sensitivityis the sum of sensitivities to each source in addition to a) and b). Low values in these plots indicate low degree of influence from the nonperturbed regions.

The emission rates in each source region were then arbitrarily adjusted by factors ranging between 0.6 and 1.7 and the simulation repeated with the assumption that the emissions were homogeneous within the region. NO_2 concentrations and sensitivities from each simulation were aggregated to column values to more closely mimic the type of available satellite data. The perturbed emissions vector and the corresponding gridded NO_2 column values became the *a priori* estimate for the inverse method ($\overline{E}_t$ and $\overline{\chi}_t^{\mathrm{mod}}$), while the base-case NO_2 columns were used as the representation of the "truth" in the inverse ($\overline{\chi}_t^{obs}$). DKF was then applied iteratively, recalculating concentration and sensitivity fields for each *k*.

During this exercise, the uncertainties in the emissions were set to be relatively high (U_E = 2.0) to allow a large range of deviation from the *a priori* emissions vector in the subsequent estimations. The uncertainty in observations was set low (U_{obs} = 0.1) to allow the modeled values to closely approach the observations. This combination of uncertainty parameters allows for the best test of the robustness of the system at arriving at the correct solution. The dependency of the solution on these assumptions is shown further.

The application of this pseudodata scenario revealed that the proposed inverse method was able to reproduce the original base-case emissions vector within only a few iterations (Figure 3). Both large emissions increases (1.8 times in Macon) and large decreases (0.6 times in Atlanta) were corrected without numerical instability. The corresponding NO_2 fields were also nearly completely corrected (within 1%) after just four iterations (Figure 4Figure 4).

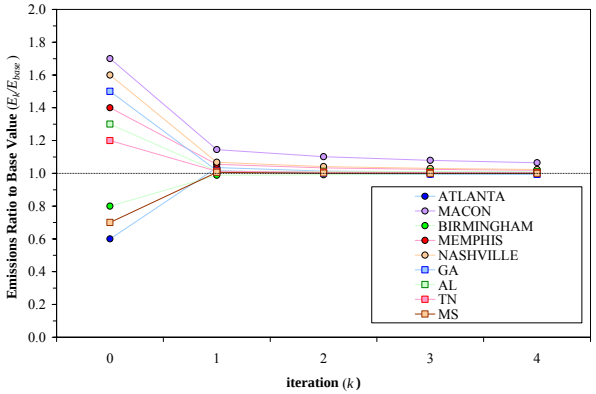

Figure 3. Aggregated regional emissions after each iteration in the pseudodata scenario normalized by the corresponding base-case values.

The response of the system to uncertainty assumptions was also evaluated by analyzing the predicted regional emissions adjustment after the first DKF iteration (*k*=1). In this pseudodata scenario, the acceptable solution was found immediately after the first iteration (Figure 1); thus it was not necessary to carry the solution further for this analysis. A range values between 0.01 and 4.0 was tested for each uncertainty parameter. As expected, large uncertainties in the observations that lead to larger values in the noise matrix (N_t) do not allow for large adjustments to the emissions fields, while large uncertainties in the *a priori* emissions estimates allow for larger adjustments through increasing the values in the initial covariance of error matrix $C_{t,k=0}$. All source regions showed this pattern (Figure 5).

Actual uncertainties in the NO_2 column density measurements from the SCIAMACHY and GOME satellite have been shown to be approximately $0.5x10^{15}$ molecules cm^{-2} +30% from various assumptions in the retrieval algorithms (Martin et al., 2002; Boersma et al., 2004) suggesting values at the lower end of the tested range. It is more difficult to arrive at estimates of emissions uncertainties in specific geographic regions, but it was shown that if there are no mathematical instabilities, larger values should be selected to arrive at a solution in fewer iterations.

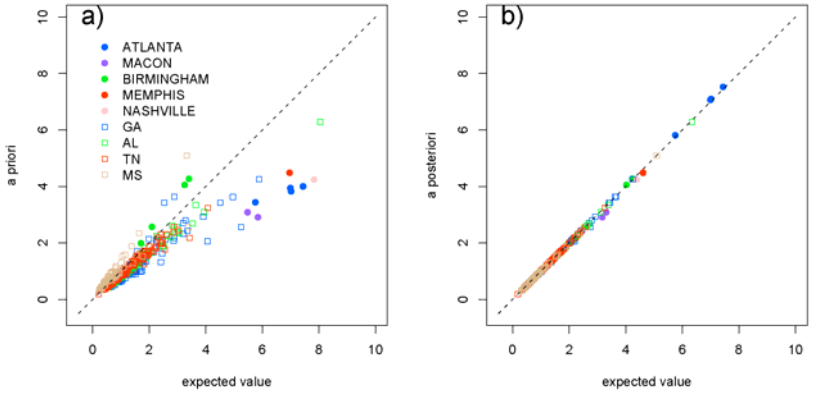

Figure 4. Comparison of expected and modeled NO_2 column values in each grid cell for a) initial perturbed simulation and b) the result of the inverse after four iterations (all values in 10^{15} molecules cm^{-2}).

4. DISCUSSION

The proposed method was successful at reproducing correct regional emissions values in the pseudodata exercise. The translation to actual satellite measurements is likely to be more complex due to the limited coverage of the observations. When retrieval is not disrupted by heavy cloud cover that the instruments are unable to penetrate, the geographical extent of what is observed is relatively small. In the pseudodata test, all grid cells in the domain were allowed to represent an "observation point," thus avoiding difficulties that can arise with real data resulting in the system being mathematically ill-posed and

therefore unconstrained. Other regional scale inverse modeling attempts have used fairly long averaging periods to allow for enough satellite observations to populate the domain (Konovalov et al., 2006). The more recently available OMI satellite has significantly better overpass frequency than older instruments and should help alleviate this problem.

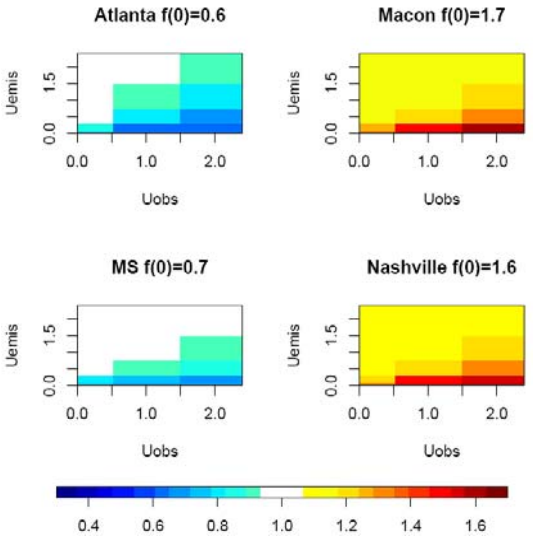

Figure 5. Regional emissions after the first DKF iteration normalized by their corresponding base-case values as a function of uncertainties in observations and uncertainties in emissions. White areas show immediate near perfect prediction. Initial perturbations are noted next to region names

Furthermore, the presented method assumes that the discrepancies in the modeled and observed NO_2 concentrations are due solely to estimates of emissions at the ground level. Uncertainties in the chemical processes, emissions aloft from lightning sources and airplanes, and meteorological predictions also contribute to differences between modeled concentrations of NO_2 and satellite observations. These uncertainties should be quantified and included in the noise matrix. The presented pseudodata analysis tests the reliability of the method before adding these complexities.

Overall, the method is computationally efficient due to the ability to calculate sensitivities directly using DDM-3D and the fact that the matrix operations required by DKF are computationally insignificant. It promises to be directly applicable to NO_x emissions inventory analysis and extendable to other species.

Disclaimer

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Future events

Harbors08

The 2nd international conference on Harbours, Air Quality and Climate Change, 29-30 May 2008 Rotterdam, the Netherlands
Deadline for the submission of abstracts has been extended to 15th of March 2008.
For more information and registration visit www.haqcc.org.

EMS8/ECAC7

The 8th Annual Meeting of the European Meteorological Society (EMS) and the 7th European Conference on Applied Climatology (ECAC) will take place in Amsterdam, The Netherlands on 29 September – 3 October 2008. The scientific programme and abstract submission are now accessible at:
http://meetings.copernicus.org/ems2008
Deadline for abstract submission: 16 May 2008.
Information on registration, accommodation, travel routes, visa requirements, social events and exhibition opportunities will also become available on this conference web site.
This event has a strong focus on all questions related to mitigation and adaptation to climate change. Sessions discussing climate change and variability are paired with a programme that explicitly addresses adaptation plans in different societal and economic areas.

The programme groups
- Forecasting the weather at all time scales – THORPEX studies, developments in NWP, applications and societal impact,
- Meteorology and society, and
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will address a wide spectrum of scientific and application topics in the atmospheric and related sciences, including the challenges to communicate results to society at large.
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Harmo12

The 12th International Conference on Harmonisation within Atmospheric Dispersion Modelling for Regulatory Purposes will be held from 6 to 9 October 2008 in hotel CROATIA, Cavtat, Croatia.
In order to get all updates on the conference please ensure that you have registered your interest via the website
http://www.meteo.hr/harmo12/PreRegistration.html
Please make a note of the following important dates: One-page abstracts by 10 March 2008; extended abstracts by 4 August 2008; Early registration by 30 June 2008.
Organizing Committee
Research and Development Division-Air Quality Research Unit
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ACCENT/GLOREAM workshop

The next ACCENT/GLOREAM workshop on atmospheric chemistry transport modelling will take place in Antwerp, Belgium. The aim of ACCENT/ GLOREAM is to investigate the processes and phenomena which determine the chemical composition of the troposphere by means of advanced and integrated modelling, both on regional (over Europe) and global scale. The workshop will start on Wednesday, 29 October 2008 at 14.00 p.m. and will end on Friday, 31 October 2008 at 14.00 p.m.
For more information on the workshop venue, call for abstracts, workshop registration, accommodation and contact information, please visit:
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NATO/SPS ITM30

The 30th NATO/SPS International Technical Meeting on Air Pollution Modelling and its Application will take place on 18 – 22 May 2009 at Holiday Inn Gateway, San Francisco, United States of America:
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