The Remote Sensing of Atmospheric Constituents from Space

ACCENT-TROPOSAT-2 (AT2): An ACCENT Integration Task

The Remote Sensing of Tropospheric Constituents from Space


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Assimilation of Tropospheric Species into a Chemistry Transport Model

A contribution to ACCENT-TROPOSAT-2, Task Group 2

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Summary

This report aims at presenting the work concerning the assimilation of chemical satellite data from present instruments into the MOCAGE-PALM chemistry-transport model. The goal is to construct assimilated fields of CO by using data from Terra/MOPITT and Aura/MLS. The assimilation methods used is the 3DFGAT scheme via the PALM software developed by the CERFACS. We present a selection of results which have been done in the last years by our lab.

Introduction

The carbon monoxide (CO) with its lifetime between a few weeks to a few months is considered as a very good tracer of pollution. It is produced by incomplete combustion and wild fires are one of the major source of CO. Moreover, CO by reacting with hydroxyl radical OH produces ozone (O₃) which is a major pollutant into the troposphere and is called a precursor of O₃. Since 2000, IR-nadir MOPITT (Measurements Of Pollution Into The Troposphere) instrument on board Terra satellite is capable of measuring CO into the troposphere with between one and two levels of information. And since 2006, we have Aura/MLS CO that covers a vertical domain including the upper troposphere and the stratosphere. The goal is to demonstrate the usefulness of the assimilation of chemical satellite data, in particular to show the different atmospheric structures related to the pollution transport. The assimilation methods used are the 3DFGAT method and the CTM is MOCAGE (Modèle de Chimie Atmosphérique à Grande Echelle). We present results of MOPITT CO assimilation during ITOP2004 (Intercontinental Transport Of Pollution) campaign and an example of dual assimilation using both MOPITT and MLS CO covering then the troposphere and the stratosphere.

The MOCAGE Chemistry Transport Model

MOCAGE is the forecast global 3-D Chemistry Transport Model developed at the Centre National de Recherches Météorologiques of Météo-France, dedicated to the numerical simulation of the interactions between dynamical, physical and chemical processes in the troposphere and lower stratosphere [Peuch et al., 1999]. It is a semi-lagrangian gridpoint model which can run with up to four levels of nested domains (two-way nesting), from the resolution of 2° (global) down to 0.2° over areas of special interest. The vertical grid is composed of 47 hybrid (σ, P) levels from the surface to 5 hPa, with typical resolutions of 40 to 400 m in the boundary layer (7 levels) and about 800 m near the tropopause and in the lower stratosphere. ARPEGE [Courtier et al., 1991] meteorological analyses (horizontal wind and temperature) are used to initialize and constrain the model every 6 hours. The vertical velocity is calculated from these horizontal components by imposing mass conservation for each vertical atmospheric column. Convective processes are simulated with the scheme of
Bechtold et al. [2001], and turbulent diffusion is calculated with the scheme of Louis [1979]. MOCAGE takes into account the chemical scheme RACMOBUS which is the combination of stratospheric and tropospheric chemical schemes, REPROBUS [Lefèvre et al., 1994] and RACM [Stockwell et al., 1997]. RACMOBUS includes 119 species, among which 89 are prognostic variables, and it considers 372 chemical reactions.

**Assimilation of chemistry satellite data into MOCAGE**

We present an example of MOPITT CO [Drummond and Mand, 1996; Deeter et al., 2004, Emmons et al., 2004] assimilation into MOCAGE CTM during ITOP campaign. ITOP experiment took place between US and Europe during summer 2004 (from July 15th to August 8th) and one of its main goals was to analyse the pollution export from US reaching Europe. During this period intense wild fires (biomass burning) spread off over Alaska producing large amounts of CO crossing the American continent before reaching Europe. However, because of the variability of fires, and no adequate emission inventory, models had difficulties to reproduce CO fields. The assimilated CO for the case of 15th of July (Figure 1) is fairly similar to daytime MOPITT CO pixels at 500 hPa; this demonstrates that assimilation corrects the default of unexpected CO sources such as biomass burning.

![Figure 1](image-url)
Dual assimilation of tropospheric Terra/MOPITT CO and stratospheric Aura/MLS CO
The Aura satellite was launched on July 15th, 2004 and placed into a near-polar Earth orbit at 705 km with an inclination of 98° and an ascending node at 13:45 hours. The main objectives of the Aura mission are to study the Earth’s ozone and climate [Waters et al., 2006]. The MLS instrument aboard Aura uses the microwave limb sounding technique to measure chemical constituents and dynamical tracers principally in the stratosphere and mesosphere as well as upper tropospheric constituents [Schoeberl et al., 2006]. It observes thermal emission from the atmospheric limb in broad spectral regions centred at 118, 190, 240, and 640~GHz, and 2.5~THz. In this study we use the first publicly available MLS dataset, version 1.5, of CO. It is retrieved between 215 and 0.0046 hPa with a vertical resolution of ~4 km.

Characterisation of the tropopause layer
To qualitatively evaluate the impact of the assimilation in the region of the tropopause, we calculated the zonal mean representative of a region centered over India. A region of strong CO outflow over India was analysed and one can notice the correction of the dual assimilation (Figure 2) by comparing with the results of the MOCAGE CTM without assimilation. The tropopause layer around 100 hPa, characterized by a strong gradient, drops more rapidly around the latitude 30° for the assimilated field and seems to be smoother compared to the dry runs. In addition, the assimilation corrects the strongest values between 200 and 150 hpa and make them relatively smaller. Above, 100 hPa a small gradient appears principally due to the information from MLS.

Future outlook
The skills we gained concerning the chemical satellite data will serve to develop a tool using assimilation and radiative transfer model to test various configurations for evaluating new concept of satellite instruments, in particular we are focusing on Air quality satellite instrument onboard a geostationary platform.

Figure 2. Zonal mean of CO calculated in the black box over India from MOCAGE without assimilation (left) and with dual assimilation (right). Grey areas represent larger values.
References


Integrating Chemical Modelling and Satellite Observations for monitoring Tropospheric Chemistry and Air Quality

A contribution to ACCENT-TROPOSAT-2, Task Group 2

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Summary

The contribution of the LISA/IPSL group aimed at exploring the potentiality of satellite observations to improve our knowledge of processes constraining the chemical composition of the troposphere and to improve regional scale air quality modelling, with a focus on Europe. Theses objectives are mainly related to work in Task Group 2, synergistic use of models and observations. In the period 2005 to 008, our work focussed especially on the use of GOME and SCIAMACHY tropospheric NO₂ and HCHO measurements for inverse modelling of European NOx and biogenic VOC emissions. Another focus was the use of POLDER and MODIS aerosol optical depths for constraining fire emissions.

Introduction

Satellite observations can help to improve pollution modelling in several ways: (1) through better model evaluation, (2) by giving constraints on pollutant emissions, and (3) through use in data assimilation to improve pollution forecast. In our contribution, we highlighted major results obtained in these directions, in particular the use of satellite observations to constrain pollutant emissions.

Scientific activities and major results

Regional chemistry-transport model evaluation with tropospheric NO₂ columns from GOME and SCIAMACHY

The multi-scale transport and chemistry model CHIMERE (http://euler.lmd.polytechnique.fr/chimere) has been built to simulate photo-oxidant and particulate matter pollution with emphasis over Europe. Computer efficient design allows for long term simulations for assessment of emission control strategies. The model is also used in an operational manner for two days in advance air quality forecast in the frame of the PREVAIR system at INERIS, France (http://www.prevair.org). In a first stage of AT2, we have compared both tropospheric NO₂ columns derived from GOME (by IUP, Univ. Bremen) and SCHIAMACHY (by KNMI) to columns simulated with the CHIMERE model [Konovalov et al., 2005; Blond et al., 2007]. During summer, NO₂ tropospheric columns are strongly correlated to surface NOx emissions; one of the conclusions of this comparison was
that differences in observed and simulated tropospheric NO$_2$ columns are expected to be to be to a large extent due to uncertainty in emission cadastres.

**Inverse modelling of NOx emissions from GOME and SCIAMACHY derived tropospheric NO$_2$ columns**

Inverse modelling studies were performed in order to highlight the additional value of satellite observations for improving anthropogenic NOx emission cadastres over Europe. We have developed an original inversion method with the following key features: (i) replacement of the CTM (CHIME) by a set of empirical models describing the relationships between tropospheric NO$_2$ columns and NO$_x$ emissions with sufficient accuracy, (ii) combination of satellite data for tropospheric NO$_2$ columns with ground based measurements of near surface ozone concentrations, which enabled consistent estimation of uncertainties in the input data together with “a posteriori” emissions and (iii) evaluation of uncertainties of the *a posteriori* emissions by means of special Bayesian Monte-Carlo experiments which are based on random sampling of errors of NO$_2$ columns, emission rates and ozone concentrations [Konovalov et al., 2006a, 2006b]. We applied this algorithm to GOME and SCIAMACHY NO$_2$ tropospheric columns satellite measurements from IUP, Bremen in order to derive correction factors for gridded emissions. Average European and the Near-East summer NOx emissions from both anthropogenic and biogenic sources are, probably, overestimated for Great Britain, Belgium, the Netherlands, Lithuania, Latvia, Poland, Bulgaria, Greece and Iraq, but underestimated for Spain, Italy, Switzerland, the Czech Republic, former Yugoslavia, Turkey, Lebanon and Iran. The emission correction factors are typically within the range 0.5 to 2.

Based on this first work, we used a decadal time series of tropospheric NO$_2$ columns from GOME and SCIAMACHY (from 1996 to 2005) in combination with a continental scale air quality model (CHIMERE$^\circ$ in order to verify and improve available estimates of multi-annual changes of emissions of nitrogen oxides (NO$_x$) in Europe and the Mediterranean area [Konovalov et al., 2008]. As a result, a measurement-based data set of NOx emissions on a 1° by 1° grid and averaged over summer months was elaborated, using an inverse modelling methodology adapted from the work presented above. The results were compared with emission data based on the EMEP emission inventory. Our data are in agreement with the EMEP estimates suggesting a general decline in the level of NO$_x$ emissions in western and central European countries (France, Germany, Great Britain and Poland, and the others, see Figure 1).

Over southern Europe and for shipping emissions, neutral to positive trends are found both for the inverted and bottom-up emissions. In contrast, considerable differences between both data sets are found in some other countries. In particular, significant negative trends instead of the positive ones in the “bottom-up” inventory are found for the Balkan countries, Russia and Turkey. The NO$_x$ emission trends derived from satellite measurements demonstrate larger spatial heterogeneity than those calculated with the EMEP data, especially in Russia and Ukraine.

The estimates obtained of the decadal trends in NO$_x$ emissions for Great Britain were found to be consistent with independent data from the U.K. Automatic Urban and Rural Network (AURN). It was also found that using our emission estimates yields better agreement of model calculations with near-surface ozone measurements of the European EMEP network.
Figure 1. Time series and linear trends in anthropogenic NO\textsubscript{x} emissions averaged over several countries. Values in round brackets are estimates of uncertainties. Values reported in square brackets are the statistical uncertainty (the standard deviation) of a linear fit to the EMEP data.

**Tropospheric HCHO columns as a constraint for biogenic VOC emissions**

Formaldehyde (HCHO) is an important intermediate compound in the degradation of volatile organic compounds (VOC) in the troposphere. Emissions of HCHO are largely dominated by its secondary production from VOC oxidation, with methane and isoprene being the main precursors in unpolluted areas. Due to the moderate lifetime of HCHO of only few hours, its spatial distribution is representative of reactive hydrocarbon emissions. However, biogenic and anthropogenic VOC emissions overlap in Western Europe and are then difficult to separate. Sensitivity studies and a source “tagging approach” using the CHIMERE CTM were used to identify specific areas mainly dominated by biogenic emissions or by anthropogenic emissions [Dufour et al., 2008]. The HCHO tropospheric column observations of SCIAMACHY from IUP, Bremen, with the corresponding CHIMERE simulations for an exceptionally hot summer (2003) and a normal summer (2005) have been compared. The main spatial structures in HCHO fields are both seen in observations and simulations, and are especially related to the variability in biogenic isoprene emissions. Adequate spatial and temporal averaging is needed to reduce noise in observations which are often near the detection limit. Remaining systematic observation errors and model errors are limiting factors for biogenic isoprene emission inversion, but nevertheless use of satellite data is shown to reduce biogenic emissions uncertainties by more than 50% in the most sensitive regions.
We have investigated effects of wildfire emissions on air quality in Europe during an intense fire season that occurred in summer 2003. A meso-scale chemistry transport model CHIMERE is used, together with ground based and satellite aerosol optical measurements, to assess the dispersion of fire emissions and to quantify the associated radiative effects [Hodzic et al., 2006]. The model has been improved to take into account a MODIS-derived daily smoke emission inventory as well as the injection altitude of smoke particles. The simulated aerosol optical properties were put into a radiative transfer model to estimate (off-line) the effects of smoke particles on photolysis rates and atmospheric radiative forcing. We have found that the simulated wildfires generated comparable amounts of primary aerosol pollutants (130 kTons of PM$_{2.5}$, fine particles) to anthropogenic sources during August 2003, and caused significant changes in aerosol optical properties not only close to the fire source regions, but also over a large part of Europe as a result of the long-range transport of the smoke [Hodzic et al., 2007]. Including these emissions into the model significantly improved its performance in simulating observed aerosol concentrations and optical properties. Quantitative comparison with MODIS and POLDER data during the major fire event (3rd–8th August 2003) showed the ability of the model to reproduce high aerosol optical thickness (AOT) over Northern Europe caused by the advection of the smoke plume from the Portugal source region. Although there was a fairly good spatial agreement with satellite data (correlation coefficients ranging from 0.4 to 0.9), the temporal variability of AOT data at specific AERONET locations was not well captured by the model. Statistical analyses of model-simulated AOT data at AERONET ground stations showed a significant decrease in the model biases suggesting that wildfire emissions are responsible for a 30 % enhancement in mean AOT values during the heat-wave episode. The implications for air quality over a large part of Europe are significant during this episode. First, directly, the modelled wildfire emissions caused an increase in average PM$_{2.5}$ ground concentrations from 20 to 200 %. The largest enhancement in PM$_{2.5}$ concentrations stayed, however, confined within a 200 km area around the fire source locations and reached as much as 40 μg/m$^3$. Second, indirectly, the presence of elevated smoke layers over Europe significantly altered atmospheric radiative properties: the model results imply a 10 to 30 % decrease in photolysis rates and an increase in atmospheric radiative forcing of 10–35 W m$^{-2}$ during the period of strong fire influence throughout a large part of Europe. These results suggest that sporadic wildfire events may have significant effects on regional photochemistry and atmospheric stability, and need to be considered in current chemistry-transport models.
Added value

ACCENT/AT2 stimulated and catalysed our research work in a variety of ways: First, the strong collaboration with satellite retrieval groups was crucial, because expertise on satellite retrieved observations is mandatory to ensure proper use of data. Moreover, for inverse modelling studies, detailed information on observation errors is needed and was provided within AT2 project. Last not least, annual AT meetings and specific workshops provided stimulating forums to present and discuss results, and to plan further studies.

References


Dufour, G., F. Wittrock, M. Camredon, M. Beekmann, A. Richter, and J. Burrows, SCIAMACHY formaldehyde observations: constraint for isoprene emissions over Europe, submitted to ACPD


**Derivation of Tropospheric Composition from Satellites using a 3-D CTM**

A contribution to ACCENT-TROPOSAT-2, Task Group 2

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**Summary**

The direct observation of tropospheric composition is difficult and for many species the overlying stratosphere causes a number of complications. Different methods have been employed for the removal of the stratospheric part of total column observations, including using model results. However this is limited by the accuracy of the model. Here we discuss a procedure for the quantitative determination of tropospheric composition through the use of data assimilation to improve model simulations of the stratosphere.

**Introduction**

In determining tropospheric composition from nadir satellite instruments the stratosphere must be properly accounted for in the retrieval process. There have been many different methods employed to try and remove the stratospheric part of the total column observed by such instruments, including using model results. Therefore in order to gain the most meaningful tropospheric information the model must simulate the stratosphere as accurately as possible.

**Scientific activities**

In this work we use a detailed, tested 3-D chemical transport model (TOMCAT/SLIMCAT) to improve the quantitative derivation of tropospheric composition. TOMCAT/SLIMCAT is used to simulate the stratosphere as accurately as possible. The model already gives a reasonable simulation of the stratosphere (following on-going improvements) but in this study we further constrain the stratosphere by assimilating longer-lived species from long-term datasets such as HALOE [Chipperfield, 2002]. The chemical data assimilation is performed using a sequential sub-optimal Kalman filter scheme [Khattatov et al., 2000] and here we assimilate HALOE CH$_4$, H$_2$O, O$_3$ and HCl. The assimilation scheme preserves tracer correlations and the overall effect is to produce a more realistic stratosphere in the CTM.

Using the stratospheric constraints from the CTM we then calculate tropospheric residuals. In the first instance we apply this method to GOME observations of NO$_2$. Retrieved slant columns are usually converted to a vertical column by an air mass factor. The calculated air mass factor is sensitive to surface albedo, cloud fraction and height and aerosol properties. Our method uses data on cloud and aerosol from the Global Retrieval of ATSR cloud Parameters and Evaluation (GRAPE) project and surface albedo is retrieved from GOME. This data in conjunction with a multiple-scattering radiative transfer model enables a more accurate air mass factor calculation.
Scientific results and highlights

Recent work has focused on the assimilation of HALOE species into the SLIMCAT/TOMCAT CTM, in order to improve model estimates of stratospheric NO$_2$. Figure 1 shows a comparison of assimilated and non-assimilated column NO$_2$ values with measurements made by ground based DOAS instruments for 4 sites from 1992 to 2000 taken from the Network for Detection of Atmospheric Composition Change (NDACC). In the standard assimilated case HALOE observations of CH$_4$, H$_2$O, O$_3$ and HCl were used in the assimilation. A sensitivity run was done without the assimilation of O$_3$. The results show an improved comparison between SLIMCAT/TOMCAT and the observations when assimilation is used. This is likely due to the assimilation correcting for errors in the model transport, which may be caused by the use of ECMWF ERA-40 winds to force the model. This is further discussed in Gunn et al. [2008].

![Figure 1. Comparison of assimilated and non-assimilated SLIMCAT/TOMCAT vertical column NO$_2$ with ground based DOAS observations for 4 NDACC sites: Jungfraujoch (45 °N), Issyk Kul (43 °N), Tenerife (28 °N) and Lauder (45 °S). The black crosses represent the observations, the dashed blue line represents the free running model and the yellow dashed line the model with assimilation. The red dashed line is a version of the assimilation model which does not assimilate O$_3$.](image)

Summary

The overall aim of this work was to improve the quantitative retrieval of tropospheric trace species by satellite. This short report has focused on how estimates of the stratospheric contribution for species such as NO2 and BrO can be estimated using a 3D model. In particular, when the model is constrained by chemical data assimilation the stratospheric component can be estimated accurately by this method.

References


Impact of Climate Change on Dynamics and Chemistry of the UTLS: Investigations with a Climate-Chemistry Model

A contribution to ACCENT-TROPOSAT-2, Task Group 2

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Summary

In recent years the climate-chemistry model (CCM) E39C has been further developed and model results have been checked with observations, in particular those derived from satellite instruments. The model evaluation was based on long-term simulations (1960-1999) which have been intensively analysed to identify and quantify the effects of climate change due to enhanced greenhouse gas concentrations and modified sea surface temperatures on stratospheric circulation and composition. The intensive comparison of modelled and measured data pointed to strengths and weaknesses of the CCM E39C. The model evaluation efforts ended up in the upgraded model version E39C-A which is clearly superior to E39C, particularly with regard to the description of dynamical and chemical processes in the upper troposphere and lower stratosphere (UTLS) region.

Introduction

A major problem of E39C, which is very similar to other CCMs and climate models, are significant deviations to observed atmospheric temperatures. E39C exhibits a severe temperature bias in the extra-tropical lowermost stratosphere (i.e. the so-called “cold bias”) as well as in polar regions below the model top layer which is centred at 10 hPa (see also Dameris et al., [2005]). The stratospheric temperature errors have further implications for model dynamics like a delayed break down of the polar vortices at the end of the winter. Furthermore, the comparison with observations indicated biases in simulated tracer fields which might be caused by deficiencies in model dynamics as well as limitations of the applied transport algorithm. For example, the atmospheric water vapour ‘tape recorder’ signal in E39C indicates a too rapid upward propagation in the tropics. E39C also has problems simulating the stratospheric chlorine (Cl\textsubscript{2}) mixing ratios and trends [Eyring et al., 2006]. The latter has an impact on simulated ozone trends which are too weak in E39C (see Chapter 6 of WMO, [2007]). The exact cause and effect relationships inducing these model deficiencies are generally not obvious and figuring them out is often challenging.

Implementing the Lagrangian advection scheme ATTILA in E39C for the transport of water vapour and cloud water as well as other chemically and radiatively active trace species was the most important step to improve the coupled model system with regard to dynamics and chemistry in the UTLS. In the following, some highlights of this new model update, i.e. E39C-A, are described. This contribution gives an extensive summary of the most important model improvements which are explicitly discussed in Stenke et al. [2008b].
Scientific activities

The most important step towards improved model dynamics has been described in the study of Stenke et al. [2008a] employing the fully Lagrangian, numerically non-diffusive, and strictly mass conserving advection scheme ATTILA [Reithmeier and Sausen, 2002] for the transport of water vapour and cloud water in E39 (model version without coupled chemistry). In the operational model version a semi-Lagrangian advection scheme is used for tracer transport which exhibits an exceptional high numerical diffusion in the presence of sharp tracer gradients. In case of atmospheric water content this leads to an artificial horizontal diffusion of water vapour from the tropical upper troposphere into the extra-tropical lowermost stratosphere and a severe overestimation of water vapour within this atmospheric region (wet bias). In turn, the simulated wet bias contributes to a cold bias in the extra-tropical lowermost stratosphere due to excessive long-wave cooling rates. The use of ATTILA results in a pronounced and consistent reduction of the modelled biases, e.g. the cold bias in the extra-tropical lowermost stratosphere is reduced to one third of its original amount. The advancements in simulated temperatures have a remarkable impact on model dynamics, e.g. on the representation of the extra-tropical tropopause or the zonal winds in the stratosphere.

In a second step the described Lagrangian approach has been extended to the CCM E39C, resulting in the upgraded model version E39C-A. A first transient reference simulation has been carried out with E39C-A for the period from 1960 to 2004. This simulation has been performed with the aim to reproduce recent atmospheric conditions as realistically as possible. The data derived from this model simulation enables a detailed evaluation with data and analyses derived from long-term observations.

Scientific results

The study of Stenke et al. [2008a] already has shown how the improvements with respect to the simulated water vapour distribution arising from the use of ATTILA directly feed back to modelled temperatures and other dynamic variables. The most obvious upgrade is found during summer months with both hemispheres showing a much better representation of the transition from westerlies to easterlies between November and January in the Southern Hemisphere and between June and August in the northern hemisphere. The former model version with the semi-Lagrangian advection scheme was not able to simulate the observed wind reversal in the stratosphere below 10 hPa in a realistic manner [Dameris et al., 2005]. The improvements are also visible in the mean annual cycle of both, the zonal mean wind and temperature in the lower stratosphere which leads to obvious advancements in interactions between planetary waves and the mean flow in terms of Eliassen-Palm fluxes and the meridional heat flux. As a consequence, transport of atmospheric tracers is in much better agreement with analyses from observations. For example, E39C-A correctly reproduces the amplitude of the observed water vapour deviation in the tropical lowermost stratosphere. In the lower stratosphere also the decay of the ‘tape recorder’ signal with height is in reasonable agreement with observations [Stenke et al., 2008b].

As a consequence, the Lagrangian transport in E39C-A leads to a significant improvement with regard to the simulated Cl\textsubscript{y} profile. Compared to E39C high stratospheric Cl\textsubscript{y} concentrations extend further downward in E39C-A which is in reasonable agreement with observations and calculated profiles of other CCMs.

To demonstrate the impact of ATTILA on the ozone distribution, two representative ozone profiles from E39C, E39C-A, and radio-sonde observations are compared in Figure 1. In this case the observations are taken from the Binary DataBase of Profiles (BDBP), a new database of high vertical resolution measurements [Hassler et al., 2008] which also includes vertically resolved measurements from satellite instruments. One main improvement of using ATTILA
on the ozone profiles is a better representation of the ozono-pause, *i.e.* the transition of low ozone concentrations in the troposphere to higher concentrations representative for the stratosphere. While in E39C this transition occurs at too high altitudes, E39C-A shows a much better agreement with observations. This effect is most evident at high latitudes, where the deviations of E39C from observations are largest, but also at middle and low latitudes the transition of ozone between troposphere and stratosphere is improved in E39C-A (not shown). One of the reasons for the downward shift of the ozono-pause is certainly the improved representation of the tropopause in E39C-A (for details see Stenke *et al.*, [2008b]).

![Figure 1](image_url)

The maximum in ozone partial pressure around 60 hPa, which is generally overestimated in E39C, is still too high in E39C-A. However, the deviation is slightly reduced in E39C-A for northern high latitudes (Figure 1, left) and for mid-latitudes (not shown). In southern high latitudes the situation is different in that E39C-A shows even higher values of ozone than E39C at all altitudes in winter (Figure 1, right). Note that in E39C, the southern high latitudes are the only region where ozone is not overestimated. This is caused by a compensation of errors, *i.e.* the strong cold bias in this region counteracts the underestimated Cl\textsubscript{y} concentrations. Since the cold bias is significantly reduced and the Cl\textsubscript{y} values are much more realistic in E39C-A, ozone levels in southern polar regions are higher, being consistent with a general overestimation of total ozone in the model.

Not only the spatial but also the temporal evolution of ozone is influenced by the use of ATTILA (Figure 2). Even though absolute ozone values are generally higher in the south polar region in E39C-A, the strong negative trend in total column ozone in spring (September to November) is larger in magnitude in E39C-A. This enhanced trend is consistent with changes in the temporal evolution of Cl\textsubscript{y}. Since E39C is clearly underestimating the loss in total column ozone [Eyring *et al.*, 2006], the changes brought upon by ATTILA are an
obvious improvement towards a more realistic simulation of the evolution of stratospheric ozone.

![Time series of total column ozone anomalies averaged over 60 °S to 90 °S and over September to November for E39C-A (red) and E39C (blue). The anomalies are calculated with respect to the 1960-1969 mean. Linear trends for the period 1970 to 1999 are added as solid lines together with their magnitude; taken from Stenke et al. [2008b].]

**Future outlook**

The recent investigations have shown that many deficiencies in E39C are not caused by the low model top centred at 10 hPa, but primarily by the exceptional high numerical diffusion of the semi-Lagrangian advection scheme and resultant model biases in the distributions of radiatively active trace species which feed back to model dynamics. This result is of general importance for global modellers. However, Lagrangian transport can not be regarded as a universal remedy for all kind of model deficiencies. For example, E39C-A still shows a low temperature bias in the polar stratosphere above 50 hPa in winter, the so-called “cold pole” problem. Nevertheless, replacing the operational semi-Lagrangian scheme in E39C with ATTILA was an important step forwards. Therefore, the next generation of climate-chemistry models, which certainly will still have coarse vertical and horizontal resolution, will need at least less diffusive advection algorithms than formerly used.

**References**


Assimilation of Satellite-Retrieved Tropospheric NO$_2$ Columns and \textit{in-situ} Observations into the CMAQ CTM Model

A contribution to ACCENT-TROPOSAT-2, Task Group 2

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Summary

Tropospheric NO$_2$ columns retrieved from the instruments GOME2 and OMI and ground-level observations of NO$_2$ from monitoring stations have been assimilated into the CMAQ mesoscale model. The 4DVar method has been used to optimize both initial conditions and emission factors.

Introduction

We used for our simulations the model pairs WRF-CMAQ and WRF-CAMx. Several simulations have been performed. Both pairs have been configured for three nested domains with horizontal resolutions 27, 9 and 3 km. As a preliminary work, a comparison of retrieved models with their model counterparts has been done with the pair WRF-CAMx [Eben \textit{et al.}, 2007]. The information contained in satellite columns for data assimilation has been investigated. Several possibilities for using this information have been found. In particular, model bias and stations with low representativeness can be detected this way. Another positive contribution of satellite columns is their lower sensitivity to daily cycles of NO$_2$ concentrations, which enables better tracking of transport phenomena.

Assimilation of satellite columns and \textit{in-situ} observations

In earlier work [Eben \textit{et al.}, 2005] it was found that correction of model concentrations achieved by assimilation of \textit{in situ} observations is effective if we want to improve, for example, an exposure index estimated from long-term off-line simulations. For the purposes of prediction this kind of data assimilation has a limited benefit as long as model bias and errors in emission inputs are present. Another cause of this behaviour is the lack of information for higher levels of troposphere.

In order to improve the forecast performance of the model, further sources of information are required and emission constraining appears to be necessary. Since the latter is hard to achieve by ensemble techniques, a CTM capable of adjoint modelling had to be selected. The adjoint operator for the model CMAQ was being developed by the CMAQ community during recent years [Hakami, 2007]. We have taken part in this development and have finalized the parallelization of the adjoint operator. We also contributed several technical improvements to the adjoint code so as to enable its use for real cases. Finally, observation operators for satellite columns have been implemented. Thus the 4D-Var method constraining both initial conditions and emissions could be used. We used a similar approach as [Elbern \textit{et al.}, 2007]. As a first step, the variability in emissions has been roughly parameterized by allowing for an emission factor, specific for each grid point.
This leads to the discrete formulation of the cost function:

\[
J(c_0,e) = (c_0 - c_B)^T B^{-1}(c_0 - c_B) + (e - e_B)^T K^{-1}(e - e_B) + \sum_{i=1}^{N} (y_i - H(M(c_0,e,t_i)))^T R^{-1}(y_i - H(M(c_0,e,t_i)))
\]

where:
- \(c_0, c_B\) are optimized and first guess concentrations in time \(t_0\)
- \(e, e_B\) are optimized and first guess emission multiplicative factors,
- \(c = M(c_0,e,t)\) are the modelled concentrations,
- \(H\) is the observation operator,
- \(y\) are the available observations, both satellite-retrieved columns and in situ observations,
- \(B, K\) and \(R\) and the covariance matrices for initial conditions, emission factors and observations errors

The cost function \(J\) is minimized utilizing the L-BFGS-B algorithm [Zhu 1997] and the gradient of \(J\) with respect to joint variable \((c_0,e)\) can be expressed as:

\[
\nabla_{(c_0,e)} J(c_0,e) = B^{-1}(c_0 - c_B) + K^{-1}(e - e_B) + \sum_{i=1}^{N} M^T H^T R^{-1}(y_i - H(M(c_0,e,t_i)))
\]

This gradient is calculated by the adjoint method. For easier problem constraining we apply a logarithmic transformation of variables:

\[
u(c_0,e) = (\ln(c_0), \ln(e))\]

The variable \(u\) can be considered as unconstrained.

The implementation of the adjoint in CMAQ is done by similar means as in the model STEM and it is described in [Sandu et al., 2003]. An experimental CMAQ adjoint code was implemented in California Institute of Technology and Virginia Polytechnic Institute. This code contains the adjoint for gas phase processes for the mechanism CB4. We have parallelized it in order to be able to process real experiments and we implemented our 4DVar assimilation above this adjoint code.

The observation operator \(H\) has two separate versions, one for treating tropospheric columns obtained by the retrieval process and the other one for handling in-situ observations. The first one accounts for the spatial intersection of the satellite instrument pixel with grid cells and for the influence of the retrieval process given by the averaging kernel operator. The second one is straightforward. If there are several stations in a grid cell, an average of all measurements has been taken for \(y\).

Similarly the matrix \(R\) is assembled from two blocks. The estimates of standard deviations of the retrieved NO\(_2\) columns are based on values supplied by the TEMIS project. These values are multiplied by a factor (0.3 in this experiment) which takes into consideration smaller time variability of these values in comparison with ground values and thus longer time representativity of these values. As for the block corresponding to the in-situ observation, the base standard deviation of the errors for the stations is calculated as 20 % of the NO\(_2\) average over all stations in the domain during a day. Representativeness errors are taken into account by multiplying this constant by a factor which depends on the type of the station. After some test runs, the multiplicative coefficients were set up to 20, 10 and 1 for urban, suburban and rural stations respectively. This rough approach will be generalized in the future. Only background stations have been taken into account.
The assimilation experiment

In our experiments we used three nested domains. The outer domain encompasses most parts of Europe (horizontal res. 27 km). The assimilation experiment was performed on a sub-window of the outer domain with $72 \times 52$ grid-points. The middle domain (res. 9 km) was used in WRF nested runs only. The fine 3 km resolution domain covers north-west part of the Czech Republic and adjacent regions of Germany ($62 \times 46$ grid-points). The emission data and model were the same as in [Eben et al., 2005].

The tropospheric NO$_2$ columns are retrieved from measurements obtained from satellite instruments OMI and GOME2 (provided by the TEMIS project). The data contain all other necessary information, in particular the averaging kernel operator.

Altogether 280 background monitoring stations have been included into the experiment.

We selected for our experiments an assimilation period of eight days from June 28$^{th}$ to July 5$^{th}$. A free run of CMAQ, 21 days on the outer domain, was performed to obtain reasonable initial and boundary conditions.

For each day sequentially, an assimilation run has been performed, assimilating both NO$_2$ columns and in-situ observations. A one-day-ahead forecast has also been computed, using emission factors and initial conditions obtained by 4D-Var for the past day.

**Results**

After the first two days of assimilation, the emission factors converged to a fairly stable solution. The map of emission factors for one day and the average from the entire assimilation period is shown in Figures 1 and 2.

![Figure 1. Optimized emission factors for July 3$^{rd}$, 2008](image1.png)

![Figure 2. The average of the optimized emission factors from June 29$^{th}$ to July 5$^{th}$, 2008.](image2.png)

Figure 3 depicts the changes between the free run and concentrations corrected by 4DVar. It is seen that both in situ observations and satellite columns contribute to the corrections. In particular, the corrections in France and Italy are induced by satellite observations only. In Figure 4 there is a sample map of the gradient of the cost function with respect to the emission factors.
Figure 3. Differences between optimized and referential concentrations of NO$_2$ for July 2$^{nd}$, 2008 at 20:00.

Figure 4. Emission factors gradient for the first iteration for July 3$^{rd}$, 2008

Table 1 contains results of the one-day-ahead forecast, compared with those of the free run. It is seen that the forecast from assimilated values outperforms the free run, but the bias of the forecast is larger. In Figure 5 there is a sample output for the fine domain, compared with the corresponding zoom of the coarse domain. Assimilation helps here to identify and localize sources in a better resolution (beyond the downscaling due to better orography and meteorology), even though the initial emission inventory is coarse.

Table 1. Mean residuals (i.e. differences of hourly observed value at a station and the model value) and absolute residuals for the free run and forecast from assimilated initial conditions and parameters. We excluded small values of NO$_2$ from the evaluation, so that only values of NO$_2$ larger than 20, either in the observation or in the model, enter the evaluation. All values are in $\mu$g/m$^3$.

<table>
<thead>
<tr>
<th>No. of observations</th>
<th>Free run mean residual</th>
<th>Forecast mean residual</th>
<th>Free run mean absolute res.</th>
<th>Forecast mean absolute res.</th>
</tr>
</thead>
<tbody>
<tr>
<td>58567</td>
<td>2.3</td>
<td>12.1</td>
<td>20.15</td>
<td>17.7</td>
</tr>
</tbody>
</table>

Figure 5. Optimized concentrations in ppmV for July 1$^{st}$, 2008 in the fine domain (left) and in the coarse domain (right).
Conclusions and outlook

The assimilation experiment shows the usefulness of satellite data for constraining emissions and initial conditions in the short term. The improvement in forecasting NO\textsubscript{2} concentrations due to assimilation of in situ and satellite observations is evident. Longer experiments, adaptations of the emission model itself and a deeper investigation of complementarity between in-situ and satellite observations are required before using the technique in operational forecasting.

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References


Eben, K., Resler, J., Jurus, P., Belda, M.: A comparison of tropospheric NO\textsubscript{2} columns retrieved from satellite instruments with model values simulated by the mesoscale model CAMx. Proc. of the 2nd ACCENT symposium, Urbino, July 2007, 5p


Adrian Sandu, Dacian Daescu, Gregory Carmichael and Tianfeng Chai: Adjoint Sensitivity: Analysis of Regional Air Quality Models, J. of Computational Physics 204 (2005), 222 – 252


4-Dimensional Variational Assimilation of Satellite Data into a Chemistry Transport Model

A contribution to ACCENT-TROPOSAT2, Task group 2

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Summary
During AT2 period a prominent suite of satellite data level 2 retrievals (GOME, SCIAMACHY and OMI NO$_2$ tropospheric columns, GOME-NNORSY ozone profiles, MOPITT CO columns, SYNAER PM$_{10}$ retrievals) has been assimilated in the 3 and 4-dimensional variational (3/4D-var) data assimilation modules of the EURAD-IV chemistry transport model and validated. While benefits can be observed for all species assimilated, the signatures of assimilation results are more sustained for aerosols, and least for NO$_2$. In summary, advantages from satellite data assimilation are emerging, especially for aerosol retrievals. However, substantial study efforts are required in optimising assimilation algorithm parameters like covariances and preconditioning measures by well established performance statistics.

Introduction
Due to sparse in situ measurement networks, there is an increasing incentive to assimilate available satellite retrievals in limited area chemistry transport models (CTM). This study was aimed to make optimal use of satellite retrievals by using the 4dimensional variational data assimilation technique (4D-var) as cutting edge method to ingest data into a model. Special emphasis is placed to apply as much sensors as possible, in order to infer not only chemical (initial) states of the atmosphere, but also optimise emission rates. The latter proofs necessary as emission rates are both important for model performance and insufficiently well known.

Scientific activities
All activities were based on EC, ESA and national projects, where the latter built both the theoretical basis, software development and validation of concepts. Practical experiences and extensions to further sensors were later performed in European consortia like ESA GSE-PROMOTE and FP7 GEMS with an additional component of preoperational application and long term evaluation.

During AT2, ingestion of GOME 1, SCIAMACHY and OMI averaging kernels for NO$_2$ tropospheric column assimilation from IFE and KNMI, GOME-NNORSY ozone profiles, MOPITT CO partial columns, and SYNAER PM10 retrievals in the 4D-var data assimilation scheme for gas phase constituents, and 3D-var assimilation scheme for aerosols. The underlying CTM and assimilation system is the EURopean Air pollution Dispersion model – Inverse Model (EURAD-IV, Elbern et al., [2007]). Most efforts were devoted to NO$_2$ tropospheric column and PM$_{10}$ assimilation validation.
Scientific results and highlights

Assimilation of gas phase data

While NNORSY data [Müller et al., 2003] and MOPPIT CO assimilation do not pose a special problem for spatio-temporal data assimilation, tropospheric column data is lumped information, which must be redistributed over height profiles, formally figuring the data assimilation problem as an ill-conditioned integro-differential equation.

An example of combined assimilation of SCIAMACHY and OMI NO$_2$ tropospheric columns with the EURAD-IV CTM for the period July 2006 is given for illustration. Both SCIAMACHY and OMI satellite retrievals from KNMI were assimilated by averaging kernels, using error information from the data provider. In an attempt to provide a horizontal model resolution comparable to the minimal OMI $24 \times 13$ km$^2$ footprints, the horizontal model resolution was refined to $15 \times 15$ km$^2$.

Figure 1 exhibits these conditions, along with retrievals ($y$), forecasted retrievals of NO$_2$ columns ($Hx_b$), and analysed tropospheric NO$_2$ columns ($Hx_a$) for an OMI overpass on July 6th, 2006.

The effect of NO$_2$ column data assimilation for the same day is presented in Figure 2. The field obtained is supplemented by difference fields for the tropospheric columns and the concentrations. Clearly major increments can be observed in western England and in the area of north-western Russia. Both these signals are visible for surface concentrations.

Figure 2. Data assimilation result in terms of tropospheric columns for July 6th, 2006. NO$_2$ model columns based on OMI and SCIAMACHY assimilation within the assimilation interval, 09-12 UTC. Units in molecules/cm$^2$ (left panel). Difference field giving implied changes for tropospheric columns by assimilation (middle panel), and induced surface concentration changes by NO$_2$ in ppb.
Retrieval results from tropospheric NO$_2$ columns are ingested into the model by means of averaging kernels, where the observation operator $H$ is constructed by the scalar product of the averaging kernel with the NO$_2$ molecular density of the model profile [Eskes, 2005]. The average fraction of the averaging kernel at the surface is roughly about 10% of the maximal amplitude in most cases, as can be observed from Figure 3, left. Recalling the significance of the averaging kernel shape as a sensitivity profile, corrections by the DA procedure are enforced at the same proportion as the sensitivity. The practical meaning is that the modification of the surface layer is by far not as affected as in comparison to the free troposphere. This must be recalled when considering the following results.

![Figure 3](image.png)

**Figure 3.** Left panel: Mean averaging kernel over the European continental scale model domain and two weeks case study. Right panel: probability density functions (pdf) of differences between observations and several model runs for July 8$^{th}$, based on OMI retrievals. Each panel displays observation differences with control run (OmC) (no data assimilation at all,) black bold line, differences between observations and forecasted values (OmF), green bold line, differences between observations and analyses (OmA), blue bold line. For comparison, Gaussian fit to OmF pdf by mean and standard deviation given by broken purple line.

Satellite data assimilation, as other assimilated data, impinges on the model, dependent on the evaluation procedure. For NO$_2$ tropospheric columns approaches of evaluation can be distinguished in the following ways:

1. **Performance improvements in terms of probability density functions (pdf) of Observation minus Forecast differences (OmF) and likewise with analyses (OmA) (following Talagrand, [2003]):** The visual result presented in Figure 2, can be corroborated quantitatively by the OmA and OmF pdfs displayed in Figure 3, right panel, where July 8$^{th}$ 2006 is presented. The marginal forecast improvement is reflected by the close coincidence of the observation-minus-forecast run pdf (OmF) with the observation-minus-control run pdf (OmC). It is however visible, that a slight bias reduction occurs, which will continue to prevail with progressing days. Clearly, the analysis shows a significant improvement in terms of both, bias and variance reduction.

2. **Performance improvements in terms of better forecasts, validated against surface data:** A clear validation result indicating forecast skills improvements by surface in situ stations is difficult to obtain. On the one hand the averaging kernel with only 10% average signal strengths at the ground (Figure 3, left) implies only minor impact in near surface layers, while on the other hand NO$_2$ is a constituent reacting too fast for displaying a direct benefit for the next days forecast. As concerned the indirect effect via ozone improvement, two cases must be distinguished: In the rural case,
ozone formation is often controlled by low NO\textsubscript{x} levels, where relative variance of NO\textsubscript{2} column retrievals, and hence also error margin, is high and the assimilation is affected by these conditions. In the case of urban areas, the spatial resolution problem of the model and its emissions prevail, as was shown by Elbern and Strunk [2006], who found an effective horizontal resolution of 6 km.

Assimilation of remote sensing aerosol data

Within the AERO-SAM project (Boundary layer AEROsol characterisation from Space by advanced data Assimilation into a tropospheric chemistry transport Model), an assimilation system for tropospheric aerosols, capable of handling both in situ and remote sensing aerosol data, is being constructed for the EURAD-IV CTM. Using the SYNAER algorithm (SYNergetic AErosol Retrieval; Holzer-Popp, [2001a,b]), delivering aerosol optical thicknesses (AOT) for several intrinsic aerosol components, the assimilation system can assimilate distinct aerosol types, without need to lump all aerosol components into a single quantity like PM\textsubscript{x} (particulate matter with a diameter less or equal to \(x\) \(\mu\)m). The SYNAER retrieval makes use of two sensors AATSR/ATSR-2 and SCIAMACHY/GOME onboard Envisat/ERS-2 and delivers AOT for five aerosol classes (WAter SOlubles, INSOlubles, soot, transported mineral dust and sea salt), which are assimilated.

To be able to assimilate data like AOT columns delivered by SYNAER, which are not directly related to a model state space variable on an advanced level, the observation operator \(H\) and its adjoint \(H^T\) for the SYNAER retrieval has to be built as a mapping between model and observation space. Furthermore, these operators have to be implemented in the existing EURAD-IV CTM aerosol module MADE (Modal Aerosol Dynamics model for Europe) with the secondary aerosol model SORGAM. The technical development of \(H^T\) (the radiative transfer model part of SYNAER) has been done during the exchange visits of two PhD students funded by AT-2. In the first stage of AERO-SAM it was implemented in a 3D-var system.

In 3D-var the assimilation is accomplished at one point in time. Minimisation of discrepancies between observations and model state delivers the most probable state of the atmosphere, called analysis. Figure 4 shows the results of 3D-var assimilation using SYNAER species resolved AOT measurements.

In comparison to an assimilation of PM\textsubscript{x}, where all species would have been adjusted by the same relative amount, it can be seen that the rather sparse measurements of SOOT (accumulation mode elemental carbon is chosen as representative EURAD species) lead to an increase, while the WAter SOlubles (represented by accumulation mode sulphate) are slightly depleted. Especially under extreme conditions like these (severe wildfire activities in Spain) it can clearly be stated that it is necessary to use an assimilation system capable of resolving aerosol types to avoid false attribution to aerosol types not involved in biomass burning. This approach aims towards the implementation of a full 4D-var aerosol assimilation scheme for the EURAD-IV.
Figure 4. Results of aerosol 3D-Var using SYNAER species resolved measurements for July 13th 2003. Left: SYNAER measurements (AOT); centre: Difference of Analysis and Background (µg/m³); right: Analysis field (µg/m³). Top row: SOOT, respectively elemental carbon in accumulation mode; bottom row: sulfate in accumulation mode as representative for WASO.

References

A Synergistic Approach for Deriving Regional Tropospheric NO$_2$

A contribution to ACCENT-TROPOSAT-2, Task Group 2

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Summary

In order to account for the high spatial variability of NO$_2$ sources in Europe a synergistic method has been developed to derive regional tropospheric NO$_2$. The approach utilizes satellite observations from SCIAMACHY as well as stratospheric and tropospheric NO$_2$ forecasts from chemical-transport models. This method enables to particularly tackle the stratospheric NO$_2$ variability and its contribution, the tropospheric vertical NO$_2$ distribution and the influence of the cloud-top height. The method combines the strengths of space borne spectrometers like SCIAMACHY and chemical transport modelling to gain accurate regional NO$_2$ tropospheric columns for Europe.

To evaluate the method some case and sensitivity studies have been performed within TROPOSAT-2. Since the cloud-top height is a critical parameter, 4 different operational algorithms have been compared. The stratospheric NO$_2$ content and its variability have been examined using data assimilation and two different MIPAS observational data sets. The temporal and spatial variability of the resulting tropospheric NO$_2$ columns has been investigated by geostatistical means.

Introduction

Since the beginning of the industrialization the chemical composition of the atmosphere has changed significantly. Among the emitted pollutants, nitrogen oxides (NO$_x$ = NO + NO$_2$) play a central role in tropospheric chemistry. NO$_x$ is mainly released by industry and traffic, biomass burning, microbiological activity in soils and lightning. Due to its short lifetime tropospheric NO$_x$ is closely linked to its sources. The high spectral resolution of the space-borne spectrometer SCIAMACHY in the UV/VIS range enables to derive column densities of NO$_2$ [Burrows et al., 1998]. Several methods to derive the tropospheric NO$_2$ content have been developed mainly using a DOAS (Differential Optical Absorption Spectroscopy) fitting technique (Platt et al. 1994). Two key problems are the quantification of the stratospheric contribution and the calculation of the tropospheric air mass factor. Velders et al. [2001] apply the tropospheric excess method [Richter and Burrows, 2002] and image processing technique [Leue et al., 2001] to GOME data. The tropospheric excess method allows to separate the tropospheric and stratospheric NO$_2$ content assuming a relatively stable (both in time and space) NO$_2$ distribution over the free Pacific Ocean. A constraint is that stratospheric (zonal) variability is neglected. Martin et al. [2002] use a tropospheric chemical-transport model to determine the tropospheric air mass factors as a function of the NO$_2$ profile shape. For this approach usually monthly means of tropospheric NO$_2$ from global models are used. This implies two disadvantages: The temporal variability related to the individual weather conditions or emission rates like the day of the week (weekend or working day) is neglected. Furthermore, the coarse resolution of global models smears out the strong gradients for NO$_2$, as it is strongly linked to the sources (i.e. cities).
Scientific activities and results

Derivation of tropospheric NO\textsubscript{2} column

In order to tackle the abovementioned problems a synergistic method has been developed utilizing tropospheric and stratospheric chemical-modelling capabilities. To quantify the stratospheric contribution in NO\textsubscript{2} slant column density retrievals from SCIAMACHY, time-referenced forecasts of the online stratospheric CTM (ROSE/DLR), driven by meteorological wind- and temperature fields, are used. A bias is avoided by scaling the stratospheric NO\textsubscript{2} forecasts to “clean” observation conditions at a reference sector. The stratospheric NO\textsubscript{2} slant column density (SCD) is then derived by applying a geometric air mass factor (AMF). A similar approach was developed by Eskes et al. [2003] using a CTM including data assimilation. For the determination of the profile dependent tropospheric AMF, chemical forecasts of the air quality model EURAD-CTM are applied [Jakobs et al., 2002]. The tropospheric AMF is then derived by weighting the height dependent air mass factors with the relative NO\textsubscript{2} concentrations. This allows us to compute the air mass factor as a function the forecasted NO\textsubscript{2} profile shape:

\[
AMF = \frac{\sum AMF_i \cdot c_i}{\sum c_i}
\]

\(AMF_i\) denotes the height dependent air mass factor for level \(i\) and \(c_i\) the NO\textsubscript{2} concentration of level \(i\). The height dependent air mass factors are taken from a pre-calculated sensor specific look-up-table. To gain the concentrations of a certain level the EURAD NO\textsubscript{2} volume mixing ratios are integrated to partial column taken into account the temperature variability. In a final step, the tropospheric NO\textsubscript{2} column can be derived using the tropospheric slant column and the air mass factor for pixels with a cloud fraction lower than 20\%. Cloud-fraction and cloud-top height are taken from SCIAMACHY, too. The air mass factor calculation further considers surface albedo and surface height. The surface albedo is taken from the climatology of Koelemeijer et al. [2001]. Error estimates for the resulting tropospheric NO\textsubscript{2} columns caused by these factors were quantified by Boersma [2004]. The resulting tropospheric NO\textsubscript{2} vertical column does not depend on the NO\textsubscript{2} column of the model forecast, but on the profile shape.

Monitoring tropospheric NO\textsubscript{2} over Europe

The near-real-time computation of SCIAMACHY tropospheric NO\textsubscript{2} VCD has been performed since August (see Figure 1). This operational service is part of the GMES Service Element Project PROMOTE (http://www.gse-promote.org). Enhanced levels of pollution are seen over the heavily industrialised regions in the central and southern parts of the UK and the Benelux countries, as well as over major cities like Madrid, Paris, London and Moscow. The tropospheric NO\textsubscript{2} columns are generally higher during wintertime. This observation is consistent with the fact that the amount of fossil fuel burnt during winter is higher than during summer due to cold weather conditions, and that the lifetime of NO\textsubscript{2} is longer during winter.

Evaluation of stratospheric NO\textsubscript{2} using MIPAS observations

MIPAS observations were assimilated using a modified version of the chemistry-transport model ROSE/DLR [Baier et al., 2005] to derive consistent global chemical analyses of the stratosphere. Two different MIPAS data products are examined: the ESA operational product [Carli et al., 2003] and the IMK (Institute for Meteorology Karlsruhe) scientific product [Stiller et al., 2002].
The two data sets differ in coverage and type of chemical species observed. Due to these differences, assimilation results differ also considerably. ENVISAT/MIPAS baseline observations of H\textsubscript{2}O, O\textsubscript{3}, HNO\textsubscript{3}, CH\textsubscript{4}, N\textsubscript{2}O and NO\textsubscript{2}, covering October to November 2003, were considered. Sequential assimilation was performed using an optimum interpolation scheme with error propagation. Improvements to model results without assimilation of MIPAS data were quantified by comparisons to HALOE observations. Results show an r.m.s. error reduction of up to 30\% for the assimilated species [Baier et al., 2005].

Cloud-top height comparison
In order to examine the influence of cloud-top height information (CTH), a comparison of four operationally available products was performed for all data of April 2007: SACURA using SCIAMACHY (Semi-Analytic CloUd top height RetrievAl) [Kokhanovski et al., 2006], Aura/OMI effective CTH retrieval algorithm [OMI, 2002], ROCINN using GOME-2 (Retrieval of Cloud Information by a neuronal network) and APOLLO using AVHRR (Avhrr Processing scheme over Land, cLoud and Ocean). Methods 1 to 3 are based on absorbing O\textsubscript{2} (respectively the O\textsubscript{2}-O\textsubscript{2}) as an indicator for the CTH. Method 4 uses meteorological analyses from ECMWF (temperature). The study yielded a good overall agreement between the different algorithms. CTH derived from OMI exhibits a low bias of 1.3 km in comparison to CTH retrieved from GOME-2/ROCINN, whereas SACURA-CTH exhibits a positive bias of 1.06 km in comparison to GOME-2/ROCINN. Due to the fact that the OMI CTH retrieval has to be regarded as a measurement of the effective CTH which includes solar irradiance penetrating into the uppermost cloud layers, the retrieved values are to be expected to yield lower altitudes. The study reveals no statistically significant dependency when considering the miss-time-error for two (or more) different instruments. Only measurements with a maximum miss-time of less than 6 hours have been considered. As a result, it can be stated that the systematic differences in CTH exceed the temporal variability of the atmosphere.
**Geostatistical analysis of tropospheric NO₂**

Information on tropospheric NO₂ VCD derived from SCIAMACHY is sparsely distributed in space and time due to the measurement geometry and cloudiness. In order to generate synoptic maps of tropospheric NO₂ VCD reflecting the variability, a geostatistical method was applied. By means of kriging the best linear unbiased estimation for interpolated values based on the statistical distribution of the measurements is achieved. Synoptic maps of tropospheric NO₂ VCD can be derived by co-kriging of averaged data from the most recent 30 days (depending on the number of successful measurements) with NO₂ VCD derived from SCIAMACHY measurements for the current day. Figure 1a shows the tropospheric NO₂ VCD as retrieved from SCIAMACHY using the synergistic approach of this report. The 30-days running mean of the tropospheric NO₂ column is used as secondary variable and is depicted in Figure 1b. The co-kriging result obtained with optimized settings is depicted in Figure 1c. Figure 1d shows the innovation due to co-kriging and an additional weighting according to the estimation variance obtained from the kriging algorithm itself. In case of a high estimation variance, only information from the running mean is ingested at this point. When the estimation variance is significantly lower than the running mean value, only information obtained from co-kriging is considered. It can be concluded that the combination of running mean values of tropospheric NO₂ VCD with values retrieved for the current day is a very promising approach to yield composites in near-real time.

**Future outlook**

The presented synergistic approach is currently applied to MetOp/GOME-2 observations, where the spatial and temporal coverage is better compared to SCIAMACHY.

**References**


Variability of Atmospheric Composition Associated with Global Circulation Patterns using Satellite Data

A contribution to ACCENT-TROPOSAT-2, Task Group 2

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Summary

In this contribution we analyze how climate variability and associated large-scale circulation modes relate to atmospheric composition and eventual atmospheric composition change, with a focus on the northern hemisphere. Our work has been twofold: on the one hand we focussed on an evaluation of tropospheric composition over Europe using the nitrogen dioxide ($\text{NO}_2$) monthly data during the 1996 to 2006 period. On the other hand total ozone daily data products from 1995 to 2005 have been used to evaluate seasonal and monthly global climatologies. Results have been compared with other total ozone column datasets available and the inter-annual variability of total ozone has also been analyzed and related to the North Atlantic Oscillation (NAO).

Introduction

Tropospheric NO$_2$ columns from GOME have been retrieved and interpreted by several research groups [Lauer $et al.$ 2002; Richter and Burrows 2002; Richter $et al.$ 2005]. Numerous studies have shown the relevance of the NAO to the winter surface climate of the Northern Hemisphere in general and over the Atlantic/European sector in particular [Van Loon and Rogers, 1978; Hurrell, 1995; Trigo, $et al.$ 2002]. Creilson $et al.$ [2003] have analyzed monthly climatologies of tropospheric ozone for five different regions across the North Atlantic and showed that distinct springtime inter-annual variability over eastern North Atlantic and western Europe is particularly evident and exhibits similar variability to the positive phase of the NAO (correlations coefficient values of + 0.61). Using a 15-year model simulation of the dispersion of passive tracers representative of anthropogenic emissions from northern hemisphere continents, Eckhardt $et al.$ [2003] studied inter-annual variability of pollution pathways from northern hemisphere continents into the Arctic and showed that the NAO exerts a strong control on the pollution transport into the Arctic, particularly in winter and spring.

Thompson and Wallace [2000] had previously investigated the signature of the annular modes in total column ozone using TOMS data for the period of record of the Nimbus-7 satellite (November 1978–April 1993, their NH analysis being restricted to March), focusing on the active seasons, that is, when the annular modes amplify with height upward into the lower stratosphere.
Thus, these are research topics of interest and the longer the data records the better these results may be investigated and confirmed.

**Aims and scientific activities**

On our work we focussed on an evaluation of tropospheric composition over Europe using the nitrogen dioxide (NO$_2$) monthly data within the 1996 to 2006 period. The Global Ozone Monitoring Experiment (GOME) onboard ESA’s ERS-2 provided global measurements of atmospheric species from space. Measurements from the GOME instrument were used to retrieve the amounts and distributions of tropospheric trace constituents from space. Since the beginning of August 2002 the SCanning Imaging Absorption spectroMeter for Atmospheric CartograpHY (SCIAMACHY), launched on board of ENVISAT, is also making global observations. Tropospheric NO$_2$ columns from GOME and SCIAMACHY derived at the Institute of Environmental Physics of the University of Bremen were kindly provided by Dr. Andreas Richter and used in this research to study both monthly and seasonal tropospheric composition variability over Europe within the 1996 to 2006 period.

On the other hand total ozone daily data products kindly provided by Dr. Mark Weber, from 1995 to 2005, used in this study have been retrieved from nadir observations of the GOME using WFDOAS algorithm approach [Coldewey-Egbers et al., 2005]. This algorithm has been extensively validated by comparison with measurements from the World Ozone and UV Radiation Data Centre and excellent agreement between WFDOAS results and ground data was found [Weber et al., 2005]. Obtained results have here been compared with total ozone data from the Earth Probe Total Ozone Mapping Spectrometer (TOMS, version 8) Instrument.

The NAO index used here is the one developed by the Climatic Research Unit (University of East Anglia, UK) and is defined, on a monthly basis, as the difference between the normalized surface pressure at Gibraltar, in the southern tip of the Iberian Peninsula) and Stykkisholmur, in Iceland [Jones et al., 1997].

Finally, attention has been devoted to the summer 2003 total ozone anomalies, at the monthly and seasonal scales, and these have been evaluated and related with corresponding anomalies of different meteorological fields: low tropospheric temperature, geopotential height, cloud cover and several radiation parameters. All meteorological fields were extracted from the NCEP/NCAR reanalysis dataset, including monthly averages of net short and long wave radiation fluxes, total cloud cover at surface and temperature and geopotential height at the surface, 850 and 500 hPa. Monthly and seasonal anomalies have been computed separately for all datasets after removing the seasonal cycle. Climatological averages were computed for the relatively short period with available data (11-year period).

**Results and highlights**

*Monthly NO$_2$*

Monthly means and anomalies were calculated on three selected regions (Iberian Peninsula, northern Europe and northern Italy). Monthly means for winter months are presenting the higher NO$_2$ values over northern Europe. The high values obtained in the North of Italy, a well know pollution hot spot just south of the Alps are remarkable. The lower monthly values are obtained for summer months. Individual trends were analyzed and the northern Europe region presents a decreasing trend while the Iberian Peninsula shows an increasing trend (Figure 1).
Figure 1. Standardized GOME and SCIAMACHY monthly anomalies in Iberian Peninsula and trends obtained separately for both datasets (Units are $10^{14}$ molecules/cm$^2$).

**Monthly and seasonal total ozone**

Monthly mean patterns of Total O$_3$ obtained with GOME data are in good agreement with patterns obtained with TOMS data. Monthly mean patterns of Total O$_3$ obtained with TOMS performed over the period 1995-2005 are also in good agreement with patterns obtained with TOMS data for the 29-year period. Similar results are obtained for seasonal mean patterns (winter patterns in Figure 2 left).

**Interannual total ozone variability and NAO**

Results obtained by separating total O$_3$ values for positive NAO ($> 0.5$) and negative NAO ($< -0.5$) reveal different spatial patterns (Figure 2 right), showing that in winter, total O$_3$ exhibits similar variability to the NAO. Results show that the springtime inter-annual variability also depends on winter NAO regime. Correlation patterns between winter NAO index and total O$_3$ values for both the winter and spring seasons suggest that this inter-annual variability may be mainly due to a large scale mechanism.

![Figure 2](image-url). Winter total ozone pattern obtained with GOME 1995-2005 global data records (left); and obtained by subtracting winter total O$_3$ values for positive NAO ($> 0.5$) and negative NAO ($< -0.5$) (right) (Dobson units, DU).

Previous results have highlighted the link between tropospheric ozone and favourable weather conditions (less cloud cover, high temperature and high short wave radiation [Solberg et al. 2008]. Here we show that this link is also valid, at least partially, when we consider the total ozone column. Unfortunately lack of data for southern Europe in July and August do not allow elaborating further on this relationship.
Achievements and future outlook

Although our research confirm most known and published results, most of these have been obtained with short available data records, and it is of the greatest importance to further obtain longer time series which allow the greatest significance of obtained conclusions. The use of longer time series would also allow the study of eventual atmospheric composition change and we will be looking forward to have climatological datasets.

References


Scientific Interpretation of SCIAMACHY CO, CO$_2$ and CH$_4$ Measurements

A contribution to ACCENT-TROPOSAT-2, Task Group 2

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Summary

In 2008 the main focus of this project has been on the interpretation of SCIAMACHY CH$_4$ measurements, in particular the use of the 4D-VAR technique to infer CH$_4$ sources and sinks from a combination of surface and satellite measurements. In comparison to an inversion with surface measurements only, the SCIAMACHY data indicated an increase of emissions over the tropical continents by more than a factor of two. We have further investigated these emissions using aircraft measurements. It turned out that the optimized emissions overestimate the in situ observed vertical methane profiles. In the meanwhile, however, considerable progress has been made on the spectroscopic parameters that are used in the SCIAMACHY CH$_4$ retrieval, which improve the agreement between the satellite-optimized model and in situ measurements.

Introduction

The SCanning Imaging Absorption spectroMeter for Atmospheric CHartographY (SCIAMACHY) instrument, which was launched on board the ENVISAT satellite on March 1$^{st}$ 2002, measures in the near-infrared (NIR) global concentrations distributions of, among others, CH$_4$, CO$_2$ and CO. These gases play an important role in tropospheric chemistry and climate change. A good knowledge of the global distributions of these gases is important to understand the present day chemical composition and to predict changes in the near future.

The aim of this project is to investigate what can be learned from the SCIAMACHY measurements. This will be done in a comparison with atmospheric transport models to identify shortcomings in our understanding of source and sink processes, which will be quantified using the inverse modelling technique. In a later stage of the project the aim is to combine the constraints that come from atmospheric remote sensing, with that of surface measurements networks and data from other satellites that measure surface properties such as FPAR (photosynthetic activity), fire counts and burned area (biomass burning), as well as emerging techniques to measure wetland area.

In this stage of the project the main emphasis is on the interpretation of the SCIAMACHY CH$_4$ product, which is of a sufficiently high quality to allow detailed scientific analysis. Even more interestingly, the data show unexpected features over the tropics, which may point to the gaps in our understanding of tropical CH$_4$ sources. In the previous report, results were shown of transport model sensitivity tests using methane emission scenarios that included elevated tropical wetland and vegetation emissions. Meanwhile we have carried out CH$_4$ inversions
including SCIAMACHY data using our 4D-VAR system, of which results will be shown in this report. In addition, we have performed high-resolution simulations of the Amazon region using the TM5 model with a zoom over South America to support the planning of an in-situ measurement campaign, and to further investigate the SCIAMACHY data and compare with other available measurements.

**Results**

*Inverse modelling of CH$_4$ sources and sinks using SCIAMACHY data*

After various inverse calculations had been carried out to test the TM5 4D-VAR, as discussed in the previous report, inverse modelling calculations have been performed using SCIAMACHY retrievals. The top panel of Figure 1 gives an example of the SCIAMACHY data that were used after bias correction. The bias correction consists of monthly polynomial functions that are fitted to the data in order to account for a seasonal and latitudinal concentration variation that was believed to be an instrument artifact. The bottom panel of Figure 1 shows the difference between the inversion-derived prior and a posterior fluxes, highlighting increased emissions over tropical forests and a reduction in the emission over South East Asia. Frankenberg *et al.* [2008a, 2008b] studied the causes for the bias correction in further detail and found out that these could largely be attributed to inaccuracies in the methane and water vapour spectroscopy.

![Figure 1. Comparison of (top panel) bias corrected SCIAMACHY-CH$_4$ measurements and (bottom panel) the difference between inversion-derived prior and a posterior methane concentrations [Meirink *et al.*, 2008].](image-url)
Regional modelling of methane sources in the Amazon region using TM5

TM5 simulations have been carried out to further investigate methane emissions in the Amazon region. Figure 2 shows a comparison of methane vertical concentration profiles at Santarem, Brazil, as measured during aircraft measurement campaigns coordinated by NOAA ESRL [Miller et al., 2007] and TM5 simulations. Model simulations are shown on the basis of prior fluxes, posterior fluxes using the surface data only and posterior fluxes derived using both surface and satellite measurements. As can be seen, the prior fluxes clearly lead to an underestimation of the methane concentrations as measured by the aircraft, pointing to concentrations that are significantly elevated above background and therefore represent contributions of regional methane sources. When surface measurements are taken into account this leads to an improved agreement between model and measurements over Santarem (note that the aircraft measurements are not used in the inversion). This is remarkable, because the surface network is virtually insensitive to emissions from the Amazon. However, emissions are nevertheless adjusted to improve the model simulated north to south concentration gradient, for which the Amazonian fluxes are a 'cheap' option since its uncertainties are very high, and therefore the corresponding prior fluxes receive low weight in the inversion. Inversions that account for both in situ and SCIAMACHY measurements tend to overestimate the methane concentration profile at Santarem. Similar results are obtained at Manaus (not shown).

Figure 2. Comparison of aircraft measured and TM5 simulated methane concentration profiles at Santarem, Brazil. Black, measurements; blue, TM5 using prior fluxes; green, TM5 using posterior fluxes obtained using surface measurements; blue and red, TM5 using surface measurements and SCIAMACHY retrievals. The differences between blue and red represent the sensitivity to prior assumptions [Meirink et al., 2008].

As explained earlier the overestimated concentrations over Santarem using SCIAMACHY data are caused in part by the spectroscopic inaccuracies reported by Frankenberg et al. [2008a, 2008b]. Additional inverse modelling calculations were carried out to assess the impact of spectroscopy on the estimated methane sources. The results are summarized in Table 1. The inversion using surface measurements (S1) increases the estimated methane flux over tropical South America by about a factor 2. When the uncorrected SCIAMACHY data are used this leads to an additional 50% increase of the methane emissions (S2). However, when the spectroscopic inaccuracies are accounted for, the emissions obtained are only slightly elevated in comparison with the inversion using in situ data only. This result is consistent with the comparisons presented in Figure 2.
Table 1. Summary of inversion-derived methane emissions. S1, inversion using *in situ* data only; S2, inversion using *in situ* data and uncorrected SCIAMACHY retrievals; S3, inversion using *in situ* data and spectroscopy corrected SCIAMACHY retrievals [Frankenberg *et al.*, 2008a].

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References


Assimilating Remote Sensing Derived Air Quality Parameters into a CTM: 
A Study of Data Suitability and Assimilation Techniques.

A contribution to ACCENT-TROPOSAT-2, Task Group 2

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Summary
The National Environmental Research Institute (NERI), Department of Atmospheric Environment (ATMI), runs and continuously develops an air pollution forecast system: Thor. Thor includes a Chemical Transport Model (CTM): The Danish Eulerian Hemispheric Model (DEHM). To further improve the prognostic capabilities and accuracy of the Thor system, work has been done to implement data assimilation of satellite derived air quality parameters into the Danish Eulerian Hemispheric Model.

NERI have started a project ThoRSDA (Thor Remote Sensing Data Assimilation) in a contract with ESA/GMES/PROMOTE, that will give access to funding as well as NRT access to data for project development and implementation. The project was started late 2007 to be completed in late 2009.

Introduction
The objectives of this work are to investigate the availability of remote sensing (RS) derived air quality parameters, to implement data assimilation of these into the CTM and to research which data and which data assimilation techniques better benefits the performance of the forecasts. The new version of the CTM, with data assimilation, is now running as scheduled and the output is in process of being evaluated and being compared with the earlier version as well as ground truth measurements from air quality measuring station networks.

Scientific activities
Considerations on which parameters would be best to include in the CTM, NO$_2$, CO, SO$_2$ and PM seems to be those most relevant to the air pollution forecast module. CO$_2$ and H$_2$O may well be good candidates as control parameters. Data availability, in NRT form, limits the options to NO$_2$ from the OMI sensor as well as NO$_2$ and SO$_2$ from the SCIAMACHY sensor.

The CTM is defined in 20 vertical layers extending well up into the stratosphere (around 20 km). The data retrieved from remote sensing (RS) data is delivered as total column concentrations. In order to compare the concentrations in the layered model with the RS totals, the model values are summed to one total column value for the troposphere. This value is then adjusted with the RS information also covering the troposphere, for example, the NO$_2$ measurements, finally to be re-distributed and fed back into the model. The re-distribution is done so the relative distribution between layers is the same after, than before, the adjustment. The adjustment is done by a simple data assimilation algorithm based on Optimum Interpolation.

First priority, based on end-user preferences, has been to optimize the O$_3$ surface concentration forecast. As the model used is mainly developed for the troposphere, and as the total column of O$_3$ is almost entirely dominated by ozone in the stratosphere, it makes no
sense to try data assimilate RS retrieved total column values of O$_3$ into the layers of the CTM. Instead RS data of NO$_2$ (from OMI) was data assimilated into the CTM. Most recent RS derived values of NO$_2$ is used to update the CTM model field, as described above, before each model run, *i.e.* at every six hours. The RS data is requested NRT, and is usually delivered 3-9 hours after a satellite overpass.

The new version of the CTM, including data assimilation, has been running parallel with the operational model without data assimilation since early June 2008 (presently for 2 months). Preliminary inspection of the resulting O$_3$ forecasts from the two versions show promising results. O$_3$ concentrations seem to be more heavily adjusted in the regions of extreme NO$_2$ concentrations in the RS data, as well as downwind.

An example of comparison of the forecast of the new RS-improved version and the currently operational version can be seen on the web www.dmu.dk/atmi/thorsda

![Sample images of the O$_3$ surface concentration over Europe](image)

Figure 1. Sample images of the O$_3$ surface concentration over Europe. Left is with the RS enhanced model version, and the right is the current operational model without data assimilation. Images are taken from the forecasts initiated at midnight on 17$^{th}$ July 2008. The figures show the situation at + 6 hours, *i.e.* later the same morning. Note that the concentrations are higher for the RS enabled version in this case, that is in the areas around Moscow, the western Alps and the central Mediterranean.

The differences between the two model versions decreases gradually with longer forecasts. The + 72 hours is the longest provided, at that point the influence of the RS DA is insignificant or zero.

Further data requirements have been prioritized. 4-D (X,Y,Z,t) data would be optimal, though 3-D (X,Y,t) total column data seems to be considerably more available. Priority will be given to collecting many data points, in time and space. This is believed to strengthen the statistical analysis of the forecast model. Secondary priority is given to collecting data sets representing high temporal data density, optimally samples every 6 hours, in 3-D, or less realistic 4-D.
Data assimilation techniques and their possibilities have been considered. There have been made experimental implementations for data assimilation of information from surface measuring station, so far focus have been on Optimal Interpolation (OI) and Kalman Filtering. The DA enabled version of the CTM uses OI.

**Scientific results and highlights**

The project in question has been defined as purely a research project, meaning that it is not part of the operational forecast produced every 6 hours. This ensures that data delivered to and used by this project are not included in the operational data base, but is kept on the research platform only. By this specification of the project we can easily meet the data confidentiality specifications in ACCENT, and hopefully then get access to more sample data.

Additionally NERI is a GMES Service Extension (GSE), and a partner in PROMOTE This opens the possibility for the project to receive data through PROMOTE for NRT processing and forecast in an operational mode.

The two data channels allow the project to operate on scientific test data from partners in scientific networks, for example, the ACCENT AT2 work groups, as well as and including NRT data from more operational network like GSE-PROMOTE. Notably the actual data provider might in some cases be the same research group.

**Future outlook**

Near future include quantification and documentation of the performance of the RS enabled version, especially compared to ground truth measurements from monitoring stations networks, with focus on surface concentrations of NO₂, O₃ and other relevant compounds.
Validation of GEM-Chemistry Modelling and Data Assimilation System: A High Resolution Study

A contribution to ACCENT-TROPOSAT-2, Task Group 2

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Summary

The Multiscale Air Quality Network (MAQNet) is funded by the Canadian Foundation for Climate and Atmospheric Science to carry out fundamental research in the area of air quality and tropospheric chemistry. The main focus of this research is the development and validation of a chemical weather and data assimilation system.

GEM-AQ (Global Environmental Multi-scale Model with Air Quality) has been exercised with a 7 year simulation (2001 to 2007) on a global uniform 1.5 × 1.5 degree resolution domain (240 × 120 grid points). The objectives of this simulation were to derive a multi-year model climatology, to examine seasonal variation and regional distribution, evaluate global emissions, and provide chemical initial and boundary conditions for high resolution model simulations. High resolution simulations were made for the July 2006 heat wave over Europe. Also, the modelling system is used to investigate the spatial and temporal distribution of hydrogen cyanide (HCN) in the upper troposphere.

Introduction

In order to develop an air quality modelling system which can accommodate various scales and processes, we used the GEM model as a computational platform and environmental processes were implemented on-line. The on-line implementation of environmental processes in the GEM model allows us to run in global uniform, global variable, and limited area configurations, allowing for multiscale chemical weather modelling. This approach provides access to all required dynamics and physics fields for chemistry at every time step. The on-line implementation of chemistry and aerosol processes allows feedback on model dynamics and physics. The use of the GEM framework permits the incorporation of chemical data assimilation techniques into the model validation and application studies in a unified fashion. The developed modelling system can be used to plan field campaigns, interpret measurements, and provide the capacity for forecasting oxidants, particulate matter and toxics. Also, it can be used to provide guidance to evaluate exposure studies for people, animals, crops and forests, and possibly for epidemiological studies.

Evaluation of the developed modelling system was done for several species and for different scales. We compared simulated ozone with ozone-sonde observations from SHADOZ as well as climatological ozone-sonde observations, GOME satellite observations and surface station data. Modelled nitrogen dioxide was compared with SCIAMACHY and aircraft campaigns such as TRACE-A observations were used to evaluate other species such as nitric acid, hydrogen peroxide and others. Modelled CO concentrations were compared with MOPITT satellite data. A full description of model simulation and results is given in Kaminski et al., [2008].
Aims and activities

The strategic objective of our project was to develop and evaluate a modelling system for tropospheric chemistry and air quality. In our design we have selected the Global Environmental Multi-scale model (GEM) as a host meteorological model for inclusion of air quality processes. The GEM model was developed at the Canadian Meteorological Centre and is used for operational weather prediction in Canada. The GEM model was augmented by implementing air quality chemistry, including the gas phase, aerosol and cloud particles, limited wet chemistry, emission, deposition and transport processes.

Results and highlights

In Figure 1 we present GEM-AQ and SCIAMACHY column data for September 2004 and January 2005 using a logarithmic scale because of the large variability of tropospheric NO\textsubscript{2}. The SCIAMACHY tropospheric column was computed by subtracting the total column over a clean reference sector in the Pacific, between 180 and 220 degree east. This column is assumed to be the stratospheric contribution only. For comparison with GEM-AQ, the SCIAMACHY data is shown on the same 1.5 × 1.5 degree grid. For the GEM-AQ results (sampled within 30 minutes of satellite overpass), the same clean reference sector method was used. The tropospheric column was also computed using the thermal tropopause and was found to be about 25 % higher than the clean sector method in relatively unpolluted regions and through the tropics. This may suggest the reference sector in GEM-AQ has an excess of tropospheric NO\textsubscript{2}, perhaps from lightning emissions.

Figure 1. September 2004 tropospheric NO\textsubscript{2} column from (a) GEM-AQ using the subtraction of the Pacific sector, and (b) SCIAMACHY. Figures (c) and (d) are for January 2005.
Over North America and Europe, where anthropogenic emissions dominate, the agreement is good for both September and January. However, column NO₂ over China is under-estimated by an order of magnitude in January 2005 and to a lesser extent in September 2004. We note that emissions over China have significantly increased since 1990 and it is not reflected in the emission data used in the present version of the model. In Africa and South America, the values are generally under-estimated. In this region, using the thermal tropopause to determine the column gives better agreement. Again, this is probably due to an excess of lightning NOₓ in the reference sector. In January, a low density plume can be seen from North America over the Atlantic by both the model and observations.

**Achievements and future outlook**

The developed modelling system has been exercised with a 7 year simulation (2001-2007) on a global uniform 1.5 × 1.5 degree grid in order to generate chemical climatology and provide chemical initial and boundary conditions for high resolution model simulations. High resolution simulations (~15 km) were made for the July 2006 heat wave over Europe [Struzewsksa and Kaminski, 2008]. Also, the modelling system is used to investigate the spatial and temporal distribution of hydrogen cyanide (HCN) in the upper troposphere. Model results are compared with HCN profiles measured by the Atmospheric Chemistry Experiment (ACE) infrared Fourier Transform Spectrometer instrument onboard the Canadian SCISAT-I satellite.

GEM-AQ will be used to study air quality in future climate in global and regional configurations. The next version of the model will have stratospheric chemistry and the top of the model will be extended to 0.1 hPa.

**References**


Synergistic Use of Satellite Data, Ground Based Observations, Back Trajectory Analysis and Global CTM Results for Studies of Tropospheric Trace Gases and Aerosols with Focus on the Mediterranean

A contribution to ACCENT-TROPOSAT-2, Task group 2

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Summary

This study is a demonstration of the synergistic use of satellite data, ground based observations, back trajectory analysis, box and global 3-D chemistry transport model results to evaluate the impact of distinct pollution sources (like urban pollution, biomass burning fires, etc.) on oxidant and aerosol in the Mediterranean, the consequences on regional climate and the seasonal and inter-annual variations. Nitrogen dioxide (NO\textsubscript{2}) and formaldehyde (HCHO) observations in spring from 1996 to 2002 enabled the characterization of the build-up of tropospheric O\textsubscript{3} and its precursors over the eastern Mediterranean. Aerosol retrievals of MERIS and SEAWIFS observed reflectances enable the evaluation of the large contribution of dust outbreaks and fire events to the AOTs over the Mediterranean. Both events have similar effects on AOT but the chemical composition of the aerosol is different since fire events are associated with high carbonaceous aerosol content.

Global source apportionment investigations were also performed to analyse glyoxal (CHOCHO) recent observations from space. The TM4-ECPL global model results were compared with the SCIAMACHY satellite observations of CHOCHO columns [Wittrock \textit{et al.}, 2006]. The global annual mean CHOCHO burden and lifetime in the model domain are estimated to be 0.02 Tg (equal to the global burden seen by SCIAMACHY over land for the year 2005) and about 3 h, respectively. However, the model did not reproduce the high CHOCHO columns observed over the tropical oceans. When accounting only for the secondary sources of CHOCHO in the model (about 56 Tg/y with 70 % from biogenic hydrocarbons oxidation), the model underestimates CHOCHO columns observed by satellites. This is attributed to an overestimate of CHOCHO sinks or a missing global source of about 20 Tg/y.

Introduction

The increasing availability of space-based remote sensing observations of tropospheric chemistry trace constituents challenges the scientific community in developing new approaches for understanding the observed changes in the atmosphere by the synergistic use of space and ground based remote sensing data, ‘traditional’ (ground-based or aircraft) observations and various types and levels of complexity numerical models of the atmosphere. In this context, the present study is an investigation of the synergistic use of satellite data,
ground based observations, back trajectory analysis, box and global 3-D chemistry transport model (CTM) results to evaluate the impact of distinct sources on atmospheric composition. Priority is given to the use of the European satellite products (like GOME (Global Ozone Monitoring Experiment) and SCIAMACHY (Scanning Imaging Absorption Spectrometer for Atmospheric CHartographY)) for trace gases and Sea-viewing Wide Field-of-view Sensor (SeaWiFS) and MEedium Resolution Imaging Spectrometer Instrument (MERIS) for aerosols. The ground-based observations consist of the gas phase, aerosol data and auxiliary measurements at Finokalia monitoring station of ECPL as well as data regarding Mediterranean area and collected during EU funded projects (ADIOS, MOZAIC (aircraft data), etc.) and of sun-photometer observations from the AERONET network. Box models that have been developed by ECPL to study Boundary Layer chemistry are applied to selected case studies to evaluate the potential of chemical built up of oxidants in the observed air masses. The global CTM TM4-ECPL is used to evaluate budgets of trace constituents and aerosols.

Outline of scientific activities

Pollution events over the eastern Mediterranean

Synergistic use of GOME, ground based and sonde observations and models – Trace gases

Nitrogen dioxide (NO₂) and formaldehyde (HCHO) observations in spring from 1996 to 2002 enabled the characterization of the build-up of tropospheric O₃ and its precursors over the East Mediterranean. These results have been presented in detail in earlier reports and published by Ladstätter-Weißenmayer et al. [2007]. They demonstrated the consistency of GOME remote sensing and ground based observations of NO₂, HCHO and O₃. The total observed variability in tropospheric O₃ has been evaluated at about 25 DU. Chemical box model calculations associated the GOME-observed NO₂ and HCHO tropospheric columns with a potential of daily photochemical enhancement in the tropospheric O₃ columns of about 0.8–1 DU over Crete and estimated the daily potential of regional photochemical buildup within upwind polluted air masses at about 2–8 DU. A Langrangian analysis attributed at most 10–20 DU of tropospheric O₃ to stratosphere – troposphere exchange. The remainder is attributed to long-range transport of O₃ from industrial regions in Central Europe.

The impact of forest fires on ozone and aerosol loading over the eastern Mediterranean.

In July 2000, large pollution events have affected the eastern Mediterranean and roughly doubled the tropospheric burden of ozone from about 25 to 50 DU. NO₂ and HCHO columns have been also increased although the impact of fires was restricted closer to the source area. GOME NO₂ observations show a significant built up of NO₂ tropospheric column over north and central Greece during the whole month of July as depicted in Figure 1 where the enhancement of NO₂ from the fires over Peloponissos is clearly shown (left panel; Dermitzaki et al, [2008]). Dilution of NO₂ high concentrations is observed after the extinction of major fires (right panel). Note the low NO₂ columns over Crete, southern Greece. O₃ columns vary between 25 for clean air masses and 53 DU for polluted situations depending on the air mass origin, the lowest values calculated for W or S air masses origins on the 16th and the 30th of July respectively. These differences are significant even when considering the large uncertainties involved in the calculations (smaller than 6 DU). Similarly high levels of NOx and aldehydes are observed downwind the forest fires over Peloponissos in August 2007 [Ladstatter-Weissenmayer et al., 2008].
Observations and CTM simulations of Aerosol Optical Thickness (AOT) over the Mediterranean during dust events

Aerosol retrievals using the BAER (Bremen Aerosol Retrieval) have been calculated and employed to investigate changes in the aerosol optical thickness (AOT) under various conditions influencing the eastern Mediterranean. The retrieved aerosol parameters indicate seasonality in the AOT over the Mediterranean with maxima AOT in the Eastern basin observed during summer as presented in earlier reports. They have been evaluated against AERONET observations and compared with TM4-ECPL global chemistry transport model results as reported in earlier years.

Figure 1. NO$_2$ columns observed over Greece by GOME – composite picture for July 13$^{th}$-16$^{th}$ (fires over Peloponissos and Samos), and for July 18$^{th}$-28$^{th}$.

Figure 2. Dust outbreak on 31$^{st}$ August 2003: (a) SeaWiFS image; (b) back trajectories arriving over Crete the same day; (c) AOTs and (d) Angstroem coefficients retrieved using the BAER algorithm.
In order to evaluate the BAER algorithm capabilities to retrieve AOT over the eastern Mediterranean during major dust events, and to evaluate the impact of these events on the levels of AOT, the dust events that affected the eastern Mediterranean during summer 2003 and during the whole year of 2002 have been studied.

Remarkably, BAER AOT retrievals from SeaWiFS observations (443 nm) are in excellent agreement with the FORTH_CRET AERONET (440 nm) station observations during August and September 2003. The scene of the 31st of August 2003 has been further used to study the impact of dust outbreaks from Africa to AOT as retrieved by BAER from the SeaWiFS observations over the eastern Mediterranean (Figure 2). No MERIS data was available for this day.

Source apportionment – glyoxal global distribution
Observations and CTM simulations of glyoxal.

Figure 3. Annual mean CHOCHO columns as computed with the TM4 model for the year 2005 (a) when accounting only the secondary sources of CHOCHO, (b) when accounting only a primary combustion source of CHOCHO of 7Tg/y, (c) Comparison of annual mean glyoxal columns from TM4-ECPL simulations and SCIAMACHY data product (in units of molecules/cm$^2$). The points show the annual mean simulated columns for the year 2005 binned every 0.25 $10^{14}$ molecules/cm$^2$ of the corresponding SCIAMACHY observations, together with their standard deviations. Dark blue circles and light blue are the binned data when both land and oceanic grids are considered for the TM4-ECPL high and low resolution simulations, respectively. Solid red and black squares are the corresponding binned data when neglecting the grid cells over the oceans. (d) Comparison of annual mean glyoxal columns from TM4-ECPL simulations and SCIAMACHY data products (in units of molecules/cm$^2$). Binned data over the continents; red triangles are for simulation considering both primary and secondary CHOCHO sources, solid gray squares for simulation accounting for all secondary sources, and solid black circles for simulation neglecting the anthropogenic secondary source. Linear fit regression lines are also plotted with the corresponding color (figures adopted from Myriokefalitakis et al., [2008], where more details are given).
In the TM4 chemical scheme, glyoxal is produced (56 Tg/y) mainly from biogenic hydrocarbons (70 %) but also from acetylene (17 %) and anthropogenic aromatic hydrocarbons (11 %). TM4 is able to simulate the global concentrations of glyoxal as observed by the SCHIAMACHY sensor [Wittrock et al., 2006]. The most recent TM4 glyoxal simulations with the updated chemical scheme with regard to glyoxal production based on Volkamer et al. [2005] kinetic experiments reproduce the observed tropical hot spots as well as the smaller enhancement of glyoxal over polluted areas in the northern hemisphere. The contribution of aromatics and acetylene to glyoxal (CHOCHO) column is limited to 3-20 % over the tropics and maximizes over the urban areas in the northern hemisphere where it reaches 20-70 %.

Comparison between model results and satellite column retrievals indicates a potentially missing source of ~20 Tg/y for glyoxal lifetime of about 3h. The agreement is better when data over land are only taken into account (Figure 3c). Consideration of a primary – combustion glyoxal source of 7 Tg/y leads to an overestimate over hot spot land areas, although on a global scale a dissent agreement between model results and observations over land is derived (Figure 3d).

Future outlook

Our study has demonstrated the large potential of synergistic use of satellite observations with box, 3-d chemistry-transport and trajectory models as well as ground-based observations, for the understanding of tropospheric processes and oxidant and aerosol build up in the troposphere.

Further work will focus on the evaluation of the contribution of sources, transport and chemistry to the observed inter-annual variability of oxidants and aerosols over the Mediterranean. Particular effort will be put on evaluating the contribution of aqueous phase chemistry and ocean emissions to the levels of glyoxal observed over the tropical ocean.

References


Global Measurements of Water Vapour in the Tropopause Region and Upper Troposphere with MIPAS/ENVISAT

A contribution to ACCENT-TROPOSAT-2, Task Group 2

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Summary

At IMK water vapour profiles are retrieved from MIPAS/Envisat infrared spectra using a non-operational science-oriented retrieval processor. For the period 18th September 2002 to 25th March 2004 MIPAS was working in its initially planned mode measuring spectrally high resolved atmospheric emission spectra in the infrared. Since January 2005 MIPAS has been operated with reduced spectral resolution. At IMK a subset of available Level 1b spectra has been processed and global distributions of water vapour have been derived. Individual measurements and zonally and globally averaged water vapour originating from these dates have been analyzed. The temporal evolution of water vapour distributions in different altitudes and regions has been observed. Among others, the tropical tape recorder has been observed in the MIPAS data, providing vertical transport velocities in agreement with earlier observations. Individual measurements obtained during various measurement campaigns like TROCCINOX or SCOUT-O₃ were used to compare to various instruments and to support the scientific questions behind the campaigns. Furthermore, the data have been validated against other measurements.

Introduction

The Michelson Interferometer for Passive Atmospheric Sounding (MIPAS) onboard the sun-synchronous orbiting ENVISAT is operating in the mid-infrared spectral region with limb geometry. The scientific retrieval processor for MIPAS/ENVISAT data, developed at IMK, is used to obtain, besides other parameters, vertical profiles of water vapour. Between 18th September 2002 and 25th March 2004 MIPAS was operating in the so-called full resolution mode (FR) (nominal spectral resolution: 0.025 cm⁻¹). For technical reasons, MIPAS has been operating with reduced spectral resolution of 0.625 cm⁻¹ (RR mode) since January 2005. A subset of days of these periods have been processed and analyzed with respect to different scientific aspects.

Scientific results and highlights

Averaged distributions and temporal behaviour of water vapour

The retrieved water vapour profiles of all processed data were averaged for individual days and for each month and analyzed. In general, the zonally averaged water vapour for different seasons shows distributions as expected from climatologies. In the troposphere below the tropopause the volume mixing ratios (VMR) increase nearly exponentially with decreasing altitude. At or slightly above the tropopause the water vapour distributions show a minimum, the so-called hygropause, and then increases with altitude in the stratosphere. Although the general structure is similar for all seasons, the individual months show distinct differences resulting from atmospheric conditions related to seasonal variations.
In particular in the tropics, the water vapour minimum shows a clear annual cycle with the lowest VMR occurring during northern hemispheric (NH) winter, when comparatively cold tropopause temperatures result in lower VMR values than in NH summer.

**Tropical Tape-recorder**

The time series of daily zonal means for tropical latitudes reveals the so-called tape recorder effect in tropical stratospheric water vapour (see Figure 1). The water vapour content of the rising air is determined by the seasonal cycle of the temperature in the tropopause region. During the year the dry and wet air, respectively, slowly rises in the stratosphere. MIPAS measurements cover the period from 2002 to 2008. The estimated ascent rates of the air masses obtained from MIPAS measurements are in good agreement with earlier observations as published in the literature.

![Figure 1. Time series of zonal mean water vapour for the latitude range 10 °S to 10 °N for the period September 2002 to January 2008. The white gap marks the period when MIPAS was out of service.](image)

**Dehydration of the polar vortex**

The water vapour distribution at polar regions shows a distinct seasonal cycle, dominated by the Antarctic winter, when the large scale distribution of PSCs over Antarctica leads to dehydration of the air masses as a consequence of the sedimentation of ice particles. The measurements by MIPAS caught the unusual sudden stratospheric warming and subsequent vortex split in September/October 2002 which lead to a comparatively weak dehydration and early recovery of the Antarctic stratospheric water vapour compared to the next years when the remnants of the dehydration were visible until late November.

During northern hemisphere winter a large scale dehydration of the stratosphere is not visible. The Arctic polar vortex is quite inhomogeneous compared to Antarctic conditions and regions with temperatures enabling the formation of PSC particles, for example, due to orographically induced waves, are restricted to small areas. Here the subsidence of comparatively wet air from higher altitudes leads to an increase of water vapour vmrs in the lower stratosphere during the northern hemisphere winter (see Figure 2).
STE processes in the Indian Monsoon

During the summer months June-July-August MIPAS measurements reveal a region over South Asia and the Himalaya with humid air masses reaching further up into the stratosphere than in the tropics. Here the Indian Monsoon circulation leads to enhanced water vapour amounts up to the lower stratosphere [Milz et al., 2005]. The involved transport processes have been further analyzed using correlations with tropospheric tracers such as C$_2$H$_2$, CO, HCN which give clear indication of the region of upward transport. While enhanced water vapour vmrs near the tropopause are found during June, July, and August, the Asian monsoon anticyclone tropopause region becomes very dry in September, although the upward transport still persist [Stiller et al., in preparation].

![Timeseries, H2O 20 km](image)

Figure 2. Time series of zonal mean water vapour at 20 km retrieval altitude for the period September 2002 to March 2004.

Inter-annual variability of zonal water vapour at 20 km

In Figure 2 the temporal development of daily zonal averaged water vapour at 20 km is shown for the period September 2002 to March 2004. Additionally to the above mentioned seasonal variations in the tropics and the polar regions there are distinct differences between the different years. Over Antarctica the vortex split up in 2002 is accompanied by a sudden increase in humidity in October which is much slower in 2003. In the tropics the lower stratosphere in the northern hemisphere autumn 2002 is much drier than in autumn 2003. A similar distribution can be seen in the time series of MIPAS temperature measurements at the same altitude which shows colder condition for 2002 than for the following year. This is in agreement with analyses from NOAA Climate Prediction Centre. The differences in lower stratospheric temperature and the observed water vapour amount can partly be explained by the influence of the QBO on the tropopause temperature. During summer/autumn 2002 the QBO showed an easterly phase which leads to a cooling of the tropopause.

Validation against other satellite measurements

During the SCOUT-O$_3$ and TWP-ICE measurement campaigns in Darwin, Australia in November and December 2005 MIPAS was operated in the reduced resolution mode. Among other parameters, temperature and water vapour distributions have been derived from the MIPAS reduced resolution data, and relative humidity over ice (RHi) has been calculated. Comparison to measurements of the Microwave Limb Sounder (MLS) on board NASA’s EOS Aura reveal lower relative humidities in MIPAS data than in MLS, which, however,
could be traced back to some problems in MLS temperature measurements [Chauhan et al., in preparation].

MIPAS water vapour profiles were compared to collocated measurements of the Japanese instrument ILAS-II which was operated from May to October 2003. ILAS was a limb sounding FTIR instrument operated in solar occultation mode. For ILAS measurements taken in the northern hemisphere, the agreement is good for altitudes up to 40 km [Griesfeller et al., 2008].

For winter 2004, just before MIPAS FR mode was discontinued, several collocated measurements with the Canadian ACE-FTS were used for validation. ACE-FTS is, similar to ILAS-II, a limb sounding FTIR operating in solar occultation. The water vapour profiles agree very well, with ACE being slightly wetter by 3 % [Carleer et al., 2008].

For the microwave instrument ODIN relative humidity was compared to MIPAS. Both RHi products show good agreement for cloud free cases [Ekström et al., 2008].

Comparisons with collocated HALOE measurements reveal a slight wet bias of 10 % of MIPAS. Taking into account that HALOE is known to have a slight dry bias, the wet bias of MIPAS is acceptable.

Comparison with the VIS/NIR instrument POAM show good agreement, considering the known wet bias of POAM water vapour profiles.

**Validation against in situ and LIDAR measurements**

During the TROCCINOX campaigns in South America in 2004 and 2005 MIPAS was operated both in FR and RR mode, respectively. During both campaigns the DLR DIAL on board the Falcon was operated. MIPAS measurements were compared with DIAL measurements in the tropical and subtropical UT/LS. The mean deviations are small with MIPAS being 8 % drier than DIAL, however, the standard deviation of the comparison ensemble is quite large (48 %) due to heterogeneous atmospheric conditions and sub-optimal co-location of the measurements. No differences between the FR and the RR mode data have been found which confirms continuation of high-quality water vapour measurements by MIPAS in the RR mode [Kiemle et al., 2008].

During the operation of MIPAS in FR mode several measurements with in situ instruments have been made, which could be used for comparison with collocated MIPAS measurements.

The FISH instruments on board of the stratospheric research aircraft Geophysica was operated in several campaigns. Comparison of profiles recorded during the aircraft’s descend have been compared to MIPAS. The agreement between the measurements is good, if the lower spatial resolution of MIPAS is taken into account for the comparisons.

During the LAUTLOS campaign in Sodankylä, Finland, several instruments such as the Russian FLASH-B and the NOAA Frostpoint hygrometer have been used to validate different radio sonde sensors. Comparison with profiles obtained by these instruments show good agreement with MIPAS measurements in the UT/LS region.
References


The Use of Satellite Based Measurements of AOD for Air Quality at Ground Level

A contribution to ACCENT-TROPOSAT-2 Task Group 2

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Summary

Throughout the duration of the ACCENT-TROPOSAT-2 project our activities were focused towards the improvement of analysed PM$_{2.5}$ fields at ground level, using satellite measurements of AOD. On the one hand this is done by using data assimilation and the LOTOS-EUROS chemistry transport model while on the other hand we have looked at the relationship between total AOD and PM$_{2.5}$ at the site of Cabauw.

Introduction

In Europe, particulate matter (expressed as aerosol mass concentrations at the surface, PM) is the most important air pollutant responsible for loss of human health. To protect against these negative health effects, the EU has set limit values for PM$_{10}$ and more recently also new limit values for PM$_{2.5}$ are being set by the European Commission, to be attained by 2010. Besides the limit values, the member state have an obligation to inform the public on the air quality situation. Further, many countries provide forecast of air pollution levels for the next day(s). Hence, as identified within for example the ESA-project PROMOTE, there is a need for accurate near real time assessments of air quality and forecasts.

In contrast to ground based monitoring sites, satellite measurements of aerosol optical depth (AOD) provide full spatial coverage (although only during daylight and in cloud-free conditions) and – in principle – consistent data for the whole European region. This suggests that satellite measurements may be useful to improve the insight in PM distributions in Europe in combination with models and ground based measurements.

Aims and activities

The principle aim of our work is to improve analysed PM$_{2.5}$ fields at ground level, making use of all available sources: a chemistry transport model, ground-based measurements and satellite measurements. The combination of these sources can be achieved by using data assimilation. Data assimilation defines a new atmospheric state by combining the observed and modelled state in an intelligent and statistically sound way.

In our work we used an ensemble Kalman filter to assimilate MODIS AOD retrievals for the year 2003, within the LOTOS-EUROS chemistry transport model [Schaap \textit{et al.}, 2008] over...
Europe. We have used random noise to the emissions of NO$_x$, SO$_x$, VOC, NH$_3$ and particles to define the ensemble and therewith the model uncertainty. Prior to the assimilation the quality and suitability of the MODIS and AATSR AOD measurements for assimilation in the LOTOS-EUROS model were tested by comparing them with ground-based measurements from the AERONET network.

The same data assimilation technique has been used to perform an Observing System Simulation Experiment (OSSE). The aim of the OSSE was to assess the added value of future satellite measurements of AOD on the analysis and forecast of PM$_{2.5}$ concentrations over Europe. The first step in the OSSE is the performance of a nature run with another model (in our case the CHIMERE model) to simulate the ‘true’ state of the atmosphere. From this nature run synthetic ground-based PM$_{2.5}$ measurements and satellite based AOD measurements are retrieved that correspond to the instrument characteristics of future satellite instruments [Siddans et al., 2007]. These are then assimilated in the LOTOS-EUROS model to investigate their added value.

The relation between Aerosol Optical Depth (AOD) and PM$_{2.5}$ was addressed using PM$_{2.5}$ from the artefact free FDMS technique at the Dutch rural site of Cabauw. At this site long term time series of AOD and a range of optical and physical aerosol parameters are obtained. Furthermore, the vertical aerosol distribution is monitored using a LIDAR. The combination of the instrumentation at Cabauw provides a unique opportunity to study the AOD-PM$_{2.5}$ relationship in the Netherlands. The study is performed together with RIVM, MNP and KNMI.

**Results and highlights**

After evaluation of MODIS collection 4 it was decided to assimilate MODIS fine mode AOD. The data were corrected for the 10% bias found when comparing to the AERONET data.

Assimilation of MODIS AOD in the LOTOS-EUROS model improves the modelled AOD in absolute sense but especially in timing. The correlation between modelled AOD and AERONET at almost all stations increases considerably after assimilation. At the boundaries of the model domain the assimilation is not effective due to the fixed boundary conditions in the model. Through the assimilation the monthly mean PM$_{2.5}$ levels are increased by about 2-3 µg (up to 50 %) in central Europe (see Figure 1).

![Figure 1. PM$_{2.5}$ fields before and after assimilation of MODIS AOD fine values.](pm25.png)

From a scientific point of view a successful application is still hampered by a number of unresolved issues. The model (without assimilation) underestimates AOD systematically. SO$_4$, NO$_3$ and NH$_4$ concentrations are probably not the cause for the underestimation of AOD.
Reliable parameterizations for emissions of fugitive dust and SOA formation do not exist and are therefore not incorporated in the model. Furthermore, primary emissions are uncertain and a previous study indicated that the concentrations of EC and OC are underestimated by a factor 2 [Schaap et al., 2004]. The AOD calculation from the separate aerosol components was also identified to be a major source of uncertainties due to the assumptions on optical properties, water uptake and (BC) mixing state. Lastly, the retrieved AOD values may be biased due to undetected glint and cloud contamination.

Figure 2 shows the added value of assimilation of half hourly satellite total AOD measurements for an Imager type instrument on analysed fields of PM$_{2.5}$ for a period with high aerosol levels over central Europe. In this situation the assimilation of total AOD measurements has a clear additional impact compared to the assimilation of hourly synthetic ground based PM$_{2.5}$ measurements only (at the locations indicated by the black squares).

![Figure 2](image_url)

**Figure 2.** Mean PM$_{2.5}$ concentrations at ground level for 25$^{th}$-28$^{th}$ February from the nature run ‘true state’ (top left), the LOTOS-EUROS model without assimilation (top right), the LOTOS-EUROS model with assimilation of synthetic ground-based PM$_{2.5}$ measurements at locations indicated by the black squares (bottom left), and the LOTOS-EUROS model with assimilation of synthetic ground-based PM$_{2.5}$ measurements and half hourly synthetic total AOD measurements (bottom right).
The impact of the AOD measurements on the analysis and forecast of PM$_{2.5}$ concentrations is dependent on the situation and on the errors provided with the AOD measurements. The OSSE experiment needs some further development to evaluate the impact of differently resolved AOD measurements (temporal, horizontal and vertical resolution).

Figure 3 shows the time series of AOD and PM$_{2.5}$ for spring 2007. The correlation of AOD with ground level PM$_{2.5}$ is very good for this period which is characterised by very nice weather, a well mixed mixing layer and no clouds. In principle, AOD data can only be obtained during clear sky conditions, which are biased towards conditions of stagnant flow and high aerosol concentrations. For such conditions, the AOD correlates reasonably well with PM$_{2.5}$ ($R^2 = 0.6$), suggesting that temporal variations of AOD can be used as a proxy for temporal variations in PM$_{2.5}$. The AOD-PM$_{2.5}$ relationship constructed from the AERONET and PM$_{2.5}$ data at Cabauw is used to produce satellite (AATSR and MODIS) derived PM$_{2.5}$ maps over the Netherlands. The mean values in the maps are similar to observed ones at Cabauw but features in the spatial distributions are distinctly different illustrating that the interpretation of these AOD maps in terms of PM is hampered. This is because spatially dependent systematic errors in AOD can be of the same order of magnitude as the real spatial variations in AOD.

![Figure 3. The time series of all AOD data points and PM$_{2.5}$ at Cabauw, the Netherlands](image)

**Achievements and future outlook**

Through AT-2 we gained knowledge on available satellite products and AT-2 allowed getting acquainted with groups working on these data.

The performed studies all indicate that satellite measurements of AOD can have added-value, particularly regarding the temporal variation of PM, when combined with atmospheric transport models and surface measurements of PM$_{2.5}$.

The model and assimilation system are used in European projects (PROMOTE, MACC) focused on producing ensemble forecasts on air quality. In future the assimilation system will be further developed using also newly available satellite aerosol data to produce improved analysed and forecasted maps of PM$_{2.5}$ over Europe. The OSSE method will be further developed to evaluate the impact of differently resolved measurements and its application to profile data.
References


Brief analysis of Precursor Emissions, Satellite Observations and Modelling of Formaldehyde Column over Europe: Perspectives for Constraints on Precursor Emissions

A Contribution to ACCENT-TROPOSAT-2, Task Group 2

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Summary

The aim of this contribution is to synthesize the current state of knowledge about European formaldehyde (HCHO) column as seen from space and as simulated with a chemistry-transport model. This constitutes the basis for potential exploitation of satellite observations of HCHO as constraint on European volatile organic compound emissions of both anthropogenic (AVOC) and biogenic (BVOC) origin.

First of all, we pose the following question: how we expect the HCHO column over Europe to be?

We will try to answer this question looking at knowledge about European land use, temperature anomalies, and fire counts.

We show land use over Europe extracted from the Global Land Cover Facility (GLCF) database¹. The land cover classification into 14 categories (see Figure caption) is derived from AVHRR imagery collected between 1981 and 1994 at 1 km spatial resolution [Hansen et al., 2000] A great fraction of land in western, central and eastern Europe is dedicated to agriculture. Scandinavia is mostly covered with evergreen forest and wooded grassland. Northern Russia is also a mix of forests. Large forested areas are also found in northern Spain, northern France, the Alps and the Apennines, the Balkans and at the eastern border of the Black Sea. Spain has an intricate mix of shrub, grass and crop land. Turkey has a great fraction of grassland.

These broad features are confirmed by the U.S. Geological Survey database² (USGS). We show European land use also derived from AVHRR imagery but with USGS Land Use/Land Cover System [Anderson et al., 1976]. The 24 original categories were merged into GLCF categories for the sake of comparison. Although the main features of European land use are consistent with GLCF database, the classification at some specific regions may be quite different (e.g. Great Britain).

We show the GLCF land-use over the United States of America. This is a useful term of comparison since satellite observations of HCHO have been used there for the first time as a constraint on isoprene biogenic emissions [Palmer et al., 2003], revealing a persistent hot spot above the South-Eastern US. We see that the region is mostly covered by forests.

¹ The University of Maryland Department of Geography generated this global land cover classification collection in 1998. http://www.landcover.org/data/landcover/
² http://edcsns17.cr.usgs.gov/glcc/globdoc1_2.html
In order to have an idea of biogenic VOC production over Europe we can cross land use information with typical emissions factors of isoprene and α-pinene (a major monoterpene emitted by vegetation) given in Table 1. Broadleaf trees and shrubs are strong isoprene emitters. Needle leaf trees are strongest α-pinene emitters, with broadleaf trees and shrubs having about half of their emission potential. Grasslands and crops are expected to have minor emissions. We might then expect high isoprene emissions above light green, orange and avana regions in southern Europe (Spain, southern France, Italy, southern Germany, the Balkans). High α-pinene emissions are expected around the Mediterranean basin.

Figure 1. European Land Use according to Global Land Cover Facility (GLCF) database.

Figure 2. European Land Use according to US Geological Survey (USGS) database. USGS Land Use/Land Cover System (Anderson et al., 1976) modified level 2. Original 24 categories were merged to 14 GLCF categories for ease of comparison.

Figure 3. United States Land Use according to GLCF database.
Table 1. Typical emission factors for isoprene and α-pinene at standard conditions as a function of emitter type. Data from the MEGAN model [Guenther et al., 2006]

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<thead>
<tr>
<th>Plant Functional Type (PFT)</th>
<th>Isoprene (µg/m²/h)</th>
<th>α-pinene (µg/m³/h)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Broadleaf Tree</td>
<td>13000</td>
<td>180</td>
</tr>
<tr>
<td>Needleleaf Tree</td>
<td>2000</td>
<td>450</td>
</tr>
<tr>
<td>Shrubland</td>
<td>11000</td>
<td>200</td>
</tr>
<tr>
<td>Grassland and crops</td>
<td>400</td>
<td>2</td>
</tr>
</tbody>
</table>

The qualitative analysis based only on land use and typical emission factors is confirmed by recent detailed calculations reported by Steinbrecher et al. [2008] for year 2000. Higher isoprene emissions are found over Iberian Peninsula, France, Italy Balkans and Greece. Monoterpene emissions are particularly elevated over Iberian Peninsula. Boreal forests in Russia are also high BVOC emitters. We point out that Steinbrecher et al. [2008] also calculated that 40% of BVOC emissions in Europe take place in July, with an approximate equal share among isoprene, monoterpenes and oxygenated VOCs. In June and August BVOC emissions are generally 35% lower than in July.

Solberg et al. [2001] reported a detailed analysis and comparison with model simulations of EMEP HCHO measurements for years 1993-1996. Formaldehyde was found to display a broad summer maximum at all selected stations and model generally underestimated HCHO levels. An analysis of sources and sinks of HCHO at the stations revealed that HCHO production is largely controlled by VOC oxidation, with isoprene being the most significant contributor. Terpenes were not included in the analysis.

In Figure 4 we show the monthly mean HCHO column observed by OMI³ (onboard NASA’s Aura satellite) over Europe in summer 2005. Isoprene is likely to play a key role on HCHO column variability in summer over Europe. Isoprene emissions are related to temperature in an exponential fashion. Also photochemical activity is strictly related to temperature. We thus include in our analysis maps of temperature anomalies in Figure 4. The source of the monthly gridded fields of temperature anomalies is the Climatic Research Unit⁴ in UK. Temperature anomalies are calculated with respect to the 1961-1990 period. Although it might be expected that temperatures in 21st century display always a positive anomaly with respect to that period because of global warming, the picture coming out is still useful to evaluate inter-annual and inter-monthly variability of temperature. In Figure 4 we can see that June 2005 was much warmer than usual with respect to July and August 2005, especially in south-western Europe.

Also fires are known to be a source of formaldehyde and other VOCs [Andreae and Merlet, 2001]. We calculated total number of fire counts on a 0.5° × 0.5° grid from the ATSR World Fire Atlas⁵. In Figure 4 we display results for June-August 2005. A large number of fires, potentially affecting HCHO column variability, have been observed only over northern Portugal during this period. A minor effect is also expected in the South of Italy.

³ OMI Level 2 HCHO Total Column products were downloaded from http://daac.gsfc.nasa.gov/data/dapool/OMI/Level2_V003/OMHCHO/ server. Data are provided in HDF5-EOS format and were processed with custom software for use in this work.
⁴ CRU, http://www.cru.uea.ac.uk/cru/data/temperature/. The variance adjusted combined land-ocean temperature anomalies are displayed (product HadCRUT3v).
⁵ http://dup.esrin.esa.int/ionia/wfa/index.asp
Figure 4: Summary of selected factors driving formaldehyde column over Europe. First column from the left: monthly average HCHO column observed by OMI in summer 2005 (June to August from first to third row) remapped on a 0.5° × 0.5° grid. Second column: number of fires per month detected by ATSR on same grid as first column. Third column: monthly temperature anomaly calculated by CRU on a 5° × 5° grid. Fourth column: total isoprene emissions calculated with MEGAN on the same grid as the first column.

In Figure 5 we show the monthly mean HCHO column observed by OMI over Europe in July 2005. Data are averaged over a 0.5° × 0.5° regular grid. A cloud fraction filter with a 40% threshold was applied. A few interesting features can be noted. Enhanced HCHO values (> 8 × 10^{15} molecules/cm^2) are observed above the heavy industrialized and populated northern Italy, Ile de France (Paris area) and western Germany, and over the relatively less polluted south-western Iberian Peninsula and south-western France. Very high (>12 × 10^{15} molecules/cm^2) are found downwind of the Marseille area (southern France on the Mediterranean Sea). Elevated HCHO column (6-12 × 10^{15} molecules/cm^2) are observed over the Mediterranean Sea, in contrast to the North Atlantic (< 8 × 10^{15} molecules/cm^2).

In Figure 6 we show monthly mean HCHO column simulated with the regional chemistry-transport model Chimere\(^6\) over the same grid and period as satellite data of Figure 6. Model values are sampled at OMI overpass time (13UTC), but an exact correspondence with OMI observations were not applied (e.g. cloudy scenes screening). Model column appears to be well spatially correlated with OMI. We can notice correspondences of HCHO enhancements in the south western Iberian Peninsula, East coast of Spain, south eastern France, Marseille

\(^6\) http://www.lmd.polytechnique.fr/chimere/. Here driven with MM5 meteorological model (http://www.mmm.ucar.edu/mm5/). Chimere is run with 20 vertical \(\sigma\)-levels from surface to 200 hPa. Boundary conditions are provided by monthly climatology of global model LMDz-INCA.
area, Italy (particularly the Po Valley) and the Balkans. The feature including Eastern France, Benelux and Western Germany is more smeared in the model then observations. However the model column is lower than OMI by $2-4 \times 10^{15}$ molecules/cm$^2$ (which is about 20-40% on the mentioned regions), and it does not predict elevated values on the Mediterranean Sea.

![Figure 5. Monthly average HCHO column observed by OMI in July 2005](image1)

![Figure 6. Monthly average HCHO column simulated at 13UTC by Chimere model.](image2)

Modelled HCHO column is largely underestimated and this makes the inversion procedure difficult: next steps will be (i) validation of HCHO simulations at ground against EMEP data, (ii) comparison of OMI with output from another model (GEOS-Chem).

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References


Overview and Preliminary Analysis of NO$_2$, Total Peroxy Nitrates ($\Sigma$PN$_s$) and Total Alkyl Nitrates ($\Sigma$AN$_s$) measured in the Borneo Forest, Malaysia

A contribution to ACCENT TROPOSAT2: Task Group 2

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Summary

Here we report the preliminary data analysis of the measurements carried out during summer 2008 in the Borneo Forest in Malaysia. In that campaign a state of the art instrument, developed by the CETEMPS has been used. The system has been used for direct observations of NO$_2$ using the Laser Induced Fluorescence (LIF) and for the observation of total peroxy nitrates ($\Sigma$PN$_s$) and total alkyl nitrates ($\Sigma$AN$_s$) using the Thermal Dissociation Laser Induced Fluorescence (TD-LIF). NO$_2$ molecules are excited with a laser at 532 nm and the fluorescence signal is measured for $\lambda > 620$ nm, this instrument has a detection limit of 3.6 pptv and allows observations to be made at 10 Hz, more details on the instrument could be found in Dari-Salisburgo et al. [2008]. The fast observations could be used to measure the flux of NOx using the eddy covariance method.

During the campaign in the Borneo forest the CETEMPS LIF measured NO$_2$, $\Sigma$PN$_s$ and $\Sigma$AN$_s$ at 8 m and NO$_2$ at 75m on the tower for the flux estimation. An overview of all the data collected is shown in Figure 1.

Figure 1. Time series of all the data collected during the campaign in the Borneo forest.
The concentrations of NO\textsubscript{2} were below 0.5 ppbv for almost all the campaign, only during 2 days the NO\textsubscript{2} levels were above 1 ppbv. These observations show that the site is very clean and not impacted from anthropogenic emissions; the level of the nitrate species confirm this hypothesis.

The diurnal cycle of NO\textsubscript{2} (Figure 2) at 8 m does not show any signature of the morning and evening peaks due to anthropogenic emission and the concentrations during daytime are lower than during night-time. At 75 m, daytime NO\textsubscript{2} concentrations are higher than during night-time, levels influenced more by transport than at 8 m.

Figure 2. Diurnal cycle of NO\textsubscript{2} observed at 8 m and at 75 m.

The diurnal cycle of \(\Sigma\text{PN}_s\) shows an increase after midday when the concentration of OH is high and the oxidation of VOC is more efficient, whereas \(\Sigma\text{AN}_s\) does not show a well definite diurnal cycle.

Figure 3. Diurnal cycle of total peroxy nitrates and total alkyl nitrates observed at 8 m.
The first analysis done is the study of the photochemical steady state. The Leighton relationship (Leighton, 1961) defines the photochemical steady state (PSS) as:

$$\Phi = \frac{J_{NO_2}}{k_{O_3NO} [NO][O_3]}$$  \hspace{1cm} (1)$$

where $J_{NO_2}$ is the photolysis rate of NO$_2$ (s$^{-1}$), $k_{O_3NO}$ is the reaction rate between ozone and NO and [O$_3$] (ppbv s$^{-1}$), and the bracket notation represents concentration or mixing ratio, if $\Phi$ is equal to 1 means that the PSS is valid.

Under PSS condition ozone is controlled mainly by the ratio of concentrations of NO$_2$ and NO:

$$[O_3] = \frac{J_{NO_2} [NO_2]}{k_{O_3NO} [NO]}$$  \hspace{1cm} (2)$$

In the upper panel of Figure 4 shows the calculated $\Phi$ parameter using the measured concentration of NO, NO$_2$, O$_3$ and the photolysis coefficient of NO$_2$. The steady state approximation is valid only for small periods of the day and for them the measured NO$_2$ is compared with calculated NO$_2$ using the relation (1). Under steady state approximation the concentration of O$_3$ is also calculated (Figure 5) and, as for NO$_2$, the agreement with measured O$_3$ is good.

![Figure 4](image-url)  

Figure 4. In the upper panel shows the steady state parameter $\Phi$ and in the lower panel the calculated NO$_2$ versus the measured NO$_2$. 

Figure 5. Calculated $O_3$ versus measured $O_3$, when the steady state approximation is valid.

More analyses are ongoing with special attention at the quantification of the NOy budget using total peroxy nitrates and total alkyl nitrates observations. These analyses will help to understand the role of the vegetation emissions on the NOx budget in an environment not impacted by anthropogenic activities.

References
