Atmos. Chem. Phys. Discuss., 7, 11621–11646, 2007 www.atmos-chem-phys-discuss.net/7/11621/2007/ © Author(s) 2007. This work is licensed under a Creative Commons License.



Summertime stratospheric processes at northern mid-latitudes: comparisons between MANTRA balloon measurements and the Canadian Middle Atmosphere Model

S. M. L. Melo^{1,2}, R. Blatherwick⁶, J. Davies⁴, P. Fogal², J. de Grandpré⁴, J. McConnell³, C. T. McElroy⁴, C. McLandress², F. J. Murcray⁶, J. R. Olson⁶, K. Semeniuk³, T. G. Shepherd², K. Strong², D. Tarasick⁴, and B. J. Williams-Rioux^{5,1}

¹Canadian Space Agency, St-Hubert, Quebec, Canada
 ²Department of Physics, University of Toronto, Ontario, Canada
 ³Department of Earth and Space Science and Engineering, York University, Ontario, Canada
 ⁴Environment Canada, Canada
 ⁵Department of Atmosphere and Ocean, McGill University, Montreal, Quebec, Canada
 ⁶Department of Physics and Astronomy, University of Denver, CO, USA
 Received: 2 July 2007 – Accepted: 3 August 2007 – Published: 8 August 2007

Correspondence to: S. M. L. Melo (stella.melo@space.gc.ca)

AC	ACPD	
7, 11621–11646, 2007		
Summertime ozone at northern mid-latitudes		
S. M. L. Melo et al.		
Title Page		
Abstract	Introduction	
Conclusions	References	
Tables	Figures	
14	►I	
•	•	
Back	Close	
Full Scr	Full Screen / Esc	
Printer-friendly Version		
Interactive Discussion		

Abstract

In this paper we report on a study conducted using the Middle Atmospheric Nitrogen TRend Assessment (MANTRA) balloon measurements of stratospheric constituents and temperature and the Canadian Middle Atmosphere Model (CMAM) in order to evaluate the ability of the model to reproduce the measured fields and to thereby test our ability to describe mid-latitude summertime stratospheric processes. The MANTRA measurements used here are vertical profiles of ozone, temperature, N₂O, CH₄, HNO₃, and HCI obtained during four campaigns, involving the launch of both ozonesondes and large balloons from Vanscoy, Saskatchewan, Canada (52° N, 107° W). The campaigns were conducted in August and September 1998, 2000, 2002 and 2004. During late summer at mid-latitudes, the stratosphere is close to photochemical control, providing an ideal scenario for the vertical diffusion coefficient in CMAM to a more physically reasonable value results in the model better reproducing the measured profiles of

¹⁵ long-lived species; (2) the existence of compact correlations among the constituents, as expected from independent measurements in the literature and from models, confirms the self-consistency of the MANTRA measurements; and (3) the 1998 ozone measurements show a narrow layer of low ozone centered near 25 km that is consistent with fossil debris from the polar vortex, suggesting that localized springtime ozone anomalies can persist through summer, affecting ozone levels at mid-latitudes.

1 Introduction

25

The science of stratospheric ozone gained a new dimension when it became clear that ozone depletion, and ultimately its recovery, is linked to climate change in a complex way. Temperature, humidity, winds, and the presence of other chemicals in the atmosphere influence ozone formation and transport, and the presence of ozone, in turn, affects those atmospheric fields through radiative processes. Although the existence

ACPD 7, 11621-11646, 2007 Summertime ozone at northern mid-latitudes S. M. L. Melo et al. **Title Page** Introduction Abstract Conclusions References Tables **Figures** ►T. Back Close Full Screen / Esc **Printer-friendly Version** Interactive Discussion

of such a link now seems intuitive, a detailed description of the processes and their relative importance remains a challenge. Understanding of the processes that control the ozone budget and the proper representation of such processes in atmospheric models are recognized in recent assessments (WMO 2006, 2005) as fundamental to forecasting ozone recovery and to further exploring the effects of ozone on climate and vice versa.

5

In contrast to the large ozone loss observed in the Antarctic spring and in the Arctic during cold winters, ozone depletion at mid-latitudes has been shown to be much smaller (3% to 6% per decade) (WMO 2006, 2005). However, it is also less well understood and more difficult to model. As first pointed out by Dobson in 1926, the ozone total column measured at different mid-latitude locations will differ depending on the synoptic meteorological conditions (Dobson et al., 1926). Therefore, small trends are difficult to detect and demand statistical analysis of long-period data sets. Similarly, the attribution of any such small trend is difficult.

- ¹⁵ Different mechanisms have been proposed to explain the observed mid-latitude ozone trends involving chemical (Solomon et al., 1998) and dynamical (Hadjinicolaou et al., 1997; Hood et al., 1997; Appenzeller et al., 2000) processes. So far, most of the work in the field focuses on a single mechanism, although it is currently accepted that a combination of mechanisms is needed to explain the observations.
- ²⁰ While chemical processes are local, dynamical processes normally involve links between mid and high latitudes. Processes such as changes in circulation and the transport of ozone-depleted air from polar latitudes have been proposed to explain the mid-latitude ozone deficit. However, the relative importance of each process remains unclear.
- ²⁵ Model experiments (Chipperfield and Jones, 1999; Millard et al., 2003) and measurements conducted in brief campaigns (Ross et al., 2004; Durry and Hauchecorne, 2005) support the hypothesis of vortex air contributing to the mid-latitude ozone deficit detected at summertime. Recently, Ross et al. (2004) and Durry and Hauchecorne (2005) report on the presence of long-lived remnants of the wintertime polar vortex in

ACPD 7, 11621-11646, 2007 Summertime ozone at northern mid-latitudes S. M. L. Melo et al. **Title Page** Introduction Abstract Conclusions References Tables **Figures** ►T. Back Close Full Screen / Esc **Printer-friendly Version** Interactive Discussion

the mid-latitude summer stratosphere in June 2000. Statistical analysis using zonally averaged total ozone data indicates that the observed summertime ozone trend over the Northern Hemisphere mid-latitudes can be entirely explained by the springtime trends there, as the photochemically damped remnant of the mid-latitude springtime trends (Fioletov and Shepherd, 2003). The importance of such results, as emphasized by the authors, is that it implies no need to invoke anomalous summertime ozone chemistry to explain summertime ozone trends. While the same relation between springtime and summertime mid-latitude trends does not hold in the Southern Hemisphere, Fioletov and Shepherd (2005) showed that the relation holds in both hemispheres when applied to the entire extra-tropics, but fails for southern mid-latitudes because of the

¹⁰ applied to the entire extra-tropics, but fails for southern mid-latitudes because of the contribution of springtime polar losses to summertime mid-latitude ozone trends, via transport. The extent of this contribution, estimated statistically, is consistent with that from detailed modelling studies.

In addition to the trends, transient decreases in column ozone levels over single stations at mid-latitudes have been observed during summertime and reported in several communications (Orsolini and Nikulin, 2006; Kar et al., 2002, and references therein). Those events have been referred to as low-ozone episodes, and although they have been associated with a conjunction of a deep tropospheric anticyclone and the displacement above the anticyclone of the mid-stratospheric pool of low-ozone column (Orsolini and Nikulin, 2006), the mechanism remains a subject of debate.

It is clear that the required level of understanding of the processes governing the midlatitude ozone balance can not be achieved by models or observations alone, but rather through a combination of both. Such a synthesis not only aids in the interpretation of the observations, but also helps identify model deficiencies and thereby leads to the improvement of the representation of physical processes in the models and hereby leads to the

improvement of the representation of physical processes in the models, and hence in their predictive capabilities.

In this paper we report on co-located measurements of ozone, temperature, and long-lived species made during four Middle Atmospheric Nitrogen TRend Assessment (MANTRA) balloon campaigns conducted in different years and compare them with

ACPD 7, 11621-11646, 2007 Summertime ozone at northern mid-latitudes S. M. L. Melo et al. **Title Page** Introduction Abstract Conclusions References Tables **Figures** ►T. Back Close

EGU

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

results from the Canadian Middle Atmosphere Model (CMAM), a coupled chemistry-climate model. Our objectives are: (1) to test the ability of CMAM to reproduce summertime mid-latitude conditions, (2) to investigate the validity of correlations among long-lived species as predicted by the model, and (3) to use CMAM as a support to
⁵ interpret the MANTRA measurements. The use of a chemistry-climate model, like CMAM, for studies involving comparisons with measurements has the advantage of allowing us to test our understanding of physical processes. MANTRA provides an ideal scenario for such a study since the experiments were conducted in late summer when the stratospheric zonal wind changes from easterly to westerly. During the summertime easterly period, planetary wave activity is minimal and the stratosphere is dynamically quiescent (e.g. Wunch et al., 2005), while photochemistry is dominant. Therefore, at turnaround, the stratosphere is close to photochemical control (Fahey et al., 2001; Fioletov and Shepherd, 2003).

2 Measurements

25

MANTRA is set of balloon campaigns aimed at investigating the changing chemical balance of the Northern Hemisphere mid-latitude stratosphere, with a particular focus on the nitrogen budget and its role in the depletion of mid-latitude ozone. A review of the MANTRA campaigns is provided by Strong et al. (2005). We provide here only a brief description of the campaigns and the instruments used to acquire the data used in this paper.

MANTRA campaigns have been conducted at Vanscoy, Saskatchewan (52° N, 107° W) during the period when the stratospheric zonal wind velocity changes its sign. Turnaround of the stratospheric winds occurs twice each year at mid-latitudes: in the early spring and in late summer (see Wunch et al., 2005, for more details). Under such conditions, the stratospheric winds are at a minimum, ensuring that the payload remains within the telemetry range (approximately 400 km) for the duration of the mission (typically 18 h). Four campaigns have been conducted so far, with a large balloon



launch dates of 24 August 1998, 29 August 2000, 3 September 2002, and 1 September 2004. Ozonesondes were also launched throughout each campaign.

The vertical profiles of temperature and ozone partial pressure discussed here were measured by the ozonesondes during all four campaigns, while the N_2O , CH_4 , HNO_3 ,

- ⁵ and HCI mixing ratio profiles were measured by the Denver University Fourier Transform Spectrometer (DU-FTS) during the 1998 campaign. For the analysis presented here we present the units in which the measurements were provided.
 - 2.1 Denver University Fourier Transform Spectrometer (DU-FTS)

This instrument, as well as the technique employed for the retrieval of each species, is
fully described in Fogal et al. (2005) and is only briefly described here. It has strong heritage since it took part in other balloon campaigns and has been used extensively as a ground-based instrument at many locations including Fairbanks, Alaska and the South Pole (for example see Murcray et al., 1980). DU-FTS is a BOMEM DA2 Michelson type interferometer-spectrometer with two-inch input optics. The optical path difference is 50 cm, resulting in an unapodized full-width half-maximum resolution of approximately 0.01 cm⁻¹. The scan time was about 80 s. The measurements extend from 700–1300 cm⁻¹ in one channel (with the mercury-cadmium-telluride detector) and from 2650–3250 cm⁻¹ in the other channel (indium antimonide detector). A biaxial solar tracking telescope was used to maintain the input solar beam on the interferometer

²⁰ window. During the 1998 MANTRA balloon flight, spectra were acquired during the sunset occultation.

The measured spectra were analyzed by fitting synthetic spectra generated with the Denver University line-by-line, layer-by-layer computer code RADCO (RADiation COde) (Blatherwick et al., 1989) to the observed spectra. Mixing ratio profiles were generated from the sunset spectra using the "onion peeling" technique. The pressure and temperature profiles used as input to RADCO were based on radiosonde data for the day of the flight, and the line parameters were taken from HITRAN96 (Rothman et al., 1998).



2.2 The ozonesondes

During all four MANTRA campaigns, ozone profiles were obtained on a nearly daily basis by electrochemical concentration cell (ECC) sensing devices carried on a standard balloon-borne meteorological radiosonde (Komhyr, 1969). Ozone partial pressure

is measured from the reaction of ozone with potassium iodide in an aqueous solution. The ECC's were prepared and the data analyzed according to the standards used in the Canadian ozonesonde network. In addition to the ozone sensor, each instrument package included a standard Vaisala RS-80 radiosonde so that pressure, temperature and humidity profiles could be recorded. The ozonesondes produce in situ ozone
 concentration with a vertical resolution of the order of 100 m. Estimation of errors for ozonesonde measurements is not a trivial exercise but inter-comparison campaigns and laboratory tests indicate in general a 5% maximum error in the measurement (Davies et al., 2000).

3 The Canadian Middle Atmosphere Model (CMAM)

- ¹⁵ CMAM is an upward extension of the Canadian Centre for Climate Modelling and Analysis General Circulation Model (CCCma GCM) up to 0.0006 hPa (roughly 100 km altitude) (Beagley et al., 1997). The model incorporates middle atmosphere radiation, interactive chemistry, gravity wave drag, as well as all the processes in the GCM. For the version used in this work (referred to as version 7) prognostic variables are computed
- ²⁰ in spectral space using T32 horizontal resolution (corresponding to about 6° latitude and longitude) and 65 vertical levels (about 2 km resolution in the middle atmosphere but higher resolution below 25 km).

CMAM includes a comprehensive representation of stratospheric chemistry (de Grandpré et al., 1997) with 31 non-advected species and 16 advected species and fam-

²⁵ ilies. Concentrations of the long-lived source gases are imposed in the troposphere. Transport of species is accomplished using spectral advection. Heterogeneous reac-

ACPD 7, 11621-11646, 2007 Summertime ozone at northern mid-latitudes S. M. L. Melo et al. **Title Page** Introduction Abstract Conclusions References **Tables Figures** ►T. Back Close Full Screen / Esc **Printer-friendly Version** Interactive Discussion

tions of CINO₃, N₂O₅ and BrNO₃ take place on sulphate aerosols. In the polar regions, heterogeneous reactions occur on stratospheric ternary solution, and water ice; no sedimentation or nitric acid trihydrate particles formation on polar stratospheric clouds is included. Profiles are generated for the model grid point closest to Vanscoy, and at various solar zenith angles.

In these simulations, climatological sea-surface temperatures are imposed which, although varying from month to month, are kept constant from year to year. Also, phenomena like the quasi-biennial oscillation and solar and aerosol variability, which are known to be significant factors affecting the inter-annual variability of the stratosphere, are not included in the model version used here. As a consequence, the inter-annual variability in the model as reported here is likely underestimated.

4 Results

10

4.1 Ozone and temperature

During the four MANTRA campaigns, a large number of ozone profiles were collected allowing calculation of average profiles that can be used for comparisons with CMAM. The measurements extend from 9 August to 11 September, being concentrated around 24 August, for which the largest number of profiles is available (four). Seven ozonesondes were launched in 1998, five in 2000, 12 in 2002, and 23 in 2004, making a total of 47 ozone and temperature profiles available for this analysis. Figure 1 shows a distribution of the measurements as a function of the day for all the campaigns.

- Measurements and model are compared qualitatively in Fig. 2, in scatter plots using all the ozonesonde measurement and the matching model days. Different altitude ranges are shown in different colors. A model day is calculated as the daily average of the model profiles for ozone and temperature, for each year of the model run. Thus,
- ²⁵ for each measurement point there are at least 20 corresponding model points (one for each model year). The dotted line represents the 1:1 correlation. From Fig. 2 we



see that the points are generally distributed around the 1:1 correlation line indicating a good overall consistency between model and measurements. There is an interesting tendency on the temperature plot shown in Fig. 2 where points corresponding to altitudes above 20 km tend to be consistently located below the 1:1 correlation line. It is

important to stress here that we use only four years of measurements, which may not be representative of climatology. Data below 10 km altitude are not used in this analysis since the model version used here does not include full tropospheric chemistry.

The relative differences between the model and the measurements ([CMAM-MANTRA]/MANTRA) are quantified and shown in Fig. 3 as histograms. The histograms

on the left side of Fig. 3 are constructed using the full altitude range. Relative deviations are calculated by linear interpolation of the ozonesondes onto the CMAM altitude grid. The statistical analyses of all the points (the number of points used is shown in each plot) show that the general agreement between CMAM and the ozone measurements is on the order of 3%, with a standard deviation of the relative difference between model and measurements of 18%. For temperature, the model and the measurements agree remarkably well, with mean difference being 0.1% and the standard deviation on the

order of 0.5%.
As for Fig. 2, the histograms are also constructed for different altitude ranges and shown in the right side of Fig. 3. Between 10 and 20 km the model reproduces the
ozone measurements within 5%, with a standard deviation of 18%. Above 20 km, the model ozone reproduces the measurements very well (within 0.7% on average). For altitudes above 10 km, the model reproduces the measured temperature within 0.15% (less than 0.5 K difference). Note that this analysis is limited to 30 km, which is the maximum altitude common to all the measurements.

²⁵ The model and measurement profiles are compared as averages in Fig. 4. For this comparison the measurements are averaged for each campaign year and compared to the model averaged profile. Furthermore, the average profiles for each year are further averaged to build what we call the averaged sonde profiles in Fig. 4, referred to as AV_sondes in the figure. The model ozone and temperature fields are first daily

ACPD 7, 11621-11646, 2007 Summertime ozone at northern mid-latitudes S. M. L. Melo et al. **Title Page** Introduction Abstract Conclusions References **Tables Figures** ►T. Back Close Full Screen / Esc

Printer-friendly Version

Interactive Discussion

averaged, as before, and further averaged for the period of 11 August to 11 September to encompass the period for which the measurements are available. Then, the 20 resulting averaged model profiles, one for each model year, are averaged again, building what we refer to as CMAM in Fig. 4. Ozone is shown as partial pressure since this is the standard ozone product. Differences between the measurement and model (in

percent for ozone and in Kelvin for temperature) as a function of altitude are quantified in the right panel in Fig. 4.

As was the case for Fig. 2, Fig. 4 shows different results for different altitude ranges. For ozone, at around 10 km the model reproduces the measurements very well. Above

- 10 10 km, differences between the model and measurements increase, reaching a maximum at around 15 km, with the model underestimating the measurements by 20%. The agreement again becomes very good, when the average of all the measurements is considered, for altitudes above about 20 km. However, the differences observed here between model and measurements are within the measurements variability, as can be seen in Fig. 4, and we can conclude that generally CMAM reproduces the MANTRA
- 15 seen in Fig. 4, and we can conclude that generally CMAM reproduces the MANTRA ozone measurements reasonably well.

One interesting feature in Fig. 4 is the measurement of a significant depletion of about 35% of ozone at the peak altitude during the 1998 MANTRA campaign. While such a feature would be rather common in individual profiles, its persistence throughout the campaign is only evident for 1998. This feature is further discussed in the next

²⁰ the campaign is only evident for 1998. This feature is further discussed in the nex section.

The averaged temperature profile agrees with the model within 2 K, except for altitudes below 10 km where the difference may reach 3 K if we take the average of the sondes for 1998. As for ozone, the differences between model and measurements be-

²⁵ low about 20 km are within the measurement variability. The shallow layer of enhanced static stability observed in Fig. 4 just above the tropopause, seen in both the sondes and the model, is the "tropopause inversion layer" identified by Birner et al. (2006), which is strongest in summer and which is a ubiquitous feature in CMAM (albeit somewhat too high and somewhat too deep). For altitudes above about 20 km, the model



Interactive Discussion

results are consistently about 2 K warmer than the measurements.

4.2 Long-lived species

During the 1998 campaign, the balloon-borne DU-FTS measured vertical profiles of N_2O , CH_4 , HCI, and HNO₃ during sunset. We compare here the measured profiles s with CMAM fields as shown in Fig. 5. As for ozone and temperature, CMAM fields shown here represent a 20-year average for the period of 11 August to 11 September. The average profile of each species is calculated by taking the daily average of the profiles, then further averaging the resulting daily profiles for the period of 11 August to 11 September and finally making the average of the 20 years. The error bars are the standard deviation of the 20 daily and then monthly (11 August to 11 September) 10 averaged profiles and is used here as representing the model variability. The differences between measurement and model (curves labeled CMAM-V7) are quantified in percent ([CMAM-MANTRA]/MANTRA) as a function of altitude, for each species and shown in the right panels of Fig. 5. Note that the tropospheric value of N₂O in CMAM was inadvertently set about 4% too high, so all CMAM nitrogen fields can be expected to also be 4% too high. When that difference is accounted for the CMAM N₂O profile agrees very well with the MANTRA observations.

It is instructive to compare this version of CMAM with a previous version (referred as CMAM – WMO in Fig. 5) which had a much higher value of the vertical diffusivity (and slightly reduced vertical resolution), namely Kzz=1.0 m² s⁻¹ rather than the current Kzz=0.1 m² s⁻¹. The smaller value is much more realistic, as discussed below, whereas the larger value is so strong as to play a first-order role in vertical transport, especially in summer when the Brewer-Dobson downwelling is so weak (Shepherd, 2007). The results shown in Fig. 5 suggest that such a change does indeed have a first-order effect on the tracer profiles, and that once the vertical diffusion is reduced to a sufficiently small value, the model agrees well with the measurements.

An exception is the HCl, with the model reproducing the measurements very well at lower altitudes but overestimating the measurements by a factor of two for altitudes



above about 20 km. From the model side, the concentrations of CFC-11 and 12 are scaled in the CMAM to take into account the production of chlorine compounds from other sources which are necessary to maintain the background level of chlorine compounds near the current level. However, the level of introduction of Cly from freons can

⁵ have a significant impact on the vertical distribution of Cly and thus affect the distribution of Cl species such as HCl which may explain some of the discrepancy obtained in the lower stratosphere.

From the measurement side, since MANTRA produced one profile, while CMAM is a climatology, it is instructive to look also at other available datasets. We choose to use HALOE dataset since it is the largest dataset available and has been extensively used in many other analyses. The HALOE climatology used here consists of a monthly mean HALOE data set constructed by harmonic regression of HALOE (v18) profile data representative of the entire HALOE record (repeating seasonal cycles were fit through a 6+ year record) (http://www.sp.ph.ic.ac.uk/haloe/userguide/uguide.html). As

- ¹⁵ can be seen in Fig. 5, both MANTRA and model HCl profiles agree well with HALOE for altitudes below 22 km. However, HALOE HCl values, although being higher than MANTRA, are still lower than CMAM values for altitudes above 22 km. Compared to HALOE, CMAM overestimates HCl above 20 km by about 40%. We also compared CH₄ (not shown here) from HALOE with both MANTRA and CMAM and found a very
- good agreement through the whole altitude range with MANTRA and CMAM values being consistent with HALOE. Since we have only one measurement profile for HCI, and there is no real coincidence as for time and location within HALOE measurements (we are using climatological profiles) it is difficult to further explore the differences in the HCI profile observed here. An analysis of satellite data that includes a suite of chlorine
- species, like the measurements by the ACE-FTS instrument on board the Canadian satellite SCISAT, could be useful but is beyond the scope of this work. Therefore, although we do not have elements to support the hypothesis that the depletion of HCI observed in Fig. 5 has geophysical origin, we report it here, given that we do not expect that such feature can be explained by uncertainties in the measurements alone.

ACPD

7, 11621–11646, 2007

Summertime ozone at northern mid-latitudes



4.3 Compact correlations

One further effort for comparing model and measurements is through correlations among long-lived species. Correlation plots between long-lived species have been used in the literature as a way of eliminating, in large part, dynamical effects by assum-

- ing different long-lived species are advected in a similar manner. Indeed the existence of a compact correlation between long-lived species has been supported both theoretically (see for example Avallone and Prather, 1997, and references therein) and experimentally (see for example Loewenstein et al., 1993; Michelsen et al., 1998).
- Recently Sankey and Shepherd (2003) have used CMAM to investigate the general conditions under which compact correlations can be expected to form. One important result of this analysis was to show that a correlation between long-lived species can be characterized even from measurements with a limited sampling. Therefore, we use correlation plots to assess internal consistency of the MANTRA measurements. Furthermore, MANTRA measurements can be compared with other dataset through correlations. We use the ATMOS measurements for mid-latitudes given the availability
 - of data for the specie of interest.

Figure 6 shows the $CH_4:N_2O$ correlation plot using MANTRA and ATMOS measurements and CMAM output. Below 25 km (i.e., where $CH_4>1$ ppm), the MANTRA measurements indeed show a linear correlation in agreement with ATMOS measurements

- and with CMAM. However, above 25 km (i.e., where $CH_4 < 1$ ppm) the slope of the correlation seems to change. Taken together with Fig. 5, the change in slope above about 25 km suggests that the MANTRA observed air mass is somehow poor in CH_4 above about 25 km. As shown before in Sankey and Shepherd (2003), the linear correlation obtained from ATMOS measurements is well reproduced by CMAM. Note that a linear
- ²⁵ correlation should not be much affected by the change in vertical diffusivity.

ACPD

7, 11621–11646, 2007

Summertime ozone at northern mid-latitudes



5 Discussion

The comparison between MANTRA measurements and CMAM shown here during the late summer when the mid-latitude stratosphere is comparatively quiescent and close to photochemical control, suggests that vertical profiles of long-lived species in the

- ⁵ model are much improved when the vertical diffusivity is reduced from $1.0 \text{ m}^2 \text{ s}^{-1}$ to $0.1 \text{ m}^2 \text{ s}^{-1}$. As discussed by Shepherd (2007), the lower value is more realistic (being consistent with the upper bound inferred from aircraft measurements of trace species) and sufficiently small to have a negligible impact on mean tracer distributions. In that sense, the mean tracer distributions are independent of the vertical diffusivity provided
- ¹⁰ the diffusivity is sufficiently small. On the other hand, the higher value of $1.0 \text{ m}^2 \text{ s}^{-1}$ is sufficiently large to have a significant effect on the mean tracer distributions. Indeed, Tegtmeier and Shepherd (2007) found that the summertime persistence of ozone anomalies, which is quite realistic in CMAM with a vertical diffusivity of $0.1 \text{ m}^2 \text{ s}^{-1}$, is rapidly lost for a diffusivity of $1.0 \text{ m}^2 \text{ s}^{-1}$.
- ¹⁵ During 1998, the measured ozone profiles show a narrow layer of enhanced ozone at about 12 km. This lower-altitude enhanced ozone layer is also seen in the average 2000 campaign profile but is not evident in 2002 or 2004. However, looking at individual profiles we see the occurrence of layers of enhanced ozone at altitudes below 15 km is rather common, with their frequency of occurrence and persistence changing from year to year. Those layers of enhanced ozone coincide with the temperature "tropopause"
- inversion layer" identified by Birner et al. (2006) and shown to be stronger in summer. Although CMAM reproduces this feature in temperature, it is not evident in ozone model data.

While the occurrence of filament structures in the ozone profile for altitudes below about 15 km seems to be rather common and in general associated with dynamics (Tarasick et al., 2005), the origin of layers of depleted ozone above 20 km, as observed in the 1998 dataset used here, seems to be less clear. Kar et al. (2002) conducted a statistical analysis of the frequency of occurrence of layered structures in the ver-

ACPD

7, 11621–11646, 2007

Summertime ozone at northern mid-latitudes



tical profiles of ozone as retrieved by the Stratospheric Aerosol and Gas Experiment (SAGE II) version 6.0 measurements. Their results show a bimodal peak in the occurrence of such layers with two maxima: one between 12 and 15 km and another between 22 and 28 km. Their analysis suggests a maximum probability of occurrence

- ⁵ of structures in the ozone profile above 20 km altitude for latitudes above 45° N during summer time. However, a caveat in their analysis is that it makes no distinction between occurrences of enhancement or depletion layers. No identification is made as for the source of those structures in ozone although the authors suggest the structures observed above 20 km altitude could be consistent with remanent air from the
- polar vortex "frozen-in" into the mean summertime easterly flow as proposed by Hess and Holton (1985), and Orsolini (2001), and supported by the analysis by Fairlier et al. (1999). Recently Durry and Hauchecorne (2005) presented evidence for long-lived polar vortex air in the mid-latitude summer stratosphere from in situ laser diode CH₄ and H₂O measurements at Gap, in southern France (44° N, 6° E) during June 2000.
 Indeed the photochemical lifetime of ozone at altitudes from 20 to 28 km is of the order
 - of 100 days or more.

The observed layer of depleted ozone at around 25 km altitude during 1998 campaign persists in the averaged profile, therefore lasting for at least one week. However, such a persistent feature is not observed during the other years. This result suggests

- that if mixing of air from the polar vortex can generate layers of depleted ozone above 20 km altitude that can persist throughout the summer, impacting the estimation of the mid-latitude ozone trend, its importance varies from year to year. Indeed, Millard et al. (2003) found that the mixing between the pole and middle latitudes varies with the meteorological conditions. During the winter of 1997/98 temperatures dropped period-
- ically below the threshold for polar stratospheric cloud formation and polar vortex was relatively disturbed with several minor warmings. The behavior of ozone and the Arctic vortex during winter 1997/98 is described in details by Langer et al. (1999). They found that the vortex was well established in mid-November 1997, weakened until the end of January 1998, when it reformed and remained strong until March. Temperatures in



February/March 1998 were considerably higher than the same period in 1997. They found evidence for stronger ozone depletion occurring in February 1998 compared to February 1997, indicating that strong chlorine activation must have been present. However, the vortex breakdown occurred near the end of March.

⁵ Finally, although the layer of depleted ozone reported here is consistent with the hypothesis of long lived "fossil" debris from the polar vortex that can persists till later summer, as modeled by Orsolini (2001), a more in-depth analysis using a climate-transport model would be necessary to confirm it.

In summary, we conclude that CMAM has achieved a level of development in which it can reproduce reasonably well not only ozone measurements but a suite of atmospheric constituents and temperature, such as those reported here.

Acknowledgements. All MANTRA campaigns have been supported by the Canadian Space Agency and Environment Canada. MANTRA 1998 also received funding from the Centre for Research in Earth and Space Technology, while MANTRA 2002 and 2004 received additional

- ¹⁵ support from the Natural Sciences and Engineering Research Council. The participation of the University of Denver was supported in part by the National Science Foundation and in part by the National Aeronautics and Space Administration. We thank Scientific Instrumentation Limited for payload and launch support, and all team members who contributed to the success of the MANTRA campaigns. The CMAM is funded by the Canadian Foundation for Climate and
- ²⁰ Atmospheric Sciences, the Canadian Space Agency, and the Natural Sciences and Engineering Research Council.

References

- Appenzeller, C., Weiss, A. K., and Stachclin, J.: North Atlantic Oscillation modulates total ozone trends, Geophys. Res. Lett., 27, 1131–1134, 2000.
- Avallone, L. M. and Prather, M. J.: Tracer-tracer correlations: Three-dimentional model simulations and xomparisons to observations, J. Geophys. Res., 102(D15), 19233–19246, 1997.
 Beagley, S. R., Grandpré, J. D., Koshyk, J. N., McFarlane, N. A., and Shepherd, T. G.:



Printer-friendly Version

Radiative-dynamical climatology of the first-generation Canadian Middle Atmosphere Model, Atmos.-Ocean, 35, 293–331, 1997.

- Birner, T., Sankey, D., and Shepherd, T. G.: The troposphere inversion layer in models and analyses, Geophys. Res. Lett., 33, L14809, doi:10.1029/2006GRL026599, 2006.
- ⁵ Blatherwick, R. D., Murcray, D. G., Murcray, F. H., Murcray, F. J., Goldman, A., Vanasse, G. A., Massie, S. T., and Cicerone, R. J.: Infrared emission measurements of morning N2O5, J. Geophys. Res., 94, 18337–18340, 1989.

Chipperfield, M. P. and Jones, R. L.: Relative influence of atmospheric chemistry and transport on arctic ozone trends, Nature, 400, 551–554, 1999.

- Davies, J., Tarasick, D., McElroy, C. T., Kerr, J. B., Fogal, P. F., and Savastiouk, V.: Evaluation of ECC Ozonesonde Preparation Methods from Laboratory Tests and Field Comparisons using MANTRA. Proc. Quadrennial Ozone Symposium, Sapporo, Japan, 2000.
 - de Grandpre, J., Sandilands, J. W., McConnell, J. C., Beagley, S. R., Croteau, P. C., and Danilin,
 M. Y.: Canadian middle atmosphere model: preliminary results from the chemical transport module, Atmos.-Ocean, 35(4), 385–431, 1997.
- Dobson, G. M. B., Harrison, D. N., and Lawrence, J.: Measurements of the amount of ozone in the Earth's atmosphere and its relation to other geophysical conditions, Proc. R. Soc. London, Ser. A, 110, 660–693, 1926.

Durry, G. and Hauchecorne, A.: Evidence for long-lived polar vortex air in the mid-latitude

summer stratophere from in situ laser diode CH_4 and H_2O measurements, Atmos. Chem. Phys., 5, 1467–1472, 2005,

http://www.atmos-chem-phys.net/5/1467/2005/.

15

- Fahey, D. W., Gao, R. S., Carslaw, K. S., et al.: The detection of large HNO3 particles in the winter Arctic stratosphere, Science, 291, 1026–1031, 2001.
- Fairline, T. D., Profitt, M. H., and Webster, C. R.: The contribution of mixing in Lagrangian photochemical predictions of polar ozone loss over the Arctic in summer 1997, J. Geophys. Res., 104, 26 597–26 609, 1999.
 - Fioletov, V. and Shepherd, T. G.: Summertime total ozone variations over middle and polar latitudes, Geophys. Res. Lett., 32, L01207, doi:10.1029/2004GL022080, 2005.
- ³⁰ Fioletov, V. E. and Shepherd, T. G.: Seasonal persistence of midlatitude total ozone anomalies, Geophys. Res. Lett., 30(7), 1417, doi:10.1029/2002GL016739, 2003.
 - Fogal, P. F., Blatherwick, R. D., Murcray, F. J., and Olson, J. R.: Infra-red FTS Measurements of CH₄, N₂O, O₃, HNO₃, HCl, CFC-11 and CFC-12 from the MANTRA Balloon Campaign,

7, 11621–11646, 2007

Summertime ozone at northern mid-latitudes

S. M. L. Melo et al.

Title Page		
Abstract	Introduction	
Conclusions	References	
Tables	Figures	
14	►I	
Back	Close	
Full Screen / Esc		
Printer-friendly Version		
Interactive Discussion		

Atmos.-Ocean, 43(4), 351-359, 2005.

5

10

15

30

Hadjinicolaou, P., Pyle, A., Chipperfield, P. M., and Kettleborough, J. A.: Effect of interannual meteorological variability on mid-latitude O3, Geophys. Res. Lett., 24, 2993–2996, 1997.

- Hess, P. G. and Holton, J. R.: The origin of temporal variance in long-lived trace constituents in the summer stratosphere, J. Atmos. Sci., 42, 1455–1463, 1985.
- Hood, L. L., McCormack, J. P., and Labitzke, K.: An investigation of dynamical contributions to midlatitude ozone trends in winter, J. Geophys. Res., 102, 13 079–13 093, 1997.
- Kar, J., Trepe, C. R., Thomason, L. W., and Zawodny, J. M.: Observations of layers of ozone vertical profiles from SAGEII (v6.0) measurements, Geophys. Res. Lett., 29(10), 1443–1446, 2002.
- Komhyr, W. D.: Electrochemical concentration cells for gas analysis, Ann. Géophys., 25, 203–210, 1969.
- Langer, J., Barry, B., Klein, U., Sinnhuber, B.-M., Wohltmann, I., and Kunzi, K. F.: Chemical ozone depletion during Arctic winter 1997/98 derived from ground based millimeter-wave observations, Geophys. Res. Lett., 26(5), 599–602, 1999.
- Loewenstein, M., Podolske, J. R., Fahey, D. W., Woodbridge, E. L., Tin, P., Weaves, A., Newman, P. A., Strahan, S. E., Kawa, S. R., Schoeberl, M. R., and Lait, L. R.: New observations of the NOy/N2O correlation in the lower stratosphere, Geophys. Res. Lett., 20(22), 2531– 2534, 1993.
- ²⁰ Michelsen, H. A., Manney, G. L., and Gunson, M. R.: Correlations of stratospheric abundances of NO_y, O₃, N₂O, and CH₄ derived from ATMOS measurements, J. Geophys. Res, 103(D21), 28 347–28 359, 1998.
- Millard, G. A., Lee, A. M., and Pyle, J. A.: A model study of the connection between polar and midlatitude ozone loss in the Northern Hemisphere lower stratosphere, J. Geophys. Res., 108(D5), 8323, doi:10.1029/2001JD000899, 2003.
 - Murcray, F. J., Goldman, A., Murcray, D. G., Cook, G. R., van-Allen, J. W., and Blatherwick, R. D.: Identification of isolated NO lines in balloonborne infrared solar spectra., Geophys. Res. Lett., 7, 673–676, 1980.
 - Orsolini, Y.: Long-lived tracer patterns in the summer polar stratosphere, Geophys. Res. Lett., 28. 3855–3858. 2001.
 - Orsolini, Y. and Nikulin, G.: A low-ozone episode during the European heatwave of August 2003, Q. J. Roy. Meteor. Soc., 132, 667–680, 2006.

Ross, D. E. M., Pyle, J. A., Harris, N. R. P., McIntyre, J. D., Millard, G. A., Robinson, A. D.,

7, 11621–11646, 2007

Summertime ozone at northern mid-latitudes

Title Page		
Abstract	Introduction	
Conclusions	References	
Tables	Figures	
14	P1	
•	•	
Back	Close	
Full Screen / Esc		
Printer-friendly Version		
Interactive Discussion		
FGU		

and Busen, R.: Investigation of Arctic ozone depletion sampled over midlatitudes during the Egrett Campaign of spring/summer 2000, Atmos. Chem. Phys., 4, 141–168, 2004, http://www.atmos-chem-phys.net/4/141/2004/.

Rothman, L. S., Rinsland, C. P., Goldman, A., Massie, S. T., Edwards, D. P., Flaud, J.-M.,

- Perrin, A., Camy-Peyret, C., Dana, V., Mandin, J.-Y., Chroeder, J., McCann, A., Gamache, R. R., Wattson, R. B., Yoshino, K., Chance, K. V., Jucks, W., Brown, L. R., Nemtchinov, V., and Varanasi, P.: The HITRAN molecular spectroscopic database and HAWKS (HITRAN Atmospheric Workstation): 1996 edition, J. Quant. Spectrosc. Radiat. Transfer, 60, 665–710, 1998.
- ¹⁰ Sankey, D. and Shepherd, T. G.: Correlation of long-lived chemical species in a middle atmosphere general circulation model, J. Geophys. Res, 108(D16), 4494, doi:10.1029/2002JD002799, 2003.

Shepherd, T. G.: Transport in the middle atmosphere, J. Meteorol. Soc. Japan, in press, 2007. Solomon, S., Portmann, R. W., Garcia, R. R., Randel, W., Wu, F., Nagatani, R., Gleason, J.,

- ¹⁵ Thomason, L. W., Poole, L. R., and McCormick, M. P.: Ozone depletion at midlatitudes: coupling of volcanic aerosols and temperatute variability to anthropogenic chlorine., Geophys. Res. Lett., 25, 1871–1874, 1998.
 - Strong, K., Bailak, G., Barton, D., Bassford, M. R., Blatheiwick, R. D., Brown, S., Chartrand, D., Davies, J., Drummond, J. R., Fogal, P. F., Forsberg, E., Hall, R., Jofre, A., Kaminski,
- J., Kosters, J., Laurin, C., McConnell, J. C., McElroy, C. T., McLinden, C. A., Melo, S. M. L., Menzies, K., Midwinter, C., Murcray, F. J., Nowlan, C., Olson, R. J., Quine, B. M., Rochon, Y., Savastiouk, V., Solheim, B., Sommerfeldt, D., Ullberg, A., Werchohlad, S., Wu, H., and Wunch, D.: MANTRA a balloon mission to study the odd-nitrogen budget of the stratosphere., Atmos. Ocean, 43(4), 283–299, 2005.
- Tarasick, D. W., Fioletov, V. E., Wardle, D. I., Kerr, J. B., and Davies, J.: Changes in the vertical distribution of ozone over Canada from ozonesondes: 1980–2001, J. Geophys. Res., 110, D02307, doi:10.1029/2004JD009693, 2005.

Tegtmeier, S. and Shepherd, T. G.: Persistence and photochemical dacay of springtime total ozone anolamies in the Canadian Middle Atmosphere Model, Atmos. Chem. Phys., 7, 485–

30 493, 2007,

http://www.atmos-chem-phys.net/7/485/2007/.

WMO 2006: Ozone Assessment, IPCC Special Report on Safeguarding the Ozone Layer, 2005.

ACPD		
7, 11621–11646, 2007		
Summertime ozone at northern mid-latitudes S. M. L. Melo et al.		
Title Page		
Abstract	Introduction	
Conclusions	References	
Tables	Figures	
I	۰	
•	F	
Back	Close	
Full Screen / Esc		
Printer-friendly Version		

Wunch, D., Tingley, M. P., Shepherd, T. G., Drummond, J. R., Moore, G. W. K., and Strong, K.: Climatology and predictability of the late summer stratospheric zonal wind turnaround over Vanscoy, Saskatchewan, Atmos.-Ocean, 43(4), 301–313, 2005.

ACPD 7, 11621–11646, 2007 Summertime ozone

at northern mid-latitudes





Fig. 1. Days on which the ozonesondes were launched, for each campaign year. Also shown is the sum of the number of ozonesondes profiles for each day, considering all four campaigns.





Fig. 2. Scatter plots for ozone and temperature, comparing MANTRA measurements with CMAM data for all four campaigns. CMAM data are daily averages (see text). The different colors correspond to ozone and temperature values in different altitude ranges: blue for altitudes between 10 and 20 km and green for altitudes above 20 km.

ACPD

7, 11621–11646, 2007

Summertime ozone at northern mid-latitudes





Fig. 3. Histogram of the relative deviation between CMAM and the sonde measurements for ozone (upper panels) and temperature (lower panels). For the plots on the left side (dark blue) all the available sonde measurements are used (from all four campaigns) while the plots on the right side are made using selected altitude ranges (20>z>10 km in blue and z>20 km in green). The model data is daily averaged, and for each model year run, the days matching the measurements are used. The points are grouped and counted for occurrence in bins of 1% relative deviation ((CMAM – MANTRA)/MANTRA) for ozone and 0.1% for temperature. For each histogram shown, "n" represents the number of points used.

ACPD

7, 11621–11646, 2007

Summertime ozone at northern mid-latitudes





Fig. 4. Ozone (upper panels) and temperature (bottom panels) sonde measurements during the four MANTRA campaigns. Also shown is CMAM climatology for August–September covering the measurement period (see text). The differences between each yearly averaged profile (measurement) to the model average for all campaigns (CMAM), for ozone (in percent: ((CMAM-MANTRA)×100/MANTRA) and for temperature (in Kelvin: CMAM-MANTRA), are shown on the right-hand panels, respectively.

ACPD

7, 11621–11646, 2007

Summertime ozone at northern mid-latitudes

S. M. L. Melo et al.





Fig. 5. The left panels show the vertical profiles of N₂O, CH₄, HCl, and HNO₃ measured with the DU-FTS during sunset as part of the MANTRA 1998 campaign (pink diamonds). The blue diamonds represents CMAM (V7) climatological 20 year run output averaged for August over Vanscoy, Saskatoon. The error bar in the model output represents the model variability (see text). The black lines represents the CMAM (WMO) climatological 20 years run also for Vanscoy, Au-The green symbols in aust. HCI plot represent HALOE climatological values for August at MANTRA coordinates. The right panels show the differences in percent (100.(CMAM (V7)-MANTRA)/MANTRA) for each constituent.

ACPD

7, 11621–11646, 2007

Summertime ozone at northern mid-latitudes

S. M. L. Melo et al.





Fig. 6. Correlation between CH_4 and N_2O mixing ratios as measured during the MANTRA campaign of 1998 compared with ATMOS measurements for mid-latitudes and CMAM.

