

Assimilation of ozone profiles and total column measurements into a global general circulation model

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Received 19 June 2001; revised 3 May 2002; accepted 15 May 2002; published 25 October 2002.

[1] Ozone profiles from the Microwave Limb Sounder (MLS) instrument flown on board the Upper Atmosphere Research Satellite (UARS) and total ozone columns measured by the Global Ozone Monitoring Experiment (GOME) on board the Second European Remote Sensing Satellite (ERS-2) have been assimilated using a troposphere-stratosphere data assimilation system. The analysis system is based on the global analysis system used for operational analysis of the stratosphere at the Meteorological Office from 1991 to 2000. Three assimilation runs have been completed for a three-week period in April 1997 to test the advantage of using a combination of MLS and GOME observations, compared with the assimilation of each observation data set separately. The statistical information produced by the assimilation system shows that the combination of MLS and GOME observations via the assimilation process produces ozone fields that show improvement compared with analysis fields produced by the assimilation of either MLS or GOME separately. Comparison of the analyzed ozone fields with independent observations (ozonesondes, Halogen Occultation Experiment (HALOE) profiles and Total Ozone Mapping Spectrometer (TOMS) total ozone column measurements) corroborates these results and shows that the combined MLS and GOME ozone analyses provide a realistic representation of the atmospheric ozone distribution. The global root-mean-square residual (difference between the analyses and independent observations) against HALOE and TOMS observations is comparable to the quoted errors in the HALOE and TOMS instruments (5% in each case). *INDEX TERMS:* 3337 Meteorology and Atmospheric Dynamics:

Numerical modeling and data assimilation; 0340 Atmospheric Composition and Structure: Middle atmosphere—composition and chemistry; 1640 Global Change: Remote sensing; 3334 Meteorology and Atmospheric Dynamics: Middle atmosphere dynamics (0341, 0342); 0341 Atmospheric Composition and Structure: Middle atmosphere—constituent transport and chemistry (3334); *KEYWORDS:* chemical data assimilation, ozone, general circulation model, Earth observation

Citation: Struthers, H., R. Brugge, W. A. Lahoz, A. O'Neill, and R. Swinbank, Assimilation of ozone profiles and total column measurements into a global general circulation model, *J. Geophys. Res.*, 107(D20), 4438, doi:10.1029/2001JD000957, 2002.

1. Introduction

[2] Ozone is one of the most important trace species in the atmosphere, and its study has generated wide scientific and public interest. In the stratosphere, there is an apparent downward trend in global ozone, punctuated by significant springtime losses in the polar regions [*World Meteorological Organization (WMO)*, 1999]. The increased UV-B radiation at the surface due to loss of stratospheric ozone is a concern due to the harmful effect of this radiation on living systems. Understanding the present state of the atmospheric ozone

field and its interaction with other meteorological variables is critical for the prediction of future ozone changes and the consequent impacts on the earth's environment.

[3] There are a number of space-based remote sensing instruments designed to provide global data sets of atmospheric ozone. These include the TOVS (Tiros Operational Vertical Sounder), SBUV (Solar Backscatter UV) and TOMS (Total Ozone Monitoring Spectrometer) series of instruments, and the Upper Atmosphere Research Satellite (UARS) spacecraft [*Reber et al.*, 1993]. UARS carries four research instruments designed to measure atmospheric trace species: Halogen Occultation Experiment (HALOE), Microwave Limb Sounder (MLS), Cryogenic Limb Array Etalon Sounder (CLAES) and Improved Stratospheric Mesospheric

Sounder (ISAMS). ESA's Global Ozone Monitoring Experiment (GOME) is an experimental nadir viewing instrument on board the Second European Remote Sensing Satellite (ERS-2) which was launched in April 1995. Future missions include ESA's Environmental Satellite, Envisat [*European Space Agency (ESA)*, 1998] due for launch on 1 March 2002 (GMT), and NASA's EOS Aura satellite, due for launch in June 2003, both of which have novel research instruments designed to measure the chemical composition of the atmosphere.

[4] Ozone data products from satellite remote sensing instruments have different strengths and weaknesses which reflect the particular characteristics of the instrument and platform. For example, instruments differ in their horizontal coverage, observation rate, vertical resolution, instrument stability and lifetime. Thus, it is advantageous to generate an estimate of the atmospheric ozone field using an analysis system which combines a number of different observation types in a way that retains the important information content in each observation data set. This is also of particular relevance to Envisat and EOS Aura as both missions have plans for the measurement of contemporaneous ozone profiles from limb instruments and ozone columns from nadir viewing instruments. The challenge is to combine these data sets in a way that retains the information content in both the limb and nadir measurements in the final analysis product.

[5] Data assimilation [*Daley*, 1991], which is at the heart of Numerical Weather Prediction (NWP) and is increasingly being used to analyze photochemical data, provides an objective way of combining the heterogeneous data sets from remote sensing instruments onboard satellites. Data assimilation brings a number of benefits to this task. These benefits include: (1) it "interpolates" the data in an "intelligent" way using the governing equations of the system (or an approximation), (2) it propagates information from data rich regions to data poor regions using the governing equations of the system, (3) it combines heterogeneous observations in a self-consistent way, (4) it quality-controls data, and (5) it provides statistical information which can be used to test assumptions on the error characteristics of the model, analyses and observations. Furthermore, assimilation of photochemical species can both test photochemical theories and provide information on unobserved species via the photochemical model equations [see, e.g., *Wang et al.*, 2001]. Finally, a General Circulation Model (GCM) based assimilation system (as used in this paper) can take account of the feedback between the dynamical, photochemical and radiative components of the system.

[6] Interpolation (bilinear or a higher order polynomial) between data points is often used to analyze data or combine heterogeneous data sets, and may be regarded as an alternative to data assimilation. However, although interpolation between data points is simpler and less expensive than data assimilation, and in data rich areas it can provide fields of similar quality, it cannot match the benefits of data assimilation. For example, linear interpolation of a tracer between data points would not be able to capture its distribution in the presence of a strong wind shear, whereas data assimilation (which incorporates the governing equations of the system) would be able to do so. Such strong wind shears occur during the winter and early-spring time in the region of the so-called polar vortex edge.

[7] In this paper we describe the assimilation of ozone measurements taken by the UARS MLS and ERS-2 GOME instruments in conjunction with all the available meteorological data, using the Meteorological Office data assimilation system. The object of this study is to demonstrate the advantage of using a combination of UARS MLS ozone profile data and ERS-2 GOME total column ozone data, compared with the assimilation of each observational data set separately. The assimilation system used is an extension of the full troposphere-stratosphere NWP system used for the real-time meteorological analysis of the stratosphere by the Meteorological Office from 1991 to 2000 [*Swinbank and O'Neill*, 1994]. This is an extension of the work completed on the assimilation of UARS MLS ozone by *Connew* [1998].

[8] This study provides, to the best of our knowledge, the first instance of the combined assimilation of ozone profile and total column ozone measurements into a GCM-based system. The results from this study will help develop the algorithms to assimilate photochemical data from Envisat [see, e.g., *van der A*, 2001].

[9] The UARS MLS [*Waters et al.*, 1999] instrument measures radiation in the 63 GHz, 183 GHz and 205 GHz bands in a limb geometry. The tangent height of the scans range from the surface to approximately 90km, with a scan being completed every 65.5 seconds. After commissioning, the 205 GHz radiometer on MLS was operated successfully from January 1992 to December 1996. After this time, MLS was only used sparingly due to power saving constraints on the UARS platform. More details of the MLS observations used in this study are given in section 2.2. An updated MLS instrument has been designed to be flown on NASA's EOS Aura spacecraft. MLS data has been assimilated by *Levelt et al.* [1998] who used the ROSE chemical transport model [*Rose and Brasseur*, 1989] forced by the Meteorological Office meteorological analyses. The ROSE model contains an extensive photochemical scheme which includes heterogeneous chemistry. The observations were assimilated using a sequential statistical interpolation approach.

[10] GOME [*Burrows et al.*, 1999], was launched on board ESA's ERS-2 in April 1995. GOME is a nadir viewing spectrometer measuring earthshine radiance in the wavelength range 240–790nm with a spectral resolution of between 0.2 and 0.45nm. The swath width of each scan is 960km with a ground pixel resolution of $40 \times 320 \text{ km}^2$ for the majority of the orbit. Global coverage is achieved in three days from 42 orbits. GOME was designed as a precursor to SCIAMACHY (SCanning Imaging Absorption spectroMeter for Atmospheric Cartography) which is due for launch on ESA's Envisat-1 platform on 1 March 2002 (GMT). Koninklijk Nederlands Meteorologisch Instituut (KNMI; the Royal Netherlands Meteorological Institute) assimilate GOME total column ozone using the TM3 chemistry transport model, in near real time (see http://www.knmi.nl/gome_fd/index.html). They use a simplified Kalman filter approach for the assimilation, with a Cariolle scheme included in the model to parameterize the ozone photochemistry [*Eskes and Jeuken*, 1998].

[11] The assimilation of a combination of profile and total column ozone measurements is described by *Riishøjgaard et al.* [2000] and *Štajner et al.* [2001], who use SBUV/2 profiles and TOMS total ozone to generate global ozone

analyses in near real time using the Data Assimilation Office (DAO) Physical Space Assimilation System (PSAS), and an off-line transport model. It is shown that over the validation period of the winter of 1992, the assimilation system performs well, with the ozone analyses in good agreement with independent ozonesonde and HALOE observations.

[12] Section 2 gives a more detailed description of the assimilation system and the observations used. Section 3 presents the results and evaluation of three assimilation runs over a three-week period in April 1997. The assimilation period is short, being only 21 days. This is because there is no Meteorological Office operational data currently available before January 1997, and there is no MLS data available after April 1997 for periods longer than two weeks (after April 1997, MLS has been off most of the time with very few measurements being made). The longest period for which there is data available from both sources is this three week period in April 1997. A longer assimilation, over seasons or years, is desirable to provide general conclusions about the accuracy of the assimilated ozone fields, but the focus of this paper is on the advantage gained by the combination of two different observation types which can be assessed using data over a relatively short period of time (but note that for NWP systems, and due to computational restrictions, periods of about three weeks are used for initial assessment). Section 3 also provides evidence which suggests that this 21 day period is adequate to assess the merit of the different combinations of observations. Observation minus analysis (O-A) residual statistics from the runs are used to evaluate the analyzed ozone fields. Observation minus background (i.e., forecast) (O-B) residual statistics are used to assess the error characteristics of the observations and the background. (Unless otherwise indicated, residual statistics refer to O-A.) Residual statistics will either refer to the bias or root-mean-square (RMS) difference. To corroborate the results of section 3 and assess whether the combined assimilation of MLS and GOME ozone provides a realistic representation of the atmospheric ozone distribution, we compare the analyses with independent observations (ozonesondes, HALOE profiles and TOMS total ozone columns) in section 4. Finally, section 5 gives some conclusions from this work and outlines future improvements to the assimilation system.

2. Assimilation System

[13] The scheme used for the assimilation of the ozone data is the Analysis Correction (AC) method, which is described by *Lorenc et al.* [1991]. This scheme was used for real time meteorological analysis of the stratosphere at the Meteorological Office from 1991 to 2000 as described by *Swinbank and O'Neill* [1994]. The analysis is performed by repeated insertion using a modified successive-correction scheme.

[14] The ozone assimilation calculated as part of the full meteorological analysis is univariate. No correlations between the ozone background error covariance matrix and the meteorological background error covariance matrix are considered in this study. Because of computational restrictions, the elements of the background error covariance matrix for the ozone assimilation are estimated empirically. A similar approach is followed by, for example, *Errera and Fonteyn* [2001]. Based on short assimilation

experiments, the background variance is chosen to be 10% of the background ozone mass mixing ratio.

[15] The horizontal background error correlations are modelled using a second order auto-regressive (SOAR) function with a nominal correlation length of 600km. This is the value used by the Meteorological Office in the AC scheme for the stratosphere (temperature and/or wind correlations). The correlations also vary as a function of time. In particular, the correlation length varies as a function of the time difference between the observations and the model, so that when the observations are first inserted they are given a low weight, but influence a large area (the horizontal correlation is increased from its nominal value). As the model time approaches the observation time, the correlation length is decreased to the nominal 600km as the weight is increased. After the observation time, the weight is decreased and the correlation length is held constant at 600km.

[16] The background horizontal correlation is given by

$$\mu_h = (1 + (r/s)) \exp(-r/s). \quad (1)$$

r is the horizontal distance and s is the correlation length (which varies in time as described above).

[17] The background vertical correlations are approximated by a Gaussian function, the argument being the log of the pressure ratio between the two levels,

$$\mu_{ij}^v = \exp(-b \log^2(p_i/p_j)). \quad (2)$$

μ_{ij}^v is the vertical correlation between level i and j , at pressures p_i and p_j respectively and b is a dimensionless parameter which varies with level and latitude. In the stratosphere, a value of $b = 4$ is used in the tropics, and a value of $b = 3$ in the extra-tropics. Note that the vertical correlations only have an effect vertically above and below the observation level.

2.1. Forecast Model

[18] The numerical forecast model which is the basis of the assimilation system is the Unified Model (UM) [*Cullen and Davies*, 1991], which has been developed for a number of applications including operational weather forecasting and climate prediction. For this study we used the HADAM3 version of the stratospheric configuration of the UM which uses a regular Arakawa B grid with 2.5° resolution in latitude and 3.75° resolution in longitude. The model has 42 levels in the vertical, the lowest 19 of which are terrain following. The vertical resolution in the stratosphere is approximately 1.6km with the top most model level at 0.28hPa [see *Swinbank and O'Neill*, 1994]. The tracer advection scheme used for the advection of the ozone is a positive definite scheme based on the flux redistribution method of *Roe* [1985] with a superbee limiter [*Cullen and Barnes*, 1997]. The performance of a troposphere-stratosphere version of the UM which uses the HADAM3 configuration has been discussed by *Lahoz* [2000].

[19] To approximate the atmospheric ozone photochemistry, the Cariolle [*Cariolle and Déqué*, 1986] ozone photochemistry parameterization was included in the UM. This scheme is based on a linearization of the ozone tendency about the local ozone mixing ratio, temperature and overhead column ozone. The ozone and temperature climatolo-

gies and their partial derivatives with respect to the ozone mixing ratio, are derived from a two-dimensional photochemical model. The scheme that we used is based on the Cariolle scheme developed for the assimilation of ozone column data at KNMI [Jeuken *et al.*, 1999]. This implementation replaces the original ozone equilibrium values with the Fortuin ozone climatology [Fortuin and Kelder, 1998]. In addition, the Cariolle scheme that we implement also calculates an estimate of the loss of ozone due to the heterogeneous processes which occur on polar stratospheric clouds (PSCs). This parameterization of heterogeneous processes has been developed by P. Braesicke at Cambridge University, and is based on the concept of an “activated chlorine tracer” which becomes active when the temperature falls below the threshold of PSC formation.

[20] The radiation scheme in the HADAM3 configuration of the UM is described by Edwards and Slingo [1996] and Ingram *et al.* [1997]. It has been modified so the ozone fields used to calculate the radiative forcings are the assimilated ozone fields rather than climatology.

2.2. UARS MLS Observations

[21] The MLS ozone data assimilated in this work was the version 4 ‘level 3AT’ retrieved product which consists of profiles of ozone mixing ratios along track. The measurements are reported on standard UARS pressure levels. The version 4 MLS product contains useful information in the pressure range 100hPa to 0.46hPa, on alternate standard UARS pressure levels (100hPa, 46.4hPa, 21.6hPa, 10.0hPa, 4.64hPa, 2.16hPa, 1.0hPa and 0.46hPa). Approximately 320 profiles are measured in a six hour period by the UARS MLS.

[22] The quality of the version 4 MLS ozone data is described in the UARS/MLS Data Quality Document (see http://mls.jpl.nasa.gov/lucien/daac_document_v4). The quoted accuracy is the retrieval uncertainty provided by the MLS team. This includes random errors and some systematic errors. The estimated percentage accuracy of the version 4 profiles is quoted as 5% over the pressure range 21.6hPa to 2.16hPa, rising to 15% at 0.46hPa and 20% at 46.4hPa.

[23] For MLS ozone (and GOME total column ozone) we do not attempt to remove a bias prior to the assimilation. Estimation of such biases is difficult (chiefly because the “truth” is not known). However, data assimilation can be used as a tool to estimate biases in the observations (as well as in the model and the analyses). Later in the paper we discuss results from the analyses which provide supporting evidence for a bias between the GOME and TOMS measurements of total column ozone.

[24] The period of this study consists of three weeks in April 1997, starting on the 10th and finishing on the 30th. For this period the MLS instrument made measurements in a mode of reverse scan for two days generating normal profiles, followed by a day in limb tracking mode where no profiles were measured, this cycle continuing for a full three weeks. This is one of the last periods for which the UARS MLS instrument made measurements over an extended time period.

[25] The latitudinal coverage of the MLS observations ranges from 80° in one hemisphere to 34° in the other, depending on the orientation of the spacecraft. UARS undertakes a yaw approximately every 36 days which

changes the hemisphere that has greater coverage. The MLS instrument was northward facing for the whole period of this study (April 1997).

[26] The MLS samples a volume with a vertical resolution of about 3km and a horizontal resolution of about 600km [see, e.g., Froidevaux *et al.*, 1996]. Because the MLS vertical resolution is comparable to the UM grid, we choose to treat the MLS observations as layer means. The vertical analysis step in the assimilation calculates an analysis increment by vertically integrating the model ozone over each observation level and subtracting this from the vertically integrated observation over the same levels.

[27] The retrieved product also contains observation errors for each profile which were used to estimate the MLS observation errors for the assimilation scheme. The quoted observation errors show little variation between profiles, days and seasons so a global error profile was used in the assimilation. This was calculated as a mean of the error profiles reported in the retrieved product. The observation errors are assumed to be uncorrelated. We also assume the mean error profiles used include the error of representativeness for the MLS measurements.

[28] MLS temperature profiles are also assimilated, see Asenek *et al.* [2000] for details.

2.3. ERS-2 GOME Observations

[29] The GOME measurements used are the GOME Data Processor (GDP) version 2.7 operational level 2 total ozone column values, derived at Deutsches Zentrum für Luft- und Raumfahrt (DLR) using the DOAS (Differential Optical Absorption Spectroscopy) algorithm (see <http://auc.dfd.dlr.de/data.html> and Balzer and Loyola [1996]). The column ozone measurements range in latitude from approximately 90°N to between 75°S and 70.1°S through the period of this study. There are three pixels per scan along the ERS-2 orbit track, each scan spanning approximately 960km. The assimilation system assumes each observation represents a point measurement made at the center of the corresponding pixel. In a six hour period approximately 7000 observations are made by the GOME instrument.

[30] The GOME level 2 total ozone columns have been evaluated against ground based measurements in two geophysical validation campaigns (see <http://auc.dfd.dlr.de/data.html> and Balzer and Loyola [1996]). The average deviation between ground based measurements and co-located GOME measurements was found to be no greater than 5% for solar zenith angles less than 60°, and no greater than 10% for solar zenith angles between 60° and 90°. Based on this information, it was decided to use for the assimilation a GOME observation error of 10% of the GOME total column measurement. This includes random and systematic components of the GOME error. The error assumed for the GOME observations includes the error of representativeness associated with the GOME observations and the errors in the observations are assumed to be uncorrelated.

[31] For each observation, the analysis increments on the model levels are calculated at the observation location using the vertical component of the standard optimal interpolation (OI) analysis equation

$$\delta \mathbf{x}_v = \mathbf{B}_v \mathbf{H}_v^T (\mathbf{H}_v \mathbf{B}_v \mathbf{H}_v^T + O_i)^{-1} (y_i - \mathbf{H}_v \mathbf{x}_{b_i}) \quad (3)$$

where $\delta\mathbf{x}_{v_i}$ is the vector of ozone increments for observation i . \mathbf{B}_{v_i} is the vertical component of the background error covariance matrix interpolated to the location of observation i . \mathbf{H}_v is the vertical component of the GOME observation operator, which in this case is a simple numerical integration scheme which assumes the ozone mixing ratio is constant over a model layer. O_i is the GOME observation error, y_i is the GOME total column measurement and \mathbf{x}_{b_i} is the background estimate of the ozone profile at the observation location. T denotes the transpose.

[32] $\delta\mathbf{x}_{v_i}$ represents the ozone increment added to the model at the observation location, for a single, isolated total ozone observation denoted i . The full increment field $\delta\mathbf{x}_N$, is determined by horizontally spreading the $\delta\mathbf{x}_{v_i}$ on model levels in the normal way within the AC scheme. The analyzed ozone field is then simply the background field plus the increment field $\delta\mathbf{x}_N$,

$$\mathbf{x}_a = \mathbf{x}_b + \delta\mathbf{x}_N. \quad (4)$$

[33] The AC scheme uses a repeated insertion method which calculates the increment field $\delta\mathbf{x}_N$ and updates the background \mathbf{x}_b at every time step of the integration (see *Lorenz et al.* [1991] for details).

2.4. Operational Observations

[34] A full range of operational meteorological observational data was used in the assimilation in addition to the MLS and GOME observations. These are National Environmental Satellite Data and Information Service (NESDIS) temperature retrievals from the NOAA polar orbiters, radiosonde temperature, winds and relative humidity, aircraft temperature and winds, geostationary satellite cloud track winds and surface pressure observations (see *Swinbank and O'Neill* [1994] for more details).

[35] The assimilation described in this paper was initialized on 10 April 1997 at 1200 GMT using the operational troposphere-stratosphere analysis provided by the Meteorological Office. The initial ozone field was set to the Fortuin April climatology [*Fortuin and Kelder*, 1998], which is a zonally symmetric field based on monthly means, derived from ozonesonde and satellite measurements.

[36] The use of this ozone climatology to initialize the model may, in principle, cause large departures from reality in the analyzed fields. However, the analysis increments (for levels in the range 100hPa - 1hPa and for total column ozone) are small relative to the analyzed fields after the two-day spin-up period (see Figure 1), suggesting that the assimilation system is behaving in a stable (and realistic) way, and that the initial conditions are not influencing the analyses in a significant way.

3. Results and Discussion

3.1. Overview

[37] Three separate runs of the assimilation system were performed over the same 21 day period (10 April - 30 April 1997): (1) MLS ozone profiles used to constrain the ozone field, (2) GOME total column ozone measurements used to constrain the ozone field, and (3) MLS and GOME measurements used together to constrain the ozone field.

[38] The same operational observations as well as MLS temperature measurements were used in all three cases for the assimilation of the meteorological fields. Comparison of the results from run 3 with those from runs 1 and 2 give an indication of any advantage gained by using a combination of profile and total ozone measurements in the assimilation.

[39] The spin-up of the ozone fields takes two days, after which time the residual statistics are stable. Unless otherwise indicated, all the statistics shown in the following sections are from 12 April 1997 through to the end of the assimilation period on 30 April 1997, excluding the initial two day spin up period.

3.2. MLS and GOME Comparisons

[40] The evaluation of the ozone analyses from runs 1, 2 and 3 is based on an assessment of the statistical information provided by the assimilation procedure. Later in section 4 we compare the ozone analyses against independent observations (i.e., not used in the assimilation procedure) to (1) corroborate the results of section 3 and (2) assess whether the combined MLS and GOME ozone analyses (run 3) provide a realistic representation of the atmospheric ozone distribution. These independent observations provide a spatially independent test of the analyses.

[41] *Talagrand* [1998] discusses the a posteriori evaluation of assimilation schemes, and introduces the information minus analysis vector as a diagnostic tool. The information minus analysis vector includes the observation minus analysis vector, which *Talagrand* shows should have the following properties: (1) The expectation value for the observation minus analysis (the bias) must be zero for an optimal system. (2) The RMS value of observation minus analysis should be less than the a priori observation error. (3) The RMS value will increase asymptotically to the observation error as the estimation error (analysis minus truth) decreases.

[42] Figure 2 shows the profile of RMS differences between the MLS observations and the runs 1, 2 and 3 analyses relative to the mean MLS profile generated from the period 12 April 1997 to 30 April 1997. The results are plotted on the seven UARS pressure levels for which the MLS observations are reported. The dashed curve is the assumed MLS ozone error profile used in the assimilation (see section 2.2).

[43] For runs 1 and 3, the RMS values for MLS follow the prescription of *Talagrand*, with the RMS values being slightly less than the a priori MLS observation errors over the whole pressure range of the observations. Within the pressure range 21.6hPa - 2.16hPa the mean difference (bias) between the MLS observations and the analyses for runs 1 and 3 is positive and less than 2% of the mean MLS profile (see Figure 1). This indicates that for these pressures the biases between MLS observations and run 1 and run 3 analyses are close to zero (and are not significant) as required by the prescription of *Talagrand*.

[44] For pressures greater than 21.6hPa the bias in the analyses for runs 1 and 3 increases in magnitude, rising to 25% at 100hPa. The bias (difference between the MLS observations and the analyses: O-A) is negative at 46.4hPa and very slightly positive at 100hPa (see Figure 1). At 100hPa, the information content in the MLS observations is low (reflected in larger MLS observation errors) and the analysis is less constrained by the observations. At 2.16hPa

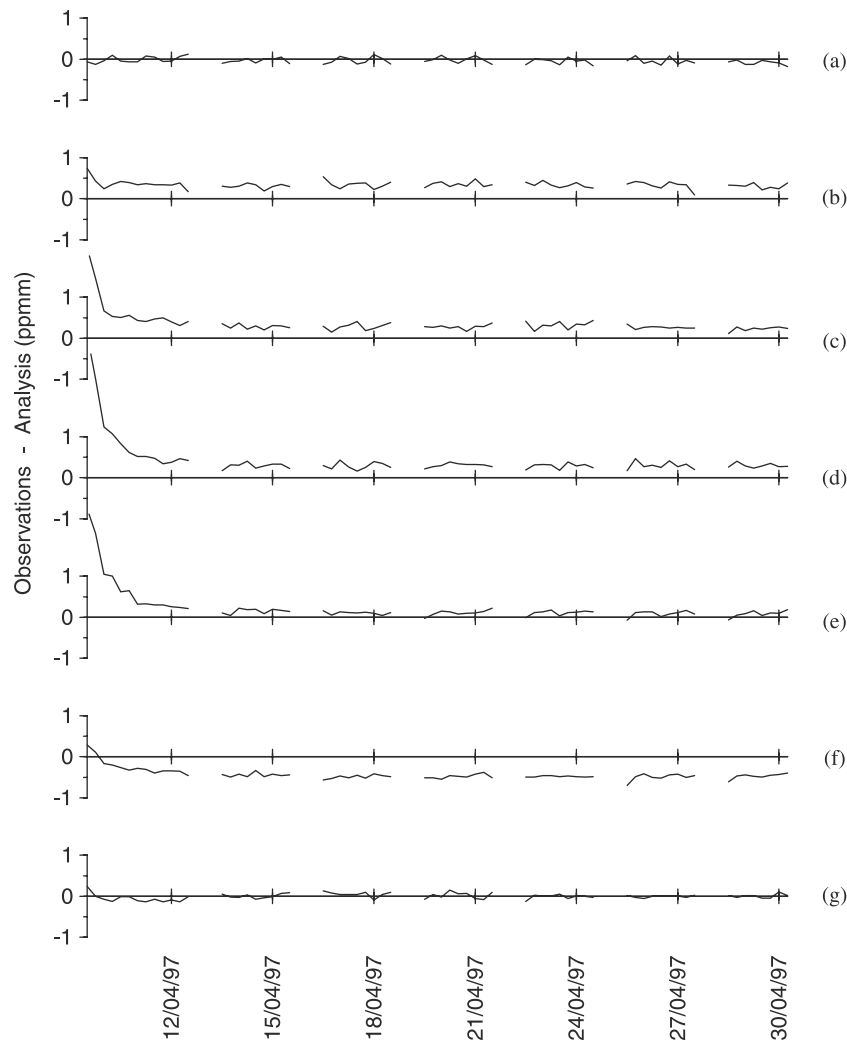


Figure 1. Globally averaged mean difference (MLS minus analysis), in parts per million by mass (ppmm), between the MLS ozone observations and co-located analyzed ozone values for run 3 for several pressure levels. Positive values indicate that the MLS observations are larger than the analyzed values. (a) 1hPa, (b) 2.16hPa, (c) 4.64hPa, (d) 10hPa, (e) 21.6hPa, (f) 46.4hPa, (g) 100hPa.

the bias is positive and at 1hPa it is very slightly negative (see Figure 1). At these pressure levels, the Cariolle scheme strongly constrains the model.

[45] Run 2, in contrast (Figure 2b), shows RMS differences which are greater than the a priori MLS observation error within the pressure range 50hPa - 1hPa. This indicates a significant difference between the ozone analyses and the MLS observations, which has been traced to a positive bias in the run 2 ozone fields with respect to the MLS observations (the average difference between the co-located run 2 ozone analyses and MLS ozone observations peaks at 0.54 parts per million by volume, ppvm, at 10hPa).

[46] Possible reasons for the bias between run 2 analyses and MLS observations include the following: (1) an incorrect prescription of the ozone background error covariance, affecting the spreading of the total ozone increment in the vertical via equation (3) in section 2.3 [see *Jeuken et al.*, 1999]; (2) a bias in the model (UM plus Cariolle scheme) that cannot be corrected by the run 2 assimilation due to the fact that no profile information is being introduced via the assimilation; and (3) a bias in the initial (unrealistic) ozone

fields (again, the assimilation is unable to correct in the vertical when only column amounts are assimilated).

[47] The bias may also result from the combination of two or more of the reasons outlined above.

[48] Further work is under way to identify all the factors influencing the vertical structure of the analyzed ozone fields from run 2. Preliminary studies suggest that the initial ozone distribution is unlikely to be a significant factor (see Figure 1 and accompanying discussion). The relatively simple background error covariance matrix for ozone (see section 2) could also be a significant factor. For example, the covariance statistics take no account of the synoptic situation in the estimate of the forecast variance. (These considerations apply to the comparison between run 2 analyses and HALOE independent observations; see section 4.2.)

[49] Figure 3 shows the profile of RMS differences between the GOME observations and the runs 1, 2 and 3 analyses as a function of latitude. The dashed curve is the mean of the GOME observation error (assumed to be 10% of the GOME observation). In this case, run 1 shows a significant difference between the observations (GOME)

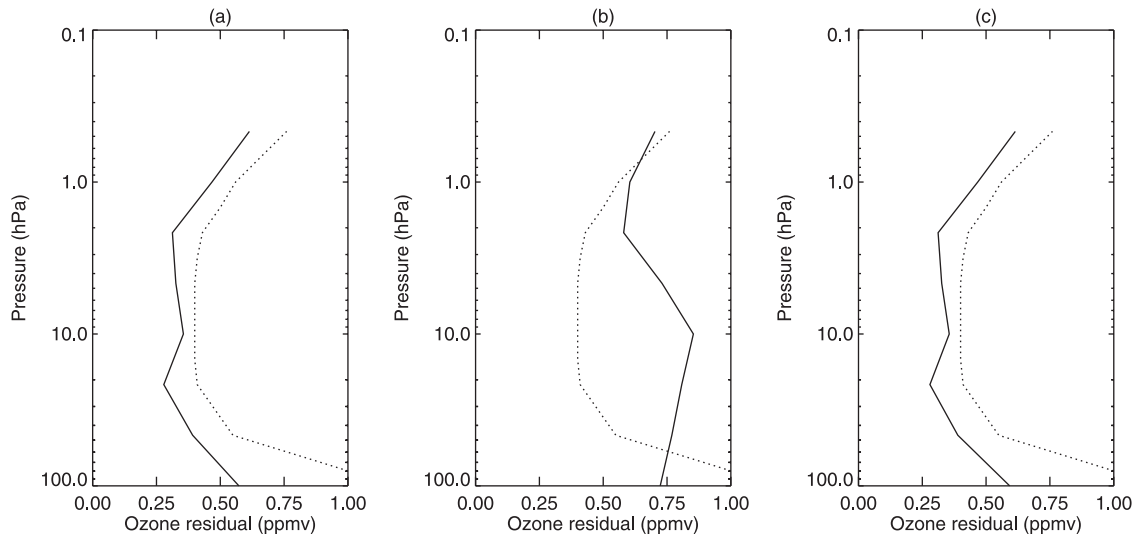


Figure 2. (a) Root-mean-square difference, in parts per million by volume (ppmv), between the MLS ozone observations and co-located analyzed ozone values for run 1. Dashed line is the assumed MLS ozone error profile in ppmv (see text for details). (b) As Figure 2a but for run 2. (c) As in Figure 2a but for run 3.

and the column ozone derived from the analysis at latitudes south of 30°S . This is not surprising because no MLS observations are present at latitudes south of 34°S over the period of this study. Excluding the region south of 30°S in run 1, the RMS values for the GOME observations are significantly lower than the a priori observation errors, particularly in the equatorial region. This suggests the assimilation is sub-optimal.

[50] For runs 2 and 3, over the whole latitude range covered by the GOME observations, the mean difference (bias) between the GOME observations and the analyses varies in sign and is less than 2% of the mean GOME measurements. This indicates that there is no significant bias between the GOME observations and the run 2 and 3 analyses as required by the prescription of Talagrand.

[51] Despite the shortcomings in the ozone background error covariance matrix, Figures 2–3 suggest that the system is adequate for demonstrating the relative merit of assimilating both column and profile information. Overall, Figures 2–3 indicate that the combination of MLS and GOME observations provides a better analysis than using each observational data set separately.

[52] Further evidence that the combination of MLS and GOME observations provides better analyses than using each observational data set separately, is provided by comparing the latitude-longitude distribution of the total column ozone analyses produced by runs 1 and 3 against independent gridded version 7 ADEOS TOMS data (obtained from the British Atmospheric Data Centre, BADC). Figure 4 shows total column ozone fields for 21 April 1997. For this day the run 3 analyses show better agreement (in both the magnitude and patterns of the fields) with the TOMS data than the run 1 analyses. For example, the relative high gradients in TOMS total ozone near 40°N and 60°W are captured by the run 3 analyses but not by the run 1 analyses. Also, the TOMS values in the Southern Hemisphere are captured better by the run 3 analyses than by the run 1

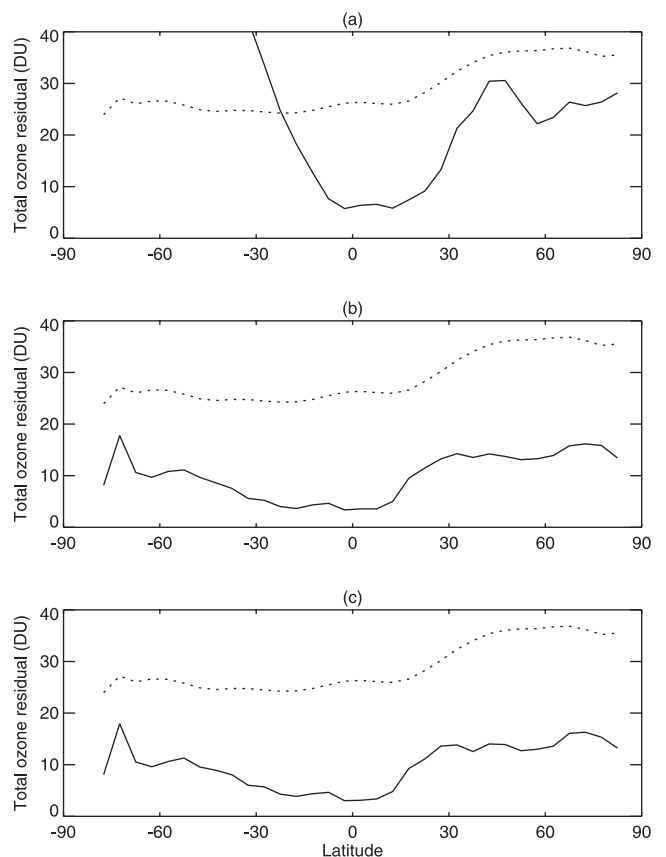


Figure 3. (a) Root-mean-square difference, in Dobson units (DU), between the GOME total ozone observations and co-located total columns derived from run 1. Dashed line is the mean GOME total ozone error in DU, assumed to be 10% of the measured value (see text for details). (b) As in Figure 3a but for run 2. (c) As in Figure 3a but for run 3.

analyses. Comparison against Earth Probe (EP) TOMS data for 21 and 29 April 1997, and ADEOS TOMS for 29 April 1997, confirms these results (not shown). In section 4 we provide a fuller comparison of the run 3 analyses against independent data (ozonesondes, HALOE and TOMS data).

[53] Even though the assimilation system uses simple approximations to the background and observation error covariances, the MLS and GOME RMS values (for run 3) are consistent with the expected results from a best linear unbiased estimate (BLUE) of the analysis as explained by Talagrand. The run 3 analyses are not inconsistent with both the MLS and GOME observation data sets.

[54] A method of evaluating whether an assimilation system is optimal is the chi-square diagnostic [Ménard *et al.*, 2000, Ménard and Chang, 2000]. Although this method provides important information for evaluating error covariance models and the validity of probabilistic assumptions used in the analyses, it has, nevertheless, some limitations [see, e.g., Štajner *et al.*, 2001]. Because this diagnostic is relatively difficult to implement with the AC scheme (an approximate iterative scheme), we do not attempt its calculation.

[55] In common with the majority of other atmospheric assimilation schemes, the AC scheme assumes the observation and background errors are normally distributed. Although we do not have any method available to directly test this assumption, a necessary but not sufficient condition for this to be true is that the O-B residual distributions are normally distributed [Štajner *et al.*, 2001].

[56] Figure 5a shows the 10hPa MLS O-B residual distribution for the run 3 analyses. This distribution has a mean of 0.11 ppmv and a standard deviation of 0.31 ppmv. The comparison between the actual O-B residual distribution and the true Gaussian function is very good, which gives us some confidence in the assumption of normally distributed observation and background errors. Similar conclusions can be drawn from the results at other levels.

[57] Figure 5b shows the 10hPa MLS O-B residual distribution for run 3, for the last 7 days of the assimilation period. As for Figure 5a, the results give us some confidence in the assumption of normally distributed observation and background errors. Furthermore, comparison between Figures 5a and 5b suggests that the error characteristics for MLS ozone observations and the ozone background are time-independent after the spin-up period. This strengthens the claim that the relatively short length of the runs does not affect the conclusions made about runs 1, 2 and 3.

[58] The GOME O-B residual distribution at 45°N for run 3 is shown in Figure 5c. This has a mean of -2.53 Dobson units (DU) and a standard deviation of 13.33 DU. For this case the O-B residual distribution does not match the true Gaussian function to the same extent as the MLS results in Figure 5a. The O-B residual distribution is too peaked at the center and the tails are too long. The MLS and operational observations were subjected to the comprehensive Bayesian quality control algorithms which are part of the assimilation cycle [Ingleby and Lorenc, 1993]. For technical reasons the GOME observations were quality controlled using a very simple algorithm which rejected observations if they differed from the background column ozone by more than 50 DU. Only a small number of GOME observations (less than

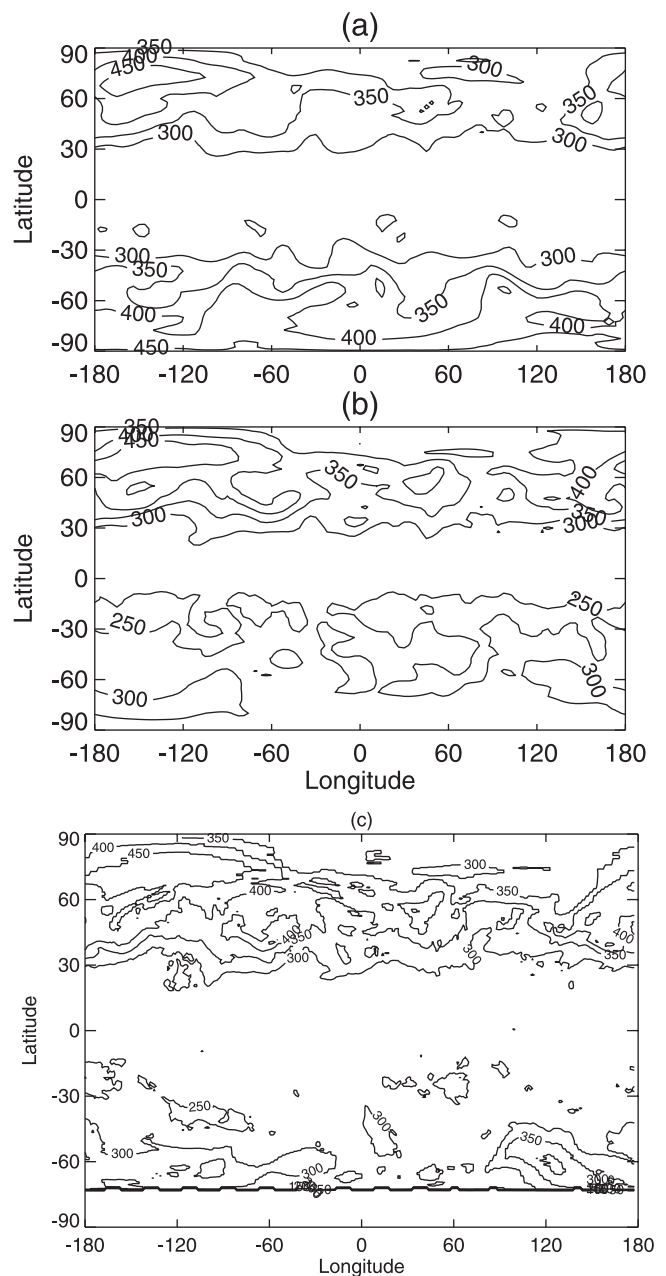


Figure 4. (a) Latitude-longitude distribution of total column ozone (DU) derived from run 1 analyses for 12 GMT 21 April 1997. (b) Latitude-longitude distribution of total column ozone (DU) derived from run 3 analyses for 12 GMT 21 April 1997. (c) Latitude-longitude distribution of total ozone (DU) from gridded version 7 ADEOS TOMS for 21 April 1997. The contour step is 50 DU for all plots.

1%) were rejected by this process. The lack of objective quality control of the GOME observations is likely to contribute to the elongated tails seen in the GOME O-B residual distribution. The rejection rate for MLS observations for the run 1 and run 3 configurations was similar (in both cases typically less than 5% in the stratosphere).

[59] The initial ozone distribution could be having a significant impact on the analyses and thus on the elongated tails of the GOME O-B residual distribution for the whole

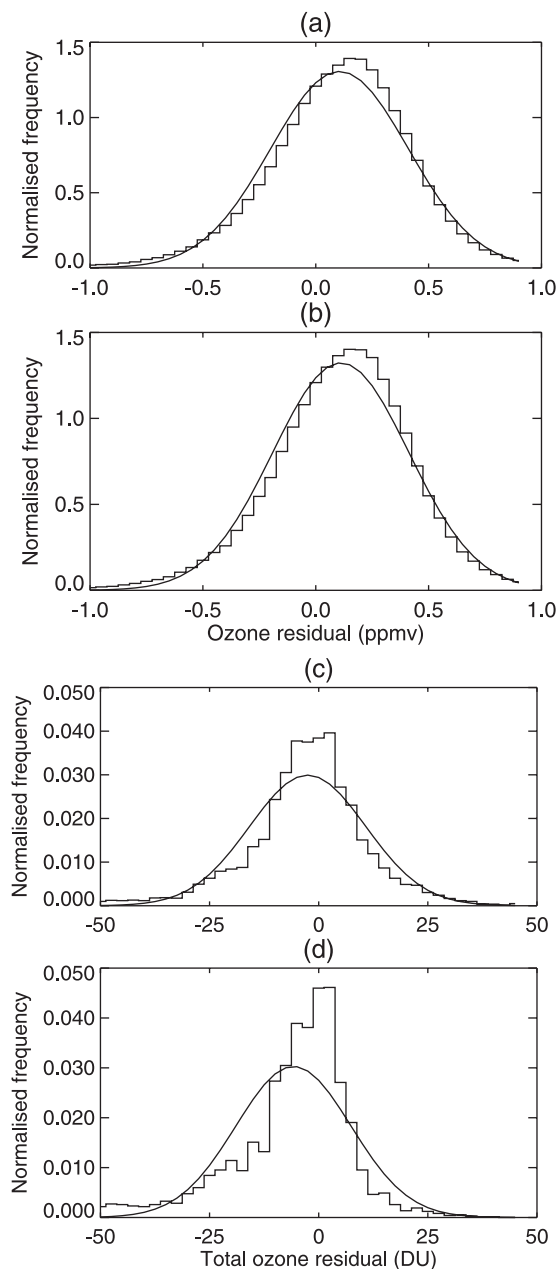


Figure 5. (a) Normalized 10hPa MLS O-B residual distribution (ppmv) for run 3. Also shown (bell-shaped curve) is the true Gaussian function with the same mean and standard deviation. (b) As in Figure 5a but for the last 7 days of run 3. (c) Normalized GOME total ozone O-B residual distribution (DU) at 45°N for run 3 with the corresponding Gaussian function. (d) As in Figure 5c but for the last 7 days of run 3.

three-week period under study (Figure 5c). However, as discussed in section 2.4, the evidence suggests that after the two-day spin-up period the impact of the initial conditions is not significant.

[60] Figure 5d shows the GOME O-B residual distribution at 45°N for run 3, for the last 7 days of the assimilation period. Although the agreement between Figures 5c and 5d is not as good as that between Figures 5a and 5b, the similarities between Figures 5c and 5d lend some support to the sugges-

tion that the error characteristics for GOME ozone observations and the ozone background are time-independent after the spin-up period. As above, this helps to strengthen the claim that the relatively short length of the runs does not affect the conclusions made about runs 1, 2 and 3.

[61] Comparison of the Gaussian distributions for Figures 5a and 5c (which are the realization of the O-B distribution in the analyses) with the expected Gaussian distribution given the choices made in the assimilation system (see section 2), suggests that (1) at 10hPa and at 45°N (and for run 3), there are small (and not significant) biases in the MLS and GOME O-B distributions, respectively, and (2) the ozone background error covariance may be too large by up to a factor of 2. The first result is consistent with the bias residuals discussed in the context of Figures 2–3. The second result is not surprising given the simple approximation to the ozone background error covariance which the assimilation system uses.

[62] The results from Figure 5 illustrate one of the benefits of data assimilation, namely that it provides statistical information which can be used to test assumptions about the error characteristics of the model, analyses and observations.

4. Comparison With Independent Observations

[63] The purpose of this paper is to assess the merit of the joint assimilation of MLS and GOME ozone data in comparison to the separate assimilation of MLS and GOME. To corroborate the results of section 3 and assess whether the combined MLS and GOME (run 3) ozone analyses provide a realistic representation of the atmospheric ozone distribution, we compare the run 3 analyses with independent observations. This will provide us with information on the performance of the assimilation system with a view to the eventual implementation of the approach in run 3 to assimilate Envisat data. The independent data used for this comparison are ozonesondes, HALOE profile data, and TOMS total column ozone data. Some caution must be taken with the comparison of assimilated fields with independent data. Biases may exist between the information used in the assimilation system and the independent data. It is outside the scope of this paper to fully address these issues with respect to the ozonesonde, HALOE and TOMS comparisons.

4.1. Ozonesonde Comparison

[64] A set of ozonesonde ascents have been compared with co-located analyses profiles. Ozonesondes provide high quality data but, over the short period of this study, there are only a limited number of ascents available. This means it is not possible to generate global residual statistics from this data set. Further, there is a difference in representativeness between the ozonesondes and the assimilated ozone values which should be considered. The ozonesonde observations are point measurements whereas the assimilated ozone fields represent the model grid box values, thus they represent slightly different quantities. For these reasons we use the ozonesonde comparisons to provide a qualitative indication of the validity of the assimilated ozone fields.

[65] Figure 6 shows four examples of co-located ozonesonde profiles, analyses profiles for assimilation run 3, and profiles for the Fortuin ozone climatology. The four loca-

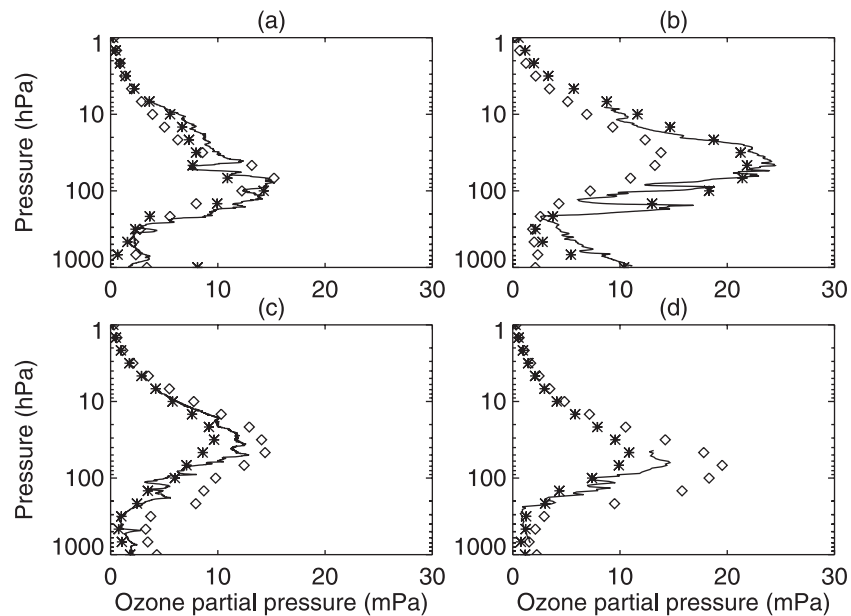


Figure 6. Four examples of co-located ozonesonde profiles, analysis profiles for assimilation run 3 and profiles of the Fortuin climatology (milliPascals, mPa). The stars represent analysis values plotted on standard UARS pressure levels. The diamonds represent Fortuin climatology values plotted on standard UARS pressure levels. (a) Ny Alesund 27 April 1997 (12 GMT). (b) Payerne 25 April 1997 (12 GMT). (c) Lauder 16 April 1997 (12 GMT). (d) South Pole 18 April 1997 (12 GMT). See text for details.

tions of the ozonesonde measurements are Ny Alesund (78.9°N, 11.9°E), Payerne (46.8°N, 7.0°E), Lauder (45.05°S, 169.7°E) and South Pole (90°S). The analyses profiles (denoted by the stars), and the profiles of the Fortuin climatology (denoted by the diamonds), are plotted on standard UARS pressure levels.

[66] For the Northern Hemisphere cases, the run 3 analyses profiles agree well with the ozonesonde profiles, the differences in most cases being within 20% of the ozone partial pressure measured by the ozonesondes. For the Ny Alesund ascent, the run 3 analyses are able to partially resolve the layer of reduced ozone measured by the ozonesonde at approximately 50hPa.

[67] For the Southern Hemisphere cases the run 3 analyses profiles agree well with the ozonesondes between 1000hPa and 100hPa, but underestimate the partial pressures between approximately 50hPa and 20hPa.

[68] Note that for Lauder and the South Pole, the run 3 analyses show qualitatively good agreement with the ozonesondes even though there is no MLS data available at Lauder, and there is no MLS and GOME data available at the South Pole. In this case, the data assimilation has propagated information from the data rich areas to the data poor areas using the governing equations of the atmosphere.

[69] Run 3 retains the ability to resolve some of the vertical structure in the ozone profile seen in the MLS only (run 1) analyses. The run 1 analyses, however, do not agree well with the sondes in the troposphere, where there is little or no information content in the MLS data, as shown by the MLS averaging kernels [Froidevaux *et al.*, 1996]. The GOME only (run 2) analyses are generally unable to capture the vertical structure seen in runs 1 and 3 because no profile information is being introduced by the assimilation system

(see Burrows *et al.* [1999] for a discussion of GOME averaging kernels).

[70] Figure 6 shows that the analyses agree better with the sondes than the Fortuin climatology.

4.2. HALOE Comparison

[71] For the HALOE comparison we use the version 19 retrieved product obtained from the BADC archive. The HALOE instrument is capable of measuring on average 15 sunrise and 15 sunset profiles each day. The retrieved HALOE measurements are reported on standard UARS pressure levels with useful information ranging from approximately 200hPa to 0.1hPa. There are only very minor differences between version 19 HALOE ozone and the previous version 18 product (see <http://haloedata.larc.nasa.gov/home.html>). HALOE ozone measurements have been validated by Bruhl *et al.* [1996] who find agreement to within 5% when compared with a number of independent observation types.

[72] For the period of this study, the latitude range covered by the HALOE instrument was 57.9°S to 35°N. For each observation, the analysis valid at the closest synoptic hour was interpolated horizontally to the HALOE location and vertically to each HALOE observation level. The difference between these two profiles (HALOE - analysis) was then taken, and the statistics of the HALOE/analysis differences accumulated over all HALOE measurements. Residual statistics on the difference between run 3 (MLS plus GOME) analyses profiles and co-located HALOE profiles, averaged over four latitude bands are plotted in Figure 7.

[73] In the latitude range north of 30°S (where there is MLS data) the run 3 analyses reproduce the independent

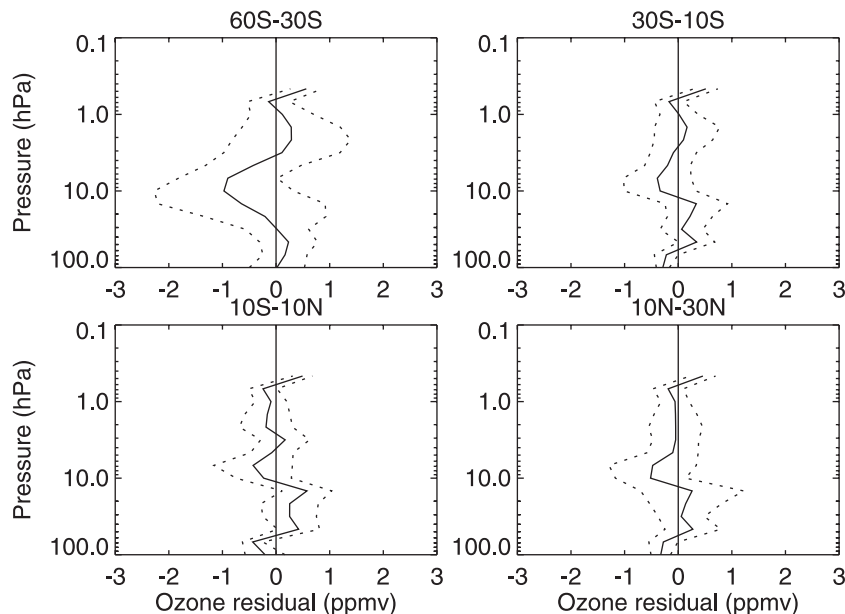


Figure 7. Comparison between co-located version 19 HALOE ozone profiles and run 3 analysis profiles, for four latitude bands spanning the range of latitudes sampled by the HALOE instrument over the assimilation period. Solid lines show residual mean (HALOE minus analysis), and dashed lines show the 1σ range of the residual distribution (both in ppmv). Positive values indicate that the HALOE observations are larger than the analyzed values.

HALOE data. The mean residuals for these latitudes tend to lie within the range of ± 0.5 ppmv, and the 1σ range of the residuals tends to lie within ± 1 ppmv of the zero-line. As a percentage of the mean HALOE volume mixing ratio, the mean residuals lie within $\pm 6\%$ between approximately 40hPa and 1hPa with the 1σ range being within $\pm 15\%$ over the same pressure range. Below 40hPa, the analyses and HALOE measurements do not agree as well and can differ by up to 200% at 100hPa.

[74] At latitudes south of 30°S the analysis is overestimating the ozone volume mixing ratio by approximately 15% at 10hPa, due to the lack of MLS measurements correcting the profile before the GOME part of the assimilation. In general the agreement is within 20%, except at 100hPa where some discrepancies remain.

[75] In general, the results from run 3 are close to those from run 1 (MLS only; not shown), except in the region where there are no MLS observations. There is a small improvement in the run 3 results at altitudes below 50hPa compared with the run 1 results, which arises because of the introduction of the GOME measurements in the assimilation. For the latitude range 60°S to 30°S the run 3 results match closely the run 2 (GOME only) results. This is because only GOME observations are directly influencing the analysis in this latitude range.

[76] The results for run 2 (not shown) are significantly worse than the equivalent statistics for the runs 1 and 3. The analyses overestimate the ozone volume mixing ratio around the 10hPa level by up to 2 ppmv in the mean. Also, between 30°S and 30°N the analyses underestimate the ozone volume mixing ratio around the 20–30hPa level with respect to the HALOE observations. The biases observed in the HALOE/run 2 residuals are relatively uniform with latitude and are similar in structure and magnitude to the MLS/run 2 biases. Section 3.2 discusses the factors which

might influence the vertical structure of the analyzed ozone fields from run 2.

4.3. TOMS Comparison

[77] We now compare total ozone calculated from our analyses with independent ADEOS TOMS measurements. We vertically integrated the analysis fields using the same observation operator as used in the assimilation (see equation (3)). The calculated total ozone columns are compared with daily gridded version 7 TOMS data obtained from the BADC. Our 1200 GMT analyses are used to calculate the total column ozone, whereas the gridded TOMS data is derived from 24 hours of accumulated observations, which means the two fields are not exactly equivalent. The TOMS product is reported on a 1° latitude by 1.5° longitude grid. We interpolate the TOMS data to the UM horizontal grid, where the differences are calculated, and residual statistics over all days of the assimilation excluding the spin-up are generated.

[78] *Krueger et al.* [1998] estimate an absolute error in the ADEOS TOMS total ozone of 3% and a random error of 2%. When compared with a network of 45 ground based stations the TOMS product was found to be consistent with the quoted uncertainties. On this basis, we assume a TOMS error (random plus systematic) of 5%.

[79] Figure 8 gives the mean and 1σ range of the total ozone residual distribution (TOMS minus analysis) as a function of latitude for run 3 (MLS plus GOME). Results for run 2 (GOME only; not shown) are similar to those of run 3.

[80] For run 3, there is good agreement between the TOMS and analyzed total ozone in the Northern Hemisphere, the absolute value of the mean difference being less than 5 DU for most of the Northern Hemisphere.

[81] In the Southern Hemisphere the comparison between the analyses and TOMS observations for run 3 is worse than

for the Northern Hemisphere (note that there is no MLS data south of 34°S). The mean difference rises to approximately 30 DU at 60°S, with the analyses underestimating the TOMS measurements. The GOME residuals at the same latitudes show a bias of less than 5 DU which implies there is a bias between the TOMS and GOME observations at these southern latitudes. A positive bias in TOMS ozone columns with respect to GOME total ozone has been reported [Corlett and Monks, 2001], in both southern and equatorial latitudes. The magnitude of this bias matches well with the bias seen in our comparison of TOMS and analyzed total ozone. This result shows that the data assimilation method has the potential to identify biases between independent observations.

[82] For latitudes north of about 50°S, the mean difference between the run 3 analyses and the TOMS measurements is less than the 1σ range of the TOMS residuals. For all latitudes where there are TOMS data, this difference is less than the 2σ range of the residuals.

[83] The total ozone comparison for run 1 (MLS only; not shown) is significantly worse than the equivalent statistics for the runs 2 and 3. The analyses underestimate the total ozone in the mean by more than 20 DU for much of the Northern Hemisphere and overestimate the total ozone in the Southern Hemisphere by up to 100 DU with respect to the TOMS data. The differences between the analyses and TOMS can mainly be ascribed to the lack of constraint on the total ozone from not using GOME observations. Nevertheless, where there is MLS data (as in the Northern Hemisphere), and despite the lack of a constraint on the total ozone, the analyses qualitatively capture the major features of the total ozone field.

5. Conclusions and Future Work

[84] In this paper we have presented results from a three dimensional data assimilation system based on the Meteorological Office troposphere-stratosphere NWP assimilation scheme. The system has been modified to assimilate UARS MLS ozone and temperature profiles and ERS-2 GOME total column ozone, in conjunction with the standard meteorological observations, and has been run for a three-week period in April 1997 to evaluate the ozone analyses produced.

[85] Three assimilation runs were performed, one run with only MLS ozone observations being assimilated, one run with GOME only and a final run with both observational data sets being used by the assimilation scheme. The paper presents evidence which suggests that the three-week period is adequate to assess the merits of the different combination of observations. Statistical information produced by the assimilation has been used to evaluate the ozone analyses.

[86] The results of the statistical information produced by the analyses indicate that the MLS and GOME configuration performs better than either the MLS only or the GOME only configuration (these results also provide confidence in the assumption of normally distributed observation and background errors). In particular, the MLS and GOME configuration is not inconsistent with both the MLS and GOME observational data sets, and is more consistent with the expected results from a BLUE [Talagrand, 1998]. These

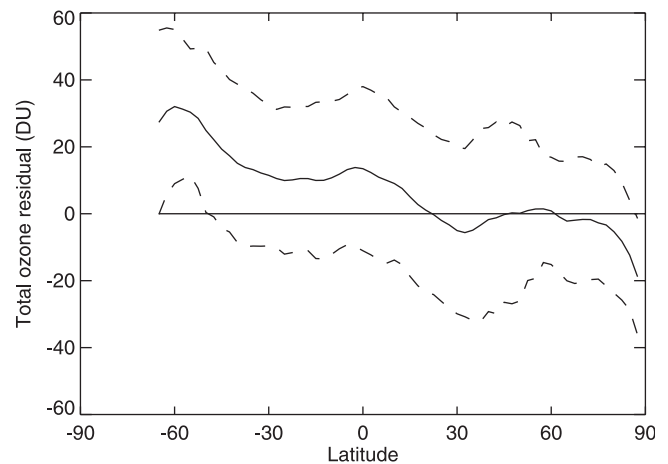


Figure 8. Comparison between gridded version 7 ADEOS TOMS total ozone and run 3 analyzed total ozone as a function of latitude. Solid line shows residual mean (TOMS minus analysis), and dashed lines show the 1σ range of the residual distribution (both in DU). Positive values indicate that the TOMS observations are larger than the analyzed values.

results are corroborated by the comparisons between the analyses from the different assimilation configurations and independent information (ozonesondes, HALOE profiles and TOMS total ozone measurements).

[87] The comparison with independent information also shows that the assimilation of MLS plus GOME analyses provides a realistic representation of the atmospheric ozone distribution in regions where there are MLS and GOME data.

[88] In regions where there are no MLS and/or no GOME data, the differences between the analyses and the independent information are somewhat larger than the differences for the regions where there are MLS and GOME data. In the regions where there are no MLS and/or no GOME data, the analyses tends to overestimate the HALOE data, and underestimate the ozonesonde and TOMS data. The negative bias against TOMS data can be explained by a bias between the TOMS and GOME measurements. Evidence suggests that the positive bias against HALOE data could be due to biases in the model. A contribution to this bias from the relatively simple background error covariance for ozone cannot be ruled out (this needs to be tested, and will be the subject of future work). The significance of the negative bias against ozonesonde data cannot be assessed given the limited number of ascents available for comparison.

[89] Overall, the combination of MLS and GOME observations via the assimilation system produces analyzed ozone fields that are consistent with both independent profile and total column measurements. The global RMS residual value (i.e., the RMS difference between the analyses and the independent observations) for the HALOE observations averaged over all levels ranges from 7% to 10% (depending on the pressure level - lowest values are in the stratosphere), and from 5% to 15% (depending on latitude - highest values are for latitudes south of 30°S where there is a bias between GOME and TOMS) for the

TOMS ozone columns. These values, which are expressed as a percentage of the HALOE and TOMS observations, are comparable to the quoted errors in the HALOE and TOMS instruments (5% in each case). The effect of combining the MLS and GOME observational data sets is to correct some of the deficiencies present when the same observations are assimilated separately.

[90] The results of this paper suggest that the combination of ozone profile and total column ozone information from instruments on board Envisat will provide better analyses than using the profile or column information separately, and that these analyses will provide a realistic representation of the atmospheric ozone distribution.

[91] Preliminary results (not shown) suggest that for the combined assimilation of profile and total column ozone, useful quantitative information on the tropospheric ozone column is being introduced via the assimilation system. Further work is required to fully characterize this result. Nevertheless, the results are encouraging, and the methodology will be applied to Envisat data sets to extract information on tropospheric ozone.

[92] There is wide scope for improvement of the assimilation system described in this paper. A number of areas of future development have been identified: (1) We intend to change the analysis system from the AC scheme to the 3D-var system currently operational at the Meteorological Office [Lorenc et al., 2000]. The 3D-var system provides a more statistically optimal use of the observations than the AC scheme. It also provides more flexibility in the use of observational data than the AC scheme. (2) We will adjust the spreading of the total ozone increments in the vertical for the GOME only component of the MLS plus GOME assimilation, to investigate potential improvements to the analyses. (3) We will investigate improvements of the estimates of the ozone background error covariances within the computational constraints of an NWP system. (4) We will investigate improvements in the calculation of the ozone photochemistry within the computational constraints of an NWP system. (5) We intend to modify the 3D-var system to incorporate ozone and other observations measured by instruments on board the Envisat platform. This will allow us to continue studying how combining different observation data sets through the assimilation process affects the final analyzed ozone fields.

[93] **Acknowledgments.** We would like to thank Henk Eskes (KNMI), who helped with the implementation of the Cariolle scheme, and Richard Rood (DAO) for his useful comments on our work. We would like to acknowledge the technical and scientific support of the Meteorological Office, particularly Penny Boorman, and the BADC for providing access to the MLS, HALOE and TOMS data (TOMS data is provided by NASA GSFC). The ozonesonde data used in this publication was obtained as part of the Network for the Detection of Stratospheric Change (NDSC) and is publicly available (see <http://www.ndsc.ncep.noaa.gov>). We would like to thank the reviewers and the editor for comments which helped improve the paper. Thanks are given to Eric Guilyardi (CGAM) for helping with one of the plots. This work was funded by the U.K. Natural Environment Research Council.

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