Scientific challenges in chemical data assimilation

Henk Eskes

Royal Netherlands Meteorological Institute
P.O.Box 201, 3730 AE De Bilt, The Netherlands
eskes@knmi.nl

ABSTRACT

This paper provides a short overview of atmospheric chemical data assimilation and satellite observation, with a focus on ozone and on tropospheric chemistry measurements. Ozone data assimilation is receiving increasing attention over the past five years. This development is related to vertical extensions of numerical weather prediction models that include the full stratosphere. In particular assimilation-based reanalysis runs that include ozone as prognostic variable are valuable for atmospheric chemistry research, protocol monitoring and NWP. Satellite ozone observations, ozone data assimilation and ozone forecasting are discussed. The focus will be on aspects related to numerical weather prediction. The performance of the KNMI ozone assimilation system is discussed in more detail. Satellite observations of the tropospheric chemistry composition is a field of active research: the retrieval uncertainties are large, related to the presence of clouds, aerosols and complicated surface properties. Very recently several new data sets have become available on tropospheric CO, CH_4, NO_2, SO_2, CH_2O and aerosols. New instruments like OMI and AIRS can deliver air-quality data on a daily basis for different trace gases. It will be a major challenge to set up data assimilation and inverse modelling analysis systems that can make optimal use of these new data sets.

1 Introduction

Data assimilation is at the core of modern numerical weather forecasting. The analysis provides a global description of the state of the atmosphere and it is the basis for a reliable medium-range weather forecast. With data assimilation the atmospheric observational data sets can be combined with knowledge of the dynamics and chemistry of the atmosphere to provide global 3D maps of the chemical composition consistent with the available observational data. Data assimilation will play an increasingly important role to rationalise the huge atmospheric composition (chemistry, aerosols) observational data base that is generated - and will be generated - by present and future satellite instruments. Reanalysis assimilation data sets, such as those from the European Centre for Medium Range Weather Forecast (ECMWF) ERA-40 project, are valuable for assessments of, in particular, the chemical and dynamical changes in the ozone layer (e.g. WMO-UNEP, 2002). The Global and regional Earth-system (Atmosphere) Monitoring using Satellite and in-situ data (GEMS, EU project 2005-2009) project is a major European effort to set up a comprehensive data assimilation system to exploit the available satellite data sets on atmospheric composition.

1.1 Chemical data assimilation

In the field of atmospheric chemistry the use of data assimilation is still new, although extensive satellite data sets are available and similar benefits can be expected. In a pioneering paper Fisher and Lary (1995) demonstrated the benefits of the 4D-Var assimilation approach to analyse measurements of several chemical species with a comprehensive chemistry model. Several 4D-Var studies with full chemistry models have been published since (e.g. Elbern and Schmidt, 2001; Khattatov *et al.,* 1999; Errera and Fonteyn, 2001).

Because of the available satellite ozone data sets a majority of the assimilation studies so far have focused on this compound. The use of the Kalman filter and sub-optimal Kalman filter techniques for the assimilation of long-lived chemical species in chemistry-transport models was discussed by Menard *et al.* (2000), by Khattatov *et al.* (2000) and by Eskes *et al.* (2003). The Data Assimilation Office of NASA has developed the GEOS ozone data assimilation system for the operational analysis of TOMS and SBUV/2 data, as described in Riishøjgaard *et al.* (2000) and Štajner *et al.* (2001).

1.2 Ozone and numerical weather prediction

Numerical weather prediction centres, such as the ECMWF and the National Centres for Environmental Prediction (NCEP) have started programs for the assimilation of satellite ozone data. The first experiences of ozone assimilation with the ECMWF model are discussed in a paper by Hólm and co-workers (1999). Ozone in the ECMWF 40-year reanalysis is discussed by Dethof and Hólm (2003). Assimilation of UARS MLS and GOME data with the UK Met Office Unified model are discussed in a paper by Struthers *et al.* (2001).

There are several benefits of the assimilation of ozone satellite data for numerical weather forecast.

1. The retrieval of temperature profiles from e.g. the TOVS instruments is influenced by ozone. Cold, stratospheric ozone absorbs and emits infrared radiation, and thereby reduces the radiance or brightness temperature as observed by the satellite instrument.
2. Ozone has a strong influence on both short and long-wave radiation and the temperature in the middle atmosphere. An accurate knowledge of the ozone distribution is expected to lead to improvements of these aspects.
3. The time evolution of ozone contains information on the wind field that transports ozone.
4. Global ozone forecasts based on ozone analyses are of direct use for (clear-sky) surface UV forecasts (e.g. Long *et al.*, 1996)

The relation between ozone and meteorological features has a long history, dating back to the work of Dobson in the 1920s. Especially at mid and high latitudes, and in Winter, ozone shows a large variability which is mainly caused by transport. The total amount of ozone has been correlated with the passage of fronts, the jet stream, the tropopause height and temperature at 100 mb. In the modern literature two relations between ozone and the meteorological fields have received much attention, namely the correlation between ozone and (potential) vorticity (Vaughan and Price, 1991; Allaart *et al.*, 1993, Jang *et al.*, 2003) and the direct influence on the wind field when ozone observations are assimilated with a modern assimilation system (4D-Var, Kalman).

A more direct way of exploiting meteorological information contained in ozone observations is by assimilation into a meteorological analysis system which contains ozone as a model variable. Since ozone concentrations are influenced by the history of the wind fields before the observation, it is important to use an advanced data assimilation scheme which includes the time dimension. Riishøjgaard (1996) showed that the analysis of simulated tracer observations in a 2D barotropic model with 4D-Var has a large and positive impact on the wind field. During the EU SODA project experiments have been performed with the ECMWF 4D-Var system (Hólm *et al.*, 1999; Stoffelen *et al.*, 1999). A 4D-Var OSSE (Observing System Simulation Experiment) with simulated TOVS ozone column data was performed with the French ARPEGE model (Peuch *et al.*, 2000). This work showed a positive impact on the winds in the troposphere when the idealised simulated TOVS columns were assumed to be very accurate. However, for more realistic TOVS observation/retrieval errors the impact is only small. The impact of ozone on the forecast quality may be improved by using high-quality ozone observation (e.g. from UV-Vis satellite instruments like TOMS, GOME) and by using height-resolved ozone measurements.

To conclude: several experiments have shown that accurate ozone data may be beneficial for both stratospheric and tropospheric forecasts. However, most of the ozone assimilation work until now has been univariate. More work is needed to quantify the impact of ozone on the other variables of the NWP models, and to judge the role of ozone for improving the forecasts. Essential for success is a good characterisation of both model and measurement related biases.

2 Satellite ozone observations

Ozone is a well-observed compound. Below we will provide examples of past and present ozone monitoring satellite instruments, categorised according to measurement technique. More information can be found for instance in the WMO/CEOS report (2001) or the IGACO report (2004).

1. *Nadir viewing UV-Visible spectrometers.*
   High quality, detailed information of the ozone distribution is available on an almost continuous basis from 1979 onwards. The successful Total Ozone Mapping Spectrometer (TOMS) spectrometers (on 4 different satellites) measures the total column of ozone with a good resolution of about 50 km, and with a nearly global coverage each day (http://www.toms.gsfc.nasa.gov, e.g. McPeters *et al.*, 1998). The Solar Backscatter Ultra Violet (SBUV, SBUV/2) instruments have been flown on the NOAA satellites for basically the same time period, and provide additional vertical profile information (e.g. Bhartia *et al.*, 1996). Since 1995 the European spectrometer GOME (Global Ozone Monitoring Instrument) on the ESA ERS-2 satellite has collected 8 years of ozone data (Burrows *et al.*, 1999). SCIAMACHY (Scanning Imaging Absorption Spectrometer for Atmospheric Cartography) is part of the Envisat payload, launched in 2002 (Bovensmann *et al.*, 1999). The Dutch-Finnish OMI (Ozone monitoring Instrument) on NASA's EOS-Aura was launched in July 2004 (Levelt, 2002; Aura special issue 2005). OMI is meant to continue the TOMS long-term ozone record.

2. *Nadir viewing infrared spectrometers.*
   Total columns of ozone are retrieved using the 9.7 μm channel of the High-Resolution Infrared Sounder (HIRS) on all of the NOAA TIROS-N operational Polar-Orbiting Environmental Satellites (Neuendorffer, 1996). The major NWP centres are assimilating the radiances of these instruments. Therefore a natural step (and a potential improvement over the ozone retrievals) is the adjustment of ozone by the HIRS radiance assimilation. The Atmospheric Infrared Sounder (AIRS) instrument on the NASA EOS-Aqua satellite (http://www-airs.jpl.nasa.gov) extends the capabilities by replacing the channels of HIRS by a high-resolution spectrum. An other example is TES on EOS-Aura, which is able to provide ozone profile information in the troposphere (Aura special issue 2005).

3. *Limb measurements*
   The limb geometry has the important advantage that it allows for the retrieval of stratospheric ozone profiles. A disadvantage is the relatively low horizontal resolution as compared to nadir measurements. Several instruments on board of NASA-UARS (Upper Atmosphere Research Satellite) measured stratospheric ozone profiles (http://umpgal.gsfc.nasa.gov/uars-science.htm). In particular the Microwave Limb Sounder (MLS) has created a long record of ozone profile measurements which has been used in data assimilation by several groups. A successor of this instrument is MLS on EOS-Aura (Aura special issue 2005). The Swedish satellite ODIN (http://www.ssc.se/ ssd/ssat/odin.html) has two instruments that measure ozone profiles, namely the UV-Vis spectrometer OSIRIS (Odin Spectrometer and InfraRed Imager System) and a microwave radiometer SMR (Sub-Millimeter Receiver). MIPAS (Michelson Interferometer for Passive Atmospheric Sounding) on Envisat derives trace gas profiles from the infrared spectra. Apart from the nadir mode, SCIAMACHY measures profiles of ozone in limb.

4. *Occultation*
   In occultation the extinction of radiation due to the presence of trace gases is measured during sunset and sunrise. The UARS-HALOE (Halogen Occultation Experiment), SAGE (Stratospheric Aerosol and Gas Experiment) and POAM (Polar Ozone and Aerosol Measurement) instruments use this technique. This technique is generally regarded to provide accurate ozone profile measurements. A disadvantage is the

small number of measurements made. The data sets are important to validate other satellite retrievals. GOMOS (Global Ozone Monitoring by Occultation of Stars) on Envisat uses stars instead of the sun, which provides a much better coverage than the solar occultation instruments. SCIAMACHY measures ozone profiles with solar occultation.

New satellite missions, e.g. NPOESS/NPP of NASA (http://jointmission.gsfc.nasa.gov/), METOP of EUMETSAT (http://www.eumetsat.int/), will continue and enhance this ozone observing capability. The ozone information available is complemented by networks of ground stations, in particular WMO-GAW (Global Atmosphere Watch) and NDSC (Network for the Detection of Stratospheric Change). The long-term measurement series of these networks are crucial to validate/calibrate the existing satellite retrievals and to derive climatological ozone data sets.

Compared to the large amount of available satellite measurements, the corresponding ozone data assimilation efforts have been very modest. An increased and co-ordinated effort on the assimilation of ozone observations is needed to make optimal use of the available observations.

2.1 Ozone measurements from UV-Vis nadir spectrometers

The TOMS and SBUV instruments have been monitoring the ozone layer since 1979. These nadir UV-Vis instruments use the strong absorption of ozone in the UV to derive ozone columns and stratospheric profiles. From 1995 Europe is contributing to this global monitoring of ozone with GOME (on ERS-2, since 1995) and SCIAMACHY (on Envisat, launched in 2002). In 2004 the Dutch-Finnish Ozone Monitoring Instrument (OMI) was launched as part of the NASA EOS-AURA satellite. These measurement series will be continued on an operational basis with the GOME-2 instruments (2005-2020) and with OMPS on NPOESS.

The GOME instrument is part of the payload of the ERS-2 satellite of ESA. A discussion of the instrument, the ozone products and retrieval techniques can be found in Burrows *et al.,* (1999). The advantage of GOME over the TOMS instruments is that it measures a detailed spectrum including the ultraviolet and visible (240-790 nm). Apart from the total column, this spectral information allows the retrieval of nadir profiles (about 5-6 pieces of profile information) and cloud parameters which are needed for accurate ozone retrievals. GOME has a global coverage in three days (apart from the dark winter pole). GOME has provided global ozone measurements from 1995 until June 2003. In June ERS-2 experienced a breakdown of the tape that records the GOME data. At the time of writing GOME data is only available for limited areas where the measurements can be transmitted directly to a receiving station on the ground.

Sciamachy extends the measurement capabilities of GOME with matching nadir and limb modes (Bovensmann *et al.*, 1999) for detailed profiling from the surface to the top of the atmosphere. The main advantages of OMI compared to GOME and SCIAMACHY are the small pixel size and global coverage in one day (Levelt, 2002).

For use in numerical weather prediction models and for ozone forecasting purposes a near-real time product is essential. Currently the ECMWF IFS is assimilating SBUV-2 observations and SCIAMACHY ozone columns retrieved by the TOSOMI algorithm (Eskes *et al.,* 2005).

The recent retrieval algorithm developments for GOME and SCIAMACHY (e.g. the TOSOMI algorithm), and similar developments for TOMS (version 8 of the TOMS processor) demonstrate that UV-Vis satellite instruments have the potential for high-quality retrievals of the total column with accuracies of a few percent and with low noise (high precision). Such accuracies are of considerable importance for trend analyses and ozone assessments based on multi-year satellite ozone measurements.

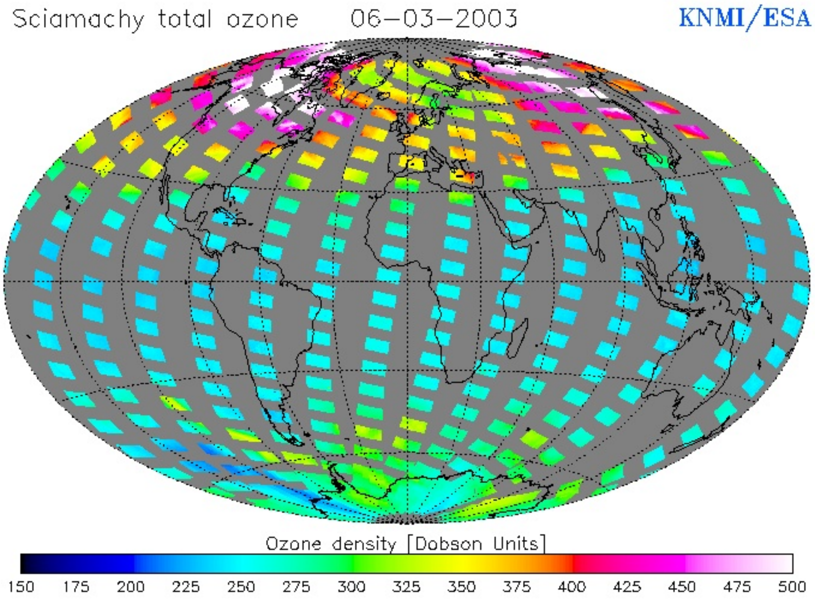

Figure 1: One day of SCIAMACHY total ozone observation, for 6 March 2003. Column in Dobson units.

3 Ozone data assimilation

Ozone data assimilation work is now performed at several institutes. In this section I will use results of the KNMI ozone analyses to illustrate the performance of such ozone data assimilation systems.

Shortly after the 60 layer stratosphere-troposphere version of the ECMWF IFS became operational in October 1999, the KNMI has started to produce ozone analyses and forecasts based on the GOME near-real-time measurements. A tracer transport model with a simplified chemistry was developed. Daily ozone runs, based on ECMWF wind fields, are performed directly after completion of the IFS forecast run. The service has been operational in the period 2000-2003.

Similar developments have occurred at other centres. In particular the ECMWF model has been extended with an ozone tracer field (Hólm *et al.*, 1999) and ozone data (SBUV, GOME) are assimilated operationally. The use of Envisat MIPAS ozone profiles in the ECMWF data assimilation system is now under investigation (A. Dethof, private communication). Starting early 2000, the NASA Data Assimilation Office have assimilated Earth Probe TOMS and SBUV/2 data on an operational basis in a transport model driven by the Goddard Earth Observing System Data Assimilation System (GEOS-DAS), see Riishøjgaard *et al.* (2000), Štajner *et al.* (2001).

3.1 The model

The KNMI ozone analyses and forecasts are based on a tracer-transport and assimilation model called TM3-DAM. The modelling of the transport, chemistry and the aspects of the ozone data assimilation are described in Eskes *et al.*, (2003a). Here we will only provide a brief overview of the model set-up. A state of the art treatment of ozone chemistry in the stratosphere involves the explicit treatment of many chemicals (typically 50 or more) and the description of heterogeneous chemical reactions on ice particles. Such models are computationally expensive, and for applications such as numerical weather prediction simplified parameterized ozone chemistry schemes have been introduced.

Ozone chemistry in our model is described by two parameterizations. One follows the work of Cariolle and Déqué (1986) and consists of a linearization of the chemistry with respect to sources and sinks, the ozone amount, temperature and UV radiation. The Cariolle scheme has been used by several groups involved in ozone data assimilation and/or forecasting, including the ECMWF. A second parametrization scheme accounts for heterogeneous ozone loss (P. Braesicke, private communications). This scheme introduces a three-dimensional chlorine activation tracer which is formed when the temperature drops below the critical temperature of polar stratospheric cloud formation. Ozone breakdown occurs in the presence of the activation tracer.

The three-dimensional advection of ozone is described by the flux-based second order moments scheme of Prather. The model follows the new ECMWF vertical layer definition, operational from the end of 1999 until the present. The 60 ECMWF hybrid layers between 0.1 hPa and the surface have been reduced to 44 in TM3-DAM by removing 16 layers in the lower troposphere. The horizontal resolution of the model version discussed here is 2.5 degree. The model is driven by 6-hourly meteorological fields (wind, surface pressure, temperature) from the ECMWF model.

Note that the largest changes in ozone on the time scale of one day to a week are related mainly to transport. Even the dramatic ozone depletion occurring at the South Pole in August–September has a time scale of a week to a month. This should be compared with the 1 to 3 days within which new satellite measurements become available to the assimilation. The parameterizations remove a large part of the bias the model would have without any chemistry, and ensure that the ozone profile shape remains realistic.

On the other hand it is important to improve the simple description of ozone chemistry as scetched above. A persistent bias will have a detrimental effect on the analysis: it may for instance lead to unrealistic vertical profile shapes or to unrealistic increments in other model variables such as the wind field. More realistic parametrisations or on-line modelling of the chemistry are possible approaches.

3.2 Assimilation results

The total ozone data is assimilated in TM3-DAM based on a sub-optimal Kalman filter technique (Eskes *et al.*, 2003a). This approach retains several aspects of the Kalman filter equations by implementing a time-dependent error covariance matrix, and allows the scheme to produce a detailed forecast error estimate.

An example of an ozone analyses based on the Fast Delivery GOME ozone columns is shown in Fig. 2, second panel. For comparison, we show an Earth Probe TOMS (McPeters *et al.*, 1998) map of ozone on 15 April 2001 in the third panel. TOMS has a nearly global coverage in one day, and the figure shows the ozone column observations gridded on a 1 by 1.25 degree grid. Because TOMS has a sun-synchronous orbit, we have constructed a 12 h local time global ozone map based on the model analysis (second panel). The date line is clearly visible at 180 degree longitude in both frames.

The small-scale features in ozone in the assimilated GOME fields correlate very well with the small scale features in the TOMS map. The image provides an impression of the level of detail in the assimilated ozone fields and the effective resolution of the model. It also demonstrates the ability of the model and the underlying ECMWF wind fields to produce realistic dynamical ozone features. On a larger scale there are also clear differences. The results are based on retrievals of several years ago (TOMS version 7 and the KNMI GOME fast delivery algorithm). Closer agreement is observed with the latest versions of the TOMS and GOME algorithms.

The observation minus forecast statistics is discussed in more detail in Eskes *et al.*, (2003a). On average the root-mean-square (RMS) observation-minus-forecast difference between GOME Fast Delivery observations (before assimilation) and the short range model forecast (between 1 and 3 days) is small: about 9 Dobson Units (DU), or roughly 3%. The bias between the model forecast and the GOME fast-delivery ozone columns is in general smaller than 1%.

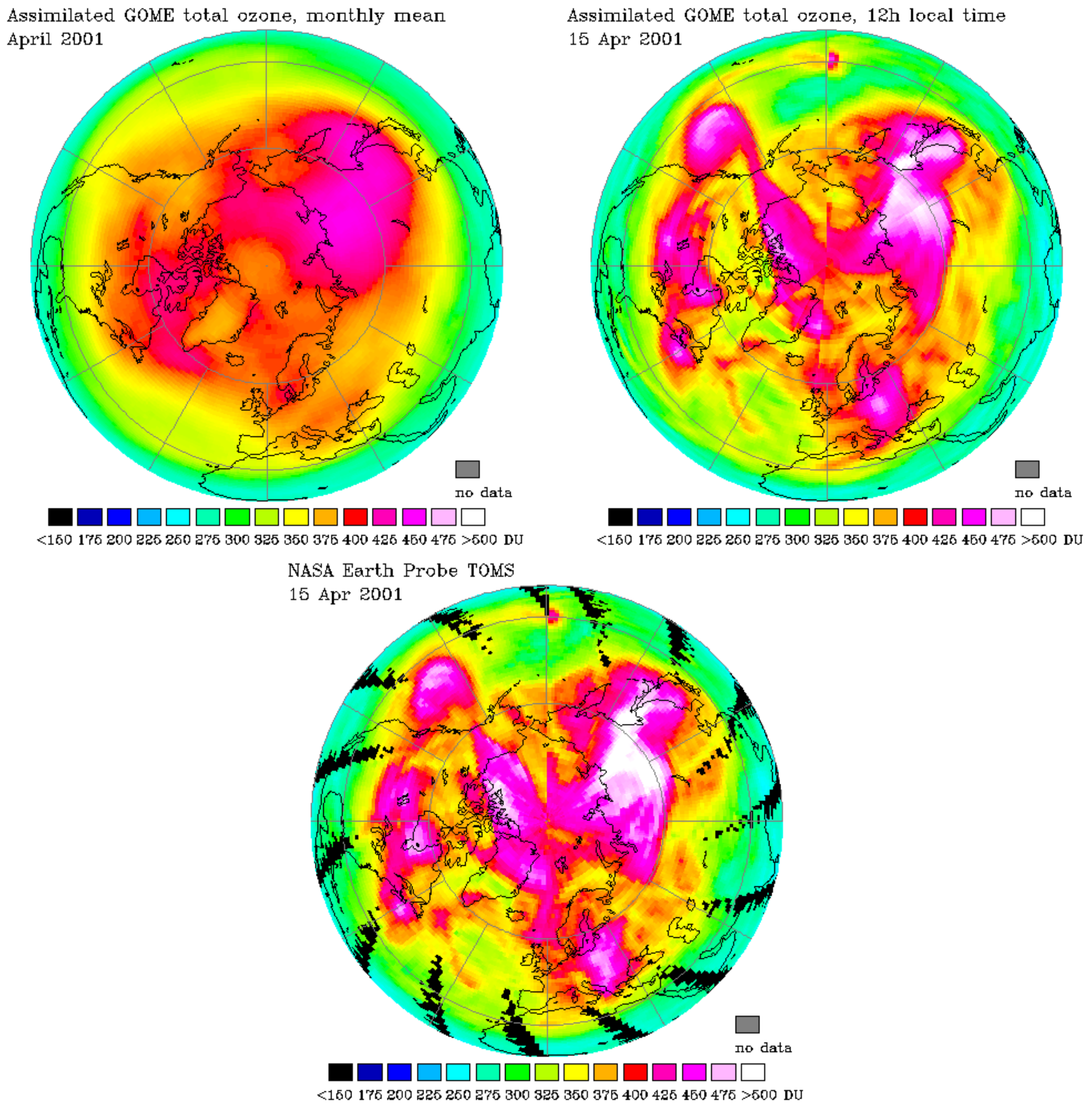

Figure 2: Total ozone distribution in the Northern hemisphere in April 2001. Left: monthly mean. Right: Analysis on 15 April, 12:00 LT. Bottom: Earth Probe TOMS observations for 15 April. Scales in Dobson units.

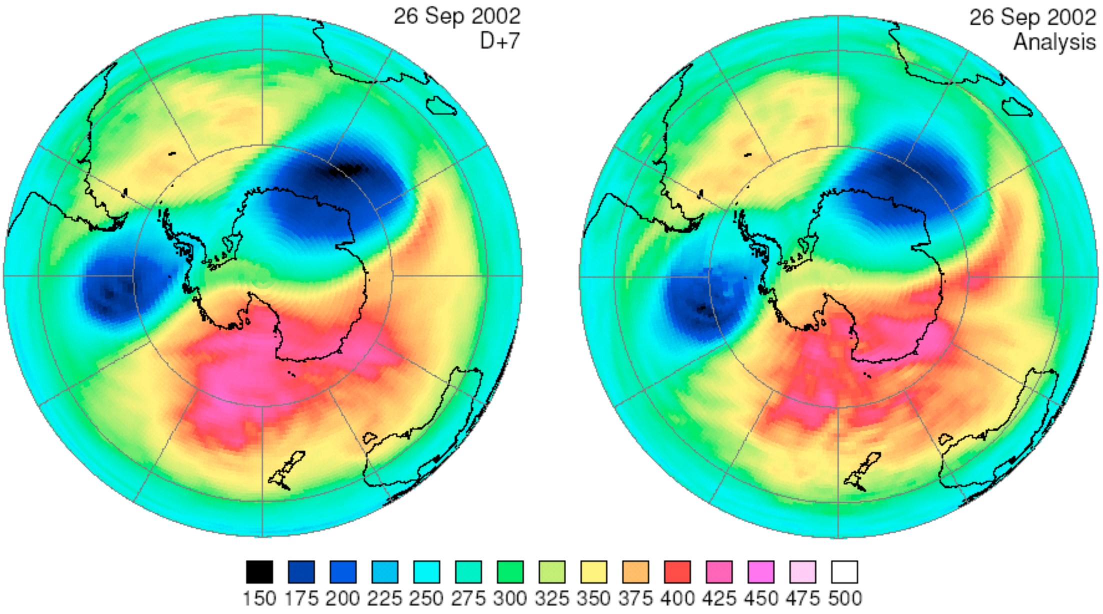

Figure 3: The ozone distribution on 26 September 2002. Left: 7-day forecast. Right: analysis based on GOME data. Scale in DU.

4 Ozone forecasts

Soon after the 60-layer model version of the ECMWF became available (October 1999), ozone forecasts were produced on a routine basis at the KNMI, based on the 10-day ECMWF forecast meteorology. The ozone forecasts are disseminated via the TEMIS project web site, http://www.temis.nl/.

The performance of the forecast system has been discussed in Eskes *et al.* (2002). Anomaly correlations and rms errors were computed for the total ozone columns. A modified anomaly correlation was introduced in which the anomalies are computed as the difference between the actual ozone column and a centred (running) monthly mean. An example of this is shown in Fig. 2: the anomaly for this day is the difference between the field in the right panel and the monthly mean shown in the left panel.

On average the ozone forecasts are found to be meaningful up to 6–7 days in the extra-tropics. The current ECMWF meteorological forecasts are characterised by 500 hPa geopotential height anomalies that cross 0.6 after about 7 days (see ECMWF technical reports), which is quite comparable to what we find for total ozone. Note that this crossing time is sensitive to the choice of the climatological reference (in our case a running monthly mean), and this dependence is one of the factors which complicates the direct comparison between the total ozone and height anomalies.

The rapid formation of the ozone hole in August/September over the South Pole, and the recovery in later months is related to an interplay between heterogeneous chemistry and dynamics (e.g. UNEP/WMO, 2002; Solomon, 1999). The stability of the polar vortex plays a crucial role during the later stage of the ozone hole. In this respect the year 2002 was very exceptional, and the forecast of this event is a good example of what present day weather forecast models can achieve.

At the end of September 2002 the Antarctic underwent a major stratospheric warming in which the polar vortex split up into two parts (Baldwin *et al.*, 2003) in a manner similar to the wavenumber-2 strong stratospheric warmings which occur in northern winter. As a result of this unprecedented event the ozone hole split into two parts. The separated ozone hole parts moved toward areas experiencing more sunlight, and one of them moved toward South America. The major warming was accompanied by a temperature enhancement of several tens of K and a zonal wind reversal at 60 S and 10 hPa similar to a northern hemispheric major stratospheric warming.

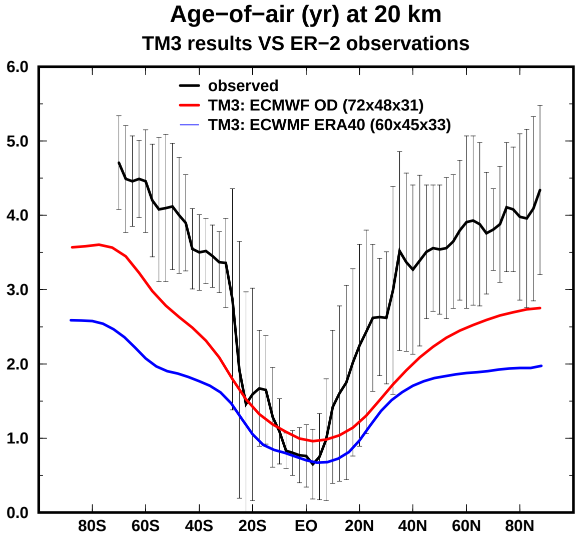

Figure 4: Observed mean age of air at 20 km altitude, compiled from all ER-2 CO_2 data from 1992 and 1998 (solid line), including the error bars ($\pm 2\sigma$) (Andrews et al., 2001). The model results are shown by the red line (winds from the operational ECMWF model) and blue line (winds from the ERA-40 reanalysis).

Global numerical weather prediction (NWP) models have been remarkably successful in accurately predicting the wind and geopotential height field during stratospheric warming events including the 2002 split-vortex event (e.g. Simmons *et al.*, 2003) in the medium-range 10-day forecasts. This success in predicting polar warming events is confirmed by our ozone forecasts, which are based on the ECMWF operational medium-range weather forecasts (Eskes *et al.*, 2003b). The various ozone forecasts produced around September 18-21 were all very consistent with each other. This is demonstrated in Fig. 3 which shows the 7-day forecasts and the analysis for 26 September.

4.1 Brewer-Dobson circulation and age of air

Numerical weather prediction models are especially well tested for changes that occur on the medium-range, a few days to a few weeks. For atmospheric chemistry the range of relevant time scales is much broader, from seconds up to years. Especially the concentration of long-lived reservoir species is crucial to understand and model the chemical composition. As a result the modelling of mixing barriers (troposphere-stratosphere, northern-southern hemisphere, tropics-subtropics) and the residence times of these species is a crucial issue to study.

Model experiments have been performed (e.g. Bregman *et al.*, 2003) to evaluate the impact of the choice of wind field on long-lived tracer transport. The age spectrum (Hall and Plumb, 1994) is obtained from a passive tracer simulation where the mixing ratio in a small tropospheric volume is set equal to a delta-function in time. One result of simulations based on ECMWF operational winds and ERA-40 winds are shown in Fig. 4.

The figure shows a significant difference between the two meteorological input fields. The ERA-40 runs show a shorter age and smaller gradients between the tropics and extra-tropics. This indicates faster transport to mid-latitudes in the lower stratosphere. Furthermore, we have investigated the dependence of the ozone flux from the stratosphere to the troposphere in the tropospheric chemistry model TM3, and find that the flux is nearly a factor two larger than runs driven by operational ECMWF meteorology (van Noije *et al.*, 2005).

It is interesting to compare these results with a recent paper by Shoeberl *et al.* (2003). This study discusses trajectories driven by different assimilation models and by the corresponding GCM. It is found that the wind fields from assimilation models show too much exchange from the tropics to mid-latitudes. When the same model is run as a GCM (without data assimilation) this exchange and the age spectrum improve and compare

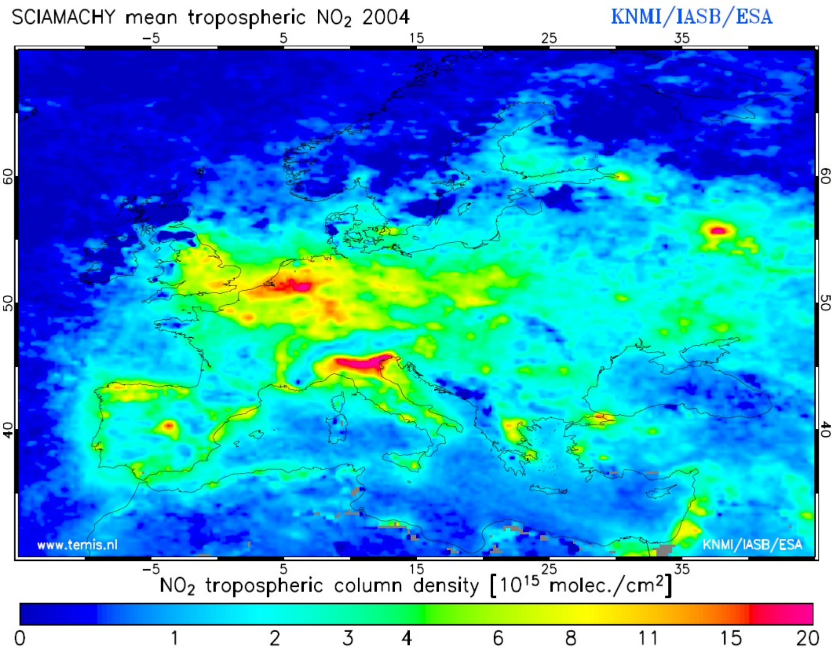

Figure 5: SCIAMACHY measurements of tropospheric columns of NO_2 *over Europe. Yearly-mean for 2004.*

much better with observations. Our findings based on ECMWF meteo are qualitatively consistent with this picture.

A very recent experiment conducted at ECMWF has focussed on this aspect. Especially the biases that exist between the model and satellite observations of temperature have been addressed more carefully (A. Simmons, private communication). The resulting age-of-air spectrum in this experiments was improved considerably and is much more in line with observations. This new result suggests that a careful treatment of data assimilation is a key to improve the residual circulation in weather models.

4.2 Tropospheric trace gases and aerosols

Dedicated satellite missions for atmospheric chemistry, in particular the NASA UARS mission, have focussed on the stratosphere. This attention for the middle atmosphere is related to the ozone layer depletion problem which became very pressing after the discovery of the ozone hole around 1984 leading to the Montreal protocol agreement. Limb and occultation satellite viewing geometries are especially well suited to probe trace gas profiles above about 10 km.

Satellite measurements of the tropospheric composition have become available only quite recently (see, e.g. the final report of the TROPOSAT project, Borrell *et al.*, 2003; the IGACO-IGOS report). Nadir-viewing instruments are most suitable for this purpose. Different wavelength ranges are exploited. The infrared provides good sensitivity in the middle troposphere and offers the possibility to obtain profile information in the troposphere. The near-infrared wavelength range involves scattered sunlight and provides a good sensitivity at the surface. Total columns of trace gases like CO, CH_4, CO2, can be measured at these wavelengths. The visible and near-UV can be used to retrieve trace gas columns like ozone and NO_2.

The retrieval of tropospheric trace gases is complicated. This is due to the presence of clouds, aerosols and complicated surface properties. Furthermore, the derived trace gas amount is often sensitive to the assumed *a priori* vertical distribution of the tracer. An other complication is the often limited availability of validation measurements (e.g. from the ground). Due to these complications the retrieval of for instance NO_2 columns from GOME or SCIAMACHY is currently characterised by errors of the order of 30-60% for polluted areas. Further improvements of the retrieval schemes and dedicated validation campaigns are challenges for the future.

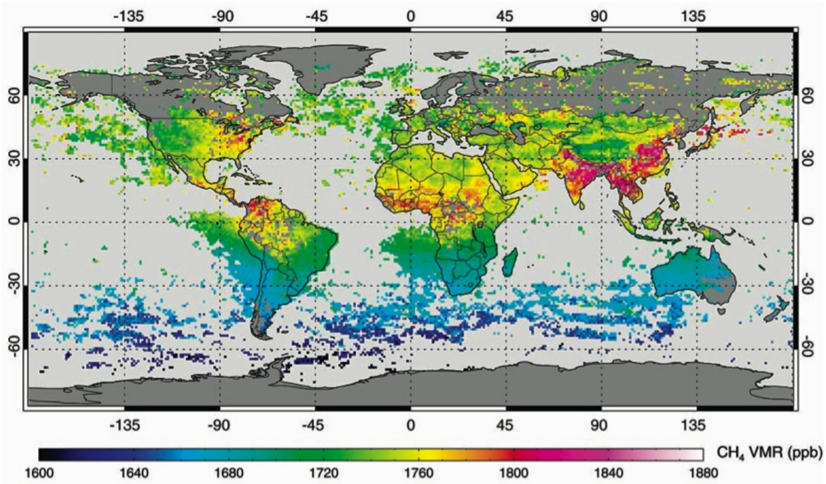

Figure 6: SCIAMACHY measurements of the column-averaged mean mixing ratio of CH_4. From Frankenberg et al., 2005.

Several key chemical species in the troposphere are presently measured from space. Examples are:
Carbon monoxide (CO). Sensors: MOPITT, AIRS, TES, SCIAMACHY.
Methane (CH_4). Sensors: SCIAMACHY.
Nitrogen dioxide (NO_2). Sensors: GOME, SCIAMACHY, OMI.
Sulphur dioxide (SO_2). Sensors: TOMS, GOME, SCIAMACHY, OMI.
Formaldehyde (CH_2O). Sensors: GOME, SCIAMACHY, OMI.
Ozone (O_3): Sensors: TES, TOMS, AIRS, GOME, SCIAMACHY, OMI.

This list of 6 species contains key trace gases for tropospheric chemistry, and most of them are subject to air-quality health regulations (O_3, NO_2, SO_2, CO). Because of this, and because of the availability of satellite observations, the GEMS reactive trace gas sub-project will first of all focus on these 6 species.

At the time of writing there exist only a limited number published data assimilation and inverse modelling studies of tropospheric chemistry based on satellite observations. These activities have focussed on MOPITT CO especially. The satellite data sets mentioned above have only recently become available or have been improved considerably over the past few years. It will be a major challenge for the coming years to integrate these satellite data sets with models by means of data assimilation techniques, and to learn about trace gas emissions and chemical and physical processes in the atmosphere. A recent observing system simulation experiment performed for SCIAMACHY methane measurements (Meirink *et al.*, 2005) suggest that a 4D-Var data assimilation approach with simultaneous concentration and emission optimisation is a powerful tool to extract information from the satellite measurements.

5 Conclusions and challenges

A short overview was given of ozone data assimilation, ozone satellite observations and tropospheric trace gas satellite observations, with a focus on aspects related to numerical weather prediction. The main conclusions and challenges are:

1. Ozone is the best documented chemical in the atmosphere. A large number of satellite instruments measure ozone, and these observations are complemented by long-term ozone records from ground stations. In comparison to this the ozone data assimilation activities with both chemistry transport models and

numerical weather prediction systems have started only quite recently. An extension of these ozone data assimilation activities is crucial to make optimal use of these valuable data sets.

2. A reliable long-term ozone data set is important to document the development and recovery of the ozone layer. The NASA TOMS measurements have been crucial to monitor ozone over the past 25 years. The European UV-visible spectrometers GOME, SCIAMACHY, OMI and GOME-2 play an important role in the continuation of this data set.
3. Recent improvements in the total ozone algorithms for GOME, SCIAMACHY, OMI and TOMS suggest that accuracies of few percent are achieved.
4. There is still a need for reliable tropospheric ozone data sets. Extracting information on tropospheric ozone from the existing data sets remains a challenge.
5. Most ozone assimilation work has been univariate. The assimilation of ozone will influence short and long wave radiation, retrievals and the wind field. A few initial studies indicate that in a multivariate approach there is a considerable impact on the winds (vorticity), and observations of ozone (in the stratosphere) may even be beneficial for the tropospheric weather forecast and e.g. storm forecasts. However, such impacts are very sensitive to biases and a realistic model and high-quality observations are required for a successful multivariate ozone assimilation. Setting up a well-balanced bias-free multivariate ozone assimilation and demonstrating a positive impact of ozone observations for the NWP wind field is a challenge for the future.
6. The stratospheric wind fields of numerical weather prediction models like the ECMWF IFS are already sufficiently accurate to describe the synoptic scale features of ozone in considerable detail. Root-mean square differences between the KNMI ozone assimilation and total ozone observations are about 3% on average, demonstrating the good agreement between modelled and measured ozone column anomalies.
7. The first stratospheric ozone (and clear-sky surface UV) forecast results are very encouraging, with forecast scores comparable to those of the 500mb height field. Extreme events such as ozone mini-holes over Europe or the break-up of the Antarctic ozone hole are well captured by the forecasts.
8. Finding efficient but accurate ways to represent stratospheric ozone chemistry is a challenge. An improved description of the chemistry (with respect to the simple parametrisations) could reduce the model bias and is of importance for instance for the multivariate coupling. Within GEMS the initial approach chosen on this point is a tight coupling between IFS and a comprehensive chemistry-transport model.
9. A realistic description of the slow residual stratospheric circulation and stratosphere-troposphere exchange is of central importance for atmospheric chemistry modelling. Assimilation models typically show a too small age of air and corresponding overestimation of the mixing between tropics and extratropics, most probably related to the assimilation process. It is a major challenge for NWP models to identify the problems and improve the model and assimilation approach.
10. New satellite instruments, new retrievals and data sets have become available for tropospheric trace gases in the past couple of years. This includes CO, CH_4, NO_2, CH_2O, SO_2 which play a central role in atmospheric chemistry and air pollution. New instruments like OMI have a high coverage and small pixels, which allows a day-to-day monitoring of air quality and of individual events such as fires and volcanoes. The improvement of the retrievals remains a challenge.
11. Only a few data assimilation/inverse modelling studies exist at this moment to exploit these tropospheric trace gas satellite observations, and more work is needed. A combined state and emission 4D-Var is a promising approach to analyse the satellite observations of tropospheric tracers.

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