The Remote Sensing of Atmospheric Constituents from Space

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

Combining satellite observations to study atmospheric composition

Report from the Fifth AT2 Workshop

held at the Department of Chemistry, University of Crete,
Voutes, Heraklion Crete,
on
Monday 3rd and Tuesday 4th July 2006

Peter Borrell
Deputy Coordinator
September 2006

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Comparison between AATSR and MODIS AOD and assimilation in a regional chemistry transport model. Renske Timmermans, Martijn Schaap, Robert Koelemeijer, Robin Schoemaker, Gerrit de Leeuw, and Peter Builites.


NO2 and aerosol observations during the DANDELIONS campaign. Ellen Brinksma, Pepijn Vreeking, Pieter Pelt, Thomas Wagner, Ossama Ibrahim, Andreas Richter, Folkard Wittrock, Hilke Oetjen, Michel van Roozendael, Caroline Fayt, Gaia Pinardi, Christian Hermans, Daan Swart, Hester Volten, Gerrit de Leeuw and Lyana Curier.

Quantitative analysis of SCIAMACHY CO and comparison with model and MOPITT. Miranda van den Broek.


Long range transport of pollution from space: a multiplatform (MOPITT, TES, ACE) analysis of CO observations. Solène Turquety.


7. CO Workshop. Miranda van den Broek.

8. Appendices. A. AT2 5th Workshop Participants. B. AT2 5th Workshop programme.
1. Workshop Plenary Session and Programme

1.1 Workshop opening. The Coordinator, John Burrows, opened the workshop at 09.00 on Monday 3rd of July 2006, by welcoming the 42 principal investigators, co-workers and guests present (Appendix A). He emphasised:

- the need for AT2 to make the best use of the satellite data to produce valid observations;
- the role that PIs should play in the three task groups;
- the need to look towards the final objectives;
- and the need, if there is to be a future project, to demonstrate the added value that ACCENT has given to the work and activities in our field.

1.2 AT2 News and feedback session. During the meeting, the Deputy Coordinator, Peter Borrell, conducted a news and feedback session to alert PIs to the latest developments and to receive comments and feedback on the programme, for the steering committee. A detailed report is given in section 2.

1.3 Invited Speakers. The following colleagues were invited to give talks on results and developments of current interest. Illustrated abstracts of these are given in section 3.

- Ulrich Platt, Uni-Heidelberg, D - Atmospheric trace substances emitted by volcanoes
- Matthias Beekmann, LISA, Uni Paris-XII, F - Estimation of emissions with satellite data.
- Gareth Thomas, Uni-Oxford, UK - GLOBAEROSOL: a multi-instrument, long term aerosol dataset from European satellites

1.4 Retrievals and Results; Task Group 1. The first scientific session was devoted to selected presentations from Task Group 1. The emphasis was on recent scientific results obtained, but an account was also given by Thomas Wagner of the recent progress with the difficult problem of estimating radiative transfer to correct retrievals, a task being undertaken by a sub-group of task group 1. Illustrated abstracts of these talks are given in section 4.

- Phillipe Ricaud, Lab. d’Aérologie, Toulouse, F - Impact of convective outflow and biomass burning into the tropical lower stratosphere as diagnosed from satellite observations and model results
- Catherine Wespes, Uni-Libre, Brussels, B - Tropospheric studies using infrared spectroscopic measurements from space
- J.-M. Flaud, B. Picquet-Varrault, A. Gratien, J. Orphal and J.-F. Doussin, LISA, Créteil, F - What about the consistency of spectral parameters when using different atmospheric instruments?
- Nicolas Theys, BIRA–IASB, Brussels, B - Total and tropospheric BrO retrieval from space and ground-based UV-VIS observations
- Thomas Wagner, Uni-Heidelberg, D - Recent Progress in the Radiative Transfer Correction Group
- Andreas Richter, IUP, Bremen, D - Glyoxal measurements with SCIAMACHY - a new tropospheric species from satellite measurements
- Thierry Marbach, Heidelberg, D - Highlights of the Third International DOAS Workshop

1.5 Modelling and observations; Task group 2. The second scientific session was devoted to selected presentations from Task Group 2. The four presentations illustrated the use of satellite data in the validation and evaluation of models and pointed in some
cases to shortcomings in both the models and the observational data. Illustrated abstracts of these talks are given in section 5.

Martin Dameris, DLR, Oberpfaffenhofen, D
Validation of tropospheric ozone values derived from the chemistry-climate model E39/C

Renske Timmermans, TNO, Apeldoorn, NL
Comparison between AATSR and MODIS AOD observations and assimilation in a chemistry transport model

Maria Sfakianaki, ECPL, Univ. Crete, GR
Observations and CTM simulations of high Aerosol Optical Thickness (AOT) over the Mediterranean during dust events.

Lori Neary, Uni-York, Toronto, Canada
Using satellite observations to evaluate an on-line global air quality model (GEM-AQ)

1.6 CO retrievals and results; Task group 3. A special session was run on retrievals of CO from MOPPITT and SCIAMACHY and their validation. In the concluding discussion Miranda van den Broek offered to formulate a "wish list" for the group to act as a guide for future work. Illustrated abstracts of these talks are given in section 6.

Miranda van den Broek, SRON, Utrecht, NL
CO satellite observations from SCIAMACHY, combined with model results and MOPITT observations

Solene Turquety, Service d’Aeronomie, Paris, F
Infrared satellite observations for the study of tropospheric composition

Bart Dils, BIRA-IASB, Brussels, B
The evaluation of SCIAMACHY CO scientific data products, using ground-based FTIR measurements

Voltaire Velazco, IUP, Bremen, D
Ground-based measurements of CO vertical profiles: satellite validation and model comparisons

1.7 Validation of satellite retrievals; Task group 3. Following the CO session, there were two further talks on validation, one being on the excellent results obtained by the DANDELIONS campaign, last year. Illustrated abstracts of these talks are given in section 4.

Ellen Brinksma, KNMI, De Bilt, NL
NO2 and aerosol observations during the DANDELIONS campaign

Remco Braak, KNMI, De Bilt, NL
Ozone Monitoring Instrument Aerosol Products and Validation

1.8 News from the Task groups. The task groups each had brief meetings to discuss progress. In their brief oral reports the three leaders reported:

TG1: Workshops were planned on retrievals of data in the infra-red region, on aerosol parameters and on water vapour. Thomas Wagner.

TG2: Seeking a way to run an intercomparison, probably in collaboration with people from the ACCENT modelling group. Within AT2, more synergy is needed with the other groups. Martin Dameris

TG3: The work in progress was reviewed. It is intended to have a follow up validation campaign (DANDELIONS-2) during the autumn and an NO2 validation workshop in 2007.
1.9 **Workshop finish.** The Coordinator concluded the workshop by:

- expressing the thanks of all those present to our hosts, Maria Kanakidou and her colleagues for their generous hospitality and the enormous efforts that had been made to ensure the smooth running of the meeting with entirely new facilities in the wonderful new building;

- thanking all those present for their participation and particularly for the high quality of the excellent work presented;

- emphasising the need, demonstrated by the work in the Dandelions campaign, for measurements of high temporal and spatial resolution;

- re-iterating the need to demonstrate how ACCENT is providing the opportunities for European integration in this field;

- encouraging PIs to use AT2 as a platform to submit applications for support when the EU Framework 7 call is issued later in the year;

- encouraging PIs to apply to AT2 for assistance in facilitating inter-laboratory exchanges.

The meeting concluded at 18.30 on Tuesday 14\textsuperscript{th} July 2006.
2. **AT2 News and Feedback Session Report**

2.1. **AT2 Report. John Burrows**, the coordinator

   a. **ACCENT progress.** ACCENT had had a second good review but all projects were asked to emphasise the added value that being part of ACCENT brought to their field.

   b. **Financial Situation.** We were close to the budgeted amounts so some rearrangement of the various workshops planned would be made to even out the expenditure.

   c. **Urbino Meeting (23rd – 27th July 2007).**
      * All PIs are urged to attend and present posters.
      * ACCENT is making available funding to support the attendance of younger research workers. PIs are urged to take advantage of this possibility.
      * In view of the ACCENT funding, AT2 will not offer any support for attendance.

   d. **7th framework Programme.** Ulrich Platt reported that there would soon be a call for proposals for the EU 7th Framework Programme. AT2 offered a special opportunity, to form consortia to apply for funding.

2.2. **Funding for meetings and PIs**

   a. **Applications approved at the Steering Group (updated after the following steering group meeting).**

<table>
<thead>
<tr>
<th>2006-7 financial year</th>
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<tr>
<td><strong>Thierry Marbach</strong></td>
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<td>Jean Marie Flaud</td>
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<td>Gerrit de Leeuw</td>
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<td>Ellen Brinksma</td>
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### 2007-8 financial year

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<th>Event Description</th>
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<tr>
<td>Diego Loyola &amp; Thomas Wagner</td>
<td>Brussels</td>
<td>H₂O vapour retrieval workshop April 2007</td>
<td>3.5 k€</td>
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<tr>
<td>Maria Kanakidou</td>
<td>Heraklion</td>
<td>International Aerosol Workshop April 2007 no proposal received</td>
<td>up to 5k€</td>
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<tr>
<td>Ankie Piters</td>
<td>not known</td>
<td>Tropospheric NO₂ workshop summer 2007 no proposal received</td>
<td>up to 15k€</td>
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<tr>
<td>Coordinator</td>
<td>Bremen</td>
<td>6th AT2 workshop 26th to 28th March 2007</td>
<td>up to 30k€</td>
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b. **Funding for student exchanges.** AT2 members are invited to apply for funds to facilitate exchanges or to support meetings. An application help sheet is available on the web site and on request together with templates for submitting proposals and reports.

2.3. **Satellite Data Available.** There was a brief presentation by Peter Borrell, Deputy coordinator. The web pages for data collection and display are live on the AT2 web page but are in need of updating.

★ The steering committee urges PIs to make tropospheric satellite data available to the community via the new web facility.

2.4 **Web Page & Portal.** The AT2 web page and the ACCENT web portal are run in synchrony, and are regularly updated. The page has recently been simplified.

2.5 **Annual reports**

**2004-5.** The report was published at the end of 2005. Although, initially, it was proposed to limit circulation and encourage PIs to buy a copy, the financial difficulties were resolved (thanks to Cloud Workshop not requiring its allocated funds) and copies were circulated to all PIs in January 2006.

**2005-6.** PI contributions were requested for May 31st 2005; the various overviews will be prepared during July and August, and it is hoped to publish the report in October.

★ Those PIs who had not yet submitted their report were asked to do so promptly.

2.6. **Future Meetings and activities.** The steering group has approved the attached schedule.

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<th>Event</th>
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<tr>
<td>ACCENT VOC Expert Meeting</td>
<td>Mon Oct. 30th – Wed. Nov. 1st</td>
<td>Peter Borrell <em>(AT2 supported meeting)</em></td>
<td>Barnsdale Hall Hotel, Rutland</td>
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<td>Tropospheric trace gas retrieval using mid-infrared spectroscopy from space</td>
<td>November 2006 Organiser Jean-Marie Flaud <em>(AT2 supported meeting)</em></td>
<td>Paris</td>
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<td>Dandelions Follow Up</td>
<td><em>date not known</em> Ellen Brinksma <em>(AT2 supported meeting)</em></td>
<td><em>venue not yet known</em></td>
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<tr>
<td>7th Steering Group Meeting</td>
<td>Friday 6th December 2006</td>
<td>ESRIN</td>
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2.7. **e-Learning Module.** Maria Kanakidou and Annette Ladstaetter-Weissenmayer reported on the AT2 e-learning workshop which had been held on the day before the AT2 scientific workshop. The workshop had tested the AT2 e-learning module and learned about the AT2 authoring tool. The approach used by the module had been universally praised. There was a lot of positive feedback and comment which should result in a satisfactory final version. It is expected that the module will be finished in September, and mounted on the ACCENT web site shortly afterwards.

2.8. **Date and Venue for the next workshop.**

Title: Observing trace substances from space and integrating the results with models.

Date and Venue: 26th to the 28th March 2007; IUP, Bremen

Style: A reporting meeting to provide everyone with both the opportunity to make a presentation and to hear about current work within the project. All PIs will be invited to participate; there will be no parallel sessions.

2.9. **Satellite News:** Colleagues were invited to report on news and developments on the satellite instruments and platforms. The following points were made.

* ENVISAT is expected to continue functioning until 2010.
* METOP will be launched in October 2006 with Global Ozone Monitoring Experiment-2 (GOME-2) and Infrared Atmospheric Sounding Interferometer (IASI) on board. Both will be of much interest to the AT2 community.
* CALIPSO was launched by NASA as part of the "A Train". It will provide aerosol information.
* GOME-1 which is nearing the end of its life will overlap with GOME-2 by about a year.
* ACE/ODIN will continue for the foreseeable future.
* NPOESS appears to be in financial trouble and may be reduced in scope.
* GeoTrope not selected for the pre-phase A studies. TRACK (asynchronous orbit) was selected.
3. Abstracts of Invited Talks

Inverse Modelling of emissions using satellite data and surface data

Contribution to the Heraklion ACCENT/AT2 workshop on Data Assimilation

Matthias Beekmann\(^1\), Igor Konovalov\(^2,1\), Jean-François Müller\(^3\), Jenny Stavrakou\(^3\), Laurent Déguillaume\(^1\), Gaelle Dufour\(^4\), Andreas Richter\(^5\), John Burrows\(^5\)

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\(^2\) Institute of Applied Physics / Russian Academy of Sciences, Nizhniy Novgorod, Russia
\(^3\) Belgian Institute for Space Aeronomy, Brussels, Belgium
\(^4\) Laboratoire De Météorologie Dynamique (LMD), IPSL, Ecole Polytechnique, Palaiseau, France
\(^5\) Institute of Environmental Physics and Remote Sensing IUP/IFE, University of Bremen, Bremen, Germany

Summary

Novel methods to derive gridded reactive gas emissions (NO\(_x\), CO, VOC) by inverse modelling from combined data sets of satellite and surface data have been derived. They have been applied both on global and regional scale. The use of these combined data sets allows deriving not only correction factors for a priori emissions, but also their a priori and a posteriori uncertainty.

Introduction

Remarkable recent progress in satellite measurements of the chemical composition of the troposphere opens a challenging perspective to use them as an independent source of information on sources and sinks of atmospheric pollutants. However, the retrieval of such information from measurement data poses serious methodological and computational problems, especially in the case of chemically reactive gases. The paper describes the development of methods that enable “top-to-down” evaluations of NO\(_x\) and CO emissions at global and regional scale (with a high spatial resolution of 0.5 degree for the latter). So called optimal emissions are obtained, which take into account available information on pre-existing (a priori) emission data bases.

Scientific Activities

The paper reports about three original methods to derive reactive gas emissions from combined sets of satellite and surface data:

1) Global scale inverse modelling of NO\(_x\) and CO and VOC emissions,
2) Regional scale inverse modelling of NO\(_x\) emissions;
3) Inverse emission modelling using Bayesian Monte Carlo analysis (NO\(_x\), VOC)

Global scale inverse modelling of NO\(_x\) and CO emissions

Ground-based observations of CO mixing ratios and vertical column abundances together with tropospheric NO\(_2\) columns from the GOME satellite instrument as constraints for improving the global annual emission estimates of CO and NO\(_x\) for the year 1997 (Müller and Stavrakou, 2005). An inverse modelling framework has been developed based on the adjoint of the three dimensional tropospheric model IMAGES (Mueller and Brasseur, 1995). This procedure allows inverting for CO and NO\(_x\) fluxes simultaneously, taking into account
their chemical interactions. The analysis scheme quantifies a total of 39 flux parameters, comprising anthropogenic and biomass burning sources over large continental regions, soil and lightning emissions of NOx, biogenic emissions of CO and non-methane hydrocarbons, as well as the deposition velocities of both CO and NOx. Comparison between observed, prior and optimized CO mixing ratios at NOAA/CMDL sites shows that the inversion performs well at the northern mid- and high latitudes, and that it is less efficient in the Southern Hemisphere, as expected due to the scarcity of measurements over this part of the globe. The inversion, moreover, brings the model much closer to the measured NO2 columns over all regions. Sensitivity tests show that anthropogenic sources exhibit weak sensitivity to changes of the a priori errors associated to the bottom-up inventory, whereas biomass burning sources are subject to a strong variability.

Regional scale inverse modelling of NOx emissions

A novel method has been developed which not only enables a computationally efficient optimisation of a large number of model parameters corresponding to the seasonally averaged NOx emissions, but also drastically diminishes the need in a priori assumptions that may lead to uncontrollable uncertainties of the results (Konovalov et al., 2006). The key features of the method are (i) replacement of a CTM by a set of statistical models describing the relationships between tropospheric NO2 columns and NOx emissions with sufficient accuracy, (ii) consistent estimation of uncertainties of the NO2 columns and the a priori emission data using independent observational data, and (iii) evaluation of uncertainties of the a posteriori emissions by means of a special Bayesian Monte-Carlo experiment which is based on random sampling of errors of both NOx columns and emission rates.

Using the CHIMERE CTM for Western Europe (Schmidt et al., 2001), the method has been applied in two successive studies with slight modifications. In a first study, NOx emissions were inverted over Western Europe for summer 2001 (Konovalov et al., 2006). NO2 columns were derived from GOME (Burrows et al., 1999) and SCIAMACHY measurements (Bovensmann et al., 1999). Insufficient spatial resolution of original GOME measurements was preliminary improved by the deconvolution with SCIAMACHY data (only available for 2003). Additionally, near-surface NO2 concentrations from EMEP ground based monitoring network were used. A second inversion study was performed for summer 2003 for a larger domain covering the entire Europe, the Mediterranean and Middle East region, using a spatially extended version of the CHIMERE model (Konovalov et al., 2005). Tropospheric vertical NOx columns derived from SCIAMACHY were used in combination with near surface ozone observations from the EMEP network.

Inverse emission modelling using Bayesian Monte Carlo analysis

The third inversion method is based on Bayesian Monte Carlo analysis applied to a regional-scale chemistry transport model, CHIMERE (Deguillaume et al., 2006, Beekmann and Derognat, 2003). This method consists in performing a large number of successive simulations with the same model but with a distinct set of model input parameters at each time. Then, a posteriori weights are attributed to individual Monte Carlo simulations by comparing them with observations from the AIRPARIF network: urban NO and O3 concentrations and rural O3 concentrations around the Paris area. The method yields a probability distribution for a posteriori emissions, for a given probability distribution of a priori emissions. A limitation is that a priori information on the spatial and temporal distribution of emissions has to available.
Figure 1: *A priori* and *a posteriori* emissions from ground based in-situ CO observations (CMDL network) and GOME tropospheric NO$_2$ columns; a) for CO emissions for different continents and sectors, (b) the same for NOx emissions.
Scientific results and highlights

Global scale inverse modelling of NOx and CO emissions

Three inversion analyses are proposed in this approach (Mueller and Stavrakou, 2005). In case study A, CO emissions are inverted using only ground-based CO measurements. In case B, ground-based CO and GOME measurements are used as constraints for improving CO and NOx emissions, whereas in case C an additional constraint on the global methane lifetime is used to bring its value close to the recommended value (IPCC, 2001) of 9.6 years. The inversion scheme performs reasonably well in all cases, as shown by the comparison between prior and posterior mixing ratios at NOAA/CMDL stations. The improvement for CO is more significant in the northern high and mid-latitudes, whereas the inversion has small impact on the CO mixing ratios at southern high latitudes. Furthermore, the optimization brings the NO2 columns much closer to the GOME observations over all geographical regions.

The top-down global annual CO flux is estimated at 2928, 2760, and 2688 Tg CO/yr in inversions A, B, and C, respectively (Figure 1a). Note the decrease in cases B and C compared to the a priori, related to the lower OH abundances predicted in these simulations in most of the Northern Hemisphere. Direct CO emissions rise by 5–10 %, due to an increase of anthropogenic emissions by about 30 % with respect to their prior value. The largest increases occur over the former Soviet Union, North America, south Asia and Europe. The biomass burning source, however, is reduced in all cases. The tropospheric methane lifetime is adjusted from 8.55 years in the a priori to 9.13 years in inversion B, and to 9.58 years when the methane lifetime is also constrained. The global annual NOx flux is estimated at 42.1 Tg N/yr, out of which 22.8 Tg N/yr are due to the anthropogenic source (Figure 1b). Evaluation of the inversion studies against independent airborne observations shows that among the three optimization studies, case B (i.e. the simultaneous optimization of CO and NOx), results in a better performance in all regions. Although the improvement in the Tropics and the Southern Hemisphere is generally not very significant, this simulation is found to give more robust results in these regions, compared to the A and C analyses.

The a posteriori errors on the control variables are smaller than the a priori error estimates in almost all cases. As expected due to the wide coverage of the CMDL network, significant error reductions are achieved for CO anthropogenic sources over North America, Far East, Former Soviet Union, and South Asia. On the contrary, African and South American anthropogenic emissions are very poorly constrained. Significant error reductions are also achieved for NOx-related control parameters.

Comparison of our results to previous inverse modelling studies is very encouraging. The inversion of CO emissions perfomed by Bergamaschi et al. (2000a,b), Petron et al. (2002), and Palmer et al. (2003), although not strictly comparable to case B, compare quite well to our results. Furthermore, the results of Martin et al. (2003) are in good agreement with our global NOx surface emissions, even though large discrepancies are found concerning regional scale emissions.
Figure 2: (a) Ratio of the a posteriori to a priori EMEP NOx emissions from combined GOME and SCIAMACHY NO$_2$ columns and EMEP NO$_2$ surface measurements for Western Europe and summer 2001;
(b) Ratio of the a posteriori to a priori EMEP NOx emissions from SCIAMACHY NO$_2$ columns and EMEP O$_3$ surface measurements for summer 2003; the western European domain corresponding to figure 2a is also indicated;
(c) EMEP NOx emissions for summer (June – August 2003) in $10^8$ mol. cm$^{-2}$ s$^{-1}$. 
Regional scale inverse modelling of NOx emissions

The inversion procedure for the Western European domain leads to a strong reduction of uncertainty in emissions averages over a summer (2001) season. While the uncertainty for a priori EMEP emissions (Vestreng, 2004) is determined as about 1.9 (logarithmic factor), the uncertainty in a posteriori emissions is reduced to about 1.4. A priori EMEP emissions are underestimated by several tenths of percent especially over Spain, southern France and Northern Italy (Figure 2a). They are overestimated in particular over southern UK, North-Western France, the Netherlands, and intense shipping regions in the Channel and over the Atlantic. Use of a posteriori emissions in CHIMERE simulations leads to improved comparison with NO2 surface measurements from the EMEP network at 15 sites out of 21 (in terms of reduced root mean squired error).

From the inversion study for the larger domain, it is seen that the a priori emissions are persistently overestimated, in particular, over Great Britain, North-West of France, Netherlands, Greece, and Iraq, but underestimated over Spain, South of France, northern Italy, Israel, Northern Turkey, Iran, Lebanon and Israel. Results obtained with both methods for Western Europe are rather similar. Use of a posteriori emissions in CHIMERE simulations leads to improved comparison with O3 surface measurements from the EMEP network at about 70% of the sites (in terms of reduced root mean squired error and correlation). Additional statistical tests prove that the improvement in the agreement between simulated and measured ozone concentrations is statistically significant.

These results are of large scientific and political relevance. First, they illustrate that satellite and surface measurement facilities not only allow to document pollutant concentrations, but also contain relevant information on pollutant sources. Second, they indicate, in which regions emission cadastres are most likely to be biased. This type of results allows then to initiate a process of revision of emission data on the one hand, and of control of the satellite data inversion procedure and modelling skill on the other hand.

Inverse emission modelling using Bayesian Monte Carlo analysis

The major results obtained from Bayesian Monte Carlo analysis are summarized here. It is shown that the observational constraints strongly reduce the a priori uncertainties in NOx and VOC emissions (Deguillaume, 2006): (1) The a posteriori probability density function for NOx emissions is not modified in its average (Figure 3a), but the standard deviation is decreased to around 20% (40% for the a priori standard deviation). (2) VOC emissions are enhanced (+16%) in the a posteriori PDFs with a standard deviation around 30% (40% for the a priori standard deviation). As a conclusion, this type of analysis has the merit to directly assess the effect of model (parameter) uncertainty on the retrieved emissions. This model uncertainty is sufficiently small to still allow a significant reduction in the a posteriori emission uncertainty compared to the a priori one.
Figure 3: A priori (transparent) and a posteriori (grey) emission distributions averaged over the Ile de France region and summer seasons 1998 and 1999; values represent logarithmic deviations from reference emissions; left for NOx, middle for VOC, right for VOC/NOx emission ratio. While a priori and a posteriori emissions are similar on the average, the distribution of a posteriori emissions is narrower.

Future outlook

Future work on emission inverse modelling will take several directions. On a regional scale, the temporal evolution of NOx emissions will be addressed by using the available set of GOME and SCIAMACHY observations over Europe since 1996. This will allow verification of EMEP trend estimates for European emissions. On both global and regional scale, the simultaneous inversion of emissions of chemically related compounds (NOx, VOC, CO) is a promising research topic, which can be tackled by applying adjoint methods. The inversion of VOC emissions is particularly difficult, as direct tracers of these emissions are only available from limb measurements (e.g. Dufour et al. 2006). Thus also observations of VOC oxidation products will have to be used, such as formaldehyde, glyoxal, CO and ozone, the latter strongly depending on VOC emissions in VOC sensitive regions).

References


Globaerosol: A multi-instrument, long term aerosol dataset from European satellites

Gareth E. Thomas, Elisa Carboni, Caroline Poulsen, Richard Siddans, Don Grainger, Brian Kerridge and Celestino Gomez

University of Oxford, UK

Introduction

Globaerosol is an ESA Data User Element project aimed at providing a 10 year aerosol climatology from European satellite radiometers. The project is making use of the ATSR-2 instrument (on board ERS-2), AATSR and MERIS (on board ENVISAT), and SEVIRI (on board Meteosat-8). The data products to be produced are:

- Aerosol optical depth at 550 and 870 nm.
- Ångström coefficient.
- Aerosol speciation (from a selection of 5 pre-defined types: Maritime, continental, desert, urban/polluted and biomass burning).

Data will be produced on a 10×10 km sinusoidal grid on both a daily and monthly composite basis, and will be available for each individual instrument and as a combined product. The dataset will run from 1995 until the end of 2005 but, due to the launch dates of the various satellites, the dataset will be produced from the follow subsets of instruments:

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<tr>
<th>Date Range</th>
<th>Instruments</th>
</tr>
</thead>
<tbody>
<tr>
<td>Mar. 2002 – Feb. 2004</td>
<td>AATSR and MERIS only</td>
</tr>
</tbody>
</table>

The products will be produced at the nominal overpass time of ENVISAT (from 2002 onwards) or ERS-2 (until 2001).

Two separate retrieval algorithms are being used to provide the aerosol products: in the case of ATSR-2, AATSR and SEVIRI, the Oxford-RAL Aerosol and Cloud retrieval is being used (Thomas et al. 2006), while MERIS results will be provided by a modified version operational MERIS level 2 algorithm (Antoine and Morrel 2005, Santer et al. 2000).

Validation and Merging

Globaerosol products will be primarily validated against measurements from the AERONET network of ground based sun photometers (Smirnov et al. 2000). Comparisons with this dataset will provide both bias and offset values for the optical depths retrieved from each instrument, and a consistent method of estimating the accuracy of each individual product.

The individual products will be merged to form the combined product using an optimal estimator:

$$\hat{x} = \left( \sum_{i}^{N} S_{x_{i}} \right)^{-1} \left( \sum_{i}^{N} S_{x_{i}}^{-1} x_{i} \right)$$  \hspace{1cm} (0.1)

Where $x_{i}$ is the $i$th state estimate and $S_{x_{i}}$ is the error covariance on that state estimate. Here $i$ corresponds to an individual aerosol retrieval, with results $x_{i}$, which have had biases against the AERONET results removed. The error covariance, which provides the weighting of each individual product, is based on the AERONET comparisons plus a time based decorrelation factor, if the results are being temporally interpolated. Since all products are being produced
on the same sinusoidal grid, no special interpolation is required (noting that the 10×10 km grid is considerably more coarse than the resolution of any of the instruments being used).

As the product is being produced for local the ENVISAT or ERS-2 overpass time, no temporal interpolation will be required for the majority of ATSR-2, AATSR and MERIS results. The exception to this occurs near at high latitudes, where consecutive satellite overpasses produce overlapping results. In this instance, each grid box will use an optimal estimate combining the two estimates either side of the nominal ENVISAT (ERS-2) overpass time for that grid box. Results from the geostationary SEVIRI instrument will be merged in the same fashion, with the two SEVIRI results (produced every 15 minutes) bracketing the ENVISAT overpass time will be combined.

Speciation will be based on which aerosol model provided the best fit to the measured radiances, given strong a priori constraints. Where MERIS results are available, its speciation will take precedence, with the AATSR and SEVIRI aerosol type being set accordingly.

**Results**

At present, Globaerosol products have been produced for a single test month (May 2000 for ATSR-2 and September 2004 for the rest) and validation has been performed on these results.

Figure 1 shows the results from the SEVIRI comparison with AERONET for the test month. In addition to AERONET comparisons, monthly mean products have also been compared with their equivalents from the NASA instruments, MODIS and MISR. The results of these comparisons show that, in general the ATSR-2, AATSR and SEVIRI retrievals are performing well. However, the MERIS retrieval algorithm is yet to function properly in its Globaerosol incarnation. There is good correlation between the AERONET and satellite optical depths, although the satellite results show a high level of scatter over the land, particularly for ATSR-2 and AATSR.
Comparison with the NASA instruments shows that the ATSR-2, AATSR and SEVIRI retrievals are producing results with similar patterns and values. Notable areas of difference include the Amazon basin, where the Globaerosol results show somewhat elevated optical depths, and off the west cost of Africa, where the Globaerosol results do not show the very high optical depths shown by the NASA instruments in the biomass burning and Saharan dust plumes. It is possible that both of these differences are due to cloud flagging issues, but further investigation is required.

Concluding remarks

Globaerosol will provide one of the first long term, global aerosol dataset derived from multiple satellite instruments. Despite the ambitious nature of the project, early results suggest that the aerosol retrieval algorithms used are working well and that the resulting products should provide a valuable addition to the available aerosol datasets. Further information on Globaerosol can be found at: http://www.globaerosol.info/.

References

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4. Abstracts of Task Group 1 Contributions

What about the consistency of spectral parameters when using different atmospheric instruments?

Contribution to AT2 Task group 1

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Optical measurements of atmospheric minor constituents are performed using spectrometers working in the UV-visible, infrared and microwave spectral ranges. For example the satellite ENVISAT has been launched with three spectrometers on board, SCIAMACHY and GOMOS working in the UV-visible spectral region and MIPAS working in the thermal infrared. In the future, the combined use of Nadir-viewing UV-visible and thermal infrared spectrometers (remote-sensing experiments such as OMI and TES onboard EOS-AURA, or GOME-2 and IASI onboard MetOP) will provide an important improvement of vertical trace gas concentration profiles. The analysis and interpretation of the corresponding atmospheric spectra require good knowledge of the molecular parameters of the species of interest as well as of the interfering species. In particular meaningful comparisons of profiles retrieved by various instruments using different spectral domains require that the spectral parameters are consistent in these spectral domains. To illustrate how this is difficult we will discuss the problems one is facing to achieve this goal in the case of the formaldehyde molecule.

For the measurement of atmospheric formaldehyde concentrations, mid-infrared and ultraviolet absorptions are both used by ground, air or satellite instruments. It is then of the utmost importance to have consistent spectral parameters in these various spectral domains. Consequently the aim of the study performed at LISA was to intercalibrate formaldehyde spectra in the infrared and ultraviolet regions. The experiments were performed by acquiring simultaneously UV and IR spectra at room temperature and atmospheric pressure using a common optical cell. The reactor (Figure 1) contains two multiple reflection optical systems interfaced to a Fourier transform infrared spectrometer and to an UV-visible absorption spectrometer. The results of the work will be presented allowing one to point out a much better agreement between the set of infrared data and one of the various UV absorption cross sections available in the literature.

![Experimental set-up](image_url)

Figure 1: General set-up of the reactor and spectrometers.
Report from the Third International DOAS Workshop

Contribution to AT2 Task group 1

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¹IUP Heidelberg; ²IUP Bremen; ³Forschungszentrum Jülich

Like the previous two international DOAS workshops of 2001 and 2003, the third international DOAS workshop covered aspects of Differential Optical Absorption Spectroscopy related to the spatially resolved DOAS-measurements. The Workshop has been hosted by the ACCENT/AT2 organization, who gracefully support the meeting costs. This funding also helped more students and oversea scientists to participate to the workshop through travel cost supports.

The third international DOAS workshop was a great success (very good feed-backs) and has allowed people working with the same technique to compare their results and develop new ways to use, elaborate, and improve the DOAS instruments. The participants also profited from the synergistic presentation of the different utilization of the DOAS technique (and other instruments) presented during the workshop. The main feeling that came out from the meeting is that the DOAS community is alive and kicking! Multi-axis applications are developing quickly. The main applications are monitoring of volcanic activity, pollution studies, and halogen chemistry. For the satellites applications the limb retrievals are improving, IR products move ahead. The exploitation of long time series just beginning and new products are possible (Glyoxal). Dense ground-based networks should help for validation. OMI data will be released soon and GOME-2 launch is approaching (17th of July)
Impact of convective outflow and biomass burning into the tropical lower stratosphere as diagnosed from satellite observations and model results

Contribution to AT2 Task group 1

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⁴ University of Saskatchewan, Saskatoon, Saskatchewan, Canada

Abstract.

Space-borne measurements of chemical species (N₂O, CH₄, H₂O and CO) and related products (temperature, cloud occurrence frequency, fire counts, winds, and outgoing longwave radiation) are used to assess the impact of tropospheric processes on the tropical lower stratosphere during the March-May 2002-2004 period. We show that tropospheric air masses characterized by high concentrations of CO, CH₄ and N₂O are uplifted to the Tropical Tropopause Layer (TTL) by strong convective outflows preferentially over Western Africa, Indonesia and Northern South America. The emission of CO and CH₄ associated with biomass burning modifies even more the distribution of these species, particularly above Western Africa and Northern South America. The rapid and strong vertical outflow over Western Africa is also responsible for the dehydration of the cold TTL. Although convection is present over Western Pacific, it does not penetrate into the TTL and, consequently, does not alter the distribution of long-lived species. This is in general agreement with the output from the three-dimensional chemical transport model MOCAGE over the same time frame.
Glyoxal measurements with SCIAMACHY - a new tropospheric species from satellite measurements

Contribution to AT2 Task group 1

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Summary

The first global simultaneous observations of glyoxal (CHOCHO) and formaldehyde (HCHO) columns retrieved from measurements by the Scanning Imaging Absorption Spectrometer for Atmospheric Chartography (SCIAMACHY) satellite instrument are presented and compared to model calculations. The global pattern of the distribution of CHOCHO is similar to that of HCHO. High values are observed over areas with large biogenic isoprene emissions (Central Africa, parts of South America, and Indonesia). Also regions with biomass burning and anthropogenic pollution exhibit elevated levels of CHOCHO. The ratio of the columns of CHOCHO to HCHO is generally of the order of 0.05 in regions having biogenic emissions, which is in reasonable agreement with the current understanding of the oxidation of hydrocarbons emitted by the biosphere.

However and in contrast to our model, high values of both HCHO and CHOCHO are also observed over areas of the tropical oceans. This is tentatively attributed to outflow from the continents and local oceanic biogenic sources of the precursors of HCHO and CHOCHO.

Introduction

Glyoxal, CHOCHO, the simplest alpha dicarbonyl organic compound, is formed from the oxidation of a variety of hydrocarbons [Volkamer et al., 2005a; Volkamer et al., 2001]. Tropospheric CHOCHO has been measured during several campaigns at levels from 0.07 ppbv in rural areas to several ppb in heavily polluted regions. The main sources are thought to be oxidation of biogenic VOCs such as isoprene, biomass burning and to a much less certain degree direct anthropogenic emissions. During the day, photolysis and reaction with OH determine the CHOCHO lifetime. This was found to be 1.3 hours for overhead sun conditions, as compared to about 1.6 hours for HCHO [Volkamer et al., 2005b]. Global observations of CHOCHO from space offer the potential of identifying photochemical hot spots in the Earth’s atmosphere [Volkamer et al., 2005b], and coupled with observations of HCHO constrain our understanding of biogenic emissions, biomass burning, and urban pollution.

Recently, the tentative identification of glyoxal in measurements from the OMI (Ozone Monitoring Instrument) satellite instrument has been reported (Kurosu et al. http://www.cfa.harvard.edu/~tkurosu/). In this study, the global fields of HCHO and CHOCHO were derived from the measurements of the satellite instrument SCIAMACHY using the method of Differential Optical Absorption Spectroscopy (DOAS). and compared with a global model including HCHO and CHOCHO chemistry are presented.
Results

In Figure 1, the first global annual composite of the CHOCHO and HCHO retrieved from SCIAMACHY for the year 2005 are depicted. Enhanced CHOCHO column amounts of $\geq 1 \cdot 10^{15}$ molec·cm$^{-2}$ are mainly observed in South America, Africa and Asia. In South America, high column values can be found in the Amazon Basin, the world’s largest tropical rain forest, and low values are found over the Andes. In Africa, enhanced CHOCHO is found over the tropical rain forests and in regions having regular biomass burning events e.g. in Ghana. In Asia, large values of CHOCHO are observed over Cambodia, Thailand, Sumatra, Borneo, south of the Himalaya Mountains in India and Nepal and above densely populated areas in China. Generally, the pattern observed for CHOCHO is similar to the global picture of HCHO, indicating common main sources.

![Figure 1 Yearly mean for glyoxal (a) and formaldehyde (b) derived from SCIAMACHY observations in 2005. Sub-figures (c) to (e) illustrate the ratio between measured CHOCHO and HCHO.](image)

Fig. 2 shows zooms into the glyoxal slant columns (in different colour scale) for some regions of high anthropogenic activity. For comparison, the respective distribution of tropospheric NO$_2$ (Beterle, 2004) is also displayed.

Several Glyoxal hotspots show up over large cities and congested areas where also very high tropospheric NO$_2$ VCDs are observed, for instance Los Angeles, US eastcoast (a), Belgium/Netherlands and the Po Valley (b), Hong Kong, Tokyo, Seoul and the Eastern China (c), or Rijad, Dschidda and the metropolises at the Persian golf (d). Hence photochemical hot spots due to anthropogenic activities can clearly be identified in the Glyoxal composite. many of the patterns are similar to those observed in the NO$_2$ columns. It has to be noted that this coincidence cannot be explained by a spectral interference what can easily be seen for fits for polar regions in summer, showing very high stratospheric NO$_2$ SCDs but no Glyoxal.
Figure 2: Zoom in for (a) the USA, (b) Europe, (c) Far East and (d) the Middle East, showing enhanced Glyoxal SCDs due to anthropogenic emissions. For (d), the Glyoxal mean for 2003 is displayed. The small images show the respective mean tropospheric VCD of NO₂.

Figure 3: Comparison of SCIAMACHY (top) and TM4 modelled (bottom) Glyoxal columns for 2005. The overall pattern agrees well for biogenic emissions, but the model does not show enhanced values over water.

The TM4-ECPL model (Tsigaridis and Kanakidou [2003]) computes column distributions of CHOCHO and HCHO (Fig. 3) that are similar to those derived from SCIAMACHY. The model calculated annual mean values maximize in the tropics for both compounds over biogenic emission areas with columns exceeding 12·10¹⁵ molec·cm⁻² for HCHO and 0.8·10¹⁵ molec·cm⁻² for CHOCHO. However, the model does not capture the enhancement of CHOCHO columns over the tropical ocean seen in SCIAMACHY observations, although it simulates reasonably well the tropical enhancement of HCHO. This may indicate the existence of primary or secondary tropical sources of CHOCHO that are neglected in the model. The ratio of the simulated CHOCHO to HCHO has its highest values over continental areas in the tropics (up to 0.04) and over the mid latitudes (ratios up to 0.024). These ratios...
are smaller than observed by SCIAMACHY and this is probably best explained by missing sources or an inadequate chemical description of the formation of CHOCHO in the model.

Conclusions
The first global observations of HCHO and CHOCHO using measurements of the SCIAMACHY instrument have been presented and discussed. The global pattern of CHOCHO columns was found to be similar to that of HCHO, indicating common atmospheric sources in particular isoprene. The ratio between CHOCHO and HCHO was found to be about one to twenty in source regions such as the tropical rain forests, similar to predictions. At some locations, larger ratios are found and this is attributed to unidentified additional sources of CHOCHO. Large CHOCHO columns are found primarily over areas having strong biogenic emissions in the tropics which appear to be the dominant global source. This is confirmed by a 3-d global CTM, which simulates the observed CHOCHO annual mean column reasonably well. During strong biomass burning events CHOCHO can also be clearly observed as well as over heavily polluted cities. More detailed comparisons between measurements and model results, coupled with retrievals of other trace gases such as NO2, will constrain our knowledge of VOC chemistry in the current generation of atmospheric models.

References
Volkamer, R. et al., DOAS measurement of glyoxal as an indicator for fast VOC chemistry in urban air, GRL 32, 2005a.
Wittrock F., et al., Simultaneous global observations of glyoxal and formaldehyde from space, GRL (accepted), 2006.
Total and tropospheric BrO retrieval from space and ground-based UV-VIS observations

Contribution to AT2 Task group 1
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Bromine monoxide (BrO) is an important component responsible for stratospheric ozone depletion at the global scale. BrO has also been found massively in the polar boundary layer during ‘Tropospheric Ozone Hole’ events. Furthermore, several evidences have been reported from satellites, ground-based and balloon platforms suggesting the presence of a few pptv of BrO in the global free-troposphere, which can have a significant impact on the tropospheric ozone budget and other chemical key parameters of the troposphere.

In the present study, we focus first on the validation of a scientific algorithm, developed at BIRA-IASB, for the retrieval of total BrO columns from GOME and SCIAMACHY nadir measurements. GOME and SCIAMACHY BrO columns are compared with correlative measurements of BrO columns resolved into their stratospheric and tropospheric contributions and photochemically matched to satellite observations. Correlative data are derived from zenith-sky measurements performed at Harestua (60°N) and multi-axis DOAS observations at Observatoire de Haute-Provence (OHP, 44°N) and Reunion Island (22°S).

The second half of the work consists to validate SCIAMACHY limb BrO scientific products for the 2002-2004 period, in the framework of the BOOST project. For this purpose, a profiling algorithm, based on the Optimal Estimation Method, has been applied to zenith-sky observations at Harestua and at Observatoire de Haute-Provence. The retrieved BrO profiles are compared to limb BrO profiles from the following groups: IUP/IFE Bremen, IUP/IFE Heidelberg and Harvard Smithsonian. The comparisons are performed in the same photochemical conditions since the profiling algorithm includes a stacked box photochemical model, enabling the retrieval of BrO profiles at any solar zenith angle.

The overall consistency of the combined satellite and ground-based measurements is investigated in order to draw conclusion on the global distribution of BrO in the atmosphere and to contribute to improve our understanding of the atmospheric bromine partitioning.
Update on the status of the comparison of Box-Air-Mass-Factors and Radiances for Multiple-Axis Differential Optical Absorption Spectroscopy (MAX-DOAS) Geometries calculated from different UV/visible Radiative Transfer Models

Contribution to AT2 Task group 1

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1. Abstract

A comparison exercise of radiative transfer models (RTM) of various international research groups was performed for Multiple AXis Differential Optical Absorption Spectroscopy (MAX-DOAS) viewing geometry. In contrast to previous comparison exercises, box-air-mass-factors (box-AMFs) for various atmospheric height layers were modelled, which describe the sensitivity of the measurements as a function of altitude. In addition, radiances were calculated allowing the identification of potential errors, which might be overlooked if only AMFs are compared. Accurate modelling of radiances is also a prerequisite for the correct interpretation of satellite observations, for which the received radiance can strongly vary across the large ground pixels.

The comparison exercises included different wavelengths and atmospheric scenarios (with and without aerosols); the results were systematically investigated with respect to their dependence on the telescope’s elevation angle and the azimuth angle. For both dependencies, a strong and systematic influence of aerosol scattering was found indicating that from MAX-DOAS observations also information on atmospheric aerosols can be retrieved. During the various iterations of the exercises, the results from all models showed a substantial convergence, and the final data sets agreed for most cases within about 5%. Larger errors were found for cases with low atmospheric optical depth, for which the photon paths along the line of sight of the instrument can become very large. The differences occurred between models including full spherical geometry and those using only plane parallel approximation indicating that the correct treatment of the earth’s sphericity becomes indispensable.

The modelled box-AMFs constitute an universal data base for the calculation of arbitrary (total) AMFs by simple convolution with a given trace gas concentration profile. Together with the modelled radiances and the specified settings for the various exercises, they can serve as test cases for future RTM developments.
2 Introduction

In June 2005, a workshop on radiative transfer modelling (RTM, we will use this abbreviation also for the term ‘radiative transfer model’) for the UV and visible spectral range was held at University of Heidelberg, Germany (http://satellite.iup.uni-heidelberg.de/index.php/RTM_Workshop). The aim of the workshop was to conduct a comparison of 9 state of the art RTM from various international research groups (see section 2). These models use different approaches to solve the atmospheric radiative transfer equations; they also treat the spatial discretisation of the atmosphere and the earth’s sphericity in different ways or operate in plane parallel geometry (see section 2). In the UV and visible spectral range, thermal emission can be neglected, and as relevant processes remain only absorption by molecules, aerosols and the earth’s surface as well as scattering (or reflection) on molecules, aerosols and the ground. Most of the participating RTMs were developed for the simulation of remote sensing observations from various platforms (e.g. ground, aircraft, balloon, satellite), which is a fundamental prerequisite for their correct interpretation. In addition, these RTMs can also be applied to investigate the energy deposition of the solar radiation in the earth’s atmosphere, especially under cloudy conditions. Current state of the art RTMs simulate the earth’s sphericity, refraction and multiple scattering processes. Some models are capable of modelling polarisation and three-dimensional scenes. The latter is in particular important for the correct interpretation of satellite observations of atmospheric trace gases, because of their large ground pixels (the pixels sizes of current instruments range from about 15x15km² to 300x40km² [European Space Agency (ESA), 1995; Burrows et al., 1999; Bovensmann et al., 1999; Levelt and Noordhoek, 2002; EUMETSAT, 2005].

This comparison exercise focuses on the simulation of the recently developed Multiple AXis Differential Optical Absorption Spectroscopy (MAX-DOAS) technique [Hönninger and Platt, 2002; Leser et al., 2003; Von Friedenburg et al., 2003; Bobrowski et al., 2003; van Roozendael et al., 2003; Hönninger et al., 2004; Wagner et al., 2004; Wittrock et al., 2004; Heckel et al., 2005; Oetjen et al., 2006; Sinreich et al., 2005; Frieß et al., 2006; Frins et al., 2006]. In contrast to the well established ground based observations of zenith-scattered sun light [Noxon et al., 1979; Solomon et al., 1987], MAX-DOAS observations are directed into the illuminated atmosphere under various elevation angles. Since for a slant viewing geometry, the absorption paths through (and accordingly the AMFs for) the lower atmosphere can become rather large, MAX-DOAS observations are especially sensitive to tropospheric trace gases. From the combination of observations at several elevation angles, also information on the vertical trace gas profile can be obtained [Hönninger and Platt, 2002; Bruns et al. 2004; Wittrock et al., 2004; Heckel et al., 2005, Sinreich et al., 2005; Bruns et al. 2006; Oetjen et al., 2006, Wittrock, 2006].

For MAX-DOAS observations, the photon path length along the line of sight is limited by the atmospheric visibility; thus their sensitivity is strongly depending on the aerosol optical depth. Therefore, the knowledge of the atmospheric aerosol properties is a prerequisite for the correct interpretation of MAX-DOAS measurements. However, in turn, from MAX-DOAS observations of trace gases with constant (and known) concentration profiles (like for the oxygen molecule O₂ or the oxygen dimer O₈ [Greenblatt et al., 1990; Wagner et al., 2002]), information on the atmospheric aerosol properties can be also retrieved [Wagner et al., 2004; Wittrock et al., 2004; Heckel et al., 2005; Sinreich et al., 2005; Frieß et al., 2006; Oetjen et al., 2006].

The simulation of the MAX-DOAS geometry exhibits a particular challenge for RTMs because of the extended light paths through the lowest atmospheric layers. For such slant line of sights, the correct treatment of the earth’s sphericity can become important. Moreover, the
optical depth with respect to Rayleigh and aerosol scattering can become very large and the correct implementation of multiple scattering becomes indispensable.

Besides the specific focus on the MAX-DOAS observation geometry, this comparison exercise differs from previous exercises [Sarkissian et al., 1995; Potylyakov et al., 2001; Loughman et al., 2004; Hendrick et al., 2006] also in further aspects. First, in addition to the calculation of AMFs, radiances were also simulated and compared. The comparison of radiances is a very sensitive tool to test the correct performance of the RTMs, because it allows the identification of errors, which might not be detectable if only AMFs were compared (for the AMF-calculation potential errors of the modelled radiances typically cancel each other). In addition, the correct calculation of radiances is of great importance for the interpretation of satellite observations, for which the brightness within an observed ground pixel can vary strongly, especially for partly clouded scenes [Wagner et al., 2005]. For this comparison exercise, all modelled radiances are expressed as normalised radiance with respect to the solar irradiance:

\[ R_{\text{normalised}} = \frac{R \cdot \pi}{I} \]  

Here \( R \) and \( I \) denote the modelled radiance and solar irradiance, respectively. The comparison of normalised radiances makes the comparison independent on the absolute values of the solar irradiance.

Another new aspect of this comparison exercise is that instead of AMFs for specific trace gas profiles, so called box-AMFs were calculated. Such Box-AMFs characterise the ratio of the partial SCD to the partial VCD of an atmospheric layer with an assumed constant trace gas concentration. It is interesting to note here, that for optically thin absorbers (optical depth \(<1\)), the box-AMFs are identical to the so called weighting functions [Rodgers, 1976; 2000]. For these cases, they can also be approximated by the average geometrical path length extension with respect to the vertical thickness of the selected layer. In this comparison exercise, box-AMFs for optically thin absorbers were calculated.

The great advantage of calculating box-AMFs is that they can serve as an universal data base to calculate appropriate (total) AMFs for arbitrary species with different height profiles. Total AMFs can be easily calculated from the box-AMFs and the respective trace gas profile as the sum of the box-AMFs over the whole atmosphere weighted by the respective partial trace gas VCD:

\[ AMF = \sum_{0}^{TOA} \frac{AMF_i \cdot VCD_i}{\sum_{0}^{TOA} VCD_i} \]  

Here \( AMF_i \) and \( VCD_i \) refer to the box-AMF and the partial VCD for layer \( i \); within the layer the trace gas concentration is assumed to be constant. The sum is carried out over all layers \( i \) (from the surface to the top of the atmosphere, TOA). The vertical discretisation chosen for the calculation of the box-AMFs within this exercise is shown in Table 1.

The particular aims of this comparison exercise include the following aspects:

A) The comparison and quantification of the differences of current RTMs from different research groups.

B) The identification of shortcomings and the assessment of the uncertainties of the model results.
C) The investigation of the sensitivity of MAX-DOAS observations for different viewing geometries with a particular focus on the influence of aerosols.

D) The provision of a consolidated set of box-AMFs for selected atmospheric conditions: these box-AMFs allow the calculation of (total) AMFs for any kind of atmospheric trace gas profiles.

E) The definition and documentation of basic atmospheric reference scenarios. These scenarios (together with the model results) are ideal test cases for future RTM developments.

Table 1  Lower boundaries and vertical extensions of the atmospheric layers selected for the box-AMF calculation. Please note that above 1000m the layers are thinner than the distances between the layers. If needed, box-AMFs for layers in between can be derived by interpolation.

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</tr>
<tr>
<td></td>
<td>20000m</td>
<td>1000m</td>
</tr>
</tbody>
</table>

3. Basic settings and tests

3.1 Definition of model parameters

In order to allow a meaningful interpretation of the RTM results, several basic properties were set to predefined values for all models. For the temperature and pressure profiles the data from the US standard atmosphere were used [United States Committee on Extension to the Standard Atmosphere, 1976]. The temperature was interpolated linearly to match the vertical discretisation of the individual models; for the pressure the logarithm was interpolated linearly. No instructions were given for the setting of numerical parameters (e.g., vertical or angular discretization). The ozone cross section from SCIAMACHY [Bogumil et al., 2003] was used (see Table 3); all other atmospheric absorbers were ignored. The exercises were carried out for five wavelengths covering regions of the UV and visible spectral range, where important trace gases show characteristic absorptions (see Table 3). To minimise any complications due to different telescope apertures, the field of view was set to very small values (<0.1°).

3.2 Participating models

Nine models from eight international research groups took part in the comparison exercise. All models included multiple scattering schemes. Besides the way they solve the radiative
transfer equation, they also differ in their treatment of the earth’s curvature and refraction. The basic features of the individual models are summarised in Table 2.

Table 2  Overview on the participating models and some important properties.

<table>
<thead>
<tr>
<th>Model / Institute</th>
<th>Type</th>
<th>Treatment of sphericity</th>
<th>Refraction</th>
</tr>
</thead>
<tbody>
<tr>
<td>MODTRAN / Switzerland</td>
<td>discrete ordinate approach</td>
<td>spherical</td>
<td>yes</td>
</tr>
<tr>
<td>MCC++ / Russia</td>
<td>Backward Monte Carlo</td>
<td>spherical</td>
<td>no</td>
</tr>
<tr>
<td>MCARaTS / Japan</td>
<td>Forward Monte Carlo</td>
<td>-plane-parallel for the direct solar beam&lt;br&gt;-spherical for the line of sight&lt;br&gt;-plane-parallel for the diffuse radiation</td>
<td>yes</td>
</tr>
<tr>
<td>PROMSAR / Italy</td>
<td>Backward Monte Carlo</td>
<td>spherical</td>
<td>no</td>
</tr>
<tr>
<td>UVspec/DISORT / Belgium</td>
<td>discrete ordinate approach</td>
<td>-spherical for direct solar beam&lt;br&gt;-plane parallel for multiple scattering&lt;br&gt;-spherical for integration along the line of sight</td>
<td>yes</td>
</tr>
<tr>
<td>VECTOR / Canada</td>
<td>technique of successive orders of scattering</td>
<td>-spherical for direct solar beam&lt;br&gt;-plane parallel for multiple scattering&lt;br&gt;-spherical for integration along the line of sight</td>
<td>no</td>
</tr>
<tr>
<td>SCIATRAN / Bremen</td>
<td>Discrete Orinate Method</td>
<td>Plane parallel</td>
<td>yes</td>
</tr>
<tr>
<td>SCIATRAN / Bremen</td>
<td>-Finite Difference method for multiple scattering (plane parallel)&lt;br&gt;- characteristics method for integration along the line of sight (spherical)</td>
<td>-spherical for direct solar beam&lt;br&gt;-plane parallel for multiple scattering?&lt;br&gt;-spherical for integration along the line of sight?</td>
<td>yes</td>
</tr>
<tr>
<td>TRACY-II / Heidelberg</td>
<td>Backward Monte Carlo</td>
<td>spherical</td>
<td>Partly (see text)</td>
</tr>
</tbody>
</table>

Table 3  Wavelengths and ozone cross sections used in the RTM comparison exercise. The data are taken from Bogumil et al. [2003]. Also shown are trace gases, which are typically analysed at the respective wavelengths.

<table>
<thead>
<tr>
<th>Wavelength [nm]</th>
<th>O₃ cross section [cm²]</th>
<th>Trace gases analysed in this spectral range</th>
</tr>
</thead>
<tbody>
<tr>
<td>310</td>
<td>9.59 · 10⁻²⁰</td>
<td>SO₂</td>
</tr>
<tr>
<td>360</td>
<td>6.19 · 10⁻²³</td>
<td>BrO, HCHO, O₄</td>
</tr>
<tr>
<td>440</td>
<td>1.36 · 10⁻²²</td>
<td>NO₂, IO</td>
</tr>
<tr>
<td>477</td>
<td>5.60 · 10⁻²²</td>
<td>O₄</td>
</tr>
<tr>
<td>577</td>
<td>4.87 · 10⁻²¹</td>
<td>O₄</td>
</tr>
</tbody>
</table>

Table 4  Aerosol properties for the different test cases (Case A1 represents a pure Rayleigh-atmosphere).

<table>
<thead>
<tr>
<th>Case</th>
<th>Aerosol extinction [km⁻¹]</th>
<th>Altitude range</th>
<th>Asymmetry parameter</th>
</tr>
</thead>
<tbody>
<tr>
<td>A1</td>
<td>0</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>A2</td>
<td>0.5</td>
<td>0-2km</td>
<td>0.68 (urban)</td>
</tr>
<tr>
<td>A3</td>
<td>0.1</td>
<td>0-1km</td>
<td>0.68 (urban)</td>
</tr>
<tr>
<td>A4</td>
<td>0.5</td>
<td>0-2km</td>
<td>0.75 (maritime)</td>
</tr>
<tr>
<td>A5</td>
<td>0.1</td>
<td>0-1km</td>
<td>0.75 (maritime)</td>
</tr>
</tbody>
</table>
Table 5  Overview of the solar zenith angles, the elevation and the relative azimuth angles of the telescope. Exercises were performed for specific combinations of these angles.

<table>
<thead>
<tr>
<th>Selected elevation angles (SZA: 20°)</th>
<th>Selected relative azimuth angles (SZA: 80°)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1°</td>
<td>0°</td>
</tr>
<tr>
<td>2°</td>
<td>30°</td>
</tr>
<tr>
<td>3°</td>
<td>60°</td>
</tr>
<tr>
<td>6°</td>
<td>90°</td>
</tr>
<tr>
<td>10°</td>
<td>120°</td>
</tr>
<tr>
<td>20°</td>
<td>150°</td>
</tr>
<tr>
<td>90°</td>
<td>180°</td>
</tr>
</tbody>
</table>

The RTM comparison was performed for five different aerosol scenarios including also a pure Rayleigh atmosphere (see Table 4). For the investigation of the dependence of the model results on the viewing direction of the telescope, specific viewing directions of the telescope were chosen as described in Table 5.

4 Selected results

In Fig. 1 the results for the vertical optical depth (with respect to Rayleigh-scattering) and the normalised radiances (taking into account Rayleigh-scattering and ozone absorption) are shown. The optical depth increases with decreasing wavelength as expected for the strong wavelength dependence of Rayleigh-scattering. Accordingly, the normalised radiances increase towards shorter wavelength, but decrease again for the shortest wavelength (310nm) because of the strong ozone absorption. While at the beginning the results showed large differences, finally they are almost identical for all RTMs.

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**Fig. 1** Modelled vertical optical depth with respect to Rayleigh-scattering (left) and normalised radiances taking into account also ozone absorption (right) as a function of wavelength. Top: results at the beginning of the comparison. Bottom: results in summer 2006.
Fig. 2  Box-AMFs for 577nm and elevation angle of 1° as a function of altitude (logarithmic scale). Left: pure Rayleigh-atmosphere. Right: including aerosol scattering (scenario A2, see Table 4). Top: results at the beginning of the comparison. Bottom: results in summer 2006.

5 Conclusions

An extended comparison exercise of 9 radiative transfer models of 8 international research groups for the UV and visible spectral range was performed. The comparison exercise focused on the recently developed MAX-DOAS method, which is particularly sensitive to tropospheric trace gases. Typical MAX-DOAS geometries with varying elevation angles and relative azimuth angles were simulated for different wavelengths and atmospheric scenarios (pure Rayleigh atmosphere or varying aerosol loads, see Table 4). In contrast to previous comparison exercises, not only air mass factors were modelled, but also (normalised) radiances. From the comparison of radiances it was particularly possible to discover errors, which might have been overlooked if only AMFs were compared. Accurate modelling of radiances is also a prerequisite for the correct interpretation of satellite observations, for which the measured radiance can strongly vary across large ground pixels.

The comparison exercise was divided into two major parts. In the first part, the dependence on the elevation angle and in the second part, the dependence on the relative azimuth angle between the directions of the telescope and the sun was investigated. For both parts, overall a very good agreement (differences <5%) for the radiances and the box-AMFs of the various RTMs was found. It should be noted that these differences might be further reduced if fixed discretisation schemes (e.g. for the vertical layering and the scattering angles) were used. Larger deviations occurred only for the consideration of a pure Rayleigh atmosphere at large wavelengths. For such cases, the path length along the line of sight becomes very large and the correct treatment of the earth’s sphericity becomes important.

Within two additional sensitivity studies, the influence of a modified ground albedo and aerosol phase function was investigated. Especially for the lowest atmospheric layers, a high ground albedo strongly enhances the observed radiance and the box-AMFs. In contrast, for the considered cases the change in the aerosol phase function has a rather small effect.
During the various iterations of the comparison exercise substantial progress was made. The comparison of the first results had exhibited large differences between the models, both for the modelled radiances and box-AMFs. Many errors were caused by simple mistakes and could be easily corrected. In this way, the RTM comparison exercise caused a consolidation of currently applied RTMs.

In addition, it turned out that for correct MAX-DOAS RTM, two aspects need special attention. First, especially close to the ground, the vertical discretisation has to be chosen fine enough (<100m); otherwise, the strong gradients close to the surface could not be treated correctly, and interpolation errors can lead to substantial errors. In addition, it was found that for low atmospheric optical depth (low aerosol load and large wavelength) and small elevation angles, the correct treatment of the earth’s sphericity becomes indispensable.

The results are summarised in a paper which will be submitted to the Journal of atmospheric chemistry and physics at the end of July 2006.

Acknowledgements
We want to express two special thanks to Karin Kreher and Greg Bodeker (NIWA, New Zealand for the initiative for this workshop and their leading role in setting up the exercise specifications. Part of the workshop was sponsored by the ACCENT project. Work performed by Hitoshi Irie and Yugo Kanaya at FRCGC/JAMSTEC was supported by Global Environment Research fund (B-051).

References
ESA Publication Division (SP-1182), GOME, Global Ozone Monitoring Experiment, users manual, edited by F. Bednarz, European Space Research and Technology Centre (ESTEC), Frascati, Italy, 1995.


In this work, we show that HNO₃ data can be accessed using satellite measurements in the thermal infrared spectral region from the nadir soundings collected by the IMG. The Optimal Estimation Method is used for the retrievals of HNO₃ for 10 successive days of IMG operation in April 1997. The measurements provide HNO₃ abundances with some vertical information, enabling us to derive, for the first time, global distributions of HNO₃ in the troposphere and the stratosphere. The emphasis is put on the study of upper tropospheric data that can be used as indicator of pollution sources. The results for the troposphere are discussed by comparison with the distribution of NO₂ obtained by the Global Ozone Monitoring Experiment (GOME) instrument for the same period in April 1997.
5. Abstracts of Task Group 2 Contributions

Validation of tropospheric ozone values derived from the chemistry-climate model E39/C

Contribution to AT2 Task group 2

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DLR Institut für Physik der Atmosphäre, Oberpfaffenhofen, Germany

Extended Abstract

The interactively coupled chemistry-climate model (CCM) E39/C has been used for ensemble simulations of the time period between 1960 and 2020 (e.g. Dameris et al., 2005; 2006). Model results are investigated with the main focus on upper tropospheric and lower stratospheric values and parameters. This presentation focus on the evaluation of simulated tropospheric ozone values, i.e. columns and profiles, with available data of ground based and radiosonde measurements.

The interaction between emission changes (especially NOx) and longer-term hemispheric and global tropospheric O3 changes derived from observations is not adequately understood. Moreover, impact of climate forcing on tropospheric O3 changes over recent years is still very uncertain. The importance of stratospheric ozone changes and its impact on tropospheric ozone concentrations needs to be investigated in more detail. Trend estimates for different stations (regions) look very different, e.g. in the Northern Hemisphere there is a different pattern of long-term changes between North America, Europe, and Japan; specific investigations of Taubman (pers. comm., 2006) for the Baltimore region seem to indicate that meteorological and temperature variability and trends are more important than increase in fuel consumption to explain tropospheric O3 changes. Southern Hemisphere mid-latitudes show an increase in tropospheric O3 whereas the South Pole region shows almost no changes in the last 30 years.

Interactively coupled CCMs provide a helpful tool to investigate individual radiative, dynamical, and chemical processes and to study their interactions and feedback mechanisms. First results of an inter-comparison between data derived from transient simulations employing the CCM E39/C (1960-2000) and radiosonde measurements are shown. The aim of this study is not only to demonstrate the model’s abilities and deficiencies to simulate the distribution of tropospheric ozone and its evolution in recent years, but also to get a better understanding of the cause-effect-chains which are responsible for the detected behaviour.

As an example, Figure 1 shows climatological means of the seasonal cycle of tropospheric ozone as derived from measurements performed at the Meteorological Observatory Hohenpeißenberg (47°N, 11°E) and corresponding results from the transient simulation using the CCM E39/C. Although the model captures the main structure, there are obvious differences in the model data. For example, the tropopause (and therefore the “ozonopause”) is located higher in the model than observed. Whereas the seasonal cycle matches well in the lower stratosphere and upper troposphere, the seasonal cycle of ozone in the middle to lower troposphere indicates a slight shift in time and obvious higher mixing ratios during winter season in the CCM E39/C.

Figure 2 displays the evolution of ozone mixing ratios at distinct tropospheric pressure levels as derived from measurements at Hohenpeißenberg and data from the CCM E39/C simulation. In the lower troposphere (here: 850 hPa) the agreement between model and observed data is
reasonable. Whereas the model data also indicate a quasi-linear enhancement of ozone mixing ratio at all other tropospheric heights, observations above 850 hPa show obvious non-linear long-term changes with a strong increase in the late 1970s and late 1980s and no clear trend after year 1995. Moreover, ozone measurements in the middle and upper troposphere show a much larger month-to-month variability than it is simulated by the CCM E39/C. This can be easily explained by the fact, that the CCM E39/C has a horizontal grid size of about 3.75° whereas the measurements belong to a single location. Another apparent reason for the inconsistent long-term behaviour of tropospheric ozone mixing ratios is that in our transient model simulation we assume a step-wise linear increase of anthropogenic NOx emissions (e.g. from traffic and industry) which only inadequately describes what was observed. To clarify the impact of prescribed insufficient emission changes, newly published emission data sets will be used for inter-comparison (e.g. from the EC RETRO-project; data available at the ACCENT-GEIA emission data base).

Figure 1. Mean annual cycle of ozone as derived from measurements at station Hohenpeißenberg (47°N, 11°E), top panel, from the CCM E39/C for the corresponding grid point, middle panel. The bottom panel shows absolute differences between model and measurements. Data used cover the time period between 1967 and 2000. Units in ppbv.
In addition to tropospheric ozone measurements of balloon- and ground-based instruments it is necessary to use tropospheric ozone data products which are derived from space-borne observations (e.g. GOME; SCIAMACHY, OMI) to get a better global view on regional and temporal changes. Therefore, we have started to cooperate with the group of Pieter Valks (DLR-IMF; project within AT2 Task Group 1: “Retrival of tropospheric ozone columns from UV-Nadir measurements by the GOME/ERS-2, GOME-2/METOP and OMI Instruments”). In figure 3 a monthly mean climatology for tropical (20°S-20°N) tropospheric ozone columns as derived from E39C for the years from 1992 to 1999 is shown. Similar data products will be
determined from satellite-instrument measurements and will be used for further inter-comparison studies.

![Figure 3. Climatology of monthly mean tropical tropospheric ozone columns derived from E39/C transient simulation [in Dobson Units] based on the years from 1992 to 1999. Each picture displays the results for one month, from January (top left) to December (bottom right).](image)

The further plan within this project is to systematically investigate data of the CCM E39/C ensemble simulations in order to detect and quantify the impact of emission changes and climate change on long-term tropospheric ozone changes on a global scale.

**Acknowledgement**

We especially thank H. Claude (Deutscher Wetterdienst Meteorologisches Observatorium Hohenpeissenberg) and G. König-Langlo (AWI Bremerhaven) to provide us ozone data records.

**References**


Evaluation of GEM-Chemistry Modelling and Data Assimilation System: Five year global simulation results from GEM-AQ

Contribution to AT2 Task group 2
L. Neary, J. W. Kaminski and the MAQNet Team

York University, Toronto, Canada

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.

The primary objective of this study is to evaluate the performance of the tropospheric chemical weather model (GEM-AQ) which is based on the Canadian operational weather prediction model, the Global Environmental Multiscale (GEM) model (Côté et al., 1998).

A base model simulation of GEM-AQ was run for 5 years (2001-2005). The purpose of this base run was to examine the ability of the model to capture seasonal variations and regional distributions of ozone and other species. It also provided means to investigate issues relating to emissions and other processes. Observations from satellites such as GOME, SCIAMACHY, ACE, AURA-MLS and MOPITT provide the opportunity to assess the system’s ability to simulate the distribution of a variety of chemical species.

This simulation was done with a horizontal resolution of 1.8x1.8 degrees and 28 vertical levels up to 10 hPa. The tropospheric chemistry package is run on-line with the meteorological model and has 50 gas phase species and 5 size-resolved aerosol types.

Comparisons with ozonesonde climatologies (Logan, 1999) have indicated reasonable agreement at many stations, although some over-prediction was seen in the upper troposphere northern hemisphere winter.

The monthly averaged tropospheric column NO₂ also compares reasonably well the observations from SCIAMACHY, indicating that the anthropogenic emissions used for the simulation are acceptable for this type of simulation.

Figure 1a: GEM NO₂ tropospheric column August 2004
Figure 1b: SCIAMACHY NO₂ column August 2004
We are currently using data from two satellite instruments to evaluate the model carbon monoxide, the MLS on AURA and the MOPITT instrument on TERRA. These initial comparisons indicate that the biomass burning emissions used are most likely too low. The comparison with AURA-MLS CO measurements at 200, 150 and 100 hPa indicate the model values at 200 hPa are generally 2 times too low and often don’t pick up the major biomass burning signatures in the tropics. At higher levels, the magnitude of the model CO values approach those observed by MLS.

Another indicator of model performance is the calculation of methane and methyl chloroform lifetimes. The lifetimes can be used to evaluate OH which is the main sink of these species. There is large variability in these numbers between different models, but the IPCC Climate Change 2001 report gives a value for methane and methyl chloroform lifetimes of 8.4 years and 4.8 years, respectively. This GEM-AQ simulation produces a methane lifetime of 8.8 years and methyl chloroform of 5.3 years.

Overall, the tropospheric chemical weather model can reproduce the general regional distributions and seasonality of the species simulated and can be used to do more detailed analyses of processes. The results from this simulation can be used to initialize higher resolution scenario runs to further study and improve model processes.

References
Comparison between AATSR and MODIS AOD and assimilation in a regional chemistry transport model

Contribution to AT2 Task group 2

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Introduction

Satellite remote sensing can be a cost-effective method to monitor the highly variable aerosol fields on regional scales (e.g. Robles-Gonzalez et al., 2003). Retrieved aerosol optical depth fields consist of data that are irregularly distributed in space and time. Data assimilation allows the calculation of continuous fields in space and time from observations that are irregularly distributed. The objective of our studies is demonstrate that we can use the assimilation of aerosol optical depth (AOD) to obtain more knowledge on the PM2.5 distribution over Europe.

Methods

Data assimilation defines a new atmospheric state by combining the observed and modelled state in an intelligent and statistically sound way. In this study we used an ensemble Kalman filter to assimilate ATSR2 for May 2000 and MODIS and AATSR AOD retrievals for the year 2003, within the LOTOS-EUROS chemistry transport model (Schaap et al., 2004). We have used random noise to the emissions of NOx, SOx, VOC, NH3 and particles to define the ensemble and therewith the model uncertainty.

Modis AND AATSR data

To study the quality and suitability of the MODIS and AATSR AOD measurements for assimilation in the LOTOS-EUROS model, we have compared datasets from both satellite instruments for the year 2003 with ground-based measurements from the AERONET network. The intercomparison reveals a seasonal bias between MODIS AOD and AERONET (Fig.1).

Figure 1. Difference between retrieved AOD from MODIS and colocated AERONET measurements as function of day in the year 2003.
This seasonality in the bias is not visible when comparing the MODIS fine fraction of AOD with AERONET. MODIS_fine shows a rather constant bias with AERONET of ~10 %. AATSR on average slightly underestimates the AERONET values but shows some suspected high values over scandinavia and Ireland. The correlation between the satellite datasets and AERONET is 0.65-0.72.

**Assimilation Results**

We illustrate the results of the ongoing study with two experiments, which were carried out to assess the performance of the system as function of the uncertainty in the ATSR AOD data. For the first experiment we assumed a lower limit for the accuracy: the variability of the AOD retrievals within a grid cell. The second experiment assumes accurate AOD data with a standard deviation of 0.03. Comparison to the ATSR AOD data and AERONET data reveals that the model (without assimilation) underestimates AOD systematically (see fig 2). Verification of SO₄, NO₃ and NH₄ concentrations does not reveal large systematic deviations from observed concentrations and are probably not the cause for the underestimation of AOD.

In experiment 1 the assimilation procedure is able to reproduce the observed AOD values where the uncertainty in the observed data is lower than the ensemble variability. Overall, in experiment 1 the gap between the free model and the observations is reduced by more than 50-60 %. By assuming highly accurate AOD data (exp. 2) the system is able to reproduce the AOD fields accurately, see Figure 2. Experiment 2 yields a somewhat better agreement to observations at AERONET sites. Through the assimilation the monthly mean PM2.5 levels are increased by about 20-50% in western and central Europe. Emissions were estimated to be 50-150% higher than inventoried for Europe, which is larger than the uncertainty estimates in the emission inventories.

A sensitivity study showed that the innovations in aerosol concentrations and emissions are strongly affected by systematic errors made in the procedure. For example, implementing a second optical parameterization yielded 50% higher AOD values based on the same aerosol column which reduced the concentration changes considerably.

![Figure 2. Comparison of the average retrieved AOD compared to the corresponding average for the model and assimilation experiments for the swaths in the area bound by lon = -5.30 and lat = 45.57.](image-url)
Discussion

We have shown that the data assimilation technique can be successfully applied to assimilate aerosol optical depth fields over Europe. However, from a scientific point of view a successful application is still hampered by a number of unresolved issues.

The systematic underestimation of the retrieved AOD data by the model can be explained by a number of factors. 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. The assumptions on optical properties, water uptake and (BC) mixing state have a large influence on the AOD calculations. Lastly, the retrieved AOD values may be biased due to undetected glint and cloud contamination.

Conclusions

The assimilation system is able to closely resemble the retrieved AOD when the AOD data would be very accurate, although this is not the status of the present day AOD data. This is illustrated by the large bias in MODIS total AOD in summer as compared to the AERONET data. Inclusion of MODIS AOD_fine appears to largely solve this bias problem for the assimilation study. Due to the systematic underestimation of AOD the assimilation scheme induces a large (and unrealistic) change in the emissions estimates. Nonetheless, we illustrate that combining modeling and satellite retrievals can be used to derive secondary information such as PM2.5 concentrations. An improvement is foreseen when the availability and quality of the AOD observations improve our knowledge on aerosol optical properties and water uptake improves and model formulations become more accurate. Moreover, constraining the model with ground based measurements of aerosols is probably a mayor step forward.

References

6. Abstracts of Task Group 3 & CO Session Contributions

Ozone Monitoring Instrument Aerosol Products and Validation
Contribution to AT2 Task group 3
Remco Braak and Ellen Brinksma

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What is the role of aerosols in climate change? Answering this question is one of the science goals of the Ozone Monitoring Instrument (OMI) on board of EOS-Aura. To this end, the aerosol index, aerosol optical thickness, and single-scattering albedo are retrieved from the near-ultraviolet and visible spectra that are obtained by OMI. Two approaches to the aerosol retrieval are discussed. The near-ultraviolet method, which uses 3 wavelengths, is a continuation of the method that has been applied to TOMS observations for many years. The multiwavelength method extends the near-ultraviolet method to up to 17 wavelengths. Ground-based data (AERONET) and satellite data (MODIS, POLDER) will be used to validate the various aerosol products. First results of the validation will be presented. Special attention will be given to similarities and differences between the methods.
NO$_2$ and aerosol observations during the DANDELIONS campaign

Contribution to AT2 Task group 3

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OMI NO$_2$ and aerosol observations are of interest for studies of tropospheric pollutants, their sources and transport. To assess the quality of these satellite data, a 9-week campaign within the DANDELIONS (Dutch Aerosol and Nitrogen Dioxide Experiments for valIdation of OMI and SCIAMACHY) project took place.

We will show results for:
1) NO$_2$ column intercomparisons between the ground instruments (including five MAXDOAS instruments, one nitrogen dioxide lidar, ground in situ NO$_2$ observations)
2) Validation of OMI and SCIAMACHY overpass data with the campaign results, also discussing the homogeneity of the tropospheric NO$_2$ field, and, quantitatively, the influence of the assumed NO$_2$ profile shape on the OMI retrievals.
3) Closure-type studies on a selected number of golden days during the campaign
And present some plans for a second campaign, which will take place in Cabauw in September of 2006.
Quantitative analysis of SCIAMACHY CO and comparison with model and MOPITT

Contribution to AT2 Task group 3
Miranda van den Broek
SRON, Sorbonnelaan 2, 3584 CA Utrecht

The satellite instrument SCIAMACHY has been measuring CO total columns for several years now, allowing to study the inter and intra-annual variability of CO. We present a quantitative and systematic analysis of SCIAMACHY CO total column measurements for the years 2003 and 2004. The CO retrievals in the near infrared are hampered by the presence of an ice layer on the detector, but a detailed correction scheme has been included in the retrieval algorithm. This results in CO total columns with a precision of 1% for monthly means under ideal circumstances (cloud free pixels, high surface albedo, spatial averaging). For lower surface albedos a precision of less than 10% is obtained. Thus, SCIAMACHY CO total column measurements are of sufficient quality to provide useful new information. The SCIAMACHY CO observations are compared with both MOPITT, which measures CO in the thermal infrared, and results of the TM4 chemistry transport model, taking into account the different averaging kernels of both instruments.
The Evaluation Of SCIAMACHY CO Scientific Data Products, using Ground-Based FTIR Measurements

Contribution to AT2 Task group 3

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Abstract

In the framework of the European EVERGREEN project, three scientific algorithms, namely WFM-DOAS, IMAP-DOAS and IMLM, have been developed to retrieve the total column amounts of key atmospheric trace gases including CO from SCIAMACHY nadir observations in its near-infrared channels. These channels offer the capability to detect trace gases in the planetary boundary layer, potentially making the associated retrieval products suited for regional source-sink studies.

The retrieval products of these three algorithms, in their present status of development, have been compared to independent data from a ground-based quasi-global network of Fourier-transform infrared (FTIR) spectrometers, for the year 2003. Comparisons have been made for individual data, as well as for monthly averages. To maximize the number of coincidences that satisfy the temporal and spatial collocation criteria, the individual SCIAMACHY data points have been compared with a 3rd order polynomial interpolation of the ground-based data with time. Particular attention has been paid to the question whether the products reproduce correctly the seasonal and latitudinal variabilities of the target species. Our overall assessment of the data quality of the currently available latest versions of the CO total column products from the three scientific retrieval algorithms indicate that while significant improvements have been made, the quality criteria for inverse modelling have not been met yet. Our assessment of the requirements regarding FTIR validation of possible future CO products is that additional FTIR sites geared specifically to the detection of tropospheric pollution would be most helpful to truly assess SCIAMACHY’s boundary layer detection capabilities.

Relative Bias of SCIAMACHY CO products as a function of station latitude
Long range transport of pollution from space: a multiplatform (MOPITT, TES, ACE) analysis of CO observations

Contribution to AT2 Task group 3

Solène Turquety

CNRS, Service d’Aeronomie, Paris VI, France

In this study, we compare the CO measurements provided by three different instruments: the nadir measurements from MOPITT/Terra and TES/Aura, and the solar occultation measurements from ACE/SCISAT-1. Observations in solar occultation allow the measurement of a series of trace gases with high vertical resolution (~3km) from the middle troposphere to the thermosphere. However, this technique implies a limited coverage and does not allow for measurements below ~6km. Observations with a nadir viewing geometry allow a good spatial and temporal coverage, but very limited vertical resolution. We explore the complementarities between the measurements for the study of the long range transport of pollution.
Ground-based Measurements of CO Vertical Profiles: Satellite Validation and Model Comparisons

Contribution to AT2 Task group 3

Voltaire Velazco\textsuperscript{1}, Justus Notholt\textsuperscript{1}, Thorsten Warneke\textsuperscript{1}, Katinka Petersen\textsuperscript{1}, Mark Lawrence\textsuperscript{2}, Holger Bremer\textsuperscript{1}, Miriam Sinnhuber\textsuperscript{1} and Otto Schrems\textsuperscript{3}

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Ground-based solar absorption measurements using Fourier transform infrared (FTIR) spectrometers provide precise information about the concentration profiles of many atmospheric trace gases. Therefore these measurements play a vital role in the validation of current and future satellite instruments that measure atmospheric trace gases. Carbon Monoxide (CO) volume mixing ratio (VMR) profiles retrieved from ship borne solar absorption measurements recorded on the Atlantic have been compared with space borne measurements. The high vertical resolution of the ground-based measurements allows to detect enhancements in the lower and upper troposphere separately. By comparison with model data, the contributions of different sources such as biomass burning, fossil fuel combustion and oxidation of methane (CH\textsubscript{4}) and non-methane hydrocarbons (NMHC) have been quantified. In addition to measurements on board the research vessel Polarstern, we will also present measurements in the tropics (Suriname, 5.8°N 55.2°W) and in the Arctic (Ny Alesund, Spitsbergen, 79°N, 12°E). We will also show measurements of strato-mesospheric CO taken in the polar regions.
7. CO workshop at the Heraklion AT2 scientific workshop

Miranda van den Broek

At the AT2 scientific workshop in Heraklion, a small but interesting workshop was held on carbon monoxide, CO. Four speakers reported on CO data from various satellite (MOPITT, TES, SCIAMACHY, ACE) and FTIR observations, sometimes combined with atmospheric chemistry transport models. These presentations were followed by a discussion on the need for extra analyses and/or measurements of CO, in order to increase our understanding of its atmospheric concentration.

A summary of the presentations and the discussion is given below.

Miranda van den Broek – Quantitative analysis of SCIAMACHY CO and comparison with model and MOPITT

SCIAMACHY has measured total column CO in the near infrared for the past years. The CO retrievals have been hampered by an ice layer on the detector, but this has been corrected for. This results in CO total columns with a precision of 1% for monthly means under ideal circumstances (cloud free pixels, high surface albedo, spatial averaging). For lower surface albedos a precision of less than 10% is obtained. Comparison with CO total columns from collocated TM4 model results show a very good agreement (see Figure 1). Regarding the MOPITT satellite instrument, comparison with SCIAMACHY is complicated because of the different averaging kernels of both instruments. First comparison of MOPITT and the TM4 model shows that the model somewhat underestimates the MOPITT CO measurements.

![Figure 1. Monthly mean SCIAMACHY CO total columns (black diamonds) from september 2003 to september 2004 over different locations (longitude and latitude are given in brackets), compared to TM4 model results. The blue triangles show the results of the TM4 model with climatological biomass burning emissions. The red asterixes show model results using an up to date biomass burning emission database.](image)

B. Dils – The evaluation of SCIAMACHY CO scientific data products, using ground-based FTIR measurements
In the framework of the European EVERGREEN project, three scientific algorithms, namely WFM-DOAS, IMAP-DOAS and IMLM, have been developed to retrieve the total column amounts of key atmospheric trace gases including CO from SCIAMACHY nadir observations in its near-infrared channels. These channels offer the capability to detect trace gases in the planetary boundary layer, potentially making the associated retrieval products suited for regional source-sink studies. The retrieval products of these three algorithms, in their present status of development, have been compared to independent data from a ground-based quasi-global network of Fourier transform infrared (FTIR) spectrometers, for the year 2003. Comparisons have been made for individual data, as well as for monthly averages. To maximize the number of coincidences that satisfy the temporal and spatial collocation criteria, the individual SCIAMACHY data points have been compared with a 3rd order polynomial interpolation of the ground-based data with time. Particular attention has been paid to the question whether the products reproduce correctly the seasonal and latitudinal variabilities of the target species. Our overall assessment of the data quality of the currently available latest versions of the CO total column products from the three scientific retrieval algorithms indicate that while significant improvements have been made, the quality criteria for inverse modelling have not been met yet. Our assessment of the requirements regarding FTIR validation of possible future CO products is that additional FTIR sites geared specifically to the detection of tropospheric pollution would be most helpful to truly assess SCIAMACHY’s boundary layer detection capabilities.

Figure 2. Average difference in CO column [%] between the three SCIAMACHY retrieval algorithms and results from the FTIR global network. The results are plotted per latitude band.

S. Turquety – Long range transport of pollution from space: a multiplatform (MOPITT, TES, ACE) analysis of CO observations

In this study, we compare the CO measurements provided by three different instruments: the nadir measurements from MOPITT/Terra and TES/Aura, and the solar occultation measurements from ACE/SCISAT-1. Observations in solar occultation allow the measurement of a series of trace gases with high vertical resolution (~3km) from the middle troposphere to the thermosphere. However, this technique implies a limited coverage and does not allow for measurements below ~6km. Observations with a nadir viewing geometry allow a good spatial and temporal coverage, but very limited vertical resolution. We explore the complementarities between the measurements for the study of the long range transport of pollution.
Table 1. Differences [%] between MOPITT and ACE (smoothed with the MOPITT averaging kernel and a-priori data) for each MOPITT level

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<th>MOPITT-ACE*(%)</th>
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<td>850</td>
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ACE*: ACE as seen by MOPITT (smoothed)

There's good general consistency between the ACE and MOPITT data. The smoothed ACE data underestimate CO compared to MOPITT. ACE results can provide information on the plume height in the upper troposphere. This is further investigated with the help of a trajectory model.

Figure 1. Model results from the flexpart back trajectory model, showing where measured CO over Asia (25N, 125E) in the 5-7 km layer originates from. Blue = low CO, red = high CO concentrations.

Nadir MOPITT measurements allow a monitoring of the transport (good spatial and temporal resolution). ACE CO profiles give accurate information on the transport in the upper troposphere and therefore complement lack of vertical information of nadir measurements like MOPITT. ACE observations provide additional information on the origin and chemical composition of the plumes in the upper troposphere. Analysis of the long range transport from China is now possible, using the available satellite observations and a global model (LMDz-INCA).

Velazco - Ground-based Measurements of CO Vertical Profiles: Satellite Validation and Model Comparisons

Ground-based solar absorption measurements using Fourier transform infrared (FTIR) spectrometers provide precise information about the concentration profiles of many atmospheric trace gases. Therefore these measurements play a vital role in the validation of current and future satellite instruments that measure atmospheric trace gases. Carbon Monoxide (CO) volume mixing ratio (VMR) profiles retrieved from ship borne solar absorption measurements recorded on the Atlantic from the research vessel Polarstern have been compared with space borne measurements (MOPITT) and model results (MATCH), see
The high vertical resolution of the ground-based measurements allows to detect enhancements in the lower and upper troposphere separately. By further comparison with the model data, the contributions of different sources such as biomass burning, fossil fuel combustion and oxidation of methane (CH4) and non-methane hydrocarbons (NMHC) have been quantified. In addition to these measurements on board the research vessel Polarstern, measurements in the tropics (Suriname, 5.8°N 55.2°W) were also presented, as well as measurements of strato-mesospheric CO taken in the polar regions.

Figure 3. CO total columns along the track of the Polarstern vessel, which carries the FTIR instrument. Model results (MATCH), FTIR results and results from the satellite instrument MOPITT are compared.

Discussion on CO – where do we stand?

The diagram above schematically shows the areas where CO is measured. Satellite instruments that measure in the thermal infrared such as MOPITT, TES and AIRS, observe CO throughout the atmosphere with more or less vertical information, but are less sensitive to the boundary layer. SCIAMACHY is able to measure CO from the ground to the stratosphere, but has a large noise error and measures total columns only. Due to the low albedo, it is also not able to measure above sea. Ground based measurements are carried out in many places and can contribute to the knowledge of CO in the boundary layer. FTIR stations are more sparse, but can observe throughout the atmosphere, which is also true for airborne campaigns. The latter can give more information on the vertical distribution of CO.

Next to and sometimes in combination with these observations, models are also used to describe or analyze the distribution of CO in the atmosphere. They can be divided into chemistry transport models, inverse models and data assimilation models.
Related to this diagram, some questions were asked to start the discussion: Is more validation of satellite instruments needed? How? Are more measurements or other sources of information needed to complement our knowledge on CO?

The outcome of the discussion was a CO- “wishlist”, summarizing what kind of further research the AT2 community would want for CO:

- Can "column" insitu measurements of be made from aircraft (or have they already been made)?
- Can the MOZAIC data be used for validation of SCIA? -> a remark was made by U. Platt that these observations are not representative for a SCIA footprint, because they are usually done in polluted areas. Comparison with MOPITT data has already been done.
- Can sun glind measurements be used to get observations above sea?
- "cloud-slicing" (selecting SCIAMACHY data on the basis of cloud top height, e.g. with CO2 or CH4) may be used in the validation of SCIA with MOPITT.
- Can we get extra information on the vertical distribution of CO by using the difference in averaging kernels between MOPITT day/night measurements? J. Remedios is already looking into this.
- Is it possible to do CO satellite validation with interferometers?
- For inverse modelling, information on the ocean background is important.

This list may be further extended, for instance by modellers.
8. Appendices

A. AT2 5th Workshop Participants

### Coordinator and Steering group (task group members are shown below)

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### Guests

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### Task Group 3

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### AT2 5th Workshop Programme

**2006**

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<td><strong>09.00 – 17.00</strong></td>
<td>e-Learning workshop (by invitation only)</td>
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<td><strong>18.00 – 21.00</strong></td>
<td><strong>Registration and welcome get together</strong></td>
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<td>Buses depart from the hotel</td>
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<td><strong>09.00</strong></td>
<td>John Burrows (Coordinator IUP, Bremen, D)</td>
<td>Workshop opening</td>
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<tr>
<td><strong>09.15</strong></td>
<td>Ulrich Platt Uni-Heidelberg, D</td>
<td>Atmospheric trace substances emitted by volcanoes</td>
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**Task Group 1**

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<tr>
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<td>Phillipe Ricaud Laboratoire d’Aérologie, Toulouse, F</td>
<td>Impact of convective outflow and biomass burning into the tropical lower stratosphere as diagnosed from satellite observations and model results</td>
</tr>
<tr>
<td><strong>10.25</strong></td>
<td>Catherine Wespes Uni-Libre, Brussels, B</td>
<td>Tropospheric studies using infrared spectroscopic measurements from space</td>
</tr>
<tr>
<td><strong>10.50 – 11.15</strong></td>
<td></td>
<td>break for coffee or tea</td>
</tr>
<tr>
<td><strong>11.15</strong></td>
<td>J.-M. Flaud, B. Picquet-Varrault, A. Gratien, J. Orphal and J.-F. Doussin LISA, Créteil, F</td>
<td>What about the consistency of spectral parameters when using different atmospheric instruments?</td>
</tr>
<tr>
<td><strong>11.40</strong></td>
<td>Nicolas Theys BIRA–IASB, Brussels, B</td>
<td>Total and tropospheric BrO retrieval from space and ground-based UV-VIS observations</td>
</tr>
<tr>
<td><strong>12.05</strong></td>
<td>Thomas Wagner Uni-Heidelberg, D</td>
<td>Recent Progress in the Radiative Transfer Correction Group</td>
</tr>
<tr>
<td><strong>12.30 – 14.00</strong></td>
<td></td>
<td>Lunch</td>
</tr>
<tr>
<td><strong>14.00</strong></td>
<td>Andreas Richter IUP, Bremen, D</td>
<td>Glyoxal measurements with SCIAMACHY - a new tropospheric species from satellite measurements</td>
</tr>
<tr>
<td><strong>14.30</strong></td>
<td>Thierry Marbach Heidelberg, D</td>
<td>Highlights of the Third International DOAS Workshop</td>
</tr>
<tr>
<td>Time</td>
<td>Speaker</td>
<td>Affiliation</td>
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<tr>
<td>15.00</td>
<td>Matthias Beekmann</td>
<td>LISA, Uni. Paris-XII, F</td>
</tr>
<tr>
<td>15.45 - 16.15</td>
<td>break for coffee or tea</td>
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<tr>
<td>16.15</td>
<td>Martin Dameris</td>
<td>DLR, Oberpfaffenhofen, D</td>
</tr>
<tr>
<td>16.40</td>
<td>Renske Timmermans</td>
<td>TNO, Apeldoorn, NL</td>
</tr>
<tr>
<td>17.05</td>
<td>Maria Sfakianaki</td>
<td>ECPL, Univ. Crete, GR</td>
</tr>
<tr>
<td>17.30</td>
<td>Lori Neary</td>
<td>Uni-York, Toronto, Canada</td>
</tr>
<tr>
<td>17.55 to 18.30</td>
<td>Task group meetings.</td>
<td></td>
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<tr>
<td>20.00</td>
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<tr>
<td>Tuesday 4th July 08.30</td>
<td>Buses depart from the hotel</td>
<td></td>
</tr>
<tr>
<td>09.00 – 10.30</td>
<td>Peter Borrell</td>
<td>P&amp;PMB Consultants, UK</td>
</tr>
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<td></td>
<td></td>
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<tr>
<td>10.30 – 11.00</td>
<td>break for coffee or tea</td>
<td></td>
</tr>
<tr>
<td>11.00</td>
<td>Gareth Thomas</td>
<td>Uni-Oxford, UK</td>
</tr>
<tr>
<td>12.00</td>
<td>Miranda van den Broek</td>
<td>SRON, Utrecht, NL</td>
</tr>
<tr>
<td>12.30 – 14.00</td>
<td>lunch</td>
<td></td>
</tr>
<tr>
<td>14.00</td>
<td>Solene Turquety</td>
<td>Service d’Aeronomie, Paris, F</td>
</tr>
<tr>
<td>14.20</td>
<td>Bart Dils</td>
<td>BIRA-IASB, Brussels, B</td>
</tr>
<tr>
<td>Time</td>
<td>Speaker/Task Group</td>
<td>Topic/Session</td>
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<tr>
<td>14.40</td>
<td>Voltaire Velazco, IUP, Bremen, D</td>
<td>Ground-based measurements of CO vertical profiles: satellite validation and model comparisons</td>
</tr>
<tr>
<td>15.00</td>
<td><strong>Discussion on CO validation</strong></td>
<td></td>
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<tr>
<td>15.30 – 16.00</td>
<td></td>
<td>break for coffee or tea</td>
</tr>
<tr>
<td>16.00</td>
<td>Ellen Brinksma, KNMI, De Bilt, NL</td>
<td>NO$_2$ and aerosol observations during the DANDELIONS campaign</td>
</tr>
<tr>
<td>16.00</td>
<td>Remco Braak, KNMI, De Bilt, NL</td>
<td>Ozone Monitoring Instrument Aerosol Products and Validation</td>
</tr>
<tr>
<td>17.00</td>
<td>Thomas Wagner</td>
<td>Task group 1 – prospects and plans</td>
</tr>
<tr>
<td>17.20</td>
<td>Martin Dameris</td>
<td>Task group 2 – prospects and plans</td>
</tr>
<tr>
<td>17.40</td>
<td>Ankie Piters, KNMI, de Bilt, NL</td>
<td>Task group 3 – prospects and plans</td>
</tr>
<tr>
<td>18.00</td>
<td>John Burrows</td>
<td>Conclusions: Where do we go from here? <strong>Workshop Finish</strong></td>
</tr>
<tr>
<td>18.30</td>
<td></td>
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</tr>
<tr>
<td><strong>Wed. 5th July</strong></td>
<td>09.00 – 12.00</td>
<td><strong>AT2 Steering Committee Meeting</strong></td>
</tr>
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