
Infra-red FTS Measurements of CH₄, N₂O, O₃, HNO₃, HCl, CFC-11 and CFC-12 from the MANTRA Balloon Campaign

P. F. Fogal*, R. D. Blatherwick, F. J. Murcray and J. R. Olson

*Department of Physics and Astronomy
University of Denver, 2112 E. Wesley Avenue
Denver, CO 80208 USA*

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ABSTRACT *The Middle Atmosphere Nitrogen TRend Assessment (MANTRA) campaign is intended to address the question of whether possible changes in the mid-latitude nitrogen budget can account for discrepancies between predicted and observed ozone loss at these latitudes. In addition, MANTRA seeks to establish consistency between old and new measurement techniques. We report here the results of infra-red spectral measurements made with a high resolution Fourier Transform Spectrometer (FTS) during the August 1998 MANTRA flight. Vertical mixing ratio profiles of methane, nitrous oxide, ozone, nitric acid, hydrogen chloride, CFC-11 and CFC-12 are presented. The error estimates on the retrieved values are larger than typical for this type of measurement, due mostly to instrument problems that occurred during flight. In addition, the CFC values are larger than expected. Possible reasons for this are discussed.*

RESUMÉ [Traduit par la rédaction] *La campagne d'Évaluation des tendances de l'azote dans l'atmosphère moyenne (MANTRA) vise à déterminer si des changements possibles dans le bilan de l'azote aux latitudes moyennes peuvent expliquer les écarts entre les pertes prédites et observées d'ozone à ces latitudes. De plus, le projet MANTRA cherche à assurer une cohérence entre les anciennes et les nouvelles techniques de mesure. Nous signalons ici les résultats des mesures spectrales infrarouge faites au moyen d'un spectromètre à transformée de Fourier (FTS) à haute résolution durant le vol MANTRA du mois d'août 1998. Nous présentons les profils de rapport de mélange vertical du méthane, de l'oxyde nitreux, de l'ozone, de l'acide nitrique, du chlorure d'hydrogène, du CFC-11 et du CFC-12. Les estimations d'erreurs sur les valeurs extraites sont plus élevées que d'ordinaire pour ce type de mesure, principalement à cause de problèmes d'instruments survenus durant le vol. Les valeurs de CFC sont en outre plus élevées que les valeurs attendues. De causes possibles pouvant expliquer ce résultat sont avancées.*

1 Introduction

It has been established that there is a decline in mid-latitude ozone (World Meteorological Organization, 1999). Current photochemical models are unable to account for all of the observed ozone loss at mid-latitudes (Marchand et al., 2003). The cause of this loss is as yet undetermined, but one possible explanation is a change in the odd nitrogen budget ($\text{NO}_y = \text{NO} + \text{NO}_2 + \text{NO}_3 + 2 \text{N}_2\text{O}_5 + \text{ClONO}_2 + \text{HNO}_3 + \text{HO}_2\text{NO}_2 + \dots$), or in the partitioning between $\text{NO}_x (= \text{NO} + \text{NO}_2)$ and the reservoirs HNO_3 and ClONO_2 . While this is the proposed mechanism most easily evaluated by our measurement capabilities, other mechanisms are also possible. They include changes in intensity of the Brewer-Dobson circulation and a subsequent effect on diabatic descent, and transport of ozone-poor air from the polar regions (Vaugh et al., 1994; Marchand et al., 2003).

A primary objective of the Middle Atmosphere Nitrogen TRend Assessment (MANTRA) campaign is to address the question of a possible change in any of the members of the

odd nitrogen family. A second objective is to provide a convincing and coherent connection to measurements made years ago by the same or similar instruments, particularly those that flew together during such balloon campaigns as the Balloon Intercomparison Campaign (BIC). The University of Denver (UDenver) 0.01 cm^{-1} resolution Fourier Transform Spectrometer (FTS), fabricated by Bomem, Inc. of Québec City, QC, Canada, had flown as part of BIC and as part of other campaigns before and after BIC. It has also been used extensively as a ground-based instrument at many locations including Fairbanks, AK and the South Pole. Establishing the consistency of measurements made with older instruments and those made using more modern techniques is crucial to ensuring that any observed changes in the state of the atmosphere over time are not merely instrumental artefacts. This activity is tied directly to the primary goal of the MANTRA experiment, the determination of a possible change in the

*Corresponding author's e-mail: pierre.fogal@utoronto.ca; current affiliation-University of Toronto, Department of Physics, 60 St. George St., Toronto ON M5S 1A7

NO_y budget, by essentially validating all of the applicable measurements made during the intervening period. Further details about the MANTRA concept are discussed in Strong et al. (this issue).

2 Instrumentation

An infra-red FTS, (sometimes referred to as an FTIR when operated in the infra-red) is a particularly capable instrument. It can provide high resolution spectra over a relatively wide bandpass. The atmospheric mid infra-red (IR) spectrum provides a wealth of spectral features present in molecular vibrational-rotational bands, and most atmospheric constituents have spectral features somewhere in it. FTSs are often operated in transmission mode, in which they use solar radiation as a source, and the atmosphere acts as the sample to be measured. Atmospheric spectral features then appear as absorptions in the solar signal. Because the Sun acts as a very hot black body to a good approximation, this provides a situation in which high signal-to-noise ratio (SNR) measurements with very good sensitivity are typical, allowing the analysis of small absorptions. The result is an instrument package that, depending on its configuration, can sample a large number of molecules important to the study of the atmosphere. The geometry of occultation measurements made from a balloon platform provides significant path enhancements, resulting in further sensitivity to absorptions by trace gases along the line of sight. One minor drawback to operation in the solar transmission mode, is the presence in the measured atmospheric spectrum of spectral features due to transitions that occur in the Sun's own atmosphere. These features sometimes interfere with atmospheric features of interest, but in this experiment they have not.

The UDenver FTS used for this experiment was a BOMEM DA2 Michelson type interferometer-spectrometer with two-inch input optics. This FTS began its operational life in 1977 as a single channel instrument, and has since been modified to sample two detector channels. The maximum optical path difference was 50 cm, resulting in an unapodized Full Width at Half Maximum (FWHM) resolution of 0.01 cm^{-1} . However, as discussed in the following, during part of the flight, scanning problems resulted in a need to truncate the interferograms resulting in a resolution of approximately 0.2 cm^{-1} (apodized). The scan time was about 90 seconds, and data were recorded in both forward (Zero Path Difference (ZPD) at the beginning of scan) and reverse (ZPD at the end of scan) directions. Optical alignment was maintained during the scans using the BOMEM dynamic alignment system, in which a dual axis servo system maintains the phase relationship on a set of photo diodes illuminated by the sampling laser. The output beam from the interferometer was divided by a dichroic beamsplitter that directed the long wavelength part of the IR signal onto an HgCdTe (henceforth, MCT) detector and the short wavelength part onto an InSb detector. The sampling interval, digital and optical filters were selected so that the bandpasses measured were $700\text{--}1300 \text{ cm}^{-1}$ in the MCT channel, and $2650\text{--}3250 \text{ cm}^{-1}$ in the InSb channel.

This instrument is one of the earliest commercial dynamical alignment systems with a 2-in aperture (DA2). It should be noted that as for most of the MANTRA instruments, it had been a number of years since it was last deployed as part of a balloon-borne instrument package, and this was its first flight in the two-detector configuration. A number of other modifications had been carried out, primarily to the electronics subsystems. It should also be noted that spectral analysis techniques have progressed somewhat in the intervening years. This instrument is similar in its general operating principals to that described by Murcray et al. (1990). Results from flights in its original single channel configuration are documented in Murcray et al. (1980), Blatherwick et al. (1980), and Goldman et al. (1981).

The input solar beam was maintained on the interferometer window using a biaxial solar tracking telescope designed and constructed at UDenver. Data were recorded onboard using a custom personal computer writing to a solid state hard disk. The FTS and ancillary hardware were controlled from the ground via a series of telecommands sent from the command centre allowing cycling of the computer, data storage, FTS, FTS heaters, FTS laser and the solar tracking telescope.

3 Flight notes

The MANTRA payload was launched at 03:25 local time, 24 August 1998 from the Environment Canada launch site at Vanscoy, Saskatchewan (52°N , 107°W) by launch services contractor Scientific Instrumentation Limited (SIL). The payload reached a float altitude of 38.8 km at approximately 07:15, shortly after sunrise. As a result of cold soaking for nearly three hours prior to sunrise, the interferometer laser feedback system experienced some problems with the laser heater controller before and during sunrise, causing instabilities in the dynamic alignment system. In addition, apparent problems with the digital electronics during this time resulted in byte magnitude errors. Consequently, the data recorded during sunrise are unusable.

The payload remained at float altitude throughout the day. (By sunset, the float altitude had decreased to about 31 km.) FTS data were recorded at high sun for about one hour, and the instrument was then turned off to conserve power and disk space. The instrument was powered up about 45 minutes before the onset of sunset. Unfortunately, the FTS experienced less than optimal performance of the dynamic alignment system that attempts to maintain (parallel) alignment between the moving and fixed mirrors, resulting in the useful length of the scans being only about 5 cm. At that point, the momentary misalignment caused interferogram points to be lost resulting in the need to truncate the interferograms to that length. However, all of the sunset occultation scans were recorded, and only one pair was completely corrupted.

4 Analysis

One of the assumptions in FTS theory is that the source and sample remain constant. If either is changing, the interferogram

is subject to a smearing effect, in which the measured interferogram is not the Fourier transform of a single spectrum. To mitigate the effects of interferogram smearing due to the air-mass changing during the time required to record a scan, we routinely average (henceforth co-add) phase corrected interferograms in reverse-forward pairs that share the same ZPD time (within about 2 seconds). This procedure was followed here, resulting in ten such spectra recorded during sunset. Table 1 lists the time, solar zenith angle, and tangent height of the sunset sequence.

The spectra were analysed by fitting synthetic spectra generated with the UDenver line-by-line, layer-by-layer computer RADiation COde (RADCO) (Blatherwick et al., 1989) to the observed spectra. Figures 1 and 2 show typical fits to the spectra produced with RADCO. Mixing ratio profiles were generated from the sunset spectra using the “onion peeling” technique, in which the mixing ratio of the retrieved molecule at the highest tangent height and above is determined first (by scaling the initial profile until the synthetic spectrum matches the observed), and thereafter held fixed. The mixing ratio for that species is then determined for the next highest tangent height, and so on, in an iterative manner. If the retrieval gas or any interfering species are diurnally varying (such as NO or NO₂), then one must adjust the previously determined mixing ratios as a function of time using a photochemical model. The temperature profile and pressure used as input to RADCO were based on radiosonde data for the day of the flight, and the line parameters were taken from the HIGH-resolution TRANsmiSSion database (HITRAN96) (Rothman et al., 1998).

Table 2 summarizes the volume mixing ratio (vmr) profiles resulting from the analyses. The numbers in parentheses are the measurement uncertainties estimated from uncertainties in the RADCO inputs (the pressure, temperature profile and the line parameters) and in the sensitivity of the quality of fit to variations in the retrieved mixing ratio (i.e., the mixing ratio in the tangent layer). The altitudes are the pressure weighted altitudes in the layers defined by the tangent heights. These errors are larger than normal for this type of measurement, and are a further manifestation of the instrument problems that occurred during the sunset occultation.

5 Results and discussion

Although the performance of the FTS was below optimal, we were still able to derive atmospheric profiles for a number of constituents. Figure 3 shows the vmr for HNO₃ and ozone measured during sunset. HNO₃ is of particular interest because of its role as a nitrogen reservoir. Our HNO₃ results are consistent with previous mid-latitude measurements, although the peak of 6.8 ppbv is somewhat less than that derived by Quine et al. (this issue) using spectra from the Meteorological Service of Canada - University of Toronto (MSC-UofT) filter radiometers that also flew on the MANTRA gondola during this flight. There are several reasons that might explain the difference. First of all, the radiometer measurements are recorded at a much lower reso-

TABLE 1. Summary of occultation times and geometry for FTS measurements.

Time of Scan (UT)	Solar Zenith Angle (degrees)	Tangent Altitude (km)
02:14:54	90.03	31.23
02:25:37*	91.55	28.82
02:28:16	91.92	27.54
02:31:00	92.30	26.05
02:33:40	92.67	24.34
02:39:00	93.42	20.15
02:41:40	93.79	17.71
02:44:21	94.16	15.28
02:49:43**	94.90	10.53

*The three scans following this one were recorded, but were not used in the profile generation because of the relatively small airmass changes between them.

**The scan at 02:47:00 was corrupted and could not be used.

lution, which makes it difficult to separate signal from overlapping features. The HNO₃ band has interference from both water vapour and CFC-11. It should be noted that the radiometer measurements are based on fitting the overall shape of the ν_9 band, whereas the FTS measurement deals with individual HNO₃ manifolds. Also, the radiometers recorded their spectra on ascent approximately 18 hours earlier. While a diurnal variation is not expected, there is a latitudinal gradient, and as the gondola was not oriented during darkness it is possible that the radiometers might have seen that gradient.

The ozone is in good agreement with the ozonesonde measurement for the same day except above 27 km, where our measurements are about 20% larger. In general, discrepancies at higher altitudes may result in part from the fact that the sonde itself begins to suffer from pump inefficiencies. The ozone retrievals from the Sun PhotoSpectrometer (SPS) instrument (Davies et al., 2000) also show similar values of ~8 ppm at 30 km, although their profile does not peak as sharply beyond that. It should be noted that the SPS profile was derived for the sunrise occultation and the sonde was flown in the early afternoon. Also shown is a HALogen Occultation Experiment (HALOE) ozone profile retrieved for 44.4°N, 97.6°W on 29 August 1998 (Russell et al., 1996). Our values agree very well with HALOE except for the point at 29.5 km.

Figure 4 shows the N₂O and CH₄ profiles retrieved for the flight. The peak in the CH₄ profile, and to a lesser degree, the N₂O profile, near 29.5 km appears to be real, and occurs at the same altitude as the peak in the ozone profile. Similar peaks have been seen by ourselves and others in mid-latitude measurements of these molecules, generally earlier in the season. The HALOE CH₄ profile for 44.4°N, 97.6°W on 29 August 1998 is also plotted on this figure and compares quite well to our measurements at altitudes below 25 km. The HALOE profile exhibits a change in slope above 25 km, slightly lower than the peak in our measurements. The N₂O and CH₄ retrieved profiles have been studied in conjunction with the Canadian Middle Atmosphere Model (CMAM) (Stella Melo, personal communication, 2004). The CMAM output for these two molecules compares very well to the retrieved profiles.

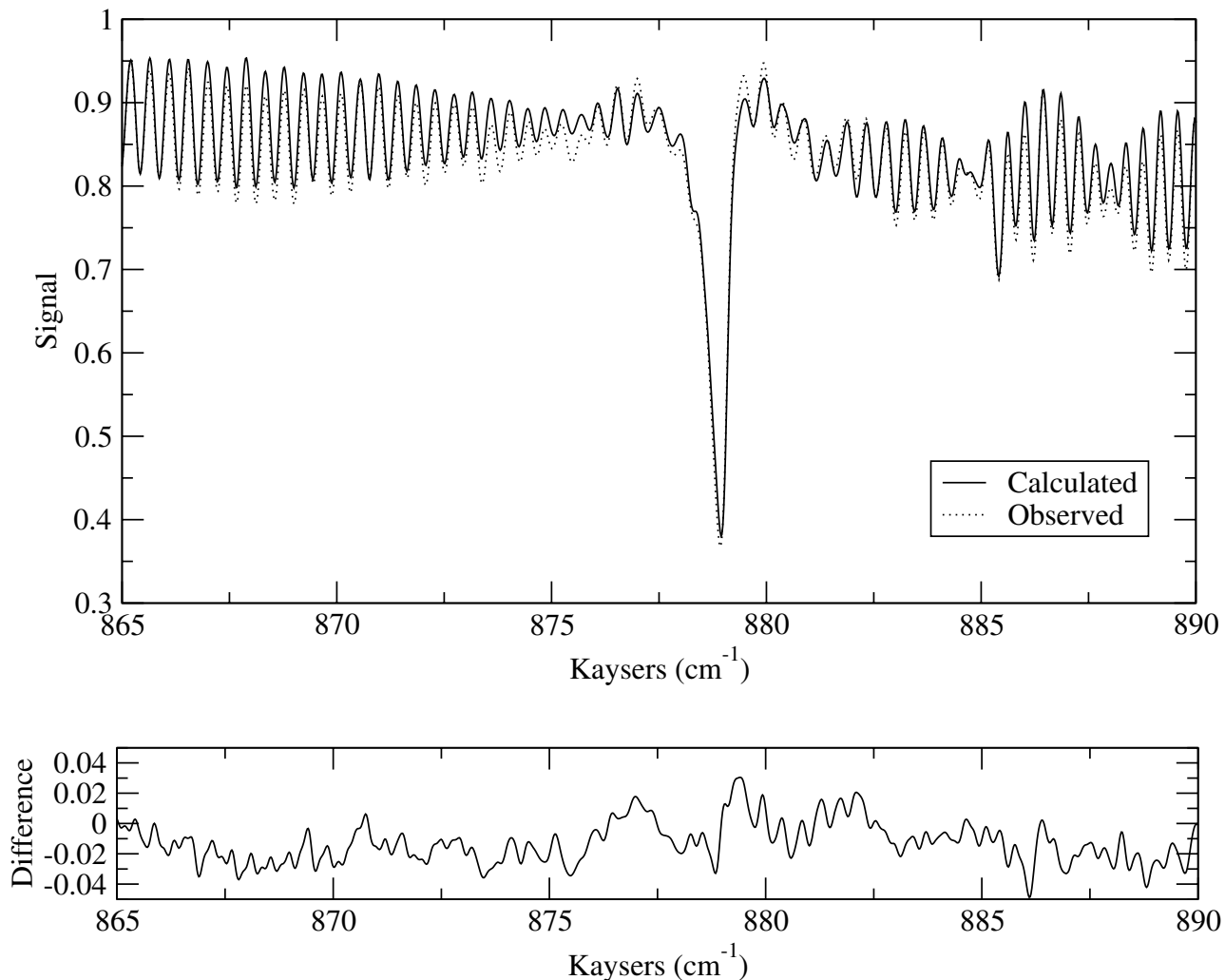


Fig. 1 Comparison of calculated spectrum to observed spectrum for a portion of the ν_9 band of HNO_3 near 11 μm .

Figure 5 shows the CFC-11, CFC-12, and HCl profiles retrieved from this flight, as well as the HALOE profile for the date and location mentioned previously. The profile for CFC-11 extends only to 23.7 km, and for CFC-12 to 25.7 km because the spectral signatures become too small to be useable above those altitudes due to CFC photolysis. The retrieved vmrs for these two gases seem somewhat high below 23 km, but agree reasonably well with the values measured with the MSC-UofT radiometers (Quine et al., personal communication, 2004). Cunnold et al. (1997) report a CFC-11 tropospheric vmr of approximately 0.265 ppm and a growth rate of essentially zero since 1992. As CFC-11 is long-lived in the troposphere and lower stratosphere, our lower stratospheric value of 0.29 (± 0.058) ppm is consistent with those values.

Engel et al. (1998) have calculated temporal trends for CFC-12 based on cryogenic sampler measurements made during balloon flights launched from Aire-sur-l'Adour in France (44°N) and Kiruna, Sweden (68°N) from 1978 to

1997. They have produced trends for CFC-12 versus N_2O for N_2O vmrs of 310, 250, and 200 ppb. From their calculated trends, it is easy to see that our value, at 308 ppb N_2O , is rather larger than the value extrapolated from their trend is likely to be. It is interesting to note that the slope decreases significantly at the end of their fit and that the change is driven primarily by the 1997 measurement. At 250 ppb N_2O , we show little change from our 310 ppb N_2O value, and even at 200 ppb N_2O our measurement remains about 10% larger than the Engel et al. (1998) values.

Possible explanations for the differences between our measurements and those of Engel et al. (1998) include: latitudinal differences, measurement based differences between the two techniques and actual differences in the atmosphere that might transport tropospheric air into the lower stratosphere. Latitudinal differences can probably be ruled out as the two sites used in the Engel et al. (1998) analyses nicely bracket our Saskatoon site. To investigate atmospheric differences we turned to the ozonesonde flights flown as part of the

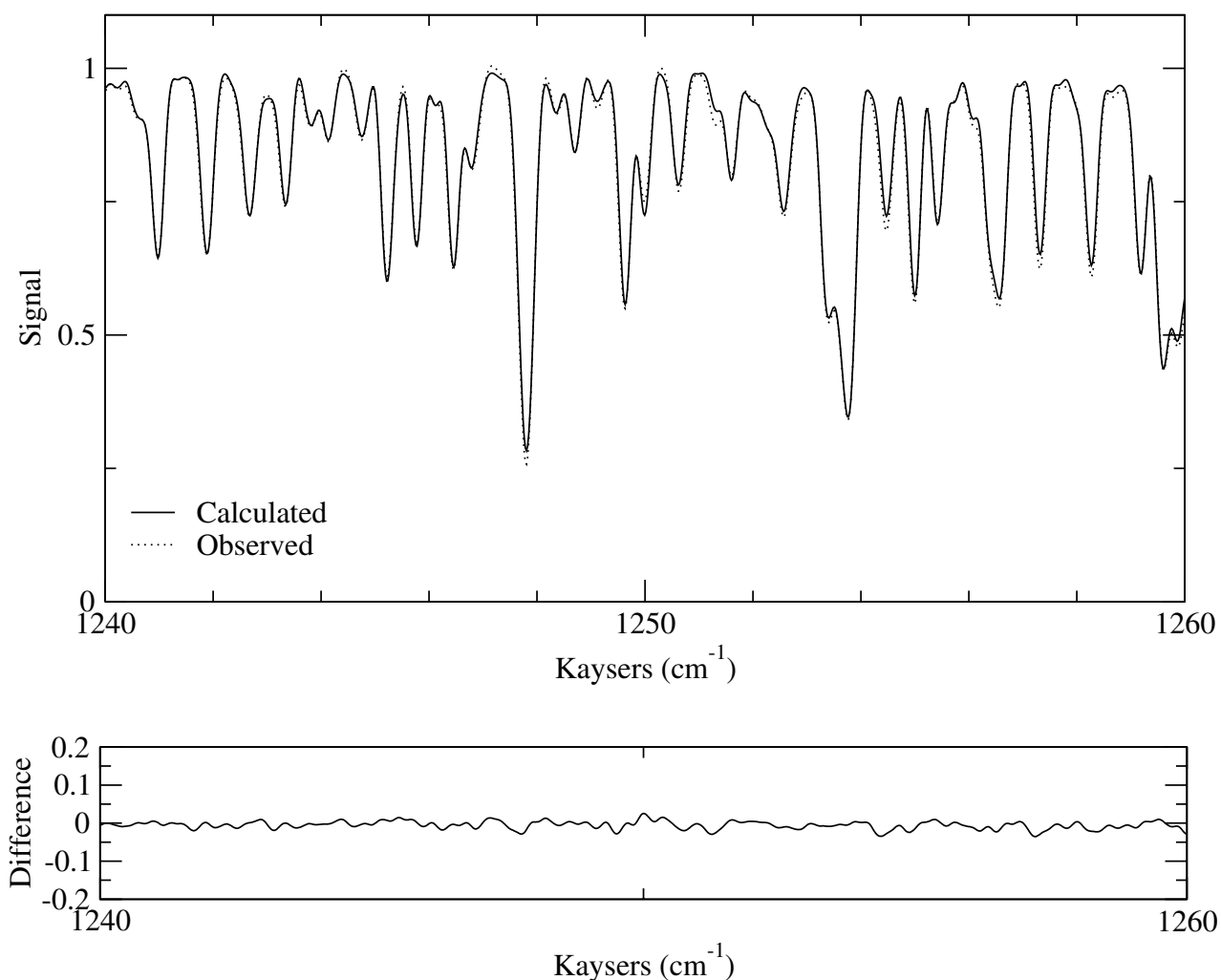

 Fig. 2 Comparison of calculated spectrum to observed spectrum for a portion of the CH₄ band near 8 μm .

TABLE 2. Mixing ratio profiles retrieved in this experiment. Absolute uncertainties are in parentheses.

Altitude (km)	HNO ₃ (ppbv)	HCl (ppbv)	CFC-11 (ppbv)	CFC-12 (ppbv)	N ₂ O (ppbv)	CH ₄ (ppmv)	O ₃ (ppmv)
11.82	0.50(0.15)	0.09(0.02)	0.29(0.058)	0.60(0.13)	308(31)	1.68(0.34)	0.20(0.5)
16.01	1.52(0.38)	0.35(0.09)	0.26(0.050)	0.65(0.14)	289(30)	1.56(0.32)	0.67(0.17)
18.45	4.88(0.73)	1.27(0.32)	0.23(0.050)	0.63(0.12)	260(39)	1.48(0.29)	1.53(0.30)
20.79	5.99(0.90)	0.99(0.20)	0.16(0.030)	0.42(0.08)	215(32)	1.32(0.27)	2.42(0.48)
22.89	6.55(0.65)	1.00(0.20)	0.13(0.030)	0.34(0.08)	167(25)	1.17(0.23)	3.60(0.72)
24.87	6.81(0.68)	0.93(0.19)	0.07(0.030)	0.28(0.07)	128(20)	1.03(0.20)	4.74(0.95)
26.52	5.39(0.80)	1.06(0.21)	–	0.22(0.05)	89(14)	0.82(0.17)	5.71(1.1)
27.95	4.82(0.72)	0.98(0.24)	–	–	72(14)	0.67(0.17)	5.44(1.1)
29.52	3.93(0.78)	1.26(0.31)	–	–	70(17)	0.96(0.24)	8.90(1.7)
32.05	1.99(0.50)	1.70(0.42)	–	–	28(7)	0.30(0.12)	7.80(2.0)

MANTRA campaign. If we investigate the behaviour of the ozonesonde flight records in the days prior to the launch of the main balloon, we see indications of a layer near 12 km. This behaviour is apparent for both temperature and ozone vmr. Figure 6 shows the temperature structure and its development from the 18 August until 24 August. Figure 7 depicts the same interval for ozone vmr. In both cases, a well-defined

layer becomes apparent on 21 August. This layer appears to broaden somewhat on 24 August. In the case of ozone vmr, this feature, coupled with the measured profile values, would seem to indicate a descent of a layer having a thickness of about 1 to 2 km, that has descended from approximately 16 km to around 12.5 km. The measured temperature profile shows a sharp inversion at the tropopause that would suggest that

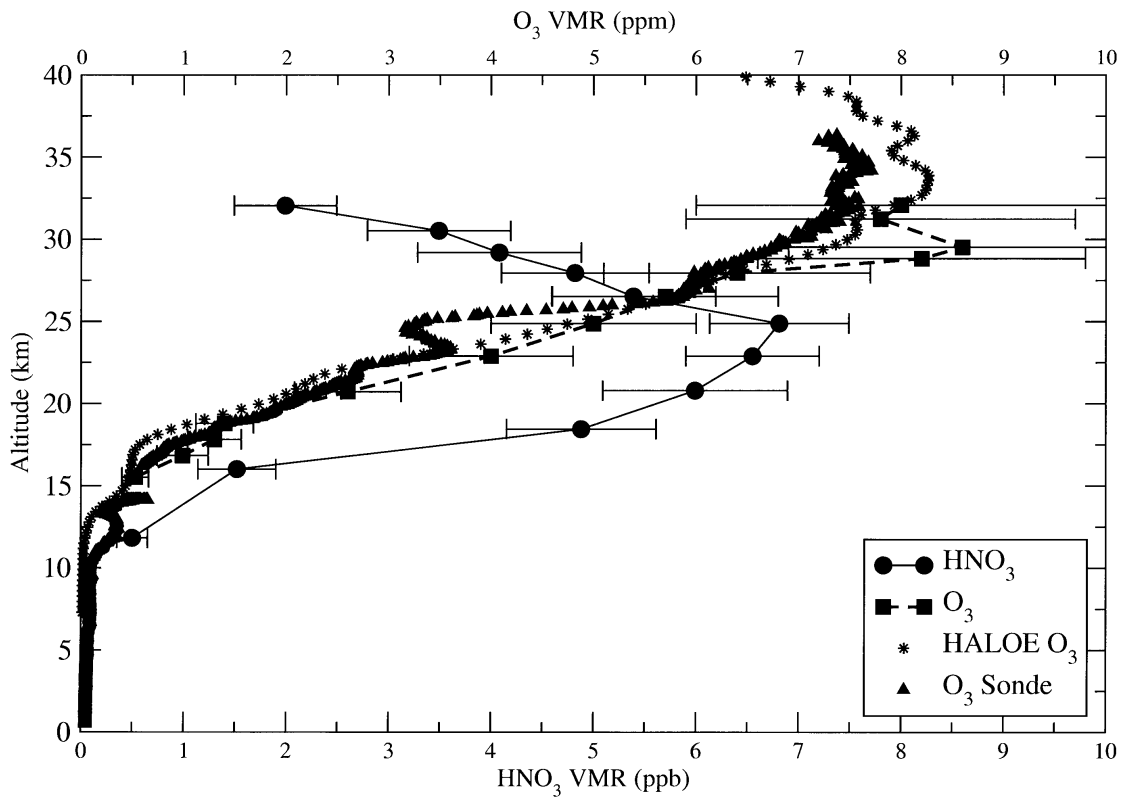


Fig. 3 Ozone and HNO₃ volume mixing ratio profiles.

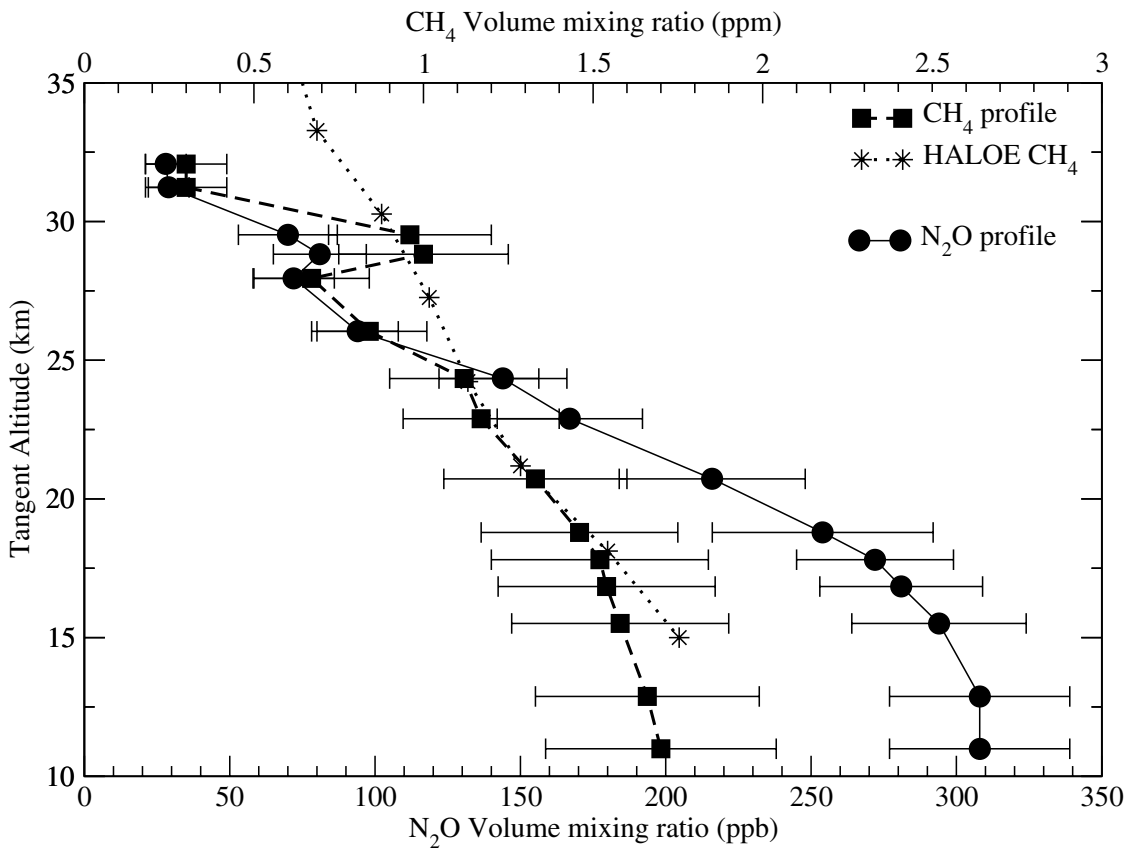


Fig. 4 N₂O and CH₄ retrieved profiles compared to nearby HALOE profiles.

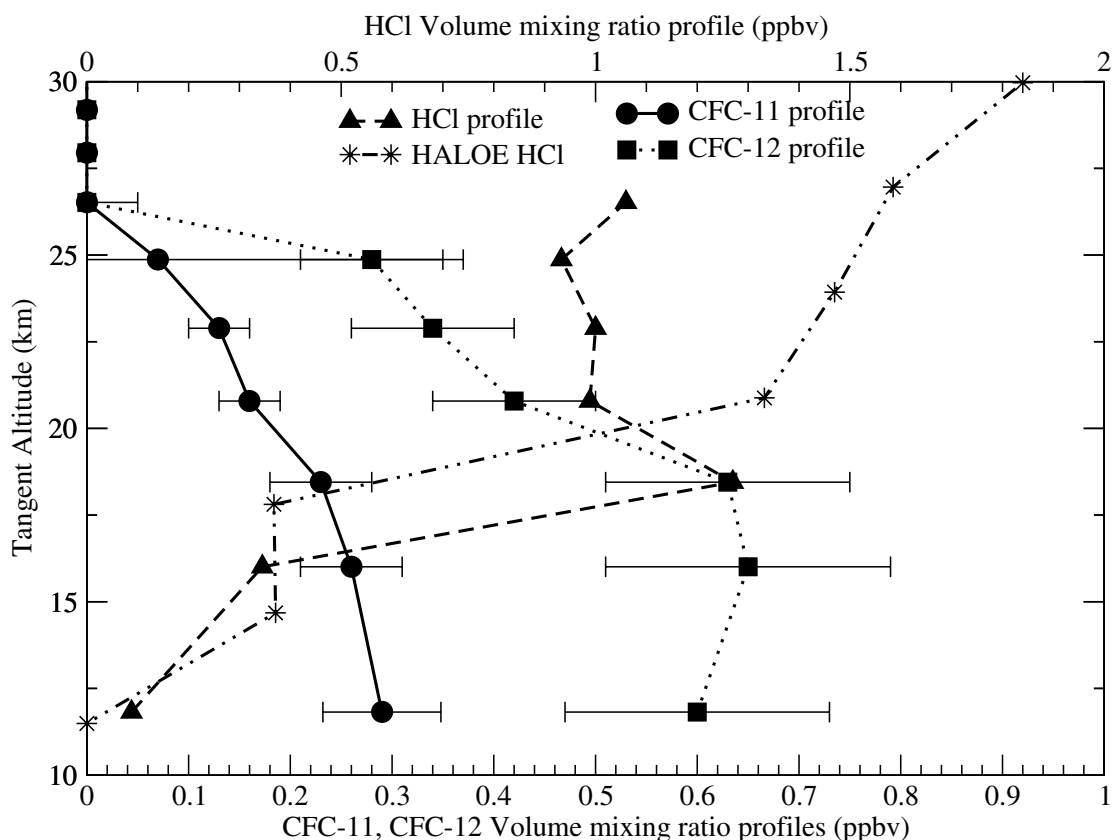


Fig. 5 CFC-11 and CFC-12 volume mixing ratio profiles shown along with the HCl profile. An HCl profile from HALOE is also shown for 44.4°N, 97.6°W on 29 August 1998.

there is, on those days, a definite boundary with little mixing across it. The small peak above tends to be 3 to 5 K warmer than the atmosphere at 16 km. It is interesting to note that from 18 August to 20 August the tropopause is not nearly so well defined. One might expect the observed enhanced CFC values in the lower stratosphere to correlate with a tropospheric fold or other upwelling tropospheric event. It is not readily apparent from the sonde data that such an event occurred, so this particular question remains open. As to differences between the two techniques, Hartmann et al. (1997) present a comparison of retrieved profiles for CFC-12 from an FTS that is very similar to ours and the in situ sampler data used by Engel et al. (1998). The agreement is deemed “satisfactory”, and we must conclude that there is little likelihood of a significant systematic difference between the two measurement techniques.

As to the MANTRA objective of creating a link between old instruments and measurements and newer measurement techniques, consider that despite being one of the earliest balloon-borne FTS instruments, the UDenver BOMEM Balloon-borne Dynamic Alignment 2-in aperture (BBDA2) is a rather more modern approach to balloon spectroscopic measurements than is the MSC-UofT filter radiometer. As such, the comparisons between the FTS and the radiometers allow new modelling and interpretation techniques to be developed to

analyse the current radiometer dataset. These techniques will then be applied to the historical radiometer data record and should allow them to be interpreted on a comparable basis to the modern measurements. The radiometers are the only other infra-red instruments on the gondola, but there is still interest in comparing the infra-red measurements to those operating in the ultraviolet-visible part of the spectrum (e.g., Davies et al. (2000)) and these comparisons will, in turn, be compared to earlier comparisons between these two classes of instruments. The primary difference between FTS measurements today and the late 1970s when this instrument first flew, is one of resolution. Unfortunately, the in-flight problems that resulted in greatly decreased resolution exacerbated this problem to such an extent that comparisons of retrievals at our intended apodized resolution of 0.02 cm^{-1} to more typical modern resolutions of better than 0.008 cm^{-1} is not possible.

6 Conclusions

The MANTRA UDenver FTS flight results are a limited success. The sunset occultation produced profiles for several molecules, and those profiles are largely consistent with the other measurements that are a part of the MANTRA effort, as well as measurements of other types. The CFC profiles are higher than expected, and remain unexplained. The HNO₃ profile is consistent with our expectations for northern mid-latitudes, as

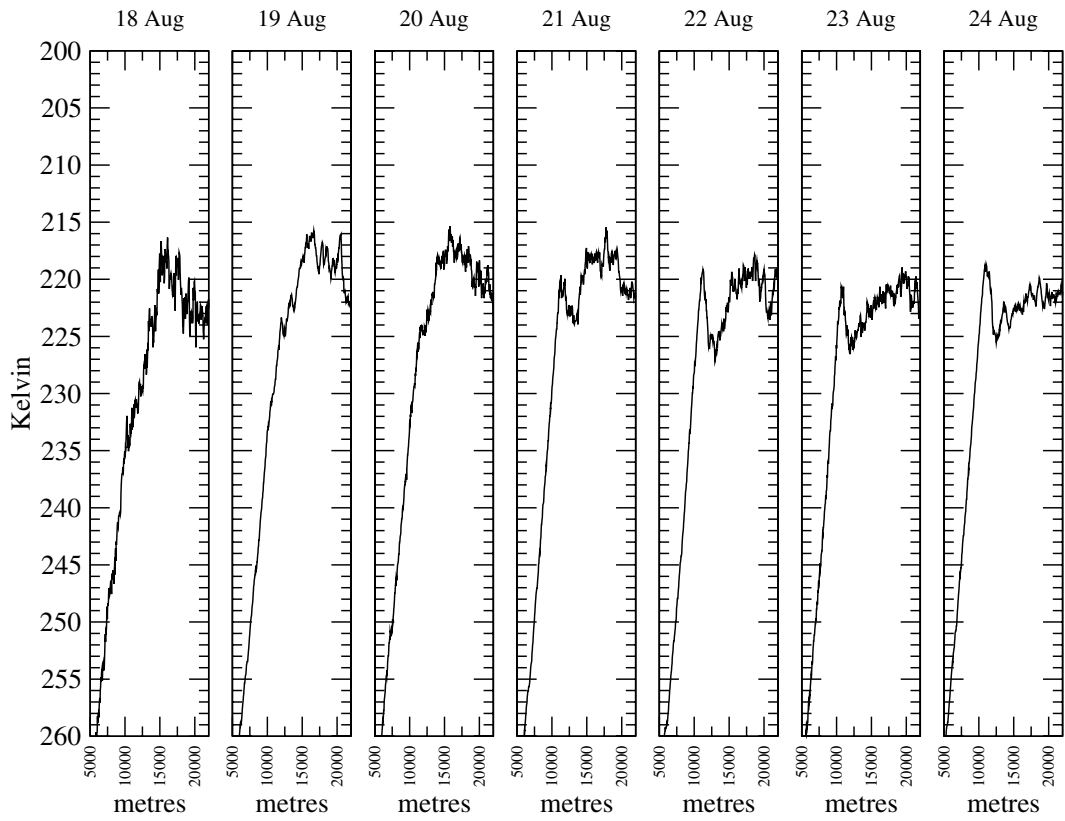


Fig. 6 Development of temperature layer with time.

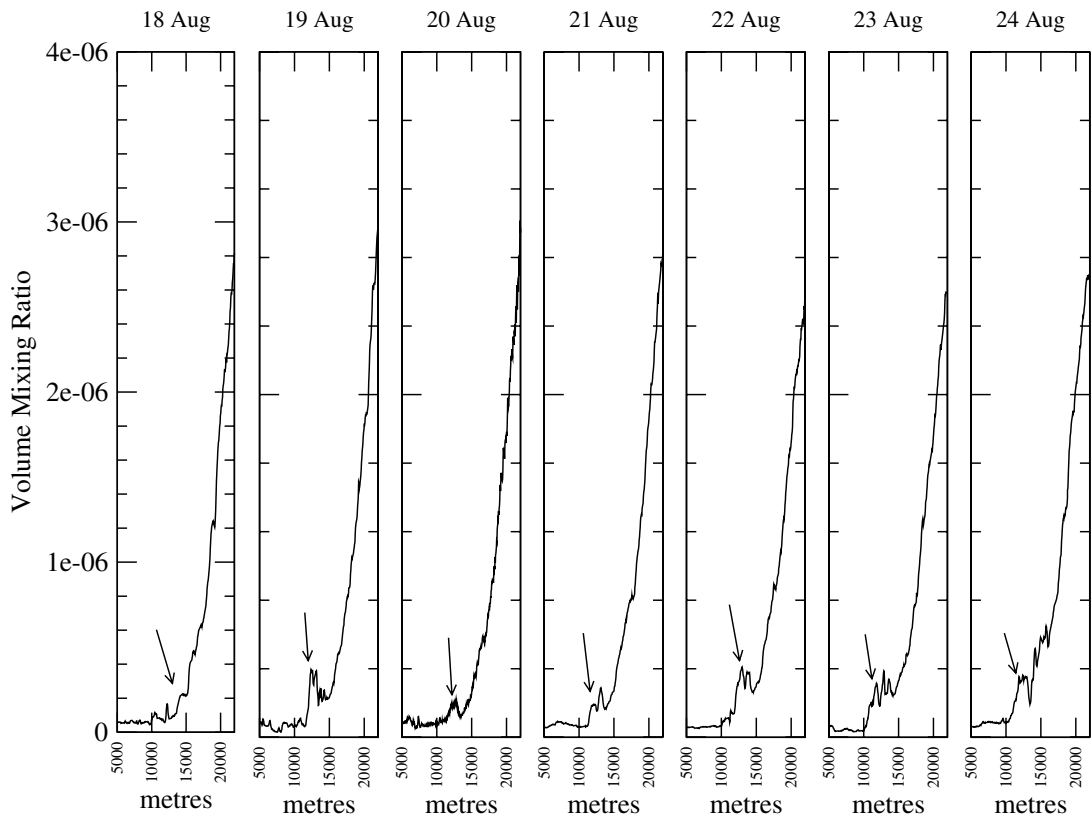


Fig. 7 Layer evolution as seen in ozone vmr.

are the HCl, CH₄ and N₂O profiles. Even though historical comparisons with similar instruments are of limited value given the problems during flight, these measurements provide a useful tool for verifying the results of the cooled radiometers that also flew as part of this mission, and the knowledge gained from this intercomparison will provide a basis from which the historical radiometer record can be re-analysed.

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