
MANTRA – A Balloon Mission to Study the Odd-Nitrogen Budget of the Stratosphere

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ABSTRACT *The Middle Atmosphere Nitrogen TRend Assessment (MANTRA) series of high-altitude balloon flights is being undertaken to investigate changes in the concentrations of northern hemisphere mid-latitude stratospheric ozone, and of nitrogen and chlorine compounds that play a role in ozone chemistry. Four campaigns have been carried out to date, all from Vanscoy, Saskatchewan, Canada (52°01'N, 107°02'W, 511.0 m). The first MANTRA mission took place in August 1998, with the balloon flight on 24 August 1998 being the first Canadian launch of a large high-altitude balloon in about fifteen years. The balloon carried a payload of instruments to measure atmospheric composition, and made measurements from a float altitude of 32–38 km for one day. Three of these instruments had been flown on the Stratoprobe flights of the Atmospheric Environment Service (now the Meteorological Service of Canada) in the 1970s and early 1980s, providing a link to historical data predating the onset of mid-latitude ozone loss.*

The primary measurements obtained from the balloon-borne instruments were vertical profiles of ozone, NO₂, HNO₃, HCl, CFC-11, CFC-12, N₂O, CH₄, temperature, and aerosol backscatter. Total column measurements of ozone, NO₂, SO₂, and aerosol optical depth were made by three ground-based spectrometers deployed during the campaign. Regular ozonesonde and radiosonde launches were also conducted during the two weeks prior to the main launch in order to characterize the local atmospheric conditions (winds, pressure, temperature, humidity) in the vicinity of the primary balloon flight. The data have been compared with the Model for Evaluating oZONE Trends (MEZON) chemical transport model, the University of California at Irvine photochemical box model, and the Canadian Middle Atmosphere Model (CMAM) to test our current understanding of model photochemistry and mid-latitude species correlations. This paper provides an overview of the MANTRA 1998 mission, and serves as an introduction to the accompanying papers in this issue of Atmosphere-Ocean that describe specific aspects and results of this campaign.

RÉSUMÉ *[Traduit par la rédaction] On a entrepris la série de lancements de ballons de haute altitude pour l'Évaluation des tendances de l'azote dans l'atmosphère moyenne (MANTRA) afin d'étudier les changements dans les concentrations de l'ozone stratosphérique des latitudes moyennes de l'hémisphère Nord de même que des composés azotés et chlorés qui jouent un rôle dans la chimie de l'ozone. Quatre campagnes ont été menées*

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jusqu'à maintenant, toutes à Vanscoy (52 01'N, 107 02'W, 511,0 m), en Saskatchewan, au Canada. La première mission MANTRA a eu lieu en août 1998 et le vol de ballon du 24 août 1998 était le premier lancement d'un gros ballon de haute altitude au Canada depuis environ 15 ans. Le ballon a transporté une charge utile d'instruments servant à mesurer la composition de l'atmosphère et a pris des mesures en flottant à une altitude entre 32 et 38 km pendant une journée. Trois de ces instruments avaient été lancés dans le cadre des vols Stratoprobe du Service de l'environnement atmosphérique (aujourd'hui le Service météorologique du Canada) dans les années 1970 et au début des années 1980, fournissant ainsi des données historiques antérieures au commencement de la perte d'ozone dans les latitudes moyennes.

Les instruments emportés par ballon ont fourni les mesures principales suivantes : ozone, NO₂, HNO₃, HCL, CFC-11, CFC-12, N₂O, CH₄, température et rétrodiffusion par les aérosols. Des mesures de l'ozone, du NO₂ et du SO₂ totaux et de l'épaisseur optique des aérosols dans la colonne ont été prises au moyen de trois spectromètres déployés au sol durant la campagne. Des sondages par radiosondes et par sondes d'ozone régulières ont aussi été effectués au cours des deux semaines précédant le lancement principal pour connaître les conditions atmosphériques locales (vents, pression, température, humidité) dans la zone de vol du ballon principal. Nous avons comparé les données au modèle de transport chimique MEZON (Model for Evaluating oZONE Trends), au modèle de la boîte photochimique de la University of California at Irvine et au modèle canadien de l'atmosphère moyenne (CMAM) pour vérifier notre compréhension actuelle des corrélations entre la photochimie des modèles et les espèces des latitudes moyennes. Cet article présente un aperçu de la mission MANTRA de 1998 et sert d'introduction aux articles d'accompagnement publiés dans le présent numéro d'Atmosphere-Ocean qui décrivent divers aspects et résultats de cette campagne.

1 Introduction

It is now generally accepted that ozone concentrations have declined significantly since about 1980 in response to the enhanced levels of chlorine in the stratosphere resulting from anthropogenic emissions of chlorofluorocarbons (e.g., Kerr, 1991; Wardle et al., 1997; Harris et al., 1998; Solomon, 1999; Staehelin et al., 2001; WMO, 1999, 2003). Average total ozone columns at northern mid-latitudes (35°–60°N) declined by about 3% between 1980 and 1997–2001, with these losses exhibiting both seasonal and regional variations (WMO, 2003). There have been statistically significant decreases at all altitudes from 12 to 45 km, with maxima of about 7.3% per decade at both 40 km and 15 km (Harris et al., 1998). Only tropical regions have not experienced a detectable decline in the ozone column.

Ozone destruction is controlled by NO_x (NO + NO₂) through both the NO_x catalytic cycle and its influence on the level of free chlorine in the lower stratosphere. It follows that changes in the amount of total odd nitrogen (NO_y = NO_x + all oxidized nitrogen species = NO + NO₂ + NO₃ + 2 × N₂O₅ + HNO₃ + HNO₄ + ClONO₂ + BrONO₂) or changes in the partitioning of nitrogen compounds between NO_x and the longer-lived constituents will have an impact on the ozone budget. While NO_x/NO_y partitioning is largely determined by ozone and aerosol concentrations (Salawitch et al., 1994; Solomon et al., 1996), there are several mechanisms that can change the concentration of NO_y. Increases in NO_y are expected due to increasing tropospheric N₂O, which has seen a rise in global average concentration from a pre-industrial value of 275 ppbv to 312 ppbv in 1996 (WMO, 1999), and as a consequence of decreasing stratospheric ozone (which increases N₂O), while a decrease in NO_y may result from a cooling of the stratosphere (e.g., McLinden et al., 2001). Decreasing temperatures in the polar lower stratosphere can lead to NO_y loss due to the formation of HNO₃ in and on polar stratospheric clouds and its subsequent sedimentation on the larger particles. Variable aerosol concentrations over the last twenty years may also

have increased the impact of denitrification. In addition, stratospheric NO_y levels may be perturbed by the current fleet of subsonic aircraft (Thompson et al., 1996).

A number of balloon campaigns and satellite instruments have measured some or all of the components of the NO_y family at mid-latitudes over the past twenty-five years (e.g., Ackerman et al., 1975; Evans et al., 1976b, 1981; Callis et al., 1985; Russell et al., 1988; Roscoe et al., 1990; Webster et al., 1990; Chance et al., 1996; Kondo et al., 1996; Morris et al., 1997; Murcray et al., 1987; Sen et al., 1998; Gao et al., 1999; Jucks et al., 1999; Toon et al., 1999). These yield a wide range of estimates for NO_y in the mid-stratosphere (30–35 km), varying between 11 ± 5 ppbv and 21 ± 2 ppbv. Long-term studies of odd nitrogen in the stratosphere are relatively few. The systematic monitoring of even the NO₂ column has only been carried out at a few locations and for only limited time intervals (e.g., Kerr and McElroy, 1976; Kerr et al., 1982; McKenzie et al., 1991). More recently, Randel et al. (1999) have analysed trends in several species from Upper Atmosphere Research Satellite data, and have identified a statistically significant increase in NO₂, as well as a trend of about -2% in HNO₃ between 1993 and 1997 which is not entirely accounted for by the change in NO₂. The eruption of Mt. Pinatubo in 1991 resulted in a positive trend in NO₂ for some years afterwards that has been linked to perturbations in dynamics and to chemical recovery of NO₂ following the repartitioning to HNO₃ caused by the eruption (Koike et al., 1994). Liley et al. (2000) have analysed eighteen years of ground-based NO₂ columns over Lauder, NZ (45°S) and derived a trend of +5% per decade, which is consistent with the observed N₂O increase of +3% per decade if trends in ozone, chlorine, and bromine are taken into account (McLinden et al., 2001).

In the 1970s and 1980s, the Atmospheric Environment Service (AES) of Canada (now the Meteorological Service of Canada (MSC)) led the Stratoprobe series of balloon flights that included measurements of NO₂ and HNO₃. These campaigns contributed to our understanding of the stratosphere and

included early estimates of the northern hemisphere mid-latitude odd-nitrogen budget (Evans et al., 1976b, 1977, 1981, 1982a, 1982b, 1985; Kerr and McElroy, 1976; Kerr et al., 1982; Ridley et al., 1976, 1984). Changes to the mid-latitude ozone layer which started in the 1980s (Bojkov et al., 1990; Stolarski et al., 1991, 1992; Kerr and McElroy, 1993; Hofmann et al., 1994) followed shortly after the end of the Stratoprobe flight series. The Stratoprobe results are notable because they: (1) predate the onset of ozone decline, (2) connect to the Arctic through flights made using the same equipment in 1978 and 1979, and (3) have been compared to satellite results from lower latitudes (e.g., Limb Infrared Monitor of the Stratosphere (LIMS) – Gille and Russell (1984)). The historical Stratoprobe measurements thus provide a useful benchmark against which current measurements can be compared to examine the possibility of detecting long-term changes.

The Middle Atmosphere Nitrogen TRend Assessment (MANTRA) series of high-altitude balloon flights has thus been undertaken to investigate changes in the odd-nitrogen budget of the northern hemisphere mid-latitude stratosphere, building on the earlier Stratoprobe balloon campaigns. The overall scientific objectives of the MANTRA project are as follows:

- (1) To conduct a series of balloon flights, each carrying a suite of instruments to measure vertical profiles of the key stratospheric species that control the mid-latitude ozone budget, particularly species in the NO_y , Cl_y , Br_y , and HO_x chemical families, along with dynamical tracers and aerosols;
- (2) To combine these measurements with those obtained from similar northern mid-latitude campaigns of the past twenty years, in order to quantify changes in the chemical balance of the stratosphere, with a focus on the odd-nitrogen budget;
- (3) To compare measurements of the same species recorded by different instruments on the balloon, and perform an intercomparison and assessment of the old and new measurement techniques; and
- (4) To use the measurements for evaluation and ground-truthing of satellite measurements, including those made by instruments on the Odin, SCISAT-1, and ENVISAT platforms.

Four campaigns have been carried out to date, all from Vanscoy, Saskatchewan, Canada (52°01'N, 107°02'W, 511.0 m above sea level). The first MANTRA mission took place in August 1998, with the primary balloon flight on 24 August being the first Canadian launch of a large high-altitude balloon in about fifteen years. Subsequent MANTRA campaigns were conducted in the late summers of 2000, 2002 and 2004. While the accurate detection of trends using limited datasets is difficult (e.g., Weatherhead et al., 1998, 2000), the intention of the MANTRA project is to contribute to the international body of knowledge needed to address the issue of changing stratospheric composition.

The MANTRA 1998 payload included three of the original Stratoprobe instruments and three newer instruments, allowing

a dual intercomparison: one between measurements obtained by the same instruments after an interval of fifteen to twenty years, and the other between old and new measurement techniques. The three older instruments were an interference-filter infra-red emission radiometer for HNO_3 (circa 1983 version of the instrument described by Evans et al. (1976a)), a high-resolution scattering spectrometer for OH (McElroy and Wardle, 1978), and a visible grating spectrophotometer for NO_2 measurements (Kerr and McElroy, 1976). In addition, NO_2 and HNO_3 , along with a number of other key trace species, were simultaneously measured using a photodiode array spectrometer (McElroy, 1995; McElroy et al., 1994, 1995), and a Fourier transform infra-red spectrometer (Murcay, 1987; Murcay et al., 1987; Goldman et al., 1988), and a new acousto-optic tunable filter spectrometer. Supporting ground-based observations were performed by a Brewer spectrophotometer, another photodiode array instrument, and a zenith-sky grating spectrometer. Regular radiosonde and ozonesonde flights were conducted during the two weeks prior to the primary balloon flight to characterize the local atmospheric conditions, and an aerosol backscatter sonde was flown on the day of the primary balloon flight. With this combination of instruments, the following measurements were made during the MANTRA 1998 campaign: vertical profiles of ozone, NO_2 , HNO_3 , HCl, CFC-11, CFC-12, N_2O , CH_4 , and aerosol backscatter, vertical columns of ozone, NO_2 , and SO_2 , and aerosol optical depth, and J-values for O^1D and NO_2 .

This paper is intended to provide an overview of the MANTRA 1998 mission, and serves as an introduction to the accompanying papers in this issue of *Atmosphere-Ocean* that describe specific aspects and results of this campaign. It briefly describes the instruments deployed on the balloon and on the ground, the balloon flight, the measurements, and the modelling results that have been obtained.

2 MANTRA instruments

a MSC Emission Radiometer

The first of the historical Stratoprobe instruments is the MSC emission radiometer, which is based on the balloon emission radiometer of Pick and Houghton (1969), which was later modified by Evans et al. (1976a) and further improved in the early 1980s. These modified versions were flown many times in the 1970s and 1980s to measure stratospheric HNO_3 . The instrument uses a liquid-nitrogen-cooled detector and optics to measure the atmospheric thermal emission at low resolution (20 cm^{-1}) in the 8–14 μm ($715\text{--}1250 \text{ cm}^{-1}$) atmospheric window during balloon ascent. The most recent version (circa 1983) of this instrument was flown during MANTRA and measures the emission using a circular variable filter to provide spectral information. Two of these emission radiometers were included on the MANTRA 1998 payload and were mounted at elevation angles of 19° and 41°, providing independent atmospheric paths and allowing an assessment of the effects of payload swing. A detailed description of the MANTRA emission radiometers and their flight preparation is given by Quine et al. (this issue).

The original analysis approach involved differentiating radiance profiles with respect to height to assign a radiance change to each layer, from which the concentration of HNO_3 was calculated (Evans et al., 1976a, 1976b, 1977). However, this required smoothing of the radiances prior to differentiating, which made the retrieved volume mixing ratios sensitive to the degree of smoothing applied. A new analysis technique was therefore developed for the analysis of the MANTRA flight data, and is described by Quine et al. (this issue). The HNO_3 concentrations obtained with this instrument are accurate to about 20% in a 5-km layer. The difference between the old and new observations should be accurate to about 30% for a 5-km-thick layer.

b *MSC High-Resolution OH Scattering Spectrometer*

The MSC high-resolution scattering spectrometer for measurements of OH was flown on Stratoprobe payloads in July 1975 from Yorkton, SK and in September 1976 from Palestine, TX (McElroy and Wardle, 1978). The OH resonance scattering technique has been used since 1975 by others (Torr et al., 1987), but without the MSC refinement of using polarization. This instrument measures polarized spectra of the sky at right angles to the Sun and at about 10° above the horizon. It observes the direct solar beam (as a reference), and strong and weak polarizations at 90° to the Sun to detect resonant scattering by OH. The spectral range and resolution of the instrument are 305 to 311 nm, and 0.04 nm, respectively. The OH signal-to-noise ratio in the altitude range 25 to 40 km is high, but the lack of sufficiently detailed, very-high-resolution data on the solar spectrum precluded an absolute calibration of the original historical measurements. This problem can be solved today, and the calibration factor will not have changed between the earlier flights and the present. Thus measurements of OH taken with this instrument during MANTRA would be directly comparable with those taken in 1975 and 1976. The expected accuracy of the OH measurements is approximately 15% based on the mode in which it was flown during the Stratoprobe campaigns, and less than 10% assuming a “best” case scenario which includes a number of improvements that should be possible now. This “best” case estimate includes: (a) improved gondola swing-detecting instrumentation that could reduce the largest uncertainty (the 10% associated with elevation angle determination) to about 2 or 3% (based on subsequent MANTRA gyroscope elevation measurement performance); (b) possible future improvements to absolute OH spectroscopic errors (from 5% to 3%); and (c) good characterization of ozone absorption effects by using reliable vertical profile data (from 5% to 3%).

The OH spectrometer was independently mounted at 90° to the solar-pointing instruments. Due to a problem with its solar tracker tilt mechanism just prior to launch, it was mounted at a fixed angle of 5.3° . It recorded polarized spectra of the sky (from 305 to 311 nm) at right angles to the Sun throughout the day, but signal levels proved to be too low for OH retrievals. However, the instrument worked well on two later flights (MANTRA 2002 and 2004) and analysis of these results is in progress.

c *MSC NO_2 Spectrophotometer*

The MSC NO_2 spectrophotometer is an antecedent of the Brewer ozone spectrophotometer and was flown on a number of balloons in the 1970s and 1980s. It is a modified Ebert-Fastie spectrograph with a mechanical chopper system that is used to multiplex the single photomultiplier detector between five wavelengths (exit slits) in the 430 to 450 nm region of the spectrum. It can be used to make solar occultation measurements (Bloxam et al., 1975; McElroy, 1976; Kerr and McElroy, 1976) and to measure light scattered from the Earth’s limb (McElroy, 1985, 1988; Roscoe et al., 1985, 1990). In earlier flights, this instrument typically provided NO_2 concentrations at about 5% precision and 10% absolute accuracy for a 2-km-thick layer. The measurements made in 1976 should be comparable to new measurements at the 10% confidence level. Just prior to the MANTRA flight, it was successfully used to make evening twilight atmospheric observations on the ground, from which a set of NO_2 slant columns was derived. Unfortunately, the NO_2 spectrophotometer failed prior to launch and so no balloon-borne measurements were obtained; therefore no further details on this instrument are provided here.

d *MSC SunPhotoSpectrometer*

The MSC SunPhotoSpectrometer (SPS) is a photodiode array spectrometer that has been flown on STS-52 and aboard the National Aeronautics and Space Administration (NASA) ER-2 aircraft as part of the NASA Upper Atmospheric Research and High Speed Research Programs (McElroy et al., 1994; McElroy, 1995). On the ER-2, the instrument is called the Composition and Photodissociative Flux Measurement (CPFM) Experiment, and is used as an absolute spectroradiometer and to provide ozone data using the technique of differential absorption spectrophotometry. The instrument has flown at 21 km nearly one hundred times with this program.

The SPS is based on an EG&G randomly addressable 1024-element photodiode array detector, situated at the focus of an $f/2$ holographic diffraction grating. Spectra are recorded from 375 to 775 nm in first order and from 188 to 388 nm in second order, with a spectral resolution varying from 1.2 to 4 nm (Full Width at Half Maximum intensity (FWHM)). The dynamic range is greater than 10^6 , and the signal-to-noise ratio is typically 1000:1. Typically, data from wavelengths down to 300 nm are used in the analysis, due to the decreased ultraviolet (UV) sensitivity of the detector coupled with the large contribution of stray light to the observed signal at shorter wavelengths. Data can be collected in both solar occultation mode and limb-scan mode. Apparent column densities of both ozone and NO_2 as a function of tangent height are retrieved using a chi-squared minimization technique, and vertical profiles are derived by applying the Chahine relaxation method (Chahine, 1972, 1977). Information about H_2O , O_2 , and aerosols can also be derived from the spectra. The errors in the retrieved ozone and NO_2 mixing ratios are estimated to be $\pm 10\%$ of the retrieved values.

A second version of this instrument (SPS#2) was flown during MANTRA to make absolute radiometric measurements

of the direct solar beam, the brightness of a point on the horizon, and the brightness of the upwelling radiation from below the balloon. These three fields can be integrated with the cross-sections of the relevant absorbers to produce J-values for reactions such as the photolysis of ozone and the photodissociation of NO_2 (McElroy et al., 1995), which are needed to partition the odd-nitrogen family properly in photochemical model simulations.

e *University of Denver Fourier Transform Spectrometer*

The University of Denver Fourier transform spectrometer (FTS) is a high-resolution Bomem DA2 interferometer that has been used in a large number of flights in the United States since 1977 (e.g., Murcray, 1987; Goldman et al., 1988; Murcray et al., 1987). The interferometer has a 50-cm path difference, which results in an unapodized FWHM resolution of about 0.01 cm^{-1} . The system uses a dichroic beamsplitter that directs the output beam from the interferometer onto two infra-red detectors. The InSb detector covers $2650\text{--}3250 \text{ cm}^{-1}$ and is used to measure HCl and ozone, while the HgCdTe detector covers $700\text{--}1300 \text{ cm}^{-1}$ and is used to measure ozone, HNO_3 , CH_4 , N_2O , ClONO_2 , CFC-11, and CFC-12. While NO could have been measured with the FTS, this would have precluded the measurement of HCl. Because NO can be derived from measurements of other species, it was decided to configure the FTS to measure HCl rather than NO for the 1998 flight.

The FTS records light from the central 70% of the solar disk using a servo-controlled solar tracking system. Spectra are recorded in solar occultation as the Sun rises or sets and analysed using a line-by-line layer-by-layer algorithm (Blatherwick et al., 1989). Vertical profiles of mixing ratio are retrieved using the “onion peeling” technique (Rodgers and Connor, 2003), as described by Fogal et al. (this issue). A large number of atmospheric constituents can be measured with this instrument. Typical accuracies in a 5-km layer are 10–15% for ozone, 15% for NO_2 , 10% for HNO_3 , 15% for ClONO_2 , and 10% for HCl. Additional species measured are N_2O , CH_4 , CFC-11, and CFC-12. Further details on the FTS instrument and data analysis are given by Fogal et al. (this issue).

f *CRESTech/York University Acousto-Optic Tunable Filter Spectrometer*

The Centre for Research in Earth and Space Technology (CRESTech)/York University acousto-optic tunable filter (AOTF) spectrometer is a new instrument that uses an MgF_2 acousto-optic filter to scan the spectrum in the visible and near-UV regions (250–400 nm). It has a wavelength-dependent resolution of 0.07 nm at 250 nm, 0.1 nm at 300 nm, 0.15 nm at 350 nm, and 0.21 nm at 400 nm. The detector is a photomultiplier tube, with very low dark count. The system throughput is $5.7 \times 10^{-4} \text{ cm}^2 \text{ sr}$ with an external field-of-view of 0.05° half angle. During flight, an on-board mercury lamp is used to calibrate the wavelength. The AOTF instrument consists of two cylinders mounted one on top of the other. The main cylinder is 1.5-m long and contains the baffle, telescope optics, a filter wheel, a mercury calibration lamp, the

AOTF itself and the detector (with its high voltage supply). All components, except the baffle and telescope, are in a sealed housing to maintain ground-level pressure. The second cylinder, 1 m long, is also airtight and contains the control and AOTF radio frequency (RF) electronics which are connected to the instrument housing by three low-voltage cables. The total mass, excluding batteries, is 45 kg. The AOTF spectrometer is designed for balloon-borne solar occultation and limb-scanning observations, in order to measure profiles of ozone and NO_2 , and to set detection limits for OCIO and BrO. Both modes require balloon pointing (azimuth control) and altitude scanning (tilting platform).

g *Ground-Based Instruments*

The primary role of the three ground-based optical instruments was to provide vertical column abundances of ozone and NO_2 to constrain and verify the vertically resolved measurements made by the balloon-borne instruments. In addition, the campaign provided the opportunity to compare the instruments against each other, to set detection limits for the slant columns of OCIO and BrO, and to investigate the accuracy of vertical profiles retrieved from twilight zenith-sky slant columns of NO_2 .

The MSC contributed a Brewer single spectrophotometer (#029) to the campaign (Brewer, 1973; Kerr et al., 1984; Kerr et al., 1988). The Brewer is fully automated, and can make quasi-simultaneous measurements of total column ozone, NO_2 , SO_2 , aerosol optical depth, and UV-B radiation, using observations at discrete wavelengths. Ozone vertical columns are typically measured at 1% accuracy and 0.5% precision. Measurements are primarily made using direct solar and lunar viewing. For additional details on the Brewer instrument and measurements, see Savastouk and McElroy (this issue).

A new portable ground-based UV-visible zenith-sky spectrometer was assembled at the University of Toronto just prior to the MANTRA campaign (see Bassford et al., 2001, this issue). This instrument is based on a “Triax-180” crossed Czerny-Turner imaging spectrometer manufactured by Instruments SA. It has an adjustable entrance slit and a triple-grating mount to provide flexibility in the available resolution and dispersion. The detector is a “Spectrum One” CCD array from Instruments SA, which uses a SITE chip of 800×2000 pixels, each $15 \mu\text{m} \times 15 \mu\text{m}$. The CCD is back-illuminated for high quantum efficiency, UV-coated for response down to 300 nm, and thermoelectrically cooled. Total column abundances are retrieved from the zenith-sky spectra using differential optical absorption spectroscopy (DOAS) (Solomon et al., 1987). This is a well-established technique, with typical accuracies of $\pm 10\%$ for total ozone and NO_2 (e.g., Hofmann et al., 1995; Vaughan et al., 1997), and $\pm 15\%$ to $\pm 35\%$ for OCIO and BrO. The zenith-sky spectrometer and data analysis are described by Bassford et al. (this issue). Vertical profiles of NO_2 can also be retrieved, following the approach of Preston (1995) and Preston et al. (1997, 1998), as described by Melo et al. (2004) and Melo et al. (this issue). The MANTRA 1998 campaign provided an opportunity to deploy

this new instrument and to evaluate its performance against simultaneous measurements of ozone and NO_2 made by a number of other instruments.

A ground-based photodiode array spectrometer, SPS-G, similar to those flown on the balloon (see Section 2d) was also deployed during MANTRA. Spectra of sunlight scattered from the zenith sky were recorded and analysed using a spectral fitting code to determine vertical columns of ozone and NO_2 .

h *Ozonesondes, Radiosondes, and Aerosol Sondes*

A series of independent ozonesonde flights was made in support of the primary balloon launch (for details, see Davies et al., 2000). Electrochemical Concentration Cell (ECC) sondes were used with Vaisala RSA-11 or T-MAX C interfaces and Vaisälä Metgraph software (Komhyr, 1969). Ozonesondes make accurate in situ measurements of the ozone profile on ascent using a reaction between ozone and potassium iodide that converts iodide to iodine, which can then be detected electrically. Typically, the ozone partial pressure is measured at 50–100 m vertical resolution with an accuracy of $\pm 10\%$, depending on altitude. The main sources of error are the pump correction (which is applied above 200 hPa) and background current in the troposphere.

Radiosonde flights were conducted prior to the primary launch using Vaisälä RS 80s with Global Positioning System (GPS) wind finding (Antikainen and Hyvonen, 1983). These provided information on wind speed and direction for flight planning, as well as pressure (0.1 mb resolution, ± 0.5 mb accuracy), temperature (0.1°C resolution, ± 0.2 °C accuracy), and humidity (1% relative humidity (RH) resolution, $\pm 2\%$ RH accuracy).

Aerosol backscatter sondes, obtained from the University of Wyoming, were also flown during the campaign, just after the primary launch. These instruments use a photodiode to measure backscattered light from a xenon flashlamp at 490 and 940 nm. Observations are made on ascent during darkness and are used to derive vertical profiles of aerosol backscatter (Rosen and Kjome, 1991, 1997).

3 The primary balloon payload

a *Payload Configuration*

The balloon gondola consisted of a lightweight aluminum frame, about 2 m \times 2 m \times 2 m in size, built of 3.8 cm \times 3.8 cm square tubing and cross braced. Solar shields were mounted on the gondola support frame to reduce heating of the instruments at float altitude. This frame carried two major structures: the University of Denver FTS with its own solar tracking system, and an elevation pointing table. Three co-aligned instruments were mounted on this table: the NO_2 spectrophotometer, SPS#1, and the AOTF spectrometer. The emission radiometers were mounted directly on the gondola structure, but oriented at 180° to the solar direction so that the Sun would not enter the instrument field-of-view. The OH scattering spectrometer was mounted on the gondola at 90° to the solar direction, while SPS#2 was suspended from the upper edge of the opposite side of the gondola, also at 90° to the solar direction. The instrument layout is shown in Fig. 1.

The payload also included the command and telemetry package, pointing systems, batteries, ballast, and crush pads, as well as the flight train and a parachute, all standard components that have been flown on previous balloon flights. Power was supplied by 28-V lithium battery packs. The mass of the instrument package (including batteries) was about 300 kg, while the total payload had a mass of about 630 kg. A GPS unit was used to provide position and altitude information. A 16-channel 115.2-kbaud serial statistical multiplexer and an S-Band transmitter were used to provide telemetry between the instruments and the ground station. A limited serial command link enabled commands to be sent in real time to the payload or a specific instrument and responses from the instrument to be received at the ground station. This was used to change instrument operation modes and to initiate in-flight calibrations. Data were recorded at the ground station in real time and required a line-of-sight view of the balloon.

b *Pointing Control*

Two pointing control systems were flown on the gondola: an existing Sun-tracking system that included both azimuth and elevation control, and a new limb-scanning system. The entire gondola was suspended from an azimuth-pointing joint so that the gondola and the elevation pointing table could be directed at or away from the Sun in a controlled fashion. Azimuth, or yaw, control was maintained by a drive linkage situated about 1 m above the suspended gondola. This system uses Sun sensors (a fine sensor having an accuracy of $\pm 0.1^\circ$ over a $\pm 1^\circ$ interval and a range of $\pm 5^\circ$, and a coarse sensor having an accuracy of $\pm 1^\circ$ over a $\pm 5^\circ$ interval and a range of $\pm 50^\circ$), as well as a coarse mechanical gyroscope mounted on the gondola to correct pointing. Drive and control are by an analogue electronics system with substantial flight heritage. Before sunrise and after sunset, a crude azimuth angle of a few degrees is maintained using a magnetic field sensor. The azimuth controller also employs a gyroscope to provide azimuth rate information to aid control stability. During periods of stable flight (at float altitudes) this system has a pointing performance of the order of $\pm 0.5^\circ$.

In addition, a stepping motor drives the pointing table in elevation (or pitch). Pointing table elevation could be adjusted in steps of 0.018° to track the Sun. Elevation is determined by Sun sensors (fine and coarse) and controlled by analogue control electronics. The system has two modes: a rally mode moves the table rapidly to Sun pointing and a fine pointing mode steps the table to track small changes in Sun pointing. This system also has a pointing performance of the order of 0.1° and is used for solar occultation measurements. However, it does not provide for roll control. Previous flight analysis indicates that during stable periods of operation there can be a gondola swing of up to 2° pivoted somewhere near the top of the flight train (30 m above the gondola) which introduces a small additional pointing error.

For the MANTRA mission, a new limb-pointing system was developed to enable off-Sun limb-scanning observations during the day. During the limb-scanning mode, knowledge

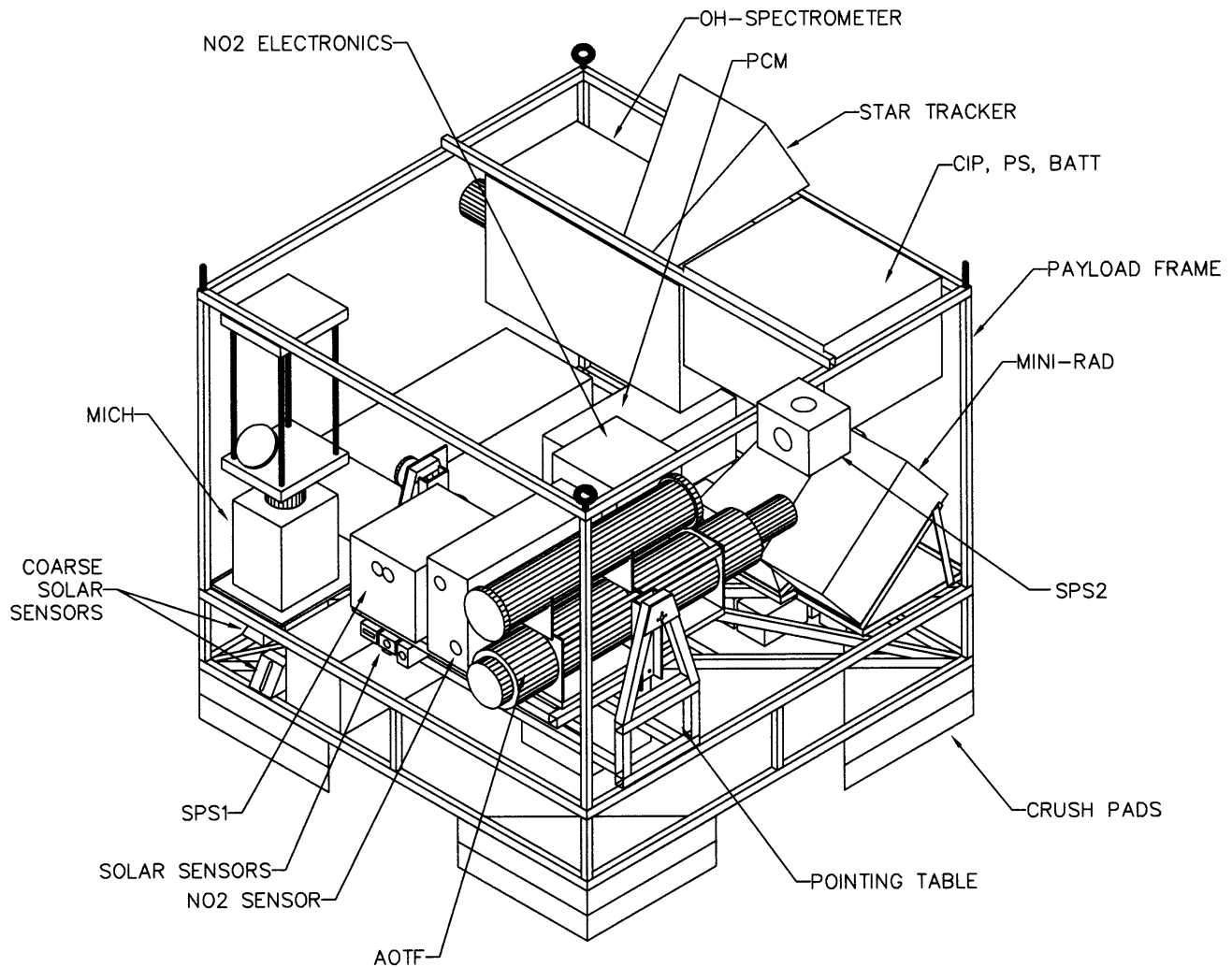


Fig. 1 Instrument layout on the gondola.

of the Sun vector does not provide useful information for controlling the tilting table elevation; therefore some other sensing technique must be employed. This system consisted of a small microprocessor controller, a Litton G2000 two-axis mechanical gyroscope, several clinometers (gravity-referenced inclinometers) and a pointing table shaft encoder. The microprocessor controller, running at 10 Hz, generates a drift-corrected integrated rate output from gyroscope readings. With drift correction, the G2000 gyroscope can achieve a drift stability of better than 1° per hour. The small long-term gyroscope drift was compensated for by using a single state linear Kalman filter that combined the gyroscope output with the clinometer data. (Because the clinometers are gravity referenced, they cannot be used directly to control elevation as they do not detect pendulum motion.) The pointing table is designed to be scanned up and down on command through a range of angles corresponding to the tangent heights at which observations are required. The pointing direction is stabilized in absolute spatial co-ordinates by feeding a vertical reference from a differential GPS system into the control system. Both

azimuth and elevation data are recorded, but only the elevation information is utilized for control.

The limb-scan pointing system achieved pointing control to $\pm 0.25^\circ$ in dynamic tests performed prior to flight. During the MANTRA flight, the system behaved nominally until just before it reached float altitude, at which time a power supply failure occurred, preventing its use for daytime limb-scanning observations. However, the existing solar tracking system allowed occultation measurements at sunrise and sunset, and a subsequent version of this pointing system was successfully flown in 2000 and is fully described by Quine et al. (2002).

4 The MANTRA balloon flight

a Launch Preparations

The launch facilities were provided by Environment Canada, which has a balloon launching station at Vanscoy, Saskatchewan, at $52^\circ 01'N$, $107^\circ 02'W$, and 511.0 m elevation. Scientific Instrumentation Limited (SIL) operates this facility, and was the launch contractor for MANTRA. The instruments

arrived at the launch facility on 3 August 1998, and underwent final commissioning, payload integration, electrical and RF interference tests, full-up electrical tests, and dynamic and balance tests, culminating in a launch dry run on 19 August.

Daily forecasts of both surface and stratospheric winds were produced throughout August using the Canadian Meteorological Centre's global weather forecast model, the Global Environmental Multiscale (GEM) model (Côté et al., 1998a, 1998b). The high-resolution version was used to provide 48-h forecasts of the surface and low-altitude winds at 15-km and 35-km horizontal resolution to aid in identifying suitable launch conditions at the surface. The global version of GEM with a model top at 10 mb provided 5-day forecasts for balloon trajectory calculations, which were used to identify the time of stratospheric wind reversal at float altitude.

An August launch date was chosen to coincide with fall turnaround, when dynamical variability is minimized in the stratosphere and stratospheric winds change direction from westward to eastward (Fahey et al., 2000; Fioletov and Shepherd, 2003; Wunch et al., this issue). This provides a brief period of winds that are sufficiently low so that the balloon remains within telemetry range (about 400 km) for a full day of measurements from float altitude. In addition, many of the historical Stratoprobe flights were also carried out at fall turnaround; it was thus preferable that the MANTRA data should be obtained at the same time to mitigate seasonal effects in any trends.

b *The Balloon Flight*

The primary balloon was launched at 3:25 AM local time (09:25:25 UTC) on Monday, 24 August 1998. An 11.62 million cubic feet (mcf) Winzen balloon made of Astrofilm E dating from May 1986 and assembled in October 1986 was used for this flight, and was capable of carrying a payload of 1040 kg to 39 km. Soon after launch, measurements by the emission radiometers were carried out on ascent. Sunrise measurements by the FTS, the SPS instruments, and the AOTF spectrometer began at 5:30 AM local time, at which time the balloon had reached an altitude of about 26 km. These solar occultation measurements continued during the remainder of the ascent, with the payload reaching its maximum altitude of 38.8 km at about 7:15 AM, shortly after sunrise. During the day, the balloon floated at altitudes between 32 and 38 km, with some limb observations made during this time. The winds at float altitude were negligible, so that the balloon remained almost directly overhead all day. The balloon may have had a small leak as it slowly descended during the day and did not maintain the expected altitude of 39 km. Sunset solar occultation measurements began at 7:40 PM local time from an altitude of about 31 km.

The balloon's flight profile and trajectory were derived from GPS data. These were reprocessed after flight to remove corrupt data, which were caused by filtering algorithms implemented in the GPS flight unit that made it unsuitable for tracking balloon location, and by an antenna that did not give adequate GPS satellite coverage. Future balloon missions

should fly a GPS that also transmits the raw data for later processing. The GPS data were evaluated using a variety of means including the bounding of possible balloon speed, forcing a monotonic time sequence, and removing obviously corrupt data points manually. About one-third of the original dataset was discarded. Position information is probably accurate to better than about 0.1° in latitude and longitude, and altitude information to better than about 500 m. The uncorrupted data have been interpolated on a 20-s time interval throughout the mission. Figure 2 shows the resulting flight altitude profile while the balloon remained within telemetry range of Vanscoy, and Fig. 3 presents the corrected location data for the balloon for the same period.

Termination was attempted at 9:16 PM local time on 24 August (03:16 UTC on 25 August), but was unsuccessful. Two explosive squib systems failed to separate the balloon from the gondola and parachute, although the telemetry monitor indicated that they had fired and successfully cut the wire cable holding the terminate mechanism together. Operation of the balloon valve to release helium at 9:45 PM also failed to bring down the payload, although telemetry indicated that the valve was opening and closing. The valve was then left in the open position. At 11:15 PM, an electronic timer was activated to cut the cable, but again, no separation was observed. The balloon subsequently descended at about 1.5 km h^{-1} overnight to an altitude of about 18 km (60,000 ft). With efforts to bring the balloon down unsuccessful, it started to drift eastwards, tracked by an SIL spotter plane during the day, and had travelled about 400 km from Vanscoy by sunset on 25 August. It eventually moved beyond telemetry range at 9:45 PM local time on 25 August (03:45 UTC on 26 August).

During the day on 26 August, the balloon was spotted by several aircraft and was seen east of Thunder Bay in the early afternoon. It reached the Gulf of St. Lawrence on 27 August, by which time it had descended to near the altitude of trans-Atlantic air lanes. At Transport Canada's request, the Department of National Defence (DND) sent two CF-18 jets from Baggottville, QC in pursuit of the balloon. At 23:00 UTC on 27 August, when the balloon was at an altitude of about 12.5 km (41,000 ft), the CF-18 jets fired 1000 rounds of 20 mm ammunition at it over the west coast of Newfoundland, trying to weaken the cables connecting the balloon and gondola. However, these efforts were unsuccessful and the balloon drifted out over the north Atlantic, tracked by DND from land and then by DND Aurora aircraft on 28 August.

Later that day, the balloon was acquired by British Air Traffic Control (ATC) and was tracked by a Royal Air Force (RAF) Nimrod aircraft, before swinging north and being acquired by Icelandic ATC at 10:45 UTC, at which time it was at an altitude of about 10 km (34,000 ft). On 29 and 30 August, tracking was continued by a United States Air Force (USAF) P3 Orion aircraft. The balloon entered Norwegian airspace on 30 August, passing west of Spitzbergen; Icelandic ATC stopped tracking at 12:12 UTC, and the last sighting was at 19:45 UTC with an unconfirmed altitude of 5.5–6.7 km (18,000–22,000 ft). The balloon then headed into Russian airspace and two

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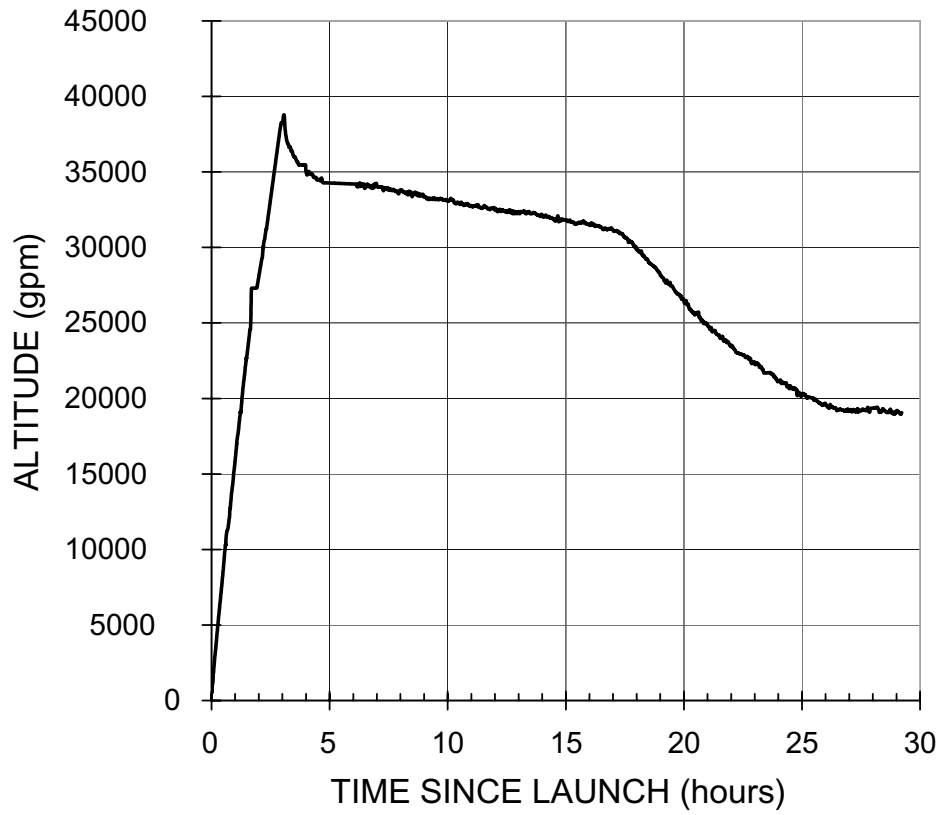


Fig. 2 The balloon flight altitude profile derived from corrected GPS readings while within telemetry range of Vanscoy.

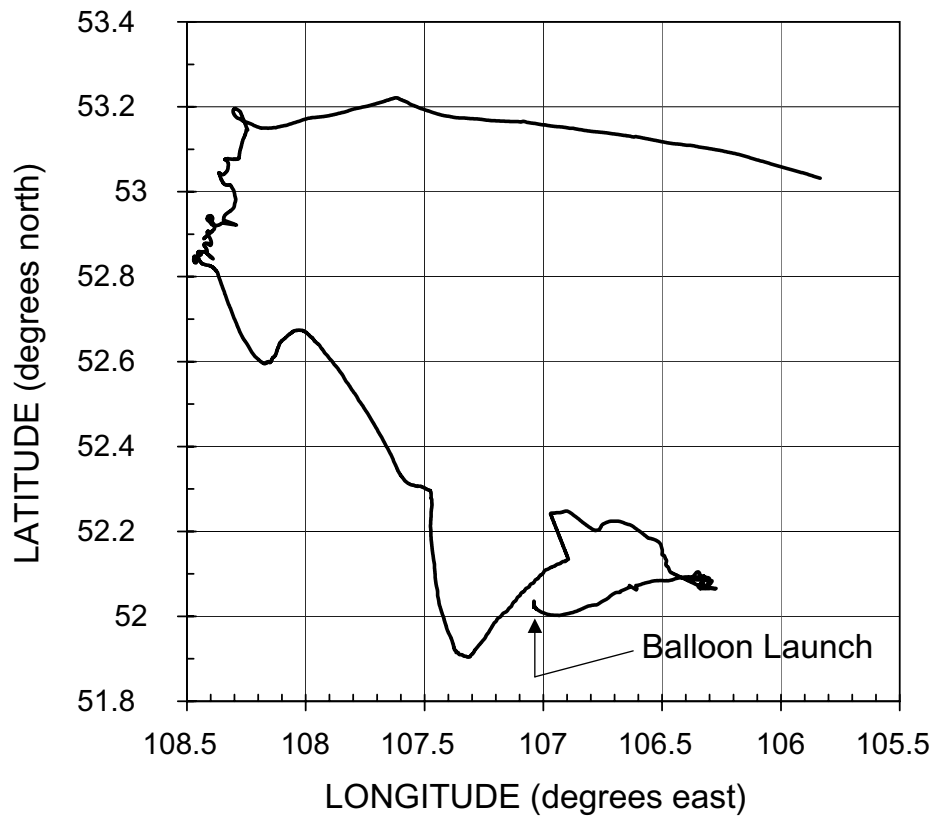


Fig. 3 The balloon trajectory derived from corrected GPS readings while within telemetry range of Vanscoy.

days later, on 1 September, there was an unconfirmed sighting from Russia. At this time, N. Elansky of the Russian Academy of Sciences located the balloon near the Kol'Skiy Peninsula, Russia and calculated its forward trajectory. Finally, at 1:30 PM local time on Wednesday, 2 September, the balloon landed near Mariehamn on the Åland Islands, Finland, in the middle of the Baltic Sea, a location that was on Elansky's projected flight path. Figure 4 illustrates the approximate path followed by the balloon on its 9000-km voyage from Vanscoy. Video footage of the descent shows that the gondola and parachute separated from the balloon at about 150 m above the surface. P. Taalas and J. Kaurola from the Finnish Meteorological Institute recovered the payload. The instruments suffered minor damage from a bullet, shrapnel, and the landing, but all are reusable.

Along with the instrumentation, the termination device and the helium valve were also recovered, enabling an investigation of the failed mechanisms. The terminate device, mounted between the parachute and the balloon, consists of two dove-tailed aluminum blocks held against a central block by a wire cable. To terminate the flight, a radio command was sent to fire two explosive cable cutters that cut the cable, allowing the balloon and parachute to separate, and pulling a ripcord that tears open the balloon to release the helium gas. Primary, back-up, and timed terminate signals were all used. On recovery of the mechanism, it was found that the primary terminate cutters had both fired, and the wire cable had been cut (in agreement with the telemetry information). Severe indentation on one wedge and scoring of other aluminum parts of the terminate device were noted. It was concluded that the release mechanism had become misaligned, causing it to jam and preventing separation when the wire cable was cut. Tests indicated that extreme force was required to move the wedge the observed 12 mm, and suggested that the explosive charge forced the wedge over or the cable pulled it over as it dragged across. Then the force of gravity (approximately 6000 N) jammed the wedge in the fitting as it moved laterally. For future flights, such a failure could be avoided by use of a harder grade of aluminum for the two wedges, lubrication of the parts, and modification to prevent lateral movement. Two failures of this type of terminate mechanism have occurred at NASA's National Scientific Balloon Facility in Palestine, Texas over the past ten years.

The helium valve was also recovered. It suffered no mechanical damage, and operated normally when powered up. It appeared that the ripcord plastic (attached to the inside of the balloon) had come loose and covered the valve opening. This may have been due to the use of inappropriate tape to hold the plastic in place, or to static build-up during the launch. In future, modification of the balloon destruct device may be needed to ensure that the plastic cannot cover the valve opening.

5 MANTRA results

a Measurements

Table 1 summarizes the measurements that were made during the MANTRA 1998 campaign. The first measurements of the

primary balloon flight were obtained by the two emission radiometers during ascent and consisted of about 140 spectral scans. Quine et al. (this issue) describe these measurements and the new forward estimation technique developed for the inversion of these radiances to obtain vertical profiles. This approach enabled the retrieval of mixing ratio profiles of HNO_3 , with the simultaneous fitting of ozone, CFC-11, CFC-12, N_2O , and CH_4 .

Solar occultation measurements began with the onset of sunrise, when the balloon was at 26 km. Measurements were made by SPS#1 (for constituents) during both sunrise and sunset, however, due to overheating of the instrument during sunset, only the sunrise data were useable. Melo et al. (this issue) present the sunrise NO_2 profiles retrieved during this flight. SPS#2 was suspended from one side of the gondola and made absolute radiometric measurements during the day for the derivation of J-values for O^1D and NO_2 .

The second instrument on the pointing table, optically co-aligned with SPS#1, was the AOTF spectrometer. It obtained both sunrise and sunset occultations, recording spectra from 250 to 400 nm, but using a filter with a bandpass from 360 to 390 nm. This enabled the 250–350 nm region to be used to derive the background signal, which proved useful in correcting for the temperature dependence of the RF amplifier and for sinusoidal variation in the baseline due to azimuthal oscillations across the Sun. A video camera mounted on the AOTF spectrometer confirmed this azimuthal swing, which was estimated to be as large as $\pm 0.5^\circ$. The AOTF instrument worked well, with a signal-to-noise ratio of approximately 100:1. Differential optical absorption spectroscopy has been used to derive NO_2 and OCIO slant columns as a function of solar zenith angle, from which vertical profiles of number density from 25 to 50 km were derived using an iterative least squares algorithm.

The FTS began making observations at sunrise using its own solar tracking system. However, these sunrise data were not useable due to instabilities in the dynamic alignment system caused by problems with the laser heater controller and the digital electronics. Further laser thermal instabilities encountered during sunset reduced the useful length of the scans from 50 cm to about 5-cm optical path difference. This degraded the spectral resolution to 0.23 cm^{-1} , and reduced the number of trace gases that could be retrieved from the spectra. However, all of the sunset occultation scans were recorded, and after co-adding of interferograms, nine good sunset spectra were obtained at tangent heights from 10 to 31 km. As described by Fogal et al. (this issue), vertical mixing ratio profiles of ozone, HNO_3 , CH_4 , N_2O , HCl , CFC-11, and CFC-12 were retrieved for sunset using a line-by-line, layer-by-layer spectral fitting and an "onion peeling" approach.

All three ground-based instruments worked well during the MANTRA campaign. Brewer spectrophotometer #029 made daily total ozone measurements from 5 to 26 August. In addition, some lunar observations were made when conditions were suitable, providing a number of night-time ozone columns. Vertical columns of SO_2 and relative aerosol optical

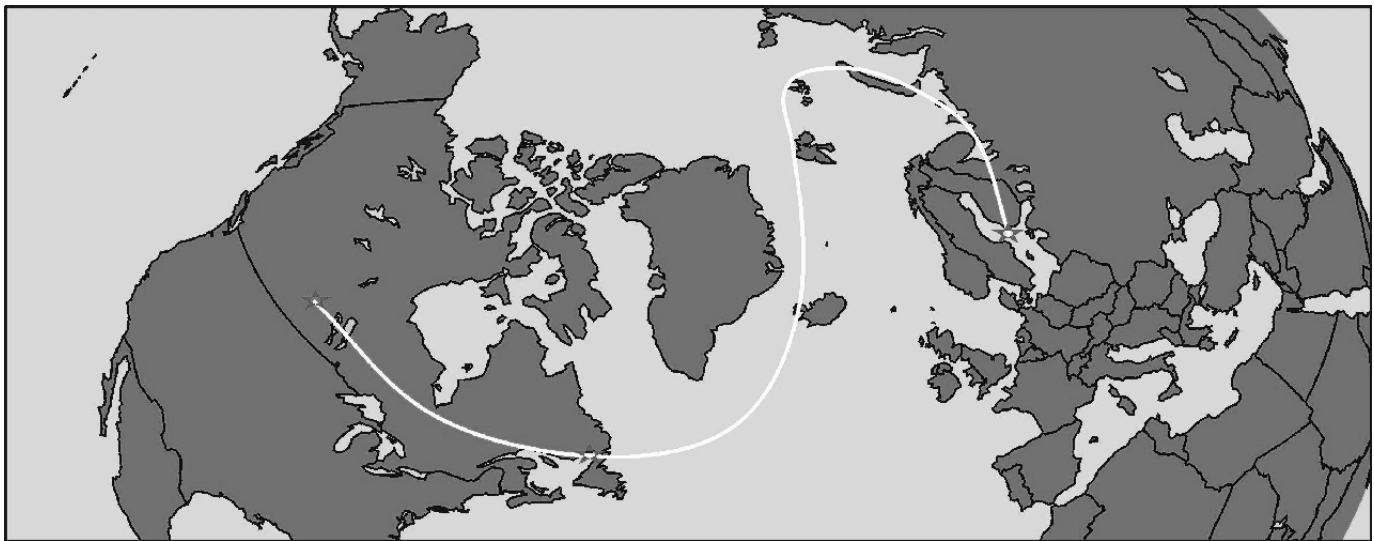


Fig. 4 The approximate path of the MANTRA 1998 balloon on its 9000-km journey.

TABLE 1. Summary of the MANTRA 1998 measurements. FTS is the University of Denver Fourier transform spectrometer, SPS#1 is the MSC photodiode array spectrometer #1 for constituents, SPS#2 is the MSC photodiode array spectrometer #2 for J-values, AOTF is the CRESTech/York University acousto-optic tunable filter spectrometer, DOAS is the University of Toronto zenith-sky grating spectrometer, GB stands for ground-based and DU stands for Dobson units.

Trace Gas	Instrument	Technique	Altitude Range (km)	Error Range (%)	Reference
Ozone	FTS	sunset occultation	12–32	20–25	Fogal et al. (this issue)
	ozonesondes	in situ chemical	0–40	10	Davies et al. (2000)
	radiometers	emission on ascent	10–36	12–100	Quine et al. (this issue)
	SPS#1	sunrise occultation	12–37	10–15	McElroy (1995), Melo et al. (this issue), C. Nowlan and C.T. McElroy (personal communication, 2005)
NO ₂	SPS#1	sunrise occultation	14–37	10–15	McElroy (1995), Melo et al. (this issue), C. Nowlan and C.T. McElroy (personal communication, 2005)
	AOTF	solar occultation	25–50	See note ^a	None
HNO ₃	DOAS	GB zenith sky	17.5–37.5	20–30	Melo et al. (this issue)
	FTS	sunset occultation	12–32	10–30	Fogal et al. (this issue)
HCl	radiometers	emission on ascent	10–30	15–100	Quine et al. (this issue)
	FTS	sunset occultation	12–32	20–25	Fogal et al. (this issue)
CFC-11	FTS	sunset occultation	12–25	20–43	Fogal et al. (this issue)
	radiometers	emission on ascent	10–22	20–100	Quine et al. (this issue)
CFC-12	FTS	sunset occultation	12–27	19–24	Fogal et al. (this issue)
	radiometers	emission on ascent	10–24	16–100	Quine et al. (this issue)
N ₂ O	FTS	sunset occultation	12–32	10–25	Fogal et al. (this issue)
	radiometers	emission on ascent	10–22	16–100	Quine et al. (this issue)
CH ₄	FTS	sunset occultation	12–32	20–40	Fogal et al. (this issue)
	radiometers	emission on ascent	10–18	15–100	Quine et al. (this issue)
J-values for O ¹ D and NO ₂	SPS#2	absolute radiometry	—	—	McElroy et al. (1995), H. Wu and C.T. McElroy (personal communication, 2005)
Aerosol optical depth	Brewer	GB direct sun	column	—	Savastiouk and McElroy (this issue)
Ozone columns	Brewer	GB direct sun	column	≤ 1	Savastiouk and McElroy (this issue)
	DOAS	GB zenith sky	column	≤ 5	Bassford et al. (this issue)
NO ₂ columns	DOAS	GB zenith sky	column	10–12	Bassford et al. (this issue)
SO ₂ columns	Brewer	GB direct sun	column	0.2–0.6 DU	Savastiouk and McElroy (this issue)

^a Estimates of NO₂ number density profiles have been derived from the AOTF solar occultation spectra, but these results require further assessment and error analysis.

depths were also derived from the Brewer spectrophotometer. Savastiouk and McElroy (this issue) describe these measurements in greater detail. The zenith-sky grating spectrometer recorded twilight and noon spectra of scattered sunlight from 18 to 25 August. These spectra were analysed using the

DOAS technique, as described by Bassford et al. (this issue) and Bassford et al. (2001), to derive vertical column amounts of ozone and NO₂. Melo et al. (this issue) also retrieved NO₂ vertical profiles from the daily slant columns. The NO₂ profile obtained at sunrise on 24 August generally agreed with

the co-located and simultaneous NO₂ profile measured by the balloon-borne SPS#1. SPS-G, the ground-based photodiode array spectrometer, was used to make daytime zenith-sky measurements of scattered sunlight from 15 to 24 August. Derivation of ozone and NO₂ columns from these spectra is continuing.

A total of fifteen radiosonde and twelve ozonesonde flights were made from Vanscoy between 6 and 24 August. In addition, ten of the twelve ozonesonde flights carried dual ozonesondes for the investigation of two different sensing solutions and their impact on the response of the sondes (see Boyd et al., 1998). These and subsequent experiments during the MANTRA 2000 campaign clearly revealed the height dependence of the slow secondary reactions, as described by Davies et al. (2000). One aerosol backscatter sonde was launched within an hour of the primary balloon launch, and provided vertical profiles of aerosol backscatter at 490 and 940 nm from the ground up to 15 km. These show a layer of enhanced backscatter in the lower stratosphere correlated with enhanced ozone that appear to be due to forest fire smoke.

The emission radiometers and the FTS measured a number of the same species, and obtained generally consistent results. Of particular interest are the HNO₃ measurements, as it was one of the primary species to be measured due to its importance as a nitrogen reservoir. Both instruments measured similar profile shapes and peak mixing ratio altitudes, but the emission radiometers obtained higher values above 20 km, with a peak mixing ratio of 9 ppbv at 24 km compared to 6.8 ppbv from the FTS, as seen in Quine et al. (this issue). As noted by Fogal et al. (this issue), this discrepancy may be the result of the large difference in spectral resolution between the two instruments, or to their sampling different paths through the latitudinal gradient in HNO₃. Five different ozone mixing ratio profiles were also obtained on 24 August: by the two emission radiometers, the FTS, SPS#1, and an ozonesonde. While these are broadly in agreement as shown by Quine et al. (this issue), there are some differences, particularly for the FTS, which measured about 20% more ozone than the sonde above 27 km; possible causes are discussed by Fogal et al. (this issue).

b Photochemical Modelling

Interpretation of the MANTRA 1998 measurements involves the use of model simulations of the stratosphere to compare retrieved and predicted trace gas abundances, as was done for the original Stratoprobe dataset. Three such models have been used to date.

The first is the MEZON (Model for Evaluating oZONe Trends) 3-D global chemistry-transport model (Rozanov et al., 1999; Rozanov and Schlesinger, 2001; Egorova et al., 2003). Melo et al. (this issue) describe how this model was used to generate NO₂ vertical profiles appropriate for MANTRA conditions. The model profile for sunrise on 24 August 1998 was compared with those retrieved from the balloon-borne SPS#1 and the ground-based zenith-sky grating

spectrometer. While the modelled and measured maximum of the NO₂ mixing ratio and its altitude agree within the measurement error bars, the model substantially underestimates NO₂ from 20- to 30-km altitude (by approximately 30% at 30-km altitude and 45% at 20 km). Comparison of MEZON sunrise and sunset NO₂ profiles from 18 to 25 August with the ground-based time series shows that the day-to-day variability in the NO₂ mixing ratio maximum is more pronounced in the observations than in the model results. Melo et al. (this issue) discuss these results and possible sources of the observed variability.

Two additional models, a photochemical box model and a general circulation model, were also compared with MANTRA 1998 measurements (S.M.L. Melo, personal communication, 2005). The former is the University of California, Irvine photochemical box-model (McLinden et al., 2000, 2001). This box model was used to derive vertical profiles of NO_y from the limited MANTRA dataset. It was locked onto the diurnal cycle of 52°N for 24 August and constrained with profiles of pressure, temperature and ozone from the ozonesonde, Cl_y and Br_y from tracer correlations with N₂O measured by the FTS, H₂O from a correlation with CH₄ measured by the FTS, and the aerosol surface area density from the Stratospheric Aerosol and Gas Experiment (SAGE) II August 1998 mean. NO_y was then derived by scaling the sum of HNO₃ as measured by the FTS and NO₂ as measured by SPS#1 with the NO_y:(HNO₃+NO₂) ratio calculated by the box model. As this ratio is dependent on NO_y, this process required three iterations and gave excellent agreement between the modelled and measured HNO₃ and NO₂ at all altitudes, including the NO₂ measured by the ground-based zenith-sky grating spectrometer at sunrise and sunset. These results indicate that the different measurements are consistent and that the box model photochemistry can adequately describe the partitioning of NO_y under MANTRA conditions. This approach yields a maximum NO_y mixing ratio of 14.3 ppbv at 35.6 km, a value that is at the low end of the wide range of estimates for NO_y in the mid-stratosphere (11 to 21 ppbv), noted in Section 1.

A different set of comparisons has been made with the Canadian Middle Atmosphere Model (CMAM), which is an upward extension of the Canadian Centre for Climate Modelling and Analysis General Circulation Model to 0.0006 hPa (roughly 100-km altitude) (Beagley et al., 1997; de Grandpré et al., 1997, 2000). The model includes a full representation of stratospheric chemistry with thirty-one non-advected species and sixteen advected species and families, and heterogeneous reactions on sulphate aerosols, stratospheric ternary solutions, and water ice, without sedimentation. For the version used here, prognostic variables were computed in spectral space using T32 resolution (corresponding to about 6° latitude and longitude) and sixty-five vertical levels (about 2-km resolution in the middle atmosphere). The goal of this study was to compare CMAM chemical fields at the nearest grid point to Vanscoy to the MANTRA measurements. For CMAM run in climate mode, the data cannot be compared in a day-by-day manner, but only statistically. The

CMAM has a stable chemical climate over 20-year simulations, which can be used to evaluate the representation of processes in the model, to assess how representative the MANTRA measurements are of the mean mid-latitude stratosphere, to test assumptions about variability of different chemical species, and to study the nitrogen partitioning at northern mid-latitudes during summer.

An extensive set of comparisons between CMAM fields and MANTRA measurements from the campaigns of 1998, 2000 and 2002 has been undertaken (S.M.L. Melo, personal communication, 2005). Comparisons with the MANTRA 1998 FTS sunset profiles, particularly those for the long-lived species N_2O and CH_4 , illustrate the sensitivity of the CMAM fields to the vertical eddy diffusion coefficient (K_{zz}), which determines the distribution of long-lived species. Reducing it from $1 \text{ m}^2 \text{ s}^{-1}$ (as used in the World Meteorological Organization (WMO) version of CMAM as described by Austin et al., 2003) to $0.1 \text{ m}^2 \text{ s}^{-1}$ significantly improved the agreement between CMAM and the measurements. CMAM also reproduces the MANTRA temperature and ozone measurements very well, indicating that CMAM climatology is representative of the northern hemisphere mid-latitude late summer stratosphere. For NO_2 , CMAM tends to agree with the sunrise NO_2 measurements, but is somewhat higher than the NO_2 measured at sunset. The MANTRA measurements are consistent with the compact correlation between CH_4 and N_2O seen in the lower stratosphere in the Atmospheric Trace Molecule Spectroscopy (ATMOS) experiment measurements (Michelsen et al., 1998), which is well reproduced by CMAM. However, CMAM appears to overestimate the $\text{NO}_y:\text{N}_2\text{O}$ correlation for mid-latitudes and overestimates NO_y during the mid-latitude summer above 20 km. The complete sunrise and sunset partitioning of NO_y from CMAM to discuss possible explanations (S.M.L. Melo, personal communication, 2005).

6 Summary and future plans

The first MANTRA balloon campaign took place in Vanscoy, SK in August 1998. The primary balloon was launched on 24 August and represented the first Canadian launch of a large high-altitude balloon in approximately fifteen years. It carried a suite of instruments to measure stratospheric composition, making measurements on ascent and from a float altitude of approximately 35 km for one day. Although not all of the instruments worked as intended, a useful dataset was collected, as summarized in Table 1. Vertical mixing ratio profiles of ozone, NO_2 , HNO_3 , HCl , CFC-11 , CFC-12 , N_2O , and CH_4 were retrieved by the combination of balloon-borne instruments (two emission radiometers, SPS#1, FTS, and the AOTF spectrometer). J-values for O^1D and NO_2 have been retrieved from SPS#2. Aerosol backscatter profiles were measured by a backscatter sonde, while a series of ozonesondes launched during the two weeks prior to the main launch provided profiles of ozone, temperature, pressure, humidity, and winds. Total column measurements of ozone, NO_2 , SO_2 , and aerosol optical depth were made by the three ground-based

spectrometers (Brewer, SPS-G, zenith-sky grating spectrometer) for the duration of the campaign. The combination of ground-based and balloon-borne measurements provided a unique opportunity to evaluate the ability to retrieve vertical profiles of NO_2 from the ground-based zenith-sky spectra. The MANTRA 1998 measurements are discussed in the accompanying papers by Bassford et al., Fogal et al., Melo et al., Quine et al., and Savastiouk and McElroy, and also by Davies et al. (2000).

The MANTRA 1998 data have been compared with three atmospheric models: the MEZON chemical transport model, the University of California at Irvine photochemical box model, and the CMAM. MEZON was used to generate NO_2 vertical profiles that were compared with those measured by SPS#1 and the ground-based zenith-sky grating spectrometer (Melo et al., this issue). The box model was used to derive vertical profiles of NO_y from the MANTRA data giving excellent agreement between the modelled and measured HNO_3 and NO_2 at all altitudes, indicating consistency between the measurements by the FTS, SPS#1, and the ground-based zenith-sky spectrometer. The box model photochemistry also adequately describes the partitioning of NO_y under MANTRA conditions, and gives a maximum NO_y mixing ratio of 14.3 ppbv at 35.6 km. Comparisons with CMAM show that it reproduces the MANTRA temperature and ozone measurements very well, indicating that CMAM climatology is representative of the northern hemisphere mid-latitude late summer stratosphere. These comparisons also revealed the sensitivity of the long-lived species to the vertical eddy diffusion coefficient used in CMAM. CMAM also appears to overestimate NO_2 and NO_y during the mid-latitude summer above about 20 km.

While this paper and the accompanying papers in this issue of *Atmosphere-Ocean* focus on the MANTRA 1998 campaign, it is the first of a series of MANTRA balloon flights to study the odd-nitrogen budget of the northern hemisphere mid-latitude stratosphere. Three additional campaigns have since been conducted at Vanscoy, in 2000, 2002, and 2004, with a fifth one tentatively planned for 2006. All have taken place in late summer, at fall turnaround, which is a time when dynamical variability is minimized in the stratosphere and stratospheric winds change direction from westward to eastward, as discussed by Wunch et al. (this issue).

The primary objective of the 2000 balloon flight was an engineering test flight of a new pointing control system in order to demonstrate its performance and capabilities for future balloon flights. It was launched on 29 August 2000 and carried a small payload, consisting of the pointing control system, two emission radiometers and one SPS, from which vertical profiles of ozone, NO_2 , and HNO_3 have been retrieved. On this, as on all the campaigns, additional column and profile measurements were made by ground-based spectrometers, ozonesondes, and aerosol backscatter sondes. The pointing control system achieved one solar occultation and one limb scan observation, and demonstrated an accuracy of 0.1° (1σ) in elevation and 3° (1σ) in azimuth, as described by Quine et al. (2002).

The MANTRA 2002 and 2004 campaigns both involved the flight of large payloads. The 2002 payload, launched on 3 September, again included two emission radiometers, the OH spectrometer, SPS#1, SPS#2, and the Denver FTS, as well as three additional instruments: another FTS, a Système d'Analyse par Observations Zénithales (SAOZ) spectrometer, and the new Measurement of Aerosol Extinction in the Stratosphere and Troposphere Retrieved by Occultation (MAESTRO)-B dual spectrometer (Strong et al., 2003). In 2004, all of these instruments were flown once more (on 1 September), with the further addition of an airglow infra-red radiometer and a third FTS, the new Portable Atmospheric Research Interferometric Spectrometer (PARIS-IR). A separate SAOZ-BrO flight took place on 24 August, while the ground-based component of the 2004 campaign involved measurements by six spectrometers from 3 August to 15 September.

Analysis of data from these campaigns is complete for some instruments and continuing for others, and will be the subject of future publications. Of the four scientific objectives of the MANTRA project, the first (to measure vertical profiles of the key stratospheric species that control the mid-latitude ozone budget) has largely been accomplished, although it has not been possible to retrieve all relevant species. Work is also continuing to improve the retrieval algorithms for some of the instruments. The second objective (to combine these measurements with historical data to quantify changes in the chemical balance of the stratosphere, with a focus on the odd-nitrogen budget) is currently being pursued. While the accurate detection of trends will be difficult, MANTRA is contributing to the body of knowledge needed to address the issue of changing stratospheric composition. In particular, we have the raw emission radiometer data from flights conducted by MSC in 1989, 1990, 1991 and 1992 (two per year) and intend to reanalyse these data with the new algorithm developed for MANTRA. Combining all of the radiometer data will produce eight years of coverage between 1989 and 2004, allowing for a careful assessment of the late summer northern mid-latitude stratospheric HNO₃ budget during this period. The third MANTRA

objective (to compare measurements of the same species recorded by different instruments) is well underway; some discrepancies have been observed between the 1998 measurements and the later campaigns are being used to investigate these. The fourth objective (to use the measurements for satellite validation) is also in progress, with comparisons to the Optical Spectrograph and InfraRed Imager System (OSIRIS) and Sub-Millimeter Receiver (SMR) on Odin, to the Global Ozone Monitoring by Occultation of Stars (GOMOS), Michelson Interferometric Passive Atmosphere Sounder (MIPAS) and Scanning Imaging Absorption Spectrometer for Atmospheric Chartography (SCIAMACHY) on ENVISAT, and to the Atmospheric Chemistry Experiment Fourier Transform Spectrometer (ACE-FTS) and MAESTRO on SCISAT-1 all underway.

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