Stratospheric ozone: satellite observations, data assimilation and forecasts

Henk Eskes

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

ABSTRACT

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 very valuable for atmospheric chemistry research, protocol monitoring and NWP. This paper provides a short overview of satellite ozone observations, ozone data assimilation and ozone forecasting. The focus will be on aspects related to numerical weather prediction. The performance of the KNMI ozone assimilation system is discussed in more detail.

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 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).

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. One of the first studies is described by Levelt *et al.* (1996) where TOVS ozone data is assimilated with a simple 2D global model. This work was extended by Eskes *et al.* (1999), who discussed the implementation of 4D-Var and used the approach to assimilate GOME ozone column data. Levelt *et al.* (1998) discusses the assimilation of MLS ozone data with the ROSE chemistry-transport model. 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, and the direct influence on the wind field when ozone observations are assimilated with a modern assimilation system (4D-Var, Kalman).

The strong correlations between total ozone and vorticity has been discussed by several authors (e.g. Vaughan and Price, 1991; Allaart *et al.*, 1993). These correlations can be used in the formulation of the background covariance matrix in data assimilation, or total ozone observations may be interpreted as vorticity observations. A recent study (Jang *et al.*, 2003) is based on such a linear relation between ozone columns and mean potential vorticity. This case study shows that the additional TOMS data was beneficial to improve the forecast of the winter storm of 2425 January 2000 at the East Coast of the USA. This indicates that ozone data contains valuable meteorological information, which may be of use to improve cyclone predictions.

A more direct way of exploiting meteorological information contained in ozone observations is by assimilation into a 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). This approach included correlations between ozone and vorticity in the background covariance matrix.

The experiments showed that both the 4D-Var coupling between ozone and wind, and the background coupling have a substantial impact on the system, comparable with other measurements. However, one puzzling result was that the analysis increment patterns related to the background correlation and the 4D-Var process were quite different.

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 an overview 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).

1. *Nadir viewing UV-Visible spectrometers.*
   High quality, detailed information of the ozone distribution is available on an almost continuous basis from 1979. 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).

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 operatinal 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.

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. 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. EOS-AURA of NASA, METOP of EUMETSAT) will continue and enhance this 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) will be launched as part of the NASA EOS-AURA satellite. This measurement series will be continued on an operational basis with the GOME-2 instruments (2005-2020).

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 are the small pixel size and global coverage in one day (Levelt, 2000).

For use in numerical weather prediction models and for ozone forecasting purposes a near-real time product is essential. Based on this realisation ESA has funded the development of a GOME fast delivery total ozone column product at the KNMI (Van der A *et al.*, 2000). This fast delivery product has been assimilated in the ECMWF IFS. The ozone assimilation work discussed in the remaining chapters are based on these fast-delivery total ozone observations and on the GOME Data Processor (GDP) version-3 ozone columns (Spurr *et al.*, 2002).

In general the comparison between the GOME fast-delivery retrieval and ground-based observations is within 5% for low- and mid-latitudes, and within 10% at high latitudes and high solar zenith angles (Valks *et al.*, 2003). The inter-comparison of GDP3 against ground-based observations and retrieval error sources are discussed in the ESA report (Lambert, 2002). The performance of the DOAS retrievals for ozone, and the causes of the observed biases is still a topic of debate.

A new ozone column algorithm for GOME has recently been developed at the KNMI. This algorithm is based on the OMI DOAS algorithm developed by P. Veefkind (Levelt *et al.*, 2000), and implements several innovations compared to the fast-delivery code. A new DOAS fit formula has been developed which implicitly accounts for the Ring effect (Johan de Haan, private communications). Compared to the conventional approach this treatment of Ring leads to larger columns by up to 10 % for large solar zenith angles and large surface albedos. Initial validation results indicate a considerable improvement of the inter-comparison with ground based observations as compared to the fast delivery code (Pieter Valks and Dimitris Balis, private communications). Work is ongoing to apply this new retrieval approach to Sciamachy.

The recent retrieval algorithm developments, 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.

3 Ozone data assimilation

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. In the chapters below we will discuss some results of this ozone assimilation system.

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 a recent paper (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 very 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 GOME 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.

3.2 Assimilation approach and forecast error modelling

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 and allows the scheme to produce a detailed forecast error estimate.

The computational problem of the evaluation of the error covariance matrix in the Kalman filter equations is avoided by fixing the spatial forecast error correlations. The forecast error covariance matrix is written as a product of a homogeneous and isotropic distance-dependent correlation function, and a time and space dependent error field. The distance dependence of the correlations is determined by studying the correlations between the observation minus forecast departures. The diagonal elements of the covariance are determined by the following procedure:
1) When new observations are incorporated in the model, the forecast error at, and near, the observation is reduced as prescribed by the Kalman filter. The spatial extent of this forecast error reduction is determined by the forecast error covariance correlation length.
2) A model error contribution is added to the forecast variance every time step. The size of the model error is fixed by investigating the observation minus forecast departures as a function of the forecast time. The observed time dependence of the forecast error is described (fitted) by a functional shape with one free error

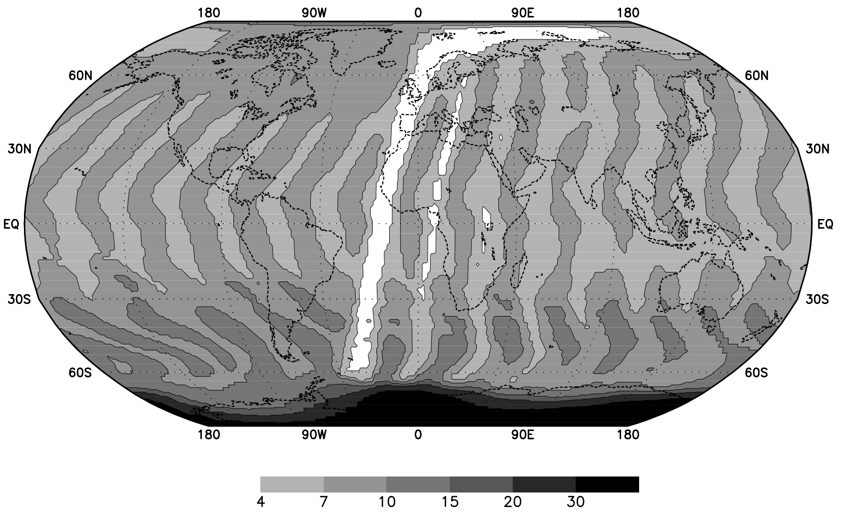

Figure 1: Forecast error distribution for the total column (in Dobson units) for August 8, 2000, 12 GMT.

growth parameter. This model error parameter is defined to be a function of latitude and month, and runs have been performed for 12 months to fix these parameters.
3) The forecast error in a pure transport problem is conserved in the Lagrangian sense. Based on this principle the three-dimensional forecast error field is advected with the transport model as a (passive) tracer.

An example of the forecast error distribution for the total column obtained with this approach is shown in Fig. 1. A first obvious feature of this distribution is the vertical bands which reflect the GOME tracks. The three aspects of the error modelling are clearly visible. First, the latest GOME track has the smallest errors because new information is added to the model which reduces it's uncertainty. Second, the larger the time difference between the current time and the over-pass time of GOME, the larger the forecast error. Third, due to the advection of the forecast error the vertical bands obtain distorted shapes.

As shown by the figure, the procedure provides a detailed time and space dependent forecast error distribution. It is easy to check whether this distribution is realistic. Together with an estimate of the observation error, the forecast error provides an estimate of the observation minus forecast departure. In figure 2 this predicted departure is plotted against the observed departure. The results are quite satisfactory: in a statistical sense the predicted small forecast errors correspond to actually observed small departures.

3.3 Assimilation results

An example of an ozone analyses based on the Fast Delivery GOME ozone columns is shown in Fig. 3, 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 agreement between TOMS and the TM3-DAM assimilated GOME field is good. The small-scale features in ozone correlate very well with the small scale features in the TOMS map. The image provides an impression of the amount of detail in the assimilated ozone fields and the effective resolution of the model. On a larger

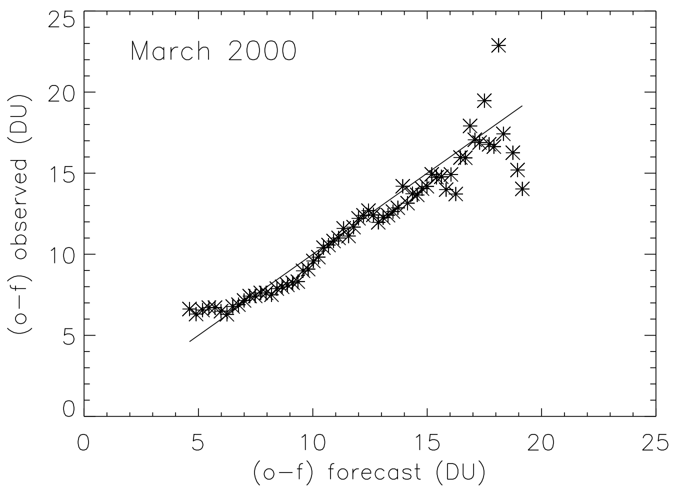

Figure 2: Observed GOME minus forecast departure as a function of the predicted GOME minus forecast departure.

scale there are also clear differences. This larger-scale offset between the two plots can for a large part be attributed to differences in the instruments and the retrieval codes for TOMS and GOME total ozone.

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 distribution of RMS differences is well approximated by a Gaussian curve, which is consistent with the Kalman filter assumption of Gaussian observation and forecast errors. The bias between the model forecast and the GOME fast-delivery ozone columns is in general smaller than 1%, a factor of 3-10 smaller than the RMS. Because of this, no additional bias corrections are applied to the forecast model. The small bias implies that the assimilation efficiently adopts the ozone levels as retrieved from the GOME spectra. Note that this small bias does not imply that the GOME data themselves have a similar small bias (see discussion in section 2).

3.4 Dependence on the meteorological wind field

In the context of the EU GOA project (GOME Assimilated and Validated Ozone and NO_2 Fields for Scientific Users and for Model Validation) a re-analysis assimilation run has been performed for the period 1995-2003. The results are available from the GOA web site, http://www.knmi.nl/goa. This 8-year run is based on the latest GOME GDP version 3 ozone columns. The meteorological input (temperature, pressure, wind) is taken from the ECMWF 40-year reanalysis (ERA-40) for the period 1995 - October 1999, and the ECMWF operational archive for the period November 1999 - 2003.

In figure 4 we show the performance of the assimilation for the month of September in 1998 and 2000. The assimilation model set-up was identical in both cases (same pre-processing of the meteorological data, same model setup, same data set, namely GOME GDP version 3). The difference is in the use of ECMWF ERA-40 data (1998) and ECMWF operational data (2000).

There is a striking difference in the performance when comparing the two meteorological inputs. In the 1998 case the O-F root-mean-square is significantly larger. We made similar plots for the other months and for other

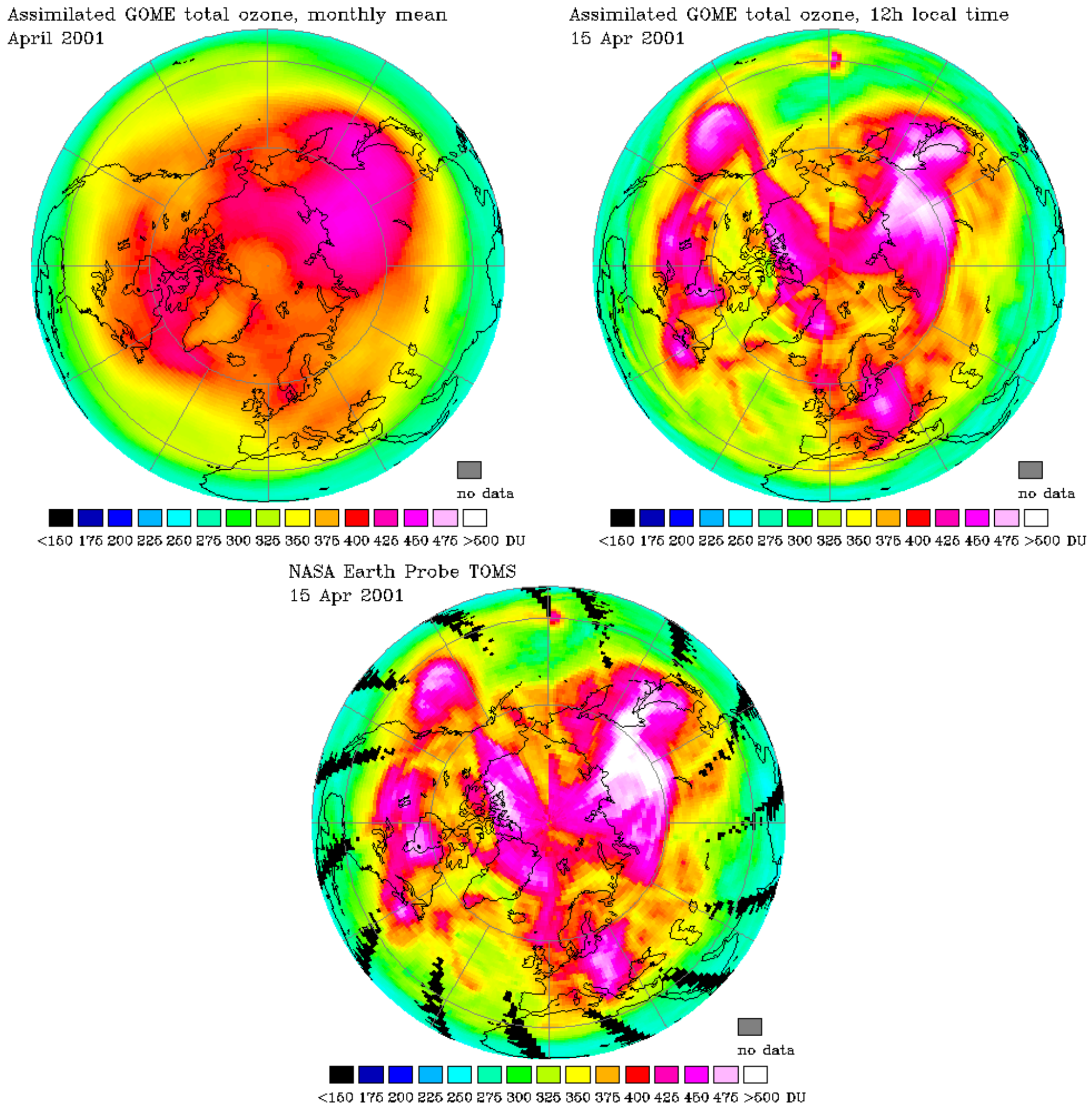

Figure 3: Total ozone distribution in the Northern hemisphere in April 2001. Left: monthly mean. Right: Analysis on 15 April, 12:00 LT. Bottom: Earth Probe TOMS observations for 15 April. Scales in Dobson units.

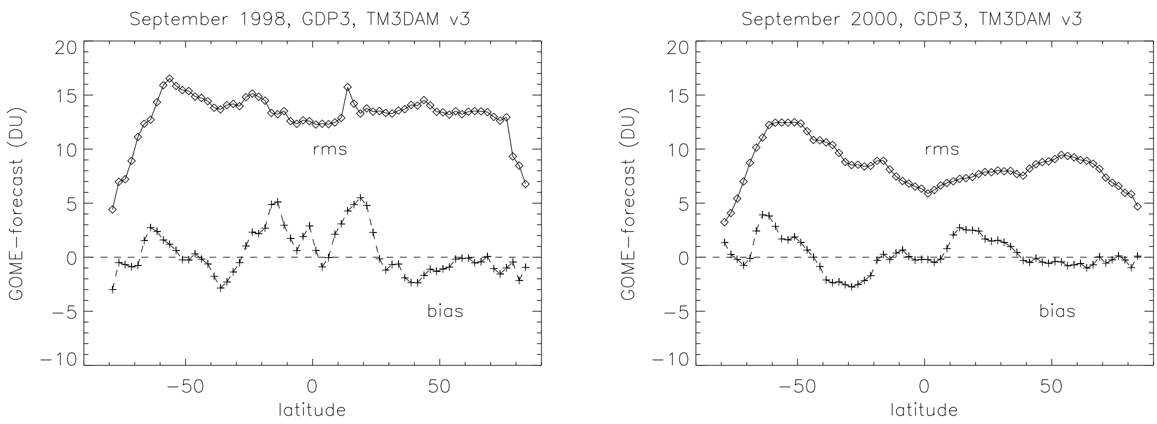

Figure 4: Observed GOME minus forecast departure as a function of latitude, for September 1998 (left) and september 2000 (right). Both plots are based on GOME GDP version 3 ozone data. The transport model is driven by ECMWF ERA-40 reanalysis (left) and with operational (right) meteorological input fields.

years, with similar conclusions. The difference in all those plots is especially pronounced in the tropics. Note that the variability in ozone is much smaller in the tropics than in the extra-tropics, and especially the ERA-40 results in the tropics are disappointing. The rms in the extra-tropics is significantly smaller than the variability, and is quite satisfactory for both operational and ERA-40 data.

A second feature is the positive bias of roughly 3 Dobson units (or 1%) at low latitudes. Again this is a features which also persists for other months and years. The positive bias implies too low ozone in the model in the tropics. Because ozone is created by sunlight mainly in the tropics, these curves suggest there is significantly enhanced transport of ozone from low to high latitudes in ERA-40 compared to the operational model.

3.5 Age of air

The residual mixing between the tropics and mid-latitudes observed in the assimilation of ozone in the previous section can be studied in a different way. Model experiments have been performed (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. The age-of-air is then the first moment of the age spectrum. The simulation was continued for 20 years, based on the repeated use of the ECMWF meteorology of a given year. One result of simulations based on ECMWF operational winds and ERA-40 winds are shown in Fig. 5.

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 that runs driven by operational ECMWF meteorology (Michiel van Weele, private communication).

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

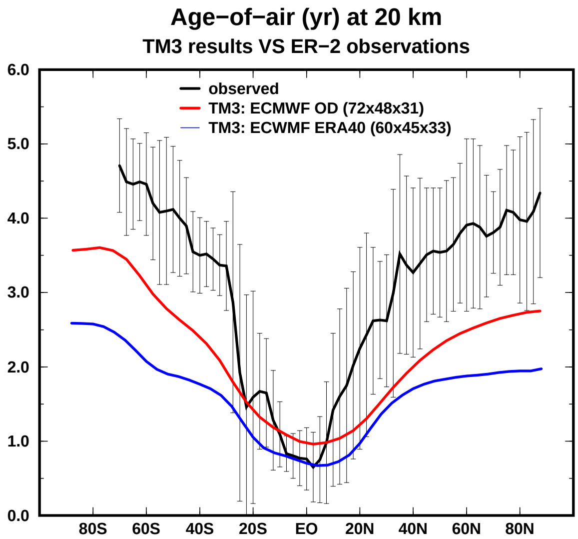

Figure 5: Observed mean age of air at 20 km altitude, compiled from all ER-2 CO_2 data from 1992 and 1998 (solid line), including the error bars ($\pm 2\sigma$) (Andrews et al., 2001). The model results are shown by the red line (winds from the operational ECMWF model) and blue line (winds from the ERA-40 reanalysis).

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

The two most pronounced differences between ERA-40 and the operational model is the difference in resolution and the use of 3D-Var versus 4D-Var. Perhaps the 4D-Var leads to more consistent analysis with smaller meridional fluxes. The work of Shoeberl suggests that also the operational data sets will show too much exchange, consistent with Fig. 5. Apart from the assimilation analysis technique, it is also important to investigate the data that enters the assimilation, i.e. the stratospheric sounder temperature data. Furthermore, problems in the pre-processing of winds for use in the transport models (Bregman *et al.*, 2003) and the numerical transport scheme used complicate the interpretation.

The issue of enhanced circulation is very relevant for atmospheric chemistry modelling and may be one of the limiting factors for realistic trace gas simulations.

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. In the period early 2000 until mid 2002 five-day forecasts were produced and the results have been made available for the scientific community via the KNMI web site (http://www.knmi.nl/gome_fd). In the summer of 2002 the forecast range has been extended to 9 days. The performance of the 5-day forecast system has been discussed in Eskes *et al.* (2002), and the results of this study will be summarised here.

The performance of the ozone forecast system has been tested with anomaly correlations and rms errors. In our case these are computed for the total ozone columns. The anomaly is normally defined as the difference between an instantaneous value and the climatological mean. This definition of the anomaly is not very useful for the total ozone column, since the mean varies considerably from one year to the next, and from one month to the next. Such ozone variations with long time scales will lead to artificially high anomaly scores. Motivated

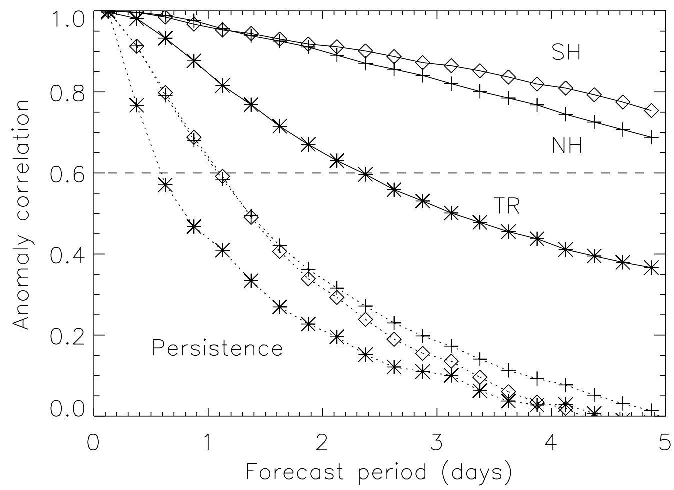

Figure 6: Modified anomaly correlation as a function of the forecast period. The top three curves represent the total ozone anomaly for latitudes north of 30 degree (NH), between −30 and 30 degree (TR) and south of −30 degree (SH). The lower three curves are the corresponding scores for persistence.

by this, we introduce a modified anomaly correlation 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. 3: the anomaly for this day is the difference between the field in the right panel and the monthly mean shown in the left panel. Note that there is a pronounced zonal variation (with pronounced wave 1 and wave 2 components) in the monthly mean map.

Figure 6 shows the modified anomaly correlation as a function of the forecast time. Plots of the rms error leads to similar conclusions (Eskes *et al.*, 2002) and are not shown here.

Extrapolation suggests that on average the ozone forecasts are 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 (A. Simmons, this proceedings), 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 c (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 plot also suggests that the southern hemisphere has a slightly better score than the northern hemisphere. The statistical significance of this is somewhat unclear, given the variability from one month to the next. The geographical distribution of the anomaly correlation shows significantly more variation in the NH than in the SH. These differences may be related to orographical effects, which are more pronounced in the NH sector.

The forecast performance is systematically lower for the tropics. The value of 0.6 is reached after about 2–2.5 days. The curves obtained show little seasonal variation: $C_t^{(m)}$ for a forecast time $t = 5$ days has a value between 0.32 and 0.42 for the individual months. There may be several reasons for this difference between the tropics and extra-tropics (see also Eskes *et al.*, 2002):

1. Ozone anomalies are very small in the tropics. Measurement noise and retrieval errors will have a larger, negative influence on the anomaly correlation.

2. Most of the ozone column variation in the tropics can be attributed to the troposphere. This is in contrast to

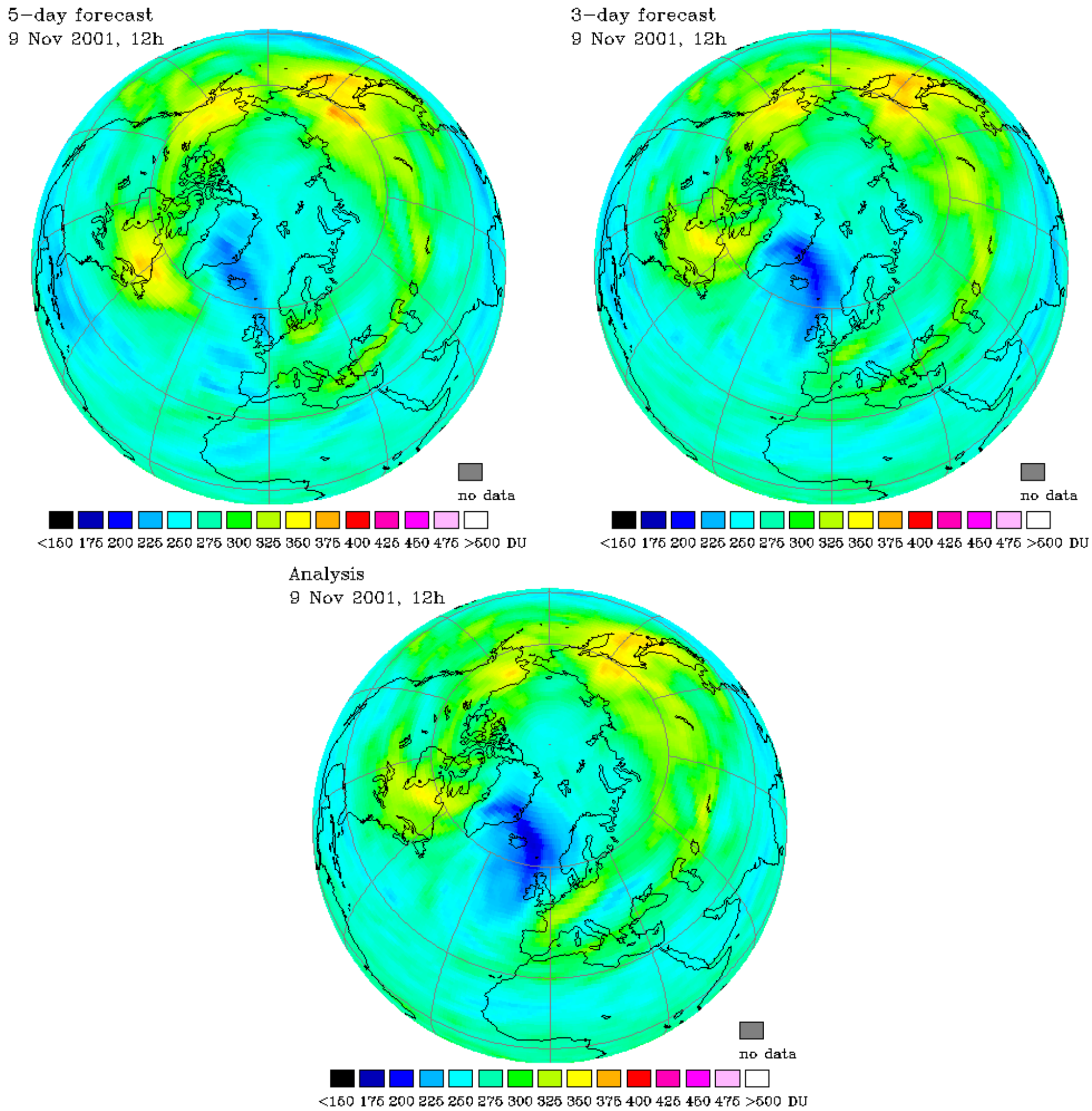

Figure 7: The first ozone mini-hole of the winter 2001–2002, on 9 November 2001. Left: 5-day forecast. Right: 3-day forecast. Bottom: Verification.

the extra-tropics, where the lower stratospheric dynamics is responsible for the bulk of the observed ozone column variability. The model gives only a very crude description of tropospheric ozone.

3. The quality of middle atmospheric winds in the ECMWF forecasts may be of lower quality that in the extra-tropics.

4.1 Ozone forecast examples

Two examples will be discussed to illustrate the potential of ozone forecasts.

Low ozone events (ozone mini-holes) over Europe have attracted considerable attention. Ozone mini-holes often occur over the Atlantic and Northern Europe. The lowest ozone values last only for about 1–2 days, and these events are mainly of dynamical origin, with transport of air with low ozone mixing ratios from the subtropics to higher latitudes.

In Autumn/Winter 2001/2002 a series of such low ozone events occurred. Figure 7 shows a forecast of the first low ozone event of this winter. The five-day forecast (first panel) predicts a thin ozone layer above Iceland and the Atlantic, in qualitative agreement with the analysis and the GOME data 5 days later. Also the ozone patterns agree well with the verifying analysis (third panel). The three day forecast (second panel) predicts even lower values at essentially the same location, and this forecast is close to the observations on 9 November with minimum ozone values below 200 DU.

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.

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.

Global numerical weather prediction (NWP) models have been remarkably successful in accurately predicting the wind and geopotential height field during the split-vortex event (e.g. Simmons *et al.*, 2003) in the medium-range 10-day forecasts. This success is related to the large scale of the event, and shows that the underlying processes are well captured by the NWP models.

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. 8 which shows three forecasts and the analysis for 26 September. The 7 and 5 day forecast are very similar to the verifying analysis. Even the 9-day forecast shows qualitatively the same development but the position of the vortex is somewhat different and the vortex has not split as pronounced as in the analysis.

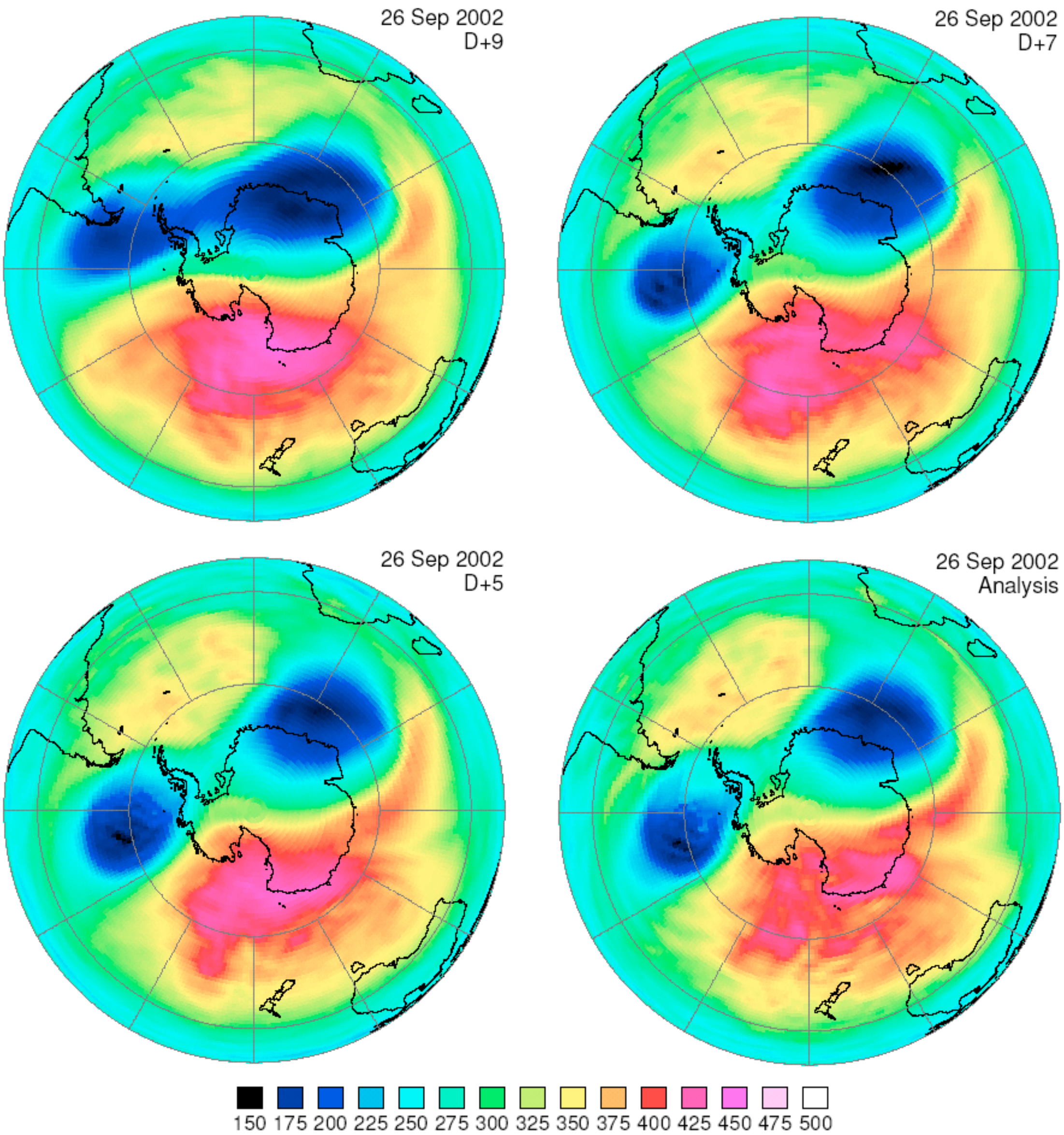

Figure 8: The ozone distribution on September 26. Top left: 9-day forecast. Top right: 7-day forecast. Bottom left: 5-day forecast. Bottom right: analysis. Scale in DU.

5 Conclusions

This paper provides a short overview of ozone data assimilation and ozone satellite observations, with a focus on aspects related to numerical weather prediction. The main conclusions 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 in numerical weather predictions have started only quite recently. An extension of these ozone 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 instruments have been crucial in the past 25 years. The European UV-visible spectrometers GOME, SCIAMACHY, OMI and GOME-2 will play an important role in the continuation of this data set.

3. The ozone retrievals are still developing. Recent improvements in the total ozone algorithms for GOME and TOMS suggest that accuracies of just a few percent are possible.

4. 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.

5. The stratospheric wind fields of numerical weather prediction models like the ECMWF IFS are sufficiently accurate to describe the synoptic scale features of ozone in considerable detail.

6. The first stratospheric ozone 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.

7. A realistic description of the residual stratospheric circulation is of large importance for atmospheric chemistry modelling. This aspect, however, is much more challenging for the NWP models than the medium-range forecast. A fundamental aspect is the observed decrease of the age of air and corresponding overestimation of the mixing between tropics and extra-tropics due to the assimilation process. The ECMWF ERA-40 reanalysis shows enhanced mixing of tropical and mid-latitude air, compared to the operational model.

Acknowledgements

This work has benefited from stimulating discussions with colleagues world wide, and from the direct input of my colleagues at the KNMI. The work described was supported by the European Union GOA project (contract no EVK2-CT-2000-00062).

References

[1] Van der A, R. J., Eskes, H. J. van Geffen, J., van Oss, R. F., Piters, A. J. M., Valks, P. J. M. and Zehner, C. (2000). GOME Fast delivery and value-Added Products (GOFAP). Proceedings of the ESA ERS - ENVISAT Symposium, Gothenburg, Sweden, 16–20 October 2000.

[2] Allaart, M.A.F., H. Kelder and L. C. Heijboer (1993). On the relation between ozone and potential vorticity, Geophys.Res.Lett., 20, 811.

[3] Allaart, M., van Weele, M., Fortuin, P., and Kelder, H. (2002). An improved UV-index as function of solar zenith angle and total ozone, preprint.

[4] Andrews, A., Boering, K., Daube, B., Wofsy, S., Loewenstein, M., Jost, H., Podolske, J., Webster, C., Herman, R., Scott, D., Flesh, G., Moyer, E., Elkins, J., Dutton, G., Hurst, D., Moore, F., Ray, E., Romanshkin, P., and Strahan, S. (2001). Mean ages of stratospheric air derived from in situ observations of CO2, CH4, and N2O, J. Geophys. Res., 106, 3229532314.

[5] Baldwin, M., T. Hirooka, A. ONeill, S. Yoden, A.J. Charlton, Y. Hio, W.A. Lahoz and A. Mori (2003). Major Stratospheric Warming in the Southern Hemisphere in 2002: Dynamical Aspects of the Ozone Hole Split, SPARC Newsletter, 20, 24-26.

[6] Bhartia, P. K., R. D. McPeters, C. L. Mateer, L. E. Flynn and C. Wellemeyer (1996). Algorithm for the estimation of vertical ozone profiles from the backscattered ultraviolet technique, Journal of Geophysical Research, 101, 18793.

[7] Bovensmann, H., J. P. Burrows, M. Buchwitz, J. Frerick, S. Nol, V. V. Rozanov, K. V. Chance, and A. P. H. Goede (1999). SCIAMACHY: Mission objectives and measurement modes, J. Atmos. Sci., 56, 127-150.

[8] Bregman, B., A. Segers, M. Krol, E. Meijer, and P. van Velthoven (2003), On the use of mass-conserving wind fields in chemistry-transport models, Atmos. Chem. Phys., 3, 447-457.

[9] Burrows, J. P., Weber, M., Buchwitz, M., Rozanov, V., Ladstätter-Weibenmayer, A., Richter, A., Debeek, R., Hoogen, R., Bramstedt, K., Eichmann, K. -U., Eisinger, M., and Perner, D. (1999). The Global Ozone Monitoring Experiment (GOME): Mission concept and first results, J. Atmos. Sciences, 56, 2, 151–175.

[10] Cariolle, D. and Déqué, M. (1986). Southern Hemisphere medium-scale waves and total ozone disturbances in a spectral generated circulation model, J. Geophys. Res., 91, 10 825–10 846.

[11] Déqué, M. (1997). Ensemble size for numerical seasonal forecasts, Tellus, 49A, 74-86.

[12] Dethof, A., and Holm, E. (2002). Ozone in ERA40: 1991-1996, ECMWF Technical Memorandum no. 377, August 2002.

[13] Elbern, H. and Schmidt, H. (2001). Ozone episode analysis by four-dimensional variational chemistry data assimilation, J. Geophys. Res., 106, 3569-3590.

[14] Errera, Q., and Fonteyn, D. (2001). Four-dimensional variational chemical assimilation of CRISTA stratospheric measurements, J. Geophys. Res., 106, 12253-12265.

[15] Eskes, H. J., Piters, A. J. M., Levelt, P. F., Allaart, M. A. F. and Kelder, H. M. (1999). Variational assimilation of total-column ozone satellite data in a 2D lat-lon tracer-transport model, J. Atmos. Sci., 56, 3560-3572.

[16] Eskes, H.J., van Velthoven, P. F. J., and Kelder, H. M. (2002). Global ozone forecasting based on ERS-2 GOME observations, Atmos. Chem. Phys., 2, 271-278.

[17] Eskes, H. J., van Velthoven, P. F. J., Valks, P. J. M. and Kelder, H. M. (2003a). Assimilation of GOME total ozone satellite observations in a three-dimensional tracer transport model, Q. J. R. Meteorol. Soc., 129, 1663.

[18] Eskes, H., Segers, A., and van Velthoven, P. (2003b). Ozone Forecasts of the Stratospheric Polar Vortex Splitting Event in September 2002, preprint, submitted to JAS special issue on the split vortex 2002.

[19] Fisher, M. and Lary, D. J. (1995). Lagrangian four-dimensional variational data assimilation of chemical species, Q. J. R. Meteorol. Soc., 121, 1681.

[20] Hall, T. and Plumb, R. (1994). Age as a diagnostic of stratospheric transport, J. Geophys. Res., 99, 10591070.

[21] Hólm, E. V., Untch, A., Simmons, A., Saunders, R., Bouttier, F., and Andersson, E. (1999). Multivariate ozone assimilation in four-dimensional data assimilation, SODA workshop on Chemical Data Assimilation, Publication 188, KNMI, De Bilt, the Netherlands.

[22] Jang, Kun-Il, Zou, X., De Pondeca, M. S. F. V., Shapiro, M., Davis, C., Krueger, A. (2003). Incorporating TOMS Ozone Measurements into the Prediction of the Washington, D.C., Winter Storm during 2425 January 2000. J. Appl. Meteor., 42, 797812.

[23] Khattatov, B. V., Gille, J. C., Lyjak, L. V., Brasseur, G. P., Dvortsov, V. L., Roche, A. E. and Waters, J. W. (1999). Assimilation of photochemically active species and a case analysis of UARS data, J. Geophys. Res., 104, 18715-18737.

[24] Khattatov, B. V., Lamarque, J.-F., Lyjak, L. V., Menard, R., Levelt, P., Tie, X., Brasseur, G. P., and Gille, J. C. (2000). Assimilation of satellite observations of long-lived chemical species in global chemistry transport models, J. Geophys. Res., 105, 29 135–29 144.

[25] Lalaurette, F. and Ferranti, L. (2001). Verification statistics and evaluations of ECMWF forecasts in 2000–2001, ECMWF Technical Memorandum 346, September 2001.

[26] Lambert, J.-C., ed. (2002). ERS-2 GOME GDP 3: Implementation and Delta Validation, ESA document ERSE-DTEX-EOAD-TN-02-0006, November 2002.

[27] Lee, A. M., Carver, G. D., Chipperfield, M. P., and Pyle, J. A. (1997). Three-dimensional chemical forecasting: a methodology, J. Geophys. Res., 102, 3905–3919.

[28] Levelt, P. F., Allaart, M. A. F. and Kelder, H. M. (1996). On the assimilation of total-ozone satellite data, Ann. Geophysicae, 14, 1111-1118.

[29] Levelt, P. F., B. V. Khattatov, J. C. Gille, G. P. Brasseur, X. X. Tie, and J. W. Waters (1998). Assimilation of MLS ozone measurements in the global three?dimensional chemistry transport model ROSE. Geophys. Res. Lett., 25, 4493?4496.

[30] Levelt, P., *et al.*, 2000. Science Requirements Document for OMI-EOS, RS-OMIE-KNMI-0001, De Bilt, The Netherlands, December 2000.

[31] Long, C. S., A. J. Miller, H. T. Lee, J. D. Wild, R. C. Przywarty, and D. Hufford (1996). Ultraviolet index forecasts issued by the National Weather Service, Bull. Amer. Meteor. Soc., 77, 729-748.

[32] McPeters, R. D., Bhartia, P. K., Krueger, A. J., Herman, J. R., Wellemeyer, C. G., Seftor, C. J., Jaross, G., Torres, O., Moy, L., Labow, G., Byerly, W., Taylor, S. L., Swissler, T., and Cebula, R. P. (1998). Earth Probe Total Ozone Mapping Spectrometer (TOMS) Data Products User's Guide, NASA Technical Publication 1998-206895, NASA Goddard Space Flight Center, Greenbelt, Maryland 20771.

[33] Menard, R., Cohn, S. E., Chang, L.-P., and P. M. Lyster (2000). Assimilation of Stratospheric Chemical Tracer Observations Using a Kalman Filter. Part I: Formulation, Mon. Wea. Rev, 128, 2654-2671.

[34] Neuendorfer, A. C. (1996). Ozone monitoring with TIROS-N operational vertical sounders, J. Geophys. Res., 101, 18807-18828.

[35] Peuch, A., J.-N. Thépaut, and J. Pailleux (2000). Dynamical impact of total-ozone observations in a four-dimensional variational assimilation, Q.J.R.Meteorol.Soc., 126, 1641-1659.

[36] Riishøjgaard, L. P.: On four-dimensional variational assimilation of ozone data in weather-prediction models, Q. J. R. Meteorol. Soc., 122, 1545–1571, 1996.

[37] Riishøjgaard, L. P., Štajner, I., and Lou, G.,-P. (2000). The GEOS ozone data assimilation system, Adv. Space Res., 25, 1063-1072.

[38] Schoeberl, M. R., A. R. Douglass, Z. Zhu, S. Pawson (2003). A comparison of the lower stratospheric age spectra derived from a general circulation model and two data assimilation systems, J. Geophys. Res., 108, doi:10.1029/2002JD002652.

[39] Simmons, A. J., Mureau, R., and Petroliagis, T. (1995). Error growth and estimates of predictability from the ECMWF forecasting, Q. J. R. Meteorol. Soc., 121, 1739-1771.

[40] Simmons, A., M. Hortal, G. Kelly, A. McNally, A. Untch, S. Uppala (2003). ECMWF analyses and forecasts of stratospheric winter polar vortex break-up: September 2002 in the southern hemisphere and related events, preprint, submitted to JAS special issue on the split vortex 2002.

[41] Solomon, S. (1999). Stratospheric ozone depletion: A review of concepts and history, *Rev. Geophys.*, **37,** 275-316.

[42] Spurr, R., W. Thomas, D. Loyola (2002). GOME Level 1 to 2 Algorithms Description, Deutsches Zentrum fr Luft und Raumfahrt, July 2002.

[43] Štajner, I., Riishøjgaard, L. P., and Rood, R., B. (2001). The GEOS ozone data assimilation system: specification of error statistics, Q. J. R. Meteorol. Soc., 127, 1069-1094.

[44] Stoffelen, A., Eskes, H., and Kelder, H. (1999). The EU SODA project, final report, KNMI, De Bilt, the Netherlands.

[45] Struthers, H., Brugge, R., Lahoz, W. A., O'Neill, A., and Swinbank, R. (2002). Assimilation of Ozone Profiles and Total Column Measurements into a Global General Circulation Model, J. Geophys. Res., 107, doi:10.1029/2001JD000957.

[46] UNEP/WMO (2002). Scientific assessment of ozone depletion: 2002, United Nations Environmental Programme and World Meteorological Organisation.

[47] Vaughan, G., and J. D. Price (1991). On the relation between total ozone and meteorology, Q. J. R. Meteorol. Soc., 117, 1281-1298

[48] Valks, P. J. M., Piters, A. J. M., Lambert, J. C., Zehner, C., and Kelder, H. (2003). A fast delivery system for the retrieval of near-real time ozone columns from GOME data, Int. J. Remote Sensing, 24, 423-436.

[49] World Meteorological Organisation, Global Atmospheric Watch (2001). WMO/CEOS Report on a strategy for Integrated Satellite and Ground-Based Observations of Ozone, Report no. 140, January 2001.