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Copernicus atmospheric service for stratospheric ozone: validation and intercomparison of four near real-time analyses, 2009–2012

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correct for the deficiency of the underlying MOZART model, which may be related to the applied meteorological fields.

Biases of BASCOE compared to ozonesonde or ACE-FTS ozone profiles do not exceed 10 % over the entire vertical stratospheric range, thanks to the good performance of the model in ozone hole conditions and the assimilation of offline MLS profiles going down to 215 hPa.

TM3DAM provides very realistic total ozone columns, but is not designed to provide information on the vertical distribution of ozone.

Compared to ozonesondes and ACE-FTS satellite data, SACADA performs best in the Arctic, but shows large biases (>50 %) for ozone in the lower stratosphere in the Tropics and in the Antarctic, especially during ozone hole conditions.

This study shows that ozone analyses with realistic total ozone column densities do not necessarily yield good agreement with the observed ozone profiles. It also shows the large benefit obtained from the assimilation of a single limb-scanning instrument (Aura MLS) with a high density of observations. Hence even state-of-the-art models of stratospheric chemistry still require the assimilation of limb observations for a correct representation of the vertical distribution of ozone in the stratosphere.

1 Introduction

The presence of a high-altitude ozone layer in the atmosphere, which protects Earth against the harmful ultraviolet (UV) light from the Sun, was first determined in the 1920s from observations of the solar UV spectrum. Systematic measurements of stratospheric ozone using ozonesondes started in the late 1950s (Solomon et al., 2005). At that time, the development of satellites just started, the first one (Sputnik) being launched in 1957. Systematic satellite measurements of ozone started in the late 1970s with the series of Total Ozone Mapping Spectrometer (TOMS) and Solar Backscatter Ultraviolet Instrument (SBUV) instruments. While the interest in stratospheric ozone started off as purely scientific curiosity (Cornu, 1879; Hartley, 1881), the

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attitude changed after the discovery of the ozone hole in 1985 (Farman et al., 1985), which led to the build-up of a comprehensive monitoring program consisting of balloon-borne ozonesondes, Brewer–Dobson spectrometers, and various satellite instruments.

The emergence and increasing capabilities of computer facilities opened at the same time the path to numeric modeling, which has permitted simulating the behavior of the atmosphere and predicting its evolution under specific conditions. The chemical processes involving ozone depleting substances, in particular the heterogeneous reactions in the polar regions, which lead to the ozone hole, are by now well known and documented in previous studies (e.g. Solomon, 1999) and ozone assessments (e.g. WMO, 2011a).

Through data assimilation (Sandu and Chai, 2011), chemistry transport models (CTM) can process information of satellite observations and use them to update the modeled 3-D chemistry field, in order to match real-world observations as close as possible and deliver more realistic datasets than pure model forecasts. The resulting analyses of atmospheric composition enable the monitoring and forecasting of the distribution of ozone and other key constituents in the earth atmosphere (Lahoz and Errera, 2010).

Succeeding to GEMS (Hollingsworth et al., 2008, Global and regional Earth-system (Atmosphere) Monitoring using Satellite and in-situ data), followed by PROMOTE (PROtocol MOniToring for the GMES Service Element: Atmosphere – <http://www.gse-promote.org>) and MACC, the EU (European Union) FP7 (Framework Project) project MACC-II (Monitoring Atmospheric Composition and Climate – Interim Implementation) is the third in a series of projects funded since 2005 to build up the atmospheric service component of the Global Monitoring for Environment and Security (GMES)/Copernicus European programme (Peuch et al., 2013). In this paper, the terminology “MACC” refers to both the MACC and MACC-II projects. MACC provides NRT tropospheric and stratospheric data for monitoring present and near-future atmospheric conditions up to 5 days ahead (Stein et al., 2012), as well as a reanalysis for the years 2003–2012 (Inness et al., 2013). The MACC stratospheric ozone ser-

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vice (<http://www.copernicus-stratosphere.eu>) deals with the evolution of stratospheric ozone and related species, and provides regular stratospheric ozone analyses and forecasts. The service currently uses a set of four independent chemical assimilation systems: IFS-MOZART (Flemming et al., 2009; Stein et al., 2012), an operational implementation of BASCOE (Errera et al., 2008; Viscardy et al., 2010), SACADA (Elbern et al., 2010), and TM3DAM (Eskes et al., 2003; van der A et al., 2010).

To ensure the reliability and suitability of the data for decision-makers and other users, assessing and documenting the quality of the delivered data products is of key importance. In this paper, we assess the ability of the MACC stratospheric ozone service to simulate stratospheric ozone for the 3 year-period September 2009–September 2012. This intercomparison paper does not aim directly at model issues, but rather documents the overall quality of the MACC stratospheric service, i.e. a full-blown intercomparison of the models is out of the scope of this paper. Intercomparison of ozone analyses has been done before by Geer et al. (2006) who evaluated the performance of different ozone data assimilation systems based on observations from Envisat (Environmental Satellite) only. Here we intercompare for the first time analyses based on data from different satellites, either total ozone column or profile observations or a combination of both.

The next section describes the different analyses in the MACC stratospheric ozone service and the reference observations used for their validation. Section 3 contains the evaluation of the total ozone columns based on Brewer/Dobson observations, while the vertical distribution of ozone is assessed in Sects. 4 and 5, through comparison with ozonesondes and ACE-FTS satellite data, respectively. In Sect. 6, we assess the performance of the MACC analyses during an event of exceptional nature: the Arctic ozone hole 2011. We additionally investigate the influence of the assimilated dataset on the performance of the analyses for one month covered by this event: March of 2011. The final section provides a summary and conclusions.

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MLS is a limb-viewing microwave radiometer, providing some 3500 daily vertical profile measurements of several atmospheric parameters, such as ozone (O₃), nitric acid (HNO₃), water vapour (H₂O), hydrochloric acid (HCl), hypochlorous acid (HOCl), and nitrous oxide (N₂O) from about 8 to 80 km (0.02 hPa to 215 hPa) with a vertical resolution of about 3 km and a horizontal resolution of 200–300 km (Waters et al., 2006). As a microwave remote sensing sounder, MLS also provides observations during the polar night, which has a positive impact on ozone analyses during the onset of the ozone hole.

MLS ozone data are delivered in near real-time by NASA/JPL (Jet Propulsion Laboratory), with a latency of only 2 to 4 h, whereas a scientific dataset, containing additionally non-ozone species, is delivered with a delay of 4 days. The former dataset is used for the assimilation of ozone by IFS-MOZART (see Sect. 2.2.1), whereas the latter is used by BASCOE (see Sect. 2.2.2) for the assimilation of O₃, HNO₃, H₂O, HCl, and N₂O (v2.2). The useful range of both datasets differs: NRT ozone profiles are only recommended for scientific use at pressure levels 68–0.2 hPa (except for the calculation of ozone columns where the 100 hPa pressure level may be used as a lower limit), while the offline MLS dataset can be used for the entire pressure range 215–0.02 hPa.

2.1.2 Envisat satellite: SCIAMACHY total columns

SCIAMACHY (SCanning Imaging Absorption spectroMeter for Atmospheric CHartography) is a UV-VIS-NIR (Near InfraRed) imaging spectrometer onboard ESA's Environmental Satellite (Envisat) launched on 1 March 2002. SCIAMACHY observed earthshine radiance in limb and nadir viewing geometry and solar and lunar light transmitted through the atmosphere in occultation viewing geometry. While spectrometers such as MLS are able to provide ozone profiles over the poles throughout the year, UV-VIS instruments such as SCIAMACHY are limited to periods with sufficient solar radiation. On the other hand, they can attain much higher spatial resolution. SCIAMACHY data have a nadir horizontal resolution of typically 32 km × 215 km and a limb vertical resolution of 3 km. After having operated five years beyond the planned mission lifetime of five years,

all communication with the Envisat satellite was lost on 8 April 2012. IFS-MOZART assimilates SCIAMACHY total ozone columns until the last date (7 April 2012). To have a clean monthly mean, it was decided to reprocess TM3DAM for the first days of April using GOME-2 from 1 April 2012, onwards. Due to a better global coverage within one day for GOME-2 (SCIAMACHY attains global coverage in 6 days), leading to an improved performance, the official MACC NRT product for SACADA already switched from SACADA-SCIAMACHY to SACADA-GOME2 on 28 October 2011.

2.1.3 MetOp-A satellite: GOME-2 total columns

The GOME-2 (Global Ozone Monitoring Experiment-2) instrument carried onboard EUMETSAT's (European Organisation for the Exploitation of Meteorological Satellites) Meteorological Operational Satellite MetOp-A (launched in October 2006) continues the long-term monitoring of atmospheric trace gases by ESA's (European Space Agency) ERS-2 (European Remote sensing Satellite-2) GOME. It is a nadir-viewing UV-VIS scanning spectrometer, which is able to achieve global coverage within one day (Munro et al., 2006). Total columns are provided with a horizontal resolution of 80 km × 40 km, while ozone profiles are given as partial ozone columns on varying pressure levels (40 levels between surface and 0.01 hPa). GOME-2 NRT products are available about two hours after sensing.

2.1.4 NOAA satellite: SBUV-2 partial columns

The SBUV-2 radiometer onboard the NOAA (National Oceanic and Atmospheric Administration) satellites continues the long-term record started by NASA's SBUV onboard Nimbus-7. SBUV-2 is a series of seven remote sensors on NOAA weather satellites, of which four are currently still operational: NOAA-16/17/18/19. It provides partial columns mostly at stratospheric levels. Six partial columns are assimilated by IFS-MOZART, located between 0.1 and 1013 hPa.

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The above data have been extensively validated against groundbased measurements: OMI by Balis et al. (2007), MLS by Froidevaux et al. (2008), SCIAMACHY by Eskes et al. (2005), GOME-2 by Loyola et al. (2011) and SBUV-2 by Bhartia et al. (1996). Total column estimates are generally in agreement within 2% (OMI, SCIAMACHY, GOME-2, MLS), except for SBUV-2, which has a somewhat larger precision of 5–15%. The uncertainty estimates of the MLS v2.2 ozone profile in the stratosphere are often of the order of 5%, with values closer to 10% at the lowest stratospheric altitudes. Tests with the improved MLS v3.4 data (Livesey et al., 2011), which have a useful range of 261 to 0.1 hPa show that a lot of those biases in the lower stratosphere disappear.

2.2 Models description and setup of the data assimilation systems

2.2.1 IFS-MOZART

Within the GEMS project, the Integrated Forecast System (IFS), operated by ECMWF, was extended to be able to simulate and assimilate the abundance of greenhouse gases (Engelen et al., 2009), aerosols (Morcrette et al., 2009; Benedetti et al., 2009), as well as tropospheric and stratospheric reactive gases (Flemming et al., 2009; Inness et al., 2009; Stein et al., 2012) from satellite retrieval products. Satellite observations for the following reactive gases can be assimilated: O₃, nitrogen dioxide (NO₂), carbon monoxide (CO), formaldehyde (HCHO), and sulphur dioxide (SO₂), but only the former three were assimilated in the operational analysis discussed in this paper. The assimilation window of IFS-MOZART is 12 h.

The version of IFS-MOZART used here was described in detail by Stein (2013). To provide concentrations and chemical tendencies of the reactive gases, the IFS was coupled to a CTM using the coupling software OASIS4 (Ocean Atmosphere Sea Ice Soil, Valcke and Redler (2006)). The IFS computes only the transport of the forementioned reactive gases while the coupled CTM provides the chemical tendencies due to chemical conversion, deposition and emission.

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The CTM selected to deliver analyses of stratospheric ozone for the MACC global monitoring and forecast system is MOZART-3 (Kinnison et al., 2007; Stein et al., 2012) because it simulates both tropospheric and stratospheric chemistry, including the catalytic destruction of ozone in the lower polar stratosphere. Inness et al. (2009) give a detailed description of the applied procedure for the assimilation of atmospheric constituents in IFS-MOZART.

During the period studied here, the IFS was run at T159L60, where T159 denotes an expansion to wavenumber 159 in the spherical-harmonic representation used by the model (corresponding to approximately 125 km horizontal resolution at the equator), and L60 denotes a vertical grid comprising 60 hybrid-pressure levels extending from 0.1 hPa down to the surface. This run uses IFS version (“cycle”) 36R1. The CTM component, MOZART-3, used the same 60 vertical levels and a regular longitude-latitude grid with $1.875^\circ \times 1.875^\circ$ horizontal resolution. Its chemical scheme includes 115 species interacting through 325 reactions (Stein, 2013).

The following satellite O₃ data were simultaneously assimilated (see Table 1): partial columns by NOAA SBUV-2, total columns by Aura OMI and Envisat SCIAMACHY, and profiles by Aura MLS down to 68 hPa. Note that all ozone data assimilated in IFS-MOZART are NRT products. Hence the MLS dataset used here (v2.2) is the product delivered 2 to 4 h after measurement, in contrast to the data assimilated by BASCOE (see Sect. 2.2.2).

The IFS-MOZART version described here was run daily (experiment f93i) from 1 September 2009 till 30 September 2012, which determined the period considered in this paper.

2.2.2 BASCOE

BASCOE (Errera et al., 2008) is a 4D-Var system developed at the Belgian Institute for Space Aeronomy, BIRA-IASB. Based on a stratospheric CTM, BASCOE assimilates satellite retrievals of O₃, H₂O, HNO₃, HCl, HOCl, and N₂O, gathered by MLS. The assimilation window is 24 h, while BASCOE produces output every three hours. The

CTM includes 57 species that interact using 143 gas-phase reactions, 48 photolysis reactions and 9 heterogeneous reactions. The system also includes a parameterization of the effects of Polar Stratospheric Clouds (PSCs) on the gas-phase species.

In this setup, the CTM is run at a horizontal resolution of 3.75° longitude by 2.5° latitude, and applies a vertical hybrid-pressure grid with 37 levels, extending from 0.1 hPa down to the surface, most of them lying in the stratosphere. It does not include any tropospheric processes and is therefore not expected to produce a realistic chemical composition below the tropopause, resulting in larger systematic error biases for the total columns and in the lower stratosphere.

The system is driven by the ECMWF operational 6 hourly analyses (winds, temperature and surface pressure). Both BASCOE and IFS-MOZART analyses assimilate Aura MLS data, but while IFS-MOZART uses the NRT retrievals v2.2 of ozone only, BASCOE uses the standard scientific, offline retrievals (level-2) v2.2 including five other species which are available with a delay of typically four days. In contrast, IFS-MOZART assimilates other satellite instruments apart from Aura MLS, but those are measuring only ozone as species relevant for stratospheric chemistry.

2.2.3 SACADA

Within the project SACADA, a 4D-Var scheme has been developed by the Rhenish Institute for Environmental Research at the University of Cologne and partners (Elbern et al., 2010) aiming at the assimilation of atmospheric Envisat data using state-of-the-art numerical methods. This system has been implemented for operational use at the Deutsches Zentrum für Luft- und Raumfahrt, DLR, who deliver routinely daily (12 h UT) trace gas analyses based on Envisat SCIAMACHY ozone columns since March 2010. In parallel, another SACADA service assimilates MetOp-A GOME-2 total column data since January 2008. In research mode, SACADA has been successfully applied to other satellite- and groundbased observations (Elbern et al., 2010; Schwinger and Elbern, 2010; Baier et al., 2013).

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The SACADA system uses an icosahedral grid (i.e. 20 equilateral triangles) on sigma-pressure levels with an approximate resolution of 250 km. The vertical grid consists of 32 model levels extending from 7 to 66 km altitude (440 to 0.1 hPa). The tropospheric ozone column is prescribed from the TOMS V8 climatology. Like IFS-MOZART and BASCOE, SACADA applies a comprehensive stratospheric chemistry scheme (see Table 2). The NRT service additionally provides information on the following unconstrained species: HNO₃, H₂O, and HCl. Contrary to the other models applied for MACC, the SACADA CTM is driven by the meteorological forecast system GME (Majewski et al., 2001), run at DLR. GME is started from ECMWF analyses data daily at 0 h UTC and provides its own 24 h forecasts. The SACADA 4D-Var assimilation uses an assimilation window of 24 h. Note that SACADA products are delivered on a standard latitude-longitude grid with 3.75° by 2.5° resolution from 147 to 0.3 hPa altitude.

Here, we investigate two independent SACADA NRT products for two consecutive time intervals (see Table 1). NRT delivery started on 4 March 2010 with SCIAMACHY-SACADA V2.0. After 28 October 2011, SACADA was switched to the GOME-2 instrument (SACADA V2.4), which has a better data coverage daily. Total columns were only provided with the data product from 13 January 2011 on.

2.2.4 TM3DAM

The TM3DAM data assimilation system is based on the TM3/TM5 tracer transport model and is driven by operational 6 hourly meteorological fields from ECMWF. The main purpose of TM3DAM is the generation of 30–45 year reanalyses of total ozone based on all available satellite datasets (van der A et al., 2010), but in MACC it has also been operated to provide real time analyses and forecasts. TM3 contains parameterized schemes for the description of stratospheric gas-phase and heterogeneous ozone chemistry.

The assimilation scheme in TM3DAM is based on a simplified Kalman-filter assimilation approach, with a time and space dependent error covariance, but with fixed correlations (Eskes et al., 2003), which considerably reduces the computational cost.

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The TM3DAM assimilation code has been updated as described in van der A et al. (2010). The model runs at a global horizontal resolution of 3° longitude by 2° latitude. It applies a vertical hybrid-pressure grid, consisting of 44 levels extending from 0 hPa to the surface (1013 hPa). From the upper troposphere upwards, the layers coincide with the ECMWF 60-layer model version.

TM3DAM assimilates near real-time level-2 total ozone column data from Envisat/SCIAMACHY until the end of March 2012 and switched to MetOp-A/GOME-2 after all communication with the Envisat satellite was lost on 8 April 2012. NRT production of daily analyses (valid at 21 h UT) in the framework of MACC started on 16 March 2010. Only total columns are available. Besides the daily analyses, TM3DAM also generates daily forecasts for up to 9 days ahead. The Observation-minus-Forecast (OmF) statistics show that the bias of the system compared to the individual satellite measurements is typically less than 1 % for a forecast period of 1 day.

2.3 Comparison of ozone background errors

The specification of the background error covariance matrix (e.g., Kalnay, 2002) is one of the most difficult parts of an assimilation system as assimilation errors are never observed directly, they can only be estimated in a statistical sense. Each of the considered analyses has a different way of dealing with background error statistics. In IFS-MOZART, the background error covariance matrix is given in a wavelet formulation (Fisher, 2006), allowing both spatial and spectral variations of the horizontal and vertical background error covariances (Inness et al., 2013). For ozone, the background error correlations were derived from an ensemble of forecast differences, using a method proposed by Fisher and Andersson (2001). The background error standard deviation profiles and the horizontal and vertical correlations can be found in Fig. 1 of Inness et al. (2009).

BASCOE analyses use a diagonal background error correlation matrix **B** with a fixed error usually between 20 % to 50 % of the background field, 30 % in this version. The diagonal setup of **B** implies that spatial correlations are neglected. Spatial correlations

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help to spread the information from the data into the model. As mentioned by Errera et al. (2008), they can be neglected in first approximation if the spatial coverage of the assimilated observations and their vertical resolution are comparable to the model resolution. This is the case here, where a maximum of three days of MLS observations are necessary to constrain all BASCOE grid points. Note that spatial correlation on the **B**-matrix has been implemented recently in BASCOE (Errera and Ménard, 2012), following the method by Hollingsworth and Lönnberg (1986).

The SACADA 4D-Var assimilation uses a flow dependent paramaterisation of the background error covariance matrix with a diffusion approach (Weaver and Courtier, 2001). The basic idea is to formulate covariances by Gaussians and approximate these Gaussians by integration of the diffusion operator over some specified time. Horizontal and vertical background error correlation lengths are fixed to 600 km and 3 km, respectively. The background variance is set to 50 %, which is quite low and allows the observations to have a strong impact on results.

In the parameterized Kalman filter approach of TM3DAM, the forecast error covariance matrix is written as a product of a time independent (i.e. fixed) correlation matrix and a time dependent diagonal variance (Eskes et al., 2003). All aspects of the covariance matrix, including the time dependent error growth and correlation length, are carefully tuned on the basis of OmF (Obervation minus Forecast) statistics. In the total ozone product a realistic time dependent error bar is provided for each location and time.

2.4 Reference ozone data

2.4.1 Brewer/Dobson observations

To assess the condition of the ozone layer, one frequently uses the total column of ozone. Roughly 150 ground stations perform total ozone measurements on a regular basis. Data are submitted into the World Ozone and UV Data Center (WOUDC), operated by Environment Canada (<http://www.woudc.org>), as part of the Global Atmo-

sphere Watch (GAW) programme of the World Meteorological Organization (WMO). The observations are predominantly taken with Dobson and Brewer UV spectrophotometers at about 60 and 70 stations respectively, but WOUDC also includes observations from UV-VIS DOAS spectrometers.

Even though Dobson and Brewer instruments are based on the same general measurement principle, previous studies have identified a seasonal bias of a few percent between their midlatitude total ozone column measurements, Brewer measurements being in slightly better agreement with satellite data than Dobson measurements. In the Northern Hemisphere, Dobson instruments exhibit a +1 % bias compared to Brewer instruments and the bias exhibits a seasonal cycle which is not the case for Brewer instruments (Scarnato et al., 2009; Lerot et al., 2013). Similar conclusions hold for the Southern Hemisphere. Since the Brewer network has not such a good coverage in the Southern Hemisphere, however, we use the Dobson instruments as a reference in the Antarctic, keeping in mind this +1 % bias compared to Brewer instruments (i.e. we did not correct the Dobsons for this bias, but instead used the original data).

In order to assess the quality of the total ozone columns (TOC) delivered by the 4 systems, we selected three stations from the WOUDC database for which the time coverage for this three year period was sufficiently large (red squares in Fig. 1: a high northern latitude station, Alert (82.49° N, 62.42° W, data gathered by the Meteorological Service of Canada), a tropical station, Chengkung (23.1° N, 121.365° E, data gathered by the Central Weather Bureau of Taiwan), and a southern latitude station, Syowa (69° S, 39.58° E, data gathered by the Japan Meteorological Agency). As indicated above, we used the observations gathered by the Brewer instruments at Alert and Chengkung, and those gathered by the Dobson spectrophotometer for Syowa. For Alert, we used the data for both Brewer instruments 019 (MKII) and 029 (MKV). The Brewer instrument (#061) at Chengkung is of type MKIV. Brewer data at $\mu > 3$ were filtered out. The Dobson instrument (#119) at Syowa was replaced on the 1st of February 2011 by a new Beck model (#122).

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2.4.2 SAOZ/DOAS observations

Conventional techniques for measuring ozone in the UV are inapplicable for SZA larger than about 80° . The SAOZ (Système d'Analyse par Observation Zenithale, Pommereau and Goutail (1988)), measuring the absorption by the atmosphere of scattered sunlight at the zenith sky, is the only instrument which is able to measure continuously and at all latitudes up to the polar circle in winter. SAOZ are designed to allow observations of NO_2 and O_3 total columns twice a day during twilight (sunrise and sunset). The retrieval is based on Differential Optical Absorption Spectroscopy (DOAS), a technique which has recently been improved by Hendrick et al. (2011). The accuracy of the zenith-sky UV-VIS spectroscopy for the measurement of total ozone has been investigated by Van Roozendael et al. (1998) by comparing against Dobson measurements. As a general result, the SAOZ O_3 measurements are between 2–8 % higher than the Dobson ones, with a scatter of about 5 % in midlatitudes and increasing at higher latitudes.

For the “Arctic ozone hole 2011” case study in this paper (see Sect. 6), the total ozone columns by the four analyses were compared with data received by three UV-VIS zenith-sky instruments at Arctic locations, which are part of the Network for the Detection of Atmospheric Composition Change (NDACC, <http://www.ndacc.org>): Zhigansk (Russia, 66.8°N , 123.4°E), Harestua (Norway, 60°N , 11°E), and Scoresby Sund (Greenland, 70.49°N , 21.98°W). Data at Harestua were gathered by BIRA-IASB (van Roozendael et al., 1995), the other two (SAOZ) instruments at Zhigansk and Scoresby Sund are owned by LATMOS/CNRS (Laboratoire Atmosphères, Milieux, Observations Spatiales/Centre National de Recherche Scientifique) respectively by CAO (Central Aerological Observatory) and by DMI (Danish Meteorological Institute).

2.4.3 Ozonesonde profiles

Balloon-borne ozonesondes measure the vertical distribution of ozone concentrations up to an altitude of about 35 km. The observed ozonesonde profiles are archived by NDACC, WOUDC, and the Southern Hemisphere ADditional OZonesondes network

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(SHADOZ, <http://croc.gsfc.nasa.gov/shadoz/>). The majority of soundings (85 %) are performed with Electrochemical Concentration Cell (ECC) sondes, while the remaining part consists of Brewer-Mast, Indian and Japanese Carbon-Iodine sondes. Optimally treated, ECC sondes yield profiles with random errors of 3–5 % and overall uncertainties of about 5 % in the stratosphere (Smit et al., 2007; Deshler et al., 2008; Stübi et al., 2008; Hassler et al., 2013). Other sonde types have somewhat larger random errors of 5–10 % (Kerr et al., 1994; Smit et al., 1996).

We use ozone observations gathered by balloon sondes at 38 locations, taken from the above-mentioned databases for the period September 2009 to September 2012: 12 in the Arctic, 19 in the Tropics, and 7 in the Antarctic (see Fig. 1). For each latitude band, we picked out one station which is representative for the general behaviour in this latitude band and for which the time coverage for this three year period was sufficiently large, for a more detailed discussion: the Arctic station at Ny-Ålesund (79° N, 12° E), the equatorial station at Nairobi (1.27° S, 36.8° E), and the Antarctic station at Neumayer (70.65° S, 8.25° W) (red dots in Fig. 1). Data are provided by the Alfred-Wegener Institute in Potsdam, Germany (for Ny Alesund and Neumayer) and by MeteoSwiss in Payerne, Switzerland (for Nairobi).

2.4.4 ACE-FTS satellite data

ACE-FTS is one of the two instruments on the Canadian satellite mission SCISAT-1 (first Science Satellite), ACE (Bernath et al., 2005). It is a high spectral resolution Fourier Transform Spectrometer operating with a Michelson interferometer. Vertical profiles of atmospheric parameters such as temperature, pressure and volume mixing ratios of trace constituents are retrieved from the occultation spectra, as described in (Boone et al., 2005), with a vertical resolution of maximum 3–4 km. Level 2 ozone retrievals (version 3.0) are used as an independent reference dataset to validate the ozone profiles of the MACC stratospheric ozone system.

It must be noted that the low spatio-temporal sampling of ACE-FTS (due to the solar occultation technique) does not deliver profiles in all latitude bands for each month.

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There are also two periods during the year where there are no measurements for a duration of almost 3 weeks due to the fact that the spacecraft is in constant sunlight: June and December (Hughes and Bernath, 2012). There are four periods per year, lasting about 1 month (Northern Hemisphere: April, June, August, December; Southern Hemisphere: February, June, October, December) with no occultation poleward of 60° (see Fig. 4 of Hughes and Bernath (2012)). At very high β angles (i.e. the angle between the orbital plane of the satellite and the Earth-sun direction $> 57^\circ$), it is common practise to skip more than half of the available measurement opportunities to avoid exceeding onboard storage capacities and overlapping command sequences. Therefore, the amount of observations in the Tropics is significantly lower than in the polar regions.

The previous version of these retrievals (version 2.2) was extensively validated against 11 other satellite instruments, ozonesondes and several types of ground-based instruments (Dupuy et al., 2009). This version reports more ozone than most correlative measurements from the upper troposphere to the lower mesosphere. Dupuy et al. (2009) found a “slight positive bias with mean relative differences of about 5% between 15 and 45 km. Tests with a preliminary version of the next generation ACE-FTS retrievals (version 3.0) have shown that the slight positive stratospheric bias has been removed.” Adams et al. (2012) additionally present an intercomparison of ACE ozone profiles (both versions 2.2 and 3.0) against groundbased observations at Eureka, confirming that the new ACE-FTS v3.0 and the validated v2.2 partial ozone columns are nearly identical, with mean relative difference of $0.0 \pm 0.2\%$ for v2.2. minus v3.0.

Standard deviations for levels where there are fewer than 20 observations are omitted for reasons of non-representativeness.

3 Validation of total ozone columns

We intercompare for the first time analyses based on data from different satellites and of different types: partial/total ozone columns, profile observations or a combination of both. For an optimal interpretation of the validation results, it is important to keep

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in mind that SACADA and TM3DAM exclusively assimilated total ozone columns, but while TM3DAM delivers only total ozone columns as output product, SACADA also provides ozone profiles. BASCOE exclusively assimilated vertical profiles of ozone (besides other species) and IFS-MOZART used a combination of total columns, partial columns and vertical profiles from various instruments.

In this section, we discuss the results obtained for the validation of the total ozone columns against Brewer observations at Alert (Arctic) and Chengkung (Tropics), and against Dobson observations in Syowa (Antarctic). The TOC datasets from the four systems were interpolated to the latitude and longitude of these stations. The resulting time series are shown in Fig. 2, side by side with the corresponding observed ground-based data. Remember that there are no SACADA total ozone products before 13 January 2011. Even though the time coverage at other locations was often poorer, the drawn conclusions are very similar (not shown).

3.1 Alert (Arctic)

The seasonal O₃ cycle at Alert is very similar each year. The only deviations from usual behaviour of the total ozone columns occur, e.g., in November 2009, when an airmass with exceptionally high ozone passed over Alert, and in February–March 2011, when 30 % of the total ozone column above Alert was destroyed by the end of March. The latter event will be studied in detail in Sect. 6 as a separate case study.

All four analyses match each other and the observed total ozone columns very closely. Peak-to-peak difference in TOC are of the order of 250 Dobson Units (D.U.), with maximum values reached during boreal winter and spring as a result of poleward and downward transport of ozone-rich air by the large-scale Brewer–Dobson circulation (Brewer, 1949; Dobson, 1956; Weber et al., 2011).

The only significant differences among the analyses occur during the O₃ maximum in northern spring (where mutual differences of maximum 50 D.U., about 10 %, are observed) and during the Arctic ozone hole season, where SACADA delivers TOC values which are about 75 D.U. (20 %) above the other analyses. Unfortunately, this coincides

TM3DAM assimilated SCIAMACHY data, but afterwards, TM3DAM switched to GOME-2, while IFS-MOZART continued to assimilate observations from SBUV/2, OMI, and MLS.

SACADA exhibits strong positive biases from observations during austral winters, right before the onset of the ozone hole (up to 30 % in 2012). Closer inspection of SACADA analyses shows that these larger differences coincide with missing SCIAMACHY and GOME-2 observations during polar night when solar zenith angles are close to or in excess of 90°. This coverage effect should especially influence models which assimilate data from UV-instruments only. The TM3DAM system is less vulnerable to data gaps than SACADA, as it performs very well under the same circumstances.

3.4 Discussion of SACADA total column results

All analyses show a realistic seasonal cycle in all three latitude bands and total ozone column values which are generally in very good agreement with independent observations. Differences between IFS-MOZART, BASCOE, and TM3DAM are usually within 5 %. Only a few exceptions were identified, i.e. larger mutual differences (up to 10 %) are found at high altitudes during polar night, and for BASCOE in the Tropics, where the model underestimates total ozone by 7–10 %.

In contrast to these three analyses, SACADA total ozone results deviate strongly from observations during certain episodes. There is a general tendency in SACADA results for positive biased ozone columns during the winter months at high latitudes compared to Alert and Syowa station data in the Northern and Southern Hemisphere, respectively. Backscatter UV-instruments provide no information for zenith angles above 90°. As recommended for SCIAMACHY data version 3 (Lerot et al., 2007), only observations with zenith angles up to 75° were used. Thus, no SCIAMACHY data were assimilated until May 2011 at the latitudes of Alert station (82.49° N). Accordingly, at Syowa station (69° S), SCIAMACHY data was not processed from end of March until end of September 2011. Data quality improved with SCIAMACHY version 5 (which is

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used for this analysis, even though recommendations for version 3 were maintained) and the coverage would significantly improve with zenith angles up to 80°.

From 28 October 2011 onwards, GOME-2 observations were assimilated by SACADA up to zenith angles of 90°. In this case, the instrument is blind from mid September 2011 to April 2012 at Alert, and from mid April to mid September at Syowa. These time periods correlate in general well with the positive bias anomalies in ozone columns found in SACADA results. The area of impact of a total column observation on assimilation results is limited by the background correlation matrix, which uses a horizontal correlation radius of 600 km. Latitudes not covered by observations can therefore only be influenced via tracer transport. In summary, we conclude that the biases evident, reflect a general tendency of the SACADA model to overestimate total ozone at high latitudes.

4 Validation of the vertical distribution of stratospheric ozone against ozonesondes

In this section, we discuss the results obtained for the validation of the ozone profiles against ozonesonde observations at Ny Alesund (Arctic), Nairobi (Tropics), and Neumayer (Antarctic).

In order to compare the ozone fields from the 3 systems with the observed ozonesonde data, the modelled ozone fields were first linearly interpolated to the geographical location of the launch sites. Even though sondes may drift long distances during their ascent, especially within the polar vortex, this often significant horizontal movement was disregarded, as tracking information is not always available. As a next step, the two analysis profiles preceding and following the measurement closest in time were linearly interpolated to the time of observation. Since the ozonesonde profiles have a much higher vertical resolution than their modelled counterparts, the ozonesonde data have been log-linearly interpolated on the coarser pressure grid, degrading the observations to the lower resolution of the model. Figure 3 shows time

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series of the monthly mean ozone bias profiles with respect to the ozonesondes at the selected sites for each of the three MACC systems.

As the ozone number density reaches its stratospheric maximum between 20 and 30 km altitude, depending on latitude (Brasseur and Solomon, 2005), we used the 50 hPa level as indicator for the system performance in the middle stratosphere. Figure 4 shows the time series of the ozone volume mixing ratio at this pressure level, allowing to identify the seasonal cycle of ozone in the middle stratosphere and to intercompare the performance of the different analyses. This time, no time interpolation has been done. Instead, 5 day moving averages of the ozone analyses have been calculated, to exclude any severe influence of differences in the original output frequency.

4.1 Arctic – Ny-Ålesund

The seasonal cycle at Ny-Ålesund is very well reproduced by the three analyses. Biases at Ny-Ålesund are generally smaller than 20 % for all MACC analyses throughout the stratosphere (Fig. 3). The time series of the ozone profiles shows alternating behavior in the vertical for IFS-MOZART, persistent over the entire 3 year period, with positive biases in the lower (below 70 hPa) and upper (above 20 hPa) stratosphere and no or only slightly negative biases (mostly 5–10 %) in the middle stratosphere. The performance of BASCOE is stable throughout the stratosphere and for the entire 3 year period, with biases mostly less than 5 %. Largest biases over the whole period for IFS-MOZART (–20 to –30 % between 50 and 70 hPa) and for SACADA (> 50 % between 35 and 65 hPa) are found for March 2011. While the ozone hole simulated by IFS-MOZART is too deep, SACADA simulates an Arctic ozone hole which is not deep enough. This special event will be discussed in detail in Sect. 6. Until March 2011, SACADA mainly overestimates ozone over the entire altitude range, while middle stratospheric ozone is mostly underestimated afterwards.

At 50 hPa (Fig. 4), IFS-MOZART and BASCOE deliver very similar results (differences < 5 %) throughout the 3 year period, in good agreement with the O₃ sonde data at this altitude, while SACADA analyses show a tendency to slightly higher mixing

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ratios until March 2011 and slightly lower mixing ratios during the period September 2011 to January 2012. The error is about 10 %. From northern spring 2012 onwards, all assimilation results are very close.

The depletion of ozone due to the Arctic ozone hole in northern spring 2011 is very prominently present (Fig. 4). IFS-MOZART and BASCOE reproduce this extraordinary behaviour very well.

4.2 Tropics – Nairobi

The O₃ bias profile time series (Fig. 3) now displays a changing performance in the vertical for all three analyses. Lower stratospheric ozone is underestimated by more than 40 % by both IFS-MOZART and BASCOE (below 80 hPa and below 100 hPa, respectively) throughout the year. For BASCOE, this is followed by a small pressure range just above (between 75 and 90 hPa) where ozone values are overestimated by more than 50 %. The results for the remaining middle to upper part of the stratosphere are almost identical to the observed ozonesonde values at Nairobi, although with a tendency to overestimate O₃ by IFS-MOZART (< 20 %). SACADA, on the other hand, overestimates ozone below 40 hPa with more than 50 %, while its performance is usually very good above. Between July 2011 and May 2012, however, SACADA underestimates O₃ with up to 20 %, and even 30 % from September to December 2011, in the pressure range between 10 and 30 hPa.

This is reflected in the time series at 50 hPa: all analyses overestimate O₃. Whereas IFS-MOZART and BASCOE produce very similar results at 50 hPa (IFS-MOZART slightly above BASCOE) with biases of about 15 % compared to the ozonesonde data at Nairobi, and even up to 30 % in the period August–November 2010, SACADA shows ozone values which are at least 35 % higher than the other two analyses, while the seasonality is well reproduced. The discontinuity in the SACADA products from 6 to 7 September 2010, is due to resumption of the assimilation after a period where SACADA ran freely (July–September 2010) due to a data gap in the assimilated SCIAMACHY. As mentioned before for the total ozone columns, the SACADA analysis tends

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to drift in the absence of UV observations for the assimilations. Once resumed, the assimilation reduces the mismatch with the other two analyses from 60 % down to only 10 %.

4.3 Antarctic – Neumayer

5 The O₃ bias profile time series show that the biases are smallest and most stable for BASCOE (usually less than 10 %). IFS-MOZART on the other hand has an annually recurrent pattern, overestimating O₃ with more than 50 % between roughly 70 and 150 hPa each Antarctic ozone hole season, from September till December, while underestimating ozone between 30 and 60 hPa in September. This indicates that
10 IFS-MOZART has problems with a correct simulation of the ozone depletion. This is a known problem of the underlying MOZART CTM in the MACC configuration which cannot be completely fixed by the data assimilation (Flemming et al., 2011; Inness et al., 2013), especially because the assimilated profile only gives information down to 68 hPa. MOZART performs better with WACCM meteorology (Kinnison et al., 2007),
15 which indicates that the chemical parameterizations are sensitive to the meteorological fields that are used to drive transport in the models. SACADA has problems to correctly simulate the ozone concentration in the lower stratosphere (below 80 hPa) . While the ozone hole depth of 2010 is underestimated (positive bias), the corresponding ozone depletion in 2011 and 2012 is overestimated by more than 50 %. This is related to the
20 premature onset and end of the ozone depletion as predicted by the model, which is reflected also in the ozone values at 50 Pa. Apart from this, the observed ozone values at Neumayer at 50 hPa are in general well reproduced by the three analyses of the MACC system. IFS-MOZART and BASCOE differ only little from each other.

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the tropopause is very hard to define in the models. Ozone concentrations are rapidly decreasing and tend to increase the relative differences.

For BASCOE, two more elements play a role in the worse performance in the lower tropical stratosphere: the low vertical resolution and aliasing errors in the horizontal wind fields, which are larger close to the UTLS and which lead to noise in the horizontal distribution of chemical tracers. This bug has been corrected in an upgraded version, which is running operationally since the beginning of 2013. The vertical grid of the model is improved and extended, from 37 levels (model top at 0.1 hPa) to 91 levels (model top at 0.01 hPa) and with a much finer resolution in the UTLS region. Comparison between both versions shows that O₃ values become lower around 80 hPa and higher lower down (which would thus correct the currently large biases in these regions).

The larger biases for IFS-MOZART in the lower stratosphere globally (i.e. not only at the Tropics, but also at the poles, especially in the Antarctic) may also result from the fact that the useful range of the NRT MLS v2.2 data is restricted to levels above 68 hPa, which means that it includes no profile information below that pressure level, in contrast to BASCOE, which assimilates the offline MLS v2.2 dataset down to 150 hPa. Tests with the improved NRT MLS v3.4 data (Livesey et al., 2011), which have a useful range of 261 to 0.1 hPa and which are used in a later version of IFS-MOZART, show that a lot of those biases in the lower stratosphere disappear (see also Sect. 6.2).

To illustrate that the selected stations at each latitude band are representative for the results at all stations and that the same conclusions hold in general, we additionally show the mean ozone profiles and ozone bias profiles for the MACC analyses compared to all considered ozonesonde measurements in each latitude band (see Fig. 1), averaged over the entire 3 year period from September 2009 to September 2012 (Fig. 5). On average, all analyses agree with the sondes mostly to within $\pm 10\%$ above 70 hPa. Larger biases are observed for IFS-MOZART in the upper stratosphere (above 10 hPa) at the poles and in the lower stratosphere with overall biases reaching 30% in the Antarctic and -40% at the equator, and for BASCOE below 150 hPa.

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Standard deviations between the MACC systems and the ozonesondes are smallest for BASCOE, and only slightly higher for IFS-MOZART, usually between 10 and 20 %, except for the region below 70 hPa in the Tropics. The standard deviations for IFS-MOZART are higher in the area between 60 and 100 hPa in the Tropics, and between 100 hPa and 200 hPa in the Antarctic.

4.4.3 Influence of the temporal and horizontal resolution

SACADA data are sampled only once a day (at 12 h UT), IFS-MOZART 6 hourly, and BASCOE data 3 hourly. This may affect its performance when compared to ozone sondes. To exclude the effect of temporal resolution, we have degraded the temporal resolution of both IFS-MOZART and BASCOE to the temporal resolution of SACADA.

Relative differences between the fine and the coarse temporal resolution datasets are usually less than 2 %, but can be as high as 10 % for some months, and at some altitudes without any clear pattern (figures not shown). The effect on the standard deviation of the differences when using the 24 h resolution dataset for all three analyses appears to be minimal (Fig. 5). Only in the lower tropical stratosphere, somewhat larger differences (compared to their original temporal resolution) in the standard deviations for IFS-MOZART and BASCOE appear, but they never reach the high values of the SACADA data.

On the other hand, also a lower horizontal resolution may lead to larger standard deviations. BASCOE and SACADA have, however, the same horizontal resolution (3.75° by 2.5°), which is coarser than for IFS-MOZART (1.875° by 1.875°). This illustrates that the differences in standard deviations between the MACC systems is not exclusively dependent of the temporal nor the horizontal resolution.

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5 Validation of the vertical distribution of stratospheric ozone against ACE-FTS

Additionally to the groundbased and ozonesonde data, the MACC ozone analyses have been compared to independent ACE-FTS satellite observations. The comparison between the measurements by ACE-FTS and the analysis output is performed in the following manner. The analyses, first regridded to a common $1^\circ \times 1^\circ$ grid, are collocated with the ACE-FTS data in space (horizontally and vertically) and time through linear interpolation. Since SACADA results are only provided every 24 h, we assume a constant composition throughout the day. Monthly mean biases of the spatial-temporal collocated data are calculated for 5 latitude bins, using 25 pressure bins based on the standard UARS fixed pressure grid (i.e., six pressure levels per decade which corresponds approximately to 2.5 km). These monthly mean biases and their associated standard deviations can be displayed as time series of the monthly mean bias (e.g. Figs. 6 and 7) or as vertical profiles (e.g. Fig. 8).

In view of the problems to constrain the model's ozone profile from total column assimilation, shown earlier, we will still include the results for SACADA in the figures, but we will not include the model in the discussion.

5.1 Partial ozone columns

The time series of the standard deviations in Fig. 6 gives a global view of how well the analyses are performing against the satellite data. The standard deviations are averaged over the entire globe (90° S– 90° N) and over the entire stratospheric area of interest (200–5 hPa). As shown earlier, when comparing with groundbased and ozonesonde observations, IFS-MOZART and BASCOE perform very similarly. Standard deviations are on average around 6–7 %, while the highest values are found around March and August each year.

Binning into a stratospheric pressure layer (100–5 hPa for the Tropics, and 200–5 hPa for all other latitude bands) shows small overall mean biases for both systems. Individual monthly mean biases for IFS-MOZART and BASCOE always remain below

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5%, which shows that these analyses have an overall stable behaviour (figure not shown).

5.2 Ozone at predefined pressure levels

Even though partial columns indicate an excellent and stable behavior for both IFS-MOZART and BASCOE, interpolation at specific pressure levels (10, 50, and 100 hPa) reveals an anticorrelation of the biases in the vertical for IFS-MOZART, both in the Arctic and Antarctic, especially in association with ozone hole conditions (Fig. 7), which was also seen earlier in the comparison with O₃ sondes (Fig. 3). These vertical oscillations in bias compensate to deliver correct (assimilated) partial or total columns (see Sect. 3).

In the Arctic, biases are, for all analyses, largest in March 2011. Biases remain low for BASCOE (<10%), but attain values up to 20% for IFS-MOZART. One obvious explanation is the occurrence of extreme conditions in the Arctic, with the appearance of the first Arctic ozone hole Manney et al. (2011). However, another issue may have an influence on the system performance in this period: on the 26 March 2011, Aura MLS stopped sending data until the 18 April. Since these data are assimilated both by IFS-MOZART and BASCOE, it means that BASCOE is running freely during this period, while IFS-MOZART is only assimilating partial or total ozone columns. The effect is, however, expected to be minimal for March, as it concerns only 5 days (out of 30 for which collocations with the ACE-FTS measurements are found) in the monthly mean. Since the assimilation of Aura MLS data was only reactivated in IFS-MOZART on the 10 May 2011, while BASCOE restarted to assimilate Aura MLS as soon as the data were back online (i.e. on 19 April), the effect of missing profile information is expected to become more apparent in April. Unfortunately, ACE-FTS did not collect any measurements in the Arctic in April 2011 (see Sect. 2.4.4), but the earlier comparison with O₃ sondes (Fig. 3) does give an indication that the larger biases disappear in April 2011. This illustrates that it are indeed the extreme Arctic conditions which lie at the basis of the larger biases in March.

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The same conclusions can be drawn for the Antarctic during the yearly ozone hole conditions. Biases for BASCOE still remain within 10 %, but are more pronounced for IFS-MOZART than in the Arctic, especially in the lower stratosphere (100 hPa), where relative differences up to almost 50 % in 2011 and even 60 % in 2010 are found in September, even now that Aura MLS data are available for assimilation. The stable behaviour of BASCOE in time may be due to the fact that BASCOE assimilates the same data all year round (day and night time), while IFS-MOZART also assimilate UV-VIS data which are not available at all times of the year at the poles. Unless the chemistry scheme of MOZART can be improved to better simulate ozone hole conditions without data assimilation, the assimilation of vertical profiles is essential for the stratospheric forecasting system.

5.3 Seasonal mean ozone profiles

Figure 8 shows seasonally averaged relative ozone biases for austral spring and boreal winter, for the three consecutive years in the studied period. BASCOE has a stable performance compared to ACE-FTS data throughout the stratosphere, very similar each year, but slightly underestimating ozone with an average of 5 % in the Arctic. While the biases vary between –10 and 0 % in austral spring 2010, the variability is larger (biases between –15 and +5 %) in austral spring 2011. The seasonal mean biases of IFS-MOZART again illustrate the oscillating behaviour of the profiles, both in the Arctic and Antarctic. Antarctic biases appear to be three times as large as those in the Arctic, and largest for the first year. The stable performance of BASCOE in the vertical can be explained by the fact that BASCOE is only assimilating profile observations, whereas IFS-MOZART highly relies on (partial and total) column observations.

6 Arctic ozone hole event 2011

6.1 Case study

Besides the overall performance of the different analyses, we want to evaluate the ability of the MACC system to capture special events, such as the yearly recurrent Antarctic ozone holes, or the exceptional Arctic ozone hole in northern winter/spring 2011 (Manney et al., 2011). Long-lasting exceptionally cold conditions prevailing over the Arctic, together with man-made ozone-depleting compounds lingering in the atmosphere, caused the destruction of almost 40 % of stratospheric ozone by the end of March (Manney et al., 2011). In this section, we address the performance of the MACC system during this particular event. Throughout the previous discussions, we have already shown that biases with respect to observations are largest at the peak of the ozone hole (i.e. March 2011), illustrating that most systems have difficulties to correctly simulate such an unexpected event. In view of the fact that SACADA did not assimilate SCIAMACHY data at high northern latitudes before May 2011 (see earlier), we have omitted the discussion of SACADA results in this particular case study (results are only shown in Fig. 11).

Figure 9 shows the evolution of the ozone depletion, as simulated by IFS-MOZART and BASCOE in the north pole vortex at 485 K potential temperature (~ 20 km, ~ 50 hPa) during the month March 2011. The vortex is determined by the potential vorticity (PV). Two contours of scaled' PV (sPV) delimit the outer and inner vortex edges, respectively using an sPV of $1.4 \cdot 10^{-4}$ (as in Manney et al., 2011) and $1.7 \cdot 10^{-4} \text{ s}^{-1}$.

Manney et al. (2011) showed that, in February–March 2011, the barrier to transport at the Arctic vortex edge was the strongest in either hemisphere for the last ~ 30 years. This barrier isolates the cold air in the vortex, preventing it from mixing with air in the mid-latitudes, causing a build-up of ozone, brought by long-range transport outside the vortex. At the same time, the return of sunlight after the polar night, releases the ozone-destroying gases trapped in the polar stratospheric cloud (PSC) particles within the vortex, breaking down ozone into individual oxygen molecules.

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From late February/early March 2011 on, reduced levels of ozone are observed inside the vortex and the ozone hole starts to develop. The largest chemical loss was recorded on the 26 March. At that time, a stretched vortex is covering Scandinavia and North-West-Russia.

As seen in Fig. 9, IFS-MOZART and BASCOE provide very similar results, BASCOE values being slightly higher and slightly noisier than the IFS-MOZART ones. The slightly higher noise is reflected in the standard deviation (Fig. 6), but has been corrected in a later version of BASCOE (see earlier).

We compared the IFS-MOZART, BASCOE, and TM3DAM analyses with data received by 3 UV-VIS SAOZ/DOAS instruments at Arctic locations, which are part of the NDACC: Zhigansk (Russia, 66.8° N, 123.4° E), Harestua (Norway, 60° N, 11° E, DOAS), and Scoresby Sund (Greenland, 70.49° N, 21.98° W) (see Sect. 2.4.2). From Fig. 9, we see that on 1, 13, and 26 March, respectively only Zhigansk, only Scoresby Sund, and only Harestua are located inside the wintertime polar vortex. The observations are well reproduced by the three analyses, with a tendency to a slight overestimation.

Comparison with ozone soundings at Ny-Ålesund (Spitsbergen), which is always located within the polar vortex, shows that both IFS-MOZART and BASCOE could correctly reproduce the ozone hole conditions with relative biases mostly less than 10% in the stratosphere (Fig. 11). IFS-MOZART has, however, problems with a correct simulation of the vertical profile, when the ozone depletion is strongest (in March 2011) and alternating positive and negative biases up to 30% can be seen.

6.2 Influence of the assimilated data on the performance of the analyses

We suspect that the significantly larger biases for the NRT stratospheric ozone products delivered by SACADA and to a lesser extent by IFS-MOZART in the vertical, compared to BASCOE are mainly due to the assimilated dataset of MLS, i.e. the fact that BASCOE assimilated Aura-MLS v2.2 Offline ozone which is valid down to 261 hPa (besides MLS HNO₃, HCl, H₂O and N₂O) while IFS-MOZART assimilated Aura-MLS v2.2 NRT ozone down to 68 hPa (and various column obs) and SACADA assimilated only

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total column data from UV observations. To test this hypothesis, we defined three new experiments in which all three models assimilate the same data, namely: Aura MLS version 3.3 offline ozone down to 261 hPa (i.e. no additional species such as HNO₃, HCl, H₂O and N₂O). We focused on the period considered in the Arctic ozone hole 2011 case study and hence, the models were run for one month (March 2011) with a short spin-up period of about one week.

Figure 12 (left) shows the mean bias and standard deviations of the differences between the NRT (i.e. the original) analyses and ACE-FTS observations, keeping only the (~200) ACE profiles within the North Pole vortex, with the vortex edge calculated with an sPV of $> 1.7e^{-4} s^{-1}$. At ozone hole level ($\theta \sim 485$ K), we see the large underestimation in IFS-MOZART and the absence of ozone depletion in SACADA. This figure is to be compared with Fig. 12 (right), which shows the results of the three new offline experiments assimilating the same dataset. Now, all analyses perform very similarly. The biases and standard deviations are smallest for IFS-MOZART, which might be related to the higher horizontal resolution. The standard deviations for SACADA are slightly larger than the ones for IFS-MOZART and BASCOE, which may be due to the lower time sampling (24 h output frequency instead of 6 h for the two other). The BASCOE experiment delivers smaller bias and standard deviations than for the original NRT analysis. This may be related to the assimilation of MLS offline v3.3 instead of MLS offline v2.2.

The results for IFS-MOZART show that the increased vertical range of MLS V3 is beneficial compared to the more limited range of the NRT MLS V2 data. An additional IFS-MOZART experiment assimilating, besides MLS V3, also the other O₃ column products (as defined in Table 1) shows that the analysis is well constrained by MLS alone in the stratosphere, while it is beneficial to have the combination of profile and total column data in the troposphere.

7 Conclusions

This paper presents the NRT stratospheric ozone service delivered in the framework of the MACC project. The service is based on four independent data assimilation systems: IFS-MOZART, BASCOE, SACADA, and TM3DAM. Two of them (SACADA and TM3DAM) assimilate only total O₃ columns, consecutively SCIAMACHY and GOME-2. The IFS-MOZART analysis is also based on SCIAMACHY data (at least until the end of Envisat), but additionally assimilates total O₃ columns of OMI, O₃ partial columns of SBUV-2, and profiles of MLS. The delayed, but more extended MLS profile dataset is the unique input for the BASCOE analyses. This paper presents the validation results of these ozone analyses against groundbased observations, ozonesondes and ACE-FTS satellite retrievals for the period September 2009 to September 2012. All intercomparisons show consistent results. Data assimilation seems to work well in all systems. When ozone columns are assimilated, columns are well reproduced. Assimilation of profiles does lead to a good representation of profiles. Model differences only show up where the systems are not well constrained by the observations, and this is where BASCOE performs best.

All analyses show a realistic seasonal cycle in the three considered latitude bands and total column values are generally in very good agreement with independent Brewer/Dobson observations. The UTLS is the most difficult range to model for all analyses. Large biases are found especially in the Tropics, where tropospheric air masses reach higher altitudes.

On the other hand, each system has its own strengths and weaknesses. IFS-MOZART assimilates the largest variety of satellite instruments simultaneously, going from partial and total ozone columns to ozone profiles. While large weight is given to total column observations in the assimilation procedure (hence the very good reproduction of total columns), the only profile information comes from the NRT MLS observations, which can only be used down to 68 hPa. Hence the input into the analysis is limited, leading to alternating positive and negative biases in the vertical compared

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to ozonesonde and ACE-FTS satellite data. Biases are largest (30 to 60 %) during ozone depletion events. This is a known problem of the underlying MOZART CTM in the MACC configuration, which improves with WACCM meteorology, but which cannot be corrected by the assimilation due to missing profile information at these altitudes.

5 The BASCOE analysis is based exclusively on ozone profiles, in this case from MLS, without any changes for the entire period, leading to stable results throughout the year. The assimilation of MLS data gives information during the polar night when the UV instruments GOME-2, SBUV/2, SCIAMACHY and OMI can not observe the ozone field because there is no backscattered solar radiation. With its purely stratospheric design,
10 larger systematic biases for the total ozone columns are expected, but still BASCOE delivers reliable TOCs. Only in the Tropics, slightly larger (negative) biases are observed. The exclusive assimilation of MLS profiles and the fact that BASCOE is specifically designed to deal with stratospheric chemistry processes leads to ozone profiles which are in very good agreement (biases less than 5–10 %) with the observed profiles delivered by ozonesondes and the ACE-FTS satellite observations throughout the
15 entire stratosphere and for the entire period between September 2009 and September 2012.

20 Compared to the other three CTMs, TM3DAM is designed to assimilate only total column observations. A bias correction is applied to the satellite observations to reduce on average the bias with the surface Brewer/Dobson observations. We showed that TM3DAM provides very realistic total ozone columns. Unfortunately, we could not compare the profiles with independent data because the 3-D model output is not provided.

25 SACADA is the only chemical data assimilation system with full chemistry which (just like TM3DAM) assimilates total column ozone only, from one UV satellite instrument at a time. As long as reliable UV-data for the assimilation are available, SACADA is able to reproduce observed total ozone columns. If the model is running unconstrained, large drifts in ozone are observed. In contrast to TM3DAM, SACADA does attempt to provide vertical profile information as well, by appropriately scaling the mixing ratios by an

altitude independent factor using the models one ozone profile. The lack of information constraining the ozone profile leads primarily to an overestimation of ozone in the lower stratosphere and in the Antarctic, especially during ozone hole conditions.

We have shown that assimilating total columns only provides total column values which are usually in good agreement with independent observations (SACADA, TM3DAM), but that these analyses are not able to provide reliable information about the ozone vertical distribution (SACADA). Even more, ozone analyses with realistic total ozone column densities (IFS-MOZART) are not necessarily in good agreement with the observed ozone profiles. This study illustrates that even state-of-the-art models of stratospheric chemistry still require the assimilation of limb observations for a correct representation of the vertical distribution of ozone in the stratosphere. This is corroborated by the results of three experimental runs, in which IFS-MOZART, BASCOE, and SACADA assimilate the same dense MLS dataset over the entire stratosphere for the month March 2011. The profile results of the three analyses are now very close, with much reduced biases. The IFS-MOZART experiment exhibits now the best performance (lowest biases and standard deviations). This illustrates the large benefit obtained from the assimilation of a single limb-scanning instrument (Aura MLS) with a high density of observations. Therefore, we can only share the serious concern about the lack of ozone-profiling capabilities at high vertical resolution at short term which has been expressed earlier on many occasions in official reviews of international monitoring capacities (e.g. WMO, 2011b). Only when ozone profile information is available, are the CTMs able to capture events of scientific interest or events of exceptional nature.

Nadir UV instruments such as GOME-2 do bring profile information into the stratosphere, but with a limited vertical resolution of about 5 km. Therefore, there is currently only little interest in the assimilation of nadir profiles, but they may provide a solution in case we lose Aura.

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Framework Programme for research and technological development including demonstration activities. MACC ran from 1 June 2009 to 31 December 2011 and was followed up by MACC II with a two-month overlap, from 1 November 2011 on, for a period of 33 months. MACC II is funded under grant agreement CP-CSA 283576. The authors thank the MACC GRG and VAL teams for constructive discussions.

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Table 1. Satellite retrievals of ozone that were actively assimilated by the four models of the MACC stratospheric ozone service. The Aura MLS data used by IFS-MOZART and BASCOE are not the same: IFS-MOZART used the MLS NRT retrievals of ozone only, while BASCOE uses the standard scientific, offline retrievals including five other species. PC stands for partial columns, TC for total columns and PROF for profiles. When two references are provided, the first refers to the satellite sensor, the second one to the retrieval algorithm.

Analysis	Satellite	Sensor	Provider	Version	Assim. data	Period	Reference
IFS-MOZART	NOAA-16/17/18	SBUV/2	NOAA	V08	PC	1 Sep 2009–30 Sep 2012	Bhartia et al. (1996)
	Aura	OMI	NASA/JPL	V003	TC	1 Sep 2009–30 Sep 2012	Levelt et al. (2006)
	Envisat	SCIAMACHY	KNMI	TOSOMI v2.0	TC	1 Sep 2009–7 Apr 2012	Eskes et al. (2005)
	Aura	MLS	NASA/JPL	V2.2, NRT	PROF, <68 hPa	1 Sep 2009–30 Sep 2012	Waters et al. (2006) Livesey et al. (2006)
BASCOE	Aura	MLS	NASA/JPL	V2.2, SCI	PROF	1 Jul 2009–30 Sep 2012	Waters et al. (2006) Livesey et al. (2006)
SACADA V2.0	Envisat	SCIAMACHY	DLR, on behalf of ESA	SGP-5.01	TC	5 Mar 2010–27 Oct 2011	von Bargaen et al. (2007)
SACADA V2.4	MetOp-A	GOME-2	EUMETSAT	GDP 4.1	TC	28 Oct 2011–30 Sep 2012	Loyola et al. (2011) van Roozendaal et al. (2006)
TM3DAM	Envisat	SCIAMACHY	KNMI	TOSOMI v2.0	TC	16 Mar 2010–31 Mar 2012	Eskes et al. (2005)
	MetOp-A	GOME-2	DLR	GDP 4.x	TC	1 Apr 2012–30 Sep 2012	Loyola et al. (2011) van Roozendaal et al. (2006)

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Table 2. Specification of the characteristics of the four assimilation systems: IFS-MOZART, BASCOE, SACADA, and TM3DAM. The horizontal and vertical resolution have been abbreviated to Hor. and Vert. resol. respectively. Freq. stands for frequency, and Assim. for assimilation.

	IFS-MOZART	BASCOE	SACADA	TM3DAM
Hor. resol.	1.875° × 1.875°	3.75° × 2.5°	250 km	3° × 2°
Vert. resol.	60 layers up to 0.1 hPa	37 layers up to 0.1 hPa	32 layers between 7 and 66 km	44 layers between 0 and 1013 hPa
Output freq.	6 hourly	3 hourly	daily, at 12 h UT	daily, at 21 h UT
Meteo input	operational hourly meteo fields from IFS	operational 6 hourly meteo analyses from IFS	24 h GME forecast initialised by IFS analyses	operational 6 hourly meteo analyses from IFS
Advection scheme	flux form semi-Lagrangian (Lin and Rood, 1996)	flux form semi-Lagrangian (Lin and Rood, 1996)	semi-Lagrangian and upstream method	flux-based second order moments scheme (Prather, 1986)
Chemical mechanism	JPL-06 (Sander et al., 2006) with some modifications as described in Stein (2013) 115 species 325 reactions	JPL-06 (Sander et al., 2006) Aerosols and PSCs 57 species 200 reactions	JPL-06 (Sander et al., 2006) Aerosols and PSCs (Damski et al., 2007) 48 species 177 reactions	Cariolle parameterization + cold tracer (2 species) (Cariolle and Teyssèdre, 2007)
Assim. method	4D-Var	4D-Var	4D-Var	Kalman Filter approach
Assim. window	12 h	24 h	24 h	24 h

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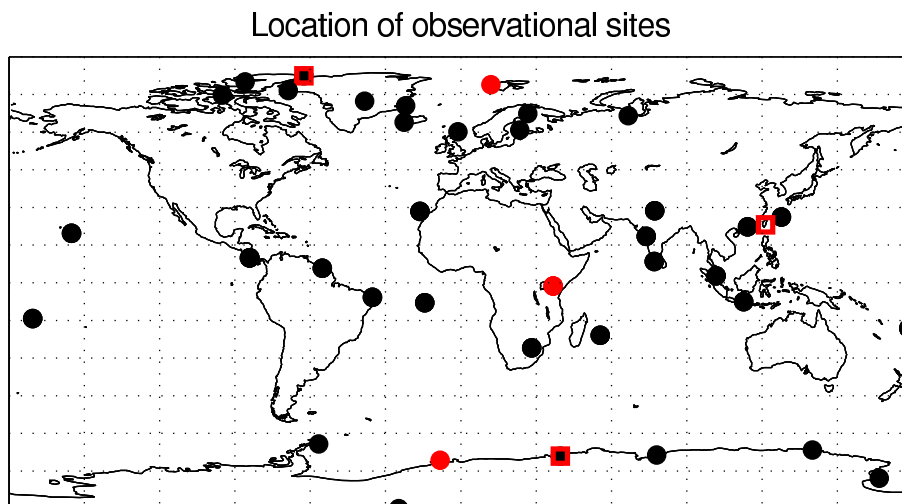


Fig. 1. Location of all stations used in this paper. O₃ sondes are indicated as filled black circles. The ones selected for a more detailed discussion have been marked in red: Ny-Ålesund (79° N, 12° E) in the Arctic, Nairobi (1.27° S, 36.8° E) in the Tropics, and Neumayer (70.65° S, 8.25° W) in the Antarctic. The three sites selected for the Total Ozone Column (TOC) discussion are indicated by the red squares.

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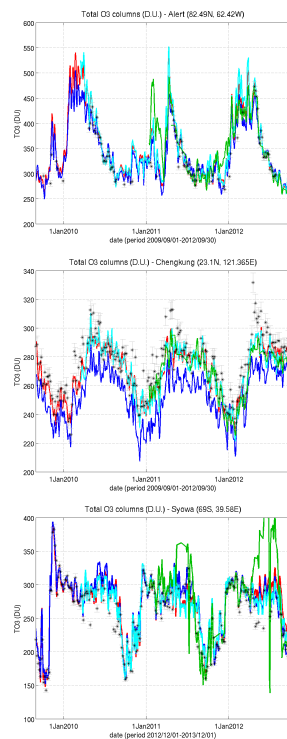


Fig. 2. Comparison between the TOC time series (5 day moving average) of the four analyses of the MACC stratospheric ozone service (IFS-MOZART in red, BASCOE in blue, SACADA in green, and TM3DAM in cyan) interpolated at a high northern latitude station (Alert, 82.49° N, 62.42° W), a tropical station (Chengkung, 23.1° N, 121.365° E) and a southern latitude station (Syowa, 69° S, 39.58° E), for the period September 2009 to September 2012. Black asterisks are 5 day moving averages of Brewer (for Alert and Chengkung) and Dobson (for Syowa) observations from the WOUDC network. A conservative observational error of 2% is indicated by gray errorbars.

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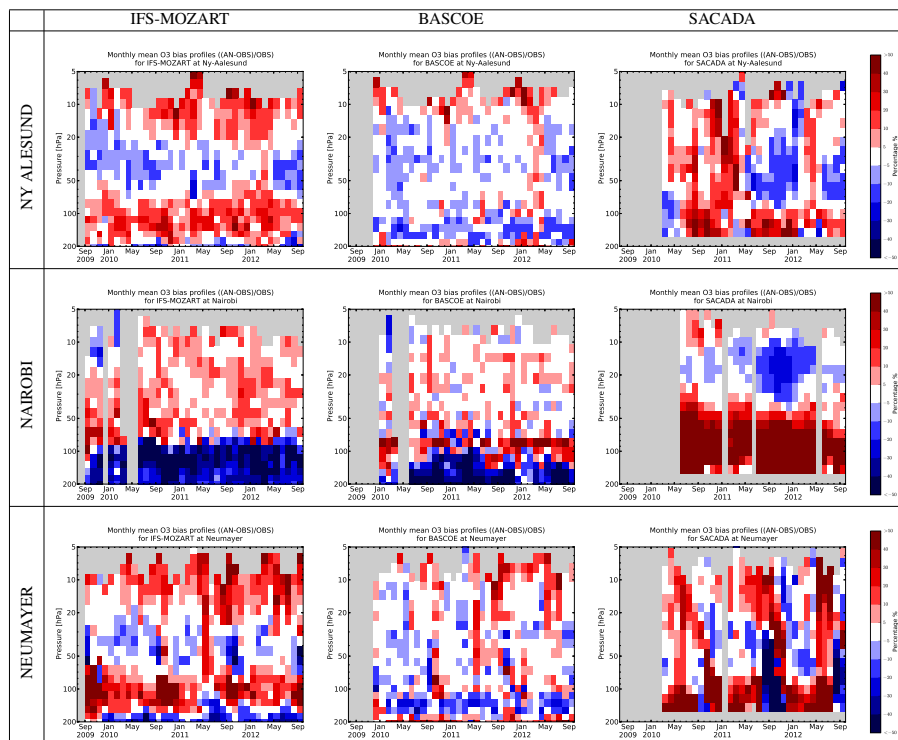


Fig. 3. Time series of monthly mean ozone biases (analysis minus observations) with respect to ozonesondes at Ny-Ålesund (top panel, 78.92° N, 11.93° E), Nairobi (middle panel, 1.27° S, 36.8° E) and Neumayer (bottom panel, 70.68° S, 8.26° W) for the period September 2009 to September 2012 in %. Left: IFS-MOZART, middle: BASCOE, right: SACADA.

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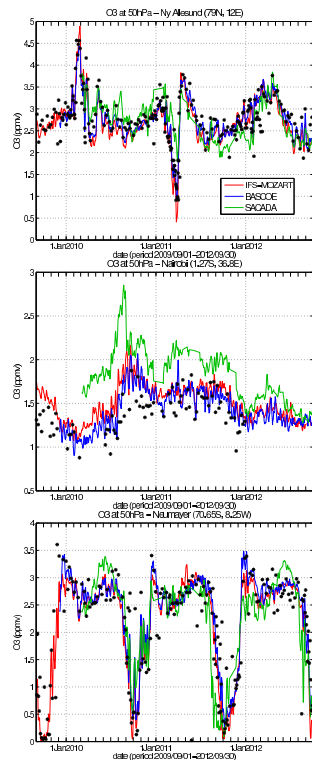


Fig. 4. Comparison between the time series (5 day moving average) of ozone at 50 hPa by IFS-MOZART (red), BASCOE (blue), and SACADA (green) interpolated at Ny-Ålesund (top panel, 78.92° N, 11.93° E), Nairobi (middle panel, 1.27° S, 36.8° E) and Neumayer (bottom panel, 70.68° S, 8.26° W), for the period September 2009 to September 2012. Black asterisks are 5 day moving averages (except for Nairobi where observations were taken at least one week apart) of O₃ sonde observations from the NDACC network.

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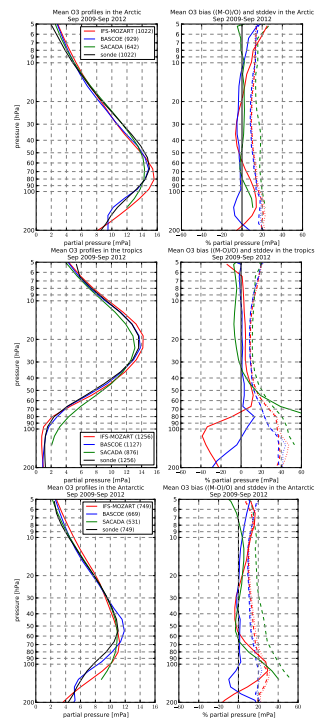



Fig. 5. Average ozone profiles (left) as partial pressures in mPa from IFS-MOZART (red), BASCOE (blue), SACADA (green) and ozonesondes (black), and the average ozone bias (solid lines) and standard deviation (dashed) in % of these analyses against the ozonesondes (right) over the period September 2009 to September 2012. Dotted lines represent the standard deviation for BASCOE and IFS-MOZART with the temporal resolution degraded to the one of SACADA. See Sect. 4.4.3.

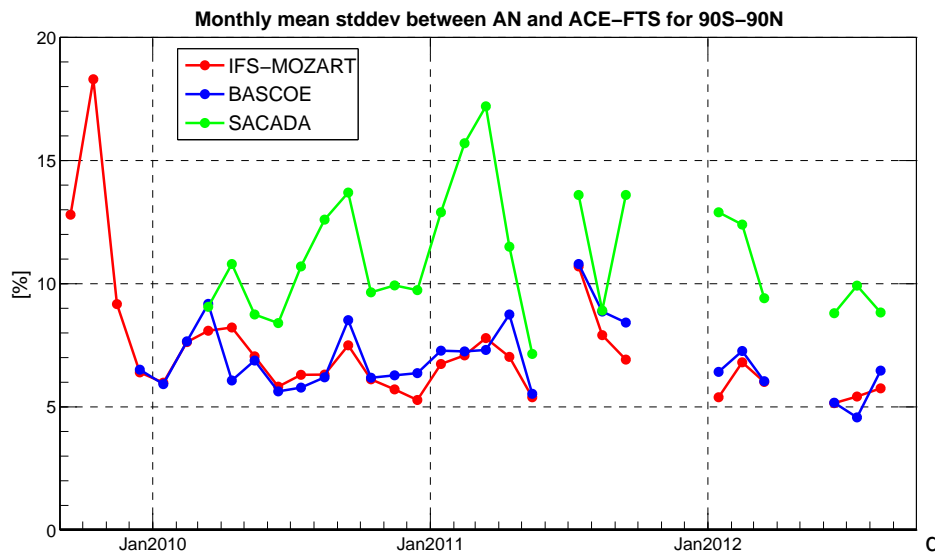


Fig. 6. Comparison of the global (i.e. from 90° S to 90° N) monthly mean standard deviation between IFS-MOZART (red), BASCOE (blue), and SACADA (green) with ACE-FTS (analysis minus observations) in %, for the [200,5]hPa pressure bin, for the period September 2009 to September 2012. Standard deviations for levels for which there are less than 20 observations are left out. Note that standard deviations are not weighted by the cosine of the latitude.

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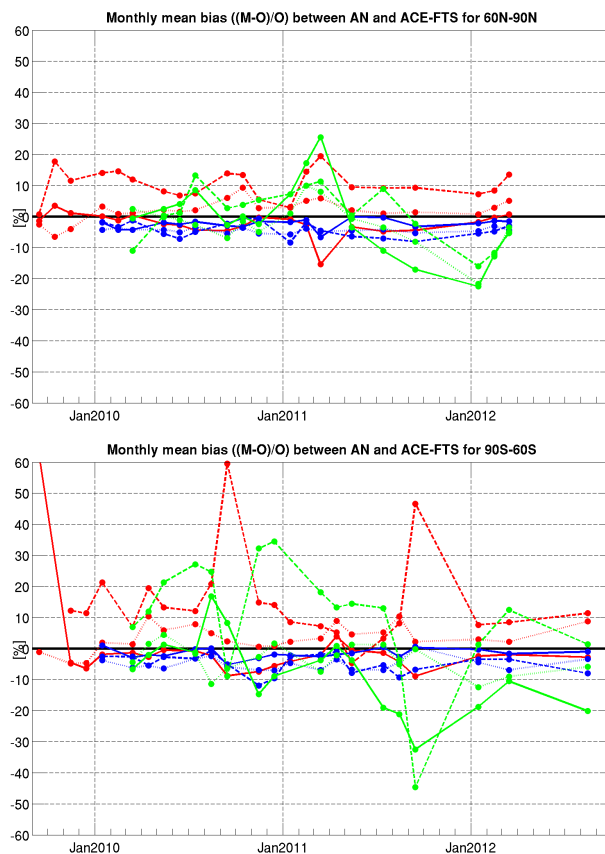



Fig. 7. Comparison of the monthly mean relative ozone biases between IFS-MOZART (red), BASCOE (blue), and SACADA (green) and ACE-FTS (analysis minus observations) in %, at 100 (dashed), 50 (full) and 10 hPa (dotted) for the period September 2009 to September 2012 for the Arctic (top) and the Antarctic (bottom).

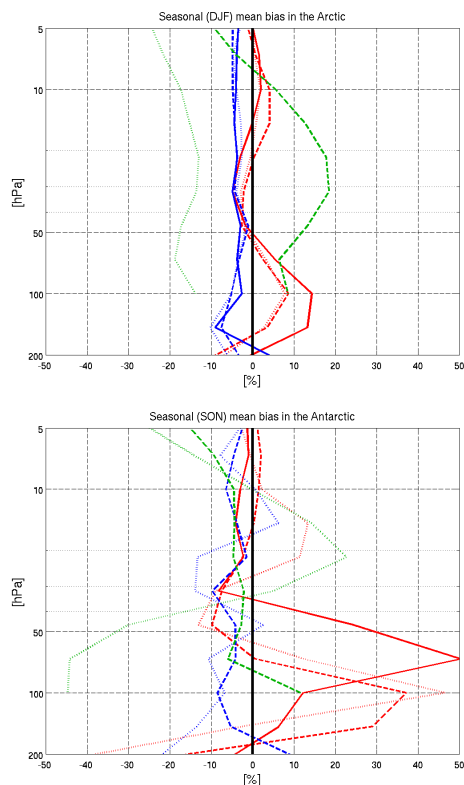


Fig. 8. Seasonally averaged relative ozone bias profiles of IFS-MOZART (red), BASCOE (blue), and SACADA (green) vs. ACE-FTS (analysis minus observations) in % for the Arctic winter (DJF i.e. months December, January, and February) 2009–2010 (full), 2010–2011 (dashed), 2011–2012 (dotted) and the Antarctic spring (SON i.e. months September, October, and November) 2009 (full), 2010 (dashed), and 2011 (dotted).

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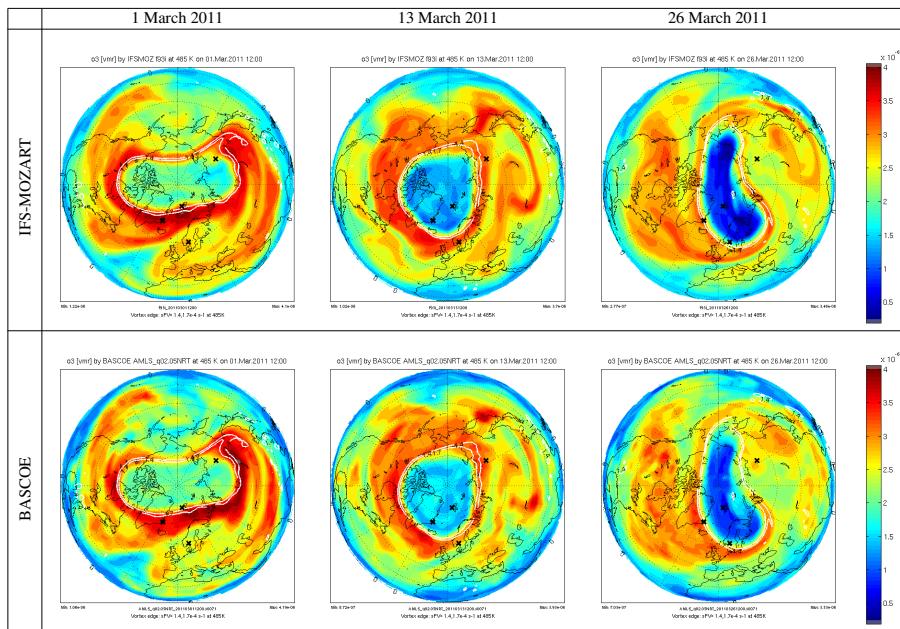


Fig. 9. Evolution of ozone volume mixing ratios at 485 K during March 2011 (left: 1 March, middle: 13 March, right: 26 March). Red/blue colours indicate respectively high/low ozone values. In white, the inner and outer polar vortex edges are indicated, calculated with an sPV of, respectively, $> 1.7 \times 10^{-4} \text{ s}^{-1}$ and $> 1.4 \times 10^{-4} \text{ s}^{-1}$. Top: IFS-MOZART, bottom: BASCOE. The location of three SAOZ stations used for the detailed total ozone column evaluation are indicated by black crosses: Zhigansk (Russia, 66.8° N , 123.4° E), Harestua (Norway, 60° N , 11° E), Scoresbysund (Greenland, 70.49° N , 21.98° W), as well as the location of the O₃ sonde at Ny-Ålesund (Spitzbergen, 78.933° N , 11.883° E).

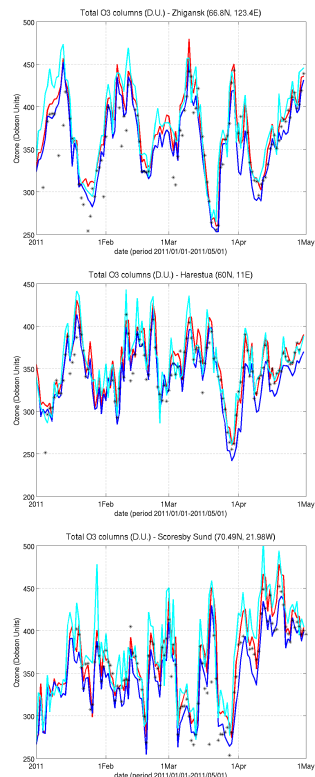


Fig. 10. Comparison of daily averaged total ozone columns (expressed in Dobson Units) for IFS-MOZART (red), BASCOE (blue), and TM3DAM (cyan) vs. ozone measurements from three SAOZ/DOAS stations: Zhigansk (66.8° N, 123.4° E, SAOZ), Harestua (60° N, 11° E, DOAS) and Scoresby Sund (70.49° N, 21.98° W, SAOZ).

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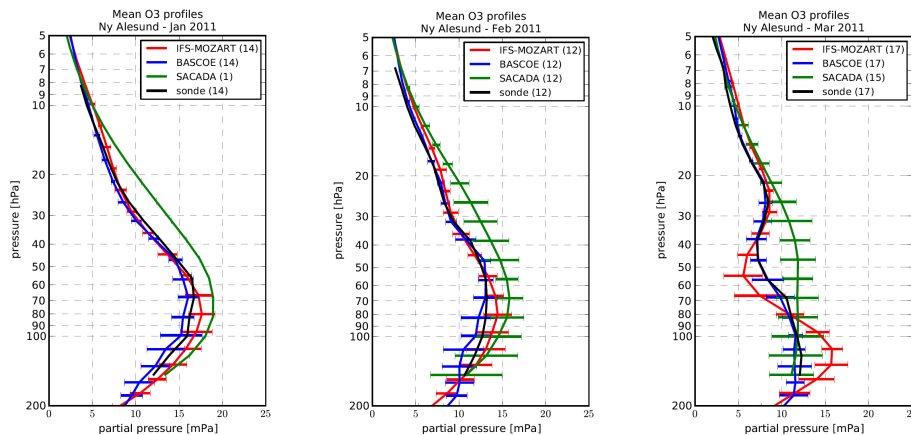



Fig. 11. Comparison of the monthly average O₃ profiles of IFS-MOZART (red), BASCOE (blue) and SACADA (green) with O₃ sonde profiles observed at Ny-Ålesund for January to April 2011. The number of available O₃ sonde profiles and the number of collocated system profiles are indicated in brackets.

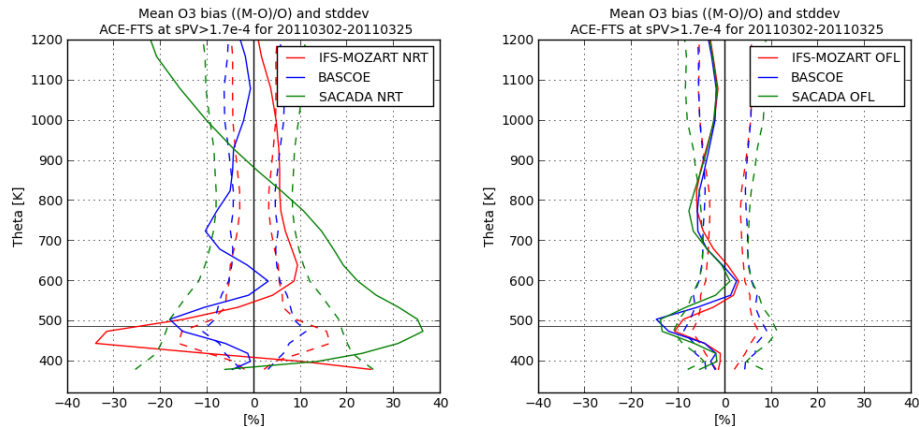


Fig. 12. Mean bias and standard deviations of the differences between the NRT analyses (left) and the offline experiments (right) of IFS-MOZART, BASCOE, and SACADA, on the one hand, and ACE-FTS observations, on the other hand, within the North Pole vortex (vortex edge calculated with an sPV of $> 1.7e^{-4} s^{-1}$) for March 2011. The ozone hole level used in Fig. 9 ($\theta \approx 485$ K) is indicated as the black horizontal line.