

**Forecasts and
analyses of the ozone
hole 2008**

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Forecasts and assimilation experiments of the Antarctic Ozone Hole 2008

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Received: 4 February 2010 – Accepted: 23 March 2010 – Published: 9 April 2010

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Published by Copernicus Publications on behalf of the European Geosciences Union.

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Abstract

The 2008 Antarctic ozone hole was one of the largest and most long-lived in recent years. Predictions of the ozone hole were made in near-real time (NRT) and hindcast mode with the Integrated Forecast System (IFS) of the European Centre for Medium-Range Weather Forecasts (ECMWF). The forecasts were carried out both with and without assimilation of satellite observations from multiple instruments to provide more realistic initial conditions. Three different chemistry schemes were applied for the description of stratospheric ozone chemistry: (i) a linearization of the ozone chemistry, (ii) the stratospheric chemical mechanism of the MOZART-3 chemical transport model (CTM) and (iii) the relaxation to a climatology as implemented in the TM5 CTM. The IFS uses the latter two schemes by means of a two-way coupled system. Without assimilation, the forecasts showed model-specific shortcomings in predicting start time, extent and duration of the ozone hole. The assimilation of satellite observations from the Microwave Limb Sounder (MLS), the Ozone Monitoring Instrument (OMI), the Solar Backscattering Ultraviolet radiometer (SBUV-2) and the SCanning Imaging Absorption spectroMeter for Atmospheric CartographY (SCIAMACHY) led to a significant improvement of the forecasts when compared with total columns and vertical profiles from ozone sondes. The combined assimilation of observations from multiple instruments helped to overcome limitations of the ultraviolet (UV) sensors at low solar elevation over Antarctica. The assimilation of data from MLS was crucial to obtain a good agreement with the observed ozone profiles both in the polar stratosphere and troposphere. The ozone analyses by the three model configurations were very similar despite the different underlying chemistry schemes. During the chemically instigated development of the ozone hole, differences quickly developed in the initialized forecasts by the different schemes. The predictions of the ozone-hole closure, which is driven mainly by dynamical processes, benefited from the initialization over a much longer forecast length. The characteristics of the individual chemistry schemes became apparent after the third forecast day but the forecasts were still close to the respective analyses. The

ACPD

10, 9173–9217, 2010

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initialization with ozone analyses was beneficial up to at least 15 days.

1 Introduction

The paper presents forecasts of the ozone hole over Antarctica in 2008, which were made with and without assimilation of ozone retrieval products from multiple sensors to provide ozone initial conditions, and which used different chemistry schemes of varying complexity.

The ozone hole in 2008 was large and long-lasting. Its size at the beginning of October was larger than 90% of the events since 1979 (WMO, 2008). The stratospheric ozone layer over Antarctica has shown severe catalytic ozone destruction in austral spring since the late 1970s and similar phenomena are observed over the Arctic (WMO/UNEP, 2007). Reductions in chlorofluorocarbon emissions imposed by the Montreal protocol and its amendments are predicted to lead to a recovery of springtime stratospheric ozone over the poles, but it will take several decades before the ozone layer will be healed (Newman et al., 2006). Hence, observations and forecasting of the stratospheric ozone depletion remain important to assess the increase in UV radiation at the surface and to monitor the recovery of the ozone layer.

Ultraviolet to visible range (UV-VIS) instruments provide the longest record of ozone measured from space, and sensors such as the Ozone Monitoring Instrument (OMI) and the SCanning Imaging Absorption spectroMeter for Atmospheric CartographY (SCIAMACHY) can provide high resolution ozone total column observations. However, the restricted sun light at the ozone-hole onset limits the use of UV-VIS sensors at this time. The Microwave Limb Sounder (MLS) does not depend on sun light and can make profile observations over Antarctica during the whole year, albeit with a low horizontal resolution. Modelling the correct timing and magnitude of the Antarctic ozone hole involves a complex interplay between dynamical phenomena, i.e. the strength of the Antarctic vortex, slow heterogeneous chemical processes during the austral winter and the rapid release of chlorine and bromine when sun light reaches the polar region

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(Solomon, 1999).

Ozone has been included in global numerical weather prediction (NWP) models as a prognostic variable since the mid-1990's (Derber and Wu, 1998) in order to assimilate satellite observations of ozone within the NWP data assimilation framework. Besides the monitoring of the man-made ozone depletion, the crucial role of ozone in the atmospheric radiation budget and its link to wind fields (Allaart et al., 1993; Riishojgaard, 1996) through its tracer characteristics motivated the introduction of the ozone variable. The underlying ozone chemistry schemes are often simplifications of stratospheric processes derived from parameterizations of Chemical Transport Model (CTM) results (Cariolle and Deque, 1986; McLinden et al., 2000 and McCormack et al., 2006) or observed climatologies (Fortuin and Kelder, 1998). For example, Geer et al. (2007) compare different ozone assimilation systems based on linear chemistry schemes. As pointed out in a review by Lahoz et al. (2007), using data assimilation systems based on CTMs (e.g. Khattatov et al., 2000; Ménard et al., 2000; Errera et al., 2008; Massart et al., 2009) has been an alternative to the NWP approach. The CTM systems can apply more complex chemical schemes and give the opportunity to study the chemical impact on other species during the ozone assimilation.

At the European Centre for Medium-Range Weather Forecasts (ECMWF), ozone has been operationally assimilated and forecast using an updated version of the linearized scheme by Cariolle and Deque (1986) since 1999 (Hólm et al., 1999), and it was included in the ERA40 re-analysis (Dethof and Hólm, 2004). Within the "Global and regional Earth-system Monitoring using Satellite and in-situ data" (GEMS) project (Hollingsworth et al., 2008), the ECMWF's integrated forecast system (IFS) was extended to be able to simulate and assimilate, using its 4D-VAR implementation, reactive gases such as ozone, carbon monoxide, nitrogen dioxide, formaldehyde and sulphur dioxide in both the troposphere and the stratosphere. This has been achieved by means of a coupled system (Flemming et al., 2009) in which the IFS is linked to one of the CTMs MOZART-3 (Horowitz et al., 2003; Kinnison et al., 2007) and TM5 (Krol et al., 2005, version KNMI-cy3-GEMS). In the coupled system IFS-CTM, the IFS

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makes use of the simulation of chemistry, emission and deposition in the CTM without directly simulating these source and sink processes in the IFS. The MOZART-3 chemical mechanism explicitly simulates ozone chemistry whereas the TM5 version used here applies a relaxation to a stratospheric ozone climatology. The new coupled system combines the NWP approach with the CTM-based approach for simulation and assimilation of atmospheric constituents in both the stratosphere and the troposphere.

An important practical application of IFS-CTM is the near-real-time (NRT) provision of boundary conditions, in particular in the free troposphere, for regional air quality forecasts (<http://www.gmes-atmosphere.eu/services/raq/>). It is therefore essential that the ozone assimilation by the coupled system IFS-CTM does not degrade the ozone values in the troposphere. This is a difficult task because of the dominance of stratospheric ozone in the total column and the limited vertical information of satellite observations. Therefore, in this study attention is also given to the tropospheric profiles in the considered region.

Ozone retrievals from OMI, SCIAMACHY, Solar Backscattering Ultraviolet radiometer (SBUV-2) and MLS have been assimilated in this study. The MLS retrievals have the potential of being important for the quality of the ozone analyses because of the ability of MLS to measure during the polar night and to provide vertical profile information between the lower mesosphere and the stratosphere. While many studies (e.g. Levelt, et al., 1998; Eskes et al., 2002; and Geer et al., 2006) focus on the assimilation of only one specific instrument, ozone profile retrievals from MLS have been assimilated together with retrievals from one UV-instrument such as SBUV-2 (Jackson and Orsolini, 2008) or OMI (Stajner et al., 2008). The use of multiple instruments raises the question of the inter-instrument biases since they could degrade the analyses. The precise quantification of these biases in ozone hole conditions, as shown in this paper, is complicated by the different sampling of the instruments, the reduced retrieval quality at low solar elevation and the pronounced horizontal ozone gradients.

In addition to evaluating the quality of the ozone assimilation and chemistry, this paper also investigates the time span over which the initialization with ozone analy-

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ses improves the forecast for each chemical scheme. This “chemical predictability” of ozone has to be distinguished from the “meteorological predictability” of the underlying dynamical processes. Most of the variability of stratospheric ozone on the global scale is caused by dynamical processes. For example, anomalies of the geopotential height in the stratosphere have been successfully used as a predictor for the ozone variability (Long et al., 1996), and Sekiyama and Shibata (2005) identify the predictability of the 100 hPa geopotential height as the limiting factor for the predictability of the ozone anomalies. Simmons et al. (2005) show that the large dynamical features of the vortex, even during the vortex split event in 2002, can be well predicted.

The lifetime of ozone in the stratosphere varies with height from a couple of hours in the upper part to a couple of weeks in the lower part according to the Chapman Cycle chemistry. Eskes et al. (2002) obtain a predictability range of 4 to 5 days based on anomaly correlations in respect to monthly means for the Southern Hemisphere. Tegtmeier and Shepherd (2007) report much longer lasting, i.e. half a year and more, correlations of stratospheric ozone anomalies in the extra-tropics, which they attribute to anomalies in odd nitrogen compounds. On the other hand, the lifetime of ozone is significantly shorter during the development of the ozone hole. Hence, the quality of the ozone hole forecast will depend much more on the correct representation of the chemical processes under these circumstances.

While the dynamical processes determine the spatial patterns of the ozone fields, the biases of the simulated ozone concentrations are more related to errors in the simulation of the sink and source processes, although an exaggerated Brewer-Dobson circulation may also lead to positive biases in the mid-latitudes of the winter-hemisphere (van Noije et al., 2004). If these biases are corrected by using ozone analyses as initial conditions, the effect might be long-lasting because of the long-lasting impact of ozone anomalies mentioned above. The correct initialization of stratospheric ozone fields is therefore not only important for NRT forecasts but also for independent CTM runs.

The remainder of the paper is structured as follows. The three different chemistry schemes and the model configuration are described in Sect. 2. The data assimila-

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tion set-up and the assimilated observations, including an investigation of the inter-instrument biases, are also introduced in this section. Section 3 contains the evaluation of the ozone forecast with and without ozone assimilation in the period from August to December 2008. The first step is the validation against total column values of ozone sondes to evaluate the analyses with independent data. In the next step, the ozone hole size below 220 Dobson Units (DU) is used as a compact parameter to study the forecasts with different forecast lead times and initializations. The last step in the evaluation looks at the shape of the forecast ozone profiles and identifies the impact of the assimilated MLS data. The paper ends with conclusions and a summary of the results in Sect. 4.

2 Model and experiment set-up

2.1 Three schemes for stratospheric ozone chemistry

The model configurations of the IFS for this study differed only in their simulation of the ozone chemistry. The IFS used (i) a linearized scheme, which is a further development of the scheme by Cariolle and Deque (1986), (ii) a full chemistry scheme implemented in the CTM MOZART-3 and (iii) a climatological approach, which is the relaxation to the ozone climatology as implemented in the CTM TM5. The latter two schemes have been made accessible to the IFS by means of a two-way coupled system that links the IFS to the CTMs MOZART-3 and TM5. The coupled system is described and validated in Flemming et al. (2009). The coupled CTMs, which are driven by IFS meteorological data, provide the IFS with the tendencies of sink and source processes which are not directly modelled in the IFS. The chemical conversion rates from the linear scheme are calculated as part of the IFS code.

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2.1.1 Linear scheme (IFS)

The parameterization is a description of the ozone chemistry in the stratosphere. The temporal change due to chemical sources and sinks of ozone is simulated in the following way:

$$\frac{dO_3}{dt} = c_1 + c_2(O_3 - \overline{O_3}) + c_3(T - \overline{T}) + c_4\left(\int_p^0 O_3 - \int_p^0 \overline{O_3}\right) + c_5(\overline{Cl_{eq}})^2 O_3$$

The change depends on the deviations of the ozone concentration O_3 , temperature T and overhead ozone columns $\int_p^0 O_3$ from the respective mean modelled 2-D photochemical equilibrium state \overline{T} and $\overline{O_3}$ (Cariolle and Teyssedre, 2007). Further, the formulae includes a parameterization of the rapid ozone loss due to the chlorine catalytic cycle which is based on a prescribed global equivalent chlorine content $\overline{Cl_{eq}}$ and which is triggered at temperatures below 195 K in daylight conditions.

2.1.2 Full scheme (IFS-MOZART)

The MOZART-3 chemical mechanism simulates the tropospheric and stratospheric chemistry with 106 species. The chemical mechanism is described in Kinnison et al. (2007) and is based mostly on the data from Sander et al. (2006). It also includes the ozone destruction caused by catalytic chemistry of halogenic compounds and their activation by heterogeneous reaction on the surfaces of polar stratospheric clouds (PSC). The PSCs simulated by the MOZART-3 are liquid binary sulphate, nitric acid trihydrate (NAT), supercooled ternary solution and water ice.

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2.1.3 Climatological scheme (IFS-TM5)

TM5 simulates tropospheric ozone with a modified CBM4 mechanism (Houweling et al., 1989). It does not apply any representation of stratospheric chemistry; instead its ozone field in the stratosphere is constrained by a climatology. This is based on a monthly varying, pressure dependent, zonal-average ozone concentration field representative for the years 1980–1991 (Fortuin and Kelder, 1998). Ozone is nudged to the climatology above 45 hPa in the tropics, and above 90 hPa in the subtropics. The relaxation times are 2.5 and 4 days, for the tropics and subtropics, respectively.

2.2 Data assimilation method

The ozone satellite observations were assimilated by ECMWF's incremental formulation of the four-dimensional variational data assimilation (4D-Var) method. In 4D-Var, a cost function is minimized to combine the model fields and the observations over a given time period in order to obtain the best possible representation of the ozone field. The resulting ozone analyses were used as initial conditions for the ozone forecast.

A description of the 4D-VAR data assimilation algorithm of the IFS is given by Mahfouf and Rabier (2000) and with focus on the assimilation of aerosol in Benedetti et al. (2009). A detailed description and examples of the application of the coupled system IFS-MOZART for the assimilation of atmospheric constituents can be found in Inness et al. (2009).

The implementation of the chemistry simulation into the 4D-VAR apparatus with the coupled system follows the assimilation of ozone with the parameterized chemistry (Dethof and Hólm, 2004), which means that the chemistry scheme is applied to determine the increments between the model and the observations but it is not included in the minimization by the adjoint and tangent linear model which is applied to these increments. The adjoint and tangent linear model accounts only for transport processes. The background errors of ozone were derived from an ensemble of forecasts (Fisher and Andersson, 2001) applying the linear ozone chemistry scheme. The background

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error matrix is represented in a wavelet formulation (Fischer, 2006). The same background error matrix was used in all assimilation runs. The 4D-Var time window had a length of twelve hours.

The assimilation of ozone data with all three model configurations is statistically univariate in the sense that no background error covariance terms between the chemical tracers or the meteorological fields are defined. The impact of the ozone data assimilation on wind and temperature fields has been switched off. A possible impact on the rest of the chemical species in MOZART-3's full chemistry scheme appeared only as a consequence of the chemical interaction in the coupled CTM, which uses the analysed ozone fields as initial conditions every 12 h (Flemming et al., 2009). A detailed investigation of the adaptation of the other chemical species has not been made but the identified influence appeared to be small.

2.3 Assimilated satellite observations

Table 1 list the assimilated satellite observations. OMI and SCIAMACHY retrievals are total column data whereas MLS and SBUV-2 retrievals are partial columns at mostly stratospheric levels. Comprehensive validation studies of the retrieval with ground based measurements were presented by Balis et al. (2007) for OMI, Froidevaux et al. (2008) for MLS, by Eskes et al. (2005) for SCIAMACHY and Bhartia et al. (1996) for SBUV-2.

As part of ECMWF operational activities, the ozone observations are monitored in terms of inter-instrument biases and their impact on the analysis before they are actively used in operational mode (Dragani, 2009). This procedure results in a quality control that prevents observations from being assimilated if their accuracy is low. Blacklist criteria have been developed to exclude data observed at low sun elevation angles (see Table 1). Further, the high-resolution data from OMI and SCIAMACHY are thinned by randomly choosing one observation within each $1^{\circ} \times 1^{\circ}$ grid box. Thinning is applied to minimise the spatial correlation between the observation errors, which is not accounted for within the 4D-VAR implementation. Retrievals from the Global Ozone

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Monitoring Instrument (GOME-2) are currently being tested for their use in the NRT experiments.

The data provider's observation-error data were used, but they were modified to be at least 10% for OMI and SBUV. The original retrieval errors were increased because they did not include the representativeness error of the observation for the respective model grid box. In the considered area between 60°–90° S, the errors of OMI and SCIAMACHY ranged from 20% to 30%. The error of the stratospheric partial columns was about 5–10% above 68 hPa for MLS and above 16 hPa for SBUV-2, and up to 25% for the partial columns below.

Figure 1 shows the number of the assimilated observations, i.e. the ones passing the quality test, for the period 1 August to 31 December 2008 between 60°–90° S. The sensors using solar UV-VIS radiation (SCIAMACHY, OMI, and SBUV) were only gradually providing more usable observations with increasing length of the solar day. The microwave observations of MLS were available in the same quantity over the whole period and were therefore the dominant information source until mid-October. The number of the SBUV-2 observations was small compared to that of the other instruments, which led to their generally very small influence during the assimilation. All observations apart from MLS were received in NRT and the hindcast experiments used these data sets without retrospective corrections. MLS data have been received and assimilated in NRT since March 2009.

The study of inter-instrument biases has to consider that the instruments measure at different times and locations, which is of importance because of the large horizontal gradients during the ozone-hole period. Figure 2 shows time series of the 24-hourly spatial averages over 60°–90° S of the assimilated total column observations from OMI and SCIAMACHY and the sum of the partial columns from MLS and SBUV-2. The typical values of the MLS partial columns in the lowest 10 levels, i.e. between 215 and 5 hPa, was about 15 DU per level and about 2 DU in the levels above. The lowest SBUV-2 partial column, i.e. below 16 hPa, contained about 75% of the total column, and most of the remaining ozone mass was part of the column above, i.e. between

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6 and 16 hPa. The data sets from all instruments showed the same overall temporal variation but with large biases until the end of October. The MLS added columns were lower than the UV sensors by about 50 DU until this point in time. The difference was mainly caused by the fact that MLS observed the area poleward to 82° S all the time. It was therefore able to capture a larger proportion of the ozone hole. The missing tropospheric contribution, i.e. below 215 hPa, in partial columns in the MLS data (see Table 1) led only to a small bias. The UV-VIS sensors showed a considerable spread until the end of October. This variability seemed also be largely caused by the different locations of the assimilated observations. This is supported by the fact that a 12-hourly accumulation of the observations, i.e. at the length of the assimilation window and not over the whole day, led to a diurnal variation of up to 50 DU. The high values from SBUV-2 on NOAA-16 in the second half of October have been identified as a sensor fault.

More detailed insight into the inter-instrument biases can be obtained from comparing the departures of the observations from a three-dimensional ozone field. By doing so the observed partial column values can better be compared with observed total column values at different locations. Figure 3 shows the frequency distribution of the analysis departures of IFS-MOZART for OMI, SCIAMACHY and MLS against latitude, averaged over one week at the end of August and the end of November. The analysis departures from the partial MLS columns were symmetric and had nearly zero average at all latitudes between 60° S-90° S for all levels as a whole (see Fig. 3) and also for the individual levels (not shown). This means that the analysis was bias-free against this data set, which covered the stratosphere only, both at the end of August and November. At the end of August, OMI's total column values had a bias of about 4 DU against the analysis with the highest bias occurring at lower solar elevation. The SCIAMACHY total columns were on average bias free but showed small negative biases at higher solar elevation at the end of August. Many more observations from OMI and SCIAMACHY had been assimilated at the end of November. At this time the OMI total columns were almost bias-free against the analysis and therefore also against MLS.

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The SCIAMACHY values had a mostly positive bias against the analysis of about 4 DU when averaged over the whole domain. When binned according to solar elevation (not shown), the SCIAMACHY bias was highest at low solar elevation where it reached an average value of 12 DU. The lowest SBUV-2 columns, i.e. below 16 hPa, had an overall negative bias of about 2 DU at the end of November. SBUV-2 on NOAA 18 showed a larger negative bias at low solar elevation.

Figure 3 shows the latitude range for which satellites observations were assimilated. While MLS always covered the area polewards to 82° S, OMI and SCIAMACHY observations had not been used south of about 70° S by the end of August. By the end of November observations over the whole domain were assimilated.

In summary, the biases between the instruments varied in time, reached values of up to 3% and were highest at low solar elevation.

2.4 Experiment set-up

The IFS forecasts evaluated in this paper (Table 2) differed in the model set-up, i.e. the choice of the applied chemical scheme, and whether ozone analyses were used to initiate the forecast. The forecasts were started at 00:00 UTC every day in the period from 1 August to 31 December 2008. The meteorological initial conditions for each forecast were derived from the operational ECMWF analysis. The ozone initial conditions were taken either from the previous forecast (FC) or from an assimilation experiment which produced analyses at 00:00 and 12:00 UTC in two 12 h 4D-VAR cycles every day (ANA).

The assimilation experiments were run for each model set-up, i.e. each chemical scheme, and the analyses were used for the forecast with the respective scheme. The assimilation experiments used observations from SCIAMACHY, SBUV-2, OMI and MLS. Additionally, the NRT forecast by IFS-MOZART (IFS-MOZ-ANA*), for which MLS data were not available, was evaluated.

To further study the influence of the chemical scheme on the forecast quality, additional one-day forecasts (FC15) were carried out which were initialized from the previ-

ous forecast as the FC runs on all days apart from the 1st and 16th day of each month, for which they were initialized by the respective ozone analysis.

The IFS was run with a T159 spectral resolution and the grid point space was represented by a reduced Gaussian grid (Hortal and Simmons, 1991), which has a grid box size of about 125 km. The vertical discretization consisted of 60 hybrid sigma-pressure levels, reaching up to 0.1 hPa. The CTMs MOZART-3 and TM5 were run in the coupled experiments on a regular latitude-longitude grid of $1.9^\circ \times 1.9^\circ$ and $2^\circ \times 3^\circ$ grid box length respectively; they used the same vertical discretization as the IFS. The emission data for MOZART-3 and TM5 were based on the year 2000 inventory from the RETRO project (<http://retro.enes.org>). Monthly averages of Global Fire Emission Database (GFEDv2, van der Werf et al., 2006) for the period 1997–2004 were used as wild-fire emissions.

3 Evaluation of the forecasts

3.1 Synoptic overview

The 2008 ozone hole started to grow rapidly after mid-August, which is about 2–3 weeks later than the average start time of the last ten years. It reached its maximum in mid-September, being at this time only slightly smaller than the biggest recorded ozone hole in 2007. The 2008 ozone hole lasted longer than most previous events and ended in December. Its size in mid-October was larger than 90% of the years since 1979 according to WMO (2008). Van Peet et al. (2009) present GOME-2 profile retrievals from September to December 2008, which agreed well with sonde profiles from Neumayer station (see Table 3).

The temperature at 50 hPa was below the 1979–2007 average on most days from June to August. The volume below the threshold temperature for the formation of PSCs type I (NAT) was well above the long-term average, in particular in early September. The vortex was stable and mostly concentric (WMO, 2008). A comprehensive evalu-

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ation of the meteorological forecasts and analyses is beyond the scope of this paper. Exemplarily, Fig. 4 shows time series of the monthly mean observed temperatures and the respective forecast errors at Neumayer Station from May to December 2008. The temperatures below the minus 85 °C threshold of the formation of ice clouds (PSC type II) occurred in July and August. The forecast temperatures profiles were very similar to the observed patterns in the region of the ozone hole between 150–20 hPa, but they were about 0.5–1 K cooler than the observed values below 20 hPa and about 2–3 K cooler above 20 hPa, in particular in July.

Humidity observations from sondes are less reliable in the stratosphere and are therefore not suited for a test of the forecast humidity. Sporadic comparison with MLS humidity retrievals (http://mls.jpl.nasa.gov/plots/mls/mls_plot_locator.php) at about 56 and 10 hPa suggest correct patterns and an underestimation of stratospheric humidity by about 1 ppbv, corresponding to about 20%.

3.2 Comparison with ozone columns from sondes

The forecasts have been evaluated against ozone soundings from seven locations (see Table 3) in the Antarctic region for the period August to December 2008. Typical uncertainties for these ozone observations in the lower stratosphere are less than 5% for both the random and the systematic error (e.g. Beekmann et al., 1994). The number of soundings varied for the different stations and most observations were available in September and October when the ozone hole was fully developed. Neumayer and South Pole station had the most complete records.

The observed ozone profiles were converted to ozone columns. The soundings stopped mostly at heights between 10 to 5 hPa and the recorded top height was also applied in the calculation of the forecast ozone columns. The total column values, i.e. up to the model top of 0.1 hPa, were about 5% larger than these vertically limited values. Figure 5 shows scatter plots of observed and forecast ozone columns by the three model systems with and without assimilation. Although there was a good correlation, the FC runs by IFS-MOZART and IFS-TM5 mostly over-predicted whereas IFS under-

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5 predicted the observations. In particular the lower values, i.e. at the time of the ozone hole, were overestimated by IFS-MOZART. When initialized with analyses, forecasts (ANA) improved to a large extent (see Fig. 5). The mean biases of the IFS-MOZART, IFS-TM5 and IFS FC run were changed from 59.9, 36.1 and -8.2 DU to 7.3, 9.9 and 9.7 DU respectively by the initialization with the analyses. Table 4 lists the biases and the observed averages for the individual stations. The ANA runs had the lowest biases at South Pole station; the highest biases occurred at Marambio (64.24° S, 56.63° E) which is located closer to the edge of the ozone hole. The initialization with analyses improved not only the biases but also reduced the standard deviation of the errors from 32.8, 32.0 and 26.1 DU to 13.3, 13.9 and 13.5 DU for IFS-MOZART, IFS-TM5 and IFS respectively. The variation among the three different chemistry schemes was greatly reduced, which means that the analyses were mainly determined by the continuous as-
10 simulation of satellite observations rather than the underlying model approach. Figure 6 shows time series of the observations, the FC and ANA runs by all three model set-ups for South Pole station. The ANA runs followed the observations closely, whereas the biases of the FC runs varied in time.

3.3 Forecast of the ozone hole size with different forecast lead times

The size of the ozone hole forecast by the ANA, FC and FC15 runs of each chemistry scheme is shown as a function of time in Fig. 7. The ozone hole was expressed as the area fraction below 220 DU between 62° S and 90° S. Figure 8 presents maps of the ozone total column fields by the different forecasts for the 30 September 2008.
20

The FC runs by IFS-MOZART predicted the right duration of the ozone hole but strongly under-predicted its size (Fig. 7, top). IFS-MOZART's under-prediction might be caused to some extent by the large over-estimation of the ozone concentrations in and around the polar vortex at the end of the austral winter (Fig. 8, top), which could be caused by the overestimation of the ozone transport from the equatorial region by an enhanced Brewer-Dobson circulation in the month before Austral spring. The use of analysed and therefore un-balanced wind fields, which then introduce artificial
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mixing, has been identified by some authors (e.g. van Noje et al., 2004) as a reason for the simulation of too little ozone depletion by CTMs. However, stand-alone runs with MOZART-3 using wind fields produced with the analysis set-up used in this study simulated a correct age of air (O. Stein, personal communication, 2009). Also Monge-Sanz et al. (2007) show that artificial mixing is much reduced in a version of the IFS, which is similar to the one used in this study.

The linearized scheme (IFS) forecast (FC) the ozone hole development three weeks too early but the slow closure from October onwards followed the analyses very well. The exaggerated ozone loss by the linear scheme could be caused by the small negative temperature bias of the analysis as shown in Fig. 4. Further, the temperature-triggered catalytic ozone loss term (see Sect. 2.1) does not take into account the delay between the conditioning of the PSCs and the activation of the chlorine. A cold tracer (Cariolle and Teyssedre, 2007; Eskes et al., 2003) may help to improve the simulation of the ozone hole start by the linear scheme.

The climatological approach (IFS-TM5, FC) correctly predicted the quick development of the ozone hole but its full extent in September was slightly under-predicted. The forecast ozone hole had already disappeared by the start of November, when still half of the area south of 62° had total ozone columns below 220 DU in the analyses.

Despite the sometimes large biases of the FC runs, the spatial patterns of the ozone total column fields were in all cases similar to the ANA and FC15 runs (e.g. see Fig. 8). Since the ANA run showed a very good agreement with observations, it can be concluded that the more dynamically driven exchange between the vortex and its surroundings was well represented by the transport scheme in the IFS.

It is apparent from Fig. 7 (middle) that the analysed ozone total columns field produced by the three different modelling approaches were very similar because the respective ANA runs did not differ to a large extent for day one. The largest differences, although only around 20 DU appeared in the vortex centre in August and September (Fig. 6), which is less observable at this time by the UV-based sensors OMI, SCIAMACHY and SBUV. These differences can be attributed to the differences in the chem-

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istry schemes. When no MLS retrievals were available, as in the IFS-MOZART-ANA* run, the observed information content was further reduced (see Fig. 1). Therefore, the forecast IFS-MOZ-ANA* in August and September was more influenced by the MOZART-3 chemical scheme, which under-predicted the ozone hole (Fig. 7, top).

5 The ANA forecast for day three followed to a large extent the ANA forecast for day one both in terms of the ozone hole size and the spatial distribution (not shown). IFS-MOZ-ANA showed the smallest differences from the analysed values over the whole period. The linear scheme (IFS) tended, as in the case of the FC run, to overestimate the ozone hole development in August. The IFS-TM5-ANA run showed a stronger drift
10 towards the FC runs. However, the positive impact of the chemical initialization could be maintained by all chemical schemes until day three and longer.

More insight into the impact of the chemical schemes on the predictability of the ozone can be obtained from the evaluation of the FC15 runs (Fig. 7, bottom). These runs are equivalent to continuous 15-day model runs, which are started from an ozone
15 analysis. The time of the biggest variability among the chemical schemes was again from mid-August to mid-September. The initialization with analyses on the 16 August delayed the simulation of the ozone hole with IFS to some extent but it did not greatly improve the results from IFS-MOZART in terms of the 220 DU threshold. However, the large bias reduction by the initialization with analyses of IFS-MOZART could mostly be
20 maintained over the 15 days. The initialization of IFS-MOZART on the 1 September led to a more realistic, but still underestimated weak, increase of the ozone-hole size over the following 15 days. Around the 15 September the ozone hole had reached its maximum size. The initialization on this day was the last one which was needed to correct the IFS-MOZART fields because from this point on the slow, more dynamically driven
25 closure of the ozone hole was forecast in very good agreement with the analyses. The same is true for the linearized scheme of the IFS; only in November and December the impact of the initializations tended to fade more quickly. The correct forecast of the dynamically driven closure of the ozone hole (see below) suggests that the transport of ozone in to the vortex was correctly simulated.

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The IFS-TM5 ozone fields are strongly constrained by the underlying climatology. Therefore the initialization with analyses had the smallest impact among the three chemical schemes. The well predicted ozone-hole increase was not changed by the initialization. The closure of the ozone hole was, as in the FC run, enforced much too early, in particular in November and December. A test with no relaxation below 5 hPa improved the simulation results of IFS-TM5-FC15 during the time of the ozone hole closure at the expense of poorer results during the rapid development of the ozone hole, which could not be reproduced anymore.

A sequence of initialized forecasts over 15 days, i.e. without the daily update of the meteorological initial conditions as in the FC15 runs, with IFS did not significantly differ from the IFS-FC15 run. This means that the large scale development of the polar vortex was correctly predicted by the IFS over this time scale. However, more dynamically complex events such as the vortex split in 2002 have a shorter meteorological predictability of about one week (Simmons et al., 2005).

3.4 Evaluation of vertical profiles

The assimilated satellite ozone retrievals (see Table 1) contained either no (OMI, SCIAMACHY and SBUV-2) or only limited information (MLS) about the shape of the ozone profile, in particular in the troposphere. Hence, ozone analyses with realistic total column values may not necessarily be in good agreement with the observed ozone profiles. In particular tropospheric concentrations, which contribute little to the total columns, may become distorted during the assimilation procedure.

For the sake of brevity, only the match with observations from the soundings at Neumayer Station (see Table 3) is discussed in this section but the findings were also true for the other locations. Figure 9 shows monthly averages of observed and forecast (day one of FC and ANA) profiles from the three model configurations. The ozone hole became visible in the observed September average in the altitude range between 150 to 20 hPa; and it was largest in October.

The FC runs by the climatological scheme of IFS-TM5 correctly reproduced the

height of the maxima and minima but underestimated the ozone loss from September onwards in the region above 200 hPa. The linear scheme of IFS produced the most realistic stratospheric monthly-averaged profiles after September but it overestimated the tropospheric values below 500 hPa and under-estimated the stratospheric concentrations in August and September. Without assimilation, the profiles of IFS-MOZ-FC did not show the indented shape typical for the ozone hole and generally underestimated the tropospheric ozone values.

The ANA runs with all available satellite data (see Table 1) by all three model configurations matched the observed profile shape in the stratosphere. The maximum error of the monthly mean at any height was about 1 mPa, and the maximum deviation of individual profiles was below 2.5 mPa. No systematic bias could be found at any level in the stratosphere. The ANA runs were in better agreement with observations than the respective FC runs in all cases. The good agreement could be attributed to the assimilation of MLS data because the NRT forecast with IFS-MOZART (IFS-MOZ-ANA*), for which MLS data were not available, produced worse results in particular in September and October. In this period an over-prediction of stratospheric ozone occurred at about 100 hPa which was accompanied by a large under-prediction of tropospheric ozone.

To get a better picture of the tropospheric performance, Fig. 10 shows the ozone profiles below 500 hPa by IFS-MOZART and IFS-TM5 for each month from August to December. The values of the linearized scheme were not considered because it does not represent tropospheric chemistry. The observed partial column up to 500 hPa was about 4% (6 DU) of the total column, which is lower than the size of the measurement error of the sondes. Typical observed values were about 30 ppb in August and decreased in the lower part of the troposphere to about 15 ppb at the end of the year.

The IFS-MOZ-FC run underestimated the observation on average by 10 to 15 ppb whereas the IFS-TM5-FC run had typical errors of 3 ppb and lower. The assimilation improved the bias of IFS-MOZ, in particular from October to December, when OMI and SCIAMACHY observations were available over the whole Antarctic region. This tropospheric improvement could be maintained in the forecasts (IFS-MOZ-FC15) over

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5 a period of about a week. In the case of IFS-TM5, the assimilation did not keep the rather correct values in August and September as the error increased up to 10 ppb in this period. The errors from October onwards were about as small as the already low errors from the corresponding FC run. The largest tropospheric bias for both IFS-MOZ-ANA and IFS-TM5-ANA occurred in September. This positive analysis bias seemed to be caused by possibly overestimated total column retrievals in this period of low solar elevation (see Fig. 3, top). The bias of the total column observations was then partly accommodated in the analysis by an increase in the tropospheric values, which were not constrained by the MLS data.

10 The largest errors occurred at the model surface where both the profiles of the FC and ANA runs showed a pronounced decrease of about 5 ppb, which could not be found in the sonde observations. The assimilation improved the forecast values for both IFS-TM5 and IFS-MOZART against one-hourly surface observations at Neumayer Station (not shown). The bias of the FC runs was reduced from minus 15 and minus 6 ppb to minus 7 and minus 4 ppb for the runs IFS-MOZ-ANA and IFS-TM5-ANA respectively.

15 The mostly positive impact on the analysis in the troposphere occurred despite the fact that the lowest MLS partial columns covered only the range from 68 to 214 hPa and that errors larger than 20% are assigned to this level. It seems that the synergy of column information from SCIAMACHY and OMI being consistent, i.e. bias free, with
20 MLS's stratospheric profile led to improved tropospheric profiles. However, the problem remains, that tropospheric values were not well constrained by the used satellite observations. The model background error specification is likely to strongly influence the tropospheric values of the analyses.

25 The generally good agreement of the ANA runs with profile observations in the stratosphere and the troposphere emphasizes the importance of the assimilation of ozone profile data, such as MLS, the Michelson Interferometer for Passive Atmospheric Sounding (MIPAS) or GOME-2. Without assimilation of height-resolved data, the vertical structure of the analysis depends entirely on the background error statistics. If the assimilating model does not simulate a realistic profile shape, as in the case of

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IFS-MOZ-ANA*, the analysed profiles can deteriorate.

4 Summary and conclusion

The paper presents forecast and assimilation experiments of the Antarctic ozone hole from August to December 2008. The focus was put on the impact of three different ways to describe stratospheric ozone chemistry and on the impact of the initialization with ozone analyses produced by assimilation of satellite data. A linearized scheme (IFS), a relaxation to a climatology (IFS-TM5) and a chemistry mechanism including heterogeneous processes on PSCs (IFS-MOZART) were used by the IFS within an otherwise identical model and 4D-VAR data assimilation configuration. Ozone sondes within the Antarctic region were used for the evaluation of the forecasts.

Without correction by ozone analyses, the forecasts differed to a large extent. The linear scheme predicted the chemically instigated ozone depletion too early but produced good results during the more dynamically driven closure of the ozone hole after its peak in September. The climatological scheme captured the development of the ozone hole correctly but did not reproduce the exceptionally long duration of the 2008 event due to the strong forcing towards climatology. The chemical scheme of MOZART-3 forecast the length of the reduced ozone levels correctly but did not simulate the sudden and strong ozone depletion between 200 and 20 hPa. The problems that CTMs (e.g. Krämer et al., 2003) and chemistry-climate models (Struthers et al., 2009) show in simulating the amount of ozone depletion correctly have been reported by some authors although more recent studies show improvements in the modelling of surface reactions on PSC which trigger the ozone depletion (e.g. Daerden et al., 2007).

The characteristics of the three different chemistry schemes were overcome by the combined assimilation of MLS, SBUV-2, OMI and SCIAMACHY ozone retrievals. One-day forecasts started from the analyses showed a considerable improvement of bias and error standard deviation when compared with columns and profiles of ozone soundings from seven locations. The analyses made with the different chemistry

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schemes hardly differed, which proved the dominating influence of the assimilated observations. Only at the time of the development of the ozone hole, defined as the area with ozone below 220 DU, did small differences appear among the schemes. At this time of the year the UV-VIS based instruments did not observe a large area south of 60° S and MLS was the main information source. The overall good agreement of the analyses is different from the finding of an inter-comparison study of linear stratospheric ozone schemes by Geer et al. (2006). They found a more important dependence of the underlying modelling approach on the global scale. However, the assimilation of four independent satellite sensors better constrained the ozone fields than the MIPAS observations used in that paper.

The assimilation of MLS partial column profiles in combination with total column retrievals from OMI and SCIAMACHY proved to be essential for the correctness of the ozone profiles in the stratosphere, troposphere and at the surface during the whole period. In the stratosphere, the maximum error of the monthly mean ozone partial pressure at any height was below 1 mPa, and the maximum deviation of individual profiles was below 2.5 mPa. The importance of the MLS retrievals was found by comparison with an assimilation run from IFS-MOZART, for which MLS data was not assimilated. This NRT run showed less realistic stratospheric profiles and deteriorated tropospheric values, despite good agreement in terms of total columns. Problems in the analysed profiles from the ERA40 re-analysis are also reported in Dethof and Hólm (2004) and attributed to missing vertical information and biases. By assimilating multiple observations the often complementary information of the sensors, such as high resolution total column observations and vertical profiles, can be gainfully combined. This has been shown in this study and by other authors (Jackson, 2007; Stajner et al., 2008; Massart et al., 2009).

The GEMS project and its successor MACC (Monitoring of Atmospheric Chemistry and Climate) aim at providing operational forecasts and analyses of atmospheric composition for the troposphere and stratosphere with one modelling system. Thus, the tropospheric performance was also studied. The tropospheric bias of IFS-MOZART was

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largely corrected by the initialization with analyses. The very small tropospheric error of IFS-TM5 without assimilation was moderately worsened by the initialization only in August and September. In the rest of the period the error with and without assimilation was of the same size. The robust performance of the assimilation with the coupled IFS-CTM system in the troposphere was a promising achievement because the ozone satellite observations are strongly dominated by the stratospheric signal. However, the analysis of tropospheric ozone remains a challenge (de Laat et al., 2009).

The multi-sensor assimilation raises the question of biases among the different sensors. These biases are more difficult to quantify in areas of large gradients because each instrument measures either partial or total columns at different places and times. The retrievals of the UV instruments are less accurate at low solar elevation. A comparison of the total column values averaged over Antarctica from OMI, SCIAMACHY and partial columns from MLS and SBUV-2 (Fig. 2) showed larger inter-instrument differences only until mid October, i.e. during the time when the UV-VIS based sensors can only observe ozone at the edges of the Antarctic area. The derived differences among the instruments were therefore mostly related to the sampling and did not constitute biases harmful to the assimilation process. A more detailed study of the biases at the end of August and the end of November confirmed that inter-instrument bias were below 3%. Both MLS and OMI were nearly bias-free against the analysis while SCIAMACHY showed a higher positive bias at low solar elevation at the end of November. Inter-sensor bias and its quantification is a common problem for data assimilation. Therefore a variational bias correction scheme (Dee, 2005) will be extended to ozone retrievals assimilated by the IFS.

This paper investigated the impact of the ozone initialization with increasing forecasts lead time. The question is how quickly the model draws from the initializations to the model-specific reference state. Providing correct initial conditions based on observations may help to better identify the cause of the under-prediction by IFS-MOZART because the slow accumulation of errors can be distinguished from the stronger, more short-term model deficiencies. The finding of this paper and others (e.g. Sekiyama and

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Shibata, 2005) indicate a long-lasting influence of stratospheric ozone initial conditions in general. Therefore it might become good practise for CTM runs over several months to be initialized with analyses.

With the correct initialization, the closure of the ozone hole from October to December was very well-predicted at least up to 15 days by the full scheme of MOZART-3 and also by the linear scheme, which only slowly drifted back to its reference state. The long-lasting impact is proof of the good simulation of the transport processes because the closure of the ozone hole is a dynamically driven process. As expected, the three-day relaxation to the climatology in TM5 wiped out the impact of the initialization very quickly, in particular in November and December. When this relaxation was switched off a much better match to the analysis was obtained during the ozone recovery phase.

The impact of the initialization with analyses lasted a much shorter time during the development of the ozone hole until mid-September. The too early forecast of the linear scheme and the underestimation of the ozone hole by the full scheme could be improved but the characteristics of the schemes became apparent after 2 days.

Acknowledgements. We thank Rossana Dragani and Martin Suttie for helpful discussions on satellite observation errors. The ozone sonde data used in this publication were obtained as part of the Network for the Detection of Atmospheric Composition Change (NDACC) and the World Ozone and Ultraviolet Radiation Data Centre (WOUDC) and are publicly available from <http://www.ndacc.org> and <http://www.woudc.org>. The ozone soundings were carried out by Alfred Wegener Institute – Neumayer (AWI-NM), University of Wyoming, Finnish Meteorological Institute (FMI), National Meteorological Service of Argentina (SNMA), Japan Meteorological Agency (JMA), Australian Bureau of Meteorology (ABM) and National Oceanic and Atmospheric Administration (NOAA). The ground based data were obtained from the Global Atmospheric Watch (GAW) network. The satellite data were downloaded from NASA, NOAA and KNMI. The work has been carried out in the GEMS project, which is funded by the European Commission under the EU Sixth Research Framework Programme, contract number SIP4-CT-2004-516099.

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Table 1. Assimilated satellite observation, specifications and blacklist criteria with respect to solar elevation (SOE) and latitude (LAT).

Sensor	Platform	Assimilated columns	Blacklist criteria	Provider	Product
SBUV-2	NOAA 16/17/18	6 (0.1–1013 hPa)	SOE<6° LAT<-80°	NOAA	V8
OMI	AURA	Total	SOE<10°	NASA	OMDOAO3 v883
SCIAMACHY	Envisat	Total	SOE<6°	KNMI	O3doas
MLS	AURA	16 (0.02–215 hPa)		NASA	MLS-Aura.L2GP-O 3 v02-23-NRT-03

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Table 2. List of experiments.

Experiment Label	Ozone initial conditions	Assimilated Sensors	scheme	Chemistry Length	FC NRT
IFS-MOZ-FC	Forecast	–	Full	96	Yes
IFS-MOZ-ANA*	Analysis	SCIA, SBUV-2, OMI	Full	72	Yes
IFS-MOZ-ANA	Analysis	SCIA, SBUV-2, OMI, MLS	Full	72	–
IFS-MOZ-FC15	Forecast, 1st and 15th day each month analysis	SCIA, SBUV-2, OMI, MLS	Full	24	–
IFS-TM5-FC	Forecast	–	Climatology	24	–
IFS-TM5-ANA	Analysis	SCIA, SBUV-2, OMI, MLS	Climatology	72	–
IFS-TM5-FC15	Forecast, 1st and 15th day each month analysis	SCIA, SBUV-2, OMI, MLS	Climatology	24	–
IFS-FC	Forecast	–	Linear	24	–
IFS-ANA	Analysis	SCIA, SBUV-2, OMI, MLS	Linear	72	–
IFS-FC15	Forecast, 1st and 15th day each month analysis	–	Linear	24	–

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Table 3. Ozone sondes used for the evaluation.

Name	Lat	Lon	Observations per month in 2008					Network/Institute
			Aug	Sep	Oct	Nov	Dec	
Neumayer	−70.65°	−8.26°	7	13	10	7	4	WOUDC & NDACC/AWI-NM
McMurdo	−77.85°	166.67°	0	8	10	0	0	NDACC/University of Wyoming
Marambio	−64.24°	56.63°	9	8	9	9	9	WOUDC/FMI-SMNA
Syowa	−69.0°	39.6°	6	5	4	5	5	WOUDC/JMA
Ushuaia	−54.85°	68.31°	2	4	6	5	1	WOUDC/SMNA
Maquari Island	−54.5°	158.94°	4	4	2	0	0	WOUDC/ABM
South Pole	−90.0°		6	11	9	6	5	NDACC/NOAA

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Table 4. Average of ozone columns in DU derived from sondes (OBS) and biases (model minus observation) for all model configurations and stations (see Table 3).

Station	OBS	IFS-MOZ-FC	IFS-TM5-FC	IFS-FC	IFS-MOZ-ANA	IFS-TM5-ANA	IFS-ANA
Neumayer	159.8	56.5	40.1	-3.1	5.0	7.7	8.4
Syowa	185.3	43.8	35.5	-4.7	3.6	8.1	3.7
McMurdo	155.3	72.3	42.2	5.5	10.7	11.5	11.0
Marambio	206.6	52.2	40.4	-15.0	13.0	12.3	12.1
Ushuaia	217.4	36.6	25.4	-22.3	9.0	10.5	12.1
Macquarie Island	304.8	40.0	33.3	-19.3	4.7	9.2	8.4
South Pole	154.3	42.7	30.2	-3.7	3.2	3.7	6.5

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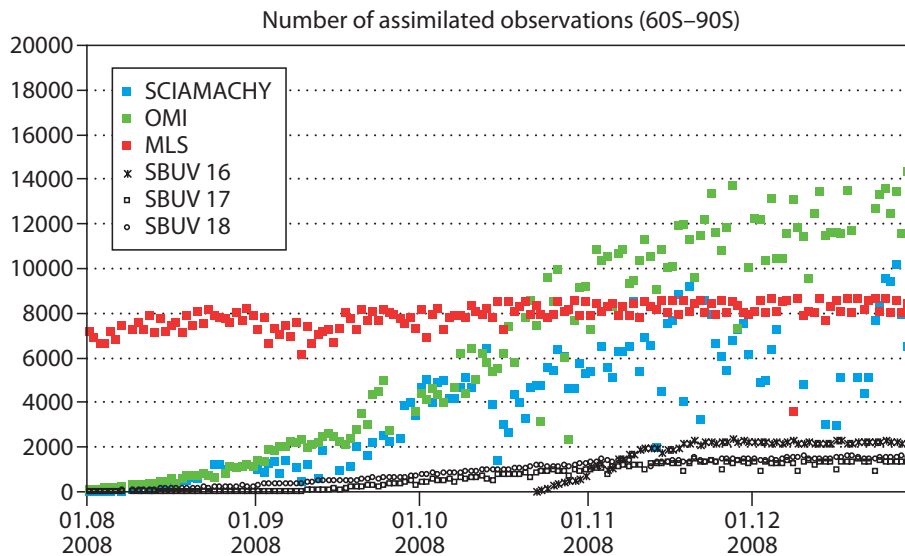


Fig. 1. Number of assimilated satellite observations per day over Antarctica (60°S – 90°S) in the period from 1 August to 31 December 2008.

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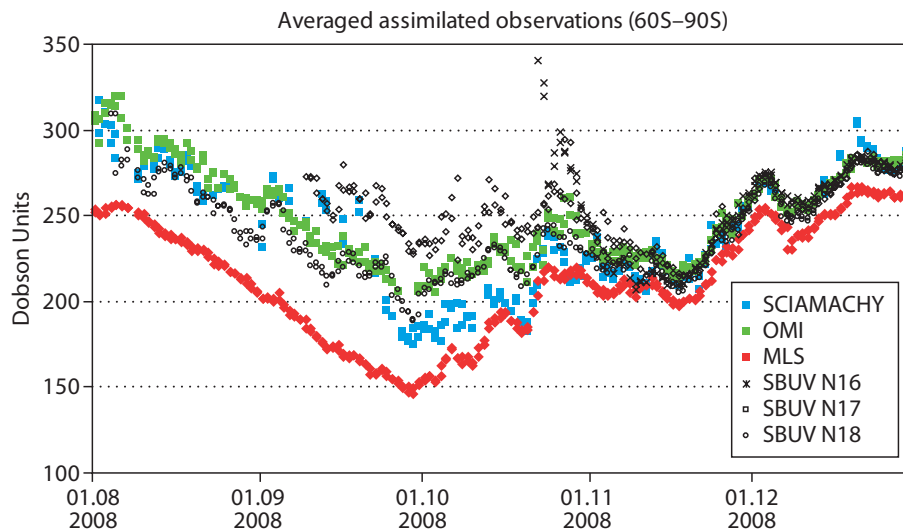


Fig. 2. Daily mean of assimilated ozone total columns from SCIAMACHY and OMI and sum of partial columns from MLS and SBUV-2 aboard NOAA 16, 17 and 18 in DU averaged over Antarctica (60° S–90° S) in the period from 1 August to 31 December 2008.

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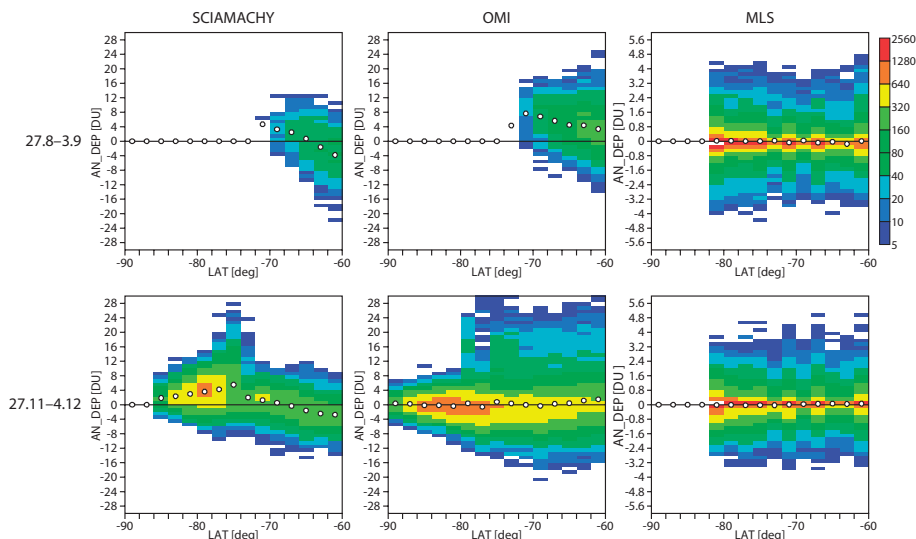


Fig. 3. Frequency distribution of analysis departures (observation minus analysis) from IFS-MOZ-ANA in DU (vertical axis) against latitude (horizontal axis) for total column observations from SCIAMACHY (left) and OMI (middle) and partial columns observations from MLS (right, different y-axis range) over Antarctica (60°S – 90°S) in the period from 27 August–3 September 2008 (top) and from 27 November to 4 December 2008 (bottom). The circles show the average for each latitude interval.

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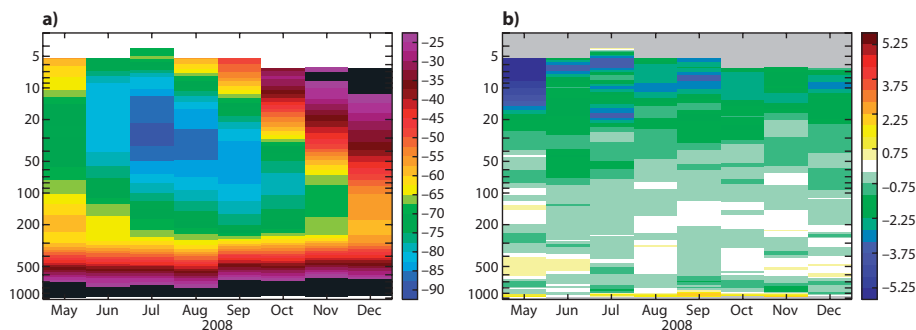


Fig. 4. Monthly mean of observed temperature profiles (left, in $^{\circ}\text{C}$) and respective forecast errors (right, in K) over Neumayer station for the period from May to December 2008.

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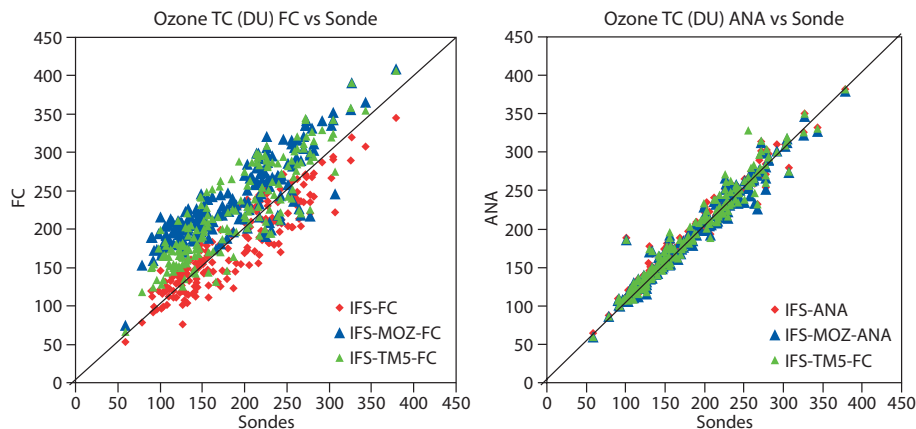


Fig. 5. Columns of ozone soundings at different locations (OBS, see Table 3) in the period from 1 August 2008 to 31 December 2008 and the corresponding predictions for the first forecast day by IFS-MOZART, IFS-TM5 and the IFS without assimilation (FC, left) and with assimilation (ANA, right).

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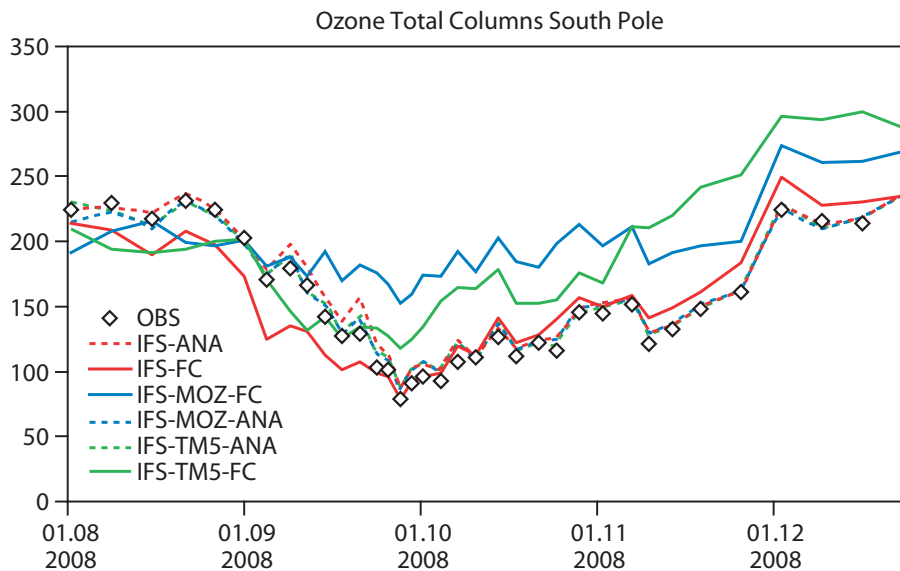


Fig. 6. Time series of observed total columns (OBS) in DU from ozone sondes at South Pole Station and from forecasts without (FC) and with assimilation (ANA) by IFS-MOZRT, IFS-TM5 and IFS. Only soundings whose top pressure was between 5 and 8.5 hPa.

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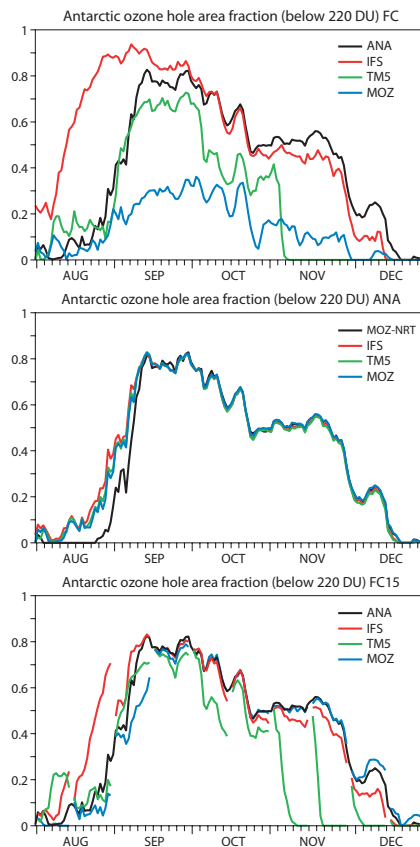


Fig. 7. Area fraction below 220 DU in the area 62°S – 90°S from the runs without assimilation (FC, top), runs with assimilation (ANA, middle) and the runs initialized only on the 1st and 15th day of each month (FC15, bottom) by IFS-MOZART (MOZ), IFS and IFS-TM5. The assimilated satellite observations were OMI, MLS, SCIAMACHY and SBUV-2 (see Table 1). MLS was not assimilated in the NRT ANA* run with IFS-MOZART (MOZ-NRT, middle panel only).

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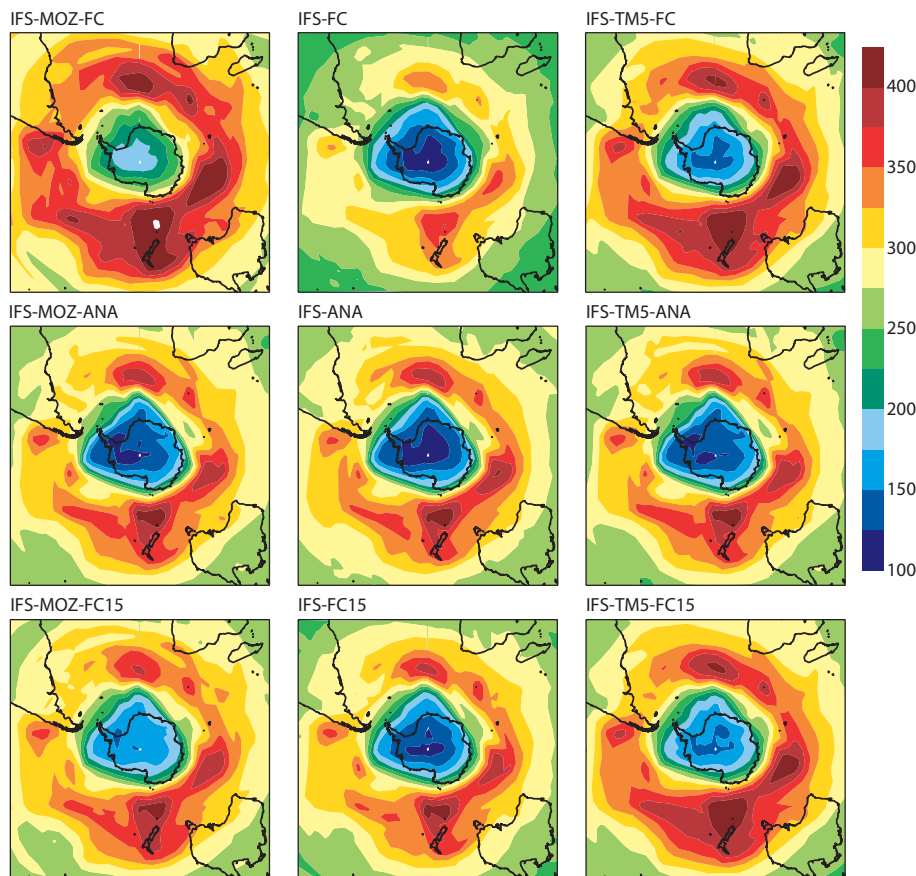


Fig. 8. Ozone total columns (DU) over Antarctica on 30 September 2008, 12:00 UTC, from the runs without assimilation (FC, top), with assimilation (ANA, middle) and from runs initialized 14 days previously (FC15, bottom) by IFS-MOZART, by IFS and by IFS-TM5.

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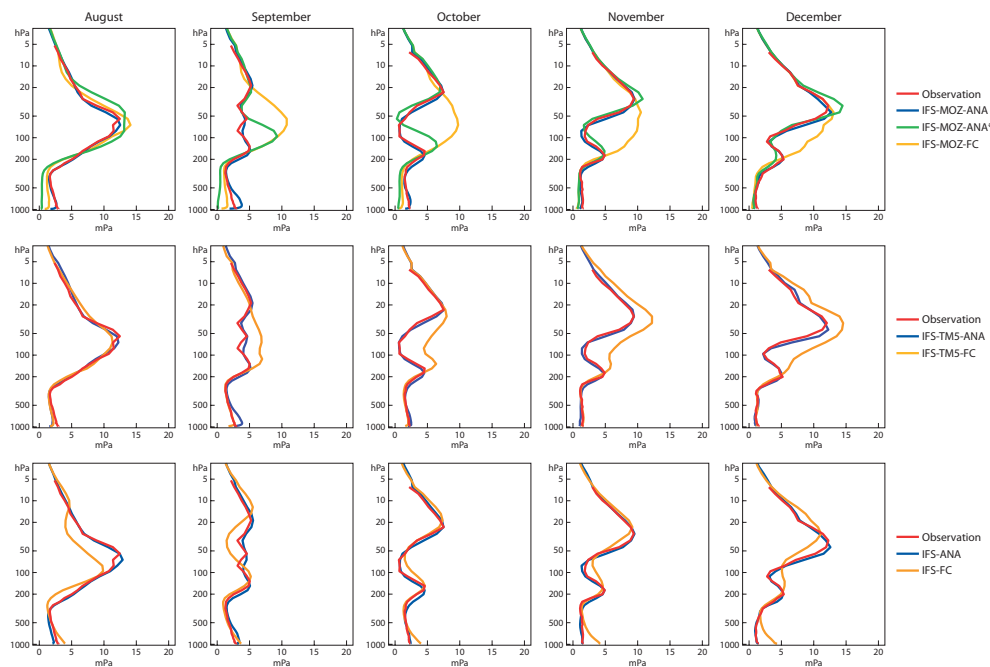


Fig. 9. Monthly averaged ozone profiles (partial pressure in mPa) forecast by IFS-MOZART (top), by IFS-TM5 (middle) and by IFS (bottom) without assimilation (FC, orange), with assimilation (ANA, blue), with assimilation but excluding MLS data (ANA*, green, IFS-MOZART only), and observations (red) at Neumayer Station from August to December 2008.

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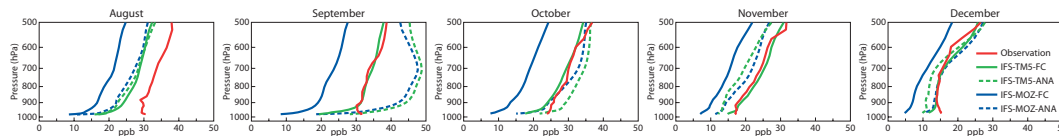


Fig. 10. Monthly averages of observed (red, solid) and forecast ozone profiles in the troposphere from IFS-MOZ (blue) and IFS-TM5 (green) without assimilation (FC, solid) and with assimilation (ANA, dashed) at Neumayer station in the period August to December 2008.

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